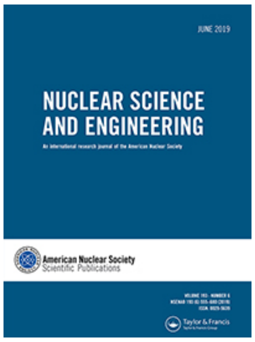

Nuclear Science and Engineering

ISSN: 0029-5639 (Print) 1943-748X (Online) Journal homepage: https://www.tandfonline.com/loi/unse20

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NUCLEAR SCIENCE AND ENGINEERING · VOLUME 193 · 555–600 · JUNE 2019

DOI: https://doi.org/10.1080/00295639.2018.1553910

Application of the Second-Order Comprehensive Adjoint Sensitivity Analysis Methodology to Compute First- and Second-Order Sensitivities of Flux Functionals in a Multiplying System with Source

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Received July 30, 2018
Accepted for Publication November 27, 2018

Abstract — *This work presents an application of the Second-Order Adjoint Sensitivity Analysis Methodology (2nd-ASAM) to the neutron transport Boltzmann equation that models a multiplying subcritical system comprising a nonfission neutron source to compute efficiently and exactly all of the first- and second-order functional derivatives (sensitivities) of a detector's response to all of the model's parameters, including isotopic number densities, microscopic cross sections, fission spectrum, sources, and detector response function. As indicated by the general theoretical considerations underlying the 2nd-ASAM, the number of computations required to obtain the first and second orders increases linearly in augmented Hilbert spaces as opposed to increasing exponentially in the original Hilbert space. The results presented in this work are currently being implemented in several production-oriented three-dimensional neutron transport code systems for analyzing specific subcritical systems.*

Keywords — *Second-Order Adjoint Sensitivity Analysis Methodology, neutron transport in multiplying systems with source, reaction rate detector response, first-order response sensitivities, second-order response sensitivities.*

I. INTRODUCTION

The computation of second-order response sensitivities to model parameters is motivated by the need to overcome the linearization limitation that is implicit in the use of first-order sensitivities. During the 1970s, the field of reactor physics provided pioneering works[1–5] for computing selected second-order response sensitivities of the system's effective multiplication factor and reaction rate ratios using the adjoint neutron transport and/or diffusion equations. These works generally indicated that the second-order sensitivities of such responses to cross-section perturbations were computationally expensive to obtain, requiring $O\left(N_\alpha^2\right)$ large-scale computations per response for a system comprising N_α model parameters, and were smaller than the corresponding first-order sensitivities. Such indications may have led to a diminishing interest in developing efficient methods for computing second-order response sensitivities for nuclear engineering systems.

While the interest in computing second-order response sensitivities practically vanished in the nuclear engineering field in the 1990s, interest in this topic became increasingly evident in other fields, driven mostly by the knowledge that second-order (Hessian) sensitivity information accelerates the convergence of optimization algorithms. In structural mechanics,[6] for example, interest has been focused primarily on the developing adjoint methods for computing second-order sensitivity of structural responses to variations of structural stiffness parameters. In atmospheric sciences,[7,8] second-order adjoint models were used to compute products between the Hessian of the cost functional and a vector (representing a perturbation in sensitivity analysis, a search direction in optimization, an

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eigenvector, etc.) to perform sensitivity analysis of the cost function with respect to distributed observations, to study the evolution of the condition number (the ratio of the largest to smallest eigenvalues) of the Hessian during minimization, and for sensitivity studies in three-dimensional atmospheric chemical transport models. In the context of parametric circuit analysis and optimization,[9] second-order sensitivities for linear circuits were also computed, albeit approximately. The availability (or unavailability) of exactly computed second-order sensitivities affects significantly many fields (e.g., optimization, data assimilation/adjustment, model calibration and validation, predictive modeling, and convergence of many numerical methods).

The methods used in the works mentioned above were all developed for specific, rather than general, applications for which they usually estimated, rather than computed exactly and inclusively, second-order response sensitivities to the model's parameters. Since the availability (or unavailability) of exactly computed second-order sensitivities affects significantly many fields (e.g., optimization, data assimilation/adjustment, model calibration and validation, predictive modeling, and convergence of many numerical methods), Cacuci[10–12] developed the generally applicable Second-Order Adjoint Sensitivity Analysis Methodology (2nd-ASAM). The 2nd-ASAM computes exactly and most efficiently all of the second-order functional derivatives of model responses to parameters and simultaneously verifies them intrinsically by computing all of the mixed partial sensitivities twice, using independently derived formulations. The application of the 2nd-ASAM for nonlinear systems[11] has been illustrated by means of a nonlinear heat conduction benchmark problem.[13] Furthermore, the 2nd-ASAM for linear systems[10,12] was applied to an illustrative linear neutron diffusion problem[14] aimed at highlighting the essential contributions of the second-order sensitivities of a detector response to changes in the underlying neutron cross sections. This illustrative problem[14] has shown that most of the second-order relative detector sensitivities to cross sections were actually larger than the corresponding first-order relative sensitivities, contrary to the tacit assumption that second-order sensitivities to cross sections are negligible in neutron diffusion problems, which was prevalent in sensitivity analysis works in the 1990s. In particular, the second-order sensitivities were shown[14] to be responsible for causing (a) asymmetries in the response distribution and (b) the expected value of the response to differ from the computed nominal value of the response. Neglecting the second-order sensitivities would nullify the third-order response correlations and hence would nullify the skewness of the response distribution. Consequently, any events occurring in a response's long and/or short tails, which are characteristic of rare but decisive events (e.g., major accidents and catastrophes), would likely be missed.

The 2nd-ASAM for linear systems[10,12] has also been applied[15,16] to compute the second-order sensitivities of the temperature distributions within a model of a test section comprising a heated rod surrounded by lead-bismuth eutectic coolant. For this model, the 6 first-order sensitivities and 21 distinct second-order sensitivities for the temperature distribution at any location within the heated rod (and/or on its surface), and a similar number of first- and second-order sensitivities for the temperature distribution at any location within the coolant, were computed using only seven independent 2nd-ASAM computations. For the thermal-hydraulic parameters used in the test section benchmark, having mean values and standard deviations typical of the conditions computed in the preliminary conceptual design of the G4M reactor,[16] the second-order sensitivities caused the temperature distributions within the rod, on the rod's surface, and in the coolant to become non-Gaussian, asymmetric, and skewed toward temperatures higher than the respective mean temperatures, as all three temperature distributions turned out to have positive skewnesses. In particular, the temperature distribution in the heated rod was skewed significantly toward higher temperatures, indicating that the conventional Gaussian-based metrics are not applicable for performing conventional risk analysis for this important safety margin indicator.

The 2nd-ASAM for linear systems[10,12] has also been applied by Cacuci and Favorite[17] to compute the second-order sensitivities of uncollided particle contributions to radiation detector responses, demonstrating once again its efficiency and accuracy. For a multiregion two-dimensional cylindrical benchmark problem, all of the benchmark's 18 first-order sensitivities and 224 second-order sensitivities of a detector's response with respect to the system's isotopic number densities, microscopic cross sections, source emission rates, and detector response function were obtained exactly by requiring only 12 adjoint large-scale transport computations. In contradistinction, 877 large-scale transport computations would have been needed to compute the respective sensitivities using central finite differences, and this number does not include the additional calculations that would have been required to find appropriate values of the parameter perturbations to use for the respective central difference expressions.

The present work extends significantly the results presented in Ref. 17 by applying the 2nd-ASAM to the neutron transport equation that models a multiplying subcritical system comprising a nonfission neutron source. Section II of this work recalls succinctly the Boltzmann transport equation describing the transport of neutrons within a finite multiplying medium with an internal nonfission source, defining this physical system's parameters and responses. Section III presents the construction of the First-Level Adjoint Sensitivity System (1st-LASS) for the transport equation. The 1st-LASS is used for the efficient computation of the first-order response sensitivities to variations in model parameters, and it serves as the basis for the construction of the Second-Level Adjoint Sensitivity System (2nd-LASS). The actual construction of the 2nd-LASS for the transport equation is presented in Sec. IV, which also presents the specific expressions for computing exactly and efficiently all of the second-order response sensitivities to variations in model parameters. Section V summarizes and concludes this work.

II. THE NEUTRON TRANSPORT EQUATION MODELING A MULTIPLYING SYSTEM WITH AN EXTERNAL SOURCE

The physical system considered in this work is a finite medium of convex volume V that contains fission and nonfission sources of neutrons. The system's outer boundary, denoted as ∂V, is considered to be perfectly well known, and the system is considered to be placed in vacuum in order to simplify the mathematical treatment by disregarding possible effects of boundary perturbations; such perturbations will be considered in subsequent work. The distribution of neutrons in such a system is modeled using the standard form of the time-independent integro-differential Boltzmann transport equation:

$$L(\boldsymbol{\alpha})\varphi(\mathbf{r},\boldsymbol{\Omega},E) \triangleq \boldsymbol{\Omega}\bullet\nabla\varphi(\mathbf{r},\boldsymbol{\Omega},E)+\Sigma_t(\mathbf{t};\mathbf{r},E)\,\varphi(\mathbf{r},\boldsymbol{\Omega},E)$$
$$-\int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\,\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})\,\varphi(\mathbf{r},\boldsymbol{\Omega}',E')$$
$$-\int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\,\chi(\mathbf{p};\mathbf{r},E'\to E)\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\varphi(\mathbf{r},\boldsymbol{\Omega}',E')$$
$$=Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)\ , \tag{1}$$

subject to the customary vacuum boundary condition, which specifies that there is no incoming flux of particles:

$$\varphi(\mathbf{r}_s,\boldsymbol{\Omega},E)=0,\mathbf{r}_s\in\partial V,\boldsymbol{\Omega}\cdot\mathbf{n}<0,0<E<\infty\ , \tag{2}$$

where $\mathbf{n}$ denotes the unit outward normal vector at any point $\mathbf{r}_s\in\partial V$ on the body's outer surface ∂V.

The macroscopic cross sections $\Sigma_t(\mathbf{t};\mathbf{r},E)$, $\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})$, and $\nu\Sigma_f(\mathbf{f};\mathbf{r},E)$; the neutron fission spectrum $\chi(\mathbf{p};\mathbf{r},E'\to E)$; and the source $Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)$ generally depend not only on the spatial variable $\mathbf{r}$, on the energy variable E, and possibly on the solid angle $\boldsymbol{\Omega}$ but also on imperfectly known scalar-valued model parameters such as atomic number densities, microscopic cross sections, and weighting functions. Specifically, the macroscopic total cross section $\Sigma_t(\mathbf{t};\mathbf{r},E)$ is considered to depend on J_t imprecisely known scalar-valued model parameters denoted as $t_i,\ i=1,\cdots,J_t$, which are considered to be the components of a vector of model parameters defined as

$$\mathbf{t}\triangleq[t_1,\ldots,t_{J_t}]^\dagger\ . \tag{3}$$

Throughout this work, the dagger symbol (†) is used to denote transposition. Similarly, the macroscopic scattering cross section $\Sigma_s(\mathbf{s};E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})$ is considered to depend on J_s imprecisely known scalar-valued model parameters denoted as $s_i,\ i=1,\ldots,J_s$, while the effective macroscopic fission cross section $\nu\Sigma_f(\mathbf{f};\mathbf{r},E')$ is considered to depend on J_f imprecisely known scalar-valued model parameters denoted as $f_i,\ i=1,\ldots,J_f$, which are considered to be the components of two vectors defined, respectively, as follows:

$$\mathbf{s}\triangleq[s_1,\ldots,s_{J_s}]^\dagger \tag{4}$$

and

$$\mathbf{f}\triangleq[f_1,\ldots,f_{J_f}]^\dagger\ . \tag{5}$$

Furthermore, the fission spectrum $\chi(\mathbf{p};\mathbf{r},E'\to E)$ is considered to depend on J_p imprecisely known scalar-valued parameters denoted as $p_i,\ i=1,\ldots,J_p$, while the source $Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)$ is considered to depend on J_q imprecisely known scalar-valued parameters denoted as $q_i,\ i=1,\ldots,J_q$, which are considered to be the component two vectors of model parameters defined, respectively, as follows:

$$\mathbf{p}\triangleq[p_1,\ldots,p_{J_p}]^\dagger \tag{6}$$

and

$$\mathbf{q}\triangleq[q_1,\ldots,q_{J_q}]^\dagger\ . \tag{7}$$

The system response considered in this work is a scalar-valued linear functional of the flux, denoted as $R(\boldsymbol{\alpha}, \varphi)$, which models a detector response of the form

$$R(\boldsymbol{\alpha}, \varphi) \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)\, \varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \ , \quad (8)$$

where $\Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)$ denotes a (macroscopic cross-section–like) function that models the interaction of the detector with the incident particles and where the vector $\boldsymbol{\alpha}$ will be defined in Eq. (12). In general, $\Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)$ depends not only on the independent variables $\mathbf{r}, \boldsymbol{\Omega}, E$ but also on J_d imprecisely known scalar-valued model parameters that are considered to be components of the vector $\mathbf{d}$, defined as

$$\mathbf{d} \triangleq [d_1, \ldots, d_{J_d}]^\dagger \ . \quad (9)$$

System responses of particular interest are (1) the scalar flux at a spatial location $\mathbf{r}_d$, in which case

$$\Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E) = \delta(\mathbf{r} - \mathbf{r}_d) \ , \quad (10)$$

where $\mathbf{r}_d$ represents the detector's location, and (2) the partial current density at a spatial location $\mathbf{r}_d$, in which case

$$\Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E) = \boldsymbol{\Omega} \cdot \mathbf{n} \delta(\mathbf{r} - \mathbf{r}_d) \ . \quad (11)$$

Since the response $R(\boldsymbol{\alpha}, \varphi)$ defined in Eq. (8) depends explicitly and/or implicitly, through the flux $\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)$, on all of the imprecisely known model parameters defined in Eqs. (3) through (9), it will be convenient for subsequent mathematical derivations to consider these imprecisely known scalar-valued model parameters as the components of a vector of model parameters, denoted as $\boldsymbol{\alpha}$ and defined as follows:

$$\boldsymbol{\alpha} \triangleq [\alpha_1, \ldots, \alpha_{J_\alpha}]^\dagger \triangleq [\mathbf{t}, \mathbf{s}, \mathbf{f}, \mathbf{p}, \mathbf{q}, \mathbf{d}]^\dagger,$$
$$J_\alpha \triangleq J_t + J_s + J_f + J_p + J_q + J_d \ , \quad (12)$$

where J_α denotes the total number of imprecisely known scalar model parameters.

The nominal values of the model parameters will be denoted as $\boldsymbol{\alpha}^0 \triangleq \left[\alpha_1^0, \ldots, \alpha_{J_\alpha}^0\right]^\dagger$. Throughout this work, the superscript 0 will be used, as needed, to denote nominal or mean values. The nominal value of the flux, denoted as $\varphi^0(\mathbf{r}, \boldsymbol{\Omega}, E)$, is obtained by solving Eqs. (1) and (2) using the nominal parameter values $\boldsymbol{\alpha}^0$. The nominal value of the detector response, denoted as $R(\varphi^0, \boldsymbol{\alpha}^0)$, is obtained by evaluating Eq. (8) at the nominal flux and parameter values.

III. THE FIRST-LEVEL FORWARD AND ADJOINT SENSITIVITY SYSTEMS FOR COMPUTING FIRST-ORDER RESPONSE SENSITIVITIES TO VARIATIONS IN MODEL PARAMETERS

The total sensitivity, denoted as $\delta R(\boldsymbol{\alpha}^0, \varphi^0; \delta\varphi, \delta\boldsymbol{\alpha})$, of the detector response defined in Eq. (8) to variations $\delta\boldsymbol{\alpha} \triangleq [\delta\alpha_1, \ldots, \delta\alpha_{N_\alpha}]^\dagger$ in the model parameters, around the nominal values $\boldsymbol{\alpha}^0$, is obtained by applying the definition of the Gateaux- (G-) differential to Eq. (8) at the nominal parameter and flux values, to obtain

$$\begin{aligned}\delta R(\boldsymbol{\alpha}^0, \varphi^0; \delta\varphi, \delta\boldsymbol{\alpha}) &\triangleq \frac{d}{d\varepsilon}\left\{\int dV \int_{4\pi} d\boldsymbol{\Omega}\right.\\ &\times \int_0^\infty dE\left[\Sigma_d^0(\mathbf{d}^0; \mathbf{r}, \boldsymbol{\Omega}, E) + \varepsilon\delta\Sigma_d(\mathbf{r}, \boldsymbol{\Omega}, \mathrm{E})\right]\\ &\times \left[\varphi^0(\mathbf{r}, \boldsymbol{\Omega}, E) + \varepsilon\delta\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\right]\varepsilon = 0\\ &= \left\{\delta R(\boldsymbol{\alpha}^0, \varphi^0; \delta\boldsymbol{\alpha})\right\}_{dir} + \left\{\delta R(\boldsymbol{\alpha}^0, \varphi^0; \delta\varphi)\right\}_{ind}, \end{aligned} \quad (13)$$

where the direct-effect term is defined as

$$\left\{\delta R(\boldsymbol{\alpha}^0, \varphi^0; \delta\boldsymbol{\alpha})\right\}_{dir} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \times \varphi^0(\mathbf{r}, \boldsymbol{\Omega}, E)[\delta\Sigma_d(\mathbf{r}, \boldsymbol{\Omega}, \mathrm{E})] \ , \quad (14)$$

and where the indirect-effect term is defined as

$$\left\{\delta R(\boldsymbol{\alpha}^0, \varphi^0; \delta\boldsymbol{\alpha})\right\}_{ind} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \times \Sigma_d^0(\mathbf{d}^0; \mathbf{r}, \boldsymbol{\Omega}, E)\ \delta\varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \ . \quad (15)$$

Since the nominal value $\varphi^0(\mathbf{r}, \boldsymbol{\Omega}, E)$ of the flux is known after having solved Eqs. (1) and (2) using the nominal parameter values $\boldsymbol{\alpha}^0$, it follows that the direct-effect term defined in Eq. (14) can already be computed at this stage. In contradistinction, however, the indirect-effect term defined in Eq. (15) can be computed only after having determined the flux variation $\delta\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)$, which is the solution of the First-Level Forward Sensitivity System[10–12,18,19] (1st-LFSS), which is derived, in turn, by G-differentiating Eqs. (1) and (2) to obtain

$$L^{(1)}\left(\boldsymbol{\alpha}^0\right)\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E) = Q^{(1)}\left(\boldsymbol{\alpha}^0,\varphi^0;\delta\boldsymbol{\alpha}\right) , \tag{16}$$

together with boundary condition

$$\delta\varphi(\mathbf{r}_s,\boldsymbol{\Omega},E) = 0, \mathbf{r}_s \in \partial V, \boldsymbol{\Omega}\cdot\mathbf{n} < 0, 0 < E < \infty . \tag{17}$$

The operator $L^{(1)}\left(\boldsymbol{\alpha}^0\right)$ and the source term $Q^{(1)}\left(\boldsymbol{\alpha}^0,\varphi^0;\delta\boldsymbol{\alpha}\right)$, which appear in Eq. (16), are defined as follows:

$$L^{(1)}\left(\boldsymbol{\alpha}^0\right)\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E) \triangleq \boldsymbol{\Omega}\cdot\nabla[\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)] + \Sigma_t^0\left(\mathbf{t}^0;\mathbf{r},E\right) \times [\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)] - \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\Sigma_s^0\left(\mathbf{s}^0;\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega}\right) \times [\delta\varphi(\mathbf{r},\boldsymbol{\Omega}',E')] - \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\chi^0\left(\mathbf{p}^0;\mathbf{r},E'\to E\right) \times \left[\nu^0\Sigma_f^0\left(\mathbf{f}^0;\mathbf{r},E'\right)\right][\delta\varphi(\mathbf{r},\boldsymbol{\Omega}',E')] \tag{18}$$

and

$$Q^{(1)}\left(\boldsymbol{\alpha}^0,\varphi^0;\delta\boldsymbol{\alpha}\right) \triangleq \delta Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E) - \delta\Sigma_t(\mathbf{t};\mathbf{r},E)\varphi^0(\mathbf{r},\boldsymbol{\Omega},E) + \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\varphi^0(\mathbf{r},\boldsymbol{\Omega}',E')[\delta\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})] + \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\delta\chi(\mathbf{p};\mathbf{r},E'\to E)\varphi^0(\mathbf{r},\boldsymbol{\Omega}',E') \times \left[\nu^0\Sigma_f^0(\mathbf{f};\mathbf{r},E')\right] + \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\chi^0(\mathbf{p};\mathbf{r},E'\to E) \times \varphi^0(\mathbf{r},\boldsymbol{\Omega}',E')\delta\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right] . \tag{19}$$

Although $L^{(1)}\left(\boldsymbol{\alpha}^0\right) \equiv L\left(\boldsymbol{\alpha}^0\right)$, as expected, and as confirmed by comparing Eqs. (18) and (1), solving the 1st-LFSS defined by Eqs. (16) and (17) is computationally expensive since the 1st-LFSS would need to be solved anew for every variation $\delta\alpha_i,\ i = 1,\dots,N_\alpha$ in the model parameters, which affects the source term $Q^{(1)}\left(\boldsymbol{\alpha}^0,\varphi^0;\delta\boldsymbol{\alpha}\right)$. The computationally expensive evaluation of the indirect-effect term by using Eq. (15) can be avoided[10–12,18,19] by expressing this indirect-effect term in terms of the solution of the 1st-LASS, which is constructed by implementing the following sequence of steps:

Step 1: In the space $L_2(V\times\Omega\times E)$ of square-integrable functions, define the inner product $\langle u(\mathbf{r},\boldsymbol{\Omega},E), v(\mathbf{r},\boldsymbol{\Omega},E)\rangle_{(1)}$ of two functions $u(\mathbf{r},\boldsymbol{\Omega},E) \in L_2(V\times\Omega\times E)$ and $v(\mathbf{r},\boldsymbol{\Omega},E) \in L_2(V\times\Omega\times E)$ as follows:

$$\langle u(\mathbf{r},\boldsymbol{\Omega},E), v(\mathbf{r},\boldsymbol{\Omega},E)\rangle_{(1)} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\; u(\mathbf{r},\boldsymbol{\Omega},E)v(\mathbf{r},\boldsymbol{\Omega},E) . \tag{20}$$

Step 2: Denote the Hilbert space endowed with the inner product defined in Eq. (20) as $\mathsf{H}_{(1)}$, and form the inner product of Eq. (18) with a yet undefined function $\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$ to obtain

$$\left\langle\psi^{(1)}, L^{(1)}\left(\boldsymbol{\alpha}^0\right)\delta\varphi\right\rangle_{(1)} = \left\langle\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E), Q^{(1)}\left(\boldsymbol{\alpha}^0,\varphi^0;\delta\boldsymbol{\alpha}\right)\right\rangle_{(1)} . \tag{21}$$

Step 3: In the Hilbert space $H_{(1)}$, define the formal adjoint operator, denoted as $A^{(1)}(\boldsymbol{\alpha})$, of $L^{(1)}(\boldsymbol{\alpha})$, through relationship

$$\langle\psi^{(1)},\, L^{(1)}\left(\boldsymbol{\alpha}^0\right)\delta\varphi\rangle_{(1)} = \langle\delta\varphi,\, A^{(1)}\left(\boldsymbol{\alpha}^0\right)\psi^{(1)}\rangle_{(1)} + P^{(1)}\left[\delta\varphi,\,\psi^{(1)}\right] , \tag{22}$$

where

$$A^{(1)}(\boldsymbol{\alpha})\psi^{(1)} \triangleq -\boldsymbol{\Omega}\bullet\nabla\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) + \Sigma_t(\mathbf{t};\mathbf{r},E)\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) - \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') - \nu\Sigma_f(\mathbf{f};\mathbf{r},E)\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\chi(\mathbf{p};\mathbf{r},E\to E')\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') , \tag{23}$$

and where the bilinear concomitant $P^{(1)}\left[\delta\varphi,\,\psi^{(1)}\right]$ is defined on the phase-space boundary $(\partial V\times\partial\Omega)$ as follows:

$$P^{(1)}\left[\delta\varphi,\,\psi^{(1)}\right] \triangleq \int_0^\infty dE \int_{\boldsymbol{\Omega}\cdot\mathbf{n}<0} d\boldsymbol{\Omega} \int_{\partial V} |\boldsymbol{\Omega}\cdot\mathbf{n}|\delta\varphi \times (\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)dA - \int_0^\infty dE \int_{\boldsymbol{\Omega}\cdot\mathbf{n}>0} d\boldsymbol{\Omega} \int_{\partial V} \boldsymbol{\Omega}\cdot\mathbf{n}\delta\varphi \times (\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)dA . \tag{24}$$

In order to simplify the notation, the superscript 0 denoting nominal values will be omitted henceforth. This simplification should not cause any loss of clarity since it will become clear from the context which quantities are to be evaluated/computed using the nominal values for the model parameters.

Step 4: Identify the term on the left side of Eq. (22) with the indirect-effect term defined in Eq. (15), and use Eq. (22) in conjunction with the boundary conditions given in Eq. (17) to construct the following 1st-LASS for the first-level adjoint function $\psi^{(1)}(r, \mathbf{\Omega})$:

$$A^{(1)}(\boldsymbol{\alpha}^0)\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E) = \Sigma_d(\mathbf{d}^0; \mathbf{r}, \mathbf{\Omega}, E) \ , \tag{25}$$

together with adjoint boundary condition

$$\psi^{(1)}(\mathbf{r}_s, \mathbf{\Omega}, E) = 0 \ , \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n} > 0 \ , \tag{26}$$

which is selected in order to cause the bilinear concomitant $P^{(1)}[\delta\varphi, \ \psi^{(1)}]$ in Eq. (24) to vanish.

Step 5: Use the 1st-LFSS defined by Eqs. (25) and (26) together with Eqs. (21) and (22) to obtain expression for the indirect-effect term [see Eq. (15)], in terms of the first-level adjoint function $\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)$:

$$\left\{\delta R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)}, \ \delta\boldsymbol{\alpha})\right\}_{ind} = \left\langle \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E), \ Q^{(1)}(\boldsymbol{\alpha}^0, \varphi^0; \delta\boldsymbol{\alpha})\right\rangle_{(1)} \ . \tag{27}$$

The Hilbert space $\mathsf{H}_{(1)}$, endowed with the customary inner product defined in Eq. (20), yields the customary adjoint Boltzmann operator shown in Eq. (23). The use of $\mathsf{H}_{(1)}$ allows the 1st-LASS to be solved by only slightly modifying the numerical methods used for solving the original Eqs. (1) and (2), namely, by reversing the sign of the solid angle and reversing the order of integration over the energy variable. As is also well known, the 1st-LASS is independent of parameter variations, so it needs to be solved just once for each particular form of the source term $\Sigma_d(\mathbf{d}; \mathbf{r}, \mathbf{\Omega}, E)$ to obtain the corresponding first-level adjoint function $\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)$. Subsequently, the indirect-effect term is computed efficiently, once $\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)$ is available, by performing the integrations (quadratures) indicated in Eq. (27).

Replacing Eqs. (27) and (14) in Eq. (13) yields the following expression for the total first-order response sensitivity in terms of the first-level adjoint function $\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)$:

$$\begin{aligned}\delta R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)}, \delta\boldsymbol{\alpha}) &= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE \ \delta\Sigma_d(\mathbf{r}, \mathbf{\Omega}, E)\varphi(\mathbf{r}, \mathbf{\Omega}, E) \\ &+ \left\langle \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E), \ Q^{(1)}(\boldsymbol{\alpha}^0, \varphi^0; \delta\boldsymbol{\alpha})\right\rangle_{(1)} \\ &\triangleq \sum_{m_1=1}^{N_\alpha} \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial\alpha_{m_1}} \delta\alpha_{m_1} \ .\end{aligned} \tag{28}$$

The partial first-order response sensitivities, denoted as $\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})/\partial\alpha_{m_1}, m_1 = 1, \ldots, N_\alpha$, to a generic parameter α_{m_1}, are obtained from Eq. (28) by identifying the quantities that multiply the various parameter variations $\delta\alpha_{m_1}$ have the following expressions:

$$\begin{aligned}\text{For } j = 1, \ldots, J_t : \ \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial\alpha_j} &\triangleq \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial t_j} \\ &= -\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE \ \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)\varphi(\mathbf{r}, \mathbf{\Omega}, E) \\ &\times \frac{\partial\Sigma_t(\mathbf{t}; \mathbf{r}, E)}{\partial t_j} \ ;\end{aligned} \tag{29}$$

$$\begin{aligned}\text{For } j = 1, \ldots, J_s : \ \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial\alpha_{J_t+j}} &\triangleq \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial s_j} \\ &= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE \ \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)\int_{4\pi} d\mathbf{\Omega}' \\ &\times \int_0^\infty dE' \ \frac{\partial\Sigma_s(\mathbf{s}; \mathbf{r}, E' \to E, \mathbf{\Omega}' \to \mathbf{\Omega})}{\partial s_j}\varphi(\mathbf{r}, \mathbf{\Omega}', E') \ ;\end{aligned} \tag{30}$$

$$\begin{aligned}\text{For } j = 1, \ldots, J_f : \ \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial\alpha_{J_t+J_s+j}} &\triangleq \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial f_j} \\ &= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE \ \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)\int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE' \\ &\times \chi(\mathbf{p}; \mathbf{r}, E' \to E)\frac{\partial[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')]}{\partial f_j}\varphi(\mathbf{r}, \mathbf{\Omega}', E') \ ;\end{aligned} \tag{31}$$

$$\begin{aligned}\text{For } j = 1, \ldots, J_p : \ \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial\alpha_{J_t+J_s+J_f+j}} &\triangleq \frac{\partial R(\boldsymbol{\alpha}, \varphi; \ \psi^{(1)})}{\partial p_j} \\ &\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE \ \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)\int_{4\pi} d\mathbf{\Omega}' \\ &\times \int_0^\infty dE' \ \frac{\partial\chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_j}\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E') \\ &\times \varphi(\mathbf{r}, \mathbf{\Omega}', E') \ ;\end{aligned} \tag{32}$$

$$\text{For } j = 1, ..., J_q : \frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial \alpha_{J_t+J_s+J_f+J_p+j}} \triangleq \frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial q_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \frac{\partial Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial q_j} \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \;\; ; \tag{33}$$

and

$$\text{For } j = 1, ..., J_d : \frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial \alpha_{J_t+J_s+J_f+J_p+J_q+j}} \triangleq \frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial d_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \frac{\partial \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_j} \varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \;\; . \tag{34}$$

The same model parameter could appear in the definitions of more than one macroscopic cross section. For example, the isotopic number density of some element, generically denoted as N_i, could be an imprecisely known model parameter that might appear in the definitions of the total, scattering, and fission macroscopic cross sections, as well as in the source term $Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)$. In such a case, the sensitivity of the response to the model parameter N_i would be the sum of the corresponding partial sensitivities computed from Eqs. (29), (30), (31), and (33), namely,

$$\begin{aligned}
\frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial N_i} &\triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \frac{\partial Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial N_i} \\
&- \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \frac{\partial \Sigma_t(\mathbf{t}; \mathbf{r}, E)}{\partial N_i} \\
&+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \; \frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E' \to E, \boldsymbol{\Omega}' \to \boldsymbol{\Omega})}{\partial N_i} \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \\
&+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \frac{\partial [\nu \Sigma_f(\mathbf{f}; \mathbf{r}, E')]}{\partial N_i} \chi(\mathbf{p}; \mathbf{r}, E' \to E)\varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \;\; .
\end{aligned} \tag{35}$$

IV. THE SECOND-LEVEL FORWARD AND ADJOINT SENSITIVITY SYSTEMS FOR COMPUTING SECOND-ORDER RESPONSE SENSITIVITIES TO VARIATIONS IN MODEL PARAMETERS

The second-order response sensitivities will be obtained by applying the 2nd-ASAM developed by Cacuci,[10–12] which relies on the construction of a 2nd-LASS for each of the first-order sensitivities defined by Eqs. (29) through (34).

IV.A. Computation of the Second-Order Sensitivities $\partial^2 R(\alpha,\varphi;\psi^{(1)})/\partial t_j \partial \alpha_{m_2}$, $j = 1,..., J_t$; $m_2 = 1,..., J_\alpha$

The second-order sensitivities $\partial^2 R(\boldsymbol{\alpha}, \varphi; \psi^{(1)}) / (\partial t_j)\,(\partial \alpha_{m_2})$, $j = 1, ..., J_t$; $m_2 = 1, ..., J_\alpha$, are obtained by determining the G-differential of the first-order sensitivity given in Eq. (29), which yields the following expression:

$$\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial t_j}\right] = \left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial t_j}\right]\right\}_{dir} + \left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial t_j}\right]\right\}_{ind}, \; j = 1, ..., J_t \;\; , \tag{36}$$

where

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})}{\partial t_j}\right]\right\}_{dir} \triangleq - \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \sum_{m_2=1}^{J_t} \frac{\partial^2 \Sigma_t(\mathbf{t}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial t_j \partial t_{m_2}} \delta t_{m_2} \; j = 1, ..., J_t \tag{37}$$

and

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)}\right)}{\partial t_j}\right]\right\}_{ind}\triangleq-\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_0^\infty dE$$
$$\times\left[\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)\right]\varphi(\mathbf{r},\boldsymbol{\Omega},E)\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},E)}{\partial t_j}$$
$$-\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_0^\infty dE\,[\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)]\;\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},E)}{\partial t_j},\quad j=1,\ldots,J_t\;. \tag{38}$$

The direct-effect term defined in Eq. (37) can be computed immediately. On the other hand, the indirect-effect term defined in Eq. (38) can be computed only after having obtained the solution $\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)$ of the 1st-LFSS and the variation $\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$ in the first-level adjoint function $\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$. It has already been discussed in Sec. III that it is computationally expensive to obtain $\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)$ since the 1st-LFSS would need to be solved anew for every variation in the model parameters. Furthermore, the function $\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$ is the solution of the system of equations obtained by G-differentiating the 1st-LASS [see Eqs. (25) and (26)], namely,

$$A^{(1)}\left(\boldsymbol{\alpha}^0\right)\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)=Q^{(2)}\left(\boldsymbol{\alpha}^0,\psi^{(1)};\delta\boldsymbol{\alpha}\right) \tag{39}$$

and

$$\delta\psi^{(1)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0\;,\mathbf{r}_s\in\partial V,\boldsymbol{\Omega}\cdot\mathbf{n}>0\;, \tag{40}$$

where

$$Q^{(2)}\left(\boldsymbol{\alpha}^0,\psi^{(1)};\delta\boldsymbol{\alpha}\right)\triangleq\delta\Sigma_d(\mathbf{r},\boldsymbol{\Omega},E)-\delta\Sigma_t(\mathbf{r},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$+\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'\,[\delta\Sigma_s(\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')]\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')$$
$$+\delta\left[\nu\Sigma_f(\mathbf{r},E)\right]\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'\,\chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}$$
$$\times(\mathbf{r},\boldsymbol{\Omega}',E')+\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'$$
$$\times\delta\chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')\;. \tag{41}$$

It is evident from Eqs. (39) and (40) that the evaluation of the function $\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$ is just as expensive computationally as determining the variation $\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)$ by solving the 1st-LFSS. The system comprising Eqs. (39) and (40) is called[10–12] the Second-Level Forward Sensitivity System (2nd-LFSS). To avoid the need for solving the 2nd-LFSS, the indirect-effect term defined in Eq. (38) will be expressed in terms of a 2nd-LASS, which will be constructed by following the general principles introduced by Cacuci,[10–12] comprising the following sequence of steps.

Step 1: Define an inner product $\langle\mathbf{u}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E),\mathbf{v}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\rangle_{(2)}$ of two vector-valued functions $\mathbf{u}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\triangleq\left[u_1^{(2)}(\mathbf{r},\boldsymbol{\Omega},E),u_2^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\right]^\dagger$ and $\mathbf{v}^{(2)}(\mathbf{r},\,\boldsymbol{\Omega},\,E)\triangleq\left[v_1^{(2)}(\mathbf{r},\,\boldsymbol{\Omega},\,E),\,v_2^{(2)}(\mathbf{r},\,\boldsymbol{\Omega},\,E)\right]^\dagger$, with $u_1^{(2)}(\mathbf{r},\,\boldsymbol{\Omega},\,E)\in\mathsf{L}_2(V\times\boldsymbol{\Omega}\times E)$, $u_2^{(2)}(\mathbf{r},\,\boldsymbol{\Omega},\,E)\in\mathsf{L}_2(V\times\boldsymbol{\Omega}\times E)$, $v_1^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\in\mathsf{L}_2(V\times\boldsymbol{\Omega}\times E)$, and $v_2^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\in\mathsf{L}_2(V\times\boldsymbol{\Omega}\times E)$, as follows:

$$\langle\mathbf{u}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E),\mathbf{v}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\rangle_{(2)}$$
$$\triangleq\sum_{j=1}^{2}\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_0^\infty dE\,u_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)v_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\;. \tag{42}$$

Step 2: Define a Hilbert space, denoted as $\mathsf{H}_{(2)}$, which is endowed with the inner product defined in Eq. (42). For a matrix-valued linear operator $\mathbf{L}^{(2)}\triangleq\begin{pmatrix}L_{11}^{(2)}&L_{12}^{(2)}\\L_{21}^{(2)}&L_{22}^{(2)}\end{pmatrix}$, define its formal adjoint operator, denoted as $\mathbf{A}^{(2)}\triangleq\begin{pmatrix}A_{11}^{(2)}&A_{12}^{(2)}\\A_{21}^{(2)}&A_{22}^{(2)}\end{pmatrix}$, through the following relationship:

$$\langle\mathbf{v}^{(2)},\,\mathbf{L}^{(2)}\mathbf{u}^{(2)}\rangle_{(2)}=\langle\mathbf{u}^{(2)},\,\mathbf{A}^{(2)}\mathbf{v}^{(2)}\rangle_{(2)}+P^{(2)}\left[\mathbf{u}^{(2)},\,\mathbf{v}^{(2)}\right]\;, \tag{43}$$

where $P^{(2)}\left[\mathbf{u}^{(2)},\,\mathbf{v}^{(2)}\right]$ denotes the corresponding bilinear concomitant on the boundary $(\partial V\times\partial\boldsymbol{\Omega}\times\partial E)$.

Step 3: Apply the definition provided in Eq. (42) to form the inner product of Eqs. (39) and (16) with a yet undefined function $\boldsymbol{\psi}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\triangleq\left[\psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E),\psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\right]^\dagger$, where $\psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\in\mathsf{L}_2(V\times\boldsymbol{\Omega}\times E)$ and $\psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\in\mathsf{L}_2(V\times\boldsymbol{\Omega}\times E)$, to obtain

$$\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) A^{(1)}(\boldsymbol{\alpha}) \delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)$$
$$+ \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) L^{(1)}(\boldsymbol{\alpha}) \delta\varphi(\mathbf{r},\mathbf{\Omega},E)$$
$$= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right)$$
$$+ \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}) \ . \quad (44)$$

Step 4: Use the relation shown in Eq. (43) to recast the left side of Eq. (44) in the following form:

$$\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) A^{(1)}(\boldsymbol{\alpha}) \delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)$$
$$+ \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) L^{(1)}(\boldsymbol{\alpha}) \delta\varphi(\mathbf{r},\mathbf{\Omega},E)$$
$$= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E) \left[A^{(1)}(\boldsymbol{\alpha})\right]^* \psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$+ \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\ \delta\varphi(\mathbf{r},\mathbf{\Omega}) \left[L^{(1)}(\boldsymbol{\alpha})\right]^* \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$+ P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ \psi_{1,j}^{(2)},\psi_{2,j}^{(2)}\right] \ , \quad (45)$$

where the symbol $[\,]^*$ indicates adjoint and $P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ \psi_{1,j}^{(2)},\psi_{2,j}^{(2)}\right]$ denotes the corresponding bilinear concomitant on the domain's boundary, similar to the bilinear concomitant shown in Eq. (24).

Step 5: Use the boundary conditions shown in Eqs. (17) and (40), and impose on the function $\boldsymbol{\psi}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq \left[\psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E),\psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\right]^\dagger$ the boundary conditions $\psi_{1,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E) = 0, \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n} > 0$ and $\psi_{2,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E) = 0,\ \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n} > 0$, to cause the bilinear concomitant $P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ \psi_{1,j}^{(2)},\psi_{2,j}^{(2)}\right]$ in Eq. (45) to vanish.

Step 6: Noting that $\left[A^{(1)}(\boldsymbol{\alpha})\right]^* = L^{(1)}(\boldsymbol{\alpha}) = L(\boldsymbol{\alpha})$ and $\left[L^{(1)}(\boldsymbol{\alpha})\right]^* = A^{(1)}(\boldsymbol{\alpha})$ and identifying the right side of Eq. (45) with the indirect-effect term defined in Eq. (38) yield Eqs. (46) through (49):

$$L(\boldsymbol{\alpha}^0)\psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq \mathbf{\Omega}\cdot\nabla\psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$+ \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)\psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) - \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'$$
$$\times \Sigma_s^0(\mathbf{s}^0;\mathbf{r},E'\to E,\mathbf{\Omega}'\to\mathbf{\Omega})\psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E')$$
$$- \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\,\chi^0(\mathbf{p}^0;\mathbf{r},E'\to E)\left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E')\right]$$
$$\times \psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E') = -\varphi^0(\mathbf{r},\mathbf{\Omega},E)\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},E)}{\partial t_j},$$
$$j = 1,\ldots,J_t \ , \quad (46)$$

subject to boundary condition

$$\psi_{1,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E) = 0, \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n} < 0,$$
$$j = 1,\ldots,J_t \quad (47)$$

and

$$A^{(1)}(\boldsymbol{\alpha}^0)\psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq -\mathbf{\Omega}\cdot\nabla\psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$+ \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)\ \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$- \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\,\Sigma_s^0(\mathbf{s}^0;\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')$$
$$\times \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}',E') - \left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E)\right]\int_{4\pi} d\mathbf{\Omega}'$$
$$\times \int_0^\infty dE'\,\chi^0(\mathbf{p}^0;\mathbf{r},E\to E')\ \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}',E')$$
$$= -\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)\frac{\partial\Sigma_t(\mathbf{t}^0;\mathbf{r},E)}{\partial t_j},\ j = 1,\ldots,J_t \ , \quad (48)$$

subject to the following boundary condition

$$\psi_{2,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E) = 0\ , \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n} > 0,$$
$$j = 1,\ldots,J_t \ . \quad (49)$$

Equations (46) through (49) constitute the 2nd-LASS for the second-level adjoint function $\boldsymbol{\psi}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq \left[\psi_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\ ,\ \psi_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\right]\ ,\ j = 1,\ldots,J_t\ ,$ which will be used to evaluate the indirect-effect term defined in Eq. (38).

Step 7: Use Eqs. (46) and (48) together with Eqs. (41) through (45) in Eq. (38) to obtain the following expression for the indirect-effect term:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)}\right)}{\partial t_j}\right]\right\}_{ind} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \times Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}),\quad j=1,\ldots,J_t\ . \tag{50}$$

Step 8: Replacing the expressions of $Q^{(2)}\left(\boldsymbol{\alpha}^0,\psi^{(1)};\delta\boldsymbol{\alpha}\right)$ and $Q^{(1)}(\boldsymbol{\alpha};\varphi;\delta\boldsymbol{\alpha})$, respectively, in Eq. (50) and subsequently using Eqs. (50) and (37) in Eq. (36) yields

$$\begin{aligned}
\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)}\right)}{\partial t_j}\right] = &-\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)\varphi(\mathbf{r},\boldsymbol{\Omega},E)\left\{\sum_{m_2=1}^{J_t}\frac{\partial^2\Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_j\partial t_{m_2}}\delta t_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left\{\sum_{m_2=1}^{J_d}\frac{\partial\Sigma_d(\mathbf{d};\mathbf{r},\boldsymbol{\Omega},E)}{\partial d_{m_2}}\delta d_{m_2}\right\} \\
&-\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)\left\{\sum_{m_2=1}^{J_t}\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_{m_2}}\delta t_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')\left\{\sum_{m_2=1}^{J_s}\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}\delta s_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left\{\sum_{m_2=1}^{J_f}\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}\delta f_{m_2}\right\}\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \\
&\times\left\{\sum_{m_2=1}^{J_p}\frac{\partial\chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_{m_2}}\delta p_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,m_1}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left\{\sum_{m_2=1}^{J_q}\frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_{m_2}}\delta q_{m_2}\right\} \\
&-\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\varphi(\mathbf{r},\boldsymbol{\Omega},E)\left\{\sum_{m_2=1}^{J_t}\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_{m_2}}\delta t_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\left\{\sum_{m_2=1}^{J_s}\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_{m_2}}\delta s_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\left\{\sum_{m_2=1}^{J_p}\frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_{m_2}}\delta p_{m_2}\right\} \\
&+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\chi(\mathbf{p};\mathbf{r},E'\to E)\left\{\sum_{m_2=1}^{J_f}\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_{m_2}}\delta f_{m_2}\right\}, \\
&j=1,\ldots,J_t\ .
\end{aligned} \tag{51}$$

The second-order partial sensitivities $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)}\right)/\partial t_j\partial\alpha_{m_2},\ j=1,\ldots,J_t;\ m_2=1,\ldots,J_\alpha,$ can now be determined by identifying in Eq. (51) the quantities multiplying the parameter variations $\delta\alpha_{m_2}$, which yields the following expressions:

$$\text{for } j, m_2 = 1, ..., J_t : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_j \partial t_{m_2}} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial^2 \Sigma_t(\mathbf{t}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial t_j \partial t_{m_2}}$$
$$- \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \left[\psi_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) + \psi_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\right]\frac{\partial \Sigma_t(\mathbf{t}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial t_{m_2}}; \tag{52}$$

$$\text{for } j = 1, ..., J_t;\quad m_2 = 1, ..., J_s : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_j \partial s_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E')$$
$$\times \frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_{m_2}} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}'$$
$$\times \int_0^\infty dE'\ \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_{m_2}}\ ; \tag{53}$$

$$\text{for } j = 1, ..., J_t;\quad m_2 = 1, ..., J_f : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_j \partial f_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right]}{\partial f_{m_2}}$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\chi(\mathbf{p}; \mathbf{r}, E \to E')\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E') + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'$$
$$\times \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\chi(\mathbf{p}; \mathbf{r}, E \to E')\frac{\partial\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_{m_2}}\ ; \tag{54}$$

$$\text{for } j = 1, ..., J_t;\ m_2 = 1, ..., J_p : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_j \partial p_{m_2}} = +\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right]$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E')\frac{\partial \chi(\mathbf{p}; \mathbf{r}, E \to E')}{\partial p_{m_2}} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]\varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_{m_2}}\ ; \tag{55}$$

$$\text{for } j = 1, ..., J_t;\ m_2 = 1, ..., J_q : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_j \partial q_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial q_{m_2}}\ ; \tag{56}$$

and

$$\text{for } j = 1, ..., J_t;\ m_2 = 1, ..., J_d : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_j \partial d_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_{m_2}}\ . \tag{57}$$

IV.B. Computation of the Second-Order Sensitivities $\partial^2 R(\alpha,\varphi;\psi^{(1)})/\partial s_j \partial\alpha_{m_2}$, $j = 1,\ldots, J_s$; $m_2 = 1,\ldots, J_\alpha$

The second-order sensitivities $\partial^2 R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})/(\partial s_j)(\partial\alpha_{m_2}).(\partial s_j)(\partial\alpha_{m_2})$, $j = 1,\ldots,J_s$; $m_2 = 1,\ldots,J_\alpha$ are obtained by determining the G-differential of the first-order sensitivity defined in Eq. (30), which yields the following expression:

$$\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})}{\partial s_j}\right] = \left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})}{\partial s_j}\right]\right\}_{dir} + \left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})}{\partial s_j}\right]\right\}_{ind}, \quad j = 1,\ldots,J_s\ , \tag{58}$$

where for $j = 1,\ldots,J_s$:

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})}{\partial s_j}\right]\right\}_{dir} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\varphi(\mathbf{r},\boldsymbol{\Omega}',E') \times \sum_{m_2=1}^{J_s} \frac{\partial^2\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j \partial s_{m_2}}\delta s_{m_2}\ , \tag{59}$$

and where for $j = 1,\ldots,J_s$:

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})}{\partial s_j}\right]\right\}_{ind} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j}\varphi(\mathbf{r},\boldsymbol{\Omega}',E') + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \times \int_0^\infty dE'\,\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j}\delta\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\ . \tag{60}$$

The direct-effect term defined in Eq. (59) can be computed immediately. On the other hand, the indirect-effect term defined in Eq. (60) can be computed only after having obtained the solution $\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E)$ of the 1st-LFSS and the solution $\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$ of the 2nd-LFSS defined in Eqs. (39) and (40). To avoid the need for solving the 1st-LFSS and the 2nd-LFSS, the indirect-effect term defined in Eq. (60) will be expressed in terms of a 2nd-LASS, which will be constructed by following the same sequence of steps as previously outlined in Sec. IV.A. Thus, applying the definition provided in Eq. (42) to form the inner product of Eqs. (39) and (16) with a yet undefined function $\boldsymbol{\theta}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \triangleq \left[\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E),\ \theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\right]^\dagger$, where $\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \in L_2(V\times\boldsymbol{\Omega}\times E)$ and $\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \in L_2(V\times\boldsymbol{\Omega}\times E)$, yields a relation that is similar to Eq. (45), except that the components of $\boldsymbol{\psi}_j^{(2)}$ are replaced by the corresponding components of $\boldsymbol{\theta}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$, namely,

$$\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)A^{(1)}(\boldsymbol{\alpha})\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \times \int_0^\infty dE\,\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)L^{(1)}(\boldsymbol{\alpha})\delta\varphi(\mathbf{r},\boldsymbol{\Omega},E) = \int dV \int_{4\pi} d\boldsymbol{\Omega} \times \int_0^\infty dE\,\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\theta_{2,j}^{(2)} \times (\mathbf{r},\boldsymbol{\Omega},E)Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}) = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \times \left[A^{(1)}(\boldsymbol{\alpha})\right]^*\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \delta\varphi(\mathbf{r},\boldsymbol{\Omega}) \times \left[L^{(1)}(\boldsymbol{\alpha})\right]^*\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) + P^{(2)}\left[\delta\varphi,\,\delta\psi^{(1)};\,\theta_{1,j}^{(2)},\theta_{2,j}^{(2)}\right]\ . \tag{61}$$

The bilinear concomitant $P^{(2)}[\delta\varphi,\,\delta\psi^{(1)};\,\psi_{1,j}^{(2)},\psi_{2,j}^{(2)}]$ in Eq. (61) will vanish by imposing the boundary conditions $\theta_{1,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E) = 0, \mathbf{r}_s \in \partial V, \boldsymbol{\Omega}\cdot\mathbf{n} < 0$ and $\theta_{2,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E) = 0,\ \mathbf{r}_s \in \partial V, \boldsymbol{\Omega}\cdot\mathbf{n} > 0$. Noting that $\left[A^{(1)}(\boldsymbol{\alpha})\right]^* = L^{(1)}(\boldsymbol{\alpha}) = L(\boldsymbol{\alpha})$ and $\left[L^{(1)}(\boldsymbol{\alpha})\right]^* = A^{(1)}(\boldsymbol{\alpha})$ and identifying the rightmost side of Eq. (61) with the indirect-effect term defined in Eq. (60) yields the following 2nd-LASS for the components of the second-level adjoint function $\boldsymbol{\theta}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$:

$$L(\boldsymbol{\alpha}^0)\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \triangleq \boldsymbol{\Omega}\cdot\nabla\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) + \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E) \times \theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) - \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\Sigma_s^0(\mathbf{s}^0;\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega}) \times \theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) - \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E')\right] \times \chi^0(\mathbf{p}^0;\mathbf{r},E'\to E)\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) = \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \times \frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j}\varphi(\mathbf{r},\boldsymbol{\Omega}',E'),\ j=1,\ldots,J_s \tag{62}$$

and

$$A^{(1)}\left(\boldsymbol{\alpha}^{0}\right)\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \triangleq -\boldsymbol{\Omega}\cdot\nabla\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)+\Sigma_{t}^{0}\left(\mathbf{t}^{0};\mathbf{r},E\right)\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)-\int_{4\pi}d\boldsymbol{\Omega}'\int_{0}^{\infty}dE'\Sigma_{s}^{0}\left(\mathbf{s}^{0};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}'\right)$$
$$\times\,\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega}',E')-\left[\nu^{0}\Sigma_{f}^{0}\left(\mathbf{f}^{0};\mathbf{r},E\right)\right]\int_{4\pi}d\boldsymbol{\Omega}'\int_{0}^{\infty}dE'\chi^{0}\left(\mathbf{p}^{0};\mathbf{r},E\to E'\right)\,\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega}',E')\;=\int_{4\pi}d\boldsymbol{\Omega}'\int_{0}^{\infty}dE'\,\psi^{(1)}$$
$$\times\,(\mathbf{r},\boldsymbol{\Omega}',E')\,\frac{\partial\Sigma_{s}(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{j}},\quad j=1,\ldots,J_{s}\;, \tag{63}$$

subject to the following boundary condition:

$$\theta_{1,j}^{(2)}(\mathbf{r}_{s},\boldsymbol{\Omega},E)=0,\boldsymbol{\Omega}\cdot\mathbf{n}<0;\;\;\theta_{2,m_1}^{(2)}(\mathbf{r}_{s},\boldsymbol{\Omega},E)=0,\boldsymbol{\Omega}\cdot\mathbf{n}>0;\;\;\mathbf{r}_{s}\in\partial V,j=1,\ldots,J_{s}\;. \tag{64}$$

Using Eqs. (61) through (64) in Eq. (60) yields the following expression for the indirect-effect term:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)}\right)}{\partial s_{j}}\right]\right\}_{ind}=\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_{0}^{\infty}dE\;\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right)+\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_{0}^{\infty}dE\,\theta_{2,j}^{(2)}$$
$$\times\,(\mathbf{r},\boldsymbol{\Omega},E)Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}),\quad j=1,\ldots,J_{s}\;. \tag{65}$$

Replacing the expressions of $Q^{(2)}\left(\boldsymbol{\alpha}^{0},\psi^{(1)};\delta\boldsymbol{\alpha}\right)$ and $Q^{(1)}(\boldsymbol{\alpha};\varphi;\delta\boldsymbol{\alpha})$ from Eqs. (41) and (19), respectively, in Eq. (65); replacing the resulting expression together with the direct-effect term from Eq. (59) into Eq. (58); and subsequently identifying the quantities multiplying the parameter variations $\delta\alpha_{m_2}\;,m_2=1,\ldots,J_\alpha\;,$ in Eq. (58) yield the following expressions for the second-order partial sensitivities $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)/(\partial s_j)(\partial\alpha_{m_2})$, $j=1,\ldots,J_s$, $m_2=1,\ldots,J_\alpha$:

$$\text{For } j=1,\ldots,J_s;\quad m_2=1,\ldots,J_t:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j\partial t_{m_2}}=-\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_{0}^{\infty}dE\Big[\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$+\,\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\varphi(\mathbf{r},\boldsymbol{\Omega},E)\Big]\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_{m_2}}\;; \tag{66}$$

$$\text{For } j=1,\ldots,J_s;\quad m_2=1,\ldots,J_s:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j\partial s_{m_2}}=\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_{0}^{\infty}dE\;\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times\int_{4\pi}d\boldsymbol{\Omega}'\int_{0}^{\infty}dE'\,\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\frac{\partial^2\Sigma_s(\mathbf{s};\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j\partial s_{m_2}}+\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_{0}^{\infty}dE\,\theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times\int_{4\pi}d\boldsymbol{\Omega}'\int_{0}^{\infty}dE'\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}+\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_{0}^{\infty}dE\,\theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times\int_{4\pi}d\boldsymbol{\Omega}'\int_{0}^{\infty}dE'\;\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}\;; \tag{67}$$

$$\text{For } j=1,\ldots,J_s;\;\; m_2=1,\ldots,J_f: \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j \partial f_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r},\boldsymbol{\Omega}',E')\chi(\mathbf{p};\mathbf{r},E'\to E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_{m_2}}\;; \tag{68}$$

$$\text{For } j=1,\ldots,J_s;\;\; m_2=1,\ldots,J_p: \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j \partial p_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right] \int_{4\pi} d\boldsymbol{\Omega}'$$
$$\times \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_{m_2}} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]\varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_{m_2}}\;; \tag{69}$$

$$\text{For } j=1,\ldots,J_s;\;\; m_2=1,\ldots,J_q: \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j \partial q_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_{m_2}}\;; \tag{70}$$

and

$$\text{For } j=1,\ldots,J_s;\;\; m_2=1,\ldots,J_d: \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j \partial d_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial \Sigma_d(\mathbf{d};\mathbf{r},\boldsymbol{\Omega},E)}{\partial d_{m_2}}\;. \tag{71}$$

It is important to note that the forward and adjoint operators appearing on the left side of the 2nd-LASS defined by Eqs. (62), (63), and (64) for the second-level adjoint function $\boldsymbol{\theta}_j^{(2)}$ are the same operators as appearing on the left side of the 2nd-LASS defined by Eqs. (46) through (49) for the second-level adjoint function $\boldsymbol{\psi}_j^{(2)}$, the forward operator being the same as on the left side of the original transport Eq. (1), while the adjoint operator is the same as that appearing in the 1st-LASS, namely, Eq. (25). Furthermore, the forward and, respectively, adjoint functions are subject to the same forward and, respectively, adjoint (vacuum) boundary conditions. Only the source terms on the left sides of the respective forward, 1st-LASS, and 2nd-LASS differ from each other. Therefore, the same forward and adjoint software packages can be used for solving numerically the various equations underlying the 1st-LASS and the 2nd-LASS. Furthermore, the formal expression of the indirect-effect term defined in Eq. (50) involving the function $\boldsymbol{\psi}_j^{(2)}$ has the same formal expression as the indirect-effect term defined in Eq. (65) involving the function $\boldsymbol{\theta}_j^{(2)}$. Therefore, these indirect-effect terms can be evaluated numerically (quantitatively) using the same software package, while inputting the corresponding second-level adjoint functions $\boldsymbol{\psi}_j^{(2)}$ and $\boldsymbol{\theta}_j^{(2)}$. Consequently, the second-order sensitivities shown in Eqs. (68) through (71) have formally the same expressions as the second-order sensitivities shown in Eqs. (54) through (57), respectively, except that the second-level adjoint function $\boldsymbol{\theta}_j^{(2)}$ in Eqs. (68) through (71) plays the role of the second-level adjoint function $\boldsymbol{\psi}_j^{(2)}$ in Eqs. (54) through (57). Thus, the software package used for computing the sensitivities shown in Eqs. (54) through (57) can also be used for computing the sensitivities shown in Eqs. (68) through (71).

The expressions of the second-order sensitivities computed using Eq. (66) must be identical to those computed using Eq. (53).

That is, for $j = 1, ..., J_s; \quad k = 1, ..., J_t$:

$$\frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}; \boldsymbol{\theta}_j^{(2)}\right)}{\partial s_j \partial t_k} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE \left[\theta_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) + \theta_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\right] \frac{\partial \Sigma_t(\mathbf{t}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial t_k}$$

$$= \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_k \partial s_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E')$$

$$\times \frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_j} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$

$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_j} \quad . \tag{72}$$

The relation shown in Eq. (72) provides an independent path for the mutual verification of the solutions $\boldsymbol{\psi}_j^{(2)}$ and $\boldsymbol{\theta}_j^{(2)}$, $j = 1, ..., J_s$, of the respective 2nd-LASS.

IV.C. Computation of the Second-Order Sensitivities $\partial^2 R(\alpha, \varphi; \psi^{(1)})/\partial f_j \partial \alpha_{m_2}$, $j = 1,..., J_f$; $m_2 = 1,..., J_\alpha$

The second-order sensitivities $\partial^2 R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}\right) / \left(\partial f_j\right)\left(\partial \alpha_{m_2}\right)$, $j = 1, ..., J_f$; $m_2 = 1, ..., J_\alpha$ are obtained by determining the G-differential of the first-order sensitivity defined in Eq. (31), which yields the following expression:

$$\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}\right)}{\partial f_j}\right] = \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}\right)}{\partial f_j}\right]\right\}_{dir} + \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}\right)}{\partial f_j}\right]\right\}_{ind}, \quad j = 1, ..., J_f \ , \tag{73}$$

where for $j = 1, ..., J_f$:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}\right)}{\partial f_j}\right]\right\}_{dir} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\chi(\mathbf{p}; \mathbf{r}, E' \to E)$$

$$\times \sum_{m_2=1}^{J_f} \frac{\partial^2 \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_j \partial f_{m_2}} \delta f_{m_2} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}'$$

$$\times \int_0^\infty dE'\ \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \sum_{m_2=1}^{J_p} \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_{m_2}} \delta p_{m_2} \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_j} \ , \tag{74}$$

and where for $j = 1, ..., J_f$:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi; \psi^{(1)}\right)}{\partial f_j}\right]\right\}_{ind} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \left[\delta\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\chi(\mathbf{p}; \mathbf{r}, E' \to E)$$

$$\times \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_j} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ [\delta\varphi(\mathbf{r}, \boldsymbol{\Omega}', E')]\chi(\mathbf{p}; \mathbf{r}, E' \to E) \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_j} \ . \tag{75}$$

The direct-effect term defined in Eq. (74) can be computed immediately. On the other hand, the indirect-effect term defined in Eq. (75) can be computed only after having obtained the solution $\delta\varphi(\mathbf{r},\mathbf{\Omega},E)$ of the 1st-LFSS and the solution $\delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)$ of the 2nd-LFSS defined in Eqs. (39) and (40). To avoid the need for solving the 1st-LFSS and the 2nd-LFSS, the indirect-effect term defined in Eq. (75) will be expressed in terms of the solution of a 2nd-LASS, which will be constructed by following the same sequence of steps as previously outlined in Secs. IV.A and IV.B. Thus, applying the definition provided in Eq. (42) to form the inner product of Eqs. (39) and (17) with a yet undefined function $\mathbf{u}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq \left[u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E), u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\right]^\dagger$, where $u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \in \mathsf{L}_2(V\times\mathbf{\Omega}\times E)$ and $u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \in \mathsf{L}_2(V\times\mathbf{\Omega}\times E)$, yields a relation that is similar to those shown in Eqs. (45) and (61), except that the components of $\boldsymbol{\psi}_j^{(2)}$ or $\boldsymbol{\theta}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E)$, respectively, are replaced by the corresponding components of $\mathbf{u}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E)$, namely,

$$\begin{aligned}
&\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)A^{(1)}(\boldsymbol{\alpha})\delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)L^{(1)}(\boldsymbol{\alpha})\delta\varphi(\mathbf{r},\mathbf{\Omega},E) \\
&= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}) \\
&= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)\left[A^{(1)}(\boldsymbol{\alpha})\right]^* u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \\
&+ \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\ \delta\varphi(\mathbf{r},\mathbf{\Omega})\left[L^{(1)}(\boldsymbol{\alpha})\right]^* u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) + P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ u_{1,j}^{(2)}, u_{2,j}^{(2)}\right] . \qquad (76)
\end{aligned}$$

The bilinear concomitant $P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ u_{1,j}^{(2)}, u_{2,j}^{(2)}\right]$ in Eq. (76) will vanish by imposing the boundary conditions $u_{1,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0, \mathbf{r}_s\in\partial V, \mathbf{\Omega}\cdot\mathbf{n}<0$ and $u_{2,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0, \mathbf{r}_s\in\partial V, \mathbf{\Omega}\cdot\mathbf{n}>0$. Noting that $\left[A^{(1)}(\boldsymbol{\alpha})\right]^* = L^{(1)}(\boldsymbol{\alpha}) = L(\boldsymbol{\alpha})$ and $\left[L^{(1)}(\boldsymbol{\alpha})\right]^* = A^{(1)}(\boldsymbol{\alpha})$ and identifying the rightmost side of Eq. (76) with the indirect-effect term defined in Eq. (75) yields the following 2nd-LASS for the components of the second-level adjoint function $\mathbf{u}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E)$:

$$\begin{aligned}
&L\left(\boldsymbol{\alpha}^0\right)u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq \mathbf{\Omega}\bullet\nabla u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) + \Sigma_t^0\left(\mathbf{t}^0;\mathbf{r},E\right)u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \\
&- \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\,\Sigma_s^0\left(\mathbf{s}^0;\mathbf{r},E'\to E,\mathbf{\Omega}'\to\mathbf{\Omega}\right)u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) - \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\,\chi^0\left(\mathbf{p}^0;\mathbf{r},E'\to E\right) \\
&\times \left[\nu^0\Sigma_f^0\left(\mathbf{f}^0;\mathbf{r},E'\right)\right]u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) = \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\ \varphi^0(\mathbf{r},\mathbf{\Omega}',E')\chi^0\left(\mathbf{p}^0;\mathbf{r},E'\to E\right)\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_j}, \quad j=1,\ldots,J_f \quad (77)
\end{aligned}$$

and

$$\begin{aligned}
&A^{(1)}\left(\boldsymbol{\alpha}^0\right)u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq -\mathbf{\Omega}\bullet\nabla u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) + \Sigma_t^0\left(\mathbf{t}^0;\mathbf{r},E\right)u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) - \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\Sigma_s^0\left(\mathbf{s}^0;\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}'\right) \\
&\times u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}',E') - \left[\nu^0\Sigma_f^0\left(\mathbf{f}^0;\mathbf{r},E\right)\right]\int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\chi^0\left(\mathbf{p}^0;\mathbf{r},E\to E'\right)\ u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}',E') \\
&= \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_j}\int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\ \psi^{(1)}(\mathbf{r},\mathbf{\Omega}',E')\chi^0\left(\mathbf{p}^0;\mathbf{r},E\to E'\right), \quad j=1,\ldots,J_f\ , \qquad (78)
\end{aligned}$$

subject to the following boundary condition:

$$u_{2,m_1}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0\ ,\mathbf{\Omega}\cdot\mathbf{n}>0;\ \ u_{1,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0,\mathbf{\Omega}\cdot\mathbf{n}<0;\ \ \mathbf{r}_s\in\partial V;\ j=1,\ldots,J_f\ . \tag{79}$$

Using Eqs. (76) through (79) in Eq. (75) yields the following expression for the indirect-effect term:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial f_j}\right]\right\}_{ind}=\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\ u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right)$$
$$+\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\,u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}),\ \ j=1,\ldots,J_f\ . \tag{80}$$

Replacing the expressions of $Q^{(2)}\left(\boldsymbol{\alpha}^0,\psi^{(1)};\delta\boldsymbol{\alpha}\right)$ and $Q^{(1)}(\boldsymbol{\alpha};\varphi;\delta\boldsymbol{\alpha})$ from Eqs. (41) and (19), respectively, in Eq. (80); replacing the resulting expression together with the direct-effect term from Eq. (74) into (73); and subsequently identifying the quantities multiplying the parameter variations $\delta\alpha_{m_2}$, $m_2=1,\ldots,J_\alpha$, in Eq. (73) yields the following expressions for the second-order partial sensitivities $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)/\left(\partial f_j\right)\left(\partial\alpha_{m_2}\right)$, $j=1,\ldots,J_f$, $m_2=1,\ldots,J_\alpha$:

$$\text{For } j=1,\ldots,J_f;\quad m_2=1,\ldots,J_t:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j\partial t_{m_2}}=-\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE$$
$$\times\left[u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)+u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\varphi(\mathbf{r},\mathbf{\Omega},E)\right]\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\mathbf{\Omega},E)}{\partial t_{m_2}}\ . \tag{81}$$

$$\text{For } j=1,\ldots,J_f;\quad m_2=1,\ldots,J_s:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j\partial s_{m_2}}=\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\,u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\int_{4\pi}d\mathbf{\Omega}'$$
$$\times\int_0^\infty dE'\,\psi^{(1)}(\mathbf{r},\mathbf{\Omega}',E')\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}+\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\,u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$\times\int_{4\pi}d\mathbf{\Omega}'\int_0^\infty dE'\ \varphi(\mathbf{r},\mathbf{\Omega}',E')\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}\ ; \tag{82}$$

$$\text{For } j=1,\ldots,J_f;\ \ m_2=1,\ldots,J_f:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j\partial f_{m_2}}=$$
$$\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)\int_{4\pi}d\mathbf{\Omega}'\int_0^\infty dE'\ \varphi(\mathbf{r},\mathbf{\Omega}',E')\chi(\mathbf{p};\mathbf{r},E'\to E)\frac{\partial^2\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_j\partial f_{m_2}}$$
$$+\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\,u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}\int_{4\pi}d\mathbf{\Omega}'\int_0^\infty dE'\chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\mathbf{\Omega}',E')$$
$$+\int dV\int_{4\pi}d\mathbf{\Omega}\int_0^\infty dE\,u_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\int_{4\pi}d\mathbf{\Omega}'\int_0^\infty dE'\,\varphi(\mathbf{r},\mathbf{\Omega}',E')\frac{\chi(\mathbf{p};\mathbf{r},E'\to E)}{4\pi}\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_{m_2}}\ ; \tag{83}$$

$$\text{For } j=1,...,J_f;\ \ m_2=1,...,J_p : \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j \partial p_{m_2}} =$$
$$\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ \varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_{m_2}} \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_j}$$
$$+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_{m_2}}$$
$$+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\ \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_{m_2}}\ ; \tag{84}$$

$$\text{For } j=1,...,J_f;\ m_2=1,...,J_q : \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j \partial q_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_{m_2}}\ ; \tag{85}$$

and

$$\text{For } j=1,...,J_f;\ m_2=1,...,J_d : \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j \partial d_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial\Sigma_d(\mathbf{d};\mathbf{r},\boldsymbol{\Omega},E)}{\partial d_{m_2}}\ . \tag{86}$$

As discussed in Secs. IV.A and IV.B, it is important to note that the forward and adjoint operators appearing on the left side of the 2nd-LASS defined by Eqs. (77), (78), and (79) for the second-level adjoint function $\mathbf{u}_j^{(2)}$ are the same operators as appearing on the left side of the 2nd-LASS for the second-level adjoint function $\boldsymbol{\theta}_j^{(2)}$ and $\boldsymbol{\psi}_j^{(2)}$; all of these second-level adjoint functions are subject to the same boundary conditions. Only the source terms on the left sides of the respective 2nd-LASSs differ from each other. Therefore, the same forward and adjoint software packages can be used for solving numerically the various forward and adjoint equations underlying the 1st-LASS and the 2nd-LASS. Furthermore, the indirect-effect terms defined in Eqs. (50), (65), and (80) involving the second-level adjoint functions $\boldsymbol{\psi}_j^{(2)}$, $\boldsymbol{\theta}_j^{(2)}$, and $\mathbf{u}_j^{(2)}$ have the same formal expression. Therefore, these indirect-effect terms can be evaluated numerically (quantitatively) using the same software package, while inputting the corresponding second-level adjoint functions $\boldsymbol{\psi}_j^{(2)}$, $\boldsymbol{\theta}_j^{(2)}$, and $\mathbf{u}_j^{(2)}$, respectively. The expressions of the second-order sensitivities computed using Eq. (81) must be identical to those computed using Eq. (54).

$$\text{That is, for } j=1,...,J_f;\ \ k=1,...,J_t : \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j\,\partial t_k} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE$$
$$\times\left[u_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)+u_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\varphi(\mathbf{r},\boldsymbol{\Omega},E)\right]\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_k}$$
$$=\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\boldsymbol{\psi}_j^{(2)}\right)}{\partial t_k \partial f_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\psi_{1,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_j} \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\chi(\mathbf{p};\mathbf{r},E\to E')\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')$$
$$+\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\psi_{2,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\chi(\mathbf{p};\mathbf{r},E\to E')\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_j}\ . \tag{87}$$

Also, expressions of the second-order sensitivities computed using Eq. (82) must be identical to those computed using Eq. (68).

$$\text{That is, for } j=1,\ldots,J_f;\quad k=1,\ldots,J_s: \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_j \partial s_k} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}'$$

$$\times \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_k} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}'$$

$$\times \int_0^\infty dE'\, \varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_k} = \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_k \partial f_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega}$$

$$\times \int_0^\infty dE\, \theta_{1,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_j} \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')$$

$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{2,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r},\boldsymbol{\Omega}',E')\chi(\mathbf{p};\mathbf{r},E'\to E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_j}\ . \tag{88}$$

The relation shown in Eq. (87) provides an independent path for the mutual verification of the solutions $\mathbf{u}_j^{(2)}$ and $\boldsymbol{\psi}_j^{(2)}$ while Eq. (88) provides an independent path for the mutual verification of the solutions $\mathbf{u}_j^{(2)}$ and $\boldsymbol{\theta}_j^{(2)}$.

IV.D. Computation of the Second-Order Sensitivities $\partial^2 R(\alpha,\ \varphi;\ \psi^{(1)})/\partial p_j \partial \alpha_{m_2}$, $j = 1,\ldots, J_p$; $m_2 = 1,\ldots, J_\alpha$

The second-order sensitivities $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\left(\partial p_j\right)\left(\partial\alpha_{m_2}\right),\ j=1,\ldots,J_p;\ m_2=1,\ldots,J_\alpha$ are obtained by computing the G-differential of the first-order sensitivities defined in Eq. (32), which yields the following expression:

$$\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial p_j}\right] = \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial p_j}\right]\right\}_{dir} + \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial p_j}\right]\right\}_{ind},\quad j=1,\ldots,J_p\ , \tag{89}$$

$$\text{where for } j=1,\ldots,J_p: \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial p_j}\right]\right\}_{dir}$$

$$\triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \nu\Sigma_f(\mathbf{f};\mathbf{r},E')\varphi(\mathbf{r},\boldsymbol{\Omega}',E') \sum_{m_2=1}^{J_p} \frac{\partial^2\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j \partial p_{m_2}}\delta p_{m_2}$$

$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ \varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j} \sum_{m_2=1}^{J_f} \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_{m_2}}\delta f_{m_2}\ , \tag{90}$$

$$\text{and where for } j=1,\ldots,J_p: \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial p_j}\right]\right\}_{ind} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \left[\delta\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)\right] \int_{4\pi} d\boldsymbol{\Omega}'$$

$$\times \int_0^\infty dE'\ \varphi(\mathbf{r},\boldsymbol{\Omega}',E')\nu\Sigma_f(\mathbf{f};\mathbf{r},E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}'$$

$$\times \int_0^\infty dE'\ [\delta\varphi(\mathbf{r},\boldsymbol{\Omega}',E')]\nu\Sigma_f(\mathbf{f};\mathbf{r},E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j}\ . \tag{91}$$

The direct-effect term defined in Eq. (90) can be computed immediately. On the other hand, the indirect-effect term defined in Eq. (91) can be computed only after having obtained the solution $\delta\varphi(\mathbf{r},\mathbf{\Omega},E)$ of the 1st-LFSS and the solution $\delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)$ of the 2nd-LFSS defined in Eqs. (39) and (40). To avoid the need for solving the 1st-LFSS and the 2nd-LFSS, the indirect-effect term defined in Eq. (91) will be expressed in terms of the solution of a 2nd-LASS, which will be constructed by following the same sequence of steps as has been outlined in Secs. IV.A, IV.B, and IV.C. Thus, applying the definition provided in Eq. (42) to form the inner product of Eqs. (39) and (16) with a yet undefined function $\mathbf{w}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq \left[w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E), w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\right]^{\dagger}$, where $w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \in L_2(V\times\mathbf{\Omega}\times E)$ and $w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \in L_2(V\times\mathbf{\Omega}\times E)$, yields the relation

$$\begin{aligned}
&\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^{\infty} dE\, w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) A^{(1)}(\boldsymbol{\alpha})\delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^{\infty} dE\, w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) L^{(1)}(\boldsymbol{\alpha})\delta\varphi(\mathbf{r},\mathbf{\Omega},E)\\
&= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^{\infty} dE\, w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^{\infty} dE\, w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha})\\
&= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^{\infty} dE\, \delta\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)\left[A^{(1)}(\boldsymbol{\alpha})\right]^{*} w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\\
&\quad + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^{\infty} dE\, \delta\varphi(\mathbf{r},\mathbf{\Omega})\left[L^{(1)}(\boldsymbol{\alpha})\right]^{*} w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) + P^{(2)}\left[\delta\varphi,\, \delta\psi^{(1)};\, w_{1,j}^{(2)}, w_{2,j}^{(2)}\right] .
\end{aligned} \tag{92}$$

The bilinear concomitant $P^{(2)}\left[\delta\varphi,\, \delta\psi^{(1)};\, w_{1,j}^{(2)}, w_{2,j}^{(2)}\right]$ in Eq. (92) will vanish by imposing the boundary conditions $w_{1,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0, \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n}<0$ and $w_{2,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0, \mathbf{r}_s \in \partial V, \mathbf{\Omega}\cdot\mathbf{n}>0$. Noting that $\left[A^{(1)}(\boldsymbol{\alpha})\right]^{*} = L^{(1)}(\boldsymbol{\alpha}) = L(\boldsymbol{\alpha})$ and $\left[L^{(1)}(\boldsymbol{\alpha})\right]^{*} = A^{(1)}(\boldsymbol{\alpha})$ and identifying the rightmost side of Eq. (76) with the indirect-effect term defined in Eq. (91) yields the following 2nd-LASS for the components of the second-level adjoint function $\mathbf{w}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E)$:

$$\begin{aligned}
L(\boldsymbol{\alpha}^0) w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) &\triangleq \mathbf{\Omega}\bullet\nabla w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) + \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E) w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\\
&- \int_{4\pi} d\mathbf{\Omega}' \int_0^{\infty} dE'\, \Sigma_s^0(\mathbf{s}^0;\mathbf{r},E'\to E,\mathbf{\Omega}'\to\mathbf{\Omega}) w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\\
&- \int_{4\pi} d\mathbf{\Omega}' \int_0^{\infty} dE'\, \chi^0(\mathbf{p}^0;\mathbf{r},E'\to E)\left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E')\right] w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\\
&= \int_{4\pi} d\mathbf{\Omega}' \int_0^{\infty} dE'\, \varphi(\mathbf{r},\mathbf{\Omega}',E')\nu\Sigma_f(\mathbf{f};\mathbf{r},E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j}, \quad j=1,\ldots,J_p
\end{aligned} \tag{93}$$

and

$$\begin{aligned}
&A^{(1)}(\boldsymbol{\alpha}^0) w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \triangleq -\mathbf{\Omega}\bullet\nabla w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) + \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)\, w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) - \int_{4\pi} d\mathbf{\Omega}' \int_0^{\infty} dE' \Sigma_s^0(\mathbf{s}^0;\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')\\
&\quad \times w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}',E') - \left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E)\right] \int_{4\pi} d\mathbf{\Omega}' \int_0^{\infty} dE'\, \chi^0(\mathbf{p}^0;\mathbf{r},E\to E')\, w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}',E')\\
&\quad = \nu\Sigma_f(\mathbf{f};\mathbf{r},E) \int_{4\pi} d\mathbf{\Omega}' \int_0^{\infty} dE'\, \psi^{(1)}(\mathbf{r},\mathbf{\Omega}',E') \frac{\partial\chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_j}, \quad j=1,\ldots,J_p ,
\end{aligned} \tag{94}$$

subject to the following boundary condition:

$$w_{2,m_1}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0\ ,\mathbf{\Omega}\cdot\mathbf{n}>0;$$
$$w_{1,j}^{(2)}(\mathbf{r}_s,\mathbf{\Omega},E)=0,\mathbf{\Omega}\cdot\mathbf{n}<0;$$
$$\mathbf{r}_s\in\partial V,\ j=1,\ldots,J_p\ . \tag{95}$$

As in Secs. IV.A, IV.B, and IV.C, the operators appearing on the left side of the 2nd-LASS defined by Eqs. (93), (94), and (95) for the second-level adjoint function $\mathbf{w}_j^{(2)}(\mathbf{r},\mathbf{\Omega},E)$ are the same operators as appearing on the respective left sides of the 2nd-LASS for the second-level adjoint function $\boldsymbol{\theta}_j^{(2)}$, $\boldsymbol{\psi}_j^{(2)}$, and $\mathbf{u}_j^{(2)}$; all of these second-level adjoint functions are subject to the same boundary conditions. Only the source terms on the left sides of the respective 2nd-LASSs differ from each other. Therefore, the same forward and adjoint software packages can be used for solving numerically the various forward and adjoint equations underlying the 1st-LASS and the 2nd-LASS.

Using the 2nd-LASS defined by Eqs. (93), (94), and (95) together with Eq. (92) into Eq. (91) yields the following expression for the indirect-effect term:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)}{\partial p_j}\right]\right\}_{ind}=\int dV\int_{4\pi} d\mathbf{\Omega}\int_0^\infty dE\ w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right)+\int dV\int_{4\pi} d\mathbf{\Omega}$$
$$\times\int_0^\infty dE\,w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}),\quad j=1,\ldots,J_p\ . \tag{96}$$

The indirect-effect term defined in Eq. (96) has the same formal expression as the indirect-effect terms defined in Eqs. (50), (65), and (80) involving the second-level adjoint functions $\boldsymbol{\psi}_j^{(2)}$, $\boldsymbol{\theta}_j^{(2)}$, and $\mathbf{u}_j^{(2)}$, respectively. Therefore, these indirect-effect terms can all be evaluated numerically using the same software package, while inputting the corresponding second-level adjoint functions $\boldsymbol{\psi}_j^{(2)}$, $\boldsymbol{\theta}_j^{(2)}$, $\mathbf{u}_j^{(2)}$, and $\mathbf{w}_j^{(2)}$, respectively.

Replacing the expressions of $Q^{(2)}\left(\boldsymbol{\alpha}^0,\psi^{(1)};\delta\boldsymbol{\alpha}\right)$ and $Q^{(1)}(\boldsymbol{\alpha};\varphi;\delta\boldsymbol{\alpha})$ from Eqs. (41) and (19), respectively, in Eq. (96); replacing the resulting expression together with the direct-effect term from Eq. (90) into (89); and subsequently identifying the quantities multiplying the parameter variations $\delta\alpha_{m_2}$, $m_2=1,\ldots,J_\alpha$ in Eq. yield the following expressions for the second-order partial sensitivities $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)/(\partial p_j)(\partial\alpha_{m_2})$, $j=1,\ldots,J_p$, $m_2=1,\ldots,J_\alpha$:

$$\text{For } j=1,\ldots,J_p;\quad m_2=1,\ldots,J_t:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j\partial t_{m_2}}=-\int dV\int_{4\pi} d\mathbf{\Omega}\int_0^\infty dE$$
$$\times\left[w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\psi^{(1)}(\mathbf{r},\mathbf{\Omega},E)+w_{2,j}^{(2)}\ (\mathbf{r},\mathbf{\Omega},E)\varphi(\mathbf{r},\mathbf{\Omega},E)\right]\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\mathbf{\Omega},E)}{\partial t_{m_2}}\quad; \tag{97}$$

$$\text{For } j=1,\ldots,J_p;\quad m_2=1,\ldots,J_s:\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j\partial s_{m_2}}=\int dV\int_{4\pi} d\mathbf{\Omega}\int_0^\infty dE\,w_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)$$
$$\times\int_{4\pi} d\mathbf{\Omega}'\int_0^\infty dE'\,\psi^{(1)}(\mathbf{r},\mathbf{\Omega}',E')\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}$$
$$+\int dV\int_{4\pi} d\mathbf{\Omega}\int_0^\infty dE\,w_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E)\int_{4\pi} d\mathbf{\Omega}'\int_0^\infty dE'\,\varphi(\mathbf{r},\mathbf{\Omega}',E')\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}\quad; \tag{98}$$

$$\text{For } j=1,...,J_p;\ \ m_2=1,...,J_f:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j \partial f_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}'$$
$$\times \int_0^\infty dE'\ \varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j} \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]}{\partial f_{m_2}} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\varphi(\mathbf{r},\boldsymbol{\Omega}',E')\chi(\mathbf{p};\mathbf{r},E'\to E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}\ ; \tag{99}$$

$$\text{For } j=1,...,J_p;\ \ m_2=1,...,J_p:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j \partial p_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}'$$
$$\int_0^\infty dE'\,\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial^2 \chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j \partial p_{m_2}}$$
$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_{m_2}}$$
$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]\varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_{m_2}}\ ; \tag{100}$$

$$\text{For } j=1,...,J_p;\ \ m_2=1,...,J_q:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j \partial q_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_{m_2}}\ ; \tag{101}$$

and

$$\text{For } j=1,...,J_p;\ \ m_2=1,...,J_d:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j \partial d_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial \Sigma_d(\mathbf{d};\mathbf{r},\boldsymbol{\Omega},E)}{\partial d_{m_2}}\ . \tag{102}$$

The expressions of the second-order sensitivities computed using Eq. (97) must be identical to those computed using Eq. (55).

$$\text{That is, For } j=1,...,J_p;\ \ k=1,...,J_t:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_j \partial t_k} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\left[w_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E)\right.$$
$$\left.+ w_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\varphi(\mathbf{r},\boldsymbol{\Omega},E)\right] \frac{\partial \Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_k} = \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\boldsymbol{\psi}_j^{(2)}\right)}{\partial t_k \partial p_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\psi_{1,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_j} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\,\psi_{2,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right]\varphi(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_j}\ . \tag{103}$$

The relation expressed by Eq. (103) provides an independent mutual verification of the second-level adjoint functions $\mathbf{w}_j^{(2)}$ and $\boldsymbol{\psi}_j^{(2)}$. Furthermore, the expressions of the second-order sensitivities computed using Eq. (98) must be identical to those computed using Eq. (69).

$$\text{That is, For } j = 1, \ldots, J_p; \quad k = 1, \ldots, J_s : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\, \psi^{(1)}; \mathbf{w}_j^{(2)}\right)}{\partial p_j \partial s_k}$$

$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_k}$$

$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_k}$$

$$= \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\, \psi^{(1)}; \boldsymbol{\theta}_j^{(2)}\right)}{\partial s_k \partial p_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{1,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E')$$

$$\times \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E \to E')}{\partial p_j} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{2,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right] \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_j} . \quad (104)$$

The relation expressed by Eq. (104) provides an independent mutual verification of the second-level adjoint functions $\mathbf{w}_j^{(2)}$ and $\boldsymbol{\theta}_j^{(2)}$. Finally, the expressions of the second-order sensitivities computed using Eq. (99) must be identical to those computed using Eq. (84).

$$\text{That is, For } j = 1, \ldots, J_p;\; k = 1, \ldots, J_f : \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\, \psi^{(1)}; \mathbf{w}_j^{(2)}\right)}{\partial p_j \partial f_k} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\; \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')$$

$$\times \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_j} \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_k} + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right]}{\partial f_k}$$

$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \chi(\mathbf{p}; \mathbf{r}, E \to E') \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E') + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, w_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$

$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \chi(\mathbf{p}; \mathbf{r}, E' \to E) \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right]}{\partial f_k} = \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\, \psi^{(1)}; \mathbf{u}_j^{(2)}\right)}{\partial f_k \partial p_j}$$

$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\; \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\; \varphi(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_j} \frac{\partial \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_k}$$

$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{1,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E \to E')}{\partial p_j}$$

$$+ \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{2,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right] \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\, \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_j} . \quad (105)$$

The relation shown in Eq. (105) provides an independent path for the mutual verification of the solutions $\mathbf{w}_j^{(2)}$ and $\mathbf{u}_j^{(2)}$.

IV.E. Computation of the Second-Order Sensitivities $\partial^2 R(\alpha, \varphi; \psi^{(1)})/\partial q_j \partial \alpha_{m_2}$, $j = 1,\ldots,J_q$; $m_2 = 1,\ldots,J_\alpha$

The second-order sensitivities $\partial^2 R(\boldsymbol{\alpha}, \varphi; \psi^{(1)})/ \times (\partial q_j)(\partial \alpha_{m_2})$, $j = 1,\ldots,J_q$; $m_2 = 1,\ldots,J_\alpha$ are obtained by computing the G-differential of the first-order sensitivities defined in Eq. (33), which yields the following expression:

$$\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)})}{\partial q_j}\right] = \left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)})}{\partial q_j}\right]\right\}_{dir} + \left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)})}{\partial q_j}\right]\right\}_{ind}, \quad j = 1,\ldots,J_q\ , \tag{106}$$

where for $j = 1,\ldots,J_q$:

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)})}{\partial q_j}\right]\right\}_{dir} \triangleq \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E) \times \sum_{m_2=1}^{J_q} \frac{\partial^2 Q(\mathbf{q}; \mathbf{r}, \mathbf{\Omega}, E)}{\partial q_j \partial q_{m_2}} \delta q_{m_2} \tag{107}$$

and where for $j = 1,\ldots,J_q$,

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)})}{\partial q_j}\right]\right\}_{ind} \triangleq \int dV \int_{4\pi} d\mathbf{\Omega} \times \int_0^\infty dE\ \ \delta\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E) \frac{\partial Q(\mathbf{q}; \mathbf{r}, \mathbf{\Omega}, E)}{\partial q_j}\ . \tag{108}$$

The direct-effect term defined in Eq. (107) can be computed immediately. On the other hand, the indirect-effect term defined in Eq. (108) can be computed only after having obtained the solution $\delta\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E)$ of the 2nd-LFSS defined in Eqs. (39) and (40). To avoid the need for solving the 1st-LFSS and the 2nd-LFSS, the indirect-effect term defined in Eq. (108) will be expressed in terms of the solution of a 2nd-LASS, which will be constructed by following the same sequence of steps as has been outlined in Secs. IV.A through IV.D. In contradistinction to the situations encountered in Secs. IV.A through IV.D, however, the indirect-effect term defined in Eq. (108) does *not* depend on the solution $\delta\varphi(\mathbf{r}, \mathbf{\Omega}, E)$ of the 1st-LFSS. Consequently, the 2nd-LASS that needed to be constructed for the alternative computation of the indirect-effect term defined in Eq. (108) will turn out to consist of a single (rather than two) operator equation to be satisfied by a second-level adjoint function that will turn out to have just a single nonzero component. Proceeding formally and applying the definition provided in Eq. (42) to form the inner product of Eqs. (39) and (16) with a yet undefined function $\mathbf{g}_j^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) \triangleq \left[g_{1,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E), g_{2,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E)\right]^\dagger$, where $g_{1,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) \in L_2(V \times \mathbf{\Omega} \times E)$ and $g_{2,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) \in L_2(V \times \mathbf{\Omega} \times E)$ yield the relation:

$$\begin{aligned}
&\int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) A^{(1)}(\boldsymbol{\alpha}) \delta\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, g_{2,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) L^{(1)}(\boldsymbol{\alpha}) \delta\varphi(\mathbf{r}, \mathbf{\Omega}, E) \\
&= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) Q^{(2)}\left(\boldsymbol{\alpha}, \psi^{(1)}; \delta\boldsymbol{\alpha}\right) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, g_{2,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) Q^{(1)}(\boldsymbol{\alpha}, \varphi; \delta\boldsymbol{\alpha}) \\
&= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \delta\psi^{(1)}(\mathbf{r}, \mathbf{\Omega}, E) \left[A^{(1)}(\boldsymbol{\alpha})\right]^* g_{1,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) + \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\ \delta\varphi(\mathbf{r}, \mathbf{\Omega}) \left[L^{(1)}(\boldsymbol{\alpha})\right]^* g_{2,j}^{(2)}(\mathbf{r}, \mathbf{\Omega}, E) \\
&\quad + P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ g_{1,j}^{(2)}, g_{2,j}^{(2)}\right]\ .
\end{aligned} \tag{109}$$

The bilinear concomitant $P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ g_{1,j}^{(2)}, g_{2,j}^{(2)}\right]$ in Eq. (109) will vanish by imposing the boundary conditions $g_{1,j}^{(2)}(\mathbf{r}_s, \mathbf{\Omega}, E) = 0, \mathbf{r}_s \in \partial V, \mathbf{\Omega} \cdot \mathbf{n} < 0$ and $g_{2,j}^{(2)}(\mathbf{r}_s, \mathbf{\Omega}, E) = 0, \mathbf{r}_s \in \partial V, \mathbf{\Omega} \cdot \mathbf{n} > 0$. Noting that $\left[A^{(1)}(\boldsymbol{\alpha})\right]^* = L^{(1)}(\boldsymbol{\alpha}) = L(\boldsymbol{\alpha})$ and $\left[L^{(1)}(\boldsymbol{\alpha})\right]^* = A^{(1)}(\boldsymbol{\alpha})$ and identifying the rightmost side of Eq. (109) with the indirect-effect term defined in Eq. (108) yield the following 2nd-LASS for the components of the second-level adjoint function $\mathbf{g}_j^{(2)}(\mathbf{r}, \mathbf{\Omega}, E)$:

$$L(\boldsymbol{\alpha}^0)g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \triangleq \boldsymbol{\Omega}\bullet\nabla g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) + \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) - \int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\,\Sigma_s^0 \times(\mathbf{s}^0;\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})\,g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) - \int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\,\chi^0(\mathbf{p}^0;\mathbf{r},E'\to E)\left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E')\right] \times\ g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) = \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_j},\ j=1,\ldots,J_q \quad (110)$$

and

$$A^{(1)}(\boldsymbol{\alpha}^0)g_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \triangleq -\boldsymbol{\Omega}\bullet\nabla g_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) + \Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)\,g_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) - \int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\Sigma_s^0 \times(\mathbf{s}^0;\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')\,g_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega}',E') - \left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E)\right]\int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\,\chi^0(\mathbf{p}^0;\mathbf{r},E\to E') \times g_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega}',E') = 0,\ j=1,\ldots,J_q\ , \quad (111)$$

subject to the following boundary condition:

$$g_{2,m_1}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0\ ,\boldsymbol{\Omega}\cdot\mathbf{n}>0;\quad g_{1,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0,\boldsymbol{\Omega}\cdot\mathbf{n}<0;\quad \mathbf{r}_s\in\partial V;\ j=1,\ldots,J_q\ . \quad (112)$$

It is evident that the unique solution of the homogeneous linear Eq. (111) subject to the linear homogeneous boundary condition Eq. (112) is

$$g_{2,m_1}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\equiv 0\ ,j=1,\ldots,J_q\ . \quad (113)$$

The nonzero component $g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$ of the second-level adjoint function $\mathbf{g}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\triangleq\left[g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E),0\right]^\dagger$ is computed using the forward transport solver with the source shown on the right side of Eq. (110). Using the 2nd-LASS defined by Eqs. (111) and (112) together with Eq. (109) into Eq. (108) yields the following expression for the respective indirect-effect term:

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\psi^{(1)})}{\partial p_j}\right]\right\}_{ind} = \int dV\int_{4\pi} d\boldsymbol{\Omega}\int_0^\infty dE\ g_{1,j}^{(2)} \times(\mathbf{r},\boldsymbol{\Omega},E)Q^{(2)}\left(\boldsymbol{\alpha},\psi^{(1)};\delta\boldsymbol{\alpha}\right),\quad j=1,\ldots,J_q\ . \quad (114)$$

Replacing the expression of $Q^{(2)}(\boldsymbol{\alpha}^0,\psi^{(1)};\delta\boldsymbol{\alpha})$ from Eq. (41) into Eq. (114); replacing the resulting expression together with the direct-effect term from Eq. (107) into (106); and subsequently identifying the quantities multiplying the parameter variations $\delta\alpha_{m_2},\ m_2=1,\ldots,J_\alpha$, in Eq. (106) yield the following expressions for the second-order partial sensitivities $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\mathbf{g}_j^{(2)}\right)/(\partial q_j)(\partial\alpha_{m_2}),\ j=1,\ldots,J_q,\ m_2=1,\ldots,J_\alpha$:

For $j=1,\ldots,J_q;\quad m_2=1,\ldots,J_t$:

$$\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j\partial t_{m_2}} = -\int dV\int_{4\pi} d\boldsymbol{\Omega}\int_0^\infty dE\,g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \times\frac{\partial\Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_{m_2}}\ ; \quad (115)$$

For $j=1,\ldots,J_q;\quad m_2=1,\ldots,J_s$:

$$\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j\partial s_{m_2}} = \int dV\int_{4\pi} d\boldsymbol{\Omega}\int_0^\infty dE\,g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \times\int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\,\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \times\frac{\partial\Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}\ ; \quad (116)$$

For $j=1,\ldots,J_q;\ m_2=1,\ldots,J_f$:

$$\frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j\partial f_{m_2}} = \int dV\int_{4\pi} d\boldsymbol{\Omega}\int_0^\infty dE\,g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}} \times\int_{4\pi} d\boldsymbol{\Omega}'\int_0^\infty dE'\chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')\ ; \quad (117)$$

$$\text{For } j=1,...,J_q;\ \ m_2=1,...,J_p:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial p_{m_2}}$$

$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right] \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E\to E')}{\partial p_{m_2}}\ ; \tag{118}$$

$$\text{For } j=1,...,J_q;\ m_2=1,...,J_q:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial q_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial^2 Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_j \partial q_{m_2}}\ ; \tag{119}$$

and

$$\text{For } j=1,...,J_q;\ \ m_2=1,...,J_d:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial d_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial \Sigma_d(\mathbf{d};\mathbf{r},\boldsymbol{\Omega},E)}{\partial d_{m_2}}\ . \tag{120}$$

The expressions of the second-order sensitivities computed using Eq. (115) must be identical to those computed using Eq. (56).

$$\text{That is, for } j=1,...,J_q;\ \ k=1,...,J_t:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial t_k} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\psi^{(1)}$$

$$\times(\mathbf{r},\boldsymbol{\Omega},E)\frac{\partial \Sigma_t(\mathbf{t};\mathbf{r},\boldsymbol{\Omega},E)}{\partial t_k} = \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\boldsymbol{\psi}_j^{(2)}\right)}{\partial t_k \partial q_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{2,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_j}\ . \tag{121}$$

The relation expressed by Eq. (121) provides an independent mutual verification of the second-level adjoint functions $\mathbf{g}_j^{(2)}$ and $\boldsymbol{\psi}_j^{(2)}$. The expressions of the second-order sensitivities computed using Eq. (116) must be identical to those computed using Eq. (70).

$$\text{That is, for } j=1,...,J_q;\ \ k=1,...,J_s:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial s_k} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E')$$

$$\times\frac{\partial \Sigma_s(\mathbf{s};\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_k} = \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_k \partial q_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \theta_{2,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_j}\ . \tag{122}$$

The relation expressed by Eq. (122) provides an independent mutual verification of the second-level adjoint functions $\mathbf{g}_j^{(2)}$ and $\boldsymbol{\theta}_j^{(2)}$. The expressions of the second-order sensitivities computed using Eq. (117) must be identical to those computed using Eq. (85).

$$\text{That is, for } j=1,...,J_q;\ \ k=1,...,J_f:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial f_k} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}$$

$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE' \chi(\mathbf{p};\mathbf{r},E\to E')\psi^{(1)}(\mathbf{r},\boldsymbol{\Omega}',E') = \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_k \partial q_j}$$

$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, u_{2,k}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E) \frac{\partial Q(\mathbf{q};\mathbf{r},\boldsymbol{\Omega},E)}{\partial q_j}\ . \tag{123}$$

The relation shown in Eq. (123) provides an independent path for the mutual verification of the solutions $\mathbf{g}_j^{(2)}$ and $\mathbf{u}_j^{(2)}$. The expressions of the second-order sensitivities computed using Eq. (118) must be identical to those computed using Eq. (101).

That is, for $j = 1, \ldots, J_q;\ k = 1, \ldots, J_p$: $\dfrac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{g}_j^{(2)}\right)}{\partial q_j \partial p_k}$

$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E)\right] \times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}', E') \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E \to E')}{\partial p_{m_2}}$$
$$= \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{w}_j^{(2)}\right)}{\partial p_k \partial q_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \times \int_0^\infty dE\, w_{2,k}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \frac{\partial Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial q_j} \ . \tag{124}$$

The relation shown in Eq. (124) provides an independent path for the mutual verification of the solutions $\mathbf{g}_j^{(2)}$ and $\mathbf{w}_j^{(2)}$.

IV.F. Computation of the Second-Order Sensitivities $\partial^2 R(\alpha, \varphi;\ \psi^{(1)})/\partial d_j \partial \alpha_{m_2},\ j = 1, \ldots, J_d;\ m_2 = 1, \ldots, J_\alpha$

The second-order sensitivities $\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}\right)/\left(\partial d_j\right)\left(\partial \alpha_{m_2}\right),\ j = 1, \ldots, J_d;\ m_2 = 1, \ldots, J_\alpha$, are obtained by computing the G-differential of Eq. (34), which yields the following expression:

$$\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}\right)}{\partial d_j}\right] = \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}\right)}{\partial d_j}\right]\right\}_{dir} + \left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}\right)}{\partial d_j}\right]\right\}_{ind},\quad j = 1, \ldots, J_d\ , \tag{125}$$

where for $j = 1, \ldots, J_d$:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}\right)}{\partial d_j}\right]\right\}_{dir} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \varphi(\mathbf{r}, \boldsymbol{\Omega}, E) \times \sum_{m_2=1}^{J_q} \frac{\partial^2 \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_j \partial d_{m_2}} \delta d_{m_2}\ , \tag{126}$$

and where for $j = 1, \ldots, J_d$:

$$\left\{\delta\left[\frac{\partial R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}\right)}{\partial d_j}\right]\right\}_{ind} \triangleq \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \delta\varphi \times (\mathbf{r}, \boldsymbol{\Omega}, E)\ \frac{\partial \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_j}\ . \tag{127}$$

The direct-effect term defined in Eq. (126) can be computed immediately. On the other hand, the indirect-effect term defined in Eq. (127) can be computed only after having obtained the solution $\delta\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)$ of the 1st-LFSS defined in Eqs. (16) and (17). To avoid the need for solving the 1st-LFSS, the indirect-effect term defined in Eq. (127) will be expressed in terms of the solution of a 2nd-LASS, which will be constructed by following the same sequence of steps as has been outlined in Secs. IV.A through IV.E. As indicated in Eq. (127), the indirect-effect term defined does *not* depend on the solution $\delta\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E)$ of the 2nd-LFSS. Consequently, the 2nd-LASS that needed to be constructed for the alternative computation of the indirect-effect term defined in Eq. (127) will (also) turn out to consist of a single operator equation, for a second-level adjoint function that will turn out to have just a single nonzero component. Proceeding formally and applying the definition provided in Eq. (42) to form the inner product of Eqs. (39) and (16) with a yet undefined function $\mathbf{h}_j^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \triangleq \left[h_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E),\quad h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\right]^\dagger$, where $h_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \in L_2(V \times \boldsymbol{\Omega} \times E)$ and $h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \in L_2(V \times \boldsymbol{\Omega} \times E)$ yield the relation:

$$\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) A^{(1)}(\boldsymbol{\alpha}) \delta\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) L^{(1)}(\boldsymbol{\alpha}) \delta\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)$$
$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) Q^{(2)}\left(\boldsymbol{\alpha}, \psi^{(1)}; \delta\boldsymbol{\alpha}\right) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) Q^{(1)}(\boldsymbol{\alpha}, \varphi; \delta\boldsymbol{\alpha})$$
$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \delta\psi^{(1)}(\mathbf{r}, \boldsymbol{\Omega}, E) \left[A^{(1)}(\boldsymbol{\alpha})\right]^* h_{1,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) + \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \delta\varphi(\mathbf{r}, \boldsymbol{\Omega}) \left[L^{(1)}(\boldsymbol{\alpha})\right]^* h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$
$$+ P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ h_{1,j}^{(2)}, h_{2,j}^{(2)}\right]\ . \tag{128}$$

The bilinear concomitant $P^{(2)}\left[\delta\varphi,\ \delta\psi^{(1)};\ h_{1,j}^{(2)}, h_{2,j}^{(2)}\right]$ in Eq. (128) will vanish by imposing the boundary conditions $h_{1,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0, \mathbf{r}_s\in\partial V, \boldsymbol{\Omega}\cdot\mathbf{n}<0$ and $h_{2,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0, \mathbf{r}_s\in\partial V, \boldsymbol{\Omega}\cdot\mathbf{n}>0$. Noting that $\left[A^{(1)}(\boldsymbol{\alpha})\right]^*=L^{(1)}(\boldsymbol{\alpha})=L(\boldsymbol{\alpha})$ and $\left[L^{(1)}(\boldsymbol{\alpha})\right]^*=A^{(1)}(\boldsymbol{\alpha})$ and identifying the rightmost side of Eq. (128) with the indirect-effect term defined in Eq. (127) yield the following 2nd-LASS for the components of the second-level adjoint function $\mathbf{h}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$:

$$L(\boldsymbol{\alpha}^0)h_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\triangleq\boldsymbol{\Omega}\bullet\nabla h_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)+\Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)h_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$-\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'\,\Sigma_s^0(\mathbf{s}^0;\mathbf{r},E'\to E,\boldsymbol{\Omega}'\to\boldsymbol{\Omega})h_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$-\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'\chi^0(\mathbf{p}^0;\mathbf{r},E'\to E)\left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E')\right]h_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)=0,$$
$$j=1,\dots,J_q \tag{129}$$

and

$$A^{(1)}(\boldsymbol{\alpha}^0)h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\triangleq-\boldsymbol{\Omega}\bullet\nabla h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)+\Sigma_t^0(\mathbf{t}^0;\mathbf{r},E)\,h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$$
$$-\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'\Sigma_s^0(\mathbf{s}^0;\mathbf{r},E\to E',\boldsymbol{\Omega}\to\boldsymbol{\Omega}')\,h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega}',E')$$
$$-\left[\nu^0\Sigma_f^0(\mathbf{f}^0;\mathbf{r},E)\right]\int_{4\pi}d\boldsymbol{\Omega}'\int_0^\infty dE'\,\chi^0(\mathbf{p}^0;\mathbf{r},E\to E')\,h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega}',E')$$
$$=\frac{\partial\Sigma_d(\mathbf{d};\mathbf{r},\boldsymbol{\Omega},E)}{\partial d_j},\ j=1,\dots,J_q\ , \tag{130}$$

subject to the following boundary condition:

$$h_{1,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0,\boldsymbol{\Omega}\cdot\mathbf{n}<0;$$
$$h_{2,j}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega},E)=0\ ,\boldsymbol{\Omega}\cdot\mathbf{n}>0;$$
$$\mathbf{r}_s\in\partial V;\ \ j=1,\dots,J_q\ . \tag{131}$$

It is evident that the unique solution of the homogeneous linear Eq. (129) subject to the linear homogeneous boundary condition shown in Eq. (131) is

$$h_{1,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\equiv0\ ,\ j=1,\dots,J_q\ . \tag{132}$$

The nonzero component $h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)$ of the second-level adjoint function $\mathbf{h}_j^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\triangleq\left[0,\ h_{2,j}^{(2)}(\mathbf{r},\boldsymbol{\Omega},E)\right]^\dagger$ is computed using the adjoint transport solver with the source shown on the right side of Eq. (130). Using the 2nd-LASS defined by Eqs. (130) and (131) together with Eq. (128) into Eq. (127) yields the following expression for the respective indirect-effect term:

$$\left\{\delta\left[\frac{\partial R(\boldsymbol{\alpha},\varphi;\ \psi^{(1)})}{\partial d_j}\right]\right\}_{ind}=\int dV\int_{4\pi}d\boldsymbol{\Omega}\int_0^\infty dE\ h_{2,j}^{(2)}$$
$$\times(\mathbf{r},\boldsymbol{\Omega},E)Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha}),$$
$$j=1,\dots,J_d\ . \tag{133}$$

Replacing the expression of $Q^{(1)}(\boldsymbol{\alpha},\varphi;\delta\boldsymbol{\alpha})$ from Eq. (19) into Eq. (133); replacing the resulting expression together with the direct-effect term from Eq. (126) into Eq. (125); and subsequently identifying the quantities multiplying the parameter variations $\delta\alpha_{m_2}, m_2=1,\dots,J_\alpha$, in Eq. (125) yields the following expressions for the second-order partial sensitivities $\partial^2R\left(\boldsymbol{\alpha},\varphi;\ \ \psi^{(1)};\mathbf{h}_j^{(2)}\right)/(\partial d_j)\,(\partial\alpha_{m_2})$, $j=1,\dots,J_d\ ,\ m_2=1,\dots,J_\alpha$:

$$\text{For } j = 1, ..., J_d,\ m_2 = 1, ..., J_t: \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial t_{m_2}} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial \Sigma_t(\mathbf{t}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial t_{m_2}}\ ; \tag{134}$$

$$\text{For } j = 1, ..., J_d;\quad m_2 = 1, ..., J_s: \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial s_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\ \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\frac{\partial \Sigma_s(\mathbf{s}; \mathbf{r}, E \to E', \boldsymbol{\Omega} \to \boldsymbol{\Omega}')}{\partial s_{m_2}}\ ; \tag{135}$$

$$\text{For } j = 1, ..., J_d;\ \ m_2 = 1, ..., J_f: \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial f_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\chi(\mathbf{p}; \mathbf{r}, E' \to E)\frac{\partial\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]}{\partial f_{m_2}}\ ; \tag{136}$$

$$\text{For } j = 1, ..., J_d;\ \ m_2 = 1, ..., J_p: \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial p_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)$$
$$\times \int_{4\pi} d\boldsymbol{\Omega}' \int_0^\infty dE'\left[\nu\Sigma_f(\mathbf{f}; \mathbf{r}, E')\right]\varphi(\mathbf{r}, \boldsymbol{\Omega}', E')\ \frac{\partial \chi(\mathbf{p}; \mathbf{r}, E' \to E)}{\partial p_{m_2}}\ ; \tag{137}$$

$$\text{For } j = 1, ..., J_d;\ m_2 = 1, ..., J_q: \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial q_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial q_{m_2}}\ ; \tag{138}$$

and

$$\text{For } j = 1, ..., J_d;\ m_2 = 1, ..., J_d: \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial d_{m_2}} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\ \varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial^2 \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_j \partial d_{m_2}}\ . \tag{139}$$

The expressions of the second-order sensitivities computed using Eq. (134) must be identical to those computed using Eq. (57).

$$\text{That is, for } j = 1, ..., J_d;\quad k = 1, ..., J_t, \quad \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial t_k} = -\int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}$$
$$\times (\mathbf{r}, \boldsymbol{\Omega}, E)\varphi(\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial \Sigma_t(\mathbf{t}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial t_k} = \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \boldsymbol{\psi}_j^{(2)}\right)}{\partial t_k \partial d_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, \psi_{1,k}^{(2)}$$
$$\times (\mathbf{r}, \boldsymbol{\Omega}, E)\frac{\partial \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_j}\ . \tag{140}$$

The relation expressed by Eq. (140) provides an independent mutual verification of the second-level adjoint functions $\mathbf{h}_j^{(2)}$ and $\boldsymbol{\psi}_j^{(2)}$. The expressions of the second-order sensitivities computed using Eq. (135) must be identical to those computed using Eq. (71).

$$\text{That is, for } j=1,...,J_d;\quad k=1,...,J_s:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{h}_j^{(2)}\right)}{\partial d_j \partial s_k}$$

$$= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r},\mathbf{\Omega}',E')$$

$$\times \frac{\partial \Sigma_s(\mathbf{s};\mathbf{r},E\to E',\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_k}$$

$$= \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\boldsymbol{\theta}_j^{(2)}\right)}{\partial s_k \partial d_s} = \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, \theta_{1,k}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \frac{\partial \Sigma_d(\mathbf{d};\mathbf{r},\mathbf{\Omega},E)}{\partial d_s}\ . \tag{141}$$

The relation expressed by Eq. (141) provides an independent mutual verification of the second-level adjoint functions $\mathbf{h}_j^{(2)}$ and $\boldsymbol{\theta}_j^{(2)}$.The expressions of the second-order sensitivities computed using Eq. (136) must be identical to those computed using Eq. (86).

$$\text{That is, for } j=1,...,J_d;\ k=1,...,J_f:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{h}_j^{(2)}\right)}{\partial d_j \partial f_k}$$

$$= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE'\, \varphi(\mathbf{r},\mathbf{\Omega}',E')$$

$$\times \chi(\mathbf{p};\mathbf{r},E'\to E) \frac{\partial\left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E)\right]}{\partial f_{m_2}}$$

$$= \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{u}_j^{(2)}\right)}{\partial f_k \partial d_j} = \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, u_{1,k}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \frac{\partial \Sigma_d(\mathbf{d};\mathbf{r},\mathbf{\Omega},E)}{\partial d_j}\ . \tag{142}$$

The relation shown in Eq. (142) provides an independent path for the mutual verification of the solutions $\mathbf{h}_j^{(2)}$ and $\mathbf{u}_j^{(2)}$. The expressions of the second-order sensitivities computed using Eq. (137) must be identical to those computed using Eq. (102).

$$\text{That is, for } j=1,...,J_d;\ k=1,...,J_p:\ \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{h}_j^{(2)}\right)}{\partial d_j \partial p_k}$$

$$= \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \int_{4\pi} d\mathbf{\Omega}' \int_0^\infty dE' \left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E')\right] \varphi(\mathbf{r},\mathbf{\Omega}',E') \frac{\partial \chi(\mathbf{p};\mathbf{r},E'\to E)}{\partial p_{m_2}}$$

$$= \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)};\mathbf{w}_j^{(2)}\right)}{\partial p_k \partial d_j} = \int dV \int_{4\pi} d\mathbf{\Omega} \int_0^\infty dE\, w_{1,k}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \frac{\partial \Sigma_d(\mathbf{d};\mathbf{r},\mathbf{\Omega},E)}{\partial d_j}\ . \tag{143}$$

The relation shown in Eq. (143) provides an independent path for the mutual verification of the solutions $\mathbf{h}_j^{(2)}$ and $\mathbf{w}_j^{(2)}$. The expressions of the second-order sensitivities computed using Eq. (138) must be identical to those computed using Eq. (120).

That is, for $j = 1, \ldots, J_d;\ k = 1, \ldots, J_q$: $\dfrac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{h}_j^{(2)}\right)}{\partial d_j \partial q_k}$

$$= \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, h_{2,j}^{(2)}(\mathbf{r}, \boldsymbol{\Omega}, E) \frac{\partial Q(\mathbf{q}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial q_k}$$

$$= \frac{\partial^2 R\left(\boldsymbol{\alpha}, \varphi;\ \psi^{(1)}; \mathbf{g}_j^{(2)}\right)}{\partial q_k \partial d_j} = \int dV \int_{4\pi} d\boldsymbol{\Omega} \int_0^\infty dE\, g_{1,k}^{(2)}$$

$$\times (\mathbf{r}, \boldsymbol{\Omega}, E) \frac{\partial \Sigma_d(\mathbf{d}; \mathbf{r}, \boldsymbol{\Omega}, E)}{\partial d_j} . \tag{144}$$

The relation shown in Eq. (144) provides an independent path for the mutual verification of the solutions $\mathbf{h}_j^{(2)}$ and $\mathbf{g}_j^{(2)}$.

IV.G. Impact of Second-Order Sensitivities on Response Expected Value, Variance, and Skewness

Knowledge of the first- and second-order sensitivities is required to compute the following moments of the response distribution:

1. *The expected value of a response R*:

$$E(R) = R\left(\boldsymbol{\alpha}^0\right) + \frac{1}{2} \sum_{i=1}^{N_\alpha} \frac{\partial^2 R}{\partial \alpha_i^2} s_i^2 ,$$

where s_i denotes the standard deviation of the model parameter α_i.

2. *The variance of response*:

$$\mathrm{var}(R) = \sum_{i=1}^{N_\alpha} \left(\frac{\partial R}{\partial \alpha_i}\right)^2 s_i^2 + \frac{1}{2} \sum_{i=1}^{N_\alpha} \left(\frac{\partial^2 R}{\partial \alpha_i^2}\right)^2 s_i^4 .$$

3. *The skewness γ_1 of response*:

$$\gamma_1(R) = \frac{\mu_3(R)}{[\mathrm{var}(R)]^{[3/2]}} ,$$

where $\mu_3(R) = 3 \sum_{i=1}^{N_\alpha} \left(\frac{\partial R}{\partial \alpha_i}\right)^2 \frac{\partial^2 R}{\partial \alpha_i^2} s_i^4$ denoted the third central moment of the response distribution.

V. MULTIGROUP APPROXIMATION EXPRESSIONS OF THE 2ND-LASS AND SECOND-ORDER RESPONSE SENSITIVITIES

In the standard multigroup approximation, the system response defined in Eq. (8) takes on the following expression:

$$R(\boldsymbol{\alpha}, \varphi) = \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, \Sigma_d^g(\mathbf{d}^g; \mathbf{r}, \boldsymbol{\Omega})\, \varphi^g(\mathbf{r}, \boldsymbol{\Omega}) , \tag{145}$$

where

$\varphi^g(\mathbf{r}, \boldsymbol{\Omega})$ = multigroup flux

G = total number of energy groups considered for representing the physical system

$\Sigma_d^g(\mathbf{d}^g; \mathbf{r}, \boldsymbol{\Omega})$ = multigroup approximation of the function that models the interaction of the detector with the incident particles,

and where each vector $\mathbf{d}^g \triangleq \left[d_1^g, \ldots, d_{J_{dg}}^g\right]^\dagger$ is considered to comprise, as components, a total of J_{dg} imprecisely known model parameters that characterize $\Sigma_d^g(\mathbf{d}^g; \mathbf{r}, \boldsymbol{\Omega})$, within each group $g = 1, \ldots, G$, where G denotes the total number of energy groups considered for representing the physical system. In general, the number of components of $\mathbf{d}^g$ may vary from group to group.

The multigroup flux $\varphi^g(\mathbf{r}, \boldsymbol{\Omega})$ appearing in Eq. (145) is the solution of the standard multigroup approximation of the forward neutron transport equation and vacuum boundary condition defined in Eqs. (1) and (2), namely,

$$L^g(\boldsymbol{\alpha})\varphi^g(\mathbf{r}, \boldsymbol{\Omega}) = Q^g(\mathbf{q}^g; \mathbf{r}, \boldsymbol{\Omega}), \quad g = 1, \ldots, G \tag{146}$$

and

$$\varphi^g(\mathbf{r}_s, \boldsymbol{\Omega}, E) = 0, \mathbf{r}_s \in \partial V, \boldsymbol{\Omega} \cdot \mathbf{n} < 0 , \tag{147}$$

where $Q^g(\mathbf{q}^g; \mathbf{r}, \boldsymbol{\Omega})$ denotes the group source and the operator $L^g(\boldsymbol{\alpha})\varphi^g(\mathbf{r}, \boldsymbol{\Omega})$ is customarily defined as follows:

$$L^g(\boldsymbol{\alpha})\varphi^g(\mathbf{r}, \boldsymbol{\Omega}) \triangleq \boldsymbol{\Omega} \bullet \nabla\varphi^g(\mathbf{r}, \boldsymbol{\Omega}) + \Sigma_t^g(\mathbf{t}^g; \mathbf{r})\, \varphi^g(\mathbf{r}, \boldsymbol{\Omega})$$
$$- \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \Sigma_s^{g' \to g}\left(\mathbf{s}^{g'g}; \mathbf{r}, \boldsymbol{\Omega}' \to \boldsymbol{\Omega}\right) \varphi^{g'}(\mathbf{r}, \boldsymbol{\Omega}')$$
$$- \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \chi^{g' \to g}\left(\mathbf{p}^{g'g}; \mathbf{r}\right) (\nu\Sigma_f)^{g'}\left(\mathbf{f}^{g'}; \mathbf{r}\right) \varphi^{g'}(\mathbf{r}, \boldsymbol{\Omega}'),$$
$$g = 1, \ldots, G . \tag{148}$$

The definition of the group total macroscopic cross section $\Sigma_t^g(\mathbf{t}^g;\mathbf{r})$, the group fission macroscopic cross section $\left(\nu\Sigma_f\right)^g(\mathbf{f}^g;\mathbf{r})$, the multigroup scattering transfer matrix $\Sigma_s^{g'\to g}\left(\mathbf{s}^{g'g};\mathbf{r},\mathbf{\Omega}'\to\mathbf{\Omega}\right)$, and the fission spectrum matrix are defined in the customary way $\chi^{g'\to g}\left(\mathbf{p}^{g'g};\mathbf{r}\right)$. Denoting the adjoint multigroup flux as $\psi^{(1),g}(\mathbf{r},\mathbf{\Omega})$, the multigroup approximation of the adjoint neutron transport operator defined in Eq. (23) is customarily defined as

$$
\begin{aligned}
A^{(1),g}(\boldsymbol{\alpha})\psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) &\triangleq -\mathbf{\Omega}\cdot\nabla\psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \\
&+\Sigma_t^g(\mathbf{t}^g;\mathbf{r})\,\psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) - \sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\Sigma_s^{g\to g'} \\
&\times\left(\mathbf{s}^{gg'};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}'\right)\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \\
&-\nu\Sigma_f^g(\mathbf{f}^g;\mathbf{r})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\chi^{g\to g'}\left(\mathbf{p}^{gg'};\mathbf{r}\right) \\
&\times\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')\,,\ g=1,\ldots,G\ .
\end{aligned}
\tag{149}
$$

Just as for the multigroup detector cross section $\Sigma_d^g(\mathbf{d}^g;\mathbf{r},\mathbf{\Omega})$, which depends on the vector of model parameters $\mathbf{d}^g$ for each group $g=1,\ldots,G$, each multigroup total macroscopic cross section $\Sigma_t^g(\mathbf{t}^g;\mathbf{r})$ is considered to depend on $\mathbf{t}^g\triangleq\left[t_1^g,\ldots,t_{J_{tg}}^g\right]^\dagger$ imprecisely known model parameters, where J_{tg} denotes the total number of model parameters that characterize $\Sigma_t^g(\mathbf{t}^g;\mathbf{r})$ in each group $g=1,\ldots,G$. Similarly, the multigroup scattering transfer matrix $\Sigma_s^{g'\to g}\left(\mathbf{s}^{g'g};\mathbf{r},\mathbf{\Omega}'\to\mathbf{\Omega}\right)$ is considered to depend on the vector $\mathbf{s}^{g'g}\triangleq\left[s_1^{g'g},\ldots,s_{J_{sg'g}}^{g'g}\right]^\dagger$, comprising $J_{sg'g}$ imprecisely known model parameters within each group $g,g'=1,\ldots,G$. The macroscopic group fission cross section is denoted as $\left(\nu\Sigma_f\right)^g(\mathbf{f}^g;\mathbf{r})$ and is considered to depend on the vector $\mathbf{f}^g\triangleq\left[f_1^g,\ldots,f_{J_{fg}}^g\right]^\dagger$, comprising J_{fg} imprecisely known model parameters within each group $g=1,\ldots,G$. Furthermore, the fission spectrum matrix is defined as $\chi^{g'\to g}\left(\mathbf{p}^{g'g};\mathbf{r}\right)$ and is considered to depend on the vector $\mathbf{p}^{g'g}\triangleq\left[p_1^{g'g},\ldots,p_{J_{pg'g}}^{g'g}\right]^\dagger$, comprising $J_{pg'g}$ imprecisely known model parameters within each group $g,g'=1,\ldots,G$. Finally, the group source is denoted as $Q^g(\mathbf{q}^g;\mathbf{r},\mathbf{\Omega})$ and is considered to depend on the vector $\mathbf{q}^g\triangleq\left[q_1^g,\ldots,q_{J_{qg}}^g\right]^\dagger$, comprising J_{qg} imprecisely known scalar-valued parameters within each group $g=1,\ldots,G$.

As in Secs. II, III, and IV, the total number of imprecisely known model parameters will be denoted by the vector $\boldsymbol{\alpha}\triangleq\left[\alpha_1,\ldots,\alpha_{J_\alpha}\right]^\dagger$. To simplify the notation, the superscripts g and g', which denote the group dependence of the vectors $\mathbf{d}^g$, $\mathbf{t}^g$, $\mathbf{s}^{g'g}$, $\mathbf{f}^g$, $\mathbf{p}^{g'g}$, $\mathbf{q}^g$, and of their components, will *not* be shown explicitly in the multigroup derivations in the remainder of this work.

In the multigroup approximation, the exact expressions given in Eqs. (29) through (34) for the first-order sensitivities take on the following approximate expressions:

$$
\begin{aligned}
\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\boldsymbol{\psi}^{(1)}\right)}{\partial t_j} &= -\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \\
&\times\varphi^g(\mathbf{r},\mathbf{\Omega})\frac{\partial\Sigma_t^g(\mathbf{t};\mathbf{r},E)}{\partial t_j}, \\
&j=1,\ldots,J_t\ ;
\end{aligned}
\tag{150}
$$

$$
\begin{aligned}
\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\boldsymbol{\psi}^{(1)}\right)}{\partial s_j} &= \sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ \psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \\
&\times\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\frac{\partial\Sigma_s^{g'\to g}(\mathbf{s};\mathbf{r},\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial s_j} \\
&\times\varphi^{g'}(\mathbf{r},\mathbf{\Omega}'),\ j=1,\ldots,J_s\ ;
\end{aligned}
\tag{151}
$$

$$
\begin{aligned}
\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\boldsymbol{\psi}^{(1)}\right)}{\partial f_j} &= \sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ \psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \\
&\times\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\frac{\partial\left[(\nu\Sigma)_f^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_j} \\
&\times\chi^{g'\to g}(\mathbf{p};\mathbf{r})\varphi^{g'}(\mathbf{r},\mathbf{\Omega}'), \\
&j=1,\ldots,J_f\ ;
\end{aligned}
\tag{152}
$$

$$
\begin{aligned}
\frac{\partial R\left(\boldsymbol{\alpha},\varphi;\boldsymbol{\psi}^{(1)}\right)}{\partial p_j} &= \sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\,\psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \\
&\times\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j} \\
&\times\left(\nu\Sigma_f\right)^{g'}(\mathbf{f};\mathbf{r})\varphi^{g'}(\mathbf{r},\mathbf{\Omega}'), \\
&j=1,\ldots,J_p\ ;
\end{aligned}
\tag{153}
$$

$$\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\boldsymbol{\psi}^{(1)})}{\partial q_j}=\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\frac{\partial Q^{g}(\mathbf{q};\mathbf{r},\boldsymbol{\Omega})}{\partial q_j}\,\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega}),\quad j=1,\ldots,J_q\;;\tag{154}$$

and

$$\frac{\partial R(\boldsymbol{\alpha},\varphi;\,\boldsymbol{\psi}^{(1)})}{\partial d_j}=\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\frac{\partial \Sigma_d^{g}(\mathbf{d};\mathbf{r},\boldsymbol{\Omega})}{\partial d_j}\,\varphi^{g}(\mathbf{r},\boldsymbol{\Omega}),\quad j=1,\ldots,J_d\;,\tag{155}$$

where the vector-valued quantity $\boldsymbol{\psi}^{(1)}(\mathbf{r},\boldsymbol{\Omega})\triangleq\left[\psi^{(1),1}(\mathbf{r},\boldsymbol{\Omega}),\ldots,\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega}),\ldots,\psi^{(1),G}(\mathbf{r},\boldsymbol{\Omega})\right]^{\dagger}$ comprises as components the multigroup adjoint fluxes $\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega})$, which are obtained as the solutions of the multigroup approximation of the 1st-LASS given by Eqs. (25) and (26), namely,

$$A^{(1),g}(\boldsymbol{\alpha})\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega})=\Sigma_d^{g}(\mathbf{d}^{0};\mathbf{r},\boldsymbol{\Omega}),\quad g=1,\ldots,G\;,\tag{156}$$

subject to adjoint boundary condition:

$$\psi^{(1),g}(\mathbf{r}_s,\boldsymbol{\Omega})=0\;,\mathbf{r}_s\in\partial V,\boldsymbol{\Omega}\cdot\mathbf{n}>0\;.\tag{157}$$

V.A. Multigroup Expressions of $\partial^2 R/\partial t_j\partial\alpha_{m_2},\ j=1,\ldots,J_t;\ m_2=1,\ldots,J_\alpha$

For the sake of simplicity, the functional dependence of the response R will be omitted henceforth.

In the multigroup approximation, the expressions of $\partial^2 R/\partial t_j\,\partial\alpha_{m_2},\ j=1,\ldots,J_t;\ m_2=1,\ldots,J_\alpha$ take on the following forms:

$$\text{For } m_2=1,\ldots,J_t:\ \frac{\partial^2 R}{\partial t_j\partial t_{m_2}}=-\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega})\varphi^{g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial^2\Sigma_t^{g}(\mathbf{t};\mathbf{r},\boldsymbol{\Omega})}{\partial t_j\partial t_{m_2}}$$
$$-\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\left[\psi_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega})+\psi_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\varphi^{g}(\mathbf{r},\boldsymbol{\Omega})\right]\frac{\partial\Sigma_t^{g}(\mathbf{t};\mathbf{r},\boldsymbol{\Omega})}{\partial t_{m_2}}\;;\tag{158}$$

$$\text{For } m_2=1,\ldots,J_s:\frac{\partial^2 R}{\partial t_j\partial s_{m_2}}=\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi}d\boldsymbol{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi}d\boldsymbol{\Omega}'\,\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}\;;\tag{159}$$

$$\text{For } m_2=1,\ldots,J_f:\frac{\partial^2 R}{\partial t_j\partial f_{m_2}}=\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi}d\boldsymbol{\Omega}'\,\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\,\chi^{g'\to g}(\mathbf{p};\mathbf{r})\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}}$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^{g}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}}\sum_{g'=1}^{G}\int_{4\pi}d\boldsymbol{\Omega}'\chi^{g\to g'}(\mathbf{p};\mathbf{r})\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')\;;\tag{160}$$

$$\text{For } m_2=1,\ldots,J_p:\ \frac{\partial^2 R}{\partial t_j\partial p_{m_2}}=\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})(\nu\Sigma_f)^{g}(\mathbf{f};\mathbf{r})\sum_{g'=1}^{G}\int_{4\pi}d\boldsymbol{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial\chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi}d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi}d\boldsymbol{\Omega}'(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}\;;\tag{161}$$

$$\text{For } m_2 = 1, \ldots, J_q : \frac{\partial^2 R}{\partial t_j \partial q_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi_{2,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) \frac{\partial Q^g(\mathbf{q}; \mathbf{r}, \mathbf{\Omega})}{\partial q_{m_2}} \; ; \tag{162}$$

and

$$\text{For } m_2 = 1, \ldots, J_d : \frac{\partial^2 R}{\partial t_j \partial d_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi_{1,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) \frac{\partial \Sigma_d^g(\mathbf{d}; \mathbf{r}, \mathbf{\Omega})}{\partial d_{m_2}} \; , \tag{163}$$

where

$\varphi^g(\mathbf{r}, \mathbf{\Omega})$ = solution of the multigroup approximation of the forward neutron transport Eqs. (146) and (147)

$\psi^{(1),g}(\mathbf{r}, \mathbf{\Omega})$ = solution of the multigroup approximation of the 1st-LASS represented by Eqs. (156) and (157), respectively

$\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega})$ = solutions of the multigroup approximation of the 2nd-LASS represented by Eqs. (164), (165), and (166)

$$L^g(\boldsymbol{\alpha})\psi_{1,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) = -\varphi^g(\mathbf{r}, \mathbf{\Omega}) \frac{\partial \Sigma_t^g(\mathbf{t}; \mathbf{r})}{\partial t_j}, \quad j = 1, \ldots, J_t; \; g = 1, \ldots, G \tag{164}$$

and

$$A^{(1),g}(\boldsymbol{\alpha}^0)\psi_{2,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) = -\psi^{(1),g}(\mathbf{r}, \mathbf{\Omega}) \frac{\partial \Sigma_t^g(\mathbf{t}^0; \mathbf{r})}{\partial t_j}, \quad j = 1, \ldots, J_t; \; g = 1, \ldots, G \; , \tag{165}$$

subject to the following boundary conditions:

$$\psi_{1,j}^{(2),g}(\mathbf{r}_s, \mathbf{\Omega}) = 0, \mathbf{\Omega} \cdot \mathbf{n} < 0; \quad \psi_{2,j}^{(2),g}(\mathbf{r}_s, \mathbf{\Omega}) = 0 \;, \mathbf{\Omega} \cdot \mathbf{n} > 0; \;\; \mathbf{r}_s \in \partial V; \; j = 1, \ldots, J_t; \; g = 1, \ldots, G \; . \tag{166}$$

V.B. Multigroup Expressions of $\partial^2 R / \partial s_j \partial \alpha_{m_2}$, $j = 1, \ldots, J_s$; $m_2 = 1, \ldots, J_\alpha$

The approximate multigroup expressions of $\partial^2 R/\partial s_j \,\partial \alpha_{m_2}$, $j = 1, \ldots, J_s$; $m_2 = 1, \ldots, J_\alpha$ are as follows:

$$\text{For } m_2 = 1, \ldots, J_t : \frac{\partial^2 R}{\partial s_j \partial t_{m_2}} = -\sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega} \left[\theta_{1,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) \psi^{(1),g}(\mathbf{r}, \mathbf{\Omega}) + \theta_{2,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) \varphi^g(\mathbf{r}, \mathbf{\Omega}) \right] \times \frac{\partial \Sigma_t^g(\mathbf{t}; \mathbf{r}, \mathbf{\Omega})}{\partial t_{m_2}} \; ; \tag{167}$$

$$\begin{aligned} \text{For } m_2 = 1, \ldots, J_s : \frac{\partial^2 R}{\partial s_j \partial s_{m_2}} &= \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi^{(1),g}(\mathbf{r}, \mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \varphi^{g'}(\mathbf{r}, \mathbf{\Omega}') \frac{\partial^2 \Sigma_s^{g' \to g}(\mathbf{s}; \mathbf{r}, \mathbf{\Omega}' \to \mathbf{\Omega})}{\partial s_j \partial s_{m_2}} \\ &+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{1,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \psi^{(1),g'}(\mathbf{r}, \mathbf{\Omega}') \frac{\partial \Sigma_s^{g \to g'}(\mathbf{s}; \mathbf{r}, \mathbf{\Omega} \to \mathbf{\Omega}')}{\partial s_{m_2}} \\ &+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{2,j}^{(2),g}(\mathbf{r}, \mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \varphi^{g'}(\mathbf{r}, \mathbf{\Omega}') \frac{\partial \Sigma_s^{g \to g'}(\mathbf{s}; \mathbf{r}, \mathbf{\Omega} \to \mathbf{\Omega}')}{\partial s_{m_2}} \; ; \end{aligned} \tag{168}$$

$$\text{For } m_2 = 1,...,J_f: \ \frac{\partial^2 R}{\partial s_j \partial f_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}} \int_{4\pi} d\mathbf{\Omega}' \sum_{g'=1}^{G} \chi^{g\to g'}(\mathbf{p};\mathbf{r},)\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \int_{4\pi} d\mathbf{\Omega}' \sum_{g'=1}^{G} \varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\chi^{g'\to g}(\mathbf{p};\mathbf{r}) \frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}}; \tag{169}$$

$$\text{For } m_2 = 1,...,J_p: \ \frac{\partial^2 R}{\partial s_j \partial p_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right] \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right] \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}; \tag{170}$$

$$\text{For } m_2 = 1,...,J_q: \frac{\partial^2 R}{\partial s_j \partial q_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})}{\partial q_{m_2}} \ ; \tag{171}$$

and

$$\text{For } m_2 = 1,...,J_d: \frac{\partial^2 R}{\partial s_j \partial d_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \theta_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_{m_2}} \ . \tag{172}$$

In Eqs. (167) through (172), the functions $\theta_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ and $\theta_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ are the solutions of the following 2nd-LASS:

$$L^g(\boldsymbol{\alpha}^0)\theta_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) = \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{r},\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial s_j} \varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\,;\ j=1,...,J_s;\ g=1,...,G \tag{173}$$

and

$$A^{(1),g}(\boldsymbol{\alpha}^0)\theta_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) = \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\ \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}{}_s(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_j}\ ,\ j=1,...,J_s;\ g=1,...,G\ , \tag{174}$$

subject to the following boundary condition:

$$\theta_{2,m_1}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega}) = 0\ ,\mathbf{\Omega}\cdot\mathbf{n}>0;\ \ \theta_{1,j}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega}) = 0,\ \mathbf{\Omega}\cdot\mathbf{n}<0;\ \mathbf{r}_s\in\partial V;\ j=1,...,J_s;\ \ g=1,...,G\ . \tag{175}$$

The expressions of the second-order sensitivities computed using Eq. (167) must be identical to those computed using Eq. (159).

$$\text{That is, for } j=1,...,J_s,\ \ k=1,...,J_t: \ \frac{\partial^2 R}{\partial s_j \partial t_k} = -\sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\left[\theta_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) + \theta_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\varphi^g(\mathbf{r},\mathbf{\Omega})\right]$$
$$\frac{\partial \Sigma_t^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})}{\partial t_k} = \frac{\partial^2 R}{\partial t_k \partial s_j} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\ \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_j}$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi_{2,k}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_j}\ . \tag{176}$$

V.C. Multigroup Expressions of $\partial^2 R/\partial f_j \partial \alpha_{m_2}$, $j = 1,...,J_f$; $m_2 = 1,...,J_\alpha$

The approximate multigroup expressions of $\partial^2 R/\partial f_j\, \partial\alpha_{m_2},\ j = 1,...,J_f;\ m_2 = 1,...,J_\alpha$ are as follows:

$$\text{For } m_2 = 1,...,J_t: \ \frac{\partial^2 R}{\partial f_j \partial t_{m_2}} = -\sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega} \left[u_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) + u_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \varphi^{g}(\mathbf{r},\mathbf{\Omega}) \right] \frac{\partial \Sigma_t^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})}{\partial t_{m_2}} \ ; \tag{177}$$

$$\text{For } m_2 = 1,...,J_s: \frac{\partial^2 R}{\partial f_j \partial s_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, u_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, u_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}} \ ; \tag{178}$$

$$\text{For } m_2 = 1,...,J_f: \ \frac{\partial^2 R}{\partial f_j \partial f_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \chi^{g'\to g}(\mathbf{p};\mathbf{r}) \frac{\partial^2 \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r}) \right]}{\partial f_j \partial f_{m_2}}$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega} u_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial \left[\nu\Sigma_f(\mathbf{f};\mathbf{r},E) \right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \chi^{g\to g'}(\mathbf{p};\mathbf{r},) \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, u_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \varphi^{g'}(\mathbf{r},\mathbf{\Omega}'E) \frac{\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{4\pi} \frac{\partial \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r}) \right]}{\partial f_{m_2}} \ ; \tag{179}$$

$$\text{For } m_2 = 1,...,J_p: \ \frac{\partial^2 R}{\partial f_j \partial p_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, \psi^{(1),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\, \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}} \frac{\partial \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r}) \right]}{\partial f_j}$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ u_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \left[(\nu\Sigma_f)^{g}(\mathbf{f};\mathbf{r}) \right] \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}$$
$$+ \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, u_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r}) \right] \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}} \ ; \tag{180}$$

$$\text{For } m_2 = 1,...,J_q: \frac{\partial^2 R}{\partial f_j \partial q_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, u_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})}{\partial q_{m_2}} \ ; \tag{181}$$

and

$$\text{For } m_2 = 1,...,J_d: \frac{\partial^2 R}{\partial f_j \partial d_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ u_{1,j}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_{m_2}} \ . \tag{182}$$

In Eqs. (177) through (182), the functions $u_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ and $u_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ are the solutions of the following 2nd-LASS:

$$L^g(\boldsymbol{\alpha}^0)u_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) = \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\chi^{g'\to g}(\mathbf{p}^0;\mathbf{r})\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_j},$$
$$j = 1,\ldots,J_f;\ g = 1,\ldots,G \tag{183}$$

and

$$A^{(1),g}(\boldsymbol{\alpha}^0)u_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) = \frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_j}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')\chi^{g\to g'}(\mathbf{p}^0;\mathbf{r})\ ,\ j = 1,\ldots,J_f;\ g = 1,\ldots,G\ , \tag{184}$$

subject to the following boundary condition:

$$u_{2,m_1}^{(2)}(\mathbf{r}_s,\boldsymbol{\Omega}) = 0\ ,\boldsymbol{\Omega}\cdot\mathbf{n} > 0;\ \ u_{1,j}^{(2),g}(\mathbf{r}_s,\boldsymbol{\Omega}) = 0,\boldsymbol{\Omega}\cdot\mathbf{n} < 0;\ \ \mathbf{r}_s \in \partial V;\ \ j = 1,\ldots,J_f;\ \ g = 1,\ldots,G\ . \tag{185}$$

The expressions of the second-order sensitivities computed using Eq. (177) must be identical to those computed using Eq. (160).

$$\text{That is, for } j = 1,\ldots,J_f;\ \ k = 1,\ldots,J_t:\ \frac{\partial^2 R}{\partial f_j\,\partial t_k} = -\sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\left[u_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega}) + u_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\varphi^g(\mathbf{r},\boldsymbol{\Omega})\right]$$
$$\times\ \frac{\partial\Sigma_t^g(\mathbf{t};\mathbf{r},\boldsymbol{\Omega})}{\partial t_k} = \frac{\partial^2 R}{\partial t_k\partial f_j} = \sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\ \psi_{2,k}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\ \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\,\chi^{g'\to g}(\mathbf{p};\mathbf{r})$$
$$\times\ \frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_j} + \sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\ \psi_{1,k}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_k}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\chi^{g\to g'}(\mathbf{p};\mathbf{r})\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')\ . \tag{186}$$

Also, expressions of the second-order sensitivities computed using Eq. (178) must be identical to those computed using Eq. (169).

$$\text{That is, for } j = 1,\ldots,J_f;\ k = 1,\ldots,J_s:\ \frac{\partial^2 R}{\partial f_j\partial s_k} = \sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\,u_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')$$
$$\times\ \frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_k} + \sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\ u_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_k}$$
$$= \frac{\partial^2 R}{\partial s_k\partial f_j} = \sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\ \theta_{1,k}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_j}\int_{4\pi} d\boldsymbol{\Omega}'\sum_{g'=1}^{G}\chi^{g\to g'}(\mathbf{p};\mathbf{r},)\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\,\theta_{2,k}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\int_{4\pi} d\boldsymbol{\Omega}'\sum_{g'=1}^{G}\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\chi^{g'\to g}(\mathbf{p};\mathbf{r})\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_j}\ . \tag{187}$$

V.D. Multigroup Expressions of $\partial^2 R/\partial p_j\partial\alpha_{m_2}$, $j = 1,\ldots,J_p;\ m_2 = 1,\ldots,J_\alpha$

The approximate multigroup expressions of $\partial^2 R/\partial p_j\,\partial\alpha_{m_2}$, $j = 1,\ldots,J_p;\ m_2 = 1,\ldots,J_\alpha$ are as follows:

$$\text{For } m_2 = 1,\ldots,J_t:\ \frac{\partial^2 R}{\partial p_j\partial t_{m_2}} = -\sum_{g=1}^{G}\int dV\int_{4\pi} d\boldsymbol{\Omega}\left[w_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega}) + w_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})\varphi^g(\mathbf{r},\boldsymbol{\Omega})\right]$$
$$\frac{\partial\Sigma_t^g(\mathbf{t};\mathbf{r},\boldsymbol{\Omega})}{\partial t_{m_2}}\ ; \tag{188}$$

$$\text{For } m_2 = 1,...,J_s: \quad \frac{\partial^2 R}{\partial p_j \partial s_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}') \times \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}} + \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega} w_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}') \times \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}\;; \tag{189}$$

$$\text{For } m_2 = 1,...,J_f: \quad \frac{\partial^2 R}{\partial p_j \partial f_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, \psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}') \frac{\partial \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j} \frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}} + \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \frac{\partial\left[(\nu\Sigma_f)^{g}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \chi^{g\to g'}(\mathbf{p};\mathbf{r}) \psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}') + \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}') \chi^{g'\to g}(\mathbf{p};\mathbf{r}) \frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}}\;; \tag{190}$$

$$\text{For } m_2 = 1,...,J_p: \quad \frac{\partial^2 R}{\partial p_j \partial p_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, \psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right] \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}') \frac{\partial^2 \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j \partial p_{m_2}} + \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \left[(\nu\Sigma_f)^{g}(\mathbf{f};\mathbf{r})\right] \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}') \frac{\partial \chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}} + \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right] \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}') \frac{\partial \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}\;; \tag{191}$$

$$\text{For } m_2 = 1,...,J_q: \frac{\partial^2 R}{\partial p_j \partial q_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \frac{\partial Q^{g}(\mathbf{q};\mathbf{r},\boldsymbol{\Omega})}{\partial q_{m_2}}\;; \tag{192}$$

and

$$\text{For } m_2 = 1,...,J_d: \frac{\partial^2 R}{\partial p_j \partial d_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) \frac{\partial \Sigma_d^{g}(\mathbf{d};\mathbf{r},\boldsymbol{\Omega})}{\partial d_{m_2}}\;. \tag{193}$$

In Eqs. (188) through (193), the functions $u_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})$ and $u_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega})$ are the solutions of the following 2nd-LASS:

$$L^{g}(\boldsymbol{\alpha}^0) w_{1,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) = \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}') (\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r}) \frac{\partial \chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_j}, \quad j = 1,...,J_p;\ g = 1,...,G \tag{194}$$

and

$$A^{(1),g}(\boldsymbol{\alpha}^0) w_{2,j}^{(2),g}(\mathbf{r},\boldsymbol{\Omega}) = (\nu\Sigma_f)^{g}(\mathbf{f};\mathbf{r}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}'\, \psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}') \frac{\partial \chi^{g\to g'}(\mathbf{p};\mathbf{r})}{(4\pi)\partial p_j}, \ j = 1,...,J_p;\ g = 1,...,G\;, \tag{195}$$

subject to the following boundary condition:

$$w_{2,m_1}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega})=0\ ,\mathbf{\Omega}\cdot\mathbf{n}>0;\ w_{1,j}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega})=0,\mathbf{\Omega}\cdot\mathbf{n}<0;\ \mathbf{r}_s\in\partial V;\ j=1,\dots,J_p;\ g=1,\dots,G\ . \tag{196}$$

The expressions of the second-order sensitivities computed using Eq. (188) must be identical to those computed using Eq. (161).

$$\text{That is, for } j=1,\dots,J_p;\ k=1,\dots,J_t:\ \frac{\partial^2 R}{\partial p_j\partial t_k}=-\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\left[w_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\psi^{(1),g}(\mathbf{r},\mathbf{\Omega})+w_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\varphi^{g}(\mathbf{r},\mathbf{\Omega})\right]$$
$$\times\frac{\partial\Sigma_t^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})}{\partial t_k}=\frac{\partial^2 R}{\partial t_k\partial p_j}=\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ \psi_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega})\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\ \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')$$
$$\times\frac{\partial\chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_j}+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ \psi_{2,k}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]\varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j}\quad . \tag{197}$$

Furthermore, the expressions of the second-order sensitivities computed using Eq. (189) must be identical to those computed using Eq. (170).

$$\text{That is, for } j=1,\dots,J_p; k=1,\dots,J_s:\ \frac{\partial^2 R}{\partial p_j\partial s_k}=\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\, w_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')$$
$$\times\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_k}+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\, w_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_k}$$
$$=\frac{\partial^2 R}{\partial s_k\partial p_j}=\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\,\theta_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega})\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_j}$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\,\theta_{2,k}^{(2),g}(\mathbf{r},\mathbf{\Omega},E)\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]\varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j}\quad . \tag{198}$$

Finally, the expressions of the second-order sensitivities computed using Eq. (190) must be identical to those computed using Eq. (180).

$$\text{That is, for } j=1,\dots,J_p;\ k=1,\dots,J_f:\ \frac{\partial^2 R}{\partial p_j\partial f_k}=\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ \psi^{(1),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\varphi^{g'}(\mathbf{r},\mathbf{\Omega}')$$
$$\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j}\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_k}+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\, w_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_k}\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\chi^{g\to g'}(\mathbf{p};\mathbf{r})\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\, w_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\chi^{g'\to g}(\mathbf{p};\mathbf{r})\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_k}$$
$$=\frac{\partial^2 R}{\partial f_k\partial p_j}=\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ \psi^{(1),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j}\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_k}$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ u_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega})\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_j}$$
$$+\sum_{g=1}^{G}\int dV\int_{4\pi} d\mathbf{\Omega}\ u_{2,k}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\left[(\nu\Sigma_f)^{g'}(\mathbf{f};\mathbf{r})\right]\varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_j}\ . \tag{199}$$

V.E. Multigroup Expressions of $\partial^2 R/\partial q_j \partial\alpha_{m_2}$, $j = 1,...,J_q$; $m_2 = 1,...,J_\alpha$

The multigroup form of the 2nd-LASS to be solved for the components $g_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ and $g_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ needed for computing the multigroup expressions of the second-order sensitivities $\partial^2 R/\partial q_j\,\partial\alpha_{m_2}$, $j = 1,...,J_q$; $m_2 = 1,...,J_\alpha$ is as follows:

$$L^g\left(\boldsymbol{\alpha}^0\right)g_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) = \frac{\partial Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})}{\partial q_j},\ j = 1,...,J_q;\ g = 1,...,G \tag{200}$$

and

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)g_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) = 0,\ j = 1,...,J_q;\ \ g = 1,...,G\ , \tag{201}$$

subject to the following boundary condition:

$$g_{1,j}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} < 0;\ \ g_{2,j}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} > 0; \mathbf{r}_s \in \partial V;\ j = 1,...,J_q;\ \ g = 1,...,G\ . \tag{202}$$

It is evident that the unique solution of the homogeneous linear Eq. (201) subject to the linear homogeneous boundary condition in Eq. (202) is

$$g_{2,j}^{(2)}(\mathbf{r},\mathbf{\Omega}) \equiv 0\ , j = 1,...,J_q;\ \ g = 1,...,G\ . \tag{203}$$

The approximate multigroup expressions of $\partial^2 R/\partial q_j\,\partial\alpha_{m_2}$, $j = 1,...,J_q$; $m_2 = 1,...,J_\alpha$ are as follows:

$$\text{For } j = 1,...,J_q;\ m_2 = 1,...,J_t:\ \frac{\partial^2 R}{\partial q_j \partial t_{m_2}} = -\sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ g_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\psi^{(1),g}(\mathbf{r},\mathbf{\Omega})\frac{\partial\Sigma_t^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})}{\partial t_{m_2}}\ ; \tag{204}$$

$$\text{For } j = 1,...,J_q;\ \ m_2 = 1,...,J_s:\ \frac{\partial^2 R}{\partial q_j \partial s_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\, g_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\ \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}') \times \frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}; \tag{205}$$

$$\text{For } j = 1,...,J_q;\ \ m_2 = 1,...,J_f:\ \frac{\partial^2 R}{\partial q_j \partial f_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\, g_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}} \times \sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\,\chi^{g\to g'}(\mathbf{p};\mathbf{r})\psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}'): \tag{206}$$

$$\text{For } j = 1,...,J_q;\ \ m_2 = 1,...,J_p:\ \frac{\partial^2 R}{\partial q_j \partial p_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\, g_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right] \times \sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\ \psi^{(1),g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}\ ; \tag{207}$$

$$\text{For } j = 1,...,J_q;\ m_2 = 1,...,J_q:\ \frac{\partial^2 R}{\partial q_j \partial q_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ \psi^{(1),g}(\mathbf{r},\mathbf{\Omega})\frac{\partial^2 Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})}{\partial q_j \partial q_{m_2}}\ ; \tag{208}$$

and

$$\text{For } j=1,\ldots,J_q;\;\; m_2=1,\ldots,J_d:\; \frac{\partial^2 R}{\partial q_j \partial d_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\; g_{1,j}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\boldsymbol{\Omega})}{\partial d_{m_2}}\;. \tag{209}$$

The expressions of the second-order sensitivities computed using Eq. (204) must be identical to those computed using Eq. (162).

$$\text{That is, for } j=1,\ldots,J_q;\;\; k=1,\ldots,J_t:\; \frac{\partial^2 R}{\partial q_j \partial t_k} = -\sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\, g_{1,j}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\psi^{(1),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial \Sigma_t^g(\mathbf{t};\mathbf{r},\boldsymbol{\Omega})}{\partial t_k}$$

$$= \frac{\partial^2 R}{\partial t_k \partial q_j} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\; \psi_{2,k}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial Q^g(\mathbf{q};\mathbf{r},\boldsymbol{\Omega})}{\partial q_j}\;. \tag{210}$$

The expressions of the second-order sensitivities computed using Eq. (205) must be identical to those computed using Eq. (171).

$$\text{That is, for } j=1,\ldots,J_q;\;\; k=1,\ldots,J_s:\; \frac{\partial^2 R}{\partial q_j \partial s_k} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\; g_{1,j}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')$$

$$\times \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_k} = \frac{\partial^2 R}{\partial s_k \partial q_j} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\,\theta_{2,k}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial Q^g(\mathbf{q};\mathbf{r},\boldsymbol{\Omega})}{\partial q_j}\;. \tag{211}$$

The expressions of the second-order sensitivities computed using Eq. (206) must be identical to those computed using Eq. (181).

$$\text{That is, for } j=1,\ldots,J_q;\;\; k=1,\ldots,J_f:\; \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\,\mathbf{g}_j^{(2)}\right)}{\partial q_j \partial f_k} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\; g_{1,j}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial f_k}$$

$$\times \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\chi^{g\to g'}(\mathbf{p};\mathbf{r})\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}') = \frac{\partial^2 R\left(\boldsymbol{\alpha},\varphi;\,\psi^{(1)};\,\mathbf{u}_j^{(2)}\right)}{\partial f_k \partial q_j}$$

$$= \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\; u_{2,k}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial Q^g(\mathbf{q};\mathbf{r},\boldsymbol{\Omega})}{\partial q_j}\;. \tag{212}$$

The expressions of the second-order sensitivities computed using Eq. (207) must be identical to those computed using Eq. (192).

$$\text{That is, for } j=1,\ldots,J_q;\;\; k=1,\ldots,J_p:\; \frac{\partial^2 R}{\partial q_j \partial p_k} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\; g_{1,j}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]$$

$$\times \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\psi^{(1),g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial \chi^{g\to g'}(\mathbf{p};\mathbf{r})}{\partial p_k} = \frac{\partial^2 R}{\partial p_k \partial q_j} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\boldsymbol{\Omega}\, w_{2,k}^{(2),\,g}(\mathbf{r},\boldsymbol{\Omega})\frac{\partial Q^g(\mathbf{q};\mathbf{r},\boldsymbol{\Omega})}{\partial q_j}\;. \tag{213}$$

V.F. Multigroup Expressions of $\partial^2 R/\partial d_j \partial \alpha_{m_2}$, $j = 1,...,J_d$; $m_2 = 1,...,J_\alpha$

The multigroup form of the 2nd-LASS to be solved for the components $h_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ and $h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$ needed for computing the multigroup expressions of the second-order sensitivities $\partial^2 R/\partial d_j\, \partial\alpha_{m_2},\ j = 1,...,J_d;\ m_2 = 1,...,J_\alpha$ is as follows:

$$L^g\left(\boldsymbol{\alpha}^0\right)h_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) = 0,\ j = 1,...,J_q;\ g = 1,...,G \tag{214}$$

and

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) = \frac{\partial\Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j},\ j = 1,...,J_q;\ g = 1,...,G\ , \tag{215}$$

subject to the following boundary condition:

$$h_{1,j}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} < 0;\quad h_{2,j}^{(2),g}(\mathbf{r}_s,\mathbf{\Omega}) = 0\ , \mathbf{\Omega}\cdot\mathbf{n} > 0;\ \mathbf{r}_s \in \partial V,\ j = 1,...,J_q;\ g = 1,...,G\ . \tag{216}$$

It is evident that the unique solution of the homogeneous linear Eq. (214) subject to the linear homogeneous boundary condition in Eq. (216) is

$$h_{1,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \equiv 0\ , j = 1,...,J_q;\quad g = 1,...,G\ . \tag{217}$$

The approximate multigroup expressions of $\partial^2 R/\partial d_j\, \partial\alpha_{m_2},\ j = 1,...,J_d;\ m_2 = 1,...,J_\alpha$ are as follows:

$$\text{For } m_2 = 1,...,J_t:\ \frac{\partial^2 R}{\partial d_j \partial t_{m_2}} = -\sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\varphi^g(\mathbf{r},\mathbf{\Omega})\frac{\partial\Sigma_t^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})}{\partial t_{m_2}}\ ; \tag{218}$$

$$\text{For } m_2 = 1,...,J_s:\ \frac{\partial^2 R}{\partial d_j \partial s_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \times \frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\mathbf{r},\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}\ ; \tag{219}$$

$$\text{For } m_2 = 1,...,J_f:\ \frac{\partial^2 R}{\partial d_j \partial f_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\chi^{g'\to g}(\mathbf{p};\mathbf{r}) \times \frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_{m_2}}\ ; \tag{220}$$

$$\text{For } m_2 = 1,...,J_p:\ \frac{\partial^2 R}{\partial d_j \partial p_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\left[\left(\nu\Sigma_f\right)^{g'}(\mathbf{f};\mathbf{r})\right] \times \varphi^{g'}(\mathbf{r},\mathbf{\Omega}')\frac{\partial\chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_{m_2}}\ ; \tag{221}$$

$$\text{For } m_2 = 1,...,J_q:\ \frac{\partial^2 R}{\partial d_j \partial q_{m_2}} = \sum_{g=1}^{G}\int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})\frac{\partial Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})}{\partial q_{m_2}}\ ; \tag{222}$$

and

$$\text{For } m_2 = 1,...,J_d: \frac{\partial^2 R}{\partial d_j \partial d_{m_2}} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ \varphi^g(\mathbf{r},\mathbf{\Omega}) \frac{\partial^2 \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j \partial d_{m_2}} \ . \tag{223}$$

The expressions of the second-order sensitivities computed using Eq. (218) must be identical to those computed using Eq. (163).

$$\text{That is, for } j = 1,...,J_d;\ \ k = 1,...,J_t: \ \frac{\partial^2 R}{\partial d_j \partial t_k} = -\sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \varphi^g(\mathbf{r},\mathbf{\Omega}) \frac{\partial \Sigma_t^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})}{\partial t_k}$$

$$= \frac{\partial^2 R}{\partial t_k \partial d_j} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ \psi_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j} \ . \tag{224}$$

The expressions of the second-order sensitivities computed using Eq. (219) must be identical to those computed using Eq. (172).

$$\text{That is, for } j = 1,...,J_d;\ \ k = 1,...,J_s: \ \frac{\partial^2 R}{\partial d_j \partial s_k} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}')$$

$$\times \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{r},\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial s_k} = \frac{\partial^2 R}{\partial s_k \partial d_j} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ \theta_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j} \ . \tag{225}$$

The expressions of the second-order sensitivities computed using Eq. (220) must be identical to those computed using Eq. (182).

$$\text{That is, for } j = 1,...,J_d;\ k = 1,...,J_f: \frac{\partial^2 R}{\partial d_j \partial f_k} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\ \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \chi^{g'\to g}(\mathbf{p};\mathbf{r})$$

$$\times \frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}(\mathbf{f};\mathbf{r})\right]}{\partial f_k} = \frac{\partial^2 R}{\partial f_k \partial d_j} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ u_{1,k}^{(2)}(\mathbf{r},\mathbf{\Omega},E) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j} \ . \tag{226}$$

The expressions of the second-order sensitivities computed using Eq. (221) must be identical to those computed using Eq. (193).

$$\text{That is, for } j = 1,...,J_d;\ k = 1,...,J_p: \ \frac{\partial^2 R}{\partial d_j \partial p_k} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega})$$

$$\times \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \left[\left(\nu\Sigma_f\right)^{g'}(\mathbf{f};\mathbf{r})\right] \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \chi^{g'\to g}(\mathbf{p};\mathbf{r})}{\partial p_k}$$

$$= \frac{\partial^2 R}{\partial p_k \partial d_j} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\, w_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j} \ . \tag{227}$$

The expressions of the second-order sensitivities computed using Eq. (222) must be identical to those computed using Eq. (209).

That is, for $j = 1, \ldots, J_d;\ k = 1, \ldots, J_q:\ \dfrac{\partial^2 R}{\partial d_j \partial q_k}$

$$= \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ h_{2,j}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})}{\partial q_k}$$

$$= \frac{\partial^2 R}{\partial q_k \partial d_j} = \sum_{g=1}^{G} \int dV \int_{4\pi} d\mathbf{\Omega}\ g_{1,k}^{(2),g}(\mathbf{r},\mathbf{\Omega}) \frac{\partial \Sigma_d^g(\mathbf{d};\mathbf{r},\mathbf{\Omega})}{\partial d_j}\ . \tag{228}$$

V.G. Second-Order Derivatives of Typical Multigroup Cross Sections with Respect to Typical Model Parameters

A generic macroscopic group cross section $\Sigma_x^g(\mathbf{t};\mathbf{r},\mathbf{\Omega})$ for a neutron interaction of type x (e.g., absorption, scattering, fission, and total) can be typically represented as follows:

$$\Sigma_x^g\left(\boldsymbol{\beta}_x^g;\mathbf{r},\mathbf{\Omega}\right) \triangleq \sum_{j=1}^{N_m} C_j^g a_j^g(\mathbf{r},\mathbf{\Omega})\ ,$$

$$C_j^g \triangleq \sum_{i=1}^{I} N_{x,j}^{g,i}\, \sigma_{x,j}^{g,i}\quad , \tag{229}$$

where for each energy group $g = 1, \ldots, G$, the various quantities are defined as follows:

$\sigma_x^{g,i}$ = imprecisely known microscopic cross section for the neutron interaction of type x, for isotope i, in group g

I = total number of distinct isotopes involved in the neutron interaction of type x

$N_{x,j}^{g,i}$ = imprecisely known isotopic atomic number density of isotope i in group g, for the neutron interaction of type x, in the j'th material contained in the heterogeneous medium under consideration

$a_j^g(\mathbf{r},\mathbf{\Omega})$ = spatial variation, in group g, characterizing the j'th material contained in the heterogeneous medium under consideration, while N_m is the total number of materials contained in this medium; boundary and interface perturbations are disregarded in this work, which means that $a_j^g(\mathbf{r},\mathbf{\Omega})$ is not subject to uncertainties, and where

$$\boldsymbol{\beta}_x^g \triangleq \Big[N_{x,1}^{g,i},\quad \sigma_{x,1}^{g,1}, \ldots,\quad N_{x,1}^{g,I},\ \sigma_{x,1}^{g,I}; N_{x,2}^{g,i},\ \sigma_{x,2}^{g,1}, \ldots,\ N_{x,2}^{g,I}, \sigma_{x,2}^{g,I}, \ldots,\ N_{N_m}^{g,1},\ \sigma_{x,N_m}^{g,1}, \ldots,\ N_{x,N_m}^{g,I},\ \sigma_{x,N_m}^{g,I}\Big]^{\dagger}$$

denotes the vector of imprecisely known scalar-valued model parameters for the neutron interaction of type x, in group g, for all isotopes and all materials involved in the definition of $\Sigma_x^g(\boldsymbol{\beta}_x;\mathbf{r},\mathbf{\Omega})$.

The partial first-order derivatives of $\Sigma_x^g\left(\boldsymbol{\beta}_x^g;\mathbf{r},\mathbf{\Omega}\right)$ can be readily obtained from Eq. (229) as follows:

$$\frac{\partial \Sigma_x^g\left(\boldsymbol{\beta}_x^g;\mathbf{r},\mathbf{\Omega}\right)}{\partial N_{x,j}^{g,i}} = \sigma_{x,j}^{g,i} a_j^g(\mathbf{r},\mathbf{\Omega}),\ \ i = 1, \ldots, I;\quad j = 1, \ldots,\ N_m \tag{230}$$

and

$$\frac{\partial \Sigma_x^g\left(\boldsymbol{\beta}_x^g;\mathbf{r},\mathbf{\Omega}\right)}{\partial \sigma_{x,j}^{g,i}} = N_{x,j}^{g,i} a_j^g(\mathbf{r},\mathbf{\Omega}),\ \ i = 1, \ldots, I;\quad j = 1, \ldots,\ N_m\ . \tag{231}$$

The nonzero partial second-order derivatives of $\Sigma_x^g\left(\boldsymbol{\beta}_x^g;\mathbf{r},\mathbf{\Omega}\right)$ can be readily obtained from Eqs. (230) and (231) as follows:

$$\frac{\partial^2 \Sigma_x^g\left(\boldsymbol{\beta}_x^g;\mathbf{r},\mathbf{\Omega}\right)}{\left(\partial N_{x,j}^{g,i}\right)\left(\partial \sigma_{x,j}^{g,i}\right)} = a_j^g(\mathbf{r},\mathbf{\Omega}),\ \ i = 1, \ldots, I;\quad j = 1, \ldots,\ N_m\ . \tag{232}$$

The macroscopic group absorption cross section can typically be represented in the form given in Eq. (229). Often, the macroscopic group fission cross section $\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]$ can also be represented in the form given in Eq. (229), namely,

$$\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right] \triangleq \sum_{j=1}^{N_m} \sum_{i=1}^{I_f} N_{f,j}^{g,i}\, (\nu\sigma)_{f,j}^{g,i} b_j^g(\mathbf{r})\ , \tag{233}$$

where I_f denotes the total number of fissionable isotopes in energy group g and in the j'th material contained in the heterogeneous medium under consideration. The partial first- and second-order derivatives of $\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]$ can be readily obtained from Eq. (233) as follows:

$$\frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial N_{f,j}^{g,i}} = (\nu\sigma)_{f,j}^{g,i} b_j^g(\mathbf{r}),\ \ i = 1, \ldots, I_f;\quad j = 1, \ldots,\ N_m\ ; \tag{234}$$

$$\frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\partial(\nu\sigma)^{g,i}_{f,j}} = N^{g,i}_{f,j} b^g_j(\mathbf{r},\mathbf{\Omega}),$$
$$i = 1, ..., I_f;\ j = 1, ..., N_m\ ; \quad (235)$$

and

$$\frac{\partial^2\left[(\nu\Sigma_f)^g(\mathbf{f};\mathbf{r})\right]}{\left(\partial N^{g,i}_{f,j}\right)\left(\partial\sigma^{g,i}_{f,j}\right)} = b^g_j(\mathbf{r},\mathbf{\Omega}), i = 1, ..., I_f;$$
$$j = 1, ..., N_m\ . \quad (236)$$

When the number of neutrons per fission ν and the respective microscopic fission cross section σ_f are provided separately, with accompanying uncertainties (e.g., standard deviations), then the derivatives provided in Eqs. (234) through (236) are to be expanded accordingly using the customary chain derivation.

For the j'th material, and energy group $g = 1, ..., G$, the material scattering cross section of order l can also be written in the form shown in Eq. (229), namely,

$$\Sigma^{g'\to g}_{s,l}(\mathbf{r}) = \sum_{j=1}^{N_m}\sum_{i=1}^{I_s} N^g_{s,i}\,\sigma^{g'\to g}_{s,l,i}\,c^g_j(\mathbf{r})\ , \quad (237)$$

where I_s denotes the total number of scattering isotopes. Often, the group source $Q^g(\mathbf{q};\mathbf{r},\mathbf{\Omega})$ and $\Sigma^g_d(\mathbf{d};\mathbf{r},\mathbf{\Omega})$ can also be represented in the form shown in Eq. (229).

VI. CONCLUSIONS

The following conclusions can be drawn based on the results that have been presented in this work:

1. As is well-known,[18,19] a single 1st-LASS needs to be solved in order to compute all first-order response sensitivities to all N_α model parameters.

2. For each model parameter, a single 2nd-LASS needs to be solved for computing the corresponding mixed second-order sensitivities. Hence, computing all of the $N_\alpha(N_\alpha+1)/2$ second-order sensitivities could theoretically require solving at most N_α 2nd-LASSs. In practice, however, the number of computations is much less, as has been shown in Refs. 11 and 13 through 17. In particular, the results in Ref. 17 show that only 12 large-scale adjoint particle transport computations were required by using the 2nd-ASAM to compute all of the detector's response to the flux of uncollided particles for the 18 first-order sensitivities and 224 second-order sensitivities, in contrast to the 877 large-scale forward particle transport calculations needed to compute the respective sensitivities using central finite differences, and this number did not include the additional calculations that were required to find appropriate values of the perturbations to use for the central differences.

3. The solution of each of the 2nd-LASSs is a two-component vector-valued second-level adjoint function, except for the 2nd-LASS that corresponds to model parameters that appear linearly in the response under consideration, in which case the vector-valued second-level adjoint function may have a null component.

4. Solving each of the 2nd-LASSs involves the inversion of the same operators as need to be inverted for solving the original transport equation and/or the 1st-LASS. Only the various source terms on the right sides of the 2nd-LASSs may differ from each other. Therefore, the same software can be used to solve both the 1st-LASS and the 2nd-LASS.

5. The computation of the second-order sensitivities involves the evaluations of integrals of the same form as those needed for computing the first-order sensitivities. Therefore, the same software can be used for computing both the first-order and the second-order sensitivities.

6. Each of the mixed second-order sensitivities is computed twice, using two distinct second-level adjoint functions. Consequently the 2nd-ASAM possesses an inherent solution verification mechanism that enables and ensures the accuracy verification of the solutions of all of the 2nd-LASSs.

7. For the reaction rate (detector) response considered in this work, it may be advantageous to compute the second-order sensitivities in the following order of increasing computational demands:

a. computation of $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\partial d_j\,\partial\alpha_{m_2}$, $j = 1, ..., J_d$, $m_2 = 1, ..., J_\alpha$
b. computation of $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\partial q_j\,\partial\alpha_{m_2}$, $j = 1, ..., J_q$; $m_2 = 1, ..., J_\alpha$
c. computation of $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\left(\partial t_j\right)\left(\partial\alpha_{m_2}\right)$, $j = 1, ..., J_t$; $m_2 = 1, ..., J_\alpha$
d. computation of $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\partial s_j\,\partial\alpha_{m_2}$, $j = 1, ..., J_s$; $m_2 = 1, ..., J_\alpha$
e. computation of $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\partial f_j\,\partial\alpha_{m_2}$, $j = 1, ..., J_f$; $m_2 = 1, ..., J_\alpha$
f. computation of $\partial^2 R\left(\boldsymbol{\alpha},\varphi;\ \psi^{(1)}\right)/\partial p_j\,\partial\alpha_{m_2}$, $j = 1, ..., J_p$; $m_2 = 1, ..., J_\alpha$.

Acknowledgments

The author gratefully acknowledges the support of, thorough review by, and constructive discussions with Jeffrey A. Favorite, of Los Alamos National Laboratory, which has provided funding for his work.

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