Nonperturbative retrieval of the scattering strength in one-dimensional media

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We examine several approaches on how to use the transmission and reflection amplitudes as functions of the modulation frequency of the laser's intensity to reconstruct the position-dependent scattering coefficient for a simple turbid medium. We explore the region where the contrast between the coefficient and its spatially averaged value is large enough such that perturbative methods fail. We show that in the case of a transillumination geometry, the knowledge of the transmission profile alone is not sufficient for unique image reconstruction, whereas the reflection spectrum allows for a complete inversion. We demonstrate the invertibility for media sampled at only a few positions.

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I. INTRODUCTION

The interaction of electromagnetic radiation with highly scattering media has been studied for several decades, encompassing a wide variety of fundamental and applied research areas including medical therapy, radar transmission and reception, astronomy, semiconductor technology, and photonics. While some applications make use of the coherence property of electromagnetic waves and their carrier frequency, others exploit the intensity of the radiation and its incoherent interaction with turbid media. Depending on the type of interaction, the prediction of the propagation properties is either based on solutions to the Maxwell theory, the Boltzmann equation, or the diffusion approximation [1,2].

In the forward problem one assumes complete knowledge of all optical parameters that characterize the highly scattering medium, denoted here by the set of parameters or functions $\{\mu\}$. The goal is to predict the properties of the scattered light, denoted by $\{S\}$. In other words, one tries to compute a function or functional F, which maps the first set of numbers into the second one, $\{S\}=F[\{\mu\}]$. In most situations, it is difficult to calculate the nonlinear functional relationship $F[\ldots]$; in fact, only the diffusion approximation permits analytical formulas, whereas the solutions of the Maxwell equations require typically numerical techniques [3,4], and the solutions of the Boltzmann equation require Monte Carlo simulations [5] based on random processes.

In the inverse problem, the general goal is to analyze the properties of the scattered light $\{S\}$ and to deduce the original scattering parameters characterizing the medium, $\{\mu\}=F^{-1}[\{S\}]$. There is a wide range of literature contesting to the intrinsic difficulties with regard to the uniqueness and the existence of this inverse function. Problems are often ill defined, or fluctuations in the input parameters $\{S\}$ for such a theory can have a drastic effect on the form of $F^{-1}[\ ]$. Inversion problems [6–9] have been studied mainly in the context of electromagnetic wave scattering of inhomogeneous dielectric media or acoustic scattering [10]. For instance, several works have explored the possibility of reconstructing the permittivity profiles for layered dielectric materials from the frequency dependency of the reflected light using iterative nonlinear optimization schemes to the Ricatti partial differential equation [11]. In this case, the scattering is induced exclusively by an index of refraction mismatch at the interfaces.

In the area of bio-optical imaging of turbid media such as tissues, a great deal of progress has been reported in the last decade [12]. Here nearly all studies focused on the diffusion approximation as the forward model and used either perturbative or optimization methods [13–15] to reconstruct the spatial dependence of the scattering coefficient, $\mu(x)$, from the dependence of the scattered light on the location of the set of detectors, $S(x)$.

Recently we proposed another controllable parameter in the input light to reconstruct $\mu(x)$ [16,17]. We showed for a simplified one-dimensional model system that the modulation frequency ω for the intensity of the laser beam can be used as a new degree of freedom for imaging. There has been a wide variety of experimental studies reported that exploit intensity-modulated light [18–22], but typically only for a single or a few modulation frequencies. The use of intensity-modulated radiation to probe the optical scattering properties is not new in bio-optical imaging. In fact, the prediction under the diffusion approximation under oscillating source terms has been studied in many areas of science [21]. In 1990 Gratton and co-workers [18] used photon density waves that were modulated in the megahertz to gigahertz region and measured the resulting shift in phase and attenuation of the ac component of the signal after propagation to predict the reduced scattering and absorption coefficients [23]. Several other approaches have used a wider set of frequencies to improve the accuracy of such a prediction [24–28]. In other studies it was shown that the photon density waves decay exponentially but have all the properties associated with typical wave forms, such as interference [29,30], refraction [31], and diffraction [32]. In none of these approaches was the continuity of measurements in the modulation frequency necessary. To the best of our knowledge, the resulting theoretical analysis used the experimental data for one modulation frequency at a time, and the measurements at different frequency values were used only to complement each other or to improve the accuracy by providing independent data.

In our analysis below we will show that derivatives of various scattering coefficients with regard to the modulation frequency permit analytical inversion formulas. In order to obtain these derivatives a continuous range of frequencies must be measured. This approach based on continuous frequency scans is similar in spirit to the work by McGill *et al.* [17], where the Fourier transform over all modulation frequencies was required to obtain inversion.

The particular model used in the study [16] restricts the light to only the forward and backward directions but displays many of the difficulties and characteristics of a three-dimensional system. It has been used to study many fundamental questions about light scattering in random media [33–37]. To demonstrate the feasibility of the modulation frequency scans, we assumed in [17] that the position-dependent scattering coefficient differs only slightly from its constant background value $\mu(x)=\mu+\delta\mu(x)$. Here $\delta\mu(x)$ represents the contrast in scattering strength associated with an object hidden inside the turbid medium. The corresponding Boltzmann equation can then be solved perturbatively to first order in the variation $\delta\mu(x)/\mu$. This approximated forward problem permitted a fully analytical solution $S(\omega)=F[\delta\mu(x)]$, which then could be inverted exactly, leading to $\delta\mu(x)=F^{-1}[S(\omega)]$. The functional form of $F^{-1}[\;]$ involved an integral transformation, and numerical examples showed the limitations of this method. Physically, the assumption about a small variation $\delta\mu(x)$ restricted the interaction due to the object to only single scattering events.

The key question that we would like to address in this note is rather fundamental. Is it at all possible to obtain a unique image of an object embedded in a turbid medium if it permits multiple scattering? Mathematically, the challenge would be to find an example of an inversion scheme that would not rely on perturbative approximations to the forward problem. Unfortunately, most known imaging schemes are based on these perturbative expansions of the diffusion approximation and therefore limit the object's response to the laser field to only single scattering. This is related to echo techniques, where the object to be detected can scatter the light only once. The question arises whether the linearization is just an advantageous mathematical method to obtain solutions or if it is actually a physical requirement for uniqueness.

In order to address this fundamental question, we use again the simplicity of the one-dimensional scattering system discussed in [16,17]. In this case, the measurable quantities are the complex reflection and transmission amplitudes $R(\omega)$ and $T(\omega)$ as a function of the modulation frequency ω of the intensity of the input beam. Is it possible to uniquely reconstruct the position-dependent scattering coefficient $\mu(x)$ from $R(\omega)$ and $T(\omega)$? If only the reflected light spectrum $R(\omega)$ is available for measurement, such as in the case of a semi-infinite medium, is a unique determination of a hidden object possible at all? Or similarly, could a transillumination geometry and the corresponding information contained in the transmission spectrum $T(\omega)$ provide sufficient information to reconstruct the medium?

Despite the relatively simple nature of this reduced dimensional system, this problem is quite challenging from a mathematical point of view, and a complete and general solution $F^{-1}[\;]$ would involve inverting an infinite set of coupled transcendental equations. Even if a complete analytical solution cannot be given at the moment, it would be desirable to find at least algorithmic solutions that could be implemented by a computer. In this work, we will report a first step toward the latter goal by examining multislab configurations with piecewise constant scattering coefficients.

The work is organized as follows. In Sec. II we review the mathematical formulation of the problem. In order to get a first insight into the feasibility of an exact inversion, we study in Sec. III a situation in which the scattering profile is sampled at only two locations, corresponding to a medium consisting of two turbid slabs. We will then show the mathematical complexity that has to be overcome to generalize this method to finer spatial sampling rates of the medium. We complete this work with an outlook on future work.

II. THE ONE-DIMENSIONAL MODEL SYSTEM

The interaction of the intensity-modulated laser field with a heterogeneous medium is described macroscopically by the Boltzmann equation. If we restrict the propagation direction to only one dimension, the resulting equations for the signal irradiance (intensity) $I(x,\pm,\omega)$ along the positive and negative x directions are

$$(-i\omega+\partial/\partial x)I(x,+,\omega)=-[\mu_s(x)+\mu_a(x)]I(x,+,\omega)+\mu_s(x)I(x,-,\omega) \tag{2.1a}$$

$$(-i\omega-\partial/\partial x)I(x,-,\omega)=\mu_s(x)I(x,+,\omega)-[\mu_s(x)+\mu_a(x)]I(x,-,\omega). \tag{2.1b}$$

The parameter ω is the modulation frequency divided by the speed of light in the host medium.

In vector notation, $\vec{\mathrm{I}}(x,\omega)\equiv\{I(x,+,\omega),I(x,-,\omega)\}$, we can rewrite this in a more compact form, $\partial/\partial x\vec{\mathrm{I}}(x,\omega)=\{[i\omega-\mu_a(x)]\boldsymbol{\sigma}+\mu_s(x)\mathbf{N}\}\;\vec{\mathrm{I}}(x,\omega)$, where we have introduced the diagonal Pauli matrix, $\boldsymbol{\sigma}\equiv\{\{1,0\},\{0,-1\}\}$ and the nilpotent matrix $\mathbf{N}\equiv\{\{-1,1\},\{-1,1\}\}$. As a result the generator takes the form $\mathbf{G}(x)\equiv[i\omega-\mu_a(x)]\boldsymbol{\sigma}+\mu_s(x)\mathbf{N}$. For a medium of total width W, we obtain the propagator solution $P\{\exp[\int_0^W dx\mathbf{G}(x)]\}$, where $P\{\;\}$ denotes the position ordering operator defined as $P\{f(x_1)g(x_2)\}\equiv g(x_2)f(x_1)\theta(x_2-x_1)+f(x_1)g(x_2)\theta(x_1-x_2)$ and where $\theta(\dots)$ denotes the Heaviside unit-step function. The operator P is the spatial analog of the time-ordering operator frequently used in the solution of nonautonomous equations of motion [38].

The propagator for the forward problem can be expressed as an infinite number of products of operators. If we divide the medium into $M\equiv W/\Delta x$ equal intervals, and approximate the corresponding scattering and absorption coefficients within each interval by constants, we can then apply the position-ordering operator to the product and obtain

$$\mathbf{K}(\omega) \equiv P\left\{\exp\left[\int_0^W dx\mathbf{G}(x)\right]\right\} = \exp[\mathbf{G}(x_M)\Delta x]\ldots\exp[\mathbf{G}(x_2)\Delta x]\exp[\mathbf{G}(x_1)\Delta x], \quad (2.2)$$

where the center positions are defined as $x_m \equiv (m-0.5)\Delta x$, for $m=1,2,\ldots M$. The four elements of each space-evolution (transfer) matrix can be obtained by diagonalizing the matrix $\mathbf{G}$ and corresponding exponentiation of the diagonal form, leading to

$$\{\exp[\mathbf{G}(x_m)\Delta x]\}_{1,1} = \cosh[\kappa(x_m)\Delta x] + [i\omega - \mu_s(x_m) - \mu_a(x_m)]\sinh[\kappa(x_m)\Delta x]/\kappa(x_m) \quad (2.3a)$$

$$\{\exp[\mathbf{G}(x_m)\Delta x]\}_{1,2} = -\{\exp[\mathbf{G}(x_m)\Delta x]\}_{2,1} = \mu_s(x_m)\sinh[\kappa(x_m)\Delta x]/\kappa(x_m) \quad (2.3b)$$

$$\{\exp[\mathbf{G}(x_m)\Delta x]\}_{2,2} = \cosh[\kappa(x_m)\Delta x] - [i\omega - \mu_s(x_m) - \mu_a(x_m)]\sinh[\kappa(x_m)\Delta x]/\kappa(x_m), \quad (2.3c)$$

where $\kappa(x) \equiv \{[i\omega - \mu_a(x) - \mu_s(x)]^2 - \mu_s(x)^2\}^{1/2}$.

We should point out that in contrast to a spatially dependent scattering parameter μ_s, for a single slab with (constant) coefficient μ_s, the corresponding transfer matrix can be expressed entirely in terms of the transmission and reflection amplitudes, T and R, with respect to light injected from the left side of the medium. Defining the right and left traveling amplitudes at the edges of the medium as $\vec{\mathrm{I}}(x=0,\omega) \equiv \{1,R\}$ and $\vec{\mathrm{I}}(x=W,\omega) \equiv \{T,0\}$, we use the propagator solution to solve for R and T. For a single turbid slab of width W we obtain

$$R(\omega) = -2\mu_s WS(\omega)/[-2\mu_s WS(\omega) - 2[C(\omega) - (i\omega - \mu_a)WS(\omega)]] \quad (2.4a)$$

$$T(\omega) = -2\mu_s WS(\omega)/[-2\mu_s WS(\omega) - 2[C(\omega) - (i\omega - \mu_a)WS(\omega)]], \quad (2.4b)$$

where

$$C(\omega) \equiv \cosh[[(i\omega - \mu_a - \mu_s)^2 - \mu_s^2]^{1/2}W] \quad (2.4c)$$

$$S(\omega) \equiv \sinh[[(i\omega - \mu_a - \mu_s)^2 - \mu_s^2]^{1/2}W]/[i(\omega - \mu_a - \mu_s)^2 - \mu_s^2]^{1/2}. \quad (2.4d)$$

As we assume complete knowledge of the scattered light spectrum, $T(\omega)$ and $R(\omega)$, we also know the precise form of the total transfer matrix, $\mathbf{K}(\omega) \equiv \{\{T-R^2/T, R/T\}, \{-R/T, 1/T\}\}$. We note that this matrix has the determinant equal to unity, but the matrix is not unitary as its two eigenvalues are not unimodular.

Let us now return to the composite system. For a medium with a spatially dependent scattering coefficient (the composite system), however, the knowledge about the total reflection and transmission amplitudes alone is not sufficient to determine all four matrix elements of the total transfer matrix $\mathbf{K}(\omega)$. The determinant of $\mathbf{K}(\omega)$ is 1, as the determinant of each product matrix in Eq. (2.2) is unity. If we solve $\{T,0\} = \mathbf{K}(\omega)\{1,R\}$ together with the determinant condition, we obtain only the general form $\mathbf{K}(\omega) = \{\{\xi, (T-\xi)/R\}, \{-R/T, 1/T\}\}$, where ξ is unspecified. The full determination of $\mathbf{K}(\omega)$ also requires the measurement of the reflection and transmission amplitudes for light that has been injected into the medium from the other (right) end of the medium, leading to $R^t(\omega)$ and $T^t(\omega)$. It should be clear that these two amplitudes are the same as obtained from light injected from the left side, but for a transposed medium, denoted as one in which the sequence of scattering coefficients has been reversed. In other words we obtain

$$R^t(\omega, \mu_{s1}, \mu_{s2}, \ldots, \mu_{sM}; \mu_{a1}, \mu_{a2}, \ldots, \mu_{aM}) = R(\omega, \mu_{sM}, \ldots, \mu_{s2}, \mu_{s1}; \mu_{aM}, \ldots, \mu_{a2}, \mu_{a1})$$

and

$$T^t(\omega, \mu_{s1}, \mu_{s2}, \ldots, \mu_{sM}; \mu_{a1}, \mu_{a2}, \ldots, \mu_{aM}) = T(\omega, \mu_{sM}, \ldots, \mu_{s2}, \mu_{s1}; \mu_{aM}, \ldots, \mu_{a2}, \mu_{a1}).$$

In contrast to the propagation of monochromatic electromagnetic radiation through dielectric media where $R^t(\omega)$ and $R(\omega)$ are directly related to each other [39], in the case of turbid media the two reflection amplitudes can be quite different. Using the measured data, we therefore can construct the entire propagator matrix using its definitions $\{T,0\} = \mathbf{K}(\omega)\{1,R\}$ and $\{R^t,1\} = \mathbf{K}(\omega)\{0,T^t\}$, from which we obtain $\mathbf{K}(\omega) = \{\{T^t - R(R^t/T^t), R^t/T^t\}, \{-R/T, 1/T^t\}\}$. As the determinant of the matrix $\mathbf{K}(\omega)$ has to be unity, it follows immediately that the transmission coefficients for the transposed and the original medium are identical, $T=T^t$ for all frequencies ω, simplifying the matrix to $\mathbf{K}(\omega) = \{\{T - R(R^t/T), R^t/T\}, \{-R/T, 1/T\}\}$. This reduces our original problem to solving this matrix equation:

$$\{[T - R(R^t/T), R^t/T], [-R/T, 1/T]\} = \exp\{[(i\omega - \mu_{aM})\sigma + \mu_{s,M}\mathbf{N}]\Delta x\}\ldots \times\exp\{[(i\omega - \mu_{a,2})\sigma + \mu_{s,2}\mathbf{N}]\Delta x\} \times\exp\{[(i\omega - \mu_{a,1})\sigma + \mu_{s,1}\mathbf{N}]\Delta x\}. \quad (2.5)$$

In summary, the goal would be to use the complete knowledge of the numerical values of the three complex values T, R, and R^t for any value of ω to reconstruct the set of M scattering coefficients $\mu_{s,1}, \mu_{s,2}, \ldots \mu_{s,M}$, and M absorption coefficients $\mu_{a,1}, \mu_{a,2}, \ldots \mu_{a,M}$. Unfortunately, we have not been able to find a generally applicable scheme to solve these infinitely many coupled transcendental equations, and we have to exploit specific properties of the functional structure of these equations to find at least solutions for simplified situations, such as those where the entire medium is described by a smaller number of coefficients. We will discuss one possible approach in the next section and explore whether the knowledge of $R(\omega)$, $T(\omega)$, and $R^{t}(\omega)$, or any combination, is suitable for reconstructing the scattering coefficients.

We should note that the discretized medium is equivalent to a layered scattering medium in which each layer has a different but constant scattering strength μ_i. As the entire medium is characterized by the same host medium, it is described by a single index of refraction. As a consequence, the different layers with different scattering strengths are perfectly index-matched and there are no additional surface reflections present that would require the Fresnel coefficients. For works on nonscattering layered dielectric media that differ by their index of refraction, see [11].

III. A SOLUTION ALGORITHM FOR THE TWO-SLAB SYSTEM

From now on let us assume that $\mu_a(x)=0$, and define $\mu_{s,m}\equiv\mu_m$ for $m=1,2,\ldots,M$ and $\Delta x=W/M$. We should note that a simultaneous determination of $\mu_a(x)$ *and* $\mu_s(x)$ is very difficult. For the special case of $\mu_s(x)=0$, it is not even in principle possible to reconstruct $\mu_a(x)$ from the transmitted light, as the solution depends only on total space-integrated absorption $\int dx'\mu(x')$ for every modulation frequency ω. The next case would evolve two nonzero, but constant absorption μ_a and a position-dependent scattering $\mu_s(x)$. The feasibility of the various methods outlined below to retrieve $\mu_s(x)$ relies on the fact that the relevant transcendental equations and their ω derivatives at $\omega=0$ reduce to algebraic equations in μ_m; this property, however, is no longer true for nonzero absorption. Our assumption that the spatial axis is discretized in equidistant steps leading to slabs with identical width $\Delta x=W/M$ is a nonrestriction assumption, as all formulas derived below can be easily extended to include slabs of nonidentical width. The drawback would be that the formulas are just more complicated. We assume, however, that we know *a priori* the total length W of the medium.

The two-slab system is an ideal system to explore our very fundamental questions about invertibility. For example, it is simple enough such that three approaches toward inversion can be evaluated and analytical formulas can be given in each case. The particular form of the solution can then be analyzed in each situation with regard to possible generalizations to systems that are sampled at more than just two points. Using the two-slab system as an example, we will show that depending on the type of measurement as well as the direction from which light is injected into the medium, various inversion schemes can be derived. We explore the corresponding functional relationships between the measured data and the scattering coefficients for these simpler systems with the hope to discover possible regularities of these schemes, which can then be generalized to media with an arbitrary number of slabs.

For a single slab with an unknown single parameter μ, the transmission and reflection coefficient takes the form of Eqs. (2.4). Due to the complicated dependence of the coefficient μ involving products of μ with hyperbolic functions, even for a system consisting of a single slab $(M=1)$, it is not possible to find a closed form solution expression for μ as a function of the measured quantities $T(\omega)$ and $R(\omega)$ for nonzero values of the frequency ω. However, for $\omega=0$ both solutions simplify significantly to $R(0)=\mu_1 W/[1+\mu_1 W]$ and $T(0)=1/[1+\mu_1 W]$, both of which can be easily inverted to obtain $\mu_1=[R/(1-R)]/W=[(1-T)/T]/W$. From now on we define $R\equiv R(\omega=0)$ and $T\equiv T(\omega=0)$.

For the two-slab system characterized by two scattering coefficients μ_1 and μ_2, we have to distinguish between the two possible directions of incoming light. Let us first assume we measure exclusively the transmission spectrum $T(\omega)$. Using the expressions for the total transmission coefficient and its derivate at $\omega=0$, denoted by $\partial_\omega T$, we obtain the following two solutions:

$$\mu_1=\{W(1-T)-[2W(i3\partial_\omega T+(1+T+T^2)W)]^{1/2}\}/[TW^2] \tag{3.1a}$$

$$\mu_2=\{W(1-T)+[2W(i3\partial_\omega T+(1+T+T^2)W)]^{1/2}\}/[TW^2], \tag{3.1b}$$

and a second set of solutions for which μ_1 and μ_2 are exchanged. This ambivalence is not surprising, as we saw above that the transposed and the original medium have the same transmission amplitude for all frequencies; thus indicating that an unambiguous determination of μ_1 and μ_2 from the transmission profile only is not possible. This can also be seen from a physical point of view. Any particular scattering path of the light through the two slabs that exits the medium on the other side is characterized by a transit time and a probability. It turns out that each particular path has a second (equivalent) one that differs only by the sequence of locations where the path gets reversed. We also remark that two different scattering systems, A and B, whose coefficients are related to each other by a simple translation, $\mu_A(x)=\mu_B(x+L)$, also lead to identical transmission amplitudes. This finding can be immediately generalized to any multislab system as the transmission profile is always identical to that of its transposed system $T=T^{t}$. Therefore a unique imaging based on the transmission spectrum only is not possible.

Let us next assume we measure only the reflection amplitude $R(\omega)$ for the two-slab system. In this case the resulting algebraic equations for R and $\partial_\omega R$ can be inverted, and we obtain for our two scattering coefficients μ_1 and μ_2,

$$\mu_1=[-(2R+3)W^{1/2}+\{-i24\partial_\omega R+(9-24R+8R^2)W\}^{1/2}]/[2(R-1)W^{3/2}] \tag{3.2a}$$

$$\mu_2=[-(2R-3)W^{1/2}-\{-i24\partial_\omega R+(9-24R+8R^2)W\}^{1/2}]/[2(R-1)W^{3/2}]. \tag{3.2b}$$

We should point out that (similar to the equation for $\partial_\omega T$) the equation for $\partial_\omega R$ is also quadratic in μ_1 and μ_2, but because of the requirement of positivity, the second pair of mathematical solutions (not given here) for (μ_1,μ_2) can be discarded as unphysical. This shows that in contrast to the transillumination geometry, an inversion solely based on the knowledge of the reflection spectrum, $R(\omega)$, permits an unambiguous recovery of the scattering coefficients for the two-slab system. However, the inversion formula (3.2) is complicated and a reliable scheme to generalize this to a multislab medium is nontrivial.

Another inversion formula based on R and T can be derived if we equate the (2,1) and the (2,2) elements of the propagator matrix at $\omega=0$ and the first derivatives with the experimentally determined one. Two of these four equations is quadratic in μ_1 and μ_2, but by adding the two equations appropriately the quadratic terms cancel out and the two unphysical solutions can be eliminated, leading to the following inversion formula:

$$\mu_1 = [W(2+3R/T) + i2\partial_\omega[(R-1)/T]]/W^2 \qquad (3.3a)$$

$$\mu_2 = [-W(2+R/T) - i2\partial_\omega[(R-1)/T]]/W^2. \qquad (3.3b)$$

In contrast to the inversion formulas Eqs. (3.1) and (3.2), which contain complicated coefficients and square roots, the formula (3.3) has a simpler structure and suggests that if $R(\omega)$ and $T(\omega)$ are measured, possibly a generalization to a multislab could be accomplished.

IV. GENERALIZATION TO MULTISLAB SYSTEMS

The general approach to inversion is twofold. First we have to find an appropriate set of equations that relate the measured transmission and reflection amplitude for the composite system to the individual scattering coefficients for each subslab. The challenge would be to identify certain regularities or schemes that permit us to generalize these equations to media with an arbitray number of slabs. The second challenge would be to find appropriate solution techniques for these coupled nonlinear equations. As we have seen in Sec. II, using just R or T as the basis leads to solutions that are very hard to generalize to multislabs.

A. $R+T$ approach

Due to the functional dependence on the frequency ω, it turns out that the quantity $R(\omega)+T(\omega)$ and its frequency derivatives provide a promising basis to obtain a sufficient number of equations from which the scattering coefficients can be determined. For zero frequency, we have $R+T=1$, and the first derivative evaluated at $\omega=0$, denoted by $\partial_\omega(R+T)$, can even be found analytically for an arbitrary number of slabs M,

$$\partial_\omega(R+T) = i(W/M)[M + (W/M)\Sigma_m(2m-1)\mu_m]/[1+(W/M)\Sigma_m\mu_m], \qquad (4.1)$$

where the summation Σ_m extends from $m=1$ to $m=M$. However, we have not been able to find similar formulas for higher derivatives. Let us briefly outline the analytical solutions obtained from this approach. For the two-slab system we obtain

$$\mu_1 = -[i2\partial_\omega(R+T) + (2+R)W]/[(R-1)W^2] \qquad (4.2a)$$

$$\mu_2 = [i2\partial_\omega(R+T) + (2-R)W]/[(R-1)W^2], \qquad (4.2b)$$

which is equivalent to the solution Eq. (3.3).

A unique determination of μ_1, μ_2, and μ_3 for a three-slab system requires at least three linearly independent equations. However, these equations are nonlinear and more than just three equations are necessary to uniquely determine the correct triplet of scattering coefficients. The derivation is extremely cumbersome and nearly impossible without symbolic manipulation software such as MATHEMATICA. The dependence of R, T, and higher derivatives of the sum of the two is nontrivial such that we omit these equations here and state only the final inversion formula,

$$\mu_1 = -D[9[\partial_\omega(R+T)]^2TW^2 + i2\partial_\omega(R+T)(4T-15)TW^3 - 17TW^4 + 8T^2W^4 + iE] \qquad (4.3a)$$

$$\mu_2 = 2D[i2\partial_\omega(R+T)(T-3)TW^3 - 2TW^4 + 2T^2W^4 + iE] \qquad (4.3b)$$

$$\mu_3 = -D[-9[\partial_\omega(R+T)]^2TW^2 + i2\partial_\omega(R+T)(4T+3)TW^3 + TW^4 + 8T^2W^4 + iE], \qquad (4.3c)$$

where D and E denote

$$D \equiv \{4T^2W^4[W + i\partial_\omega(R+T)]\}^{-1} \qquad (4.3d)$$

$$E \equiv [T^2W^4[135[\partial_\omega(R+T)]^4 - 432i[\partial_\omega(R+T)]^3W + 108[\partial_\omega^2(R+T)]TW^2 + 2[(\partial_\omega(R+T)\ ]^2(-195+24T + 44T^2)W^2 + (15-8T-88T^2)W^4 + i4[\partial_\omega(R+T)] \times\{27[\partial_\omega^2(R+T)]TW + (31-14T-44T^2)W^3\}]^{1/2}. \qquad (4.3e)$$

The complexity of this result certainly suggests that a generalization to more than three slabs is again nontrivial.

B. Q-Q^t approach

So far we have seen that an approach that is based on the knowledge of $R(\omega)$ *and* $T(\omega)$ leads to a formally easier inversion formula than those based on either $R(\omega)$ *or* $T(\omega)$. We will discuss now an approach that even relies on the full knowledge of the entire propagator matrix $\mathbf{K}(\omega)$. In other words, $R(\omega)$, $T(\omega)$ as well as $R^t(\omega)$ need to measured. As we discussed in Sec. II, this requires a scattering setup for which the input light is injected into the medium from both directions. We derived in Sec. II the form of the propagator matrix $\mathbf{K}(\omega)=\{\{T^t\text{-}RQ^t), Q^t\},\{-Q, 1/T^t\}\}$, where we denote the quotients $Q(\omega)\equiv R(\omega)/T(\omega)$ and the corresponding quotient of the transposed medium $Q^t(\omega)\equiv R^t(\omega)/T(\omega)$. We will now describe a scheme that uses these ratios to reconstruct the scattering coefficients. It is based again on the observation that the propagator $\mathbf{K}(\omega)$ becomes algebraic for the special case of an unmodulated laser field, $\omega=0$. It should be clear that the knowledge of only two matrix elements of $\mathbf{K}(\omega=0)$ does not provide sufficient information for a unique determination of all scattering coefficients. In the following we will examine whether a coupled system involving the derivatives of the matrix $\mathbf{K}(\omega=0)$ with regard to the frequency ω evaluated at $\omega=0$ can be used to provide the additional constraints necessary to uniquely determine the set of $\{\mu\}$:

$$\mathbf{K}(\omega=0)=\mathbf{1}+(\mu_M+\ \dots\ \mu_2+\mu_1)\mathbf{N}W/M \tag{4.4a}$$

$$\partial_\omega\mathbf{K}(\omega)=\partial_\omega\{\exp[(i\omega\sigma+\mu_M\mathbf{N})W/M]\times\exp[(i\omega\sigma+\mu_{M-1}\mathbf{N})W/M]\dots\times\exp[(i\omega\sigma+\mu_2\mathbf{N})W/M]\times\exp[(i\omega\sigma+\mu_1\mathbf{N})W/M]\} \tag{4.4b}$$

$$(\partial_\omega)^J\mathbf{K}(\omega)=(\partial_\omega)^J\{\exp[(i\omega\sigma+\mu_M\mathbf{N})W/M]\times\exp[(i\omega\sigma+\mu_{M-1}\mathbf{N})W/M]\dots\times\exp[(i\omega\sigma+\mu_2\mathbf{N})W/M]\times\exp[(i\omega\sigma+\mu_1\mathbf{N})W/M]\}. \tag{4.4c}$$

Here we define the (Jth) derivative operator as $(\partial_\omega)^J f(\omega)\equiv\partial^J/\partial\omega^J f(\omega)|_{\omega=0}$ evaluated at $\omega=0$. A general form for the derivative of each single factor, $(\partial_\omega)^J\exp[(i\omega\sigma+\mu_m\mathbf{N})\Delta x]\equiv(\partial_\omega)^J E_m$ is given in the Appendix. Using the formula for the derivative of each single subslab propagator matrix, we can then compute the derivatives of the (noncommuting) exponential product matrices,

$$(\partial_\omega)^J(E_M\dots E_2E_1)=\Sigma_\alpha[M!/(\alpha_1!\alpha_2!\,.\,.\,\alpha_M!)]\times(\partial_\omega)^{\alpha_M}E_M\dots(\partial_\omega)^{\alpha_2}E_2(\partial_\omega)^{\alpha_1}E_1, \tag{4.5a}$$

where the sum Σ_α extends over all non-negative values of the M parameters α_m, constrained only by $\alpha_1+\alpha_2\dots+\alpha_M=J$.

Let us illustrate this scheme for $M=1$, 2, and 3. For a single slab, $M=1$, Eq. (4.4a) is sufficient for the inversion, $\mathbf{K}(\omega)=1+\mu_1\mathbf{N}W$, leading to

$$\mu_1=Q. \tag{4.6}$$

From now on we will use notation $Q\equiv Q(\omega=0)$ and $(\partial_\omega)^J Q\equiv\partial^J/\partial\omega^J Q(\omega)|_{\omega=0}$. We note that the matrix $\mathbf{K}(\omega=0)$ is real, and we have used the conservation law that $R(\omega=0)+T(\omega=0)=1$. Correspondingly, in this case either $R(\omega=0)$ or $T(\omega=0)$ would be sufficient to uniquely determine the value for μ_1.

Let us illustrate how the scheme works for $M=2$. Using the formula from the Appendix we have the following two matrix equations:

$$\mathbf{K}(\omega=0)=\mathbf{1}+(\mu_2+\mu_1)W\mathbf{N}/2 \tag{4.7a}$$

$$\partial_\omega\mathbf{K}(\omega=0)=iW\sigma-iW^2(\mu_1+\mu_2)\mathbf{1}/4-i[\mu_1^2+\mu_2^2+6\mu_1\mu_2]W^3\mathbf{N}/24+iW^2\mu_1\sigma\mathbf{N}/4+iW^2\mu_2\mathbf{N}\sigma/4. \tag{4.7b}$$

As we need only the two solutions (μ_1,μ_2), not all of these eight coupled equations can be linearly independent of one another. In fact, each of the four equations in Eq. (4.7a) restricts only the sum, $(\mu_2+\mu_1)$, such that at least one more equation from the set Eq. (4.7b) needs to be taken into account. However, these equations are quadratic and therefore when solved by themselves, lead to an additional (unphysical) solution. This additional solution depends on which matrix element of $\partial_\omega\mathbf{K}(\omega=0)$ is used in addition to Eq. (4.7a). If we add the equations for $\partial_\omega\{\mathbf{K}(\omega 0)\}_{1,2}$, and $\{\partial_\omega\mathbf{K}(\omega=0)\}_{1,2}$ in a suitable way, the quadratic terms can be eliminated and the resulting set of equations becomes linear in μ_1 and μ_2, leading to the remarkable simple final solution

$$\mu_1=Q/W+(i/W^2)\partial_\omega(Q-Q^t) \tag{4.8a}$$

$$\mu_2=Q/W-(i/W^2)\partial_\omega(Q-Q^t), \tag{4.8b}$$

showing that also the two-slab system can be inverted exactly from the experimental data $\mathbf{K}(\omega)$.

To solve the three-slab inversion problem, we have to equate the (1,2) and (2,1) element of the measured $\mathbf{K}(\omega)$, and its first and second frequency derivative, $\partial_\omega\mathbf{K}$ and $\partial_\omega^2\mathbf{K}$, with the corresponding analytical expressions. This set of six coupled nonlinear equations is complicated but algebraic and can be solved for the triplet (μ_1,μ_2,μ_3), leading to

$$\mu_1=9[2Q\partial_\omega(Q-Q^t)+i[\partial_\omega(Q-Q^t)]^2/W-3\partial_\omega^2\partial_\omega^2\times(Q-Q^t)/W]/[8W\partial_\omega(Q-Q^t)] \tag{4.9a}$$

$$\mu_2=3[-2Q\partial_\omega(Q-Q^t)+9i\partial_\omega^2(Q-Q^t)/W]/[4W\partial_\omega(Q-Q^t)] \tag{4.9b}$$

$$\mu_3=9[2Q\partial_\omega(Q-Q^t)-i(\partial_\omega(Q-Q^t))^2/W-3i\partial_\omega^2(Q-Q^t)/W]/[8W\partial_\omega(Q-Q^t)]. \tag{4.9c}$$

Even though this sequence of solutions based on Q and Q^t given by Eqs. (4.6), (4.8), and (4.9) takes the most promising form with respect to a possible generalization to M slabs, the corresponding formula for the four-slab systems is extremely complicated. Here μ_1, μ_2, and μ_3 can be expressed as reasonable functions of μ_4. The analytical form of the latter, however, is too complicated to be reproduced here. We point out that the derivation required a division by the difference Q-Q^t, formula (4.9), has convergence problems for $Q=Q^t$, in other words for those symmetric media for which $\mu_1=\mu_3$.

C. Numerical examples for both approaches

The form of the inversion formulas presented here is a consequence of the specific functional ω dependence of the scattering amplitudes for a system with three slabs. As a numerical test, we have assumed specific values for (μ_1,μ_2,μ_3) and then computed the optical responses $R(\omega)$, $T(\omega)$, and $R^t(\omega)$ for various frequencies. We then used the symmetric three-point finite difference formulas to compute the first and second frequency derivatives, e.g., $\partial_\omega R\approx[R(\omega=\Delta)-R(\omega=-\Delta)]/(2\Delta)$, etc., and inserted the results into the inversion formulas to reconstruct the scattering coefficients. As this was just a consistency check, the scattering parameters were exactly reproduced by all formulas.

As real media, however, are typically described by a continuous position dependence $\mu=\mu(x)$, we can examine whether we can extend the range of applicability of the three-slab approximation-based inversion formulas numerically to

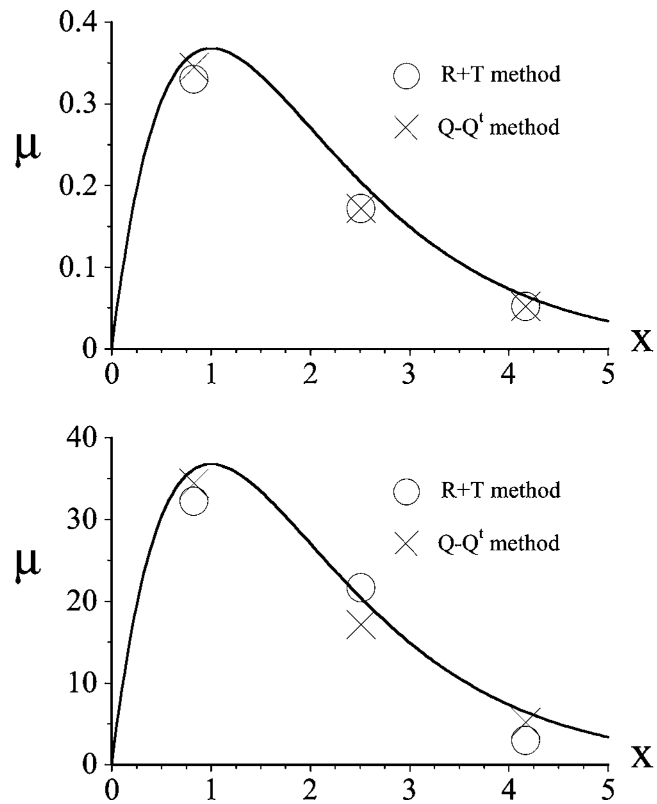

FIG. 1. Application of the three-slab based, nonperturbative imaging scheme to a medium with continuously changing scattering strength as a function of the position x (in arbitrary units) [(a) $\mu(x)=x\exp(-x)$, (b) $\mu(x)=100x\exp(-x)$]. The dots are the reconstructed results from the $(R+T)$ method outlined in Eqs. (4.3), whereas the crosses are the predictions according to the $(Q\text{-}Q^t)$ method by Eqs. (4.9). To obtain the propagator matrix $\mathbf{K}(\omega)$ for the medium it was sampled at ten points.

these systems. As the frequency dependence of a three-slab propagator can be functionally completely different than one obtained for a continuous medium, it is not clear *ab initio* whether our inversion formulas are applicable at all, as they could predict sets of unphysical negative or even complex scattering coefficients.

As our first test case let us examine a medium with small scattering coefficient where the diffusion approximation would be inapplicable. We assume the scattering coefficient is given by $\mu(x)=x\exp(-x)$ for $0<x<5$. In order to obtain the total propagation matrix with practically arbitrary precision, we can use the finite-product expansion Eq. (2.2) and the discretized form given by Eq. (2.3). This approach gives us the numerical values $T(\omega)$, $R(\omega)$, and $R^t(\omega)$ for our medium for any frequency ω. In Fig. 1(a) we display the original $\mu(x)$ together with the predictions of the $(R+T)$ method (dots) and the $(Q\text{-}Q^t)$ method (crosses). The reconstructed values (μ_1,μ_2,μ_3) turn out to be quite reasonable in this nondiffusive regime.

To stress the nonperturbative character of this approach to inversion, we have used another medium, $\mu(x)=100x\exp(-x)$. If we define an average scattering strength as $\langle\mu(x)\rangle=19$, the width of $W=5$ contains roughly 100 inverse scattering lengths, making this medium highly scattering. The data presented in Fig. 1(b) show again good agreement in this highly nonperturbative regime. Comparing the data in Figs. 1(a) and 1(b) suggests that despite the nonlinear character of the inversion formulas, the predicted triplet seems almost to be linear in the scattering strengths.

We should complete this section with an interesting comment about the nonlocal as well as nonlinear character of both inversion schemes. This is best demonstrated for a six-slab system with a given set of scattering coefficients $(\mu_a,\mu_b,\mu_c,\mu_d,\mu_e,\mu_f)$ and $W=6$. As expected for the particular set (5,5,4,4,3,3), both approaches reproduce the correct values $(\mu_1,\mu_2,\mu_3)=(5,4,3)$, as this particular six-slab matrix $\mathbf{K}(\omega)$ can be matched exactly by a $\mathbf{K}(\omega)$ for a three-slab matrix (with $\Delta x=2$ used as a basis for our inversion). Let us now repeat this inversion for (5,7,4,4,3,3), in which only the second slab was chosen different. One could expect that now only the first value μ_1 is affected by this change. However, the $(R+T)$ method predicts (5.4,4.7,2.7) and the $(Q\text{-}Q^t)$-method predicts (6.4,2.8,3.7). The fact that all three predicted scattering parameters are different shows that the mapping from $(\mu_a,\mu_b,\mu_c,\mu_d,\mu_e,\mu_f)$ to the triplet (μ_1,μ_2,μ_3) is not only nonlinear but also nonlocal, in other words, changes of the scattering strength μ_b at the entry region of the medium result in different values not only for μ_1, but for each of the three effective scattering parameters.

We also note that an increase from 5 to 7 in the second slab leads to an effective reduction from $\mu_3=3$ to $\mu_3=2.7$ in the third slab for the $(R+T)$ method, whereas the $(Q\text{-}Q^t)$ method predicts an increase from 3 to 3.7. As the $(Q\text{-}Q^t)$ method relies on the measurement of the scattered light based on both the injected light entering the medium from the left as well as from right side, the inversion results for the two media (5,7,4,4,3,3) and (3,3,4,4,7,5) are symmetric, (6.4,2.8,3.7) and (3.7,2.8,6.4), respectively. The $(R+T)$ method, however, is biased toward a single input direction (light coming in from the left side) and therefore predicts for the transposed medium an entirely different result, (2.9,4.3,5.7), compared to (5.4,4.7,2.7) for the original medium.

V. DISCUSSION AND OUTLOOK

The present work serves only as a proof of concept to demonstrate that—in principle—the frequency of the intensity modulation can be exploited as a controllable degree of freedom for imaging. We have used a simple one-dimensional system to illustrate the inherent difficulties to invert a system that is described by the Boltzmann equation. In this case even the solutions of the forward problem can be expressed only as a limit of an infinite number of products of single-slab propagator matrices. We have outlined several approaches, among which the most promising one, permitting a possible generalization to those systems that are sampled at arbitrary numbers of positions, was based on the difference of the ratios of the reflection and transmission coefficients of the original and the transposed system and required a measurement for injection of light from both directions into the medium. It turns out that the corresponding equations are transcendental, but for zero modulation frequency they become algebraic. In order to have a sufficient number of linearly independent equations to generate solutions for all the scattering coefficients of each subslab of the composite system, higher-order derivatives with respect to ω and evaluated at $\omega=0$ also had to be considered. Correspondingly, this particular approach cannot be used to identify which range for the modulation frequency is best suited for imaging with high resolution. We were able to demonstrate its applicability for a medium sampled at 1, 2, 3, and 4 positions. Many of the results reported here could only be obtained from symbolic manipulation software packages such as MATHEMATICA.

We point out that for nonzero absorption, however, this method does not convert the transcendental equations into algebraic ones and one needs to study other methods. For instance, one could try to obtain iterative schemes that are based on the reduction of the properties from an M-slab system to that of an (M-1) slab system. The formulas worked out in Sec. II could be applied as building blocks for those iterative schemes in which sequentially the leftmost subslab is obtained. As a first step in such an iterative scheme the exact matrix $\mathbf{K}(\omega)$ of the composite system could be approximated by a product of only two single-slab propagator matrices for which the equations derived in Sec. II provide the relationships to obtain these effective scattering coefficients. Then our formulas in Sec. II could be used to find the effective scattering coefficients for the leftmost subslab. The corresponding inverse of the propagator matrix of the leftmost slab could then be multiplied with the total propagator matrix, therefore reducing the problem to finding only the remaining (M-1) subslabs. The feasibility and convergence of these schemes is presently not clear, as the propagator matrix for a twoslab system has a functionally different dependence on the frequency ω as an effective single-slab system. We will report on the merits of such an approach elsewhere.

An important future challenge might also be to explore how results from the one-dimensional model system can be generalized to three-dimensional systems. For instance, the observation that a transmission-only based imaging cannot be unique generalizes immediately to media of more than one dimension. On the other hand, the present system is more than just a (nontrivial) mathematical exercise as even the reduced dimensional model system can be studied experimentally based on only plane parallel dielectric layers [40] such as multiple layers of thin films.

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APPENDIX

The derivatives of the exponential of a sum of two noncommuting operators, $\exp(\omega\mathbf{A}+\mathbf{B})$, can generally be given only as an infinite sum of nonexponential operators [41]. In our case, however, one of the two operators in the exponent is nilpotent, $\mathbf{B}\equiv\mu\mathbf{N}\Delta x$, and as a result, finite expansions of the derivatives can be given. To abbreviate our notation, we use $\mathbf{A}\equiv i\sigma\Delta x$. In order to examine the derivatives, let us first expand the exponential operator,

$$\begin{aligned}\mathbf{f}(\omega) &\equiv \exp(\omega\mathbf{A}+\mathbf{B})\\ &= \sum_{\mathrm{n}=0}^{\infty}(\omega\mathbf{A}+\mathbf{B})^{\mathrm{n}}/\mathrm{n}!\\ &= \mathbf{1}+(\omega\mathbf{A}+\mathbf{B})+(\omega\mathbf{A}+\mathbf{B})(\omega\mathbf{A}+\mathbf{B})/2!\\ &\quad+(\omega\mathbf{A}+\mathbf{B})(\omega\mathbf{A}+\mathbf{B})(\omega\mathbf{A}+\mathbf{B})/3!+\cdots . \end{aligned} \tag{A1}$$

With this expansion we obtain the derivatives for each term

$$\partial_\omega^{\mathrm{N}}\mathbf{f}(0)=\sum_{\mathrm{j}=0}^{\mathrm{N}+1}\wp(\mathbf{A}^{(\mathrm{N})},\mathbf{B}^{(\mathrm{j})})/(j+N)!. \tag{A2}$$

Note that the summation is finite and contains only $(N+2)$ terms. The function $\wp(\mathbf{A}^{(\mathrm{N})},\mathbf{B}^{(\mathrm{j})})$ represents the sum of those unique products, that can be obtained by arranging N operators $\mathbf{A}$ and j operators $\mathbf{B}$. For example, for $N=3$ and $j=1$ we obtain $\wp(\mathbf{A}^{(3)},\mathbf{B}^{(1)})=\mathbf{AAAB}+\mathbf{AABA}+\mathbf{ABAA}+\mathbf{BAAA}$. As $\mathbf{AA}=-\Delta x^2\mathbf{1}$ is proportional to the identity matrix, this expression can be simplified to $2\mathbf{A}^2(\mathbf{AB}+\mathbf{BA})$. Similarly, for $N=3$ and $j=2$ we obtain

$$\begin{aligned}\wp(\mathbf{A}^{(3)},\mathbf{B}^{(2)}) = (&\mathbf{AAABB}+\mathbf{AABAB}+\mathbf{AABBA}+\mathbf{ABABA}\\ &+\mathbf{BAABA}+\mathbf{ABAAB}+\mathbf{ABBAA}+\mathbf{BABAA}\\ &+\mathbf{BAAAB}+\mathbf{BBAAA}).\end{aligned}$$

Also, this expression simplifies significantly as $\mathbf{B}^2=\mathbf{0}$, and we obtain $\mathbf{AAABB}=\mathbf{AABBA}=\mathbf{BAABA}=\mathbf{ABAAB}=\mathbf{ABBAA}=\mathbf{BBAAA}=\mathbf{0}$. As a consequence, $\wp(\mathbf{A}^{(3)},\mathbf{B}^{(2)})$ reduces to $3\mathbf{A}^2\mathbf{BAB}+\mathbf{ABABA}$.

With these considerations we obtain immediately

$$\mathbf{f}(0)=\mathbf{1}+\mathbf{B}+\mathbf{B}^2/2!-\mathbf{B}^3/3!+\ldots=\mathbf{1}+\mathbf{B}. \tag{A3}$$

The first derivative has to take the noncommutativity of $\mathbf{A}$ and $\mathbf{B}$ into account, and we obtain the sum of only three nonzero terms:

$$\partial_\omega\mathbf{f}(0)=\mathbf{A}+(\mathbf{AB}+\mathbf{BA})/2!+(\mathbf{BAB})/3! \tag{A4}$$

Any possible higher term is zero. For example, the fourth-order term $(\omega\mathbf{A}+\mathbf{B})(\omega\mathbf{A}+\mathbf{B})(\omega\mathbf{A}+\mathbf{B})(\omega\mathbf{A}+\mathbf{B})/4!$ can be expressed as a sum of sixteen products. If we take the derivative and evaluate at $\omega=0$, each of the products contains the vanishing term $\mathbf{BB}$. Similarly, in the corresponding permutations of all higher terms where the number of factors $\mathbf{B}$ exceeds the number of factors $\mathbf{A}$, the occurrence of the product $\mathbf{BB}$ is unavoidable. Using (A2), similarly higher derivatives can be derived, such as

$$(\partial_\omega)^2\mathbf{f}(0) = \mathbf{A}^2/2! + (\mathbf{AAB} + \mathbf{ABA} + \mathbf{BAA})/3! + (\mathbf{ABAB} + \mathbf{BABA})/4! + \mathbf{BABAB}/5! \tag{A5}$$

$$(\partial_\omega)^3\mathbf{f}(0) = \mathbf{A}^3/3! + 2\mathrm{A}^2(\mathbf{AB} + \mathbf{BA})/4! + (3\mathbf{A}^2\mathbf{BAB} + \mathbf{ABABA})/5! + (\mathbf{ABABAB} + \mathbf{BABABA})/6! + \mathbf{BABABAB}/7!. \tag{A6}$$

The functional dependence for the general scheme in obtaining the higher derivatives should be obvious.

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