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Dispersion theory of meromorphic total reflectivity

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The dispersion theory of meromorphic total reflectivity is considered by dealing with the poles of the complex reflectivity function in the upper half of the complex angular frequency plane. Modified Kramers-Kronig relations and corresponding static limit sum rules are derived for the degenerate third-order nonlinear reflectivity. A high-frequency sum rule is also suggested.

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I. INTRODUCTION

Kramers and Kronig [1–3] derived dispersion relations, KK relations, which have had many far-reaching consequences and applications. The underlying important physical concept of the existence of the KK relations is causality, which has been clearly pointed out, e.g., in the paper of Toll [4] and later in the study of Remacle and Levine [5]. The mathematical concept corresponding to causality is the holomorphicity of a complex physical function in the upper half of the complex angular frequency plane, and sufficient asymptotic falloff of the function at high frequencies. These KK relations provide a basic tool in optical data inversion in linear optical spectroscopy [6]. If we, for instance, measure the wavelength-dependent light transmission of a transparent material with the aid of a spectrophotometer, the corresponding wavelength-dependent refractive index change can be calculated using the appropriate KK relation [6], which actually is a principal-value integral. If the material is opaque then the complex refractive index is obtained by a KK phase-retrieval procedure, provided that the reflectance is measured with a reflectometer from a smooth surface of a condensed matter. The surface condition is important since Fresnel's theory of light reflection is the basis of data analysis. In the case of liquids the measurement is performed using a prism reflectometer.

The validity of the KK relations for reflectivity has been described, e.g., by Velicky [7] and Smith [8], and recently by Lee [9] for metals, which obey Drude's dispersion model [10]. In his elegant paper Lee avoided the principal-value integral using the dilogarithm function [9]. Few years ago Nash *et al.* [11] paid attention to a feature of a KK relation related to a reflectivity in the regime of Drude's theory (here reflectivity means a theoretical quantity whereas reflectance means a measured one), which is frequently used in the description of the permittivity of metals. According to them the conventional KK relation for phaseretrieval has to be modified by a constant term in cases where the extinction coefficient of the medium is different from zero at zero angular frequency. The modification stems from the appearance of a pole at zero frequency. Smith [8] has also pointed out the existence of a branch point of complex reflectivity in the case of conductors. The paper of Lee and Sindoni [12], which was devoted to the mathematical study of a logarithmic kernel in data inversion, confirmed and generalized the result of Nash *et al.* Note that the constant phase angle of Nash *et al.* is canceled in Ahrenkiel's [13] singly subtractive KK analysis of reflectance data, as well as, in the multiply subtractive KK analysis of Palmer *et al.* [14] in the case of odd number of reference (''anchor'') points.

Sum rules have much importance in linear optical spectroscopy since they provide the means to estimate the validity of the measured data and also the consistency of theoretical models. Outstanding works of Altarelli *et al.* [15], Altarelli and Smith [16], Smith and Manogue [17], and King [18,19] form the basis of sum rules, which are valid in linear optical spectroscopy.

The theory of dispersion relations in nonlinear optical spectroscopy was investigated a relatively long time ago [20–26], but their experimental validity for nonlinear susceptibility was postponed mainly due to experimental complexities until the work of Kishida *et al.* [27], who investigated third-harmonic generation from polysilane. The principle of causality was at first considered as the necessary and sufficient condition for the existence of KK relations also in nonlinear optics but Kircheva and Hadjichristov [28] have shown that this is not the case in the four-wave-mixing spectroscopy. Nevertheless, the principle of causality is of course valid also in four-wave-mixing spectroscopy. The invalidity of the KK relations in nonlinear optics was shown also for the total susceptibility of the two-level atom treated by Yariv [29]. Moreover, the validity and invalidity of the KK relations in femtosecond spectroscopy was investigated by Tokunaga *et al.* [30–32].

Peiponen [24], Vartiainen and Peiponen [33], and Peiponen *et al.* [34–36] studied the dispersion theory of so-called meromorphic nonlinear susceptibilities, i.e., susceptibilities that are not holomorphic due to singular points both in the upper- and lower-half planes. The singular points in the complex angular frequency space can in the general case be poles, zeros, and/or branch points. Usually these are isolated singularities, but the number of poles can be countably infinite, especially in the quantum-mechanical picture. The total susceptibility of Yariv is a simple example of a meromorphic function that has one pole and one zero in the upper-half plane. Despite the pathological case of a meromorphic total susceptibility, phase retrieval is possible, not by a KK dispersion relation, but by a phase-retrieval procedure based on the exploitation of the maximum entropy

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model for the modulus of Yariv's complex susceptibility [35]. This meromorphism of degenerate third-order nonlinear susceptibility appears also in the context of nanocomposites [37], which are expected to have importance in the field of optoelectronics and spectroscopy of nanostructures [38–46].

Peiponen [24,47] initiated the study of sum rules for nonlinear susceptibilities followed by Bassani and co-workers [25,48–53] and by Chernyak and Mukamel [54]. Recently, Peiponen [55] investigated dispersion relations and sum rules for the meromorphic complex refractive index, i.e., an index that has linear and nonlinear contribution.

As far as we know, neither modified Kramers-Kronig dispersion relations nor basic sum rules for complex meromorphic nonlinear normal reflectivity have been presented until now. Such a theory is expected to have importance in nonlinear reflection spectroscopy, especially when involving self-action processes, e.g., in nanocomposites. The investigation of the dispersion theory of optically nonlinear materials possessing nanostructures is important because such materials may provide all-optical switching, modulating, and computing devices [56,57]. Especially, materials possessing large third-order nonlinearity and having a quantum wire or dot structure have a key role in the optoelectronic device development. The aim in nonlinear optical nanostructure engineering is usually to find optimal constituents so that the nonlinear absorption of incident light is negligible. Unfortunately, this cannot always be achieved and in some cases it is even desirable such as in two-photon absorption induced two-photon fluorescence from microvolume, which has a great potential in drug discovery [58].

II. MEROMORPHIC SUSCEPTIBILITIES

Here we concentrate on the dispersion theory of insulators. The meromorphic third-order nonlinear susceptibility [6,59] ($\chi^{(3)}$) is degenerate in the sense that it involves simultaneously positive and negative angular frequency variables (ω), i.e., $\chi^{(3)}=\chi^{(3)}(\omega,\omega,-\omega)$. The real part of this type of degenerate nonlinear susceptibility is proportional to the nonlinear refractive index of a medium, whereas the imaginary part is proportional to the two-photon absorption coefficient [59]. Expressions for the meromorphic nonlinear susceptibility can be found from Refs. [6,52]. A simple expression, which is based on the anharmonic oscillator model, can be written as follows:

$$\chi^{(3)}(\omega,\omega,-\omega)=\frac{\mathrm{A}}{D(\omega)^3 D(-\omega)}, \tag{1}$$

where A is a constant and

$$D(\omega)=\omega_0^2-\omega^2-i\Gamma\omega. \tag{2}$$

In Eq. (2) ω_0 is the resonance frequency of an anharmonic oscillator and Γ is the damping parameter. Note that in the classical Drude's dispersion model for metals, we have to set $\omega_0=0$. If in Eqs. (1) and (2) we replace the real angular frequency variable with a complex one, say, Ω we observe that the corresponding nonlinear susceptibility function has poles simultaneously, i.e., $D(\Omega)=0$ and $D(-\Omega)=0$, in the upper- and lower-half planes. In the upper-half plane there appear first-order poles whereas in the lower-half plane the poles are of third order. This means that this susceptibility is not a holomorphic but a meromorphic function [60]. Therefore, we cannot apply the KK relations to such a function because of the lack of holomorphicity. Usually the contribution of the nonlinear response of the system is small compared with the linear one. Nevertheless, this usually small nonlinear but meromorphic contribution has drastic consequences to the properties of the total susceptibility as can be observed, e.g., in the case of Yariv's total susceptibility, which can be expressed as follows:

$$\chi_{\text{total}}(\omega)=C\frac{\omega_0-\omega+iT_2^{-1}}{(\omega_0-\omega)^2+\left(1+\dfrac{I}{I_{\mathrm{s}}}\right)T_2^{-2}}, \tag{3}$$

where C is a constant, T_2 the dephasing time, I the pump irradiance, and I_{s} the saturation irradiance. Inspection of Eq. (3) shows that the total susceptibility has two symmetric poles in opposite half planes and in addition one zero in the upper-half plane. Hutchings *et al.* [26] stated in their paper that the KK relations are valid for the total susceptibility of Eq. (3) in the regime of linear optics. This is not quite true since the total susceptibility of Yariv has no symmetry properties either in the nonlinear or in the linear case ($I=0$). That is to say the real and imaginary parts of Yariv's susceptibility are not even or odd functions of angular frequency. Therefore, only Hilbert transforms hold (as concerns the complex causal function and the Hilbert transform, see Ref. [5]) for the total susceptibility of Yariv. This means the unavoidable principal-value integration from $-\infty$ to $+\infty$, whereas the KK relations require less restrictive, and physically reasonable, integration from zero to infinity.

In the general case we can always express the total meromorphic susceptibility of the system as a sum of the holomorphic linear and the meromorphic nonlinear susceptibility as follows:

$$\chi_{\text{total}}(\omega)=\chi^{(1)}(\omega)+\chi^{(3)}(\omega,\omega,-\omega)I, \tag{4}$$

where $\chi^{(1)}$ is the linear susceptibility and I is the intensity of light. In the following section we consider the meromorphic complex reflectivity using the general expression of Eq. (4).

III. MODIFIED KRAMERS-KRONIG RELATIONS FOR MEROMORPHIC NORMAL REFLECTIVITY AND SUM RULES

We restrict our consideration on normal light incidence, since the dispersion theory in linear reflection spectroscopy is devoted to such a case. Nevertheless, the present theory can be generalized also to the case of oblique light incidence but it usually involves much more complicated algebra. The meromorphic complex normal reflectivity is

$$r(\omega)=\frac{1-\sqrt{1+\chi_{\text{total}}(\omega)}}{1+\sqrt{1+\chi_{\text{total}}(\omega)}}, \tag{5}$$

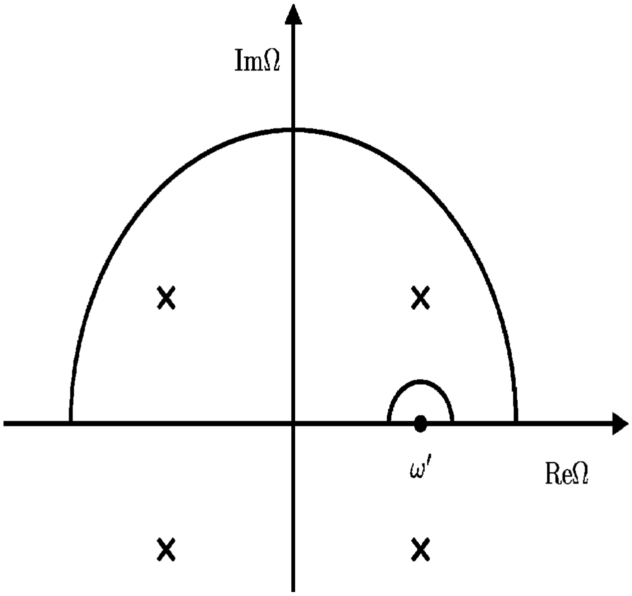

FIG. 1. Contour for derivation of modified KK relations (× indicates a pole).

where the meromorphic total susceptibility is the one defined in Eq. (4). The meromorphism of the total susceptibility induces the meromorphism of the complex reflectivity. The meromorphic nature can be observed rather easily for small nonlinearity by expressing the square roots in Eq. (5) using a binomial expansion.

On one hand, for example, in the case of the meromorphic total susceptibility of Eq. (3), and at the complex frequency for which it holds that $\chi_{\text{total}}=0$, the reflectivity would be real and equal to 0. In other words such an (physically unreasonable) angular frequency would mean total transparency of the medium. On the other hand, if the square root in Eq. (5) could have the value zero, i.e., if there could be branch points, then at the complex angular frequency matching the branch point, the medium would be totally reflecting. Generally speaking in the frame of Eq. (4), the existence of branch points can be ruled out due to the assumption that $\chi^{(3)}(\omega,\omega,-\omega)I\ll\chi^{(1)}(\omega)$, which is valid in usual experiments for real dense media including also nanocomposites. So we can expect that in most typical cases the meromorphism of the reflectivity is due only to the poles in both half planes. Making such an assumption we can perform complex contour integration in the upper-half plane as shown in Fig. 1. The reflectivity of Eq. (5) can be split in to a sum of linear and nonlinear contributions in the following way:

$$\begin{aligned} r(\omega)&=r^{(1)}(\omega)+r^{(3)}(\omega,\omega,-\omega)I\\ &=|r^{(1)}(\omega)|e^{i\varphi(\omega)}+|r^{(3)}(\omega,\omega,-\omega)|e^{i\phi(\omega,\omega,-\omega)}I, \end{aligned}\tag{6}$$

which is analogous to Eq. (4). The complex numbers are now expressed in their polar forms. Then the contour integration, which makes use of Cauchy's integral theorem [60] and the theorem of residues [61], provides the complex form of the Hilbert transform as follows:

$$\begin{aligned} i\pi r(\omega')=\mathrm{P}\int_{-\infty}^{\infty}\frac{|r^{(1)}(\omega)|e^{i\varphi(\omega)}}{\omega-\omega'}d\omega &+\mathrm{P}\int_{-\infty}^{\infty}\frac{|r^{(3)}(\omega,\omega,-\omega)|e^{i\phi(\omega,\omega,-\omega)}I}{\omega-\omega'}d\omega\\ &-2i\pi\sum_{\text{poles}}\mathrm{Res}\left[\frac{|r^{(3)}(\Omega,\Omega,-\Omega)|e^{i\phi(\Omega,\Omega,-\Omega)}I}{\Omega-\omega'}\right], \end{aligned}\tag{7}$$

where ω' is real and the summation is over the poles in the upper-half plane. The symbol P denotes the Cauchy principal value. Note that since the linear reflectivity is holomorphic the contribution of the residue term involves only the nonlinear reflectivity. For the total susceptibility we must require the fundamental symmetry properties [see Eq. (6) in Ref. [52]]

$$\chi^{(1)}(-\omega)=[\chi^{(1)}(\omega)]^{*},\tag{8}$$

$$\chi^{(3)}(-\omega,-\omega,\omega)=[\chi^{(3)}(\omega,\omega,-\omega)]^{*},\tag{9}$$

where (*) denotes complex conjugate. These symmetry relations express the reality of fields and induce corresponding symmetry relations for the linear and nonlinear reflectivities given in Eq. (6). Due to the symmetry relations the real parts of the linear and nonlinear reflectivities are even functions, whereas the corresponding imaginary parts are odd functions. Using this information and Euler's equation for a polar presentation of a complex number we get conventional KK relations for the linear reflectivity from Eq. (7) as follows:

$$|r^{(1)}(\omega')|\cos\varphi(\omega')=\frac{2}{\pi}\mathrm{P}\int_{0}^{\infty}\frac{\omega|r^{(1)}(\omega)|\sin\varphi(\omega)}{\omega^{2}-\omega'^{2}}\omega,\tag{10}$$

$$|r^{(1)}(\omega')|\sin\varphi(\omega')=-\frac{2\omega'}{\pi}\mathrm{P}\int_{0}^{\infty}\frac{|r^{(1)}(\omega)|\cos\varphi(\omega)}{\omega^{2}-\omega'^{2}}d\omega\tag{11}$$

and the modified KK relations for the nonlinear reflectivity,

$$\begin{aligned} &|r^{(3)}(\omega',\omega',-\omega')|\cos\phi(\omega',\omega',-\omega')\\ &\quad=\frac{2}{\pi}\mathrm{P}\int_{0}^{\infty}\frac{\omega|r^{(3)}(\omega,\omega,-\omega)|\sin\phi(\omega,\omega,-\omega)}{\omega^{2}-\omega'^{2}}d\omega\\ &\qquad-\mathrm{Im}\left\{2i\sum_{\text{poles}}\mathrm{Res}\left[\frac{|r^{(3)}(\Omega,\Omega,-\Omega)|e^{\phi(\Omega,\Omega,-\Omega)}}{\Omega-\omega'}\right]\right\}, \end{aligned}\tag{12}$$

$$|r^{(3)}(\omega',\omega',-\omega')|\sin\phi(\omega',\omega',-\omega') = -\frac{2\omega'}{\pi}\mathrm{P}\int_0^{\infty}\frac{|r^{(3)}(\omega,\omega,-\omega)|\cos\phi(\omega,\omega,-\omega)}{\omega^2-\omega'^2}d\omega + \mathrm{Re}\left\{2i\sum_{\text{poles}}\mathrm{Res}\left[\frac{|r^{(3)}(\Omega,\Omega,-\Omega)|e^{\phi(\Omega,\Omega,-\Omega)}}{\Omega-\omega'}\right]\right\}. \tag{13}$$

The KK relations of Eqs. (10) and (11) were already given by Smith and Manogue [17] but the modified relations, Eqs. (12) and (13), are fresh in the context of nonlinear reflectivity. We remark that in the case of meromorphic total reflectivity there is little use to formulate dispersion relations, such as those of Smith and Manogue [17] involving powers of r, i.e., r^k or functions such as $\omega^j r^k$, where j and k are appropriate integers. The reason is that higher powers would involve products of holomorphic ($r^{(1)}$) and meromorphic ($r^{(3)}$) functions, which usually implies a complex expression of the dispersion relations for such functions.

The conventional procedure in phase-retrieval problems in optical spectroscopy is to treat the logarithm of the latter equality in Eq. (6). Unfortunately, then we cannot resolve separately the linear and nonlinear parts such as those given in Eqs. (10)–(13). Furthermore, we have to remember that the poles of $r^{(3)}$, which are located in the upper-half plane are singular points of the logarithm. This brings additional complexity. If the total reflectivity has also complex zeros in the upper-half plane a term, the so-called Blaschke [4] product that takes into account the complex zeros must be included (see also the papers of Young [62] and Lee [63]). Such dispersion relations involving the logarithm of the reflectivity would certainly be important when trying to invert measured reflectance data. However, we recall that the residue terms such as those in Eqs. (12) and (13), which would appear also in the logarithm formalism, are usually complicated since they require the knowledge of the resonance points (complex numbers) of the system and involve a complex function of a complex angular variable. Evidently investigations are needed to find out the existence of the dispersion relations concerning the logarithm of the meromorphic reflectivity. Such a topic is beyond the present studies.

Sum rules for the total reflectivity can be found by allowing $\omega'=0$ in Eqs. (12) and (13). We assume that there is no dc phase shift of the electric field at $\omega'=0$ in the nonlinear case, analogous to the linear case [8,19]. This means that $\phi(0,0,0)=0$. Then we find out two sum rules. One of them is a nonlinear correction of the static reflectivity, i.e.,

$$|r^{(3)}(0,0,0)| = \frac{2}{\pi}\mathrm{P}\int_0^{\infty}\frac{|r^{(3)}(\omega,\omega,-\omega)|\sin\phi(\omega,\omega,-\omega)}{\omega}\mathrm{d}\omega - \lim_{\omega'\to 0}\mathrm{Im}\left\{2i\sum_{\text{poles}}\mathrm{Res}\left[\frac{|r^{(3)}(\Omega,\Omega,-\Omega)|e^{\phi(\Omega,\Omega,-\Omega)}}{\Omega-\omega'}\right]\right\} \tag{14}$$

and the other one is as follows:

$$\lim_{\omega'\to 0}\mathrm{Re}\left\{2i\sum_{\text{poles}}\mathrm{Res}\left[\frac{|r^{(3)}(\Omega,\Omega,-\Omega)|e^{\phi(\Omega,\Omega,-\Omega)}}{\Omega-\omega'}\right]\right\}=0. \tag{15}$$

It is obvious from Eq. (14) that the meromorphicity of the reflectivity introduces a residue term, which is a fresh feature if we compare to the static nonlinear correction given by Bassani and Scandolo [25] in the context of nonlinear permittivity. The sum rule of Eq. (15) is also interesting since the limiting process requires the real part of the sum of the residues to vanish.

The high-frequency limit of the linear reflectivity can be applied [17] to find out an *f*-sum rule by applying Eq. (10) and the falloff of the linear reflectivity proportional to ω'^{-2}. In the case of meromorphic total reflectivity, the existence of an *f*-sum rule is more problematic due to the residue term in Eq. (12), which explicitly falls off proportional to ω'^{-1}. However, if we restrict the consideration to the Hilbert transform of the total reflectivity of Eq. (7) we may write for the third-order reflectivity the following relation:

$$i\pi r^{(3)}(\omega',\omega',-\omega') = \mathrm{P}\int_{-\infty}^{\infty}\frac{r^{(3)}(\omega,\omega,-\omega)}{\omega-\omega'}d\omega - 2i\pi\sum_{\text{poles}}\mathrm{Res}\left[\frac{r^{(3)}(\Omega,\Omega,-\Omega)}{\Omega-\omega'}\right]. \tag{16}$$

Now we can quite reasonably expect that there is no nonlinear reflection of light at extremely high angular frequencies (the amplitude of the electric field is finite). In other words the medium acts just like vacuo. This means that when ω' tends to infinity, we can approximate

$$0\leftarrow\int_{-\infty}^{\infty}\frac{r^{(3)}(\omega,\omega,-\omega)}{-\omega'}d\omega - 2i\pi\sum_{\text{poles}}\mathrm{Res}\left[\frac{r^{(3)}(\Omega,\Omega,-\Omega)}{-\omega'}\right]. \tag{17}$$

Then we get a sum rule by multiplying the relation of Eq. (17) by ω', i.e.,

$$\int_{-\infty}^{\infty}r^{(3)}(\omega,\omega,-\omega)d\omega = +2i\pi\sum_{\text{poles}}\mathrm{Res}[r^{(3)}(\Omega,\Omega,-\Omega)]. \tag{18}$$

Evidently it is possible to separate sum rules both for real and imaginary parts in Eq. (18) but rigorous knowledge of the complex function $r^{(3)}(\Omega,\Omega,-\Omega)$ is then necessary.

IV. CONCLUSIONS

In this paper we have presented modified Kramers-Kronig relations and related static sum rules and a high-frequency sum rule for the meromorphic nonlinear reflectivity. The meromorphism of the nonlinear reflectivity stems from the degenerate nonlinear susceptibility of the medium. Such a susceptibility, which is related to a self-action process, has poles simultaneously in the upper- and lower-half planes.

We derived a sum rule, which gives the correction of the static nonlinear reflectivity. Other sum rules are related to the residues of the reflectivity. In principle, the real part of Eq. (18) yields a sum rule, which is analogous to the average sum rule for the linear refractive index given by Altarelli *et al.* [15].

In a more general case of a meromorphic total reflectivity, wherein complex zeros of the reflectivity are allowed in the upper-half plane, the dispersion relations and sum rules have to be revised. This means that at least one has to take into account the Blaschke product [4], which involves information about symmetric zeros. However, such a case is beyond the present study.

The present theory of meromorphic reflectivity is believed to have significance in the testing of optical spectra and models for the media related to degenerate third-order nonlinear susceptibility, which appears in the context of nonlinear refractive index and two-photon absorption. The spectral properties of nonlinear refractive index have importance, e.g., when developing optoelectronic devices based on the use of nanocomposite structures involving quantum wells, wires and dots. Wavelength-dependent two-photon absorption in turn is believed to be of crucial importance in drug discovery based on the use of nanoparticles for bioaffinity assays [58], and also in optical monitoring of controlled drug delivery by nanoparticles [64].

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