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On the dispersion theory of meromorphic optical constants

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Abstract

In nonlinear optical spectroscopy, the self-action of an intense light beam has an effect on the optical constants of a medium, which can be described in terms of a meromorphic function as defined in complex analysis. Here the dispersion theory of meromorphic optical constants is considered and general expressions for dispersion relations and sum rules are derived for meromorphic optical constants.

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1. Introduction

The interaction of light with a medium depends on the intrinsic optical constants (refractive index and extinction coefficient) of the medium. During the past century the intrinsic optical properties of media such as insulators, metals and semiconductors have been thoroughly investigated in experiments and theories. Nowadays the optical constants of media are relevant in many interdisciplinary fields of science, including physics, chemistry, engineering, medicine and biology. During the history of experiments on optical constants, new devices, for example, the ellipsometer [1], have been invented. With the theory of optical constants, the principle of causality has been crucial in describing the dispersion of light [2, 3] and, moreover, in the formulation of important dispersion relations known as Kramers–Krönig (KK) relations [4, 5]. KK relations, which result from the principle of causality, do not just yield information about optical constants or changes in these constants, but also give information about other intrinsic optical properties of media via sum rules [6], which cannot be obtained, for example, by ellipsometric measurements. In the case of nonlinear optics, the interaction of intense light with media is relatively complicated. Nevertheless, the validity of the KK relations for nonlinear susceptibilities of media has been known for a relatively long time [7–10]. Due to the experimental complexities of nonlinear optics, the validity of KK relations was shown as recently as 1992 by Kishida *et al* [11] for the case of a third-harmonic wave from polysilane film. Bassani and Scandolo [12] were the first researchers who explicitly derived the KK

relations for the optical constants of nonlinear media within the framework of pump and probe configurations.

In the case of so-called nonlinear meromorphic susceptibilities [13,14] and in femtosecond spectroscopy [15–17], however, it has been shown that KK relations are invalid. Lorentz's and Drude's [18] classical dispersion theory of linear susceptibility, in addition to the quantum mechanical description by Kramers and Heisenberg [18], allows the number of poles of complex linear susceptibility to be countably infinite. Also, the complex linear susceptibility diverges at the poles. We can state that such a complex-valued function behaves like a holomorphic function almost everywhere. In the context of linear optical spectroscopy the term 'analytic function', which is one of the important properties needed for the existence of the KK relations, is usually used instead of 'holomorphic function' in the literature [6]. Nevertheless, both terms have the same meaning, but in nonlinear optics (where two or more complex angular frequency variables may appear) the term 'holomorphic' is more appropriate according to the theory of several complex variables [19]. Now, however, if we consider the linear susceptibility in the whole complex angular-frequency plane, it means that we must call the susceptibility a meromorphic function. Fortunately, in the normal derivation of KK relations, it is possible to avoid the lower half plane where the poles of the linear susceptibility are located, by dealing with a function that is holomorphic in the upper half plane. The situation with nonlinear susceptibilities is very similar to the one above. However, nonlinear optical processes that demand the simultaneous existence of poles in both half planes require the generalization of the treatment of the complex nonlinear susceptibility and dispersion relations, because now the nonlinear susceptibility is a meromorphic function in the upper half plane. It is important to note the importance of the dispersion relations in the context of the spectra measurements. If the imaginary part, such as the absorption of light, can be measured then the real part, such as the refractive index, can be calculated and *vice versa*. In other words KK dispersion relations can be used for data inversion. The success of measurements and data inversion can be tested using sum rules [6,20]. In addition sum rules can be used for testing the consistency of some particular dispersion models of a medium [20].

In this paper, for the very first time the dispersion theory of meromorphic optical constants is considered. It is shown that KK relations for meromorphic optical constants are invalid. There is, however, no conflict with the principle of causality.

2. Dispersion relations for meromorphic optical constants

The equivalence of causality and dispersion relations for linear optical constants was shown by Krönig [21] and later by Toll [22] (see also [23]). The mathematical essence of causality is that it provides a *necessary and sufficient* condition for the existence of KK relations. The detailed derivation of KK relations, obtained by complex analysis, can be found, for example, in Peiponen *et al* [20]. There is no doubt about the validity of KK relations in the field of linear optics. We just mention here that the absolute values of the refractive index and the extinction coefficient are obtained by a KK-phase retrieval procedure based on the measurement of reflectance from a medium. If the absorption of light is measured by transmission, it is possible to obtain information with the aid of a KK relation about the change in the refractive index as a function of the light wavelength. Much of our present data on the optical constants of media are based on the use of KK relations.

In nonlinear optical spectroscopy we usually stimulate nonlinear processes using high-intensity laser beams impinging upon a medium. The complex refractive index can then be

given, for instance in a pump and probe experiment, as follows:

$$N(\omega_1, \omega_2, E) = N_L(\omega_1) + N_{NL}(\omega_1, \omega_2, E) \tag{1}$$

where ω_1 is the angular frequency of a weak probe beam, ω_2 is the angular frequency of a pump beam and E is the electric field of the pump beam. The linear complex refractive index is denoted in equation (1) by $N_L = n_L + ik_L$, where n_L is the real refractive index, k_L is the extinction coefficient, the corresponding nonlinear complex contribution is $N_{NL} = n_{NL} + ik_{NL}$ and i is the imaginary unit. The KK relations can then be given, using the property of the holomorphicity of the complex refractive index in the upper half of the complex angular frequency plane and an asymptotic fall off of $N = n + ik$ for high frequency, which is governed by the linear complex refractive index. The KK relations can be written as follows [12]:

$$n(\omega'_1, \omega_2, E) - 1 = \frac{2}{\pi} P \int_0^\infty \frac{\omega_1 k(\omega_1, \omega_2, E)}{\omega_1^2 - \omega'^2_1} d\omega_1 \tag{2}$$

$$k(\omega'_1, \omega_2, E) = -\frac{2\omega_1}{\pi} P \int_0^\infty \frac{n(\omega_1, \omega_2, E) - 1}{\omega_1^2 - \omega'^2_1} d\omega_1 \tag{3}$$

where P denotes the Cauchy principal value. KK relations similar to the standard forms of equations (2) and (3) are valid for most of the optical constants obtained by nonlinear processes. However, if we consider a third-order (or higher) nonlinear single-wave self-action process at frequency, ω , then the situation is completely different. The real part of the nonlinear complex refractive index is proportional to the real part of the third-order susceptibility, i.e. [24]

$$\text{Re} \{N_{NL}(\omega, \omega, -\omega)\} = \frac{3}{4\varepsilon_0 n_L^2 c} \text{Re} \{\chi^{(3)}(\omega, \omega, -\omega)\} I \tag{4}$$

where ‘Re’ denotes the real part, ε_0 is the vacuum permittivity, c is the velocity of light in vacuum and I is the intensity of light. The imaginary part of the third-order nonlinear susceptibility is related to two-photon absorption. In that case we can write [24]

$$\text{Im} \{N_{NL}(\omega, \omega, -\omega)\} = \frac{3\omega}{2\varepsilon_0 n_L^2 c^2} \text{Im} \{\chi^{(3)}(\omega, \omega, -\omega)\} I \tag{5}$$

where ‘Im’ stands for imaginary part. It is obvious that in both equations (4) and (5) there appears a minus sign in front of one angular frequency variable. This property has drastic consequences. We can describe the angular frequency dependence of the third-order susceptibility with the aid of a function $D(\omega) = (\omega_0^2 - \Omega^2 - i\Gamma\Omega)^{-1}$, where ω_0 is a resonance angular frequency of the medium, Ω is the complex angular frequency and Γ is related to the lifetime of an excited electron state. The third-order nonlinear susceptibility can be expressed using the products [20] of the D functions. Due to the minus sign of the angular frequency, the self-action third-order susceptibility involves a function $D^*(\omega) = (\omega_0^2 - \Omega^2 + i\Gamma\Omega)^{-1}$. The latter being the complex conjugate of D . It is evident that D indicates the existence of complex poles of $\chi^{(3)}$ and N in the lower half plane, while D^* indicates the simultaneous existence of poles of $\chi^{(3)}$ and N in the upper-half plane. This means that we have to give up the property of holomorphicity with N , which was a crucial condition for the existence of the KK relations. However, we can now deal with N as a meromorphic [25] function, which is holomorphic almost everywhere except on a numerable set of complex poles and zeros. The symmetry of the poles was recently described by Bassani and Lucarini [26]. Now the theorem of residues can be applied to yield dispersion relations, which are not of the KK form

$$n(\omega', \omega', -\omega') - 1 = \frac{2}{\pi} P \int_0^\infty \frac{\omega k(\omega, \omega, -\omega)}{\omega^2 - \omega'^2} d\omega - \text{Im} \left(2i \sum_{\text{poles}} \text{Res} \left[\frac{N(\Omega, \Omega, -\Omega)}{\Omega - \omega'} \right] \right) \tag{6}$$

$$k(\omega', \omega', -\omega') = -\frac{2\omega'}{\pi} P \int_0^\infty \frac{n(\omega, \omega, -\omega) - 1}{\omega^2 - \omega'^2} d\omega - \text{Re} \left(2i \sum_{\text{poles}} \text{Res} \left[\frac{N(\Omega, \Omega, -\Omega)}{\Omega - \omega'} \right] \right) \tag{7}$$

where the symmetry relations $N_L(-\omega) = N_L^*(\omega)$ and $N_{NL}(-\omega, -\omega, \omega) = N_{NL}^*(\omega, \omega, -\omega)$ are used. The residue terms in equations (6) and (7), i.e. the series expansions, are calculated for poles located in the first quadrant of the complex angular frequency space. Unfortunately, the residue terms involve complex functions; and such information cannot usually be obtained from measured optical spectra. Furthermore, in order to calculate the residues, we have to know the resonance points of the medium via optical spectrum. This may be an overwhelming task especially in the case of adjacent overlapping resonance lines in the spectrum.

Causality is always valid; however, it is evident from the above discussion that in a general case, which includes the meromorphism of optical constants (see [27] concerning the causality and dispersion relations of third-order degenerate nonlinear susceptibility), causality is necessary but *not a sufficient* condition for the existence of KK relations. An implication of this is that, as shown by equations (6) and (7), KK relations are invalid for meromorphic optical constants. A similar situation for the invalidity of KK relations can be found in femtosecond spectroscopy in the event of the simultaneous incidence of a probe and pump light pulse [15] upon a nonlinear medium. In such a case the third-order nonlinear susceptibility is meromorphic. It is important to emphasize that in femtosecond spectroscopy where pump and probe pulses are exploited the strict causality is broken if the pump arrives before the probe. That is to say, the medium is already responding to a cause (pump) before the probe beam is incident on the medium. Therefore, as pointed out by Tokunaga *et al* [16, 17] for third-order nonlinear susceptibility, the validity of the KK relations is questionable. The phase retrieval from meromorphic nonlinear susceptibility is possible, not by equations analogous to equations (6) and (7), but by using a maximum entropy model [20], which was exploited [28] to meromorphic third-order nonlinear susceptibility of a nanocomposite material representing a two-phase Maxwell–Garnett system [29].

3. Sum rules for meromorphic optical constants

Sum rules for meromorphic optical constants can be derived by the inspection of equations (6) and (7). For example, the DC-sum rule is obtained by setting $\omega' = 0$ in equation (6). Other sum rules are obtained by inspection; for example, a function $\omega^m [N(\omega, \omega, -\omega) - 1]^m$, where m is an integer. Then it holds, for example, for the odd m that

$$\omega^m \text{Im} \{N^m(\omega', \omega', -\omega')\} = -\frac{2}{\pi} P \int_0^\infty \frac{\omega^{m+1} \text{Re} \{N^m(\omega, \omega, -\omega) - 1\}}{\omega^2 - \omega'^2} d\omega - \text{Re} \left(2i \sum_{\substack{\text{poles, Im \{pole\} and} \\ \text{Re \{pole\} > 0}} \text{Re } s \frac{\Omega^m N^m(\Omega, \Omega, -\Omega)}{\Omega - \omega'} \right). \quad (8)$$

Now if we set $m = 1$ and let $\omega' \rightarrow 0$, we find that

$$\int_0^\infty [n(\omega, \omega, -\omega) - 1] d\omega = -\lim_{\omega' \rightarrow 0} \text{Re} \times \left(2i \sum_{\substack{\text{poles, Im \{pole\} and} \\ \text{Re \{pole\} > 0}} \text{Re } s \frac{\Omega N(\Omega, \Omega, -\Omega)}{\Omega - \omega'} \right) \quad (9)$$

provided that the limit is finite. The sum rule of equation (9) is analogous to the average sum rule of the linear refractive index given by Altarelli *et al* [6]. The right-hand side of equation (9) is equal to zero in the linear optics regime. A sum rule analogous to the f -sum rule can be found using the asymptotic property $N(\omega, \omega, -\omega) - 1 \rightarrow n_L(\omega) - 1 \rightarrow -1/2(\omega_p^2/\omega^2)$, which is valid when the angular frequency tends to infinity and where ω_p is the plasma frequency of the system. Thus, from equation (6) we obtain

$$\int_0^{\infty} \omega k(\omega, \omega, -\omega) d\omega = \frac{\pi}{4} \omega_p^2 - \lim_{\omega' \rightarrow \infty} \left(\text{Im } 2i \sum_{\substack{\text{poles, Im \{pole\} and Re \{pole\} > 0}} \text{Re } s \left(\frac{N(\Omega, \Omega, -\Omega)}{\Omega - \omega'} \right) \right). \quad (10)$$

Obviously, the second term on the right-hand side of equation (10) tends to zero. This is an important result since the f -sum rule is hereby a universal property irrespective of the linearity or nonlinearity or holomorphicity or meromorphicity of the complex optical constant of the system. This last observation complements the result of Bassani and Scandolo [12] who showed that the f -sum rule in the pump and probe system in nonlinear optics yields zero for the imaginary part of nonlinear susceptibility. Here we have shown that the f -sum rule holds for the meromorphic extinction coefficient, which includes both the linear and nonlinear extinction coefficients. The validity of the f -sum rule was proven to hold for the two-phase Maxwell–Garnett nanocomposite in a linear regime [30] and the result of equation (10) generalizes this.

Finally, we wish to emphasize that, due to the various practical applications of high-power lasers, for example, in technology and medicine, the spectroscopy of meromorphic optical constants will play an important role.

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