Modified Kramers-Kronig relations and sum rules for meromorphic total refractive index

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Modified Kramers-Kronig relations and corresponding sum rules are shown to hold for the total refractive index that can be presented as a sum of complex linear and nonlinear refractive indices, respectively. It is suggested that a self-action process, involving the degenerate third-order nonlinear susceptibility, can yield a negative total refractive index at some spectral range.

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I. INTRODUCTION

Nonlinear susceptibilities of a medium obey either the normal or the modified Kramers-Kronig (KK) relations [1–6]. Most of the nonlinear susceptibilities are holomorphic [7] functions in the upper half of the complex angular frequency plane, such as harmonic generation susceptibilities, for example, and obey the normal KK relations. However, in numerical data inversion of holomorphic nonlinear susceptibilities the multiply subtractive Kramers-Kronig (MSKK) relations [8] are more practical than the KK relations due to the strong convergence of the principal value integrals. Recently, Saarinen [9] gave a rigorous mathematical proof of the validity of the KK relations for the moments of arbitrary order harmonic generation susceptibilities. A meromorphic [7] nonlinear susceptibility, which obeys the modified KK relations, has poles simultaneously in both the upper and the lower half planes of the complex angular frequency space. Such a meromorphic nonlinear degenerate susceptibility appears in the context of a self-action process and involves only one input light beam.

In this paper we deal with a meromorphic total refractive index; that is to say, a refractive index that is a sum of the linear light intensity independent and the nonlinear light intensity dependent indices. We give modified KK relations for the total refractive index where the linear and nonlinear contributions are separated. Furthermore, sum rules for the meromorphic total refractive index are given. We also briefly point out the possibility of a negative real part of the total refractive index in nonlinear optical spectroscopy in the context of the self-action process. The negative refractive index has lately been a hot topic [10–22], but in the regime of linear spectroscopy.

II. DISPERSION THEORY OF TOTAL REFRACTIVE INDEX

The complex refractive index $N(\omega)$ of a homogeneous or effective medium obeys the familiar relation

$$N(\omega)=n(\omega)+\mathrm{i}\kappa(\omega)=\sqrt{\varepsilon(\omega)\mu(\omega)}$$
$$=\sqrt{[\varepsilon_1(\omega)+\mathrm{i}\varepsilon_2(\omega)][\mu_1(\omega)+\mathrm{i}\mu_2(\omega)]}, \qquad (1)$$

where $n(\omega)$ is the real refractive index, $\kappa(\omega)$ is the extinction coefficient, and $\varepsilon_{1,2}(\omega)$ and $\mu_{1,2}(\omega)$ are the real and imaginary parts of the relative permittivity and permeability, respectively. Note that in linear optical spectroscopy the KK relations hold separately for both the permittivity and the permeability [23]. It is well known from the theory of Maxwell that the strength of the electromagnetic field can affect the value of the permittivity and the permeability of a medium. In other words, these material parameters can be expressed as follows:

$$\varepsilon=\varepsilon_{\mathrm{L}}+\sum_{n=2}^{\infty}\chi_{\mathrm{NL},E}^{(n)}E^{n-1}, \qquad (2)$$

$$\mu=\mu_{\mathrm{L}}+\sum_{n=2}^{\infty}\chi_{\mathrm{NL},H}^{(n)}H^{n-1}. \qquad (3)$$

Here L stands for a linear contribution, $\chi_{\mathrm{NL}}^{(n)}$'s denote complex nonlinear electric and magnetic susceptibilities, and E and H denote electric and magnetic fields, respectively. Evidently, substitution of the expressions of Eqs. (2) and (3) into Eq. (1) makes the calculations of the complex refractive index somewhat complicated. However, we may assume that the medium behaves like an insulator, for which in the optical angular frequency range $\mu=1$. Furthermore, suppose that the insulator is under a third-order nonlinear self-action process, which involves only one incident light beam. This means that the complex total refractive index of the medium can be approximated as follows:

$$N(\omega)=N_{\mathrm{L}}(\omega)+N_{\mathrm{NL}}(\omega,\omega,-\omega)$$
$$=N_{\mathrm{L}}(\omega)+\chi_{\mathrm{NL},E}^{(3)}(\omega,\omega,-\omega)|E|^2. \qquad (4)$$

The imaginary part of the nonlinear contribution in Eq. (4) is related to the two-photon absorption [24]. If we resolve the real part of the total refractive index, we find that

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$$n(\omega)=n_{\mathrm{L}}(\omega)+\mathrm{Re}\{\chi^{(3)}_{\mathrm{NL},E}(\omega,\omega,-\omega)\}|E|^2. \tag{5}$$

The expressions in Eqs. (2) and (3) are more general than that of Eq. (5) in the sense that they allow higher order nonlinearities. Nevertheless, the higher order nonlinear processes are much weaker than the lower order processes; hence, Eq. (5) should be sufficient. Therefore, we may expect that Eq. (5) can be considered as a relatively good approximation. However, in the case that higher order nonlinear processes also contribute to some extent to the total refractive index, and thus allow fine-tuning of the sign of the total refractive index, Eq. (5) has to be revised. This means that higher terms in a series expansion in Eq. (5) have to be included.

Now if the linear refractive index is positive but almost equal to zero and the real part of the degenerate third-order susceptibility is negative, then, in theory, a negative refractive index can be obtained for high intensity radiation. A negative real part of the third-order degenerate nonlinear susceptibility at some wavelength range can be realized with homogeneous media [25] and nanocomposites [26]. A relatively high negative nonlinear refractive index has been observed with, e.g., π-conjugated polymers [27] and InGaAsP [28]. An interesting possibility with light intensity induced refractive index change is switching between positive and negative total refractive index just by tuning the intensity of a laser beam.

Unfortunately, the degenerate third-order nonlinear complex susceptibility, which may allow also the existence of a left-handed medium, has poles in both the lower and upper half planes [7]. This in turn means that the total complex refractive index is a meromorphic function. However, the total complex refractive index can be split, according to Eq. (4), into a sum of the always holomorphic linear and the meromorphic nonlinear refractive index, respectively. The former obeys the KK and MSKK relations, whereas the latter obeys so-called modified KK relations [6]. Modified KK relations, which can be derived with the aid of complex contour integration [7], were given purely for the total refractive index itself in Ref. [5]. The following relations resolve the linear and nonlinear contributions for the total refractive index:

$$\begin{aligned} n(\omega')-1=&\frac{2}{\pi}\mathrm{P}\int_0^{\infty}\frac{\omega\kappa_{\mathrm{L}}(\omega)d\omega}{\omega^2-\omega'^2}\\ &+\frac{2}{\pi}\mathrm{P}\int_0^{\infty}\frac{\omega\,\mathrm{Im}\{\chi^{(3)}_{\mathrm{NL},E}(\omega,\omega,-\omega)\}}{\omega^2-\omega'^2}d\omega\\ &-\mathrm{Im}\left\{2i\sum_{\mathrm{poles}}\mathrm{Res}\left[\frac{\chi^{(3)}_{\mathrm{NL},E}(\Omega,\Omega,-\Omega)}{\Omega-\omega'}\right]\right\}, \end{aligned} \tag{6}$$

$$\begin{aligned} \kappa(\omega')=&-\frac{2\omega'}{\pi}\mathrm{P}\int_0^{\infty}\frac{[n(\omega)-1]}{\omega^2-\omega'^2}d\omega\\ &-\frac{2\omega'}{\pi}\mathrm{P}\int_0^{\infty}\frac{\mathrm{Re}\{\chi^{(3)}_{\mathrm{NL},E}(\omega,\omega,-\omega)\}}{\omega^2-\omega'^2}d\omega\\ &+\mathrm{Re}\left\{2i\sum_{\mathrm{poles}}\mathrm{Res}\left[\frac{\chi^{(3)}_{\mathrm{NL},E}(\Omega,\Omega,-\Omega)}{\Omega-\omega'}\right]\right\}, \end{aligned} \tag{7}$$

where P denotes the Cauchy principal value, Ω is a complex angular frequency variable, ω' is a singular point on the real angular frequency axis, and the summation is over the poles in the upper half plane. A problem with the modified dispersion relations is the calculation of the residue terms, which require knowledge of the nonlinear susceptibility at complex frequencies and *a priori* knowledge of the resonance points of the system. Therefore, the relations (6) and (7), at the present stage, have little practical utility in the sense of numerical data inversion in comparison to the KK and MSKK relations and sum rules given for holomorphic nonlinear susceptibilities [1,2,7,8]. However, the dispersion relations (6) and (7) provide a frame to test theoretical dispersion models of electronic systems and incorporated nonlinear optical properties that may be suggested to hold for nonlinear media. Next we generalize our previous result [25] and give a sum rule for the powers of the total meromorphic refractive index using the theorem of residues as follows:

$$\int_{-\infty}^{\infty}[N(\omega)-1]^k d\omega=2\pi i\sum_{\mathrm{poles}}\mathrm{Res}[N(\Omega)-1]^k, \tag{8}$$

where $k=1,2,\ldots$. Let us first consider the case $k=1$. The real part of the linear refractive index of an insulating medium satisfies the well-known Altarelli-Dexter-Nussenzveig-Smith (ADNS) sum rule [29]

$$\int_0^{\infty}[n_{\mathrm{L}}(\omega)-1]d\omega=0, \tag{9}$$

which means that the linear refractive index averaged over all frequencies must be unity. Since $n_{\mathrm{L}}(\omega)$ is an even function of the angular frequency variable, i.e., $n_{\mathrm{L}}(-\omega)=n_{\mathrm{L}}(\omega)$, it holds that

$$\int_{-\infty}^{\infty}[n_{\mathrm{L}}(\omega)-1]d\omega=0. \tag{10}$$

Moreover, $\kappa_{\mathrm{L}}(\omega)$ is an odd function, i.e., $\kappa_{\mathrm{L}}(-\omega)=-\kappa_{\mathrm{L}}(\omega)$, and therefore we obtain

$$\int_{-\infty}^{\infty}\kappa_{\mathrm{L}}(\omega)d\omega=0. \tag{11}$$

From Eqs. (8), (10), and (11) we can deduce that

$$\int_{-\infty}^{\infty}\mathrm{Re}\{N_{\mathrm{NL}}(\omega,\omega,-\omega)\}d\omega = \mathrm{Re}\left\{2\pi i\left|E\right|^{2}\sum_{\mathrm{poles}}\mathrm{Res}[\chi^{(3)}_{\mathrm{NL},E}(\Omega,\Omega,-\Omega)]\right\} \tag{12}$$

and

$$\int_{-\infty}^{\infty}\mathrm{Im}\{N_{\mathrm{NL}}(\omega,\omega,-\omega)\}d\omega = \mathrm{Im}\left\{2\pi i\left|E\right|^{2}\sum_{\mathrm{poles}}\mathrm{Res}[\chi^{(3)}_{\mathrm{NL},E}(\Omega,\Omega,-\Omega)]\right\}. \tag{13}$$

Equation (12) is the counterpart of the ADNS sum rule (9) but in nonlinear optics. However, the sum rule of Eq. (9) is universal in the sense that it is independent of material parameters, whereas the sum rule of Eq. (12) is dependent on the resonance points of the nonlinear medium and on the intensity of the light. The sum rule (13) gives the nonlinear contribution of the integrated area of the extinction curve due to the two-photon absorption. In the case when $k\geqslant 2$ the nonlinear sum rules become rather complicated if the real and imaginary parts are resolved. Such sum rules can be simplified, but only a little, using the sum rules for the powers of linear optical constants given by Altarelli and Smith [30]. Finally, we remark that modified dispersion relations and corresponding sum rules can also be given for the meromorphic function $\omega^{j}[N(\omega)-1]^{k}$ by proper choice of the integers j and k. Unfortunately, such relations are also complicated due to the mixing of linear and nonlinear optical constants.

III. CONCLUSIONS

First we proposed that a negative real part of the total refractive index may be obtained in the context of a nonlinear self-action process, which involves the degenerate nonlinear third-order susceptibility. Generally speaking, the nonlinear susceptibility describing the self-action process is a meromorphic function, which means that the conventional KK relations have to be revised for it. We gave the modified KK relations for the total refractive index by separating the linear and nonlinear contributions. We also derived sum rules for the meromorphic total refractive index. The present theory can be applied in testing theoretical models that are suggested to describe the optical properties of self-action processes in insulating nonlinear media.

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