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Derivation of general dispersion relations and sum rules for meromorphic nonlinear optical spectroscopy

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Dispersion relations and sum rules for nonlinear susceptibilities are derived using complex analysis and especially the concept of a meromorphic function. The dispersion relations and sum rules provide frames to investigate the consistency between the theory and experiments.

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I. INTRODUCTION

Dispersion relations have been widely recognized as a powerful tool in spectroscopy that may, in particular, enable us to reveal the linear and nonlinear refraction coefficient from the relevant absorption spectrum. In linear spectroscopy, the dispersion theory approach is based on the Kramers-Kronig (KK) relations [1,2], which connect the real and imaginary parts of the linear susceptibility. Actually, much of the present spectral data related to the linear optical properties of the media, among others such as metals and semiconductors, have their origin in the spectral data inversion based on the exploitation of the KK relations and sum rules [3]. The extension of KK relations to nonlinear spectroscopy has been proposed in the early 1960s [4–6]. However, detailed study revealed a number of fundamental difficulties, which originate from the multiwave nature of the nonlinear optical interactions. This has stimulated an extensive search for proper formulation of the dispersion relations in nonlinear optics and their practical implementation in nonlinear optical spectroscopy [7–15]. In particular, consistency between the predictions of the dispersion theory and experimental data was verified by Kishida *et al.* [16] who measured the real and imaginary part of the third-order nonlinear susceptibility of polysilane using third harmonic generation process. Very recently Lucarini and Peiponen [17] showed that the experimental data obtained from the measurements of the third-order nonlinear susceptibility in polymers are consistent with the so-called generalized KK relations. We wish to emphasize that the range of available wavelengths for recording spectra is much more restricted in nonlinear optical spectroscopy than in the case of recording linear optical spectra. Therefore, any tests including sum rules, which can be used for checking the reliability of the measured and inverted data, are welcome. Unfortunately, KK relations and sum rules have found little applications in practical data analysis or checking the consistency of the measured data as concerns nonlinear optical spectra. One reason may be the tedious measurements, such as those of Kishida *et al.* [16] that require tunable dye lasers in order to obtain as broad spectral range as possible. Another reason may be that the full potential of KK relations and sum rules is not so familiar among spectroscopists in the field of nonlinear optics. However, we believe that recent progress in technology and dispersion theory in the field of nonlinear optical spectroscopy will enhance the applications of dispersion relations and sum rules in practical spectra analysis. An important step for analyzing nonlinear optical spectra at narrow spectral range was the introduction of the concept of multiply subtractive KK relations, which greatly improve the reliability of data inversion as has been demonstrated for polymer [18,19]. Furthermore, then sum rules for finite wavelength range become more practical as it was demonstrated, e.g., in the case of coherent anti-Stokes Raman spectrum (CARS) of nitrogen Q branch [20].

One general requirement for the validity of KK relations for nonlinear susceptibilities is that they have sufficient asymptotic fall off at high frequencies. By generalizing the Kubo's [21] linear-response theory to nonlinear systems [11] one can show that asymptotic behavior of the nonlinear susceptibilities at high frequencies satisfies the KK-type dispersion relations. However, the general analysis shows that there is an additional and more restrictive general requirement. Specifically, the nonlinear susceptibility should be a holomorphic [7] function of the complex frequency. In linear and nonlinear optics the susceptibility is holomorphic when it has poles only in the other (typically lower) half of the complex frequency plane. In particular, the nonlinear optical susceptibilities describing the generation of the optical harmonics satisfy this condition.

If the poles of the nonlinear susceptibility are located in the lower half plane then it is possible to write integral relations, i.e., so-called Hilbert-transforms [7] that connect the real and imaginary parts of the susceptibility. If the poles were located only in the upper half plane then also Hilbert transforms can be written but then the sign of the transform is switched [22] to negative if compared to the case of susceptibility having poles only in the lower half plane. Nevertheless, symmetry relations, under the assumption of real response function imposed on holomorphic linear and nonlinear susceptibilities, make it possible to rewrite the Hilbert transforms so that they take the familiar forms of the KK relations.

If the poles were located in both upper and lower half planes, the susceptibility can be termed as a meromorphic function of the frequency. One of the simplest examples of

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*Author to whom correspondence should be addressed. Electronic address: kai.peiponen@joensuu.fi such a function is the nonlinear susceptibility, which describes self-action of the light beam or frequency degenerate pump-probe spectroscopy. In time-resolved measurements, the applicability of the KK-type dispersion relations was first questioned by Tokunaga *et al.* [23]. They have shown both theoretically and experimentally that the presence of the poles in both half planes simultaneously makes these dispersion relations invalid for zero time delay between the pump and probe pulses. However, meromorphic functions have been shown to satisfy the generalized dispersion relations [24] allowing one to develop a consistent dispersion theory of the nonlinear optical susceptibilities even for frequency degenerate nonlinear interactions.

In this paper, by employing the theory of the meromorphic functions we obtain dispersion relations for nonlinear optical susceptibilities in Sec. II. In Sec. III we derive sum rules, which can be used to interpret experimental data, when the conventional KK-type relations are invalid. We wish to emphasize that some of the sum rules have not been, as far as we know, presented in the literature until now. Finally, we present our conclusions and the Appendix related to the derivation of sum rules.

II. CAUSALITY AND DISPERSION RELATIONS

Causality [25] is the primary reason for the existence of KK relations in the field of linear optics, but in the field of nonlinear optics it cannot be taken for granted that causality is the necessary and sufficient condition for the validity of the KK relations [22]. Indeed in the case of degenerate nonlinear susceptibility it has been shown that KK relations are invalid [24]. One thing is sure; causality works always also in the cases where the pump beam arrives before the probe in pump and probe experiments. The observation of Kircheva and Hadjichristov [22] concerning the invalidity of the traditional treatment of the causality by means of response function, due to a complicated relationship between the nonlinear polarization and excitation fields, has fundamental importance. So how to proceed in order to realize as general dispersion relations as possible? In a sense the case of meromorphic nonlinear susceptibility presents a more general case than that of the holomorphic one. The reason is that holomorphic nonlinear susceptibility can be considered as a special case of a meromorphic nonlinear susceptibility. Therefore, below we present results for meromorphic nonlinear quantities and show how these results work with holomorphic cases. We assume the most general case that the response function or correlation function, as it may be called depending on its context, can be a complex one [22,26]. In such a case there is no assumption of specific symmetry of the real and imaginary parts, which is consistent with the observations made by Remacle and Levine [26]. Note that Tokunaga *et al.* [23] observed simultaneously real and imaginary parts of susceptibility, which both were even functions at zero delay. In context with real response function the real and imaginary parts of the susceptibility obey always even and odd parity, respectively. We remark that one can find causal correlation function despite of the invalidity of the KK relations [22].

The *n*th order nonlinear susceptibility in general case can be expressed as $\chi^{(n)}=\chi^{(n)}(\Sigma_{j=1}^{n}\,\omega_j;\omega_1,\omega_2,\cdots,\omega_n)$. In the case that all frequencies are allowed to change simultaneously, one can write dispersion relations and sum rules but they are not of practical utility. Indeed, usually in experiments there is little sense to scan simultaneously the wavelength, for instance, of the probe and pump beams in pump and probe spectroscopy. Nevertheless, the theory presented below can be generalized to such cases of multivariate wavelengths by using the concept of several complex frequency variables in a similar manner as it was done in Ref. [27]. In order to understand the difference between holomorphic and meromorphic nonlinear susceptibilities and to realize what kind of nonlinear spectroscopies may be involved when we wish to utilize dispersion relations and sum rules we consider as an example the case of third-order nonlinear susceptibility in pump and probe experiments. The usual experimental situation is that the frequency (ω_2) of the pump beam is fixed, while the frequency (ω_1) of the probe beam is scanned. In fact all possible third-order nonlinear (and also higher order but weaker) processes are competing together, and the total nonlinear susceptibility is the sum of different nonlinear susceptibilities related to different competing processes (this matter is nicely described and illustrated, e.g., in the paper of Bassani and Lucarini [28]). Then, for instance, $\chi^{(3)}(2\omega_1-\omega_2;\omega_1,\omega_1,-\omega_2)$ is a holomorphic function of ω_1 and it presents nonlinear susceptibility of CARS. On the contrary, the degenerate nonlinear susceptibility $\chi^{(3)}(\omega_1;\omega_1,-\omega_1,\omega_1)$ is meromorphic. This susceptibility has importance in degenerate four-wave mixing process and in self-action processes (involving one incident light beam), the last mentioned is related to an important optical property of medium, namely, nonlinear refractive index. We remark that one can easily distinguish the meromorphic nonlinear susceptibility from the holomorphic one. In the case of meromorphic nonlinear susceptibility the same variable frequency appears simultaneously with positive and negative signs just like in the case of the degenerate nonlinear susceptibility we mentioned above. For the sake of simplicity we denote the arbitrary-order nonlinear susceptibility by the function f, and omit the other fixed frequencies except one, which is allowed to vary and denote it by x. This function is assumed to be a complex function of real variable, x, $f(x)=u(x)+iv(x)$, where x is a frequency. Next we utilize the theory of complex analysis and consider $f=f(z)$ as a meromorphic function of the complex variable z. We perform complex contour integration as shown in Fig. 1(a). Then we can write

$$\oint_C \frac{f(z)}{z-x'}dz=\mathrm{P}\int_{-R}^{R}\frac{f(x)}{x-x'}dx+\int_{\Gamma}\frac{f(z)}{z-x'}dz+\int_{A}\frac{f(z)}{z-x'}dz, \tag{1}$$

where P denotes the Cauchy principal value. The next phase is to let the radius R to tend to infinity. The integration along the closed contour on left-hand side of Eq. (1) gives according to the theorem of residues usually a nonzero contribution due to the poles located in the upper half plane. The first integral on right-hand side of Eq. (1) is the integral that is the

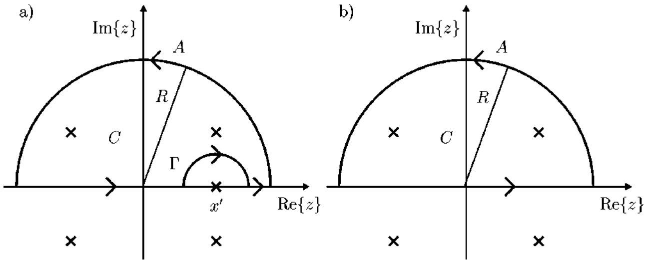

FIG. 1. Contour for the derivation of (a) the dispersion relations and (b) the sum rules (=pole).

origin of Hilbert transforms. The second integral on right-hand side in turn gives a nonzero contribution. Finally the last integral on right-hand side of Eq. (1) is equal to zero (rigorous mathematical proof is presented in the paper of Saarinen [13]). Thus we get the following results by separating real and imaginary parts as follows:

$$u(x') = \frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{v(x)}{x-x'}dx - 2\,\mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\mathrm{poles}}\mathrm{Res}\,\frac{f(z)}{z-x'}\right\} \quad (2)$$

and

$$v(x') = \frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{u(x)}{x-x'}dx - 2\,\mathrm{Im}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\mathrm{poles}}\mathrm{Res}\,\frac{f(z)}{z-x'}\right\}. \quad (3)$$

If there are no poles in the upper half plane, then the residue terms in Eqs. (2) and (3) are zero. Thus we have the more familiar case of a holomorphic quantity, i.e., Hilbert transforms. In the case of frequency dependent nonlinear susceptibility this means that the response function is real and automatically symmetry relations can be written, which imply KK relations. In the case on frequency variable the physical reality requires positive frequencies. Equations (2) and (3) can be written in other forms, allowing only positive frequency, by resolving the u and v functions to the sums of even and odd parts as follows:

$$u(x) = u_{\mathrm{even}}(x) + u_{\mathrm{odd}}(x), \quad (4)$$

$$v(x) = v_{\mathrm{even}}(x) + v_{\mathrm{odd}}(x), \quad (5)$$

which hold always. Hence, we may write

$$u(x') = -\frac{2x'}{\pi}\mathrm{P}\int_0^{\infty}\frac{v_{\mathrm{even}}(x)}{x^2-x'^2}dx + \frac{2}{\pi}\mathrm{P}\int_0^{\infty}\frac{x v_{\mathrm{odd}}(x)}{x^2-x'^2}dx - 2\,\mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\mathrm{poles}}\mathrm{Res}\frac{f(z)}{z-x'}\right\} \quad (6)$$

and

$$v(x') = \frac{2x'}{\pi}\mathrm{P}\int_0^{\infty}\frac{u_{\mathrm{even}}(x)}{x^2-x'^2}dx - \frac{2}{\pi}\mathrm{P}\int_0^{\infty}\frac{x u_{\mathrm{odd}}(x)}{x^2-x'^2}dx - 2\,\mathrm{Im}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\mathrm{poles}}\mathrm{Res}\,\frac{f(z)}{z-x'}\right\}, \quad (7)$$

where $x' > 0$. As we may observe the integrals in Eqs. (6) and (7) are now of KK type. The parity of the function f can be revealed by Fourier analysis. It is the residue term that may cause problems, since it requires the complex function of complex variable. Usually the poles are the resonance points of the system. Then if there is information on the transition frequencies and the lifetimes of the electronic states of the medium then poles can be estimated. Thus one can try to construct the complex nonlinear susceptibility function which is holomorphic almost everywhere except at the poles. We remark that the most general case of meromorphic function allows also the existence of complex zeros with f. Furthermore, it is also important to remark that the dispersion theory above can be applied also for the powers of the function f, f^j, where j is an integer, and also to the appropriate moments, $x^k f^j$, where k and j are integers.

Usually wavelength dependent spectrum in nonlinear optics is recorded. If we get information only on the modulus f then the real and imaginary parts can be retrieved both in linear and nonlinear optical spectroscopy by maximum entropy method [7,29,30]. Then the calculation of the correlation function, in the case of weak probe beam, may be based on the procedure presented by Remacle and Levine [26]. However, such a study is beyond the scope of this paper.

If the experimental conditions are arranged so that definitely holomorphic nonlinear susceptibility is dominant in the measured spectrum then the data inversion is usually relatively simple. Thus, the most reliable way for data inversion between the real and imaginary parts of the nonlinear susceptibility is based on the application of singly or multiply subtractive KK relations, and subsequent sum rules [17–19]. However, if the spectrum (total nonlinear susceptibility) is a combination of holomorphic and meromorphic susceptibilities, none of them being dominant, or meromorphic only, then the only reasonable way for practical data inversion is based on the application of the phase retrieval by maximum entropy method. In this latter case sum rules of practical utility are needed to check the consistency between theory and experimental data. The following section describes the derivation of sum rules for meromorphic nonlinear susceptibilities. These sum rules are believed to find applications at least in degenerate four-wave mixing and self-action spectroscopies.

III. SUM RULES

Sum rules have been exploited for a long time in linear optical spectroscopy as constraints for self-consistence study of experimental and calculated data. Unfortunately, the same is not true in the field of nonlinear optics probably because this field has developed much later than linear optics.

We obtain immediately the dc-sum rules from Eqs. (2) and (3) as follows:

$$u(0)=\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{v(x)}{x}dx-2\ \mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ \frac{f(z)}{z}\right\} \tag{8}$$

and

$$v(0)=-\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{u(x)}{x}dx-2\ \mathrm{Im}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ \frac{f(z)}{z}\right\}. \tag{9}$$

In the case of the dispersion relations of Eqs. (6) and (7) for $x'=0$ the first integrals on the right-hand side of these equations vanish and only the odd parts of the functions contribute to the dc-sum rules. dc-sum rules constitute constraints that the meromorphic nonlinear susceptibilities have to obey. If f were holomorphic then Eq. (9) yields $v(0)=0$, because u has to be an even function of x. Equation (8) in turn yields the familiar sum rule known already in linear optics

Next we derive another set of sum rules by making use of the complex contour integration shown in Fig. 1(b). Thus we can write the following equation:

$$\oint_C f(z)dz=\int_{-R}^{R}f(x)dx+\int_A f(z)dz. \tag{10}$$

Again letting the radius R to tend to infinity (in a symmetric manner leading to a principal value integration) and noting that then integral along the arc A vanishes, it follows from the theorem of residues that

$$\mathrm{P}\int_{-\infty}^{\infty}u(x)dx=-2\pi\ \mathrm{Im}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ f(z)\right\} \tag{11}$$

and

$$\mathrm{P}\int_{-\infty}^{\infty}v(x)dx=2\pi\ \mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ f(z)\right\}. \tag{12}$$

If the partition of even and odd functions, Eqs. (4) and (5), is exploited then the sum rules (11) and (12) change at the left-hand side of these equations so that they involve integration only on semi-infinite positive real axis and the integrands involve only even functions. Sum rules such as those given by Eqs. (11) and (12) provide other constraints that the meromorphic nonlinear susceptibility has to obey. In the case of a holomorphic quantity the only sum rule follows from Eq. (11), i.e.,

$$\mathrm{P}\int_{-\infty}^{\infty}u(x)dx=\int_{-\infty}^{\infty}u(x)dx=0. \tag{13}$$

Unfortunately, the sum rules above involving residue terms, so far, have importance only in testing of theoretical models, whereas sum rules that would involve measured data only would have crucial importance. Therefore, we now take the first step towards more practical sum rules for the meromorphic nonlinear susceptibility. For this purpose we consider Eqs. (2) and (3) and integrate them as a principal value integral with respect to x'. Then we find that

$$\begin{aligned}\mathrm{P}\int_{-\infty}^{\infty}u(x')dx' &= \mathrm{P}\int_{-\infty}^{\infty}\left[\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{v(x)}{x-x'}dx\right]dx' \\ &\quad -\mathrm{P}\int_{-\infty}^{\infty}2\ \mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ \frac{f(z)}{z-x'}\right\}dx' \\ &= \mathrm{P}\int_{-\infty}^{\infty}\left[\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{v(x)}{x-x'}dx\right]dx' \\ &\quad -2\pi\ \mathrm{Im}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ f(z)\right\}\end{aligned} \tag{14}$$

and

$$\begin{aligned}\mathrm{P}\int_{-\infty}^{\infty}v(x')dx' &= \mathrm{P}\int_{-\infty}^{\infty}\left[-\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{u(x)}{x-x'}dx\right]dx' \\ &\quad -\mathrm{P}\int_{-\infty}^{\infty}2\ \mathrm{Im}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ \frac{f(z)}{z-x'}\right\}dx' \\ &= \mathrm{P}\int_{-\infty}^{\infty}\left[-\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{u(x)}{x-x'}dx\right]dx' \\ &\quad +2\pi\ \mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\text{poles}}\mathrm{Res}\ f(z)\right\}.\end{aligned} \tag{15}$$

The principal value integral is needed because of the residue terms in Eqs. (14) and (15), which would otherwise diverge logarithmically. In the calculation of Eqs. (14) and (15) we exploited the partial fraction for the meromorphic function [31] of the form $c_{ik}/(z-a_i)^k$, where a_i are the poles of the function, k is a positive integer, and c_{ik} are complex constants. As far as we know in all cases of nonlinear susceptibilities the order of the poles appearing in the upper half plane is $k=1$, whereas the order of poles appearing in the lower half plane can be higher, see, e.g., [28] as concerns the third-order nonlinearities. In the Appendix we outline the calculation of the integral, which involves the residue term.

The order of the double integration can be changed only in the case of uniform convergent integrals. In such a case one can integrate the function $(x-x')^{-1}$ separately in Eqs. (14) and (15). King [32] used such a strategy when he derived sum rules for optical constants in the field of linear optical spectroscopy.

Now if we compare Eqs. (11), (12), (14), and (15) we find that

$$\mathrm{P}\int_{-\infty}^{\infty}\left[\mathrm{P}\int_{-\infty}^{\infty}\frac{v(x)}{x-x'}dx\right]dx'=0 \tag{16}$$

and

$$\mathrm{P}\int_{-\infty}^{\infty}\left[\mathrm{P}\int_{-\infty}^{\infty}\frac{u(x)}{x-x'}dx\right]dx'=0. \tag{17}$$

As far as we know, the sum rules (16) and (17) have not been presented before until now. We obtained sum rules that do not involve the cumbersome residue terms but integrals of quantities that may be measured.

The integrations in Eqs. (16) and (17) cover also negative frequencies. However, when we substitute the expressions of Eqs. (4) and (5) into (16) and (17) we get sum rules, which take the forms

$$\mathrm{P}\int_0^{\infty}\left[\mathrm{P}\int_0^{\infty}\frac{xv_{\mathrm{odd}}(x)+x'v_{\mathrm{even}}(x)}{x^2-x'^2}dx\right]dx'=0 \tag{18}$$

and

$$\mathrm{P}\int_0^{\infty}\left[\mathrm{P}\int_0^{\infty}\frac{xu_{\mathrm{odd}}(x)+x'u_{\mathrm{even}}(x)}{x^2-x'^2}dx\right]dx'=0. \tag{19}$$

It is noteworthy that the sum rules above, although describing meromorphic nonlinear susceptibility, have an analogy in linear optics. Indeed, following King's [32] argumentation concerning average index sum rule for complex refractive index $N(x)=n(x)+ik(x)$, where n is the real refractive index and k is the extinction coefficient of the medium, it holds that

$$\int_0^{\infty}[n(x)-1]dx=\mathrm{P}\int_{-\infty}^{\infty}\left[-\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{xk(x)}{x^2-x'^2}dx\right]dx'=0. \tag{20}$$

Naturally in the case of Eq. (20) the well known crossing relations $n(-x)=n(x)$ and $k(-x)=-k(x)$ are valid. In Eqs. (18) and (19) the integrand is an even function just like in the case of the sum rule (20).

IV. CONCLUSIONS

In this paper we have presented general dispersion relations and sum rules for nonlinear optical spectroscopy. We derived dispersion relations and sum rules that are valid both for holomorphic (analytic) or meromorphic nonlinear susceptibilities.

The theory is expected to have importance in interpretation of measured spectra and proposed theoretical models related to light interaction with nonlinear optical systems. A typical quantity is the frequency-dependent nonlinear susceptibility to describe the response of the system. The theory can be applied both for isotropic and anisotropic media, and even in the case that the effective susceptibility is a linear or nonlinear combination of the primitive susceptibilities. The symmetry of the medium as well as the polarization state of the light define the symmetry properties of dispersion relations, but here we have made no assumptions about the symmetry of the system.

In the case of holomorphic nonlinear susceptibilities the KK analysis and related sum rules work much as in the case of linear optical spectroscopy. The dispersion relations and sum rules presented in this paper are valid for holomorphic nonlinear susceptibilities so that the residue terms are equal to zero, while they are nonzero for meromorphic nonlinear susceptibilities. The sum rules (16)–(19) are expected to have importance at least in degenerate four-wave mixing and self-action spectroscopies. Self-action processes and also linear processes are finding applications in the study of the optical properties of nanostructures [33,34,30], which will have various impacts in future technology. Insofar, the investigation of the optical dispersion properties of nanostructures has been concentrated mainly on their theoretical properties. Their dispersion properties provide information, which is important for instance in the optimization of nonlinear all-optical switchers [35] that are operated tuning the intensity of the laser light. Another application is related to the optimization of the signal for two-photon absorption induced fluorescence in bioassays [36], which has great importance in drug discovery, and which is based on the utilization of nanoparticles. In this latter case the dispersion study of meromorphic nonlinear susceptibility of nanoparticles is of crucial importance.

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APPENDIX

The calculation of e.g., the integral

$$I=\mathrm{P}\int_{-\infty}^{\infty}2\,\mathrm{Re}\left\{\sum_{\mathrm{Im}\{z\}>0}^{\mathrm{poles}}\mathrm{Res}\,\frac{f(z)}{z-x'}\right\}dx' \tag{A1}$$

is based on the use of the partial fraction $\mathrm{c}_{ik}/(\mathrm{z}-a_i)^k$. It is sufficient to demonstrate the calculation for one residue term because the other terms are obtained in a similar manner. Then we find out that when $k=1$, we have

$$\begin{aligned}
&\mathrm{P}\int_{-\infty}^{\infty}\mathrm{Re}\left\{\frac{c_{i1}}{a_i-x'}\right\}dx'\\
&\quad=\mathrm{P}\int_{-\infty}^{\infty}\mathrm{Re}\frac{\mathrm{Re}\{c_{i1}\}+i\ \mathrm{Im}\{c_{i1}\}}{\mathrm{Re}\{a_i\}-x'+i\ \mathrm{Im}\{a_i\}}dx'\\
&\quad=\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\{c_{i1}\}+[\mathrm{Re}\{a_i\}-x']+\mathrm{Im}\{c_{i1}\}\mathrm{Im}\{a_i\}}{(\mathrm{Re}\{a_i\}-x')^2+(\mathrm{Im}\{a_i\})^2}dx'\\
&\quad=\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\{c_{i1}\}[\mathrm{Re}\{a_i\}-x']}{(\mathrm{Re}\{a_i\}-x')^2+(\mathrm{Im}\{a_i\})^2}dx'\\
&\quad\quad+\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\{c_{i1}\}\mathrm{Im}\{a_i\}}{(\mathrm{Re}\{a_i\}-x')^2+(\mathrm{Im}\{a_i\})^2}dx'\\
&\quad=\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\{c_{i1}\}\mathrm{Im}\{a_i\}}{(\mathrm{Re}\{a_i\}-x')^2+(\mathrm{Im}\{a_i\})^2}dx'\\
&\quad=\pi\ \mathrm{Im}\{c_{i1}\}.
\end{aligned} \tag{A2}$$

The corresponding integration of the imaginary part of the residue term can be obtained by similar procedure as above.

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