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LETTER TO THE EDITOR

Sum rules for non-linear optical constants

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Abstract. Sum rules for non-linear optical constants are given, using the concept of complex contour integral and a symmetry relation imposed on non-linear optical constants.

The non-linear susceptibilities can be described as functions of several complex angular frequency variables [1]. Furthermore, they obey a dispersion relation [1] which resembles that for their linear counterpart. The dispersion relation for a non-linear susceptibility is valid provided that it is a holomorphic function of complex frequencies and it falls off quickly enough for high frequencies. The requirements are not severe and are fulfilled in most cases in non-linear optical processes. Sum rules were derived [1–4] for non-linear optical constants and the derivations for non-linear susceptibilities were based on the use of the dispersion relation and the model of the anharmonic oscillator.

In this work we give an alternative simple derivation of sum rules that does not rest on the dispersion relation and the anharmonic oscillator model. The derivation is based on the calculus exploited in the derivation of sum rules for linear optical constants in [5].

Let us assume that a non-linear optical constant (here we denote non-linear susceptibility reflectivity or refractive index as a non-linear constant) of nth order, $f^{(n)}(\hat{\omega}_1, \ldots, \hat{\omega}_n)$, is a holomorphic function with respect to complex angular frequencies $\hat{\omega}_i, i = 1, \ldots, n$, or some of them. Furthermore, we assume that $f^{(n)}$ is holomorphic in complex half-planes with respect to all variables $\hat{\omega}_i$ or some of them. According to the theory of several complex variables each variable $\hat{\omega}_i$ can be treated independently. In other words $f^{(n)}$ can be considered as a function of one complex variable with respect to certain $\hat{\omega}_i$; the other frequencies are considered as constants with respect to $\hat{\omega}_i$. It must be emphasized that the physically meaningful frequencies are the real frequencies Re $\hat{\omega}_i$.

The variety of frequencies appearing in the non-linear optical constant $f^{(n)}$ can take into account multiphonon processes, i.e. cases where several independent laser beams are incident on the non-linear material. The simplest case appears in connection with harmonic wave generation, where one laser beam is involved. As an example of a typical multiphonon process we mention the coherent anti-Stokes Raman spectra

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(CARS) [6], which are obtained using two lasers. One laser has a fixed wavelength, whereas the wavelength of the other laser is tuned. In such a case we certainly have one complex angular frequency variable. An example of harmonic wave generation is the SHG from a metal surface; SHG is observed when a high-energy laser pulse is incident on the metal [7].

Next we give a sum rule by considering a function that is defined as follows:

$$g(\hat{\omega}_1, \hat{\omega}_2, \ldots, \hat{\omega}_n) = \hat{\omega}_1^{\alpha} \hat{\omega}_2^{\beta} \cdots \hat{\omega}_n^{\nu} [f^{(n)}(\hat{\omega}_1, \hat{\omega}_2, \cdots, \hat{\omega}_n)]^N$$

where $\alpha, \beta, \ldots, \nu$ and N are positive integers. The function g is certainly holomorphic with respect to $\hat{\omega}_i$ if $f^{(n)}$ is holomorphic with respect to $\hat{\omega}_i$. We can choose the integers in such a way that $g = \mathrm{O}(|\hat{\omega}_i|^{-1-\delta})$, $\delta > 0$, as $|\hat{\omega}_i| \to \infty$. Now we can treat g as a function of one complex variable. Then, as shown in [5], we may write

$$\int_{-\infty}^{\infty} g(\omega_1, \ldots, \omega_i, \ldots, \omega_n) \, \mathrm{d}\omega_i = \lim_{R \to \infty} \left(\oint_{\mathrm{C}} g(\hat{\omega}_1, \ldots, \hat{\omega}_i, \ldots, \hat{\omega}_n) \, \mathrm{d}\hat{\omega}_i \right) - \int_{\mathrm{A}} g(\hat{\omega}_1, \ldots, \hat{\omega}_i, \ldots, \hat{\omega}_n) \, \mathrm{d}\hat{\omega}_i. \tag{1}$$

The frequency variables in the left-hand side of the equation (1) are real numbers. There are no poles inside the closed semicircle C, which is located in a complex half-plane. The arc of the closed contour C is denoted by A and its radius is given by R. According to our assumptions we can use the results of Cauchy's integral theorem and Jordan's lemma [8] to observe that the integrals on the right-hand side of (1) vanish. This means that we can write a sum rule

$$\int_{-\infty}^{\infty} g(\omega_1, \ldots, \omega_i, \ldots, \omega_n) \, \mathrm{d}\omega_i = 0. \tag{2}$$

Sum rules are practical when they are given for the spectral range $[0,\infty)$. This is accomplished by making use of the symmetry relation [5] $f^*(\omega_1, \omega_i, \ldots, \omega_n) = f(-\omega_1, \ldots, -\omega_i, \ldots, -\omega_n)$ where * denotes the complex conjugate. The symmetry relation is generally valid and does not need the support of the anharmonic oscillator model. For an example of a sum rule we consider the SHG and set $\alpha = 0$, $\beta = 0$ and $N = 1$. We then have $f(\omega, \omega) = \mathrm{Re}\ \chi^{(2)}(\omega, \omega) + \mathrm{i}\ \mathrm{Im}\ \chi^{(2)}(\omega, \omega)$. Now we can write a sum rule

$$\int_0^{\infty} \mathrm{Re}\ \chi^{(2)}(\omega, \omega) \, \mathrm{d}\omega = 0 \tag{3}$$

which was obtained also in [1], but after using the dispersion relation and the anharmonic oscillator model. The anharmonic oscillator model is not necessary for the derivation of sum rules. Such a model guarantees appropriate asymptotic fall-off of the non-linear susceptibility when $|\omega_i| \to \infty$. This asymptotic fall-off appears, regardless of the anharmonic model, always for high energies of the electromagnetic field.

Unfortunately, in many non-linear optical processes the information about the real and imaginary parts of the non-linear optical constant is hidden in the measured intensity, which is usually proportional to the modulus of the non-linear optical constant. In such cases one may try to derive sum rules for the modulus itself, as was done in recent studies [9, 10]. One possibility for resolving the real and imaginary parts is to try to calculate the phase φ of $f^{(n)}$, $f = |f^{(n)}|e^{j\varphi}$, in order to produce sum rules. In principle the calculation of phase angle is possible with the aid of the Hilbert transform [11] or Kramers–Kronig relation [12, 13]. The latter relation is widely used for the purpose of calculating linear optical constants with the aid of an intensity reflectance spectrum.

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