Sum rules for the nonlinear susceptibilities in the case of sum frequency generation

K. -E. Peiponen
Väisälä Laboratory, Department of Physics, University of Joensuu, SF-80100, Finland
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Some sum rules are given for nonlinear susceptibilities in the case of sum frequency generation. The derivation is based on the classical model of the anharmonic oscillator and the theory of several complex variables is used.

INTRODUCTION

Sum rules for physical variables can be used for testing the consistency between theory and experiment. Most applications of sum rules concern strongly interacting particles. The best known sum rule in optics is perhaps the f sum rule of classical dispersion theory. A remarkable set of sum rules for linear optical constants were given by Altarelli *et al.*[1] and also by Altarelli and Smith.[2] Some of them follow directly from various kinds of dispersion relations[3] and some can be derived with the aid of the superconvergence theorem.[1,4]

Besides the fact that the sum rules are useful for testing the validity of experimental data, they also characterize in a very simple way the nature of the optical constants. The simplest sum rule of Altarelli *et al.* for insulators can be given as follows:

$$\int_0^\infty [n(\omega)-1]d\omega=0 , \tag{1}$$

where $n(\omega)$ is the frequency-dependent real refractive index.

Sum rules similar to Eq. (1) can also be given for the powers of the complex refractive index.[2] In this paper we will present some sum rules for the nonlinear susceptibilities in the case of sum frequency generation.

I. SUM RULES

In the classical theory of nonlinear susceptibilities the starting point is the equation of motion of a classical electron oscillator with an anharmonic force. In one dimension the equation of motion is as follows:

$$\frac{d^2x}{dt^2}+\Gamma\frac{dx}{dt}+\omega_0^2x+ax^2=F . \tag{2}$$

Here F is a driving force due to the incident light interacting with the material. The parameter a describes the anharmonicity. In the classical approximation the nonlinear susceptibilities based on Eq. (2) involve essentially frequency dependences similar to those of the Lorentz oscillator, i.e., $D(\omega_i)=(\omega_0^2-\omega_i^2-i\Gamma\omega_i)^{-1}$. Then, for instance in the case of sum frequency generation, the second-order nonlinear susceptibility is proportional to

$$\chi^{(2)}(\omega=\omega_1+\omega_2)\sim D(\omega_1)D(\omega_2)D(\omega_1+\omega_2) .$$

It is obvious that $D(\omega_i)=O(\omega_i^{-2})$ as $\omega_i\to\infty$ for each ω_i. This is a property essential to the following derivation of the sum rules. Actually sum rules can be given even if $\chi^{(n)}=O(\omega_i^{-1})$ for large values of ω_i provided that only sum frequency generation is considered. In the case of difference frequency generation the nonlinear susceptibilities based on the classical approximation may simultaneously have poles in the upper and lower half of the complex frequency plane. In such cases the procedure of this paper will not be applicable for derivation of sum rules.

It is well known that the complex linear optical constants of insulators are holomorphic functions in the upper half of the complex frequency plane. Also the nonlinear susceptibilities are holomorphic functions in the case of sum frequency generation. But now we have to deal with nonlinear susceptibilities as holomorphic functions of several complex independent angular frequency variables $\hat\omega_1,\hat\omega_2,\ldots,\hat\omega_n$. The domain where $\chi^{(n)}$ is holomorphic is the "upper half plane" $\mathrm{Im}\hat\omega_1\geqq 0\wedge\mathrm{Im}\hat\omega_2\geqq 0\wedge\cdots\wedge\mathrm{Im}\hat\omega_n\geqq 0$.

The derivation of the classical Kramers-Kronig dispersion relation for linear quantities is based on the use of the Cauchy integral theorem and integral formula.[5] The same theorems hold also for a holomorphic function of several complex variables.[6] Then for instance the Cauchy integral formula reads for the nonlinear susceptibility in the case of sum frequency generation as follows:

$$\chi^{(n)}(\hat\omega'_1,\hat\omega'_2,\ldots,\hat\omega'_n)=\frac{1}{(2\pi i)^n}\int\int_A\cdots\int\frac{\chi^{(n)}(\hat\omega_1,\hat\omega_2,\ldots,\hat\omega_n)}{(\hat\omega_1-\hat\omega'_1)(\hat\omega_2-\hat\omega'_2)(\cdots)(\hat\omega_n-\hat\omega'_n)}d\hat\omega_1 d\hat\omega_2\cdots d\hat\omega_n , \tag{3}$$

where A is the integration domain. In our physical application we are interested in the poles lying on the real axes $-\infty\leqq\mathrm{Re}\hat\omega_1\leqq+\infty,\ldots,-\infty\leqq\mathrm{Re}\hat\omega_n\leqq+\infty$. We form the Cauchy principal value in a similar way as in the linear case. The poles on the real axes are excluded by deforming the integration path by a semicircular detour of infinitesimal radius. The integration and limiting processes involved are performed with respect to one variable at a time.

Smet and Smet[7] have given a relation for the second-order nonlinear susceptibility based on Eq. (3) as follows:

$$\chi^{(2)}(\omega_1',\omega_2')=-\frac{1}{\pi^2}\mathrm{P}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\chi^{(2)}(\omega_1,\omega_2)}{(\omega_1-\omega_1')(\omega_2-\omega_2')}d\omega_1 d\omega_2\ , \tag{4}$$

where P (Ref. 8) denotes the Cauchy principal value and the integral has simple poles ω_1' and ω_2' on the real axes.

We proceed by observing that also the functions $[\chi^{(n)}(\hat{\omega}_1,\hat{\omega}_2,\ldots,\hat{\omega}_n)]^m$ and $[\hat{\omega}_1\hat{\omega}_2\ldots\hat{\omega}_n]^r[\chi^{(n)}(\hat{\omega}_1,\hat{\omega}_2,\ldots,\hat{\omega}_n]^m$, where $r\leqq m$, $r=1,2,\ldots$, $m=1,2,\ldots$, are holomorphic functions satisfying the Cauchy theorem and formula of several complex variables. Thus we can write the "dispersion relations:"[9]

$$[\chi^{(n)}(\omega_1',\omega_2',\ldots,\omega_n')]^m=\frac{1}{(i\pi)^n}\mathrm{P}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\frac{[\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega_n)]^m}{(\omega_1-\omega_1')(\omega_2-\omega_2')(\cdots)(\omega_n-\omega_n')}d\omega_1 d\omega_2\cdots d\omega_n \tag{5}$$

and

$$(\omega_1'\omega_2'\ldots\omega_n')^r[\chi^{(n)}(\omega_1',\omega_2',\ldots,\omega_n')]^m=\frac{1}{(i\pi)^n}\mathrm{P}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\frac{(\omega_1\omega_2\cdots\omega_n)^r[\chi^{(n)}(\omega_1,\ldots,\omega_n)]^m}{(\omega_1-\omega_1')(\cdots)(\omega_n-\omega_n')}d\omega_1 d\omega_2\cdots d\omega_n\ . \tag{6}$$

By setting $\omega_1'=\omega_2'=\ldots=\omega_n'=0$ in Eq. (5) we obtain a similar kind of sum rule as in the linear case. The same procedure applied in Eq. (6) gives the sum rule:

$$\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}(\omega_1\omega_2\cdots\omega_n)^r[\chi^{(n)}(\omega_1,\ldots,\omega_n)]^m d\omega_1 d\omega_2\cdots d\omega_n=0$$
$$r=1,2,\ldots,\quad m=1,2,\ldots,\quad r\leqq m\ . \tag{7}$$

Actually it is enough that for some $n=j$ $\omega_j'=0$ to make the left-hand side of Eq. (6) zero. Especially if we set $r=1$, then

$$\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}[\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega_n)]^m \times d\omega_1 d\omega_2\cdots d\omega_n=0\ . \tag{8}$$

For $n=1$ Eqs. (7) and (8) yield the famous sum rules of Altarelli and Smith for linear susceptibilities.

Let us examine a sum rule for the second-order nonlinear susceptibility in a case where $r=m=1$. Then

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\chi^{(2)}(\omega_1,\omega_2)d\omega_1 d\omega_2\ . \tag{9}$$

In the case of the generation of second harmonic we have $\omega_1=\omega_2=\omega$. Then from Eq. (9) we obtain

$$\int_{-\infty}^{\infty}\chi^{(2)}(\omega,\omega)d\omega=0\ . \tag{10}$$

Further,

$$\int_0^{\infty}\mathrm{Re}\chi^{(2)}(\omega,\omega)d\omega=0\ , \tag{11}$$

where the symmetry relation $\chi^{(2)*}(\omega,\omega)=\chi^{(2)}(-\omega,-\omega)$ has been used.

II. DISCUSSION

We have presented some sum rules for nonlinear susceptibilities in the case of sum frequency generation. The derivation was based on the simple classical model of Lorentz oscillator. The conditions for the sum rules stated were that $\chi^{(n)}\sim\omega_i^{-2}$ with respect to each angular frequency ω_i as $\omega_i\to\infty$ and further that $\chi^{(n)}$ is a holomorphic function. These conditions are not very strong. In that sense one could expect that sum rules could also be given for more realistic models than that of the present paper. One could also expect further that derivation of sum rules is possible for nonlinear reflection coefficients.

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[8]The definition of the Cauchy principal value, e.g., in the case of a function of three variables, is as follows. An integrable function f which has three simple poles ω_1', ω_2', and ω_3' exists as a Cauchy principal value if the limit

$$\mathrm{P}\int_0^{\infty}\int_0^{\infty}\int_0^{\infty}f(\omega_1,\omega_2,\omega_3)d\xi=\lim_{\phi\to 0}\lim_{\beta\to 0}\lim_{\alpha\to 0}\left[\int_0^{\omega_3'-\phi}\int_0^{\omega_2'-\beta}\int_0^{\omega_1'-\alpha}f(\omega_1,\omega_2,\omega_3)d\xi+\int_{\omega_1'+\phi}^{\infty}\int_{\omega_2'+\beta}^{\infty}\int_{\omega_3'+\alpha}^{\infty}f(\omega_1,\omega_2,\omega_3)d\xi\right]$$

exists.

[9]We prefer here the notation $\chi^{(n)}(\omega_1,\omega_2,\ldots,\omega_n)$ consistent with the theory of several complex variables rather than the physically more relevant notation $\chi^{(n)}(\omega=\omega_1+\omega_2+\cdots+\omega_n)$.