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Kramers-Kronig type of dispersion relation in nonlinear optics

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Abstract. The applicability of Kramers-Kronig (K-K) relation under nearly sharp resonant transition regime in narrow-gap semiconductors has been established and consequently, a generalized dispersion relation for nonlinear optical susceptibility of a dielectric is derived. This relation can be employed in the study of nonlinear optical processes in solids as well as in plasmas over a wide frequency spectrum.

Keywords. Kramers-Kronig dispersion relation; nonlinear optical susceptibility; nonlinear refraction; nonlinear absorption.

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1. Introduction

Kramers-Kronig (K-K) dispersion relations which connect the dispersive and absorptive aspects of matter-radiation interaction processes are extremely useful in many areas of physics. In particular, the utility of K-K relations in the determination of different optical constants by reflectivity data as well as in the derivation of sum rules is well known and needs very few physically well-founded assumptions (Landau and Lifshitz 1959; Wooten 1972).

The extension of K-K relations to the nonlinear regime is regarded as a difficult and subtle problem because optical nonlinearity of a medium has been the origin of a variety of phenomena in contrast to the simple absorptive and dispersive aspects of the linear material response of the medium. Nonlinear refraction and absorption are two of the vital properties of the medium which play critical roles in the observation of optical bistability, optical phase conjugation via degenerate four-wave mixing and pulse shaping, etc. These phenomena have large technological potential in integrated optics. The investigations of such nonlinear optical effects could be much simplified if an analytical tool similar to the K-K relations is available. Unfortunately, no such relations seems to have yet been established.

The present literature reveals that there is a controversy about the applicability of K-K relations in the nonlinear regime. Flytzanis (1975) commented that these relations are applicable even in the nonlinear optical effects but are not so useful as in linear optics. On the other hand, according to Yariv (1975), these relations are invalid because of the presence of poles in both the upper and lower halves of complex ω-plane and violate the causality principle in the nonlinear regime. K-K relations were used by Miller *et al* (1981) probably for the first time in nonlinear optics under the approximation that the quantum mechanical phases of the carriers are randomized by thermalizing collisions. They developed a semi-empirical theory that fits well with the experimentally observed band-gap resonant nonlinear refraction in III-V semiconductors, viz InSb.

Keeping in view the above discussion, we have dealt with a couple of fundamental questions: (i) whether the intensity-dependent optical susceptibility (χ) can be regarded as causal or not; (ii) what would have been the form of the K-K relation for the nonlinear optical susceptibility of a crystal, that could, under band-gap resonant transition, yield nonlinear refraction (via $\chi_r^{(3)}$) in terms of the measured nonlinearity in absorption (via $\chi_i^{(3)}$); (iii) derivation of a generalized form of dispersion relation which can enable one to estimate the nonlinear refractive index simply from the knowledge of nonlinear absorption coefficient and vice versa over an arbitrarily wide photon energy spectrum in a centrosymmetric crystal.

2. Theoretical formulations

To start with, we consider the generalized form of the nth order polarization in terms of the response function as (Ducuing 1977)

$$P_\sigma^{(n)}(t)=\int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2 \ldots \int_0^\infty \mathrm{d}\tau_n R^{(n)}_{\sigma,\lambda_1,\lambda_2,\ldots,\lambda_n}(\tau_1,\tau_2,\ldots,\tau_n)E_{\lambda_1}(t-\tau_1) \times E_{\lambda_2}(t-\tau_2)\ldots E_{\lambda_n}(t-\tau_n). \tag{1}$$

Here, $R^{(n)}_{\sigma,\lambda_1,\lambda_2,\ldots,\lambda_n}$ is the response function of nth order and $E_{\lambda_1}, E_{\lambda_2}, \ldots, E_{\lambda_n}$ are the electric field amplitudes of the spatially uniform interacting waves. For an arbitrary decomposition of the electric field, one can also have

$$P_\sigma^{(n)}(t)=\varepsilon_0 \int_{-\infty}^\infty \mathrm{d}\omega_1 \int_{-\infty}^\infty \mathrm{d}\omega_2 \ldots \int_{-\infty}^\infty \mathrm{d}\omega_n\ \chi^{(n)}_{\sigma,\lambda_1,\lambda_2,\ldots,\lambda_n}(\omega_1,\omega_2,\ldots\omega_n) \times E_{\lambda_1}(\omega_1)\,E_{\lambda_2}(\omega_2)\ldots E_{\lambda_n}(\omega_n)\ \exp[i(\omega_1+\omega_2+\omega_3\ldots+\omega_n)t], \tag{2}$$

$\chi^{(n)}_{\sigma,\lambda_1,\lambda_2,\ldots,\lambda_n}$ being the nth order susceptibility tensor. This generalized expression for the nth order polarization characterizes almost all possible types of nonlinearities like nonlinear refraction and absorption (viz., $\omega; \omega; -\omega; \omega$), harmonic generation ($\omega_n = n\omega$), sum and difference frequency mixing (viz., $\omega_1=\omega_2\pm\omega_3$), etc.

A relation between the optical susceptibility and the response function can be obtained by comparing (1) and (2) whence one gets

$$\chi^{(n)}_{\sigma,\lambda_1,\lambda_2,\ldots,\lambda_n}(\omega_1,\omega_2,\ldots,\omega_n) = \int_0^\infty \mathrm{d}\tau_1 \int_0^\infty \mathrm{d}\tau_2 \ldots \int_0^\infty \mathrm{d}\tau_n R^{(n)}_{\sigma,\lambda_1,\lambda_2,\ldots,\lambda_n}(\tau_1,\tau_2,\ldots,\tau_n) \times \exp[-i(\omega_1\tau_1+\omega_2\tau_2+\ldots+\omega_n\tau_n)]. \tag{3}$$

Equation (3) clearly indicates that the properties of the reponse function $R^{(n)}$ will be reflected in the corresponding properties of $\chi^{(n)}$.

We now restrict ourselves to the area of nonlinear refraction and absorption in the semiconducting crystals duly irradiated by lasers. The total induced polarization can be expressed as

$$P(\omega)=\varepsilon_0\chi(\omega)E(\omega)=\varepsilon_0[\chi^{(1)}+\chi^{(3)}|E|^2+\ldots]\,E(\omega), \tag{4}$$

where $E(\omega)$ is the Fourier transform of the electric field $E(t)$ of the pump laser. We also assume that the medium is centrosymmetric in nature such that the nonlinear optical susceptibilities of even orders (viz., $\chi^{(2)}$, $\chi^{(4)}$, ...) vanish under parity considerations. Nonlinearity expressed by (4) can have its origin in different types of mechanisms like (i) band nonparabolicity (Khan *et al* 1980), (ii) intensity-dependent free electron-hole pair generation (Jain 1982), (iii) interband transition saturation (Wherrett and Higgins 1982a), (iv) coherent radiation-exciton interaction (Sen 1982), etc.

While addressing ourselves to the question of developing a generalized dispersion relation in nonlinear optics, we have considered $\chi(\omega)$ to be intensity-dependent and given by

$$\chi(\omega)=\frac{G_1}{X+i\gamma}\left(1+\frac{A|E|^2}{X^2+\gamma^2}\right)^{-1}. \tag{5}$$

Equation (5) can be regarded as a model representation of $\chi(\omega)$ irrespective of its origin. The symbols G_1, A, X and γ have been defined in terms of the material parameters of the crystals in table 1. It can be shown without much difficulty that the usage of table 1 leads one to obtain the correct expression for $\chi(\omega)$ for both the models (iii) and (iv) discussed in the preceding paragraph. This equation yields the various odd-ordered optical susceptibility components only if the excitation intensity $I(=\frac{1}{2}\varepsilon_0\eta_0 c|E|^2)$ is

Table 1. Definition of terms used in equation (5).

Parameter	Wherrett and Higgins (1982b)[a]	Sen (1984)[b]
G_1	$\left\lvert\frac{e\varepsilon\cdot\mathbf{p}_{cv}}{m\omega}\right\rvert^2\frac{1}{\hbar\pi^2}\int_0^\infty k^2\mathrm{d}k$	$\frac{N\mu^2}{\varepsilon_0\hbar}\lvert\psi_1(0)\rvert^2$
A	$4T_1T_2^{-1}\lvert ep_{cv}/(m\omega_g\hbar)\rvert^2$	$\mu^2\lvert\psi_1(0)\rvert^2/4\hbar^2$
X	$\omega_{cv}(k)-\omega$	$(\omega_1-\Omega_r)-\omega$
γ	T_2^{-1}	γ_r

[a]equations (12) and (19); [b]equations (17) and (18).

$\left\lvert\frac{e\varepsilon\cdot\mathbf{p}_{cv}}{m\omega}\right\rvert$ is the transition dipole moment where ε is the radiation polarization and $\mathbf{p}_{cv}$ is the momentum matrix element. m is the electron rest mass. $|\mu|$ is the magnitude of the transition dipole moment and $|\psi_1(0)|$ corresponds to the $1s$ Wannier-Mott exciton wave function. T_1 and T_2 are the population relaxation life time and dephasing time, respectively. $\omega_{cv}(k)=\omega_g+\hbar k^2/2m_r$ with ω_g being the band-gap frequency and m_r is the electron-hole pair reduced mass. ω_1 is the excitonic effect incorporated band-gap frequency, γ_r is the renormalized damping constant due to the self-energy of the excited pair state while Ω_r appears due to the Stark broadening of the band edge.

considerably small such that $A|E|^2 \ll (X^2+\gamma^2)$ whence (5) reduces to

$$\chi(\omega)=\frac{G_1}{X+i\gamma}-\frac{G_1A|E|^2}{(X^2+\gamma^2)\,(X+i\gamma)}+\;\ldots\;+\frac{(-1)^nG_1A^n|E|^{2n}}{(X^2+\gamma^2)^n(X+i\gamma)} \tag{6}$$

with $n=0,1,2,3,\ldots$ For $A|E|^2 \gtrsim (X^2+\gamma^2)$, the present model breaks down. From (6), it can be immediately recognized that the first term corresponds to $\chi^{(1)}(\omega)$ while the second one yields $\chi^{(3)}(\omega)$ and so on. The response function $R^{(2n+1)}(t)$ can be obtained by taking the inverse Fourier transform of (6) as

$$R^{(2n+1)}(t)=(-1)^n\int_{-\infty}^{+\infty}G_1A^n|E|^{2n}\exp(-i\omega t)/[(X^2+\gamma^2)^n(X+i\gamma)]\mathrm{d}\omega \tag{7}$$

where we have taken

$$\chi^{(2n+1)}(\omega)=\frac{(-1)^nG_1A^n|E|^{2n}}{(X^2+\gamma^2)^n\,(X+i\gamma)}.$$

Before proceeding further with $R^{(2n+1)}(t)$, one has to check whether $\chi^{(2n+1)}(\omega)$ is a causal function or not. For $t<0$, $R^{(2n+1)}(t)$ must be evaluated in the upper half-plane where $\exp(-i\omega t)$ is bounded. It can be easily shown that $R^{(1)}(t)$ has no singularity in the upper half of the complex ω-plane and hence it vanishes. $R^{(3)}(t)$ and other higher-order response functions have poles in both the lower and upper half-planes. But at $t<0$, $E=0$ such that the integrands disappear. This leads one to conclude that the generalized response function $R^{(2n+1)}(t)$ as well as $\chi^{(2n+1)}(t)$ satisfy the principle of causality.

We now attempt to establish the validity of the nonlinear K-K relations in obtaining nonlinear refraction from the knowledge of nonlinear absorption and vice-versa in terms of complex

$$\chi^{(2n+1)}(\omega)\;(=\chi_r^{(2n+1)}(\omega)+i\chi_i^{(2n+1)}(\omega)).$$

From (6) and (7), one can find

$$\chi_r^{(2n+1)}(\omega)|E|^{2n}=\frac{(-1)^nA^n|E|^{2n}G_1X}{(X^2+\gamma^2)^{n+1}} \tag{8a}$$

and

$$\chi_i^{(2n+1)}(\omega)|E|^{2n}=\frac{(-1)^{n+1}A^n|E|^{2n}G_1\gamma}{(X^2+\gamma^2)^{n+1}}. \tag{8b}$$

While applying K-K relations to the third-order nonlinear optical susceptibility under certain approximations, Miller *et al* (1981) found the results to agree well with experimental observations. Thus we first examine the correctness of equation (8) for $n=1$ in obtaining $\chi_r^{(3)}$ from the knowledge of $\chi_i^{(3)}$ and vice-versa. We obtain

$$\frac{1}{\pi}\,\mathrm{P.V.}\int_{-\infty}^{\infty}\frac{\chi_i^{(3)}(\omega')\mathrm{d}\omega'}{\omega'-\omega}=(\tfrac{3}{2}+\tfrac{1}{2}X^2/\gamma^2)\,\chi_r^{(3)}(\omega) \tag{9a}$$

and

$$-\frac{1}{\pi}\text{P.V.}\int_{-\infty}^{+\infty}\frac{\chi_r^{(3)}(\omega')\mathrm{d}\omega'}{\omega'-\omega}=\tfrac{1}{2}(1+X^2/\gamma^2)\,\chi_i^{(3)}(\omega). \tag{9b}$$

From equations (9), it may be noted that under near band-gap resonant transitions with $X^2 \ll \gamma^2$, approximate K-K dispersion relations can be established for the nonlinear regime and may be regarded as the mathematical expression of the approximation taken by Miller *et al* (1981). The applicability of K-K relations in nonlinear optics is thus severely restricted within a very narrow frequency spectrum ($X^2 \ll \gamma^2$). This has stimulated our interest in developing a generalized dispersion relation which can be employed in the study of nonlinear optical effects over a broad frequency spectrum.

We proceed along a line similar to that followed usually to derive K-K relations in linear optics and arrive at the generalized K-K relations applicable to nonlinear optical susceptibility of any arbitrary order. It is well known that K-K relations are obtainable by using a Cauchy integral formula under the causal limit. Consequently, these relations are restricted only to analytic response functions. Although $\chi^{(2n+1)}(\omega)|E|^{2n}$ is causal it does not satisfy the Cauchy-Reimann conditions of analyticity. It is due to this non-analyticity of $\chi^{(2n+1)}(\omega)|E|^{2n}$ that exact K-K relations could not be obtained resulting in (9). To avoid this shortcoming, we define a new function

$$\chi'^{(2n+1)}(\omega)=(X^2+\gamma^2)^n\chi^{(2n+1)}(\omega) \tag{10}$$

which is causal and satisfies the Cauchy-Reimann conditions for analyticity as well. The application of Cauchy integral formula to this function finally leads to the following relations:

$$(X'^2+\gamma^2)^n\chi_r^{(2n+1)}(\omega')=\frac{1}{\pi}\text{P.V.}\int_{-\infty}^{\infty}\frac{(X^2+\gamma^2)^n\,\chi_i^{(2n+1)}(\omega)\mathrm{d}\omega}{(\omega-\omega')} \tag{11a}$$

and

$$(X'^2+\gamma^2)^n\,\chi_i^{(2n+1)}(\omega')=-\frac{1}{\pi}\text{P.V.}\int_{-\infty}^{\infty}\frac{(X^2+\gamma^2)^n\,\chi_r^{(2n+1)}(\omega)\mathrm{d}\omega}{(\omega-\omega')} \tag{11b}$$

where X' represents X with ω being replaced by ω'. For $n=0$, (11) yields

$$\chi_r^{(1)}(\omega')=\frac{1}{\pi}\text{P.V.}\int_{-\infty}^{\infty}\frac{\chi_i^{(1)}(\omega)\mathrm{d}\omega}{(\omega-\omega')} \tag{12a}$$

and

$$\chi_i^{(1)}(\omega')=-\frac{1}{\pi}\text{P.V.}\int_{-\infty}^{\infty}\frac{\chi_r^{(1)}(\omega)\mathrm{d}\omega}{(\omega-\omega')}. \tag{12b}$$

Equations (12) are the well known K-K dispersion relations in linear optics. For $n=1$, we obtain the dispersion relations for the complex third-order susceptibility $\chi^{(3)}$ and so on for $n=2,3,\ldots$

3. Results and discussion

Equations (11) are applicable over an arbitrarily chosen frequency spectrum although the pump power has to be restricted within a certain limit such that the condition $A|E|^2 \ll (X^2+\gamma^2)$ is satisfied. The validity of (11) can be tested by applying these to the well-known phenomena of nonlinear optical effects in solids as well as in plasmas. As an example, we consider the phenomena of nonlinear refraction and absorption under low power resonant excitation regime in direct-gap semiconductors like InSb, GaAs, GaSb and InAs where the coherent radiation-exciton interaction model has been employed (Sen 1984, 1986). One finds (Sen 1984, equation (22))

$$\chi_i^{(3)}(\omega) = -\frac{N\mu^4|\psi_1(0)|^4}{\varepsilon_0\hbar^3}\,\frac{\gamma_r}{[(\omega-\omega_{gr})^2+\gamma_r^2]^2}. \tag{13a}$$

Subsequently, one may use this equation in (11a) choosing $\omega-\omega_{gr}=X$ and $\gamma_r=\gamma$ to obtain

$$\chi_r^{(3)}(\omega) = \frac{N\mu^4|\psi_1(0)|^4}{\varepsilon_0\hbar^3}\,\frac{\omega-\omega_{gr}}{[(\omega-\omega_{gr})^2+\gamma_r^2]^2}. \tag{13b}$$

Equation (13b) is exactly identical with that obtainable from (19) of Sen (1984). The results obtained by using the expression for $\chi^{(3)}(\omega)$ of the interband transition saturation model (Wherrett and Higgins 1982b) also lead one to the same conclusion and confirms the validity of (11) in the study of nonlinear dispersion in semiconducting crystals. The same equations can also be successfully employed in correlating the nonlinear magneto-absorption and magneto-refraction studied earlier (Sen 1983, 1986). Quite interestingly, the nonlinear plasma dispersion arising due to the relativistic complex oscillatory electron fluid velocity depending upon various powers of the pump intensity is another important area where (11) can be employed satisfactorily.

It may be inferred that (11) are valid for all such kinds of nonlinear optical processes for which the nonlinearity resembles with the form obtainable from (5) irrespective of the nature of its origin. However, equation (11) is restricted to the study of nonlinear refraction and absorption in solids duly irradiated by moderately low power lasers. The possibility of extending the present formulations to two-photon absorption and nonlinear wave-wave coupling will be the subject matter of a future publication.

4. Conclusions

It may be noted that the model relations represented by (11) can be regarded as the appropriate dispersion relations dealing with nonlinear optical properties of centrosymmetric solids and plasmas. This may be of immense help in the study of nonlinear refraction and absorption processes which are the origins of a class of nonlinear phenomena like optical bistability, optical phase conjugation, laser pulse shaping, etc.

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