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RELATION BETWEEN THE REAL AND IMAGINARY PART OF THE POLARIZABILITY OF A NONLINEAR RESONANT MEDIUM

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The dispersion properties of the macroscopic polarizability of a nonlinear resonant medium are considered. The Kramers-Kronig relations are extended to nonlinear resonant media with an arbitrary inhomogeneous broadening of the operative transition line and identically oriented molecules. From the derived relations it follows, in particular, that the saturation field of the antihermitian part of the polarizability is always equal to the saturation field of a single molecule and does not exceed the saturation field of the hermitian part of the polarizability. Dispersion relations are also found for resonant media with inhomogeneously broadened lines of symmetric shape.

1. The polarizability of a molecule placed in a strong monochromatic field $Ee^{i\omega t}$ of frequency ω near one of the eigenfrequencies of the molecule is given by the following well-known expression[1]:

$$\alpha_{ik}=\frac{id_i^* d_k n_0[i(\omega_0-\omega)+\tau_2^{-1}]}{h[(\omega-\omega_0)^2+\tau_2^{-2}+4\tau_1|dE|^2/\tau_2\hbar^2]}. \quad (1)$$

Here n_0 is the equilibrium difference in population density which would exist in the absence of the field; τ_1 and τ_2 are the relaxation parameters; d is a nondiagonal element of the dipole moment matrix for a resonant pair of levels. The macroscopic polarizability $\chi_{ik}(\omega, E)$ of a medium consisting of such "two-level" molecules can be obtained by averaging α_{ik} over the distribution functions with respect to ω_0 and d in an infinitesimal physical volume element.

In the present paper we confine ourselves to the investigation of the general properties of resonant media with identically oriented molecules, without specifying the form of the distribution function with respect to ω_0.

The function χ_{ik}, defined by the expression

$$\chi_{ik}(\omega, E)=\int_{-\infty}^{+\infty} f(\omega_0)\alpha_{ik}\,d\omega_0 \quad (2)$$

($f(\omega_0)$ is the normalized distribution function of the molecules over the transition frequencies) can differ substantially from α_{ik} in both its frequency dependence and field dependence. It is possible, however, to obtain several general relations for the function χ_{ik} by using the causality principle and the properties of the resonant molecule.

2. We calculate first the integral of χ_{ik} over all frequencies[1)]:

$$\int_{-\infty}^{+\infty}\chi_{ik}(\omega, E)\,d\omega=\int_{-\infty}^{+\infty}d\omega\int_{-\infty}^{+\infty}f(\omega_0)\alpha_{ik}\,d\omega_0. \quad (3)$$

We change the order of integration with respect to ω and ω_0 in the right-hand side of this equation; when this is done, the inner integral can be calculated easily and turns out to be independent of ω_0:

$$\int_{-\infty}^{+\infty}\frac{i(\omega_0-\omega)+\tau_2^{-1}}{(\omega-\omega_0)^2+\tau_2^{-2}+4\tau_1|dE|^2/\tau_2\hbar^2}\,d\omega=\frac{\pi}{r}.$$

$$r=\sqrt{1+4\tau_1\tau_2|dE|^2/\hbar^2}.$$

The integral of the distribution function over ω_0 is equal to unity. Hence, independently of the form of $f(\omega_0)$, we find that

$$\int_{-\infty}^{+\infty}\chi_{ik}(\omega, E)\,d\omega=\frac{id_i^* d_k\pi n_0}{\hbar r} \quad (4)$$

or, dividing χ_{ik} into hermitian and antihermitian parts,

$$\int_{-\infty}^{+\infty}\chi_{ik}^h\,d\omega=0, \quad (5)$$

$$\int_{-\infty}^{+\infty}\chi_{ik}^a\,d\omega=\frac{i\pi n_0 d_i^* d_k}{\hbar r}. \quad (6)$$

It is obvious that for a real d the hermitian part of the polarizability χ_{ik}^h and the antihermitian part of the polarizability χ_{ik}^a coincide respectively with the real and imaginary parts of χ_{ik}.

The formulas (4) and (5) allow us to determine the parameters of one molecule if the function $\chi_{ik}(\omega, E)$ is known, even if only approximately. We notice that the relation (4) can also be written in another form:

$$\int_{-\infty}^{+\infty}\chi_{ik}(\omega, E)\,d\omega=\frac{1}{r}\int_{-\infty}^{+\infty}\chi_{ik}(\omega, 0)\,d\omega. \quad (7)$$

This equality gives an integral relation between the linear and nonlinear polarizabilities of the resonant medium.

3. It is clear that the Kramers-Kronig relations are not valid for $\alpha_{ik}(\omega, E)$, because this function has a singular point (a pole) both in the upper and in the lower complex ω planes. These relations, of course, are also not valid for $\chi_{ik}(\omega, E)$. Nevertheless, it turns out to be possible to find the relation between the real and imaginary parts of χ_{ik}, at least for the case of identically oriented particles considered here.

[1)] The conditions of applicability of the infinite integration limits coincide, both here and in the rest of the paper, with the conditions of applicability of the two-level model for the molecules of the medium [1]. One of these conditions, in particular, is that $f(\omega_0)$ decrease sufficiently rapidly for increasing ω_0.

We now introduce a new auxiliary function $\tilde{\chi}_{ik}(\omega, \mathbf{E})$, equal to

$$\tilde{\chi}_{ik} = \frac{id_i^* d_k n_0}{\hbar} \int_{-\infty}^{+\infty} f(\omega_0) \frac{d\omega_0}{[i(\omega - \omega_0) + r/\tau_2]} \tag{8}$$

This function can be obtained from the linear polarizability by replacement of the parameter $1/\tau_2$ with the quantity r/τ_2. It is easy to see the relation between $\tilde{\chi}_{ik}$ and the true polarizability χ_{ik} namely

$$\tilde{\chi}_{ik}{}^h = \chi_{ik}{}^h, \tag{9}$$

$$\tilde{\chi}_{ik}{}^a = r\chi_{ik}{}^a. \tag{10}$$

However, according to its definition, the auxiliary polarizability has no singularities in the lower complex ω half-plane and consequently the Kramers-Kronig relations apply here:

$$\tilde{\chi}_{ik}{}^h(\omega, \mathbf{E}) = \frac{i}{\pi} \int_{-\infty}^{+\infty} \frac{\tilde{\chi}_{ik}{}^a(x, \mathbf{E})}{x - \omega} dx, \tag{11}$$

$$\tilde{\chi}_{ik}{}^a(\omega, \mathbf{E}) = \frac{i}{\pi} \int_{-\infty}^{+\infty} \frac{\tilde{\chi}_{ik}{}^h(x, \mathbf{E})}{x - \omega} dx. \tag{12}$$

Inserting (9) and (10) into these relations, we obtain the following formulas:

$$\chi_{ik}{}^h(\omega, \mathbf{E}) = \frac{i}{\pi} r \int_{-\infty}^{+\infty} \frac{\chi_{ik}{}^a(x, \mathbf{E})}{x - \omega} dx, \tag{13}$$

$$\chi_{ik}{}^a(\omega, \mathbf{E}) = \frac{i}{\pi} \frac{1}{r} \int_{-\infty}^{+\infty} \frac{\chi_{ik}{}^h(x, \mathbf{E})}{x - \omega} dx. \tag{14}$$

We recall that the coefficient r can be found from formula (7). Thus knowledge of χ_{ik}^a as a function of the frequency and of the field is sufficient both for the determination of the parameters of the molecule (see (6)) and of the hermitian part of the macroscopic polarizability χ_{ik}^h (see (13)).

We note one important property of the resultant formulas, connected with the character of the field dependences of the hermitian and antihermitian parts of the nonlinear polarizability. It follows from (13) that the saturation field for χ_{ik}^a coincides with the minimal of the saturation fields χ_{ik}^h, and for the factor 1/r the latter, obviously, is equal to the saturation field for the polarizability of one molecule. At the same time, the saturation field for χ_{ik}^h is always larger than or equal to the saturation field for the polarizability of one molecule.

Thus, the characteristic field which changes χ_{ik}^a appreciably coincides with the saturation field for the polarizability of a single molecule

$$E_{\text{н}}{}^a \sim \hbar / d\sqrt{\tau_1\tau_2} \tag{15}$$

and is less or even much less than the saturation field for χ_{ik}^h. The last possibility was demonstrated in the paper by Javan and Kelly[2] for the particular case when the inhomogeneous broadening of the working medium line is of Lorentz form.

In the rather frequent case when the saturation field of the hermitian part of the polarizability is given by

$$E_{\text{н}}{}^h \gg \hbar / d\sqrt{\tau_1\tau_2} \tag{16}$$

for fields which are not too strong, i.e., for

$$E \ll E_{\text{н}}{}^h, \tag{17}$$

we can replace the polarizability χ_{ik}^h in the expressions (12) and (13) with its linear limit,

$$\chi_{ik}{}^a(\omega, \mathbf{E}) \approx \frac{i}{\pi} \frac{1}{r} \int_{-\infty}^{+\infty} \frac{\chi_{ik}{}^h(x, 0)}{x - \omega} dx. \tag{18}$$

Next, using the Kramers-Kronig relations for $\chi_{ik}^h(\omega, 0)$ and $\chi_{ik}^a(\omega, 0)$ we obtain the relation between the antihermitian part of the nonlinear polarizability $\chi_{ik}^a(\omega, \mathbf{E})$ and its linear limit:

$$\chi_{ik}{}^a(\omega, \mathbf{E}) \approx \frac{1}{r} \chi_{ik}{}^a(\omega, 0), \tag{19}$$

4. There is a case of certain practical interest, namely when the eigenfrequency distribution function of the molecules is symmetric with respect to a certain frequency ω_p. For such $f(\omega_0)$, it follows directly from the definition of $\chi_{ik}(\omega, \mathbf{E})$ that

$$\chi_{ik}(\omega - \omega_p, \mathbf{E}) = -\chi_{ki}{}^*(\omega_p - \omega, \mathbf{E}). \tag{20}$$

Separating the hermitian and antihermitian parts of this equation, we find that χ_{ik}^a and χ_{ik}^h are respectively an even and odd function of the frequency variance, $\Omega = \omega - \omega_p$.

It is possible now to derive for $\chi_{ik}(\Omega, \mathbf{E})$ several new relations which are analogous to the formulas for $\epsilon(\Omega)$ for linear media (see, for example,[3], Sec. 62). It follows from the symmetry relation (20) and from formulas (13) and (14) that

$$\chi_{ik}{}^h(\Omega, \mathbf{E}) = \frac{2i\Omega}{\pi} r \int_0^{\infty} \frac{\chi_{ik}{}^a(x, \mathbf{E})}{x^2 - \Omega^2} dx, \tag{21}$$

$$\chi_{ik}{}^a(\Omega, \mathbf{E}) = \frac{2i}{\pi} \frac{1}{r} \int_0^{\infty} \frac{x\chi_{ik}{}^h(x, \mathbf{E})}{x^2 - \Omega^2} dx. \tag{22}$$

We shall derive a formula expressing the values of $\chi_{ik}(\Omega)$ on the imaginary axis in terms of the values of $\chi_{ik}^h(\Omega)$ on the real axis. For this purpose we first relate the values of $\chi_{ik}(i\Omega)$ to the values of the auxiliary function $\tilde{\chi}_{ik}(i\Omega)$, Comparing the corresponding expressions (1) and (8) with imaginary arguments, we find that

$$\chi_{ik}(i\Omega) = \frac{1+r}{2r} \tilde{\chi}_{ik}(i\Omega) + \frac{1-r}{2r} \tilde{\chi}_{ik}(-i\Omega), \tag{23}$$

where $r = \sqrt{1 + 4\tau_1\tau_2 |d\mathbf{E}|^2/\hbar^2}$, as before.

Changing Ω into Ω in this equation, we obtain

$$\chi_{ik}(-i\Omega) = \frac{1-r}{2r} \tilde{\chi}_{ik}(i\Omega) + \frac{1+r}{2r} \tilde{\chi}_{ik}(-i\Omega). \tag{24}$$

It is easy to eliminate $\tilde{\chi}_{ik}(i\Omega)$ from these relations. Then for $\tilde{\chi}_{ik}(-i\Omega)$ we have

$$2\tilde{\chi}_{ik}(-i\Omega) = (1 + r)\chi_{ik}(-i\Omega) - (1 - r)\chi_{ik}(i\Omega). \tag{25}$$

On the other hand, $\tilde{\chi}_{ik}(-i\Omega)$ can be obtained by calculating the integral

$$\int \frac{x\tilde{\chi}_{ik}(x)}{x^2 + \Omega^2} dx,$$

over a contour consisting of the real axis and an infinite half-circle in the lower half-plane. In view of the analyticity of $\tilde{\chi}_{ik}$ in the lower half-plane, this integral is equal to the residue of the integrand at the point

$x = -i\Omega$, so that we obtain

$$\int_{-\infty}^{+\infty} \frac{x\tilde{\chi}_{ik}(x)}{x^2+\Omega^2}\,dx = -i\pi\tilde{\chi}_{ik}(-i\Omega).$$

On the left-hand side of the equation, the antihermitian component of the integrand does not contribute to the integral because of its oddness. Thus

$$\tilde{\chi}_{ik}(-i\Omega) = \frac{2i}{\pi}\int_{0}^{\infty} \frac{x\tilde{\chi}_{ik}{}^{h}(x)}{x^2+\Omega^2}\,dx.$$

Replacing $\tilde{\chi}^{h}_{ik}$ by χ^{h}_{ik} (see (9)) and expressing $\tilde{\chi}_{ik}(-i\Omega)$ by $\chi_{ik}(\pm i\Omega)$ (see (25)), we finally obtain

$$(1+r)\chi_{ik}(-i\Omega) - (1-r)\chi_{ik}(i\Omega) = \frac{4i}{\pi}\int_{0}^{\infty} \frac{x\chi_{ik}{}^{h}(x)}{x^2+\Omega^2}\,dx \qquad (26)$$

(Ω is a real positive number).

As a simple corollary of the above result, we have the integral relation of the form

$$(1+r)\int_{0}^{\infty} \chi_{ik}(-i\Omega, \mathrm{E})\,d\Omega = (1-r)\int_{0}^{\infty} \chi_{ik}(i\Omega, \mathrm{E})\,d\Omega. \qquad (27)$$

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[1] V. M. Faĭn and Ya. I. Khanin, Kvantovaya Radiofizika (Quantum Radio-Physics), Sov. Radio, 1965.

[2] A. Javan and P. L. Kelley, IEEE QE-2, No. 9, 470 (1966).

[3] L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Fizmatgiz, 1959 (English Trans. Addison-Wesley, Reading, Mass., 1960).

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244