GENERAL PROPERTIES OF AVERAGED POLARIZABILITIES IN NONLINEAR QUASI-OPTICS

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General properties of averaged susceptibilities in the opposed-wave mode are considered for a polarizability that depends on a squared field modulus. The differential relation between the forward and backward wave susceptibilities is determined. The problem of the structure of stationary one-dimensional opposed waves in a medium with arbitrary susceptibility $\chi(|E|^2)$ is reduced to quadratures with the aid of the obtained relationships. It is shown that in this case the direction of polarization of the opposed waves depends on the coordinate and the polarizations of both waves rotate in one direction.

1. INTRODUCTION

THE quasi-optical approximation that is customary in the optics of nonlinear media is applicable to the case when the characteristic length of field variation with respect to one variable (not necessarily Cartesian) is of the order of the wave-length and the field varies much more slowly with respect to the remaining variables. The Maxwell equations are reduced here to equations for slowly varying complex opposed-wave amplitudes $C_{1,2}$ and $P_{1,2}$ that are related to field $\mathbf{E}$ and polarization $\mathbf{P}$ by

$$\mathbf{E} = \mathbf{C}_1 e^{ikz} + \mathbf{C}_2 e^{-ikz}, \quad \mathbf{P} = \mathbf{P}_1 e^{ikz} + \mathbf{P}_2 e^{-ikz}. \tag{1}$$

The dependence on the "fast" coordinate is represented here by the factors $e^{\pm ikz}$ (the field and the dipole moment are assumed to be quasi-monochromatic and the factor $e^{i\omega t}$ is dropped).

Under these conditions the material equation

$$\mathbf{P} = \chi \mathbf{E}$$

is replaced by the system

$$\mathbf{P}_1 = \langle\chi\rangle \mathbf{C}_1 + \langle\chi e^{-2ikz}\rangle \mathbf{C}_2,$$

$$\mathbf{P}_2 = \langle\chi\rangle \mathbf{C}_2 + \langle\chi e^{2ikz}\rangle \mathbf{C}_1 \tag{2}$$

(angle brackets denote averaging over z for the period $2\pi/k$). Thus, instead of the nonlinear susceptibility, we must find, generally speaking, three functions relating the amplitudes of the opposed-wave field and the polarizations, while the dependence of the latter on $\mathbf{C}_1$ and $\mathbf{C}_2$ can be even more involved than the dependence of χ on $\mathbf{E}$. The nonlinear relationship between the opposed waves obviously makes it difficult to study the field structure, while such a study is hardly feasible when the coefficients of $\mathbf{C}_1$ and $\mathbf{C}_2$ are arbitrary.

In the present work we note the fact that the dependence of averaged susceptibilities on $\mathbf{C}_1$ and $\mathbf{C}_2$ cannot be arbitrary. This has been demonstrated for the case of an isotropic medium for which χ depends on the squared field modulus $\mathbf{E}$: $\chi = \chi(|\mathbf{E}|^2)$.

It is determined that the single fact of χ depending on the real quadratic form alone gives rise to the general relations between $\langle\chi\rangle$ and $\langle\chi e^{\mp 2ikz}\rangle$. These relations allow us to simplify the procedure of finding the sought functions. Moreover the obtained relationship between the opposed-wave susceptibilities simplifies in many cases the solution of nonlinear equations for the field. As an example, the obtained relationships were used to reduce to quadratures the problem of the structure of one-dimensional stationary opposed waves in a medium with arbitrary $\chi(|\mathbf{E}|^2)$ which until now could be solved only for certain special cases[1-4].

2. RELATIONS BETWEEN DIRECT AND MIXED SUSCEPTIBILITIES

We first introduce some designations: $m_{1,2} = |\mathbf{C}_{1,2}|^2$; $w = m_1 + m_2 = |\mathbf{C}_1|^2 + |\mathbf{C}_2|^2$ is the average field energy density; $u = (\mathbf{C}_1, \mathbf{C}_2^*)$; $v = |u|^2 = |(\mathbf{C}_1\mathbf{C}_2^*)|^2$ is the reciprocal energy of opposed waves.

The squared field modulus has the following form in the new notation:

$$|\mathbf{E}|^2 = w + ue^{2ikz} + u^* e^{-2ikz}.$$

The definition of the averaging operation shows directly that the averaging result does not depend on a change of origin of the "fast" variable z. It follows that the "direct" susceptibility $\langle\chi\rangle$ and the normalized "mixed" susceptibilities $\langle\chi e^{-2ikz}\rangle/u$ and $\langle\chi e^{+2ikz}\rangle/u^*$ are functions only of w and v, since there is such an origin of the averaging variable for which the result is independent of the phase of u.

The result of averaging does not change for the above functions if the averaging variable z is replaced by $-z$ with a simultaneous substitution of $u \rightleftarrows u^*$. On the other hand the application of this operation to $\langle\chi e^{-2ikz}\rangle/u$ transforms it into the function $\langle\chi e^{2ikz}\rangle/u^*$ which indicates their identity.

Consequently we reduced the three coefficients of vectors $\mathbf{C}_1$ and $\mathbf{C}_2$ in (2) to two functions of w and v: the direct susceptibility

$$a(w, v) = \langle\chi\rangle \tag{3}$$

and the normalized mixed susceptibility

$$b(w, v) = \langle\chi e^{-2ikz}\rangle / u. \tag{3'}$$

We now show that also these two functions are not independent. For this purpose we differentiate the direct susceptibility with respect to u. From the definition of v it follows that

$$\frac{\partial a}{\partial u} = u^* \frac{\partial a}{\partial v};$$

on the other hand

$$\frac{\partial a}{\partial u} = \frac{\partial \langle \chi \rangle}{\partial u}.$$

The derivative with respect to u can obviously be placed inside the averaging brackets; then

$$\left\langle \frac{\partial \chi}{\partial u} \right\rangle = \left\langle \frac{\partial \chi}{\partial |\mathbf{E}|^2} \frac{\partial |\mathbf{E}|^2}{\partial u} \right\rangle = \left\langle \frac{\partial \chi}{\partial |\mathbf{E}|^2} e^{2ikz} \right\rangle$$

Furthermore, substituting differentiation with respect to $|\mathbf{E}|^2$ by the derivative with respect to w and removing differentiation with respect to w outside the averaging brackets we obtain

$$\frac{\partial \langle \chi \rangle}{\partial u} = \frac{\partial}{\partial w} \langle \chi e^{2ikz} \rangle.$$

Hence considering the definition of a mixed susceptibility (3′) we find

$$\frac{\partial a}{\partial u} = u^* \frac{\partial b}{\partial w}.$$

Comparing this relation with the identity for $\partial a/\partial v$ we obtain

$$\partial a / \partial v = \partial b / \partial w. \tag{4}$$

In a similar manner, differentiating a with respect to w we obtain the second relation

$$\partial a / \partial w = \partial (vb) / \partial v. \tag{5}$$

Equations (4) and (5) are the sought relationships between the direct and mixed susceptibilities.

We note that the dependence of χ on $|\mathbf{E}|^2$ is the only significant consideration employed in the derivation of these formulas.

3. RELATION BETWEEN TRAVELING-WAVE AND OPPOSED-WAVE SUSCEPTIBILITIES

Equations (4) and (5) are sufficient for the derivation of the relationship between the traveling-wave susceptibility (or orthogonal opposed-wave) $\chi(w)$ and the averaged susceptibilities a(w, v) and b(w, v) of arbitrary opposed waves. We first note that relations (4) and (5) considered as equations in terms of the functions a and b can be easily provided with boundary conditions. In the limit as $v \to 0$ (the traveling wave or the polarizations of opposed waves are perpendicular) the susceptibility $\chi(|\mathbf{E}|^2)$ does not depend on the "fast" variable and obviously coincides with the direct susceptibility, i.e.,

$$a(w, 0) = \chi(w). \tag{6}$$

The corresponding condition for the mixed susceptibility follows from (5) and (6)

$$b(w, 0) = d\chi(w) / dw. \tag{7}$$

We take the Fourier transforms with respect to the energy w in (4)–(7). Then (4) and (5) turn out to be ordinary differential Bessel equations with respect to the variable v for $\bar{a}(s, v)$ and $\bar{b}(s, v)$ ($\bar{a}$ and $\bar{b}$ are Fourier images of a and b in terms of w) with the boundary conditions

$$\bar{a}(s, 0) = \bar{\chi}(s), \qquad \bar{b}(s, 0) = is\bar{\chi}(s),$$

where $\bar{\chi}(s)$ is the Fourier image of $\chi(w)$.

Solving the corresponding Bessel equation we finally arrive at the following formulas for Fourier images of the direct and mixed susceptibilities and $\bar{\chi}(s)$[1)]:

$$\bar{a}(s, v) = J_0(2s\sqrt{v})\bar{\chi}(s), \tag{8}$$

$$\bar{b}(s, v) = \frac{i}{\sqrt{v}} J_1(2s\sqrt{v})\bar{\chi}(s). \tag{9}$$

The obtained relations can in a number of cases significantly simplify the procedure of finding the direct and mixed susceptibilities since they allow us to use the Fourier transformation in place of the averaging operation.

An important consequence of (4) and (5) is the possibility to introduce potential functions Y(w, v) and K(w, v) which yield the direct and mixed susceptibilities by simple differentiation:

$$a = \frac{\partial Y}{\partial v}, \quad vb = \frac{\partial Y}{\partial w}; \tag{10}$$

$$a = \frac{\partial K}{\partial w}, \quad b = \frac{\partial K}{\partial v}. \tag{11}$$

These formulas, together with the definition of a and b, lead directly to expressions for Y and K in terms of $\chi(|\mathbf{E}|^2)$:

$$K = \left\langle \int_0^{|\mathbf{E}|^2} \chi(\xi)\, d\xi \right\rangle, \tag{12}$$

$$Y = \frac{v}{u} \left\langle e^{-2ikz} \int_0^{|\mathbf{E}|^2} \chi(\xi)\, d\xi \right\rangle. \tag{13}$$

Thus the two averaging operations necessary to obtain a and b can be replaced by one to derive K or Y with the subsequent differentiation with respect to the energy and reciprocal energy.

This result does not exhaust the role of potential functions. We show below that the quantity Re Y(w, v) is conserved in the interaction of opposed waves.

4. CONSERVATION LAWS FOR ONE-DIMENSIONAL STATIONARY WAVES

We use the obtained result to study the structure of one-dimensional monochromatic opposed waves in an isotropic medium whose polarizability depends on $|\mathbf{E}|^2$. Using the same averaging method as in[1-4] we readily obtain equations of complex amplitudes $C_{1,2}$:

$$\frac{dC_1}{dz} = aC_1 + ubC_2,$$

$$\frac{dC_2}{dz} = -aC_2 - u^* bC_1. \tag{14}$$

For the sake of simplicity we use a new χ, from now on equal to

$$\chi = \frac{2\pi i k}{\varepsilon} \chi_{\text{old}}$$

and correspondingly

$$a = \frac{2\pi i k}{\varepsilon} a_{\text{old}} \qquad b = \frac{2\pi i k}{\varepsilon} b_{\text{old}}$$

As noted in the introduction, equations involving

1)It is of interest to note that the Kramers-Kronig relations with respect to the variable s hold for $\bar{\chi}(s)$.

opposed-wave amplitudes were discussed in the literature for two particular relationships of $\chi(|\mathbf{E}|^2)$ and for such media as have the polarization of the vector $\mathbf{E}$ and consequently of the vectors $\mathbf{C}_{1,2}$ fixed in space. We can readily see that $\mathbf{C}_{1,2}(z)$ with a corresponding χ found in the above papers are particular solutions of (14). The set of solutions of (14) is significantly richer since there are possible solutions for the isotropic medium where the polarization plane of $\mathbf{C}$ depends on z.

The system (14) comprises eight nonlinear first-order equations and cannot be solved for arbitrary coefficients of $\mathbf{C}_1$ and $\mathbf{C}_2$. To some extent this is the reason for the comparatively particular nature of the solutions obtained in[1-4]. As noted above however the dependence of a and b on $\mathbf{C}_1$ and $\mathbf{C}_2$ is not arbitrary. Furthermore, the determined properties of the averaged susceptibilities are sufficient to find the integrals of (14).

Adding the right-hand vector product of the first equation by $\mathbf{C}_2$ to the left-hand vector product of the second by $\mathbf{C}_1$ we obtain

$$d[\mathbf{C}_1\mathbf{C}_2]/dz = 0.$$

This results in the first invariant for the field:

$$[\mathbf{C}_1\mathbf{C}_2] = \mathbf{A}, \tag{15}$$

where $\mathbf{A}$ is a constant vector.

We show that the second conservation law for the field is the relation

$$\operatorname{Re} Y(w, v) = B = \text{const}. \tag{16}$$

For this purpose we first derive an equation for the energy density w and reciprocal energy v.

We take the scalar product of the first equation in (14) by $\mathbf{C}_1^*$ and add it to its complex conjugate. As a result we obtain an equation for m_1:

$$\frac{dm_1}{dz} = 2m_1 \operatorname{Re} a + 2v \operatorname{Re} b; \tag{17}$$

Similarly for m_2:

$$\frac{dm_2}{dz} = -2m_2 \operatorname{Re} a - 2v \operatorname{Re} b. \tag{18}$$

Using the definition of u and the identity

$$\frac{du}{dz} = \mathbf{C}_2^* \frac{d\mathbf{C}_1}{dz} + \mathbf{C}_1 \frac{d\mathbf{C}_2^*}{dz},$$

we readily obtain the equation for u:

$$\frac{du}{dz} = -u(a - a^* + m_2 b - m_1 b^*),$$

whence follows the relation for v:

$$\frac{dv}{dz} = 2v(m_2 - m_1) \operatorname{Re} b, \tag{19}$$

and from (17) and (18) an expression for w:

$$\frac{dw}{dz} = 2(m_1 - m_2) \operatorname{Re} a. \tag{20}$$

From the equations for v and w we see that

$$v \operatorname{Re} b\,dw + \operatorname{Re} a\,dv = 0,$$

Finally, using the properties of the potential function (10) we arrive at the sought conservation law (16).

Thus two opposed monochromatic waves in an isotropic weakly nonlinear medium can vary the energy density, mutual orientation, and phase so as to conserve the quantities

$$B = v \operatorname{Re}\left(\frac{1}{u}\left\langle e^{-2ikz} \int_0^{|\mathbf{E}|^2} \chi(\xi)\, d\xi \right\rangle\right),$$

$$\mathbf{A} = [\mathbf{C}_1\mathbf{C}_2].$$

The last invariant leads to a relation for m_1, m_2, and v of the form

$$m_1 m_2 - v = \text{const}. \tag{21}$$

The invariants B and $\mathbf{A}$, constituting the first two integrals of the equation system for m_1, m_2, and v, allow us in principle to reduce to quadratures the problem of finding $m_{1,2}(z)$ and $v(z)$ for any $\chi(|\mathbf{E}|^2)$. In turn, knowing $m_{1,2}$ and v we can determine all the characteristics of vectors $\mathbf{C}_1$ and $\mathbf{C}_2$. In particular, in the case of a purely active medium and linearly polarized opposed waves (real vectors $\mathbf{C}_1$ and $\mathbf{C}_2$) we can easily determine the direction of their polarization.

According to (15) the sine of the angle φ between vectors $\mathbf{C}_1$ and $\mathbf{C}_2$ is inversely proportional to the square root of the product of their amplitudes, i.e.,

$$\sin^2\varphi = A^2/m_1 m_2. \tag{22}$$

The equation for angular velocity of rotation of $\mathbf{C}_1$ vector poarization is obtained from the first equation of (14) after vector multiplication by $\mathbf{C}_1$; it has the form

$$m_1 \frac{d\varphi_1}{dz} = -\sqrt{v}\, bA. \tag{23}$$

Here φ_1 is the angle of rotation of vector $\mathbf{C}_1$ measured clockwise for the chosen direction of the z axis. Similarly for the rotation of vector $\mathbf{C}_2$ we have

$$m_2 \frac{d\varphi_2}{dz} = -\sqrt{v}\, bA. \tag{24}$$

It is of interest to note that the opposed-wave polarization rotates in one direction only. This effect is particularly clearly reflected in the rotation of the bisector of the angle between vectors $\mathbf{C}_1$ and $\mathbf{C}_2$. The variation of this average direction of polarizations is described by the equation

$$\frac{d\theta}{dz} = -\sqrt{v}\, bA \frac{w}{2(v + A^2)}, \tag{25}$$

where $\theta = (\varphi_1 + \varphi_2)/2$. Hence it follows that any symmetric system 2L long with symmetric distribution of energy w(z) and reciprocal energy v(z) produces a "curling" of the mean direction of polarization to one side through the angle

$$\theta = -A \int_{-L}^{+L} \frac{\sqrt{v}\, wb}{2(v + A^2)}\, dz. \tag{26}$$

We evaluate the order of magnitude of the angular velocity of rotation of vectors C). The quantity mb is proportional to the second spatial harmonic of χ and is of the order of a under strong saturation.[2)] In turn, a is of the order of $\frac{1}{2}d \ln m/dz$. Thus under optimal conditions (strong saturation, $m_1 \sim m_2$, $\varphi = \varphi_1 - \varphi_2 \sim \pi/4$) the angle of rotation of $\mathbf{C}_1$ along the length L

[2)]It can be shown that the ratio of the second harmonic of χ to the fundamental under strong saturation is in the gaseous medium of the order of the ratio of the "natural" line width to the Doppler line width.

is

$$\varphi_1(L) - \varphi_1(0) \sim \frac{1}{4} \ln \frac{m(L)}{m(0)}.$$

For a solid with a length of 40 cm, $\varphi_1(L) - \varphi_1(0) \sim 180^\circ$. The large magnitude of the effect offers a promise of practical utilization.

5. CONCLUSION

Equations (4) and (5) can be generalized also to anisotropic media. In this case they are valid for each component of the tensor χ_{ik}. The fact that the latter depends on the quadratic form $E_m \alpha_{mn} E_n^*$ rather than on $|E|^2$ merely calls for a suitable reevaluation of w and v.

Of interest is the derivation of analogous relations for the interaction of waves with significantly different frequencies and wave numbers.

Although (4) and (5) are obtained for the quadratic dependence of χ on the field, similar relations undoubtedly exist also in the case of other types of nonlinearities.

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