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INSTABILITY OF SELF-FOCUSING OF LIGHT

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It is shown that self-focused light beams that are uniform along the propagation direction are unstable in a nonlinear transparent medium. It is also shown that the superposition of two monochromatic light waves is unstable.

1. INTRODUCTION

As is well known[1-3], self-focusing of light can occur in some nonlinear media; stationary trapping of light in the form of a plane or cylindrical beam of constant cross section is possible in these media. However, as shown by Bespalov and Talanov[4], in the same media a plane monochromatic light wave is unstable and has a tendency to break up into individual light beams. This raises the natural question—is a self-focused (plane or cyclindrical) beam of light stable?

The present paper is devoted to a proof that self-trapping of light in the form of a plane or cylindrical beam of constant cross section is unstable to small perturbations. We actually formulate the question in a more general form. In a nonlinear medium, we consider a near-monochromatic light field the envelope of which moves without shape distortion with a velocity close to the group velocity at the carrier frequency. We call such a field the stationary envelope wave. The self-focused beams whose envelope depends only on the variables perpendicular to the wave propagation direction are particular cases of such waves; stationary waves whose envelope depends on the longitudinal variable are also possible. Stationary waves whose envelope depends only on the longitudinal variable were considered by Ostrovskiĭ[5]. We note that stationary envelope waves can exist not only in those media where self-focusing is possible, but also in arbitrary transparent nonlinear media with dispersion.

We investigate in this paper the stability of stationary envelope waves. The investigation can be carried through to conclusion for one class of envelope waves—namely, for waves whose phase is constant in all of space. Plane and cylindrical self-focusing beams belong just to this class. To investigate the stability of waves of this type we shall use a variational method, which makes it possible to deduce the instability of the wave without knowing the explicit expression for its envelope. It becomes possible to prove here that all the stationary envelope waves with constant phase are unstable in media in which self-focusing is possible. It is interesting that in defocusing media stationary envelope waves can also be unstable. This fact is connected with the more general fact of instability of a biharmonic field, i.e., a superposition of two monochromatic waves which we shall show to take place in arbitrary nonlinear media.

2. FUNDAMENTAL EQUATIONS

Let us consider the propagation of light in an isotropic transparent medium with cubic nonlinearity, described by a dielectric constant in the form $\epsilon = \epsilon_0(\omega) + \epsilon_1|\mathbf{E}|^2$. For simplicity we shall assume the light to be plane-polarized. In addition, we confine ourselves to a case in which the light dispersion law $\omega(\mathrm{k})$ is a function that is convex downward ($\omega''(\mathrm{k}) > 0$). Assume that a wave with an average wave number k_0 propagates in such a medium in the x direction. For the complex envelope of the field-intensity vector we have the equation (see[6])

$$i\left(\frac{\partial E}{\partial t}+v_{\mathrm{gr}}\frac{\partial E}{\partial x}\right)+\frac{1}{2}\omega''(k_0)\frac{\partial^2 E}{\partial x^2}+\frac{v_{\mathrm{gr}}}{2k_0}\Delta_\perp E = q|E|^2E. \tag{1}$$

Here

$$v_{\mathrm{gr}}=\frac{\partial\omega}{\partial k}\Big|_{k=k_0},\quad q=-\frac{\varepsilon_1\omega^2}{(\omega^2\varepsilon_0(\omega))'}\Big|_{k=k_0}.$$

Self-focusing is possible in the medium if $\epsilon_1 > 0$ and accordingly $q < 0$.

We introduce new variables

$$\xi_1=(x-v_{\mathrm{gr}}t)\sqrt{\frac{1}{\omega''(k_0)}},\quad \xi_2=y\sqrt{\frac{k_0}{v_{\mathrm{gr}}}},\quad \xi_3=z\sqrt{\frac{k_0}{v_{\mathrm{gr}}}}.$$

We obtain

$$i\frac{\partial E}{\partial t}+\frac{1}{2}\Delta E=q|E|^2E,$$
$$\Delta=\frac{\partial^2}{\partial\xi_1^2}+\frac{\partial^2}{\partial\xi_2^2}+\frac{\partial^2}{\partial\xi_3^2}. \tag{2}$$

By introducing $E = Ae^{i\Phi}$ we transform (2) into the system

$$\frac{\partial}{\partial t}A^2+\operatorname{div}A^2\nabla\Phi=0,\quad \frac{\partial\Phi}{\partial t}+\frac{1}{2}(\nabla\Phi)^2=-qA^2+\frac{\Delta A}{2A}. \tag{3}$$

The stationary envelope waves constitute the solutions of Eq. (2) in the form

$$E=E_0(\xi-\mathbf{v}t)\exp\{-i(s+{}^1\!/_2v^2)t+i\mathbf{v}\xi\}. \tag{4}$$

We have for them the equation

$$sE_0+{}^1\!/_2\Delta E_0=q|E_0|^2E_0, \tag{5}$$

or, writing $E_0 = A_0\exp(i\Phi_0)$ —a system of equations

$$\operatorname{div}A_0^2\nabla\Phi=0,\quad \frac{1}{2}(\nabla\Phi_0)^2=s-qA_0^2+\frac{1}{2A_0}\Delta A_0.$$

We note that Eq. (1) is written with a certain degree of arbitrariness, namely, the choice of the carrier wave number k_0 is arbitrary. To exclude this arbitrariness, we introduce the additional requirement that the phase Φ be bounded in all of space. We then have for the function Φ the representation $\Phi = -(\boldsymbol{\xi}\cdot\mathbf{v}) + \tilde{\Phi}$, where $\tilde{\Phi}$ is bounded.

The class of solutions of interest to us, with constant phase, is determined by the condition Φ = const. Obviously, the function A_0 then satisfies the equation

$$\tfrac{1}{2}\Delta A_0 + sA_0 = qA_0^3, \qquad (6)$$

and the propagation velocity is v = 0, so that waves of this type propagate with a velocity equal to the group velocity at the carrier frequency.

The simplest solution of (5) is the constant $E_0 = \sqrt{s/q}$. This solution is a plane wave of finite amplitude. It is therefore possible to treat s as a frequency shift due to the nonlinear interaction. We call attention to the fact that Eq. (2) coincides in form with the equation for the Heisenberg operators, describing in the classical wave limit a weakly non-ideal Bose gas [7]. A plane wave of finite amplitude is the analog of a Bose condensate. Let us consider small perturbations against the background of a plane wave:

$$A = E_0 + \delta A e^{-i\Omega t + i\mathbf{k}\boldsymbol{\xi}},\ \Phi = -st + \delta\Phi e^{-i\Omega t + i\mathbf{k}\boldsymbol{\xi}}.$$

Substitution in (3) and linearization yield

$$\Omega^2 = q|E_0|^2k^2 + k^4/4. \qquad (7)$$

Formula (7) coincides with the known Bogolyubov formula for the spectrum of a Bose gas [8]. When $q < 0$ the plane wave experiences an instability [4] that is perfectly analogous to the instability of the condensate in a Bose gas with attraction.

Let us consider those solutions of (6) which depend only on one variable

$$\xi = \alpha_1\xi_1 + \alpha_2\xi_2 + \alpha_3\xi_3 \quad (\alpha_1^2 + \alpha_2^2 + \alpha_3^2 = 1).$$

They satisfy the equation

$$\tfrac{1}{2}d^2A/d\xi^2 + sA_0 = qA_0^3, \qquad (8)$$

which coincides formally with the equation of motion of the particle in a field with a potential

$$U(A_0) = sA_0^2 - \tfrac{1}{4}qA_0^4.$$

The character of the solution depends on the value of the energy integral of (8)

$$\mathscr{E} = \tfrac{1}{2}(dA_0/d\xi)^2 + U(A_0).$$

We consider first solutions for which $s < 0$. Obviously, they can exist only when $q < 0$. The energy integral for them lies in the range $-s^2/q < \mathscr{E} < \infty$. When $\mathscr{E} = -s^2/|q|$, the solution goes over into a plane wave of finite amplitude, and when $\mathscr{E} > -s^2/q$ it is a periodic modulation against the background of a plane wave. With increasing $\mathscr{E}$, the depth and period of the modulation increase, and when $\mathscr{E} = 0$ the period becomes infinite. In this case the solution takes the form

$$A_0 = \sqrt{\frac{2s}{q}}\frac{1}{\operatorname{ch}\sqrt{2s}\,\xi}. \qquad (9)$$

If $\xi \perp x$, then this solution is a plane self-focused beam [2,3], and if $\xi \parallel x$, this is an elongated wave packet [5]; when $\mathscr{E} > 0$, the solution remains periodic and the period decreases with increasing $\mathscr{E}$. The amplitude of the wave goes through zero in these cases.

We now consider the case $s > 0$. In this case the solution is a periodic function, which goes over for sufficiently small $\mathscr{E}$ into a purely harmonic solution:

$$A(\xi) = \text{const}\cdot\cos\sqrt{2s}\,\xi.$$

Thus, this solution is the nonlinear analog of a superposition of two plane waves with identical amplitudes and opposite wave vectors.

The behavior of the solution with increasing $\mathscr{E}$ depends on the sign of q. When $q > 0$, periodic solutions exist for arbitrarily large $\mathscr{E}$, and their period decreases with increasing $\mathscr{E}$. When $q > 0$, the period decreases with increasing $\mathscr{E}$, and V becomes infinite when $\mathscr{E} = s^2/2q$. In this case we obtain the purely aperiodic solution

$$A(\xi) = \sqrt{s/q}\,\operatorname{th}\sqrt{s}\,\xi. \qquad (10)$$

In analogy with the one-dimensional solution, there can exist also cylindrically-symmetrical and spherically-symmetrical solutions of (6). The cylindrically-symmetrical solution corresponds to a cylindrical self-focusing beam was obtained by Chiao, Garmire, and Townes [3].

3. INSTABILITY OF ENVELOPE WAVES WITH CONSTANT PHASE

We consider the stability of stationary envelope waves with constant phase. We seek the solution of (3) in the form

$$\Phi = -st + \delta\Phi e^{-i\Omega t},\quad A = A_0 + \delta A e^{-i\Omega t}. \qquad (11)$$

Linearizing in the small quantities $\delta\Phi$ and δA, and eliminating $\delta\Phi$, we arrive at the eigenvalue equation

$$\frac{1}{2A_0}\operatorname{div}A_0^2\nabla\frac{1}{A_0}\left[\frac{1}{2}\Delta\delta A + s\delta A - 3qA_0^2\delta A\right] = \Omega^2\delta A. \qquad (12)$$

We introduce the operators

$$L_1 = -\frac{1}{2A_0}\operatorname{div}A_0^2\nabla\frac{1}{A_0},\quad L_0 = -\frac{1}{2}\Delta - s + 3qA_0^2.$$

The operators L_1 and L_0 are self-adjoint. The operator L_1 is positive-definite, since

$$\langle\psi|L_1|\varphi\rangle = -\int\frac{\psi^*}{2A_0}\operatorname{div}A_0^2\nabla\frac{\psi}{A_0}d\mathbf{r} = \frac{1}{2}\int A_0^2\left(\nabla\frac{\psi^*}{A_0},\nabla\frac{\psi}{A_0}\right)d\mathbf{r} > 0$$

and consequently there is a bounded inverse operator. For the inverse operator we have the estimate

$$\langle\psi|L_1^{-1}|\psi\rangle < \lambda_1^{-1}\langle\psi|\psi\rangle, \qquad (13)$$

where $\lambda_1 > 0$ is the smallest eigenvalue of the operator L_1.

Equation (11) can be rewritten in the form

$$L_0\delta A = \Omega^2L_1^{-1}\delta A. \qquad (14)$$

It is possible to apply to (14) a variational principle, namely, it can be stated that the first eigenvalue is the minimum of the functional

$$\Omega^2 = \langle\psi|L_0|\psi\rangle/\langle\psi|L_1|\psi\rangle. \qquad (15)$$

Let λ_0 be the minimum eigenvalue of the operator L_0. We then get from the relation

$$\langle\psi|L_0|\psi\rangle \geqslant \lambda_0\langle\psi|\psi\rangle \qquad (16)$$

the estimate

$$\Omega^2 \leqslant \lambda_0\lambda_1. \qquad (17)$$

Inasmuch as λ_1 is always positive, to prove the instability it is sufficient to establish that $\lambda_0 < 0$. We consider the equation

$$L_0\psi = -\tfrac{1}{2}\Delta\psi - s\psi + 3qA_0^2\psi = \lambda_0\psi. \qquad (18)$$

This is a Schrödinger equation with a potential $V = 3qA_0^2 - s$.

Differentiating (6) with respect to an arbitrary direction, we obtain

$$-\frac{1}{2}\Delta\frac{\partial A}{\partial\xi} - s\frac{\partial A}{\partial\xi} + 3qA_0^2\frac{\partial A}{\partial\xi} = 0, \qquad (19)$$

from which we see that $\partial A/\partial\xi$ is the eigenfunction of (18) with eigenvalue $\lambda_0 = 0$. This is the consequence of the fact that Eq. (6) is invariant to a shift by an arbitrary vector. We must still determine whether the eigenvalue $\lambda_0 = 0$ is the smallest one. The smallest eigenvalue should be nondegenerate. Therefore only one-dimensional solutions can be stable, since differentiation with respect to different directions yields different nonvanishing eigenfunctions in the case of multidimensional solutions. In additions, the wave function of the ground state should not have any zeroes. Therefore only one-dimensional solutions for which $A_0(\xi)$ is a monotonic function can be stable. It follows therefore that, regardless of the sign of q, all periodic solutions with constant phase are unstable. As to aperiodic solutions, when $q < 0$ the only aperiodic solution is the self-focused beam (9), which is unstable because the function $A_0(\xi)$ is nonmonotonic. When $q > 0$ the aperiodic solution is (10), which is stable by virtue of its monotonic character.

Let us estimate the increment of the instability of a plane self-focused beam. The envelope of the beam is given by (9). We choose a perturbation in the form

$$\delta A = \psi(z)\, e^{i(k_1x+k_2y)}, \quad k_1^2 + k_2^2 = k^2.$$

For the operators L_0 and L_1 we have

$$L_0 = -\frac{1}{2}\frac{d^2}{dz^2} + \frac{k^2}{2} + s - \frac{6s}{\mathrm{ch}^2\sqrt{2s}\,z}, \qquad (20)$$

$$L_1 = -\frac{1}{2}\frac{d^2}{dz^2} + \frac{k^2}{2} + s + \frac{2s}{\mathrm{ch}^2\sqrt{2s}\,z}. \qquad (21)$$

The smallest eigenvalues of these operators are (see[9])

$$\lambda_0 = -3s + k^2/2, \quad \lambda_1 = k^2/2.$$

We put $s = qE_0^2/2$, where E_0 is the amplitude at the center of the beam. According to (17) we have the estimate

$$\Omega^2 < -\tfrac{3}{4}|q|E_0^2k^2 + k^4/4. \qquad (22)$$

Comparing (22) with (7) we see that the instability increment of the self-focused beam is of the same order of magnitude as the instability increment of the plane wave.

The wave function of the perturbation can be approximated by the smallest eigenfunction of the operator L_0:

$$\psi_0 = 1/\mathrm{ch}^2\sqrt{2s}\,z.$$

The most intense instability takes place at $k \approx (qE_0^2)^{1/2}$. It follows therefore that a plane self-focused beam breaks up in "bunches" with dimension $L \sim (1/k_0)(qE_0^2/\omega)^{-1/2}$, and these bunches have a tendency to contract without limit.

We note that it follows from the foregoing results that instability should set in against perturbations with k = 0, but more accurate estimates are necessary to obtain its increment.

It also follows from the foregoing results that a self-focused beam entering into a semi-infinite medium should have spatial instability against small perturbations of the condition on the boundary. Indeed, if we discard from (1) the time derivative and neglect the second derivative with respect to x, we obtain an equation of the type (2), but with a two-dimensional Laplacian. All the preceding arguments remain in force, and we find that an arbitrarily small "entry inaccuracy" will cause the self-focused beam to be completely broken up in the medium within a distance

$$L \sim \frac{1}{k_0}\left(\frac{qE_0^2}{\omega}\right)^{-1/2},$$

i.e., the self-focusing distance. We note also that for a cylindrical beam the picture of the instability is qualitatively the same.

The results raise doubts concerning the possibility of stationary self-focusing of light in nonlinear media.

4. INSTABILITY OF A BIHARMONIC FIELD

It was shown in the preceding section that periodic envelope waves with stationary phase are unstable regardless of the sign of q. In particular, waves with $s > 0$ and with sufficiently small amplitude are unstable. These waves are the nonlinear analog of a superposition of plane monochromatic waves (biharmonic field). In this section we shall show that the biharmonic field is unstable in a wide class of transparent nonlinear media.

Let us take the Fourier transform of (2) with respect to the coordinates:

$$i\frac{\partial E_{\mathbf{k}}}{\partial t} - \frac{k^2}{2}E_{\mathbf{k}} = q\int E_{k_1}^{*}E_{k_2}E_{k_3}\delta(\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3)\,d\mathbf{k}_1\,d\mathbf{k}_2\,d\mathbf{k}_3. \qquad (23)$$

Equation (23) can be obtained by variation of the Hamiltonian

$$H = \frac{1}{2}\int k^2E_{\mathbf{k}}E_{\mathbf{k}}^{*}d\mathbf{k} + \frac{q}{2}\int E_{\mathbf{k}}^{*}E_{\mathbf{k}_1}^{*}E_{\mathbf{k}_2}E_{\mathbf{k}_3}\delta(\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3) \times d\mathbf{k}\,d\mathbf{k}_1\,d\mathbf{k}_2\,d\mathbf{k}_3$$

in accordance with the rule

$$\partial a_{\mathbf{k}}/\partial t = -i\delta H/\delta a_{\mathbf{k}}^{*}. \qquad (24)$$

Let us consider a more general model of a nonlinear medium, described by a Hamiltonian

$$H = \int \omega(\mathbf{k})\,a_{\mathbf{k}}a_{\mathbf{k}}^{*}dk + \tfrac{1}{2}\int V(\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)\,a_{\mathbf{k}}^{*}a_{\mathbf{k}_1}^{*}a_{\mathbf{k}_2}a_{\mathbf{k}_3} \times \delta(\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3)\,d\mathbf{k}\,d\mathbf{k}_1\,d\mathbf{k}_2\,d\mathbf{k}_3. \qquad (25)$$

Here $a_{\mathbf{k}}$ is a complex variable describing the medium, $\omega(\mathbf{k})$ the wave dispersion law, $V(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ is a function describing the interaction (the Hamiltonian for electromagnetic waves if calculated in the Appendix).

Varying the Hamiltonian, we obtain

$$\frac{\partial a_{\mathbf{k}}}{\partial t} + i\omega(\mathbf{k})\,a_{\mathbf{k}} = -i\int V(\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)\,a_{\mathbf{k}_1}^{*}a_{\mathbf{k}_2}a_{\mathbf{k}_3}\delta(\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3) \times d\mathbf{k}_1\,d\mathbf{k}_2\,d\mathbf{k}_3. \qquad (26)$$

The biharmonic field is an approximate solution of this equation, in the form

$$a_{\mathbf{k}} = A_1e^{-i\omega_1t}\delta(\mathbf{k}-\mathbf{k}_1) + A_2e^{-i\omega_2t}\delta(\mathbf{k}-\mathbf{k}_2),$$

where

$$\omega_1 = \omega(\mathbf{k}_1) + 2V(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_2, \mathbf{k}_1)|A_2|^2 + V(\mathbf{k}_1, \mathbf{k}_1, \mathbf{k}_1, \mathbf{k}_1)|A_1|^2,$$
$$\omega_2 = \omega(\mathbf{k}_2) + 2V(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_2, \mathbf{k}_1)|A_1|^2 + V(\mathbf{k}_2, \mathbf{k}_2, \mathbf{k}_2, \mathbf{k}_2)|A_2|^2.$$

It is assumed that terms proportional to the squares of the amplitudes are small.

We subject this field to a perturbation of the type

$$\delta a = \alpha(t)e^{-i\omega' t}\delta(\mathbf{k}-\mathbf{k}') + \beta(t)e^{-i\omega'' t}\delta(\mathbf{k}-\mathbf{k}''),$$

where

$$\omega' = \omega(\mathbf{k}') + 2|A_1|^2V(\mathbf{k}', \mathbf{k}_1, \mathbf{k}_1, \mathbf{k}') + 2|A_2|^2V(\mathbf{k}', \mathbf{k}_2, \mathbf{k}_2, \mathbf{k}'),$$
$$\omega'' = \omega(\mathbf{k}'') + 2|A_1|^2V(\mathbf{k}'', \mathbf{k}_1, \mathbf{k}_1, \mathbf{k}'') + 2|A_2|^2V(\mathbf{k}'', \mathbf{k}_2, \mathbf{k}_2, \mathbf{k}''),$$

with $\omega_1 + \omega_2 = \omega' + \omega''$, $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}' + \mathbf{k}''$. Then $\alpha(t)$ and $\beta(t)$ are connected by the equations

$$\frac{\partial\alpha}{\partial t} = -2iV(\mathbf{k}', \mathbf{k}'', \mathbf{k}_1, \mathbf{k}_2)\beta^*, \quad \frac{\partial\beta}{\partial t} = -2iV(\mathbf{k}', \mathbf{k}'', \mathbf{k}_1, \mathbf{k}_2)\alpha^*.$$

These equations yield

$$\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2, \quad \omega(\mathbf{k}) = \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2).$$

We see therefore that the biharmonic field is unstable for any law of interaction and for any law of dispersion. In particular, a standing wave is unstable in any nonlinear transparent medium.

Application to Eq. (2) yields instability of a stationary envelope wave when $s > 0$ regardless of the sign of q, and the order of the instability increment is $\gamma \sim \omega\epsilon_1|E_0|^2/\epsilon_0$, where E_0 is the characteristic amplitude of the wave. An estimate of the increment by a variational method gives the same result. When $\mathbf{k}_1 \to \mathbf{k}_2$, this instability goes over into the plane-wave instability considered in[10].

APPENDIX

ELECTROMAGNETIC WAVE INTERACTION HAMILTONIAN

Interaction Hamiltonians of the type (25) describe either nonlinear media whose equations contain no quadratic nonlinearity, or media in which the wave dispersion laws are such that three-wave process of the type

$$\alpha, \beta \sim e^{\gamma t}, \quad \gamma^2 = 4|V(\mathbf{k}', \mathbf{k}'', \mathbf{k}_1, \mathbf{k}_2)|^2|A_1|^2|A_2|^2$$

are impossible.

Even in these cases, however, the interaction Hamiltonians of the electromagnetic waves have a more complicated form than (25), since, generally speaking, it is necessary to take simultaneously into account the interaction of waves with different polarizations. We confine ourselves to the simplest case, for the description of which a Hamiltonian of the type (25) is suitable. We consider electromagnetic waves in an isotropic medium without spatial dispersion, and assume that the electric intensity vector is directed along the z axis and depends only on the variables x and y. The equation of the medium takes the form

$$\left(k^2 - \frac{\omega^2}{c^2}\varepsilon(\omega)\right)E_{k\omega} = \frac{\omega^2}{c^2}\int \varepsilon^{(1)}(\omega, \omega_1, \omega_2, \omega_3)E_{\mathbf{k}_1\omega_1}E_{\mathbf{k}_2\omega_2}E_{\mathbf{k}_3\omega_3}$$
$$\times\,\delta(\omega + \omega_1 - \omega_2 - \omega_3)\delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3)\,d\omega_1 d\omega_2 d\omega_3 d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3. \quad \text{(A.1)}$$

Here ϵ and $\epsilon^{(1)}$ are respectively the linear and nonlinear parts of the dielectric constant. In a transparent medium $\epsilon(\omega)$ is real, and $\epsilon^{(1)}(\omega, \omega_1, \omega_2, \omega_3)$ is not only symmetrical in the indices ω_1, ω_2, and ω_3, but is subject to an additional symmetry relation

$$\varepsilon^{(1)}(\omega, \omega_1, \omega_2, \omega_3) = \varepsilon^{(1)}(-\omega_1, -\omega, \omega_2, \omega_3)$$

and to the relation

$$\varepsilon^{(1)}(-\omega, -\omega_1, -\omega_2, -\omega_3) = \varepsilon^{*(1)}(\omega, \omega_1, \omega_2, \omega_3),$$

which follows from the fact that the vector E(z, t) is real.

Let $\omega(\mathbf{k}) > 0$ be the wave dispersion law determined from the equation $c^2k^2 = \omega^2\epsilon(\omega)$. We represent the Fourier component of the electric field in the form

$$E_{\mathbf{k}\omega} = (a_{\mathbf{k}\omega} + a^*_{-\mathbf{k}-\omega})\frac{\omega(\mathbf{k})}{[\omega^2\varepsilon(\omega)]'_{\omega=\omega(\mathbf{k})}} \quad \text{(A.2)}$$

(the prime denotes differentiation with respect to ω), $a_{\mathbf{k}\omega}$ being different from zero only when $\omega > 0$ and having a short maximum at $\omega = \omega(\mathbf{k})$. We substitute (A.2) in (A.1) and assume that $\omega \approx \omega(\mathbf{k})$. We can then expand the left side of (A.1) in powers of $(\omega - \omega(\mathbf{k}))$, confining ourselves to the first term. The integrand in the first part breaks up into a sum of terms, in each of which the integration is over a region near the surface defined by the equations

$$\omega(\mathbf{k}) \pm \omega(\mathbf{k}_1) \pm \omega(\mathbf{k}_2) \pm \omega(\mathbf{k}_3) = 0, \quad \mathbf{k} \pm \mathbf{k}_1 \pm \mathbf{k}_2 \pm \mathbf{k}_3 = 0.$$

Since the dispersion law is such that processes of the type $\omega(\mathbf{k}) = \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2)$, $\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2$ are forbidden, then processes of the type $\omega(\mathbf{k}) = \omega(\mathbf{k}_1) + \omega(\mathbf{k}_2) + \omega(\mathbf{k}_3)$, $\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3$ are all the more forbidden. Therefore the only terms remaining under the integral sign are those corresponding to processes of the type

$$\omega(\mathbf{k}) + \omega(\mathbf{k}_1) = \omega(\mathbf{k}_2) + \omega(\mathbf{k}_3),$$
$$\mathbf{k} + \mathbf{k}_1 = \mathbf{k}_2 + \mathbf{k}_3.$$

Gathering all these terms and replacing the arguments in $\epsilon^{(1)}(\omega, \omega_1, \omega_2, \omega_3)$ by their values at the maxima, we obtain

$$(\omega - \omega(\mathbf{k}))a_{\mathbf{k}\omega} = \int W(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)a^*_{\mathbf{k}_1\omega_1}a_{\mathbf{k}_2\omega_2}a_{\mathbf{k}_3\omega_3}$$
$$\times\,\delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3)\delta(\omega + \omega_1 - \omega_2 - \omega_3)\,d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 d\omega_1 d\omega_2 d\omega_3 \quad \text{(A.3)}$$

or, after taking the inverse Fourier transform with respect to time,

$$\frac{\partial a_{\mathbf{k}}}{\partial t} + i\omega_{\mathbf{k}}a_{\mathbf{k}} = -i\int W(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)a_{\mathbf{k}_1}{}^*a_{\mathbf{k}_2}a_{\mathbf{k}_3}$$
$$\times\,\delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3)\,d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3. \quad \text{(A.4)}$$

Here

$$W(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) = \frac{6\omega(\mathbf{k})\omega(\mathbf{k}_1)\omega(\mathbf{k}_2)\omega(\mathbf{k}_3)}{[\omega^2\varepsilon(\omega)]'[\omega_1^2\varepsilon(\omega_1)]'[\omega_2^2\varepsilon(\omega_2)]'[\omega_3^2\varepsilon(\omega_3)]'} \quad \text{(A.5)}$$
$$\times\,\varepsilon^{(1)}(\omega(\mathbf{k}), -\omega(\mathbf{k}_1), \omega(\mathbf{k}_2), \omega(\mathbf{k}_3)).$$

Obviously, $W(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ is symmetrical with respect to the substitutions $\mathbf{k} \leftrightarrow \mathbf{k}_1$ and $\mathbf{k}_2 \leftrightarrow \mathbf{k}_3$, and goes over into its complex conjugate when the $\mathbf{k}$, $\mathbf{k}_1$ pair is replaced by $\mathbf{k}_2$, $\mathbf{k}_3$. These relations show that Eqs. (A.4) can be obtained by a variational method using formula (24). The Hamiltonian is then given by (25), for which purpose it is necessary to substitute for V the value of W from (A.5).

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