PROPAGATION AND EXCITATION OF ELECTROMAGNETIC WAVES IN A NONLINEAR MEDIUM

V. B. KRASOVITSKIĬ and V. I. KURILKO

Physico-technical Institute, Academy of Sciences, Ukrainian S.S.R.

Submitted to JETP editor July 15, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 48, 353-357 (January, 1965)

The problem treated is that of the propagation of a transverse wave of large amplitude in a two-level system. A nonlinear dispersion equation for such waves is derived and studied. It is shown that under certain conditions the transverse wave is unstable with respect to parametric excitation of a longitudinal wave with the same phase velocity. The problem of excitation of a two-level system by an electron beam is studied. It is shown that if the condition for the anomalous Doppler effect is satisfied, the system can be transformed to the inverted state. Coupled nonstationary longitudinal-transverse oscillations, in the system composed of the beam and the active medium, are treated.

1. As is known, appreciable electromagnetic field intensities are now obtained in the optical frequency range by means of coherent sources. At such field intensities, nonlinear effects of the interaction of the field with the material are important. Some of these effects have been discussed by a number of authors.[1-4]

In the present work we consider the problem of the effect of these nonlinearities on the propagation of electromagnetic waves in the medium, and also the possibility of excitation of nonlinear electromagnetic oscillations in the medium by a beam of charged particles. Nonlinear effects show up especially strongly near a resonance, when the frequency of the wave is close to one of the characteristic frequencies of the molecules of the medium. Under these conditions, the most important nonlinear effect is the change of populations of the levels under the influence of the field of the wave. The influence of this effect on the propagation and excitation properties of electromagnetic waves can be treated by using, as a model of the medium, a system with two energy levels, $E_2 - E_1 = \hbar\Omega$.

The problem under consideration reduces to the solution of Maxwell's equations jointly with the nonlinear equations that describe the properties of the medium:[3]

$$\frac{\partial^2 \mathbf{P}}{\partial \tau^2} + \mathbf{P} = -q^2 W \mathbf{E}, \qquad \frac{\partial W}{\partial \tau} = \mathbf{E}\frac{\partial \mathbf{P}}{\partial \tau}; \tag{1}$$

$$\operatorname{curl} \mathbf{E} = -\frac{\partial \mathbf{H}}{\partial \tau}, \quad \operatorname{curl} \mathbf{H} = \frac{\partial \mathbf{E}}{\partial \tau} + 4\pi \frac{\partial \mathbf{P}}{\partial \tau}. \tag{2}$$

Here $q = 2d^2N/\hbar\Omega$; the polarization vector **P** and the field vectors **E** and **H** are expressed in units Nd (d is the dipole moment of a molecule, and N is the density of active molecules); W is the density of the energy stored in molecules of the active material (in units N^2d^2); and $\Omega = (E_2 - E_1)/\hbar$.

We shall seek solutions of these equations dependent only on one coordinate, $\xi = \Omega z/c$, which coincides with the direction of propagation of the wave. In the general case, the nonlinear longitudinal and transverse waves in such a medium are coupled, since the difference of populations of the levels, qW, is determined by both components of the vector polarization:

$$qW = (1 - \mathbf{P}^2 - \dot{\mathbf{P}}^2)^{1/2}. \tag{3}$$

The effects of interaction of longitudinal and transverse waves will be treated below. In the present section we limit ourselves to the investigation of purely transverse waves. For such waves, the relation between the electric field amplitude and the polarization vector can be found from Maxwell's equations:

$$E_\perp(\eta) = -\mu P_\perp(\eta);$$
$$\mu = 4\pi / (1 - k^2/\omega^2), \qquad \eta = \omega\tau - k\xi. \tag{4}$$

Upon substituting (3) and (4) into (1), we get the equation for the polarization $P_\perp$:

$$\ddot{P}_\perp + P_\perp = q\mu(1 - P_\perp^2 - \dot{P}_\perp^2)^{1/2} P_\perp \tag{5}$$

(here dots indicate partial derivatives with respect to time τ).

A first integral of Eq. (5) has the form

$$(1-P_{\perp}^2-\dot{P}_{\perp}^2)^{1/2}=q(C-{}^1/_2\mu P_{\perp}^2), \qquad (6)$$

where $C = W(0) + {}^1/_2\mu P_{\perp}^2(0)$. We suppose that the constant C does not depend on the coordinate ξ.

In a first approximation with respect to the parameter q, which is practically always small in comparison with unity, the dispersion relation can be investigated by use of Eq. (5). On substituting into Eq. (5) the value $P_{\perp} = a_{\perp}\cos\eta$ and keeping terms of the first order of smallness in q and in $\omega-1$, we find

$$\omega^2-1=-\mu q(1-a_{\perp}^2)^{1/2}. \qquad (7)$$

For $a_{\perp}^2 \ll 1$, this equation goes over to a linear dispersion equation $((1-a_{\perp}^2)^{1/2}\approx -1)$. Thus with increase of the amplitude $a_{\perp}$, the influence of the medium on the propagation of the transverse wave decreases; in other words, increase of amplitude of the vector polarization is equivalent to decrease of the density of active material.

However, this equation is inapplicable in strong fields, when $1-a_{\perp}^2 \to 0$, for in the derivation of this equation we neglected the quantity $\omega-1$ in comparison with $1-a_{\perp}^2$ in the radical on the right side of (7). In this case, however, the effect of nonlinearity is considerably smaller, because the number of particles interacting with the field decreases.

We now consider the effects of interaction of transverse waves with longitudinal. The presence of a transverse wave of large amplitude leads to the result that the difference of populations of the levels is modulated at twice the frequency of the transverse wave. Under definite conditions, such modulation can lead to parametric excitation of a longitudinal wave. In fact, Eq. (1) for small oscillations of the longitudinal polarization vector can be be written in the form (if $P_{\parallel}^2 + \dot{P}_{\parallel}^2 \ll P_{\perp}^2 + \dot{P}_{\perp}^2$)

$$\ddot{P}_{\parallel}+\Omega_{\parallel}^2[1+h\cos 2(\omega\tau-k\xi)]P_{\parallel}=0,$$

$$\Omega_{\parallel}^2=1-4\pi q^2C+q^2\pi\mu a_{\perp}^2, \qquad h=\pi q^2\mu a_{\perp}^2. \qquad (8)$$

The solution of this equation for $h \ll 1$ has the form

$$P_{\parallel}(\tau,\xi)=a_{\parallel}(0)e^{s\tau}\cos(\omega\tau-k\xi),$$

where $s^2 = h^2/4 - 4(\omega-\Omega_{\parallel})^2$. When $|h| > 4|\omega-\Omega_{\parallel}|$, the amplitude of the longitudinal oscillations increases exponentially with increase of τ. The largest logarithmic increment s will occur when $\omega = \Omega_{\parallel}$; then the increment is equal to $s_{max} = h/2$. For $a_{\perp} \sim 1$, $\Omega \approx 3\times10^{15}\ \text{sec}^{-1}$, $N \sim 5\times10^{19}\ \text{cm}^{-3}$, and $\mu \sim 4\pi$, the value of $s_{max} \approx 2\times10^{-5}$.

The growth of the amplitude of the longitudinal wave will continue until this amplitude becomes comparable with the amplitude of the transverse wave. Since the longitudinal wave thus amplified has the same phase velocity as the transverse wave, the effect treated above can be used for acceleration of charged particles.

2. For coherent generation of light waves, it is necessary to insure an inversion of the populations of the levels. Here we treat the problem of excitation of an active medium by means of an electron beam. We shall take into account the effects of collective interaction of the beam with the active medium, neglecting effects of collisions of pairs.

We shall suppose that the energy density in the beam is large enough so that the energy expended by the beam in excitation of oscillations is large in comparison with the density of the energy that can be stored in molecules of the medium: $\gamma m n_0 v_0^2 \gg N\hbar\Omega$ (n_0 is the density of the beam, v_0 is the velocity of the beam, and γ is the increment of growth of the oscillations[5]). In this case it is sufficient to take account of oscillations in the beam in the linear approximation.

Since we are interested in nonstationary processes in the system composed of the beam and the active medium, we shall seek a solution of Eqs. (1) and (2) and also of the linearized equations of motion of the beam, in the form of waves with amplitude and phase varying slowly with time:

$$P_{\parallel}(\tau,\xi)=a_{\parallel}(\tau)\cos(\omega\tau-k_{\parallel}\beta_0\xi+\vartheta_{\parallel}).$$

We express $E_{\parallel}$ in terms of $P_{\parallel}$, from Maxwell's equations and the equations of motion for the beam:

$$E_{\parallel}(\tau,\xi)=-4\pi P_{\parallel}(\tau,\xi)+4\pi\omega_0\int_0^{\tau}\sin[\omega_0(\tau-\tau')]\times P_{\parallel}[\tau',\xi-\beta_0(\tau-\tau')]\,d\tau'$$

($\omega_0 = (4\pi n_0 e^2/m\Omega^2)^{1/2}$ is the plasma frequency of the beam). On substituting this result in Eq. (1), we get the following first-order equations for $a_{\parallel}$ and $\vartheta_{\parallel}$:

$$\frac{da_{\parallel}}{d\tau}=-\operatorname{sign}W_0\cdot\pi q\omega_0\sqrt{1-a_{\parallel}^2}\int_0^{\tau}a_{\parallel}(\tau')\,d\tau', \qquad (9)$$

$$\frac{d\vartheta_{\parallel}}{d\tau}=-\operatorname{sign}W_0\cdot 2\pi q\sqrt{1-a_{\parallel}^2}. \qquad (10)$$

In the derivation of (9) and (10), the following conditions were assumed to be satisfied:

$$q \ll \omega_0, \qquad \omega-1=1-k_{\parallel}\beta_0+\omega_0=0.$$

The last requirement coincides with the condition for the anomalous Doppler effect ($v_{ph} = \omega/k < v_0$). It is easy to see that when $\operatorname{sign}W_0 = -1$ (a majority of the molecules at the initial moment are in the lower level), growth of amplitude of the longitudinal oscillations occurs. Then in the small-amplitude range ($a_{\parallel}^2 \ll 1$), the growth is of exponential character, with an increment $\gamma = (\pi q \omega_0)^{1/2}$ determined by the linear theory.

For treatment of large amplitudes, it is convenient to introduce the substitution $a_{\parallel} = \sin\varphi$. Then Eq. (9) takes the following form:

$$d^2\varphi / d\tau^2 - \gamma^2 \sin\varphi = 0. \tag{11}$$

This is a known equation, which determines the oscillations of a nonlinear pendulum with the equilibrium position at the topmost point. Thus in the system under consideration, there takes place a periodically recurring transfer of the energy of the longitudinal oscillations of the field into internal energy of the matter. An important fact is that the difference of populations of the levels, $qW = -\cos\varphi(\tau)$ $(\cos\varphi(0) > 0)$, can as a result of this process assume positive values. The time that the system remains in the inverted state during each period of the oscillations is, in order of magnitude, $T \sim \gamma^{-1}$; the period of the oscillations increases logarithmically with decrease of the initial amplitude of the oscillations.

It should be mentioned that the relations obtained in this section are valid when there is negligible thermal spread of the electrons of the beam with respect to velocity. In the case of the anomalous Doppler effect, which we have considered, this condition is satisfied if $|\omega - k_{\parallel}\beta_0| = \omega_0 \gg k_{\parallel}\beta_T$, where $c\beta_T = v_T$ is the thermal velocity of the electrons of the beam. The last inequality, and also the condition for negligibility of nonlinear effects in the equations of motion of the beam $(m^2 v_0^2 d^2 \omega_0^{5/2} \gg 4\pi^{1/2}\hbar^2 e^2 q^{1/2})$, can be satisfied, evidently, only in the microwave region.

We now consider the problem of the development, with time, of small fluctuations of the transverse field in the system under consideration (beam and active medium). For small transverse oscillations it may be supposed that the internal energy of the medium changes with time according to the law (11) determined by purely longitudinal oscillations.

As in the case of longitudinal oscillations, we shall seek a solution for the field $E_{\perp}$ and the polarization $P_{\perp}$ in the form of a wave with slowly varying amplitude and phase:

$$P_{\perp}(\tau, \xi) = a_{\perp}(\tau)\cos(\omega\tau - k_{\perp}\xi + \vartheta_{\perp}).$$

Then the relation between $E_{\perp}$ and $P_{\perp}$, in accordance with Maxwell's equations, has the form

$$E_{\perp}(\tau,\xi) = -4\pi P_{\perp}(\tau,\xi) + 4\pi k_{\perp}\int_0^{\tau} \sin[k_{\perp}(\tau-\tau')]P_{\perp}(\tau',\xi)\,d\tau'. \tag{12}$$

On substituting into (1) the value of $E_{\perp}$ from (8) and $qW = -\cos\varphi$, we get the following equation for the amplitude of the transverse oscillations when $\omega_0 \ll 1$:

$$\frac{da_{\perp}}{d\tau} = -\pi q \cos[\varphi(\tau)]\int_0^{\tau} a_{\perp}(\tau')\,d\tau'. \tag{13}$$

From Eq. (13) it is clear that the amplitude of the transverse field begins to grow only upon transition of the system to the inverted state ($\cos\varphi < 0$). Since $\varphi(\tau)$ changes appreciably over a time of order $(\pi q \omega_0)^{-1/2} \gg (\pi q)^{-1/2}$, a solution of Eq. (13) can be found by the WKB method:

$$a_{\perp}(\tau) = a_{\perp}(\tau_0)\left|\frac{\cos\varphi(\tau)}{\cos\varphi(\tau_0)}\right|^{1/4} \times \exp\left\{\int_{\tau_0}^{\tau}[\pi q\cos\varphi(\tau')]^{1/2}\,d\tau'\right\}. \tag{14}$$

This expression is valid up to τ_0's for which $\cos\varphi(\tau_0) < 0$ and $\omega_0 \ll |\cos\varphi(\tau_0)|$. The increase of amplitude of the transverse oscillations will continue until the transverse wave begins to have an appreciable influence on the populations of the levels, that is until a time τ_m determined by the condition $|\cos\varphi(\tau_m)| \lesssim a_{\perp}(\tau_m)$.

Study of the equations of coupled longitudinal-transverse oscillations of the system shows that the system cannot be in a state in which the population of the levels or the direction of the polarization vector does not change with time. This conclusion is easily reached by studying the small oscillations in the system

$$\frac{d}{d\tau}\frac{\dot a_{\parallel}}{(1-a_{\parallel}^2-a_{\perp}^2)^{1/2}} = \pi q \omega_0 a_{\parallel};$$

$$\frac{d}{d\tau}\frac{\dot a_{\perp}}{(1-a_{\parallel}^2-a_{\perp}^2)^{1/2}} = -\pi q a_{\perp}. \tag{15}$$

By means of the substitution $a_{\perp} = \sin\varphi\sin\vartheta$, $a_{\parallel} = \sin\varphi\cos\vartheta$ it can be shown that the frequencies of small oscillations about the equilibrium position $\varphi = \varphi_0$ and $\vartheta = \vartheta_0$ are $\omega_1^2 = \cos\varphi_0$, $\omega_2^2 = -\omega_0\cos\varphi_0$. Thus the small oscillations are always unstable.

It is easily seen that in the absence of the beam, the amplitude of longitudinal oscillations $a_{\parallel}$ remains constant, but the amplitude of transverse oscillations changes according to the law of nonlinear oscillations of a pendulum. In fact, upon introducing into (15) the substitution $a_\perp = \sin\psi$ and supposing that $a_{||}(0) = 0$, we get the following equation for ψ:

$$d^2\psi / d\tau^2 - \text{sign } W_0 \cdot \pi q \sin\psi = 0. \qquad (16)$$

In the case in which sign $W_0 = -1$ (a majority of the particles are in the lower level), Eq. (16) describes a nonlinear pendulum with the equilibrium position at the bottom point. If $\sin\psi \sim \psi$ (linear oscillations), then $a_\perp(\tau)$ is modulated with frequency $(\pi q)^{1/2}$. When sign $W_0 = +1$ (a majority of the particles, at the initial moment, are in the upper level), (16) becomes the equation of oscillations of a pendulum with the equilibrium point at the top position.

The authors are grateful to Ya. B. Faĭnberg for proposing the topic and for useful discussions, and also to R. V. Khokhlov and V. D. Shapiro for discussion of the results of the work.

---

[1] A. P. Kazantsev and V. S. Smirnov, JETP **46**, 182 (1964), Soviet Phys. JETP **19**, 130 (1964).

[2] L. A. Ostrovskiĭ and E. I. Yakubovich, JETP **46**, 963 (1964), Soviet Phys. JETP **19**, 656 (1964).

[3] E. T. Jaynes and F. W. Cummings, Proc. I.E.E.E. **51**, 89 (1963); V. M. Faĭn, JETP **33**, 1290 (1957), Soviet Phys. JETP **6**, 991 (1958).

[4] Armstrong, Bloembergen, Ducuing, and Pershan, Phys. Rev. **127**, 1918 (1962).

[5] V. D. Shapiro and V. I. Shevchenko, JETP **42**, 1515 (1962), Soviet Phys. JETP **15**, 1053 (1962); V. N. Tsytovich and V. D. Shapiro, ZhTF **34**, 764 (1964), Soviet Phys. Tech. Phys. **9**, 583 (1964).

Translated by W. F. Brown, Jr.
46