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* We use atomic units with πa_0^2 equal to unity for σ.

** According to (8), only the ^2P term is excited in the LS coupling scheme.

SELF-ACTION OF LIGHT IN CRYSTALS

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1. The possibility of observing self-focusing [1-5] and other "self-action" effects of light [6] is presently connected almost exclusively with the cubic nonlinearity of the dependence of the polarization on the field in liquids, since the latter are characterized by large values of the increment to the nonlinear polarizability [7,8]. For crystals, the corresponding values are usually much lower. In crystals without inversion center, however, strong self-action is possible as a result of the second-harmonic reaction. In this case the effects due to anisotropy are distinctive and of interest. We consider below the peculiarities of such a mechanism and present very simple estimates.

The interaction of waves having frequencies ω and 2ω in a quadratic medium leads to the appearance, at the corresponding frequencies, of the nonlinear-polarization vectors

$$P_i^{(2\omega)} = \chi_{ikl}^{(2\omega)} E_k^{(\omega)} E_l^{(2\omega)} (a); P_i^{(\omega)} = \chi_{ikl}^{(2\omega-\omega)} E_k^{*(\omega)} E_l^{(2\omega)} (6). \qquad (1)$$

By determining $\vec{E}^{(2\omega)}$ from (1a) in the given field approximation (which is valid everywhere except in the synchronism directions), and substituting the result in (1b), we obtain for $P_i^{(\omega)}$ in a quasi-plane wave an expression of the form

$$P_i^{(\omega)} = \theta_{iklm} A_k^* A_l A_m e^{-i\mathbf{k}_1\mathbf{r}} + P_i' e^{-i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}}. \qquad (2)$$

Here $\vec{A}$ is the amplitude of the fundamental wave, and $\vec{k}_{1,2}$ are the values of the wave vector at the frequencies ω and 2ω respectively; $\hat{\theta}$ is a tensor proportional to the product of the tensors $\hat{\chi}^{(2\omega)}$ and $\hat{\chi}^{(2\omega-\omega)}$; P_i' is the polarization connected with the proper waves at the frequency 2ω and determined from the boundary conditions. Inasmuch as the crystal dimensions are usually much larger than the coherence length $l_{coh} \sim \pi/|2\vec{k}_1 - \vec{k}_2|$, the term P_i' can be discarded when averaging over the period $2\pi/|k_1|$. As a result there is produced a third-order stimulated polarization, which is synchronous with the wave and which leads to an amplitude dependence of the refractive index and consequently to self-action.

2. Let us consider first the propagation of the extraordinary wave in a uniaxial crystal of the KDP type, in which the following components of the tensor $\hat{\chi}$ differ from zero: $\chi_{1,23} = \chi_{2,13} \approx \chi_{3,21} = \chi$, with $\hat{\chi}^{(2\omega)} = \hat{\chi}^{(2\omega-\omega)}$ (3 - optical axis, 1,2 - the other two crystallographic axes). If the wave propagates along the z axis and is polarized along x ($x \perp 3$), then the vector $\vec{P}^{(2\omega)}$ is directed along the optical axis (cf. [8]) and excites an extraordinary wave at the frequency 2ω. Since in optics we have almost always $|2k_1 - k_2| \ll k_{1,2}$ even far from synchronism, the component $P_x^{(\omega)}$ is equal to

$$P_x^{(\omega)} = \frac{4\pi\chi^2 \sin^2 2\phi \sin\gamma \sin\theta \epsilon_\perp A_x |A_x|^2}{(n_1^2 - n_2^2)\cos(\gamma-\theta)(\epsilon_\perp \sin^2\theta + \epsilon_{\parallel}\cos^2\theta)} = \frac{\epsilon'}{4\pi}|A_x|^2 A_x. \quad (3)$$

Here θ is the angle between the z axis and the optical axis, γ the angle between the latter and the **ray** vector of the extraordinary wave, φ the azimuthal angle of the beam in the (12) plane; n_1 and n_2 are the refractive indices of the ordinary wave at frequency ω and of the extraordinary wave at frequency 2ω, respectively. The other components of $\vec{P}^{(\omega)}$ excite also an extraordinary wave at frequency ω, but owing to the anisotropy its field is not in synchronism with E_x and is small.

Thus, the ordinary wave propagates in a crystal in the same way as in an isotropic medium with nonlinearity of the type $\epsilon_{eff} = \epsilon_0 + \epsilon'|A_x|^2$, where ϵ' is given by (4). This nonlinearity leads to effects which have been investigated for isotropic media.* Here, however, not only the magnitudes of the effects, but also their character exhibit an appreciable dependence on the direction. Thus, the sign of ϵ' depends on the difference $n_1^2 - n_2^2$, i.e., whereas self-focusing is possible on one side of the synchronism cone, the wave becomes defocused on its other side.

To estimate the value of ϵ' in (3), it is necessary to take into account the fact that ϵ' increases on approaching synchronism, but (3) is valid so long as the dispersion effects prevail over the nonlinear effects, i.e., when the quantity $\xi \sim |\chi A|/|n_1 - n_2|$ is small. Far from synchronism ($\theta = \pi/2$, $\varphi = \pi/4$) the value of ϵ' is 2.5×10^{-14} cgs esu for KDP and exceeds 10^{-12} cgs esu for $LiNiO_3$. Near the synchronism angles $|\epsilon'|$ reaches values not lower than 10^{-11} - 10^{-10} cgs esu, and simultaneously the condition $\xi \ll 1$ is satisfied at a beam power on the order of 10^6 - 10^7 W/cm^2.** (We note that in nonlinear liquids ϵ' does not exceed 10^{-11} cgs esu). The characteristic length of filament formation [7] amounts to several centimeters in this case.

On coming closer to synchronism, when $\xi \gtrsim 1$, the given-field approximation used in (2) is no longer valid; synchronous frequency doubling takes place in this region. But even here self-action effects apparently play a significant role in the case of propagating beams having a finite angular divergence.

3. New effects arise in cases when the linear properties of the crystal are isotropic and the waves of both polarizations can interact in synchronism. In a cubic crystal, the

nonvanishing components of $\hat{\chi}$ are the same as in KDP. Assuming for concreteness that the wave vector (z axis) lies in the (12) plane (1,2,3 - directions of the crystallographic axes) and using Eqs. (1) - (3), we get

$$P_x^{(\omega)} = \frac{\alpha}{4\pi} A_x \left(\frac{1}{2}|A_x|^2 + |A_y|^2\right); \; P_y^{(\omega)} = \frac{\alpha}{4\pi} A_y |A_x|^2, \tag{4}$$

where $\alpha = 32\pi^2\chi^2\sin^2\varphi/(\epsilon_1 - \epsilon_2)$, the x axis lies in the 12 plane, the y axis coincides with 3, φ is the angle between the axes z and 1, $\epsilon_1 = \epsilon(\omega)$, and $\epsilon_2 = \epsilon(2\omega)$. In the derivation of (4) we discarded, as before, terms of the order of $(\epsilon_1 - \epsilon_2)/\epsilon_2$.

The propagation of a stationary paraxial beam is described here by a system of diffusion equations for A_x and A_y with right-hand side parts of (4). This system has a particular solution in the form of a stationary plane wave $A_i = A_i\exp(-i\gamma_i z)$, where

$$\gamma_x = \frac{\alpha k_1}{2\epsilon_1}\left(\frac{1}{2}|A_x|^2 + |A_y|^2\right), \; \gamma_y = \frac{\alpha k_1}{2\epsilon_1}|A_x|^2. \tag{5}$$

The propagation velocities of the different components coincide only when $|A_x|^2 = 2|A_y|^2$. In all other cases, the phase difference between A_x and A_y varies with the distance, with a period $\Lambda = 2\pi/|\gamma_x - \gamma_y|$. The character of the polarization of the wave therefore alternates, with the same period, from linear to elliptic (circular) and back to linear, but the latter is turned relative to the initial linear polarization, etc. This effect can be used, for example, for the construction of nonlinear shutters. We note that in an isotropic liquid only rotation of the polarization ellipse is possible, without change of eccentricity [9], and that the linearly polarized wave does not change.

The peculiarities of self-focusing become clearer when the stability of a plane wave is investigated. By finding a solution [10] in the form $A_i = (A_i + a_i)\exp(-i\gamma_i z)$, where the a_i are small, and putting $a_i \sim \exp(-ihz - i\vec{\kappa}\cdot\vec{r})$, we can obtain a dispersion equation in the form

$$\left(\frac{2k_1 h}{\kappa}\right)^2 = \kappa^2 - s; \; s = \frac{\alpha k_1^2}{2\epsilon_1}|A_x|^2\left(1 \pm \sqrt{1 + 16|A_y|^2/|A_x|^2}\right) \tag{6}$$

which determines, for specified $\vec{\kappa}$, four values of h. If $s > \kappa^2 > 0$, then $h^2 < 0$ and the plane wave is unstable (breaks up into filaments). It is important that such an instability is possible for any sign of α (and not only when $\alpha > 0$, as in an isotropic liquid). It can be shown that when $\alpha > 0$ the growing perturbations are those polarized in the same direction of the wave and when $\alpha < 0$, those polarized perpendicular to the wave field.

The greatest instability increment corresponds to $\kappa^2_{opt} = s/2$ and is equal to $s/4k_1$. The value of s itself at fixed wave amplitude $A^2 = |A_x|^2 + |A_y|^2$ depends on the polarization direction. If $\alpha > 0$ the maximum of s is attained when $|A_x|^2 = 2|A_y|^2 = (2/5)A^2$ (when $\gamma_1 = \gamma_2$ and the polarization of the fundamental wave does not change) and is equal to $(4/3\epsilon_1)k_1^2\alpha A^2$, and when $\alpha < 0$ the maximum occurs at $|A_x|^2 = (2/3)|A_y|^2 = (2/5)A^2$ and is equal to $(4/5\epsilon_1)k_1^2\alpha A^2$.

For cubic crystals in optics, the usual case is $\alpha < 0$ $(\epsilon_1 < \epsilon_2)$. The effective value

$|\epsilon^1|$, corresponding to the same $|h|_{max}$ as in an isotropic crystal, is then equal to 0.4 $|\alpha|$. In spite of the absence of synchronism, ϵ^1 can reach large values, owing to the electrooptical effect. In crystals of the GaAs type $|\epsilon^1| \gtrsim 10^{-10}$ cgs esu when $\lambda_0 = 1\mu$; the length of filament formation in powerful beams ($P \sim 10^7 - 10^8$ W/cm^2) is here not larger than 1 cm.

Similar conclusions can be drawn with respect to the temporal instability of a monochromatic wave with respect to modulated perturbations (space-time self-focusing) [6].

We note that self-action is made possible by the reaction of the static field component during the detection of light. However, in view of the large values of ϵ_0 at low frequency, and in view of the absence of synchronism, the magnitude of the corresponding effects is apparently much smaller.

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* The case when the fundamental wave is extraordinary calls for a special analysis.

** It is obvious that when P is reduced it is possible to come closer to synchronism by leaving ξ small. Therefore the attained value $\epsilon^1 P \sim \epsilon^1 |A|^2$ which determines the self-focusing length is in fact proportional to $\sqrt{P}$ and not to P if ξ remains constant.

DEPENDENCE OF THE WIDTH OF A SELF-FOCUSING LIGHT BEAM ON THE POWER

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It is shown in the classical paper [1] that the geometric configuration of a self-focused cylindrical light beam is described by the normalized equation

$$d^2E^*/dr^{*2} + \frac{1}{r^*} dE^*/dr^* - E^* + E^{*3} = 0. \quad (1)$$

Equation (1) in itself does not yield an estimate of the beam width, since the normal-