

NONLINEAR OPTICAL EFFECTS CAUSED BY LIGHT-INDUCED RESONANT ORIENTATION OF TUNNELING CENTERS

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The influence of self-induced anisotropy on the light propagation in impure cubic crystal is considered. The anisotropy is due to optical orientation of tunneling centers caused by absorption of polarized resonant radiation. The ellipsoid of polarization rotates through the large angle and the degree of polarization increases substantially under relatively weak fields when the crystal is thick enough. The effects depend strongly on the detuning of the radiation frequency from the centre of the impurity absorption band. The resonant self-transparency is analysed.

The orientation of tunneling defects (off-center impurity ions or molecules) in cubic crystals induced by resonant linearly polarized radiation was observed in a number of experiments [1,2]. The possibility of resonant optical orientation (ROO) at comparatively weak fields is due to the tunneling rate in the excited state (where the impurity appears after light absorption) being in large excess over that in the ground state. ROO results in the anisotropy of impure cubic crystals. The anisotropy in its turn influences the propagation of orienting radiation. Consequently in the range of resonant impurity absorption nonlinear optical effects are to become apparent at weak fields.

To calculate ROO self-consistently we shall use the simple model of a multiwell tunneling center [3]. It is supposed in this model that at sufficiently low temperature the impurity is in the ground state in one of equivalent wells and resonant light may induce the transition to the excited state in the same well only (when radiation is absent the impurities are equally distributed over the wells; the tunneling has been ignored yet). The direction of the dipole transition moment d_n in the well n is supposed to coincide with the symmetry axis of the intrawell potential. Then, neglecting the host crystal absorption in the range of resonant impurity absorption, Maxwell equations for slowly varying envelope E of the radiation electric field under stationary conditions may be presented as

follows

$$(k \nabla) E = 2\pi i [k^2 P - k(kP)];$$

$$P = (\kappa/2\pi k) \sum_n d_n (d_n E) \rho_n(E) / (fd^2),$$

$$d^2 = |d_n|^2, \quad f = \sum_n d_{nx}^2 / d^2.$$

Here k is the wave vector, $k^2 = (\omega^2/c^2)\epsilon$, κ is the resonant impurity contribution to the complex refractive index in the absence of ROO, $|\kappa| \ll k$, $\rho_n(E)$ denotes field-dependent population of the n th well ($\sum_n \rho_n = 1$). Eq. (1) is correct when the field distribution is smooth and second derivatives may be neglected.

In fact the function $\rho_n(E)$ describes the impurity orientation by resonant field. It is shown in [3] that the tunneling having been taken into account the ROO kinetics is determined by diffusion equation^{*}

$$\partial \rho_n / \partial t = - \sum_m (C_{mn} \rho_n - C_{nm} \rho_m), \quad \rho_n = \rho_n(E) \quad (2)$$

for a variety of strongly and weakly bound tunneling

* Such a description is valid if interwell transitions are jump-like, i.e. the duration of the reorientation act itself is small as compared with the characteristic time C_{mn}^{-1} of the well population change.

centers. In the range of relatively weak fields the occupation of excited states is small and

$$C_{mn} = A_{mn}^{(0)} + A_{mn}^{(1)} |(d_m E)|^2 \kappa''/N, \quad \kappa'' = \text{Im} \kappa. \quad (3)$$

The reorientation probability (3) presents formally the sum of probabilities of interwell jumps in the ground ($A^{(0)}$) and excited ($A^{(1)}$) intrawell states ($|(d_m E)|^2 \kappa''/N$ determines the occupation if the m th well excited state, κ''/N is the impurity concentration). In fact parameters $A^{(0)}$ and $A^{(1)}$ may have another sense [3]. For example eqs. (2), (3) are valid if the energy of the excited state exceeds the interwell barrier height and excited impurity is delocalized: practically it is simultaneously in all wells.

The wells equivalent with respect to inversion are evidently equally occupied in the high-frequency field. Then it follows from the symmetry arguments that the ROO kinetics of the cubic crystal defects with the symmetry of wells $\langle 100 \rangle$ or $\langle 111 \rangle$ is described by two independent parameters $A^{(0)}$ and $A^{(1)}$ only [3], while the stationary distribution $\rho_n(E)$ is described by the single parameter $a = (A^{(1)}/A^{(0)}) \kappa'' a^2/N$. The field-induced changes in the occupation of wells being essential the nonlinear optical effects are not small. However, the characteristic length where both the linear (absorption) and nonlinear effects manifest themselves appears to be large, $|\kappa|^{-1} \gg \kappa^{-1}$. Therefore if the incident light beam is wide enough, $\nabla E^2 < \sqrt{\kappa/k} E^2$ and eq. (1) is valid, the most prominent nonlinear effects are the rotation and change of the ellipsoid of polarization and the self-focusing. Self-induced bending of the light beam is negligible. It may be shown easily that $\text{div } k \sim \kappa^2$, i.e. the light propagation direction is changed when the propagation length is of the order of k/κ^2 and hence exceeds substantially the absorption length $1/\kappa''$.

To illustrate the change of polarization we shall consider the radiation propagating in the $\langle 001 \rangle$ direction (z -axis). Taking into account the expressions for ρ_n [3] and eq. (1) one obtains for $\langle 111 \rangle$ centers the following equations

$$\frac{dE_\mu}{dz} = i\kappa E_\mu \frac{1 + 2a|E_\mu|^2}{1 + a|E|^2}, \quad E_\mu = \frac{1}{\sqrt{2}}(E_x - (-1)^\mu E_y),$$

$$|E|^2 = |E_1|^2 + |E_2|^2, \quad \mu \neq \mu'; \quad \mu, \mu' = 1, 2, \quad (4)$$

while for 100 centers

$$\frac{dE_\mu}{dz} = i\kappa E_\mu \frac{1 + 3a|E_\mu|^2/2}{1 + a|E|^2 + 3a^2|E_1 E_2|^2/4},$$

$$E_1 = E_x, \quad E_2 = E_y, \quad \mu \neq \mu'; \quad \mu, \mu' = 1, 2. \quad (5)$$

The solution of eqs. (4) is

$$E_\mu(z) = \bar{E}_\mu(z) \exp[i\varphi_\mu(z)]; \quad \text{Im} \bar{E}_\mu = 0;$$

$$\bar{E}_1(z) \bar{E}_2(z) = \bar{E}_1(0) \bar{E}_2(0) \exp(-2\kappa'' z).$$

$$\bar{E}_2(z)/\bar{E}_1(z) = [\bar{E}_2(0)/\bar{E}_1(0)]$$

$$\times \exp\{-a[\bar{E}_2^2(z) - \bar{E}_1^2(z) - \bar{E}_2^2(0) + \bar{E}_1^2(0)]\}. \quad (6)$$

$$\varphi_1(z) + \varphi_2(z) - [\varphi_1(0) + \varphi_2(0)] = 2\kappa' z, \quad \kappa' = \text{Re} \kappa.$$

$$\varphi_{12}(z) - \varphi_{12}(0) = as\{[\bar{E}_2^2(z) - \bar{E}_1^2(z)]$$

$$- [\bar{E}_2^2(0) - \bar{E}_1^2(0)]\}, \quad \varphi_{12} = \varphi_1 - \varphi_2, \quad s = \kappa'/\kappa''.$$

Parameter s characterized the detuning Ω of radiation frequency from the resonance. For weakly bound centers ($\Gamma \ll \omega_D$) $s = \Omega/\Gamma$, where Γ and ω_D are the half-width of the impurity absorption band and Debye frequency. For strongly bound centers

$$s = \frac{2}{\sqrt{\pi}} \int_0^{\Omega/\Gamma} \exp(x^2) dx \quad (\Gamma \gg \omega_{LO}).$$

It follows from eq. (6) that at $\Omega = 0$ the polarization plane of linearly polarized radiation turns to the nearest direction among $\langle 110 \rangle$, $\langle 1\bar{1}0 \rangle$:

$$\bar{E}_2(z)/\bar{E}_1(z) = [\bar{E}_2(0)/\bar{E}_1(0)]$$

$$\times \exp\{a[\bar{E}_2^2(0) - \bar{E}_1^2(0)]\}, \quad \kappa'' z \gg 1.$$

To understand this rotation it should be taken into account that different field components are absorbed by the centers in wells with different orientations. The absorption of a component is proportional to the occupation of corresponding wells which in its turn is approximately inversely proportional to the intensity of the component (cf. eqs. (2), (3)). Hence the difference

in the absorption of components increases with rising difference in their intensities. The increase in the ratio of components causes monotone (at $\Omega = 0$) growth of the polarization degree (eccentricity $\epsilon(z)$) in case of elliptically polarized radiation. It is seen from fig. 1 that $\epsilon \rightarrow 1$ with the rise in field even at $s \neq 0$, but increases slower at small s .

The eccentricity $\epsilon(z)$ and the angle $\alpha(z)$ between semimajor axis of the ellipse and the direction $\langle 110 \rangle$ in case of $\langle 111 \rangle$ centers or $\langle 100 \rangle$ centers (we have supposed $\bar{E}_1(0) > \bar{E}_2(0)$) are determined by the expressions

$$\epsilon(z) = \left(\frac{1 - \{ |E|^2 - [(\bar{E}_1^2 - \bar{E}_2^2)^2 + 4\bar{E}_1^2\bar{E}_2^2 \cos \varphi_{12}]^{1/2} \}^{1/2}}{|E|^2 + [(\bar{E}_1^2 - \bar{E}_2^2)^2 + 4\bar{E}_1^2\bar{E}_2^2 \cos \varphi_{12}]^{1/2}} \right)^{1/2}$$

$$\alpha(z) = \frac{1}{2} \arctg \left(\frac{2\bar{E}_1^2\bar{E}_2^2 \cos \varphi_{12}}{(\bar{E}_1^2 - \bar{E}_2^2)} \right), \quad \bar{E}_{1,2} = \bar{E}_{1,2}(z),$$

$$\varphi_{12} = \varphi_{12}(z). \quad (7)$$

It follows from eq. (6) that for $\langle 111 \rangle$ centers $\bar{E}_{1,2}(l)$

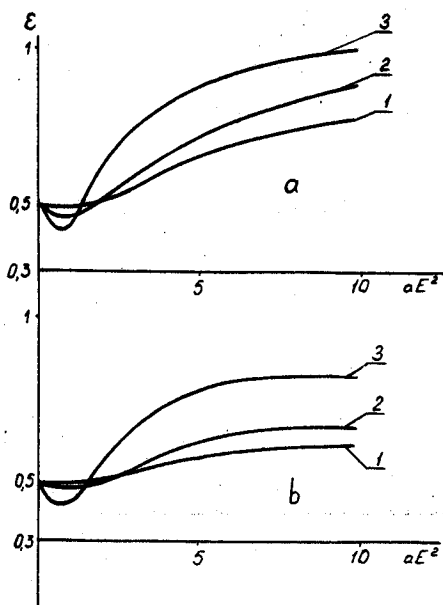


Fig. 1. The dependence of the eccentricity of the ellipsoid of polarization on the incident field intensity for a) $\langle 111 \rangle$ centers; b) $\langle 100 \rangle$ centers. The crystal thickness $l = 1.5/\kappa''$ and $\alpha(0) = 30^\circ$. The curves 1 to 3 correspond to $s = 1, 2, 4$.

$\approx \bar{E}_{1,2}(0) \exp\{a[\bar{E}_{1,2}^2(0) - \bar{E}_{2,1}^2(0)]\}$ in sufficiently thick slabs where $|E(l)|^2 \ll |E(0)|^2$. When a $[\bar{E}_1^2(0) - \bar{E}_2^2(0)] \gg 1$, $\epsilon(l) \approx 1$, $\alpha(l) \approx 0$. The dependences of ϵ and α both on the field strength and the crystal thickness are seen from figs. 1, 2 to be nonmonotonous at $s \neq 0$. Although the oscillations increase with the increase in s , the numerical calculations give $\epsilon(z) > 0.9$ at $|s| \leq 4$ both for $\langle 100 \rangle$ and $\langle 111 \rangle$ centers when incident radiation is linearly polarized. In the case of linearly polarized incident radiation $|\alpha(z)|_{\max} \leq |\alpha(0)|$ according to eqs. (4), (5), (7). In the case of elliptically polarized radiation this is not true but $|\alpha(z)|_{\max} < \pi/4$ (when $\bar{E}_2^2(0) > \bar{E}_1^2(0)$ $\pi/4 < |\alpha(z)| < 3\pi/4$). The dependence of α on the field strength is given in fig. 3 for $\langle 100 \rangle$ centers. Under strong fields ($a\bar{E}_{1,2}^2 \gg 1$) practically all $\langle 100 \rangle$ centers go to those of equivalent wells which are oriented along $\langle 001 \rangle$ or $\langle 00\bar{1} \rangle$ directions and cease to absorb radiation (it is seen from fig. 4 that the self-transparency is much more pronounced for $\langle 100 \rangle$ centers than for $\langle 111 \rangle$ centers). That is why asymptotic values of α and ϵ at strong fields coincide with $\alpha(0)$ and $\epsilon(0)$ (cf. figs. 1, 3). According to fig. 3 (it may be seen also from eq. (7)) α changes much faster with the increase in field in the range of relatively weak fields.

The ROO of $\langle 100 \rangle$ tunneling centers is often in-

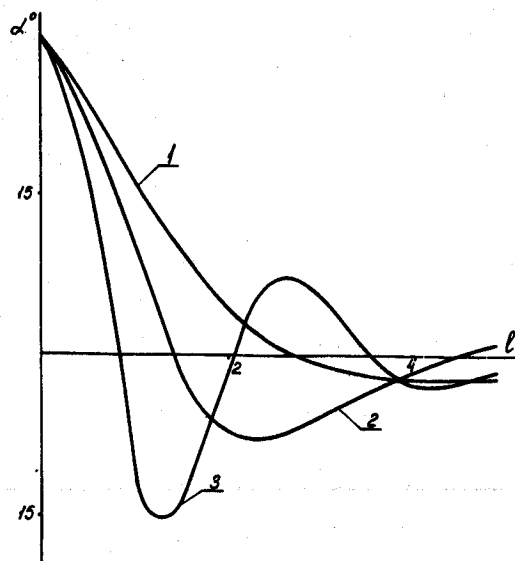


Fig. 2. The dependence of the angle included between the semimajor axis of the ellipsoid of polarization and the $\langle 110 \rangle$ direction on the crystal thickness for $\langle 111 \rangle$ centers. Incident radiation is linearly polarized and $aE^2(0) = 12$. The curves 1 to 3 correspond to $s = 1, 2, 4$.

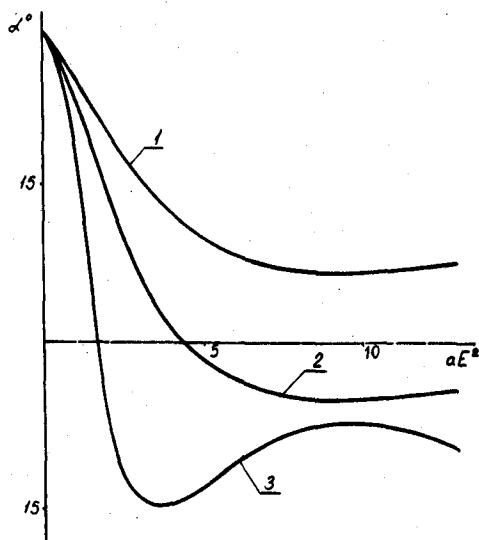


Fig. 3. The dependence of the angle included between the semi-major axis of the ellipsoid of polarization and the (100) direction on the incident field intensity. Incident radiation is linearly polarized. The crystal thickness $l = 1.5/\kappa''$. The curves 1 to 3 correspond to $s = 1, 2, 4$.

vestigated when they are excited to the two-fold degenerate level. The dipole moment of the corresponding intrawell transition is perpendicular to the intrawell potential symmetry axis. The light propagation in (001) direction is described in this case by the equation

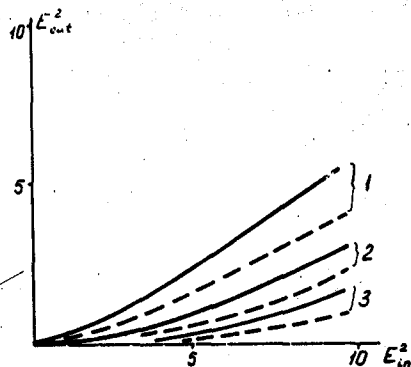


Fig. 4. The dependence of the outgoing radiation intensity on the intensity of incident radiation at $\alpha(0) = 30^\circ$. The full curves refer to (100) centers while the dotted curves refer to (111) centers. In the range of weak fields $E_{out}^2 \approx 0.05 E_{in}^2$. The curves 1 to 3 correspond to $l = 0.75/\kappa''; 1.5/\kappa''; 2.25/\kappa''$.

$$\frac{dE_\mu}{dz} = i\kappa E_\mu \frac{3(1 + a|\bar{E}_\mu|^2) [1 + a|E|^2 + a|E_\mu|^2]}{3 + 4a|E|^2 + a^2[|E|^4 + |E_1 E_2|^2]},$$

$$E_1 = E_x, \quad E_2 = E_y, \quad \mu \neq \mu'; \quad \mu = 1, 2. \quad (8)$$

Eq. (8) is valid if the transitions between degenerate intrawell states of the excited impurity due to relaxation processes [4] occurs much more frequently than interwell transitions. According to eq. (8) the electric vector of linearly polarized radiation turns to the nearest direction among (100), (010) at $s = 0$ (this is true also for the light-induced transitions to non-degenerate level when the rotation is determined by eq. (5)). In the range of weak fields ($a\bar{E}^2 < 1$) the rotation of polarization is much slower when impurities are excited to the degenerate level than to the nondegenerate one.

The change of the radiation polarization is caused by the decrease in absorption of the strong field due to ROO. This mechanism causes also a peculiar self-focusing: the cross-section of the beam propagating in crystal decreases. We shall consider the change of a shape of the one-dimensional ($\partial E(x, y, 0)/\partial y = 0$) linearly polarized beam at $\Omega = 0$. If $E_2(z = 0) = 0$ and $dE_1/dx \ll E_1 \sqrt{\kappa\kappa'}$, we obtain from eqs. (4), (5):

$$\frac{\partial E}{\partial x} = \frac{dE_0}{dx} \frac{E}{E_0} \frac{1 + a|E_0|^2}{1 + a|E|^2},$$

$$E = E_0 \exp[-\kappa''z + \frac{1}{2}a(\bar{E}_0^2 - \bar{E}^2)],$$

$$E_0 = E_0(x, z)|_{z=0} \quad (9)$$

Formula (9) refers also to the cylindrical beam shape, the coordinate x having been replaced by r . It follows from eq. (9) that the self-focusing takes place when $(aE_0^2) > 1$ only. At $z = \frac{1}{2}[aE_0^2 - 1 + \ln(aE_0^2)]$ ($aE^2 = 1$) the beam is the most sharp and then with the increase in z it broadens.

The self-induced anisotropy may be used both to investigate the orientation of the tunneling centers [4] and to find the parameters of tunneling splitting and relaxation in both the ground and excited states of impurities which are difficult to be determined by other means. It should be noticed that this anisotropy restricts the application of ROO for volume holography.

References

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