

Self-induced periodic structures in anisotropic crystals

E. F. Martynovich

Scientific-Research Institute of Applied Physics, Irkutsk State University

(Submitted 18 February 1989; resubmitted 5 May 1989)

Pis'ma Zh. Eksp. Teor. Fiz. **49**, No. 12, 655–658 (25 June 1989)

The mechanisms for the self-excitation of steady-state and time-varying spatially periodic structures during the interaction of anisotropic crystals with optical radiation have been identified. The conditions for observing these structures have been found.

Anisotropic crystals have been finding significantly wider applications in science and technology in recent years. New optical materials, high-temperature superconductors, and laser active media containing quantum systems with a symmetry different from that of the crystal lattice sites have appeared. Among these quantum systems are extrinsic and intrinsic defects with local charge neutralization and color centers. The processes by which optical radiation interacts with crystals and quantum systems of this type have not been studied adequately. The spatial characteristics of the field and the material have not received the study they require. It turns out that they have some interesting features.

In birefringent crystals the polarization state of the resultant light wave varies periodically over distance as a result of a difference between the velocities of natural optical modes.¹ The excitation of absorbing quantum systems under these conditions should lead to a spatially periodic variation of the absorption, accumulation, and emission of energy by the crystal. In an effort to determine whether this method for inducing periodic structures is feasible in principle and to determine the conditions for observing these structures experimentally, we carried out a theoretical study of the distance dependence of the time-average absorbed power $P(r)$, the luminescence intensity in the wave zone, $J(r)$, the concentrations of absorbing centers, and other characteristics. After this theoretical study, we carried out experiments with optically uniaxial crystals, Al_2O_3 and MgF_2 , containing color centers. For comparison we carried out experiments with Al_2O_3 crystals containing cation-substituted ions (Ti^{3+} , Cr^{3+} , etc.). Here are the results of this study.

1. The optical power $P(r)$ absorbed in the linear regime, i.e., at intensities far from the saturation of the transition, does not undergo any periodic changes over distance which would correspond to periodic changes in the polarization state. On the other hand, for each individual orientation of the transition dipole moments in the absorbing centers ($1-3$ at the right in Fig. 1), there are such changes (curves $1-3$). When a sum is taken over all of the orientations allowed by the symmetry laws of the crystals, however, these changes cancel each other out. The total power absorbed by the crystal thus does not oscillate in space (curve 4). This result means that the periodicity of the optical excitation of quantum systems in anisotropic crystals is of a latent nature.

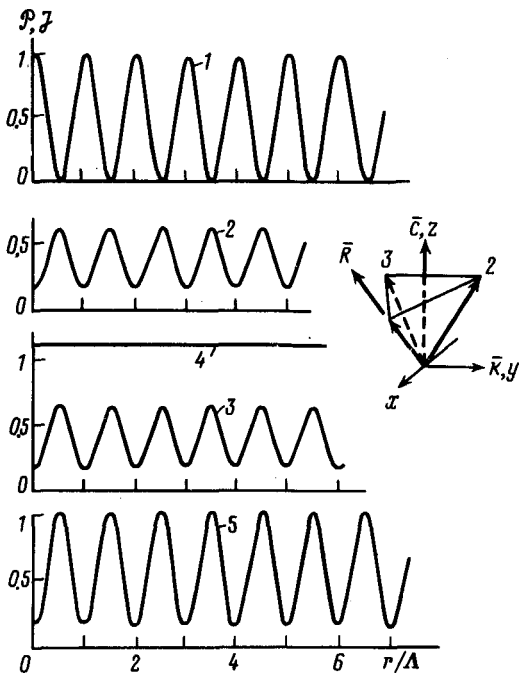


FIG. 1. Spatial profiles of (1-4) the absorbed power and (5) the luminescence intensity for $\beta_\mu = \pi/4$, $\beta_R = \pi/4$, and $\eta_R = 0$.

2. A periodicity of this sort can conveniently be observed by a luminescence method. The intensity of the luminescence over a distance R significantly greater than the dimensions of the crystal is described by

$$J(r) = \frac{3AT_1}{8\pi R^2} \sum_{i=1}^p P^{(i)} q(\beta_R, \eta_R, \beta_\mu^{(i)}, \eta_\mu^{(i)}), \quad (1)$$

where A is the Einstein coefficient for the spontaneous emission, T_1 is the time constant of the dipole-lattice relaxation, p is the number of possible orientations of the centers, and q is the weight factor for orientation i of the centers, given by

$$q = 1 - [\cos \beta_R \cos \beta_\mu^{(i)} + \sin \beta_R \sin \beta_\mu^{(i)} \cos(\eta_R - \eta_\mu^{(i)})]^2. \quad (2)$$

Here β_R and $\beta_\mu^{(i)}$ are the angles between the \bar{c} optical axis and the directions in which the luminescence is observed and the direction of the transition dipole moment in orientation i , respectively; and η_R and $\eta_\mu^{(i)}$ are the angles between the x axis and the projections of the same directions onto the xy plane. The weight factor has different values for different orientations of the centers, so the oscillations corresponding to the individual orientations do not cancel out when a sum is taken in (1). The luminescence intensity thus differs from the absorbed power in that it varies periodically over distance (curve 5).

3. The oscillation period found through a joint solution of the equations of motion for the coordinate components of the dipole moment and the wave equation for an

arbitrary angle β_k between wave vector \mathbf{k} and the optical axis is

$$\Lambda = \frac{2\pi c}{\omega n_{\perp}} \left| 1 + \frac{\chi'_{\perp}}{2n_{\perp}^2} - n_{\parallel} \frac{(n_{\perp}^2 + \frac{n_{\perp}^2}{2n_{\parallel}^2} \chi'_{\parallel}) \sin^2 \beta_k + (n_{\parallel}^2 + \frac{n_{\parallel}^2}{2n_{\perp}^2} \chi'_{\perp}) \cos^2 \beta_k}{(n_{\perp}^2 \sin^2 \beta_k + n_{\parallel}^2 \cos^2 \beta_k)^{3/2}} \right|^{-1}, \quad (3)$$

where n_{\perp} and n_{\parallel} are the principal refractive indices of the crystal when the absorbing centers are ignored, χ'_{\perp} and χ'_{\parallel} are diagonal components of the real part of the dielectric susceptibility tensor χ of the absorbing centers in the coordinate system constructed on the principal axes of the dielectric constant tensor ϵ , ω is the frequency, and c is the velocity of light in vacuum.

4. For Al_2O_3 crystals containing color centers with an absorption band at 455 nm and a known orientation of the transition dipole moment, at an angle of 39° from the \bar{c} axis,² the yellow-green luminescence excited by the beam from a helium-cadmium laser (441.3 nm) consists of a sequence of emitting layers which are transverse with respect to the excitation direction. At small angles β_k , this sequence can easily be observed visually, since the period Λ is quite large. In the case $\mathbf{k} \perp \bar{c}$, observations were carried out with a microscope. A similar picture was observed for the blue luminescence of color centers in MgF_2 (the absorption band at 370 nm), excited by the beam from a nitrogen laser, and also for the yellow and red luminescence excited in various MgF_2 samples by the beam from a helium-cadmium laser. The changes in the oscillation period upon variations in the angle β_k and the frequency ω correspond to expression (3).

5. Under completely identical experimental conditions, we observed no oscillations in the luminescence of Al_2O_3 crystals with cation-substitution impurities. This result is explained on the basis that the transition dipole moments in centers of this sort, having the symmetry of the lattice sites, are oriented either perpendicular or parallel to the optical axis. For such cases ($\beta_k = 0, \pi/2$), the depth of the modulation of the luminescence intensity is zero, as follows from the theoretical curve in Fig. 2. These results show that the orientation of the unknown centers can be determined from the characteristics of the periodic structures, and certain conclusions can be drawn regarding their nature.

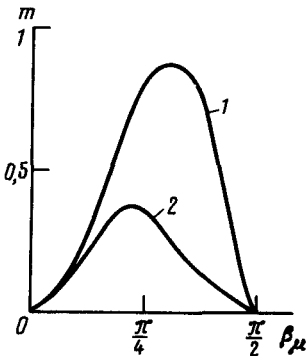


FIG. 2. Depth of the spatial modulation of the luminescence intensity versus the orientation of the transition dipole moment. 1— $\beta_k = \pi/4$, $\eta_R = 0$; 2— $\pi/4, \pi$.

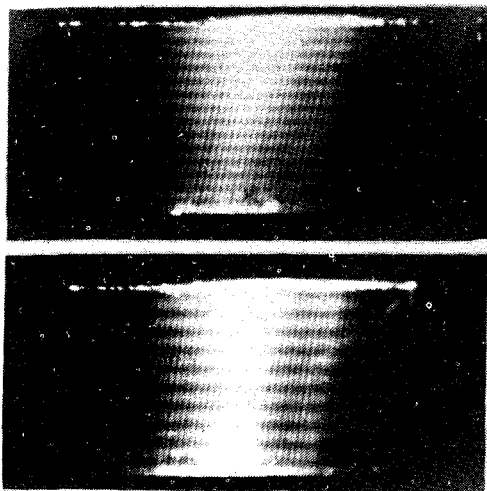


FIG. 3. Periodic structures observed in the luminescence. The optical axis is oriented along the vertical in the plane of the page. Top— $\beta_k = 0.28$ rad; bottom— 0.23 rad.

6. The power absorbed by the crystal begins to oscillate over distance if the natural distribution of the concentrations of centers with respect to orientation is changed through a saturation of the transitions, through a photoburning of the centers, or by some other method. The results caused by these agents are equivalent to a deflection of the principal axes of the tensors ε and χ , which may be thought of as the condition for the occurrence of oscillations. Steady-state periodic structures have been formed at photochemically produced color centers which are responsible for the yellow luminescence of MgF_2 . After the writing, such structures can be seen well, as a sequence of brightly emitting layers of a crystal which is illuminated by unpolarized light from an incandescent lamp through a violet optical filter.

7. The two-dimensional lattices in Fig. 3 were formed at color centers in Al_2O_3 . The horizontal bands formed as a result of the self-induction effect described above, while the vertical bands formed as a result of ordinary interference. We see that a variation in the angle of incidence leads to a change in the period Λ .

¹A. V. Shubnikov, *Fundamentals of Optical Crystallography*, Izd. Acad. Nauk SSSR, 1958, p. 164.

²E. F. Martynovich *et al.*, *Opt. Commun.* **53**, 257 (1985).

Translated by Dave Parsons