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Self-induced oscillations of the polarization of light in KCl crystals containing F_A(Li) centers

S. A. Boĭko, M. Ya. Valakh, M. I. Dykman, M. P. Lisitsa, G. G. Tarasov, and A. M. Shpak

Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR, Kiev

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A new nonlinear effect in crystal optics in the form of self-induced oscillations of the polarization of radiation in cubic crystals was observed experimentally and investigated. It was found that the effect could be used to investigate inversion of the imaginary and real parts of the susceptibility of impurity centers.

It is shown in Refs. 1 and 2 that the orientation and eccentricity of the polarization ellipse of strong radiation in nongyrotropic cubic crystals generally oscillates with the thickness. This effect is associated with the specific features of a self-induced anisotropy of refraction in crystals in which there are certain preferred directions, contrary to isotropic media.[3] Self-induced oscillations of the polarization of radiation traveling along threefold and higher-order axes are a symmetry property and the magnitude of the effect depends on the mechanism of the optical nonlinearity and on the intensity and polarization of the incident radiation. An anisotropy of the nonlinear response of cubic crystals is particularly strong under resonance conditions.[1] It is manifested both in refraction and absorption. In the case of an exact resonance the absorption anisotropy is strongest; it results in a self-induced rotation of the plane of polarization.[4,5] The nonlinear refraction anisotropy and the associated self-induced oscillations of the polarization appear on detuning from resonance. Therefore, by varying the frequency of light within an absorption band we can influence such polarization oscillations and, for a fixed thickness of a crystal, alter the eccentricity and orientation of the polarization ellipse of the output radiation: such changes may oscillate on increase in the frequency. An investigation of the polarization characteristics of the output radiation makes it possible not only to detect the self-induced oscillations of the polarization, but also to determine the spectrum of the real and imaginary parts of the resonant nonlinear susceptibility of a crystal.

We carried out an analysis of the spectral dependence of the polarization characteristics of radiation transmitted by a KCl crystal containing F_A(Li) centers. We selected crystals of sufficiently high optical density ($kd \geq 5$ at the impurity absorption maxima). A self-induced rotation of the plane of polarization of radiation of frequency corresponding to the maximum of a long-wavelength impurity band was reported earlier[6] and such rotation was found to be 40°. In this case we observed self-induced oscillations of the polarization in the wings of the impurity absorption bands and we investigated them.

Our experiments were carried out at 77 K on samples 5 mm thick cleaved along (100) crystallographic planes. Impurity F_A(Li) centers were created in a crystal by a method described in Ref. 7. The source of radiation was a tunable dye laser of the LZhI-504 type covering the relevant range 0.53-0.71 μ. Linearly polarized laser radiation was directed along the [001] axis of a crystal. A standard method was used to determine the following characteristics of the elliptically polarized output radiation: the angle α between the major axis of the polarization ellipse and the $x \parallel [100]$ axis; the angle Ψ governing the ratio of the field components $|E_y|$ and $|E_x|$ ($\tan\Psi = |E_y/E_x|$; $y \parallel [101]$ axis); the difference between the phases of the components $\phi = \arg(E_y/E_x)$.

The spectral dependences of the parameters α, Ψ, and ϕ determined under steady-state conditions for three orientations of the plane of polarization of the incident light $\alpha_0 = \Psi_0$ are represented by points in Figs. 1-3. We can see that the self-induced anisotropy makes the output radiation elliptically polarized and the orientation of the ellipse oscillates with the frequency. It is important to note that in certain spectral intervals the angle α becomes negative (the major axis of the ellipse passes through the [100] axis) and then the difference between the phases of the components ϕ exceeds $\pi/2$. This is evidence of the self-induced oscillations of the polarization. In fact, as shown earlier[5,8] and demonstrated in Fig. 1, the ratio of the amplitudes of the field components $|E_y|$ and $|E_x|$ varies monotonically with the thickness of the crystal and it is found that $\tan\Psi = |E_y/E_x|$ decreases if $\tan\Psi_0 < 1$. Therefore, in the case of a fairly thick crystal the eccentricity of the polarization ellipse tends to unity and the major axis of the ellipse becomes aligned closer to the [100] axis. Consequently, the change in the sign of α at some finite thickness of a sample is clear evidence of the oscillations of the polarization.

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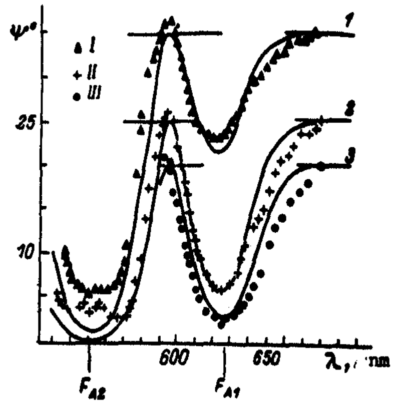

FIG. 1. Spectral dependences of the polarization parameter for radiation transmitted by a KCl crystal containing F_A(Li) centers. Theoretical curves 1-3 are plotted for three azimuths of the linear polarization of the incident radiation Ψ_0 = 35, 25, and 20°, respectively (ℓ = 0.98). Experimental values of Ψ_0 (degrees): I) 35; II) 25; III) 20.

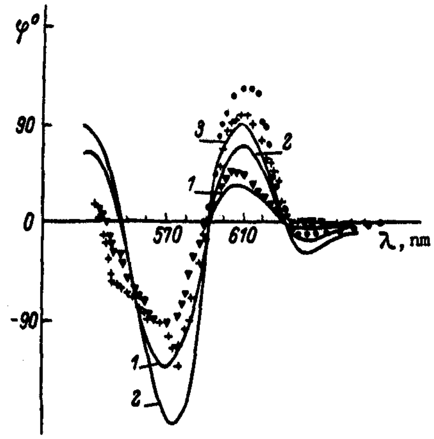

FIG. 3. Spectral dependences of the phase shift ϕ for three different values of Ψ_0. Curves 1-3 have the same meaning as in Fig. 2.

The qualitative difference between the self-induced oscillations of the polarization and the known (Ref. 9) oscillations of the polarization in birefringent crystals is that in the self-induced anisotropy case both the value of the phase shift between the field components E_y and E_x and the sign of this shift are governed by the relationship between these components (since the directions [100] and [010] are equivalent), whereas in the case of birefringence the phase shift in linear optics is governed only by the difference between the refractive indices of the ordinary and extraordinary rays. The change in the sign and in the magnitude of the phase shift due to the self-induced anisotropy are plotted in Fig. 4 as a function of the orientation of the plane of polarization of the incident radiation.

The observed self-induced optical anisotropy is due to the characteristic features of the susceptibility of the F_A(Li) centers in KCl. Optical excitation may result in reorientation of such centers[7] and polarized radiation produces a nonuniform distribution of the centers in respect of the orientation, i.e., it makes the crystal optically anisotropic as a whole. A description of the spectral dependences of the parameters Ψ, α, and ϕ is given by a theory developed for the investigated system in Refs. 5 and 8. The theoretical results based on the Gaussian approximation for the absorption band profiles F_{A_1} and F_{A_2} with the parameters found in Ref. 7, deduced allowing for the breakdown of the selection rules in the F_{A_1} band caused by an induced local vibration,[8] are shown as continuous curves in Figs. 1-4. According to Ref. 8, the change in the ratio of the moduli of the components $\tan\Psi = |E_y/E_x|$ during propagation in a crystal is independent of the phase relationships and is governed by the ratio $\sigma_\parallel/\sigma_\perp$ of the cross sections for the absorption of light polarized along and across the axes of the centers [it is this parameter that determines the anisotropy of the distribution of the F_A(Li) centers in respect of the orientations]. The advance of the phase ϕ depends both on $\sigma_\parallel/\sigma_\perp$ and on the difference $(\chi'_\parallel - \chi'_\perp)$ between the real parts of the susceptibility. The complete change in ψ and ϕ in a crystal is governed by the ratio of its thickness to the impurity absorption length $\ell \propto (2\sigma_\parallel + \sigma_\perp)(\sigma_\parallel - \sigma_\perp)^{-2}$. The relationship is only the fitting parameter used in the calculation of the curves plotted in Figs. 1-4.

In accordance with the above description, in the vicinity of the isotropic point where $\sigma_\parallel = \sigma_\perp$,

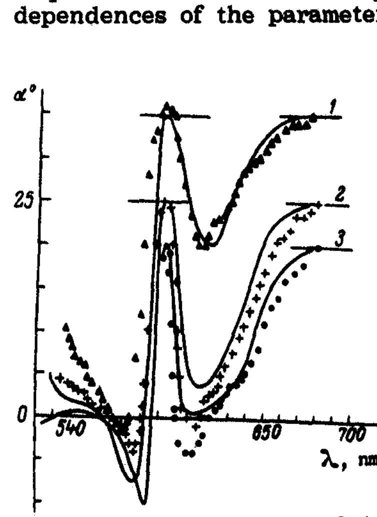

FIG. 2. Spectral dependences of the orientation α of the major axis of the polarization ellipse of radiation transmitted by a crystal for three azimuths of the linear polarization of the incident radiation. Ψ (degrees): 1) 35; 2) 25; 3) 20.

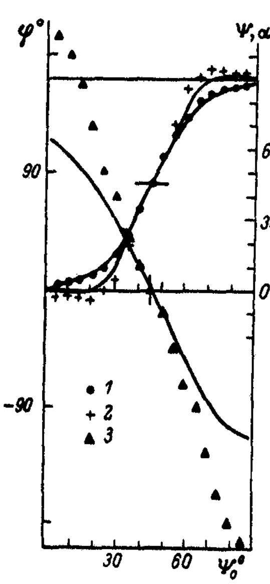

FIG. 4. Dependences of the parameters Ψ, α, and ϕ measured at the wavelength of λ = 610.5 nm on the angle ψ_0 between the plane of polarization of the incident radiation and the [100] axis of a crystal. The experimental values of: 1) Ψ; 2) α; 3) ϕ. The continuous curves are theoretical dependences base on Ref. 8.

the polarization of the radiation in a crystal changes only slightly: $|\Psi-\Psi_0| \propto |\alpha-\alpha_0| \propto |\varphi| \ll 1$. This spectral dependence of the phase shift ϕ has a characteristic form of a dispersion curve for a system with two absorption peaks. It should be pointed out that a determination of ϕ makes it possible to reconstruct directly [see Eq. (6) in Ref. 8] the spectrum of the difference between the real parts of the susceptibility of a center (in a theoretical description it is found using the Kramers–Kronig relationship).

A high-frequency maximum of the change in Ψ (Fig. 1) coincides with the center of the FA_2 band and the low-frequency maximum is shifted relative to the center of the FA_1 band toward shorter wavelengths. The general form of the spectral dependence of the angle α (Fig. 2) is close to the spectral dependence of Ψ, but there are also several important differences. The self-induced oscillations of the polarization can make the angle α negative (in the range $\phi > \pi/2$). Maxima of $|\alpha - \alpha_0|$ are shifted significantly relative to the centers of the FA_1 and FA_2 bands, because they are associated both with the real and imaginary parts of the susceptibility of the centers. The positions of the maxima of $|\alpha_0 - \alpha|$ and of the maxima of $|\phi|$ depend strongly on the orientation of the plane of polarization of the incident radiation α_0.

In spite of the fact that all these features and the maxima of $|\varphi|$, $|\Psi-\Psi_0|$, $|\alpha-\alpha_0|$ in Figs. 1-3 and that the dependences of Ψ, α, and ϕ on Ψ_0 in Fig. 4 are in qualitative agreement with the simple theory of Ref. 8, we can still see some discrepancies. For example, in the long-wavelength wing of the FA_1 band a calculation predicts a steeper fall of $|\Psi - \Psi_0|$ and $|\alpha - \alpha_0|$ than that found experimentally and at the center of the FA_2 band the theoretical values of $|\Psi - \Psi_0|$ are overestimated. We can also see that in the vicinity of the isotropic point ($\sigma_\parallel = \sigma_\perp$) the plane of polarization found experimentally deviates toward the nearest [110] axis. More significant are the differences between the theoretical and experimental values of the phase shift. The deviations of the experimental results from the theory of Ref. 8 may be due to the presence in the investigated crystals not only of the F_A(Li) centers, but also of a relatively large number of defects of other types. Such defects influence on the one hand F_A(Li) centers (the associated inhomogeneous broadening effect is manifested quite differently in the FA_1 and FA_2 bands) and on the other hand they themselves may contribute additionally to the self-induced anisotropy in the investigated frequency range.

Defects in the form of M_A and M_A^+ centers may be important: they are complexes formed from the F_2 or F_2^+ centers and the Li^+ impurity ions. This was confirmed in additional experiments on dichroism induced in an isolated M_A band ($\sim 0.87\ \mu$) by polarized radiation of 0.55 μ frequency in the F band. Calculations indicate that in the vicinity of the isotropic point of the F_A(Li) centers these defects rotate – for the investigated values of Ψ_0 – the plane of polarization of the radiation toward the nearest [110] axis, as indeed found experimentally.

It follows from the results of the present study that the self-induced oscillations of the polarization make it possible to investigate directly the spectral dependence not only of the imaginary but also of the real parts of the susceptibility of the impurity centers in cubic crystals. The nature and magnitude of the self-induced oscillations of the polarization depend strongly both on the nature of the centers and on the characteristics of their dynamics. Therefore, the self-induced oscillations of the polarization are of interest not only as a new effect in nonlinear crystal optics, but also as a possible means for the identification and investigation of impurity centers.

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