SELF-INDUCED POLARIZATION CHANGE IN RESONANT RADIATION IN A DOPED CUBIC CRYSTAL

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

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Some specific effects accompany the propagation of fairly intense radiation in a crystal, which are absent in the nonlinear optics of isotropic media. Some of them occur even in cubic crystals, including the effect considered in [1,2]: self-induced rotation of the plane of polarization SIRPP for resonant radiation and self-induced polarization oscillations SIPO. These effects have some features related to the symmetry, while they are also substantially dependent on the optical-nonlinearity mechanism, and therefore, as shown below, they can be used to determine crystal properties and parameters.

In phenomenological discussion of the optical nonlinearity, the SIRPP and SIPO arise already when one incorporates a term of $E_x|E_x|^2$ type into the expression for the x-component of the nonlinear polarization of third order $\mathbf{P}^{(3)}(\mathbf{r})\exp(-i\omega t)$ plus complex conjugate:

$$P_x^{(3)}(\mathbf{r}) = A_1 E_x |E|^2 + A_2 E_x \mathbf{E}^2 + A_3 E_x |E_x|^2, \quad \mathbf{E} = \mathbf{E}(\mathbf{r}) \tag{1}$$

($\mathbf{E}$ is the field envelope, with the coordinate axes along the fourth-order ones). It is readily shown that purely dissipative nonlinearity, $\mathrm{Re}A_1 = 0$, in the resonant case, as in two-photon absorption if the frequency lies at the center of the absorption band, has the term proportional to A_3 leading to SIRPP, with the plane of polarization propagating in the [001] rotated towards the nearest axis of <100> type for $\mathrm{Im}A_3 < 0$, while it rotates towards the nearest axis of <110> type for $\mathrm{Im}A_3 > 0$.

In the case of nondissipative nonlinearity ($\mathrm{Im}A_1 = 0$), the term proportional to A_3 leads to SIPO. The oscillation period is proportional to $\lambda|A_2E^2|^{-1}$ (λ is the wavelength of the radiation in the crystal). This is very much dependent not only on the parameter ratio A_3/A_2 but also on the degree of polarization and on the orientation of the polarization ellipse for the incident radiation relative to the crystallographic axes [3]. For radiation propagating along the [001] direction, the polarization ellipse oscillates around axes of <110> type for $|1 + A_3/A_2| < 1$, while for $|1 + A_3/A_2| > 1$ it oscillates around axes either of <100> type or <110> type in accordance with the polarization of the incident radiation. In a certain range in the optical-nonlinearity at polarization parameters for the incident radiation, the SIPO is replaced by rotation of the ellipse dependent on the thickness, which is accompanied by pulsations in the degree of polarization (the pulsations are absent in an isotropic medium [4]).

The microscopic theory of SIRPP and SIPO has been given in [1,2] for the case where the radiation frequency lies in the impurity absorption band. SIPO arises at a certain distance from the band center. Because of the absorption, the polarization oscillations are damped as the thickness increases [2].

Optical nonlinearity is particularly large on resonant excitation of tunneling centers, including self-induced optical anisotropy, i.e., for centers having several equivalent equilibrium positions in the unit cell. As the resonant absorption in a center is anisotropic, and the centers usually reorient much more readily in the excited state, a polarized resonant radiation enables one to produce an uneven distribution over the equivalent positions, which in turn influences the propagation of the radiation itself.

SIRPP has been observed for crystals containing tunneling centers [5,6]; the measure-

ments in [5,6] were made for KCl crystals containing F_A(Li) centers, which were resonantly excited by an He-Ne laser.

We have examined the spectral dependence of SIRPP for F_A(Li) centers in KCl and have observed a new effect: self-induced polarization oscillations as the exciting frequency varies.

A F_A(Li) center in KCl is a combination of a F center and a lithium ion replacing an adjacent K^+. The absorption spectrum contains two bands F_A1 and F_{A2}, which correspond to transitions to nondegenerate and doubly degenerate excited electronic states. In a simple model, these transitions are excited by radiation polarized parallel and perpendicular to the axis of the center correspondingly. However, it has been shown for SIRPP produced by radiation lying in the F_{A1} band [5] that violations of the polarization-selection rules are important.

To elucidate the selection-rule violation mechanism, we examined the SIRPP spectrum; theoretically, self-induced polarization change for radiation propagating along the [001] axis is described by the following equations when allowance is made for the self-induced refraction anisotropy at the band flanks:

$$\frac{\operatorname{tg} 2\psi(d)}{\operatorname{tg} 2\psi(0)}\left[\frac{\cos 2\psi(0)}{\cos 2\psi(d)}\right]^{\beta} = \exp(-d/l),$$

$$\varphi(d)=\varphi(0)+\frac{\zeta_1'-\zeta_2'}{\zeta_1''-\zeta_2''}\ln[\operatorname{tg}\psi(d)/\operatorname{tg}\psi(0)], \qquad (2)$$

$$\beta=\frac{1}{4}\,\frac{(\zeta_1''-\zeta_2'')^2}{\zeta_2''(\zeta_2''+2\zeta_1'')}, \quad l=\frac{2\zeta_1''+\zeta_2''}{3(\zeta_1''-\zeta_2'')^2}, \quad d=\frac{1}{2}\operatorname{arctg}(\operatorname{tg} 2\psi\cos\varphi).$$

Here $\operatorname{tg}\psi=|E_y/E_x|$; $\varphi=\operatorname{Arg}(E_y/E_x)$ (the x and y axes lie along [100] and [010]), α is the angle between the major axis of the polarization ellipse and the [100], d is crystal thickness, and the parameters ζ_i' and ζ_i'' are proportional to the real and imaginary parts of the susceptibility χ_i of a center with respect to radiation polarized along the axis of the center ($i = 1$) and perpendicular to it ($i = 2$), with $\zeta_i=\zeta_i(\omega)=\frac{4\pi\hbar\omega}{c\sqrt{\varepsilon}}C_0\chi_i$ (C_0 is the center concentration). It is evident from (2) that the $|E_y/E_x|$ increases or decreases if it is initially larger of less than one, i.e., ψ approximates to the closest of the values 0 and $\pi/2$. Also, (1) indicates that there is no polarization change if the absorption cross section becomes zero for light polarized perpendicular to the axis.

We see from (2) that the phase shift between the components alters for $\zeta_{1,2}' \neq 0$; if however the reduced thickness of the crystal is small, i.e., $d/l \ll 1$, while the incident radiation is linearly polarized ($\varphi(0) = 0$), then $|\varphi(d)| \ll 1$, and the degree of polarization p hardly alters ($\delta p \sim \varphi^2$). At the same time, according to [2], the rotation of the plane of polarization is of the first order in d/l. The spectrum of the maximum angle of rotation (with respect to $\varphi(0)$) is

$$\delta\psi=\frac{d}{4l}(\beta+1)^{-1/2} \qquad (3)$$

and is shown in Fig. 1 for small d/l.

In the region of the F_{A2} band (near the left peak in Fig. 1), the SIRPP spectrum reproduces the absorption band for the F_A(Li) centers [8], which agrees with (2) and (3), since here $\zeta_2'' \gg \zeta_1''$. A characteristic feature of the SIRPP spectrum is the dip down to zero at $\zeta_2'' = \zeta_1''$, i.e., at the frequency where the absorption cross section is isotropic. A further major feature is that the $\delta\psi$ spectrum differs from the absorption spectrum of F_A(Li) centers near the peak in the F_{A1} band. The selection-rule violation mechanism leading to a resonant spectrum for $\delta\psi$ in the region of the F_{A1} band in agreement with experiment is mixing of the wave functions for the degenerate excited electron state of a tunneling center and the wave function of the nondegenerate excited state, which is induced by local vibration.

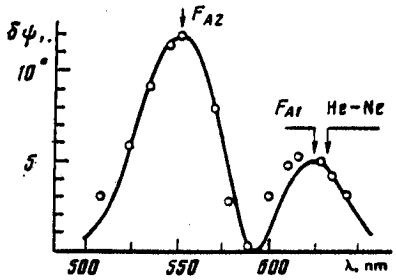

Fig. 1

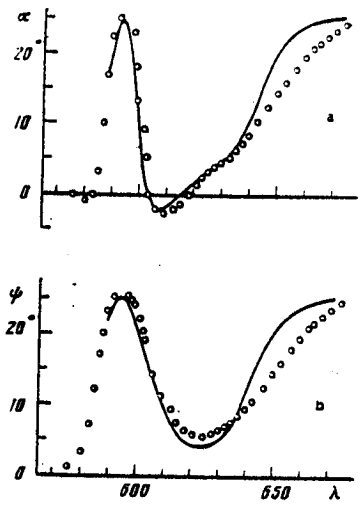

Fig. 2

Fig. 1. Spectrum of the maximum rotation in the plane of polarization for a crystal containing F_A centers; the points are from experiment, while the solid curve is from theory [7]. Relative thickness $d/l = 0.65$.

Fig. 2. Spectrum dependence for the position of the major axis of the ellipse (a) and of the azimuth of the recovered polarization (b) in transmitted radiation for the KCl containing F_A centers for $d/l = 3$; points from experiment, solid line from theory.

At high optical densities, the phase shift $\phi(d) - \phi(0)$ becomes considerable, and one gets self-induced polarization oscillations decaying as the thickness increases. The phase difference is very much dependent on frequency in the resonant region (see (2)), so the polarization of the transmitted radiation in that case oscillates with frequency. If the optical density is $d/l \approx 1$ at the frequency maximum, the frequency dependence of the angle α is aperiodic. It is important however that the quantity can change sign, i.e., the major axis of the polarization ellipse may pass through the <100> direction, which is the limiting one for ψ (the direction in which the plane of polarization rotates at exact resonance).

Figure 2 shows measurements on the oscillation in polarization with frequency, which were obtained for the range covering the F_{A1} band and the low-frequency flank of the F_{A2} one. A LZhI-504 tunable dye laser was used. Linearly polarized radiation having $\psi(0) = 25°$ was incident on the crystal. In the general case, the transmitted radiation was elliptically polarized. We measured three characteristics: the component ratio $|E_y|E_x| \equiv \mathrm{tg}\psi(d)$ (the axes of the Babinet-Soleil compensator were set parallel to the [100] and [010] crystallographic directions), together with the orientation $\alpha(d)$ of the major semiaxis and the phase difference $\phi(d)$.

Figure 2b shows the spectral dependence of ψ, which represents the pure absorption dichroism contribution to SIRPP (see (1)). At the wavelength $\lambda \approx 5925$, which corresponds to the isotropic point $\zeta_1' = \zeta_2'$ we have $\psi(d) \approx \psi(0)$ (compared Fig. 1). The phase difference $\phi(d) - \phi(0)$ changes sign on passage through this point.

Figure 2a shows the spectral dependence of α. In a certain range on the high-frequency flank of the F_A1 band, α is negative, which is an unambiguous sign of the SIPO effect. Here $\phi(d)$ exceeds $\pi/2$.

The relationships are closely described by (2), in which the shape of the absorption bands was taken as gaussian (we used the results of [7] for ζ_2'' in the region of the F_{A1} band, while ζ_1' was expressed in terms of ζ_1'' by means of the Kramers-Kronig relation).

These results show that these new nonlinear optical effects (SIRPP and SIPO) not only enable one to determine the resonant-absorption spectra without measuring it directly but also to examine detailed features of impurities that lead to violation of the selection rules for resonant absorption in the particular case of F_A(Li) centers in KCl.

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Semiconductors Institute, Academy of Sciences
of the Ukrainian SSR