

Saturation of absorption and rotation of polarization of radiation by local vibrations in cubic crystals

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(Submitted December 22, 1976)
Zh. Eksp. Teor. Fiz. 72, 2246-2255 (June 1977)

The separation of the absorption of linearly polarized resonant radiation by anharmonic local vibrations of cubic symmetry is analyzed by the method of the quantum kinetic equation. It is shown that as a result of the symmetry-induced degeneracy of the energy levels the absorption saturation is accompanied by rotation of the radiation polarization towards one of the symmetry axes of the crystal. The direction and rate of rotation are determined by the ratio of the relaxation parameters of the local vibration. An analogous rotation is investigated in the case of tunnel centers. The extreme direction of the polarization is determined in this case by orientation of the centers.

PACS numbers: 63.90.+t

I. INTRODUCTION

Absorption of resonant radiation by a two-level system decreases with increasing intensity of the radiation, owing to equalization of the level populations.^[1] Inasmuch as the absorption saturation in the absence of level degeneracy is determined by the level lifetime, this saturation is frequently used to find the relaxation parameters of the impurity state in the crystals. Lee and Faust^[2] have attempted to measure by this method the lifetime of the local vibration (LV) in the $\text{CaF}_2:\text{H}^-$ system. This LV has cubic symmetry^[3] and the crystal is isotropic in weak fields. However, owing to the level degeneracy of the LV, the observed absorption saturation,^[2] as will be shown below, should be accompanied generally speaking by rotation of the plane of polarization of the strong^[1] resonant radiation.

Self-induced resonant polarization rotation (SRPR) is due to the specifics of the relaxation of systems with a degenerate energy spectrum. We analyze the SRPR using as an example a local vibration of cubic symmetry (LVCS). The first LVCS excited level is triply degenerate, and its wave functions transform like x, y, z (Fig. 1). Assume that the radiation propagates in the z direction, $E_z = 0$. If the states $|x\rangle$ and $|y\rangle$ were to relax independently of each other, then their occupation would be determined by the E_x and E_y components of the field, respectively. Since in this case the weaker of the components would be more strongly absorbed, the field intensity vector would rotate, in the course of propagation in the crystal, towards the closer of the directions x or y . Actually the probability of field-induced transitions in the presence of degeneracy depends not only on the occupation numbers but also on the ratio of the phases of the wave functions (cf. the harmonic-oscillator "paradox").^[4,5] Therefore the velocity and the direction of the SRPR are determined by the ratio of the relaxation times of the occupation numbers of the states and their mutual coherence.

The interaction of LVCS with radiation can be treated within the framework of a two-level system if the anharmonicity of the LM is relatively large, so that the associated non-equidistance of the levels is $\Delta\omega \gg \gamma$ (γ is

the characteristic level width). The field frequency ω_f should be close to the frequency ω_0 of the transition from the ground to the first excited level, $|\omega_f - \omega_0| \sim \gamma$, and the field must not be too strong, so as to be able to neglect the occupation of the higher levels.

To describe the relaxation we use in the present study the model of a weakly-coupled local (quasilocal) vibration^[6,7] (see also^[3]). The impurity concentration is assumed small, and their interaction is disregarded. In Sec. 2 we obtain and analyze the quantum kinetic equation at $T \ll \hbar\omega_0$. We consider the saturation of the absorption and the interference of the states of the LVCS. In Sec. 3 we investigate the rotation of the polarization of strongly resonant radiation in cubic crystals containing LVCS or tunnel centers. In Sec. 4 is analyzed an experiment on absorption saturation.^[2] It is shown that the parameters of the LVCS and the orientation of the tunnel centers can be determined with the aid of SRPR.

2. KINETIC EQUATION. ABSORPTION SATURATION

There are several known mechanisms of broadening of the spectral distribution of local vibrations. For high-frequency vibrations ($\omega_0 > \omega_m$, where ω_m is the maximal oscillation frequency of the continuous spectrum of the crystal), the most important role^[3,6-9] is played by the decay of the LV with phonon emission and by broadening due to elastic or quasi-elastic scattering of the phonons by the LV (the modulation mechanism). The kinetic equation for the density matrix, $\hat{\rho}(t)$ of the anharmonic LVCS, can be determined in second order in the interaction with the phonons by using the method of integral operator equations,^[10] as was done previously for LV that are not degenerate in frequency.^[11,12] At $T \ll \hbar\omega_0$ this equation is of the form

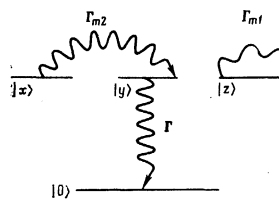


FIG. 1. Ground and first excited levels of local mode of cubic symmetry. The wavy lines show the types of relaxation processes.

$$\begin{aligned}
\frac{\partial \rho}{\partial t} = & -\Gamma \sum_{\kappa} (n_{\kappa} \rho + \rho n_{\kappa} - 2a_{\kappa} \rho a_{\kappa}^{\dagger}) - \hat{\Gamma}_{m} \rho - iV_1 \sum_{\kappa} [n_{\kappa} (n_{\kappa} - 1), \rho] \\
& - iV_2 \sum_{\kappa \neq \kappa'} [n_{\kappa} n_{\kappa'}, \rho] - iV_3 \sum_{\kappa \neq \kappa'} [a_{\kappa}^{\dagger 2} a_{\kappa'}^2, \rho] \\
& + i \sum_{\kappa} [f_{\kappa}^*(t) e^{-i\omega_{\kappa} t} a_{\kappa} + f_{\kappa}(t) e^{i\omega_{\kappa} t} a_{\kappa}^{\dagger}, \rho]; \\
\rho = & \exp \left[i\omega_0 t \sum_{\kappa} n_{\kappa} \right] \bar{\rho} \exp \left[-i\omega_0 t \sum_{\kappa} n_{\kappa} \right], \quad n_{\kappa} = a_{\kappa}^{\dagger} a_{\kappa}.
\end{aligned} \quad (1)$$

Here $\kappa = 1, 2, 3(x, y, z)$ number the local vibrations of the LVCS; $f_{\kappa} = qE_{\kappa}(2m\omega_0\hbar)^{-1/2}$ (q is the effective charge of the LVCS). The field will henceforth be regarded as linearly polarized: $\text{Im}(f_{\kappa} f_{\kappa}') = 0$.

The meaning of the parameters of the decay and modulation broadenings, Γ and Γ_m respectively, is clear from Fig. 1 (an explicit expression for the operator $\hat{\Gamma}_m \rho$ is given in the Appendix). Γ_{m1} describes the phonon scattering at which the initial and final states of the LVCS coincide; Γ_{m2} describes the transfer of the excitation from one state to the other (with the same energy); the term Γ_{m3} in (A.2) is of interference origin.

Accurate to the terms $V_{1,2,3}$ and $\hat{\Gamma}_m \rho$, Eq. (1) coincides with the equation of a relaxing harmonic oscillator in an external field.^[4] When LV are considered, however, the fourth-order anharmonicity must be taken into account^[3,6-9] (renormalization of $V_{1,2,3}$ and ω_0 as a result of the interaction with the phonons and of the terms $(a_{\kappa} + a_{\kappa}^{\dagger})^3$ in the Hamiltonian of the isolated LV is assumed). In the derivation of the kinetic equation we have discarded small corrections of the type $|V|/\omega_0$ and $(\Gamma + \Gamma_m)/\omega_0$ as well as $|f_{\kappa}|/\omega_0$ (at $|\omega_f - \omega_0| \sim \Gamma + \Gamma_m$, $|V|$). Equation (1) enables us to calculate both the absorption by the local vibrations of a strong field f_{κ} , and the absorption (or enhancement) of an additional weak field^[11] in a stationary or nonstationary regime and at arbitrary

$$|V|/\Gamma, \Gamma_m/\Gamma, |f_{\kappa}|/(\omega_f - \omega_0 + i\Gamma).$$

Consider a monochromatic field $f_{\kappa}(t) = f_{\kappa} \exp(-i\omega_f t)$. We assume the non-equidistance of the LV levels to be relatively large, $|V| \gg \Gamma$ and $V \gg \Gamma_m$ (but $|V| \ll \omega_0$), and the field to be resonant, $|\Omega| \ll |V|$, $\Omega = \omega_f - \omega_0$. Then only the first excited level is occupied at $|f_{\kappa}|^2 (\Gamma + \Gamma_m) / \Gamma V^2 \ll 1$ and $T \ll \hbar \omega_0$. In the stationary regime, the nonzero matrix elements of the operator ρ

$$\rho_{0\kappa} = e^{i\Omega t} \langle 0 | \rho a_{\kappa}^{\dagger} | 0 \rangle, \quad \rho_{\kappa\kappa} = \langle 0 | a_{\kappa} \rho a_{\kappa}^{\dagger} | 0 \rangle, \quad \rho_0 = \langle 0 | \rho | 0 \rangle \quad (2)$$

do not depend on the time ($|0\rangle$ is the ground state of the LVCS). From (1) and (2) we obtain for these elements the system of 10 linear equations

$$\begin{aligned}
(\Gamma_0 - i\Omega) \rho_{0\kappa} = & i \sum_{\kappa'} f_{\kappa'} \rho_{\kappa\kappa'} - i f_{\kappa}^* \rho_0; \\
\Gamma_0 = & \Gamma + \Gamma_{m1} + 2\Gamma_{m2}; \\
2\rho_{\kappa\kappa} [\Gamma + 2\Gamma_{m2} + (\Gamma_{m1} - \Gamma_{m3})(1 - \delta_{\kappa\kappa'})] - & 2\Gamma_{m2} \rho_{\kappa\kappa'} (1 - \delta_{\kappa\kappa'}) \\
- 2\Gamma_{m2} \delta_{\kappa\kappa'} \sum_{\kappa'' \neq \kappa} \rho_{\kappa\kappa''} = & i f_{\kappa} \rho_{0\kappa} - i f_{\kappa}^* \rho_{\kappa 0}; \\
\rho_0 + \sum_{\kappa} \rho_{\kappa\kappa} = & 1.
\end{aligned} \quad (3)$$

The system (3) greatly simplifies in the case when the field is oriented along one of the symmetrical directions. In particular, if $f_{\kappa} = f \delta_{\kappa 1}$, then

$$\begin{aligned}
\rho_{22} = \rho_{33} = & \Gamma_{m2} \rho_{11} / (\Gamma + \Gamma_{m2}); \quad \rho_{\kappa\kappa'} = 0, \quad \kappa \neq \kappa'; \\
\rho_{11} = & \frac{\sigma |f|^2 (\Gamma + \Gamma_{m2})}{\Gamma (\Gamma + 3\Gamma_{m2}) + 2\sigma |f|^2 (\Gamma + 2\Gamma_{m2})}; \quad \sigma = \frac{\Gamma_0}{\Gamma_0^2 + \Omega^2}; \\
\text{Re}(f \rho_{01}) = & -\frac{\Omega}{\Gamma_0} \text{Im}(f \rho_{01}); \\
\text{Im}(f \rho_{01}) = & -\sigma |f|^2 / \left[1 + \frac{2\sigma |f|^2}{\Gamma} \beta_{100} \right]; \\
\rho_{02} = \rho_{03} = & 0; \quad \beta_{100} = (\Gamma + 2\Gamma_{m2}) / (\Gamma + 3\Gamma_{m2}).
\end{aligned} \quad (4)$$

At $f_{\kappa} \sim \delta_{\kappa 1}$ the field induces transitions only into one of the three degenerate excited LV states. This interaction with the phonons makes it possible for the excitations to go over from one degree of freedom to another (see Fig. 1); therefore $\rho_{22} = \rho_{33} \neq 0$ at $\Gamma_{m2} \neq 0$.

In the strong-field limit we have

$$\rho_0 = \rho_{11} = \frac{\Gamma + \Gamma_{m2}}{2(\Gamma + 2\Gamma_{m2})}; \quad \rho_{22} = \rho_{33} = \frac{\Gamma_{m2}}{2(\Gamma + 2\Gamma_{m2})}; \quad \sigma |f|^2 \gg \Gamma. \quad (5)$$

The populations of the states between which the field produces transitions are equal to each other, $\rho_0 = \rho_{11}$. If $\Gamma_{m2} \ll \Gamma$, then $\rho_0 = \rho_{11} = \frac{1}{2}$ (nondegenerate two-level system). On the other hand, if $\Gamma_{m2} \gg \Gamma$, then $\rho_0 = \rho_{11} = \rho_{22} = \rho_{33} = \frac{1}{4}$.⁽²⁾

If the orientation is not symmetrical, the field induces directly the occupation of all the states of the excited LVCS levels. In the case of a strong field, the detailed-balance equations without allowance for the interference of the degenerate states take the form $\rho_0 = \rho_{\kappa\kappa}$. In the case of a more rigorous analysis of the transition probability, which are determined by the matrix elements $\rho_{0\kappa}$ in (3), depends not only on the populations ρ_0 and $\rho_{\kappa\kappa}$ but also on the off-diagonal matrix elements $\rho_{\kappa\kappa'}$. The distinguishing feature of the decay broadening is that, as seen from (1), in the absence of external fields the phase differences of the degenerate LVCS states do not relax at $\Gamma_m = 0$. If the system is acted upon by linearly polarized radiation, then one of the normal coordinates of the LVCS can be directed along the field by a canonical coordinate transformation. Since the operator of the decay broadening transforms like $x^2 + y^2 + z^2$, its form does not change and formulas (4) and (5) are valid also in the new coordinates. It is clear therefore that at $\Gamma_m = 0$ all the degenerate LVCS states are coherent and the system is isotropic. In view of the coherence, the detailed-balance equations for the states are not satisfied and in strong fields we have $\rho_0 = \sum_{\kappa} \rho_{\kappa\kappa}$ (this follows from the unitarity of the indicated coordinate transformation). Modulation broadening leads to a phase mismatch, and at $\Gamma_m \gg \Gamma$ in a strong field we have $\rho_0 = \rho_{\kappa\kappa}$ for all κ . At arbitrary Γ_m/Γ , the expressions for the populations are complicated, for example, at $f_1 = f_2 = f/\sqrt{2}$ and $\sigma |f|^2 \gg \Gamma$ we have

$$\rho_{11} = \rho_{22} = \frac{\Gamma_{\phi} (\Gamma + 2\Gamma_{m2})}{(3\Gamma + 8\Gamma_{m2}) \Gamma_{\phi} + \Gamma (\Gamma + 3\Gamma_{m2})}; \quad \rho_{33} = \frac{2\Gamma_{m2}}{\Gamma + 2\Gamma_{m2}} \rho_{11}; \quad (6)$$

$$\Gamma_{\phi} = \Gamma + \Gamma_{m1} + \Gamma_{m2} - \Gamma_{m3}.$$

The parameter Γ_{ϕ} in (6) determines the damping of the

off-diagonal elements $\rho_{\kappa\kappa'}$.

In the case of a weak field, the power absorbed by the LV is equal to $\sigma|f|^2$. The parameter Γ_0 , which enters in the cross section σ , is the half-width of the weak-field IR absorption line. The saturation of the absorption at $f_{\kappa} \sim \delta_{\kappa 1}$ is described by a relation of the same type as for a nondegenerate two-level system, but the longitudinal relaxation time $(2\Gamma)^{-1}$ is replaced in (4) by $(2\Gamma)^{-1} \cdot (\Gamma + 2\Gamma_{m2})/(\Gamma + 3\Gamma_{m2})$.

If $f_1 = f_2 = f/\sqrt{2}$ or $f_1 = f_2 = f_3 = f/\sqrt{3}$ (polarization along the [110] and [111] axes), the nonlinear susceptibility of the LVCS has also only a diagonal component with imaginary parts $\sigma/[1 + 2\sigma|f|^2\beta/\Gamma]$, where

$$\beta_{111} = \frac{\Gamma + 2\Gamma_0}{3\Gamma_0}; \quad \beta_{110} = \frac{\Gamma_0(3\Gamma + 8\Gamma_{m2}) + \Gamma(\Gamma + 3\Gamma_{m2})}{4\Gamma_0(\Gamma + 3\Gamma_{m2})}. \quad (7)$$

Both at $\Gamma \ll \Gamma_m$ and $\Gamma \gg \Gamma_m$ the saturation of the absorption for different symmetrical polarizations is the same, $\beta_{100} = \beta_{110} = \beta_{111}$. More sensitive to the relation between the parameters is the polarization rotation considered in the next section.

3. SELF-INDUCED ROTATION OF POLARIZATION OF RESONANT RADIATION

If the propagation direction of a plane transverse electromagnetic wave \vec{z} is normal to the symmetry plane of the crystal, then no longitudinal field component is produced. Assuming that $E_s(\vec{z}, t) = \vec{E}_s(\vec{z}) \exp(i\vec{k}\vec{z} - i\omega_f t)$ and regarding the contribution \mathbf{P} of the impurities to the crystal polarization as small, we obtain in the stationary regime

$$\frac{dE_s}{dz} = i \frac{2\pi\omega_f}{c\sqrt{\epsilon}} P_s; \quad P_s = nq (\hbar/2m\omega_0)^{-1} \sum_{\kappa} s_{\kappa} \rho_{\kappa 0}, \quad s_{\kappa} = 1, 2; \quad (8)$$

$$\left| \frac{dE_s}{dz} \right| \ll \frac{\omega_f}{c\sqrt{\epsilon}} |E_s|,$$

where n is the impurity concentration, s_{κ} is the cosine of the angle between the directions of E_s and κ , and κ is the permittivity of the host crystal.

In weak fields the LVCS polarizability tensor has only a diagonal component, $P_s \sim E_s$, and the light propagates in the same manner as in an isotropic medium. On the other hand if the field is strong, the direction of the vector \mathbf{P} does not coincide generally speaking with the field direction, and it is seen from (8) that as the radiation propagates the vector \mathbf{E} will rotate. Let us examine this rotation in the simplest case, when \mathbf{E} lies in the xy plane and $|\Omega| \ll \Gamma_0$, $\Omega = \omega_f - \omega_0$ (exact resonance).

If $\Omega = 0$ and the radiation is linearly polarized, $\text{Im}(E_x E_y^*) = 0$, then, as seen from (3), $\text{Re}(f_1 \rho_{01}) = \text{Re}(f_2 \rho_{02}) = 0$. From this, with allowance for (8) it follows that the ratio of the phases E_x and E_y remains unchanged, i. e., the linear polarization is preserved. The change of the intensity of the κ components of the field with changing distance, according to (8), is equal to

$$\frac{d|E_{\kappa}|^2}{dz} = \frac{4\pi\omega_f n \hbar}{c\sqrt{\epsilon}} \text{Im}(f_{\kappa} \rho_{0\kappa}), \quad \kappa = 1, 2. \quad (9)$$

Solving the system (3) at $E_z = 0$ and $\Omega = 0$, we obtain

$$\begin{aligned} \text{Im}(f_{\kappa} \rho_{0\kappa}) = & -(\Gamma(\Gamma + 3\Gamma_{m2}) |f_{\kappa}|^2 [|f|^2 + a |f_{\kappa}|^2 \\ & + 2\Gamma_0 \Gamma_0] \{ (|f|^2 + 2\Gamma_0 \Gamma_0) [2(\Gamma + 2\Gamma_{m2}) |f|^2 \\ & + \Gamma_0(\Gamma + 3\Gamma_{m2})] + a |f_{\kappa}|^2 |f_2|^2 (3\Gamma + 8\Gamma_{m2}) \}^{-1}; \\ & \kappa, \kappa' = 1, 2; \kappa \neq \kappa'; |f|^2 = |f_1|^2 + |f_2|^2; \\ & a = 2(\Gamma_{m1} - 2\Gamma_{m2} - \Gamma_{m3}) / (\Gamma + 3\Gamma_{m2}). \end{aligned} \quad (10)$$

Equations (9) and (10) make it possible to calculate with a computer the propagation of resonant radiation in a crystal for arbitrary values of the LVCS parameters. The rotation of the polarization can be investigated, however, analytically by the phase-trajectory method. Dividing the equations (9) by each other, we obtain

$$\frac{du_2}{du_1} = \frac{u_2 [u_2 + u_1(a+1) + 2\Gamma_0 \Gamma_0]}{u_1 [u_2(a+1) + u_1 + 2\Gamma_0 \Gamma_0]}; \quad (11)$$

$$u_{\kappa} = |f_{\kappa}|^2 \infty |E_{\kappa}|^2.$$

At $a = 0$ we have $u_2/u_1 = \text{const}$ and the polarization plane does not rotate. The solution of (11) at $a \neq 0$ is

$$u_1 = u_1(0) \left(\frac{1-v}{1-v_0} \right)^{-(a+2)/a} \left(\frac{v}{v_0} \right)^{1/a} + \frac{2\Gamma_0 \Gamma_0}{a} \frac{v^{1/a}}{(1-v)^{(a+2)/a}} \int_0^v dx \frac{(1-x)^{2/a}}{x^{(a+1)/a}}; \quad (12)$$

$$v = u_2/u_1; \quad v_0 = u_2(0)/u_1(0).$$

The variation of the parameter $v = |E_y(z)/E_x(z)|^2$ determines the rotation of the polarization plane as the radiation propagates in the crystal. In the derivation of (11) and (12) it was assumed that $v_0 < 1$, and the condition $a > -\frac{4}{3}$ was used (the latter follows from the inequality $\Gamma_{m1} \geq |\Gamma_{m3}|$, see the Appendix). Then, as seen from (12), $v < 1$ and $u_1 + u_2(a+1) + 2\Gamma_0 \Gamma_0 > 0$, so that the transition from (9) to (11) is possible (at $v_0 > 1$ it is necessary to interchange the positions of u_2 and u_1 in (11) and (12)).

The polarization rotation is fastest in a strong field:

$$\left(\frac{1-v}{1-v_0} \right)^{a+2} = \frac{v}{v_0} \left[\frac{u_1(0)}{u_1} \right]^a; \quad \sum_{\kappa} |f_{\kappa}|^2 \gg \Gamma_0 \Gamma_0. \quad (12a)$$

If $a < 0$, then $v \rightarrow 1$ as $u_1 \rightarrow 0$, i. e., the polarization rotates to the nearest [110] axis as the light propagates in the crystal. On the other hand if $a > 0$, then $v \rightarrow 0$ as $u_1 \rightarrow 0$, i. e., the polarization rotates to the nearest [100] axis. The rate of rotation depends essentially on the parameter a and on the initial angle between the polarization vector and the [100] axis (see Fig. 2).

It is seen from (10) and (12) that the rotation of the polarization is due to modulation processes (as already noted, if $\Gamma_m = 0$ then the LVCS is isotropic at arbitrary Ω). The rotation direction is determined by the ratio of the parameters $\Gamma_{m1} - \Gamma_{m3}$ and Γ_{m2} . At $\Gamma_{m2} > \Gamma_{m1}$ ($a < 0$) the transfer of excitation between degenerate states is faster than the relaxation of their phases. Consequently, an energy flux is produced from the state excited by the larger of the components E_x or E_y to the state excited by the smaller component. Accordingly, the smaller component of the field is less strongly absorbed than the larger component and the field intensity vector is rotated towards the nearest direction [110] (at $a < -1$,

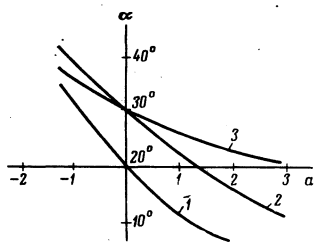


FIG. 2. Dependence of the angle of rotation of the polarization on the parameter $\alpha = 2(\Gamma_{m1} - 2\Gamma_{m2} - \Gamma_{m3})/(\Gamma + 3\Gamma_{m3})$. Curves 1 and 2 correspond to a decrease of the larger of the field components E_x or E_y by a factor of 2, while curve 3 corresponds to a decrease by a factor $\sqrt{2}$. The direction of the polarization of the incident radiation is determined by the value of α at $\alpha = 0$.

the intensity of the smaller component in a definite angle interval increases, although on the whole the field energy is always absorbed according to (10)). If $\Gamma_{m1} \gg \Gamma_{m2}$ and the transfer of excitation can be neglected, then an anharmonic LVCS is equivalent to an aggregate of three non-interacting two-level systems. As noted in the Introduction, in this case the field intensity rotates towards the nearest direction [100].

The SRPR considered here differs qualitatively from the self-induced polarization rotation in liquids.^[13] The latter takes place only for elliptically polarized light, is not a resonance effect, and is observed in the transparency region. SRPR takes place for linearly polarized radiation and is not a rotation but a turning of the polarization toward a definite direction in the crystal. It is interesting to note that as the radiation propagates the degree of polarization of the radiation at $\Omega = 0$ increases (it remains practically unchanged if $\Gamma \gg \Gamma_m$ or $\Gamma \ll \Gamma_m$), and at $\Omega \sim \Gamma_0$ depolarization takes place.

In concluding this section we point out that self-induced polarization rotation takes place also when radiation interacts with local (quasi-local) vibrations of tunnel centers (impurity molecules or noncentral ions).^[14] Since the anharmonicity of such LV is relatively large, it follows that, if tunneling is neglected, the interaction with the resonant radiation can be described within the framework of the two-level model (the time of tunneling of the transition in the ground and in the excited states is assumed to be large in comparison with the energy relaxation time $(2\Gamma)^{-1}$ and the laser-pulse duration). We consider the case when the orientation of the center coincides with the direction of the dipole moment \mathbf{j} of its LM (for example, non-central ions or linear molecules). To calculate the contribution \mathbf{P} of the impurities to the crystal polarization it is necessary to average over the equivalent directions of \mathbf{j}

$$P_x = \frac{\alpha n}{r} \sum_{(j)} j_x \frac{\sum_{m1} j_{m1} E_{m1}}{1 + (2 \text{Im } \alpha / \hbar \Gamma) (\sum_{m1} E_{m1} j_{m1})^2}, \quad (13)$$

where α is the polarizability of one LM, and r is the number of equivalent orientations of \mathbf{j} .

In the case of exact resonance $\text{Re } \alpha = 0$ and the turning of the plane of polarization of the radiation can be analyzed by the phase-trajectory method, as was done above for the LVCS. The extreme directions to which the po-

larization of the strong field ($|E|^2 \gg \hbar \Gamma / \text{Im } \alpha$) turns are listed in Table I for different orientations of the centers and for different radiation propagation directions.

4. CONCLUSION

The determination of the decay and modulation broadening parameters Γ and $\Gamma_{m1,2,3}$ for high-frequency LVCS in a wide temperature interval is of considerable interest, inasmuch as a large number of such impurity vibrations are presently known.^[15] The methods of IR absorption and Raman scattering of light do not make it possible to obtain Γ and Γ_m , since the line half-width at the fundamental frequency is $\Gamma_0 = \Gamma + \Gamma_{m1} + 2\Gamma_{m2}$, and certain other relaxation mechanisms can contribute to the half-width of the overtones.^[3,7] To measure the parameters Γ and Γ_m we can use resonant effects that are nonlinear in the field—separation of the absorption and SRPR, which require relatively low intensities ($|E|^2 \sim m \hbar \omega_0 \Gamma_0^2 / q^2$), which do not influence the host crystal and do not change the energy of the interaction of the local vibrations with the continuous-spectrum vibrations.

The saturation of LVCS absorption was observed by Lee and Faust.^[2] To describe the propagation of the resonant radiation of the crystal they used the expression for nondegenerate two-level systems

$$\frac{d|E|^2}{dz} = -\frac{4\pi\omega_l}{cV\epsilon} \left[\alpha_0'' + \frac{n\alpha''}{1+w|E|^2} \right] |E|^2; \quad w = \frac{2\alpha''}{\hbar\Gamma}, \quad (14)$$

where α_0'' and α'' are the imaginary parts of the susceptibilities of the host crystal and the LM ($n\alpha'' \gg \alpha_0''$). Formula (14) is valid for LVCS, strictly speaking, either in the case when the field is oriented along one of the symmetrical directions of the crystal (this must be a stable direction, i. e., the ones to which the polarization turns), or in the absence of modulation broadening. Lee and Faust^[2] had $\Gamma_m > \Gamma$ at $T = 90$ K. However, (14) describes fairly well the experimental dependence of the crystal transformation on the intensity of the incident radiation, corresponding to two neighboring frequencies of a high-power laser. Assuming the maximum power to be the same at both frequencies, we obtained $\alpha''(\omega_1)/\alpha''(\omega_2) = w(\omega_1)/w(\omega_2)$ with an error less than 10% (for the resonance curve $|\omega_1 - \omega_0| < \Gamma_0/2$, we have $w(\omega_1) |E|^2_{\text{max}} \approx 3.9$, which is approximately 11 times larger than the

TABLE I. Self-induced radiation-polarization rotation due to tunnel centers.

Direction of radiation propagation	Center orientation	Limits of the interval containing the direction of the incident-radiation polarization	Extremal direction of the polarization
[001]	[100]	[110], [110]	[100]
	[110]	[- $\alpha 10$]*, [$\alpha 10$]	[010]
	[111]	[$\alpha 10$], [1 $\alpha 0$] [100], [010]	[110] [110]
[110]	[100]	[111], [001]	[001]
	[110]	[111], [110]	[110]
	[111]	[110], [11 β]**	[111]
	[111]	[11 β], [001] [001], [111] [111], [110]	[001] [112] [110]

* $\alpha = (\sqrt{3} - 1)/\sqrt{2}$.

** $\beta = \sqrt{13}$.

value given by Lee and Faust,^[2] and we therefore do not regard their value of Γ as reliable). The applicability of formula (14) to transmission curves is due apparently to the following: at resonance ($\omega_1 \approx \omega_0$) is so strong (> 98%) that in a greater part of the crystal the polarization is oriented in the symmetrical direction; on the nonresonant curve we have $|\omega_2 - \omega_0| \approx 3\Gamma_0$ and $w|E|^2 < \frac{1}{2}$; since $\Gamma_{m2} > \Gamma$ in this case, it can be shown on the basis of (3) that the polarization dependence is weak.

The rotation of the polarization of strong resonant radiation at $\Gamma_m \neq 0$, as seen from Fig. 2, should occur over the absorption length. The SRPR method is very sensitive to the relation between the parameters of the modulation broadening of the LVCS. By measuring the dependence of the turning angle on the field and on the direction of polarization of the incident radiation, it is possible to obtain from formulas (12) and (12a) the values of $\Gamma_0(\Gamma + \Gamma_{m1} + \Gamma_{m2} - \Gamma_{m3})$ and $a = 2(\Gamma_{m1} - 2\Gamma_{m2} - \Gamma_{m3})/(\Gamma + 3\Gamma_{m2})$. The quantities Γ/β and Γ_0 (see (4) and (7)) can be determined from the frequency dependence of weak-field IR absorption and the field dependence of the saturation of the absorption in the symmetrical configuration. Thus, the aggregate of the methods listed above makes it possible to measure all four relaxation parameters of LVCS.

The SRPR method can be used also to determine the orientation of tunnel centers in cubic crystals. As seen from the table, the limiting direction of the polarization in the case of slow tunneling is uniquely determined by the orientation of the centers. To observe the polarization direction it is necessary to have $T < \hbar\omega_0$, whereas to observe the lowering of the symmetry of the center as a result of application of a static field, the temperature must be lower than the level splitting in the field.

The integrated (with respect to frequency) intensity of the lines of IR absorption by the local vibrations changes somewhat with temperature because of the interaction with the continuous-spectrum vibrations.^[3,16] In the case of a one-dimensional high-frequency LV one can predict that inclusion of the temperature dependence in the effective charge describes the dynamics of the LV in both a strong and a weak field. For LVCS this does not take place because of the scrambling of the degenerate states upon relaxation.

The self-induced resonant turning of the polarization by degenerate two-level impurities in cubic crystals, considered in this article, can be observed not only in the case of local vibrations, but also in the case of electronic centers. To this end it is necessary that the ratio $p/\hbar\gamma(p)$ is the dipole moment of the electronic transition, γ is the width of the weak-field absorption line) be as large as possible and that the saturating field ($|E| \sim \hbar\gamma/p$) not be strong enough to damage the crystal.

The authors thank M. A. Krivoglaz for a discussion of the work.

APPENDIX

Modulation broadening in second order in the interaction is due to the terms

$$H_i = \hbar \sum_{kk'} V_{kk'} a_{k'}^+ a_k^+ a_k a_{k'} \quad (\text{A. 1})$$

in the LVCS Hamiltonian, which is weakly connected with the vibrations of the continuous spectrum (the latter are labeled by the subscript k).^[7] When the symmetry is taken into account, the corresponding operator in the kinetic equation (1) is of the form

$$\hat{\Gamma}_m \rho = \Gamma_{m1} \sum_x \{[n_k^2, \rho]_+ - 2n_k \rho n_k\} + \Gamma_{m2} \sum_{kk'} \{[n_k(n_k+1), \rho]_+ + [a_k^+ a_{k'}^2, \rho]_+ - 2a_k^+ a_{k'} \rho (a_k^+ a_{k'} + a_k a_{k'}^+) \} + \Gamma_{m3} \sum_{kk'} \{[n_k n_{k'}, \rho]_+ - 2n_k \rho n_{k'}\}, \quad (\text{A. 2})$$

$$[A, B]_+ = AB + BA,$$

where

$$\Gamma_{m1} = \sum_{kk'} V_{11kk'}^2 f_{kk'}; \quad \Gamma_{m2} = \sum_{kk'} V_{12kk'}^2 f_{kk'};$$

$$\Gamma_{m3} = \sum_{kk'} V_{11kk'} V_{22kk'} f_{kk'}; \quad f_{kk'} = \pi \bar{n}_k (1 + \bar{n}_k) \delta(\omega_k - \omega_{k'}). \quad (\text{A. 3})$$

Different Γ_m are due to the interaction with the vibrations of the continuous spectrum, which transform in accordance with the different irreducible representations of the symmetry groups. By virtue of the Cauchy inequality we have $\Gamma_{m1} \geq |\Gamma_{m3}|$.

- ¹⁾The field is resonant and strong only for the impurity; the nonlinear polarizability of host crystals can be neglected.
- ²⁾This seems to explain the experimentally observed^[2] absence of a polarization dependence of the occupation of the excited LVCS states (the experiment of^[2] was carried out at temperatures at which $\Gamma_m \gg \Gamma$ according to the estimates of^[8]).
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Translated by J. G. Adashko