[7]V. V. Gladkii and I. S. Zheludev, Kristallografiya 10, 63 (1965) [Sov. Phys. Crystallogr. 10, 50 (1965)].

[8]S. Haussühl, Acta Crystallogr. A 34, 547 (1978).

[9]L. Onsager, D. L. Staebler, and S. Mascarenhas, J. Chem. Phys. 68, 3823 (1978).

[10]V. Dvorak and J. Ishibashi, J. Phys. Soc. Jpn. 41, 548 (1976).

[11]N. R. Ivanov, L. F. Kirpichnikova, V. P. Konstantinova, L. V. Soboleva, and L. A. Shuvalov, Kristallografiya 23, 788 (1978) [Sov. Phys. Crystallogr. 23, 443 (1978)].

Translated by A. Tybulewicz

Spectral dependence of self-induced rotation of the plane of polarization of light in KCl crystals containing F_A(Li) centers

S. A. Boĭko, M. Ya. Valakh, M. I. Dykman, M. P. Lisitsa, G. Yu. Rud'ko, and G. G. Tarasov

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

(Submitted March 20, 1984)

Fiz. Tverd. Tela (Leningrad) 26, 2888–2894 (October 1984)

Theoretical and experimental investigations were made of the spectral dependence of self-induced rotation of the plane of polarization in the case of resonance excitation of reorientable impurity centers in cubic crystals. A complex nonmonotonic spectral dependence was observed. The experimental results on KCl crystals with F_A(Li) centers were in quantitative agreement with the theory.

Propagation of resonance radiation in crystals with reorientable impurity centers is accompanied by self-induced rotation of the plane of polarization (SRPP).[1] The reason for this rotation is self-induced dichroism due to optical alignment of impurity centers.[2,3] The effect was first observed experimentally for F_A(Li) centers in KCl crystals.[4] These centers are of interest because the reorientation time in the ground state of these centers is extremely long even at liquid nitrogen temperatures. However, the probability of reorientation of a photoexcited center is ~ 1/2 (Ref. 3). Therefore, the SRPP appears in such a system even in very weak nonlaser fields.[4,5]

We shall report a study of the spectral dependence of the SRPP and analyze the physical mechanisms responsible for the effect in the case of F_A(Li) centers in KCl. The SRPP mechanism involves breakdown of the selection rules in the case of absorption in an F_{A1} band. Without allowance for this breakdown, the dipole moment of the relevant transition is parallel to the axis of a center, i.e., to one of the crystallographic $\langle 100 \rangle$ axes. Therefore, radiation traveling along the [001] axis should not excite the F_A centers oriented along the axis. Consequently, all the F_A centers would be oriented along the [001] axis (if the field has components along the [100] and [010] axes). This would have resulted in a complete bleaching of a crystal and suppression of the SRPP, which is contrary to the experimental evidence.[4-6]

One of the reasons for the breakdown of the selection rules may be the noncentral position of the Li$^+$ ion in an F_A(Li) center[4] discovered in Ref. 7. In fact, if the equilibrium position of Li$^+$ is displaced from a lattice site, then the local symmetry of the center decreases and light polarized perpendicular to the axis of the center is absorbed in the F_{A1} band (the axis of the center passes through an F center and a lattice site near which an Li$^+$ ion is located).

However, in practice the breakdown of the selection rules need not be due to a noncentral position of the ion in a center. In particular, especially at liquid nitrogen temperatures, it may be associated with phonon-induced mixing of the wave functions of nondegenerate and degenerate excited levels and also with the influence of defects of other types. The spectral dependence of the SRPP allows us to analyze the form of the absorption band forbidden in respect of the polarization.

1. OPTICAL ORIENTATION OF F_A(Li) CENTERS AND SELF-INDUCED CHANGE IN THE POLARIZATION IN A DOPED CRYSTAL

It is shown in Ref. 1 that optical orientation of strongly coupled centers is described by the rate equations

$$\frac{\partial \rho_n}{\partial t} = -\sum_m (C_{nm}\rho_n - C_{mn}\rho_m), \qquad \sum_n \rho_n = 1, \tag{1}$$

where ρ_n is the population of the n-th equilibrium position in a unit cell. The probability of reorientation of C_{nm} then consists of the probability of reorientation in the ground and excited states:

$$C_{nm} = A_{nm} + B_{nm} \sum_{\varkappa\varkappa'} E_\varkappa^* \hat{\alpha}_{\varkappa\varkappa'} (n, \omega) E_{\varkappa'}; \qquad (\varkappa, \varkappa' = x, y, z). \tag{2}$$

Here, $\mathbf{E}$ is the envelope of the electric field of the radiation [the total field is $E = \mathrm{Re}\,\mathbf{E} \exp(-i\omega t)$]; $\hat{\alpha}(n, \omega)$ a tensor which describes the absorption of light at a frequency ω by a center in the n-th orientation [$(\mathbf{E}^*\hat{\alpha}\mathbf{E})$ is the probability of photoexcitation per unit time].

Equations (1) and (2) were derived in Refs. 1 and 8 for the case of resonance excitation when the frequency ω lies within the impurity absorption band. In the case of different absorption bands the relative probabilities of reorientation in an excited B_{nm} state are generally different. In considering the F_A(Li) centers in KCl we shall ignore the difference between B_{nm} for the nondegenerate (F_{A1}) and degenerate (F_{A2}) bands. It is shown in Ref. 3 that the quantum efficiency of the reorientation process is the same for both bands and it amounts to $\approx 1/2$. Moreover, as in Refs. 4 and 5 we shall ignore reorientation in the ground state, i.e., we shall assume that $A_{nm} = 0$.

In the absence of inhomogeneous broadening due to the random field of other defects (neglect of inhomogeneous broadening is justified at moderately low temperatures), the tensor $\alpha_{\chi\chi'}(n, \omega)$ for an F_A(Li) center becomes diagonalized in a coordinate system linked to the axes of the centers and is described by two functions α_1 and α_2 $[\alpha_{1,2} \equiv \alpha_{1,2}(\omega)]$. The parameters α_1 and α_2 govern the absorption cross section of light polarized parallel and perpendicular to the axis of a center. Without allowance for the breakdown of the selection rules we find that α_1 differs from zero for the F_{A1} band, whereas in the case of α_2 this is true of the F_{A2} band. The corresponding maxima of the coefficients $\alpha_{1,2}$ occur at different frequencies. It is important to stress that because of breakdown of the selection rules the coefficient α_2 has a small peak (see the Appendix) in the F_{A1} band.

Since the [100] and [$\bar{1}$00] directions are optically equivalent, it follows that the F_A centers have only three inequivalent orientations along the [100], [010], and [001] axes (along which it is convenient to direct the coordinate axes x, y, and z). It follows from the symmetry considerations that the matrix of probabilities of reorientation C_{nm} has only one independent component and for $A_{nm} = 0$, we have

$$C_{nm} = \tilde{a}_1 |E_n|^2 + \tilde{a}_2 (|E|^2 - |E_n|^2), \quad E_n = (En). \tag{3}$$

Here, $\tilde{a}_{1,2} \equiv \tilde{a}_{1,2}(\omega) = \alpha_{1,2}B_{nm}$; the direction **n** is selected along the axis of a center in the n-th orientation. Equations (1) and (3) obtained allowing for the explicit spectral dependence ρ_n^{st} make it possible to find the distribution of the orientations of the centers as a function of the polarization and frequency of the incident light; clearly, the steady-state distribution ρ_n^{st} is then independent of the intensity.

The reduced Maxwell equations for a doped crystal are of the following form under steady-state conditions:

$$\frac{\partial E_n}{\partial z} = -3E_n[(\xi_1 - \xi_2)\rho_n^{st} + \xi_2]. \tag{4}$$

If, as pustulated below, light travels along the [001] axis, then n = x or y in Eq. (4). The quantities $\xi_{1,2}$ are governed by the resonance polarizability of the reorientable centers. They are related to the absorption cross sections $\alpha_{1,2} \sim \bar{\alpha}_{1,2}$ by

$$\xi_{1,2}' \equiv \mathrm{Re}\,\xi_{1,2} = \frac{4\pi\hbar\omega}{c\sqrt{\varepsilon}}C_0\alpha_{1,2}, \tag{5}$$

where ε is the permittivity of the host crystal and C_0 is the impurity concentration.

The orientation of the centers has the consequence that $\rho_x^{st} \neq \rho_y^{st}$ in Eq. (4) and, consequently, the absorption coefficients and the phase velocities for the field components E_x and E_y are different. Solving Eqs. (1), (3), and (4) subject to Eq. (5), we obtain the following simple transcendental equations describing the SRPP:

$$\varphi(d) = \varphi(0) + \frac{\xi_1'' - \xi_2''}{\xi_1' - \xi_2'} \ln [\mathrm{tg}\,\psi(d)/\mathrm{tg}\,\psi(0)], \tag{6}$$

where

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

$$\beta = \frac{1}{4}\frac{(\xi_1' - \xi_2')^2}{\xi_2'(\xi_2' + 2\xi_1')}, \quad l = \frac{2\xi_1' + \xi_2'}{3(\xi_1' - \xi_2')^2}.$$

Here, the quantities $\psi(d)$, $\psi(0)$ and $\varphi(d)$, $\varphi(0)$ describe respectively the ratio of the absolute values of the field components E_y, E_x and the difference between the phases of these components at the exit (z = d) and entry (z = 0) of the crystal:

$$\psi = \mathrm{arctg}\,|E_y/E_x|, \quad \varphi = \mathrm{Arg}\,(E_y/E_x). \tag{7}$$

The parameters ψ and φ depend in a known manner[9] on the Stokes parameters which describe the polarization of the incident light.

It is clear from Eqs. (6) and (7) that the ratio E_y/E_x varies monotonically with the thickness because $\beta > 0$. Then, irrespective in which band the resonance absorption takes place (i.e., to which of the frequencies $\omega_{1,2}$ is the frequency of light ω closest), the ratio $|E_y/E_x|$ increases if $|E_y(0)/E_x(0)| > 1$ and decreases in the opposite case. Under exact resonance conditions when $\omega = \omega_1$ or $\omega = \omega_2$ and $|\xi_{1,2}''/(\xi_1' - \xi_2')| \ll 1$, the difference between the phase components φ does not change and if the incident light is linearly polarized, it does not become depolarized. The angle between the plane of polarization and the [100] axis deduced from Eq. (7) is ψ for $\varphi = 0$ and -1 for $\varphi = \pi$. It is clear from Eq. (6) that in the course of propagation of light its plane of polarization rotates to the nearest crystallographic axis of the $\langle 100\rangle$ type.[1,4,8] On detuning from resonance the value of φ varies monotonically with the thickness. If $|\xi_1'' - \xi_2''|/|\xi_1' - \xi_2'|$ is sufficiently large, the polarization ellipse oscillates.[8]

The following nontrivial property of the self-induced anisotropy follows from Eq. (6): although a change in the ratio of the field component tan ψ in the $F_{A1}(\omega \approx \omega_1)$ band occurs in the same way as in the F_{A2} ($\omega \approx \omega_2$) band in the range of frequencies where the absorption cross sections α_1 and α_2 are similar, the anisotropy is weak ($l \to \infty$ when $|\xi_1' - \xi_2'| \to 0$). This is due to the fact that the polarized light corresponding to different bands orients the centers in different ways.

The expression (6) simplifies if the reduced crystal thickness d/l is small. Then, the increments in the angles ψ and φ are given by

$$\begin{aligned}
\delta\psi &= \psi(d) - \psi(0) \simeq -\frac{d}{4l}\frac{\sin 4\psi(0)}{1 + \beta \sin^2 2\psi(0)}, \\
\delta\varphi &= \frac{\xi_1'' - \xi_2''}{\xi_1' - \xi_2'}\frac{\delta\psi}{\cos^2\psi(0)}, \quad d/l \ll 1,
\end{aligned} \tag{8}$$

and the maximum (for a fixed crystal thickness and a fixed light frequency) value of $\delta\psi$ is

$$|\delta\psi_{max}| = (d/4l)(\beta + 1)^{-1/2}. \tag{9}$$

Since in the case of a high degree of polarization of the incident light the ellipticity is a quadratic function of the phase mismatch φ (Ref. 9), it is clear from Eq. (8) that the linear polarization of light is hardly affected in the case of weakly anisotropic absorption (d/$l \ll 1$) even far from a resonance. Equations (8), (9), (6), and (5) describe explicitly the spectral dependence of the SRPP if we allow for the familiar (see Ref. 10 and the Appendix) frequency dependences of the absorption cross sections in the F_{A1} and F_{A2} bands.

2. EXPERIMENTAL METHOD

The spectral dependence of the SRPP in KCl crystals with F_A(Li) centers was investigated using samples of

8 × 8 × 6 mm dimensions cleaved along (100) crystallo-
graphic planes. The source was a 30-W incandescent lamp
from which light was directed via a UM-2 monochromator,
a polarizer (Glan-Thompson prism), and a long-focus on
a sample immersed in liquid nitrogen. The F_A(Li) cen-
ters were created in radiation-colored KCl:Li crystals
(20 Mrad dose) by photostimulated conversion of the F to
the F_A centers.[3] The low intensity of the incandescent
lamp light transmitted by the monochromator made it
necessary to employ crystals with a low optical density in
the region of the absorption bands of the investigated cen-
ters so as to ensure an acceptable time for the attainment
of an equilibrium distribution of the orientations of the
centers (we aimed to ensure that the time in question did
not exceed 1 h).

Measurements were carried out as follows. Linear-
ly polarized light traveled along the [001] direction. At
each wavelength we determined the dependence of the
SRPP angle on the polarization azimuth at the entry to a
crystal (a similar curve recorded using an He-Ne laser
of the wavelength λ = 633 nm was reported in Ref. 4).
These curves were used to determine the maximum angle
of rotation at a given wavelength and a relevant point was
then plotted. The experimental error in the determination
of the angle did not exceed 1°. In this situation there was
practically no depolarization.

3. DISCUSSION OF RESULTS

Since the experimentally determined SRPP angles
were small, the results could be interpreted using the
simple and clear expression (9). The validity of Eq. (9)
was confirmed by the practical absence of depolarization
of light even in the wings of the absorption bands. The
experiments confirmed also all the other qualitative fea-
tures of the SRPP predicted by Eqs. (9), (6), and (5) and
by the results in the Appendix. In fact, the experiments
demonstrated clearly two wide SRPP peaks corresponding
to the F_{A1} and F_{A2} bands. These bands were separated
by a dip right down to zero and, according to Eq. (6), this
dip corresponded to the equality of the absorption cross
sections in the two bands (so that l^{-1} vanished even for
overlapping bands). According to Eqs. (9), (5), and (6),
near the maximum of the F_{A2} band ($\omega \approx \omega_2$), and in its
high-frequency wing, where $\alpha_2 \gg \alpha_1$, we should have

$$|\delta\psi_{max}| \approx \frac{3}{2\sqrt{5}}\,\xi_2' \sim a_2, \qquad a_2 \gg a_1. \tag{10}$$

Therefore, the SRPP spectrum and the position of its
maximum should be identical with the corresponding char-
acteristics of the absorption cross section α_2. Using the
results of Ref. 3, we easily showed that this indeed took
place.

Characteristic features were observed in the SRPP
spectrum in the region of the F_{A1} band. As pointed out in
the Introduction, the SRPP in the F_{A1} band is due to
breakdown of the selection rules. This follows directly
from Eqs. (9) and (6): if we ignore the absorption at $\omega \approx \omega_1$
in the degenerate band ($\xi_2' \to 0$), it follows that $\beta \to \infty$ and
$|\delta\psi_{max}| \to 0$. If the absorption in the degenerate band is
weak, so that $\xi_2'/\xi_1' \ll 1$, then

$$|\delta\psi_{max}| \approx \frac{3}{2\sqrt{2}}\,d\,(\xi_1'\xi_2')^{1/2} \sim (a_1 a_2)^{1/2}, \qquad a_1 \gg a_2. \tag{11}$$
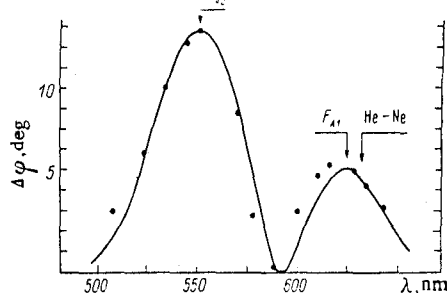

FIG. 1. Spectral dependence of the self-induced rotation of the plane of
polarization in KCl with F_A(Li) centers. The arrows identify the positions
of the absorption band maxima of the F_A(Li) centers at the He-Ne laser
wavelength. The theoretical (continuous) curve is plotted using Eqs. (6) and
(9). The relative thickness is d/l =0.65.

It follows from the results in the Appendix that the ab-
sorption cross section $\alpha_2 \sim \sigma_{\lambda\lambda}(\omega)$ [see Eq. (A.3)] has a
maximum in the F_{A1} band. Even when the simple model
represented by Eq. (A.1) is used, the profile of this band
is different from the profile of the $\alpha_1 \propto \sigma_1(\omega)$ maximum.
However, the difference between these profiles is slight.
Therefore, the spectral dependence of Eq. (11) should be
close to the spectral dependence of the absorption coeffi-
cient of the nondegenerate band $\alpha_1(\omega)$. This is indeed ob-
served experimentally. However, the spectral maximum
of the SRPP is shifted somewhat toward higher frequen-
cies relative to the $\alpha_1(\omega)$ maximum. It follows from the
results in the Appendix that such a shift may be associated
with the nonadiabatic mechanism of breakdown of the
selection rules. The sign of the shift is described cor-
rectly by Eqs. (A.3)-(A.5) and the magnitude of the shift
is greater than that of the frequencies of the local vibra-
tions near an F_A(Li) center[6] (by a factor of about 1.5).
Such a difference may be associated with the approximate
nature of the model described by Eqs. (A.1) and (A.2) or
with the experimental errors.

We shall base a quantitative description of the experi-
mental curves (such as that shown in Fig. 1) on Eqs. (5),
(6), and (9); we shall use a very rough approximation. In
fact, we shall assume that the functions $\alpha_1(\omega)$ and $\alpha_2(\omega)$
in Eq. (5) are Gaussian in the vicinity of the frequencies
$\omega_{1,2}$ and the parameters of the F_{A1} and F_{A2} bands are
those given in Ref. 3. In the range $\omega \lesssim \omega_{min}$ (where ω_{min}
is the position of the dip of the SRPP curve) the expres-
sion for α_2 is that given by Eq. (A.3) with a suitable fac-
tor. The theoretical curve in Fig. 1 is obtained using two
fitting parameters. One of them is the effective thickness
of a crystal d (the concentration of the centers was not
determined experimentally) and the other is the ratio $A_1/$
$(A_0 \triangle_1)$ in Eq. (A.3) representing the shift of the $\alpha_2(\omega)$ peak
relative to the $\alpha_1(\omega)$ peak in the F_{A1} band. The value of
the parameter $A_1/(A_0\triangle_1)$ is assumed to be 0.1 (which cor-
responds to the shift $\delta\omega$, somewhat smaller than the local
vibration frequency). The value of the parameter A_0 is
determined in such a way that the ratio ξ_2'/ξ_1' at the He-
Ne laser frequency is equal to 0.04 obtained in Refs. 4
and 5.

It is clear from Fig. 1 that such a simple approxima-
tion not only provide a qualitatively correct description of
the complex spectral dependence of the SRPP, but also
gives a fairly satisfactory quantitative description of the

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positions of the maxima and dip, of the widths of the maxima, and of their relative intensities.

It is clear from the results of our investigation that a study of the SRPP spectrum provides an effective means of finding the spectral positions of the SRPP maxima and minima (which are of interest in various applications) and it can also be used to determine the microscopic properties of reorientable centers. In particular, in the case of the F_A(Li) centers in KCl the results show that breakdown of the selection rules in the F_{A_1} absorption band is probably due to a phonon-induced mixing of the wave functions of the degenerate and nondegenerate excited electron states.

APPENDIX

We shall consider breakdown in the selection rules for the absorption of light of frequency ω located within the F_{A_1} band. As already mentioned, in the simplest case it is related to the displacement of the equilibrium position of the Li^+ ion from a site in a unit cell.[7] At moderately low temperatures the characteristic times τ_r for the reorientation of Li^+ between equivalent potential minima in a plane perpendicular to the axis of the F_A center are small compared with the reciprocal of the photoexcitation probability and $C_{nm}\tau_r \ll 1$. On the other hand, we have $\omega\tau_r \gg 1$. Therefore, light "senses" an Li^+ ion in an instantaneous displaced equilibrium position and the selection rules break down. The symmetry of a center averaged over a time $> \tau_r$ is C_{4v} and the absorption process can be described by two coefficients α_1 and α_2.

An essentially similar situation arises also in the case when the Li^+ equilibrium position is located at a lattice site. In this case the selection rules break down because of nonadiabaticity.

If we consider only the electron–phonon interaction, which is linear in respect to the phonon creation and annihilation operators b_k^+ and b_k, the Hamiltonian of the systems becomes

$$
\begin{aligned}
&H = H_0 + H_m + H_i + H_i', \\
&H_0 = \omega_1 a_1^+ a_1 + \omega_2 \sum_{\lambda=1,2} a_{2,\lambda}^+ a_{2,\lambda}, \quad H_m = \sum_k \omega_k b_k^+ b_k, \\
&H_i = a_1^+ a_1 \left(\sum_k u_k b_k + \text{H.c.} \right) + \sum_{\lambda,\lambda'} (a_{2,\lambda}^+ a_{2,\lambda'} u_{\lambda,\lambda',k} b_k + \text{H.c.}), \\
&H_i' = \sum_\lambda a_1^+ a_{2,\lambda} v_\lambda + \text{H.c.}, \quad v_\lambda = \sum_k (v_{\lambda k}' b_k + v_{\lambda k}'' b_k^+), \\
&\hbar = 1.
\end{aligned}
\tag{A.1}
$$

Here, a_1^+, a_1, $a_{2,\lambda}^+$ and $a_{2,\lambda}$ are the electron creation and annihilation operators for a nondegenerate and one of two doubly degenerate states (λ labels the directions perpendicular to the axis of a center); ω_1 and ω_2 are the energies of the corresponding levels. The interaction H_i is adiabatic, whereas H_i' is nonadiabatic. It is H_i' that mixes the wave functions of the degenerate and nondegenerate levels and therefore causes breakdown of the selection rules if the Li^+ ion is noncentral.

In the case of a strong band splitting $\omega_2 - \omega_1$ (compared with the band widths $\Delta_{1,2}$) the profile of an absorption peak near the frequency ω_1 for light polarized in the λ direction is described by[10]

$$
\sigma_{\lambda\lambda}(\omega) = \frac{1}{\pi}(\omega_2 - \omega_1)^{-2} \text{Re} \int_0^\infty dt \langle a_1(t) v_\lambda(t) a_1^+(0) v_\lambda^+(0) \rangle, \quad \omega \approx \omega_1. \tag{A.2}
$$

In the strong electron–phonon interaction case when $\Delta_{1,2} \gg \omega_D$ (ω_D is the Debye frequency), the profile of a peak of the function $\sigma_{\lambda\lambda}(\omega)$ can be found quite readily employing the usual method.[10] In the first order with respect to ω_D/Δ_1 the result is

$$
\begin{aligned}
&\sigma_{\lambda\lambda}(\omega) \approx \left(A_0 - A_1 \frac{\partial}{\partial\omega} \right) \sigma_1(\omega), \\
&A_0 = \sum_k (f_{\lambda k}' + f_{\lambda k}''), \quad A_1 = \sum_k \omega_k (f_{\lambda k}' - f_{\lambda k}''), \\
&f_{\lambda k}' = (\omega_2 - \omega_1)^{-2} |v_{\lambda k}'|^2 (\bar{n}_k + 1), \quad f_{\lambda k}'' = (\omega_2 - \omega_1)^{-2} |v_{\lambda k}''|^2 \bar{n}_k, \\
&\bar{n}_k = [\exp(\omega_k/T) - 1]^{-1}.
\end{aligned}
\tag{A.3}
$$

The function $\sigma_1(\omega)$ in Eq. (A.3) gives the profile of a nondegenerate absorption band. When a correction $\sim\omega_D/\Delta_1$ is included, it becomes

$$
\begin{aligned}
&\sigma_1(\omega) = \left(1 - A_3 \frac{\partial^3}{\partial\omega^3} \right) \sigma_1^{(0)}(\omega), \quad \sigma_1^{(0)}(\omega) = (2\pi\Delta_1)^{-1/2} \exp\left[-\frac{(\omega - \omega_1)^2}{2\Delta_1^2} \right], \\
&\Delta_1 = \left[\sum_k |u_k|^2 (2\bar{n}_k + 1) \right]^{1/2}, \quad A_3 = \frac{1}{6} \sum_k |u_k|^2 \omega_k.
\end{aligned}
\tag{A.4}
$$

In Refs. 10 and 11 the spectrum of forbidden transitions was analyzed by the method of moments in the more general case of transitions to degenerate levels. We can show that the moments of the distributions (A.3) and (A.4) are identical with those found in Ref. 11. In the specific case when the nonadiabatic interaction H_i' depends only on the momenta of ions in a crystal, we have

$$
A_1 = (\omega_2 - \omega_1)^{-2} \sum_k \omega_k |v_{\lambda k}'|^2, \quad (|v_{\lambda k}'| = |v_{\lambda k}''|). \tag{A.5}
$$

It is clear from Eq. (A.3) that the profile of the peak which is forbidden in respect of the polarization of the absorption in the nondegenerate band is close to the profile of the $\sigma_1(\omega)$ peak. On the other hand, the peak $\sigma_{\lambda\lambda}(\omega)$ is more asymmetric and shifted relative to $\sigma_1(\omega)$ by

$$
\delta\omega_1 \approx +A_1/A_0. \tag{A.6}
$$

The order of magnitude is given by $\delta\omega_1 \sim \omega_D$. When a local vibration of frequency ω_l appears near an impurity, we have $\delta\omega_1 \sim \omega_l$ [according to Ref. 6, this applies to the F_A(Li) centers]. If breakdown of the selection rules is associated with the noncentral position of the Li^+ ion, then the shifts $\delta\omega_1 \sim \tau_r^{-1}$ and, generally, $\delta\omega_1$ are less than for the nonadiabatic mechanism of the selection rule breakdown.

[1]M. I. Dykman and G. G. Tarasov, Zh. Eksp. Teor. Fiz. 74, 1061 (1978) [Sov. Phys. JETP 47, 557 (1978)].

[2]P. P. Feofilov and A. A. Kaplyanskii, Usp. Fiz. Nauk 76, 201 (1962) [Sov. Phys. Usp. 5, 79 (1962)].

[3]F. Lüty, in: Physics of Color Centers (ed. by W. B. Fowler), Academic Press, New York (1968), p. 181.

[4]M. Ya. Valakh, M. I. Dykman, M. P. Lisitsa, G. Yu. Rudko, and G. G. Tarasov, Solid State Commun. 30, 133 (1979).

[5]M. Ya. Valakh, M. I. Dykman, M. P. Lisitsa, E. V. Pidlisnyi, G. Yu. Rud'ko, and G. G. Tarasov, Fiz. Tverd. Tela (Leningrad) 23, 418 (1981) [Sov. Phys. Solid State 23, 236 (1981)].

[6]B. Fritz, in: Localized Excitations in Solids (Proc. First Intern. Conf., University of California at Irvine, 1967, ed. by R. F. Wallis), Plenum Press, New York (1968), p. 496.

[7]F. Rosenberger and F. Lüty, Solid State Commun. 7, 983 (1969).

[8]M. I. Dykman and G. G. Tarasov, Opt. Commun. 26, 122 (1978).

[9]M. Born and E. Wolf, Principles of Optics, 4th ed., Pergamon Press, Oxford (1970).

[10]Yu. E. Perlin and B. S. Tsukerblat, Effects of the Electron–Vibration Interaction in Optical Spectra of Paramagnetic Impurity Ions [in Russian], Shtiintsa, Kishinev (1974).

[11]L. M. Kushkulei and B. S. Tsukerblat, Fiz. Tverd. Tela (Leningrad) 21, 2254 (1979) [Sov. Phys. Solid State 21, 1297 (1979)].

Translated by A. Tybulewicz