Local electric field in crystals

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Matrix elements of dipole transitions in impurity centers in crystals located in an external electric field are discussed. A feature of the analysis is that the "effective" field in the crystal is considered. It is shown that the usual Lorentz-field approximation is not, in general, valid. Thus, for impurity centers of large radius, the effective field is approximately equal to the external field. In all other cases, the two fields are different. The evaluation of the matrix element is illustrated by considering a simple model. The local Lorentz-field approximation is valid only in the limiting case of a sufficiently small state radius. The relevant criteria are given. A method of determining local electric fields in ferroelectric materials using EPR data obtained in external electric fields is proposed. The method is illustrated by an experiment on EPR in the virtual ferroelectric $KTaO_3$ with a manganese impurity. The observed changes in the EPR line intensities due to the reorientation of compensating vacancy in the external electric field have been used to determine the local field at the sites of the perfect crystal. A theoretical interpretation of the observed effects is given.

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The matrix elements of the dipole moment, which involve the induced local field, play an important role in most physical effects in crystals. We have evaluated these matrix elements in a sufficiently general form and have examined the conditions under which the local field produces an appreciable contribution to the matrix elements. There is, however, a group of effects in which the local field itself plays a crucial role (for example, in phase transitions in ferroelectric materials). In this paper, we propose a method for the approximate determination of the local field.

Our account therefore consists of two parts. The first part is concerned with the evaluation of the electric dipole matrix element for localized impurity electrons, whereas the second part gives a method for determining the local electric fields at the crystal lattice sites.

1. MATRIX ELEMENTS OF ELECTRIC DIPOLE TRANSITIONS

1. The matrix elements of electric dipole transitions in external (variable and constant) electric fields can be written in the form

$$M_{\alpha\beta}=\int \psi_\alpha^*\psi_\beta U(x,y,z)\,d\tau,\quad U(x,y,z)=e\int_s \mathbf{E}(\mathbf{r}')\,d\mathbf{r}', \tag{1}$$

where $U(x, y, z)$ is the work expended in taking a charge e from a chosen origin to the point of observation, s is the path of integration, ψ_α, ψ_β are the wave functions of the impurity center, and $\mathbf{E}(\mathbf{r})$ is the field acting at the point $\mathbf{r}$.

The function $\mathbf{E}(\mathbf{r})$ is usually unknown and, to evaluate the matrix elements given by (1), one uses the Lorentz-field approximation. In this approximation, the field $\mathbf{E}(\mathbf{r})$ is replaced, without adequate justification, by the sum of the external macroscopic field $\mathbf{E}$ and the uniform Lorentz field[1)] $\mathbf{E}_L$ (see, e.g.[1]). In this approximation, the matrix element (1) becomes

$$M_{\alpha\beta}=e(E+E_L)\int \psi_\alpha^* z\psi_\beta\,d\tau, \tag{2}$$

where it is assumed that the external field is parallel to the z axis.

It is clear from (1) that the magnitude of the matrix element is determined by an integration region of the order of the state radius of the localized electron. We note that, for small impurity centers, this may be of the order of a few tens of lattice constants. Moreover, $\mathbf{E}(\mathbf{r})$ is equal to $\mathbf{E}+\mathbf{E}_L$ only at points of high crystal symmetry (lattice sites). This clearly shows that the Lorentz-field approximation is not valid in calculations of matrix elements such as those based on (1).

We shall give an analysis of the role of the effective field in evaluations of matrix elements of electric dipole transitions. We shall define a class of problems for which the Lorentz field is unimportant. We shall show that (2) can only be used in the limit of an infinitesimally small state radius of the localized electron. To illustrate our method, we shall evaluate the matrix elements with the aid of (1), using the following approximations: polarized lattice in the form of a system of point dipoles and hydrogen-like wave functions.

2. The components of the field acting on a localized electron in the crystal placed in a uniform electric field $E=E_z$ can be written in the form

$$E_i(\mathbf{r})=(E+E_L)\delta_{iz}+E_{di}(\mathbf{r}), \tag{3}$$

where $\mathbf{E}_d(\mathbf{r})$ is the field due to the dipoles inside the Lorentz sphere, which appear as a result of the polarization of the crystal.

The matrix element of the dipole transition between state α and β of the local center is

$$\begin{gathered} M_{\alpha\beta}=eEz_{\alpha\beta}+eE_Lz_{\alpha\beta}+\int_{-\infty}^{\infty}\psi_\alpha^*\psi_\beta\int_s \mathbf{E}_d(\mathbf{r}')\,d\mathbf{r}'\,d\tau, \\ z_{\alpha\beta}=\int_{-\infty}^{\infty}\psi_\alpha^* z\psi_\beta\,d\tau. \end{gathered} \tag{4}$$

It is convenient to evaluate the contour integral in (4) along the following path: parallel to the x axis, then parallel to the y axis, and, finally, parallel to the z axis. Since the result of integration along the first two segments is independent of z and $\psi_\alpha^*\psi_\beta$ is an odd function of z, these segments do not contribute to the matrix element and (4) can be rewritten in the form

$$M_{\alpha\beta}=eEz_{\alpha\beta}+eE_Lz_{\alpha\beta}+\int_{-\infty}^{\infty}\psi_\alpha^*\psi_\beta\,d\tau\int_0^z E_{dz}(x,y,\zeta)\,d\zeta. \tag{5}$$

Next, we use the point-dipole approximation in (5). We then have

$$\begin{aligned}\int_0^z E_{dz}(x,y,\zeta)\,d\zeta=&-\sum_k p_{kz}(z-z_k)\left[(x-x_k)^2+(y-y_k)^2+(z-z_k)^2\right]^{-3/2}\\&-\sum_k p_{kz}z_k\left[(x-x_k)^2+(y-y_k)^2+z_k^2\right]^{-3/2},\end{aligned} \tag{6}$$

where k labels the lattice ions and p_k is the dipole moment of the k-th ion.

The qualitative features of the problem can readily be exhibited by replacing the wave functions of the local center with hydrogen-like functions. We note that this approach can be justified for a number of problems in the theory of local centers. The restriction introduced by using the hydrogen-like functions is not fundamental because the method which we shall describe can be used with any wave functions of local centers.

In the chosen approximation, the position matrix element becomes

$$\begin{aligned}M_{\alpha\beta}=&\,eEz_{\alpha\beta}+eE_Lz_{\alpha\beta}\\&-ez_{\alpha\beta}\frac{\gamma^3}{24}\sum_k p_{kz}(1+\gamma r_k)\exp(-\gamma r_k);\end{aligned} \tag{7}$$

where $\gamma=\alpha+\beta$; α and β are the indices of the exponentials in the wave functions of the ground and excited states.

Subject to the adopted approximations, (7) enables us to obtain an exact value for the matrix element. It is clear that this not equal to its value when the matrix element involves only the external field or when the effective field is $\mathbf{E}+\mathbf{E}_L$. These two cases are realized only in the limiting situations of small state radii ($\gamma a \gg 1$) and large state radii ($\gamma a \ll 1$), where a is the distance between the local center and the nearest ion lattice.

It is clear from (7) that, in the first case, in the limit as $\gamma a\to\infty$

$$M_{\alpha\beta}=eEz_{\alpha\beta}+eE_Lz_{\alpha\beta}, \tag{8}$$

i.e., the situation reduces to the replacement of the external field by the sum $\mathbf{E}+\mathbf{E}_L$. For the opposite inequality, we have to a good approximation,

$$\sum_k p_{kz}(1+\gamma r_k)\exp(-\gamma r_k)=4\pi P\int_0^\infty(1+\gamma r)r^2\exp(-\gamma r)\,dr=\frac{32\pi P}{\gamma^3}, \tag{9}$$

where P is the dipole moment per unit volume.

Substitution of (9) in (7) leads to the cancellation of the last two terms. The result is

$$M_{\alpha\beta}=eEz_{\alpha\beta}. \tag{10}$$

It is clear that, for local centers with very large state radii, the Lorentz field provides no contribution at all.

It is interesting to consider intermediate situations in which the effective field differs from the limiting values E and $E+E_L$. Figure 1 shows the ratio of the matrix element (7) to the matrix element in the external field alone as a function of γa, where $\gamma a=(1/r_0+1/r_1)a$, and r_0 and r_1 are the radii of the ground and excited states of the local electron center, respectively. The results shown in the figure were obtained for a simple monatomic cubic crystal by including eighteen coordination spheres surrounding the local center. The contribution of all the other ions of the crystal was taken into account macroscopically (by replacing summation with integration).

Figure 1 illustrates the role of the effective field in the evaluation of the matrix elements. It turns out that, for state radii of the order of the lattice constant, the Lorentz field is already unimportant. It will therefore be necessary to reexamine the results of a large number of papers[1,2] in which the Lorentz-field approximation was used for local centers of sufficiently large state radius (greater than the lattice constant) such as, for example, the F centers.

The opposite situation (external field replaced by $\mathbf{E}+\mathbf{E}_L$) is realized only for state radii amounting to roughly a fifth of the distance to the nearest ion. This may occur, for example, in the case of deep impurity centers belonging to the iron group, and for the ions in perovskite-type lattices. In the latter case, the deviation from cubic symmetry ensures that the local field at the lattice sites is fundamentally different from E_L. A method of obtaining the magnitudes of local fields is described below.

2. DETERMINATION OF LOCAL ELECTRIC FIELDS IN FERROELECTRIC MATERIALS USING THE RESULTS OF EPR MEASUREMENTS IN EXTERNAL ELECTRIC FIELDS

1. As already noted, it is important to know the local electric fields at the lattice sites in the physics of phase transitions in ferroelectric materials and in a number of other branches of physics. Microcalculations of

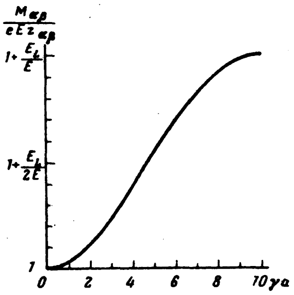

FIG. 1. Ratio of the matrix element (7) to the matrix element for the external yield alone as a function of γa.

these local fields are difficult, and it is therefore particularly important to develop a method capable of deducing this field from experimental data. We shall describe below a method which will enable us to calculate the local field from data on the effect of external electric fields on the EPR line intensities. These intensity variations turn out to be due to reorientations of compensating vacancies in the external electric field. The relative populations of the six types of paramagnetic center oriented parallel, antiparallel, and perpendicular to the electric field depend on the amount of work done during the reorientation of vacancies. It will be shown that this work can be expressed in terms of the local field at the sites of a perfect crystal. The experiments were performed on $KTaO_3$ with Mn^{2+} impurities.

2. EPR of Mn^{2+} in $KTaO_3$ was investigated experimentally in a standard RE-1301 microwave spectrometer at 77 °K. We used rectangular and cylindrical resonators with H_{101} and H_{011} modes. Control measurements were performed in the waveguide. The $KTaO_3$ crystals were grown as before,[3] but the reduced growth rate resulted in an improvement in the crystal quality as well as a lower amount of free carriers. The dopant was MnO_2 in amounts between 0.1 and 1 wt.%. The uncontrollable amount of niobium in the initial material was less than 1 at.%.

We observed the EPR spectrum of the paramagnetic center consisting of Mn^{2+} with an oxygen vacancy in the immediate environment. The EPR lines were identified as in the work of Hannon.[4]

For an arbitrary direction of the magnetic field, we observed three spectra corresponding to different orientations of the axes of the paramagnetic center relative to the constant magnetic field (Fig. 2). The field direction was chosen to be at 90° to the axis of centers 5 and 6 and 75° to the axis of the centers 1 and 2. It will be important to remember that the dependence of the EPR line intensities on the direction of the magnetic field is very slight.

The electric field was applied along the axis of the center 1. Figure 3 shows the EPR line intensities of centers 1 and 2 and 5 and 6 for the $|-5/2 \leftrightarrow -3/2|_\perp$ transition in the presence of the electric field (45 kV/cm). It is clear that the intensity of the lines due to centers whose axes are parallel to the field are increased whereas, for centers perpendicular to the field, the intensity is reduced by a factor of roughly two.

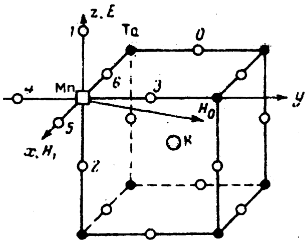

FIG. 2. Structure of potassium tantalate in the immediate environment of the paramagnetic center; E, H_0, and H_1 are, respectively, the electric field, constant magnetic field, and the magnetic component of the microwave field.

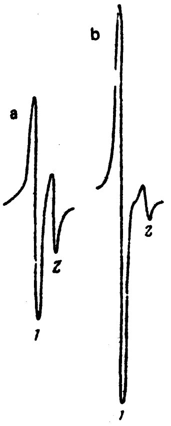

FIG. 3. EPR line intensities due to the first (2) and fifth (1) centers: a—$E=0$; b—$E=45$ kV/cm.

At the same time, the spectra of centers 1 and 2 are shifted in opposite directions (this is the electric-field effect). This enables us to observe changes in the line intensities due to centers that are parallel and antiparallel to the field. The splitting of the transition $|-3/2 \leftrightarrow -1/2|_\perp$ is sufficient for observation only in a narrow range of fields $E \approx$ kV/cm, whereas the line intensity due to centers parallel to the field is much lower than the intensity of centers antiparallel to the field but is not, nevertheless, zero.[2)]

In fields in excess of 45 kV/cm, the EPR line intensities of centers 5 and 6 are reduced still further until they vanish altogether for $E = 100$ kV/cm. This was found to be accompanied by an increase in the line intensities due to centers 2. The reduction in the areas under lines due to centers 5 and the corresponding increase in the areas under lines due to centers 2 can be regarded as evidence for the reorientation of the vacancies. The reorientation of vacancies 1 is, of course, also taken into account.

The variation in the EPR line intensities with external electric field relative to the intensities in zero field is determined by the ratio of the corresponding populations, as follows:

$$\frac{I(E)}{I(0)} = \frac{N_5+N_6}{{}^1/_3 N} = \frac{6\exp(-U_1/kT)}{1+4\exp(-U_1/kT)+\exp(-U_2/kT)}. \qquad (11)$$

where U_1 and U_2 are, respectively, the work done in displacing the vacancies from type 5 and type 1 positions to type 2 positions. The quantities U_1 and U_2 can be determined by using (11) for two different values of the external electric field. The parameter U_1/kTE is equal to 0.0154 esu^{-1} and U_2/kTE is equal to 0.019 esu^{-1}.

Figure 4 shows the relative intensity $I(0)/I(E)$ of EPR lines as a function of the external field. The solid curve was calculated from (11) and the points are experimental.

3. We shall now calculate U_1 and will show that it can, in fact, be expressed in terms of the local field at the sites of a perfect crystal. The work done in displacing a vacancy from one lattice site to another is given by

$$U_1 = Ze\int \mathbf{E}(\mathbf{r})\,d\mathbf{r}, \qquad (12)$$

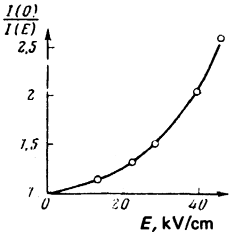

FIG. 4. EPR line intensity of Mn^{2+} in $KTAO_3$ (the $|-5/2 \leftrightarrow -3/2|_\perp$ transition) as a function of the external electric field.

where $\mathbf{E}(\mathbf{r})$ is the local field acting on the vacancy, s is the path over which the vacancy is displaced, and Ze is the charge on the vacancy. The local field can conveniently be written in the form

$$E_i(\mathbf{r}) = E[\delta_{iz} + \eta_{iz}(\mathbf{r}) + \lambda_{iz}(\mathbf{r})], \quad i = x,\ y,\ z. \tag{13}$$

The external field $E = E_z$ is parallel to the z axis and $\eta_{iz}(\mathbf{r})$ and $\lambda_{iz}(\mathbf{r})$ are tensorial coefficients representing the polarization of the perfect lattice and of the defect, respectively.

Equation (13) is easily interpreted if the crystal with the substituent impurity atom is "perfected" by adding the missing atom so that the resulting defect consists of the impurity "atom" and the lattice "antiatom" with charge and polarization of different sign.

Using (13), we can then rewrite (12) in the form

$$U_1 = ZeE \int_s [\delta_{iz} + \eta_{iz}(\mathbf{r}) + \lambda_{iz}(\mathbf{r})] d\mathbf{r}_i. \tag{14}$$

Following Grachev,[5] it can be shown that

$$\int_s \eta(\mathbf{r}) d\mathbf{r} = 0. \tag{15}$$

The point-dipole model was used to calculate the approximate contribution of λ_{iz} to (14). For this model,

$$E\lambda_{iz} = -\nabla_i \frac{p_z z}{r^3}.$$

p is the dipole moment of the defect, $p_z = (\alpha_1 - \alpha_2)E(0)$, where α_1 and α_2 are the polarizabilities of the impurity and of the replaced ion, respectively, and $E(0)$ is the local field at the site at which the replacement was made. The work done in displacing the vacancy from site 5 to site 2 is

$$U_1 = ZeEa\left[\frac{\alpha_1 - \alpha_2}{a^3}\frac{E(0)}{E} - 1\right], \tag{16}$$

where a is the distance between neighboring sites.

The work done in displacing the vacancy from 5 to 1 is also given by (16), but with the opposite sign.

The experimental value of U_1 given above suggests that the first term in brackets in (16) is greater than the second. Consequently, for $(\alpha_1 - \alpha_2)E(0) > 0$, we have the preferential orientation of vacancies antiparallel to the field. For $(\alpha_1 - \alpha_2)E(0) < 0$, the vacancies are parallel to the field.

It is convenient to compare the local field with the Lorentz field. In a cubic crystal, it is equal to $\frac{1}{3}(\epsilon - 1)E$. Suppose that $E(0) = \frac{1}{3}\gamma(\epsilon - 1)E$, so that (16) can be rewritten in the form

$$U_1 = ZeEa\left[\frac{\alpha_1 - \alpha_2}{a^3}\gamma\frac{\varepsilon - 1}{3} - 1\right]. \tag{17}$$

If, as before, we use the dipole approximation, we have $U_2 = 2U_1$. The inclusion of higher multipoles yields $U_2 \approx 1.3U_1$, which is in agreement with the experimental value of the ratio U_1/U_2.

To a good approximation, the field $E(0)$ can be regarded as the local field at a perfect-crystal site. Its distortion by the vacancy field can be estimated as follows:

$$\Delta E' = E'(0) - E(0) \approx \alpha_v E(a)/a_3,$$

where $E'(0)$ is the local field in the presence of the vacancy, (a) is the local field in the vacancy, and α_0 is the polarization of the vacancy. It is clear that $E'(0) \approx E(0)$ even if $E(0)$ is of the same order as $E(a)$. For $KTaO_3$, the field $E(a)$ is smaller by rougly two orders of magnitude than $E(0)$. This follows from our calculations performed by the Slater method.[6]

The expression given by (17) and the experimental values of U_1 and U_2 enable us to determine γ, i.e., $E(0)$. The polarization difference between the impurity and the replaced atom must, of course, be known. These polarizabilities can be obtained from experimental data on the dependence of the permittivity ϵ on impurity concentration and the relationship between ϵ in the crystal containing the impurity and the polarization difference $\alpha_1 - \alpha_2$. This quantity can easily be calculated by the Slater method.[6] The measurements were carried out on $KTaO_3$ with 0.5 at.% Mn at 77°K. In this case, $\epsilon = 900$ and the value of ϵ for pure $KTaO_3$ was assumed to be 829, in accordance with the data reported by Wemple.[7] The difference $\alpha_1 - \alpha_2$ turns out to be 0.0375×10^{-24} cm^3. Since $(\alpha_1 - \alpha_2)E(0) > 0$, the vacancy is oriented antiparallel to the field. The value obtained for $\alpha_1 - \alpha_2$ then enables us to find γ from (17). In fact, the value of γ turns out to be roughly six. When (17) was used in the calculations, account was taken of the higher order multipoles. The final value is in good agreement with the results of theoretical calculations which we have carried out in accordance with the Slater scheme.[6] It is important to emphasize that the local field is greater by a factor of 1600 than the external field. The values of the polarizability used in the calculations were, of course, approximate and this restricted the accuracy with which the local fields were calculated.

It is thus clear that the local field must be much greater than the external field if the experimental data are to be satisfactorily described. This situation may obtain in crystals with large ϵ, for example, in ferroelectrics for which the second term in (17) is greater than the first. The situation is different in crystals with more usual values of ϵ, where the second term *is* negligible in comparison with unity. When this is so, the effect is determined by the external field. One can now understand the results published in the literature[8-10] for the alkali halide crystals, which correspond to the second case.

[1] M. Bonciani and U. M. Grassano, Phys. Status Solidi B 67, 371 (1975); G. Chiarotti, U. M. Grassano, G. Margaritondo, and R. Rossei, Nuovo Cimento B 64, 159 (1969); F. Rosemberg and F. Lüty, Solid State Commun. 7, 249 (1969).
[2] M. Bonciani, U. M. Grassano, and R. Rossei, Phys. Rev. B 8, 5855 (1973); F. Lüty, Surface Sci. 37, 120 (1973); U. M. Grassano, G. Margaritondo, and R. Rossei, Phys. Rev. 132, 3319 (1970); L. D. Boga and D. B. Fitchen, Phys. Rev. B 1, 4122 (1970); G. Chiarotti, U. M. Grassano, and R. Rossei, Phys. Rev. Lett. 17, 1043 (1966); C. R. Rhyner and J. R. Cameron, Phys. Rev. 169, 710 (1969); L. A. De Werd, Phys. Status Solidi 52, 207 (1972).
[3] M. F. Deĭgen, M. D. Glinchuk, I. N. Geifman, A. A. Sytikov, and B. K. Krulikovskiĭ, Izv. Akad. Nauk SSSR Ser. Fiz. 39, 919 (1975).
[4] D. M. Hannon, Phys. Rev. B 3, 2153 (1971).
[5] V. G. Grachev, M. F. Deĭgen, and S. I. Pekar, Fiz. Tverd. Tela (Leningrad) 15, 2155 (1973) [Sov. Phys. Solid State 15, 1431 (1974)].
[6] J. C. Slater, Phys. Rev. 78, 748 (1950).
[7] S. H. Wemple, Phys. Rev. 137, A1575 (1965).
[8] Yu. A. Sherstkov, V. A. Vazhenin, and K. M. Zolotareva, Fiz. Tverd. Tela (Leningrad) 17, 2757 (1975) [Sov. Phys. Solid State 17, 1830 (1975)].
[9] V. M. Maevskiĭ, E. N. Kalabukhova, B. K. Krulikovskiĭ, and Kh. K. Maksimovich, Fiz. Tverd. Tela (Leningrad) 18, 2136 (1976) [Sov. Phys. Solid State 18, 1247 (1976)].
[10] W. Dreybrodt and G. Pfister, Phys. Status Solidi 34, 69 (1969).

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Role of the various types of transitions in three-photon absorption in InAs

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The frequency dependences of the three-photon absorption (TPA) probability and the linear-circular dichroism (LCD) associated with TPA in InAs are investigated theoretically and experimentally. The role of the various types of transitions involved in TPA is analyzed. The obtained experimental data are in satisfactory agreement with the results of a calculation performed in the two-band model with allowance for both allowed-allowed-allowed and allowed-forbidden-forbidden transitions.

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1. The observation of three-photon absorption (TPA) of CO_2-laser radiation in indium arsenide is reported in Ref. 1. During the study of the polarization characteristics of the TPA probability, there was observed—in accord with the predictions made in Refs. 2 and 3—a large linear-circular dichroism (LCD), $\Lambda = W_l^{(3)}/W_c^{(3)}$, near the TPA edge ($W_{l,c}^{(3)}$ are the TPA probabilities for linear and circular polarizations, respectively). The theoretical results of Refs. 2 and 3 were obtained in the lowest approximation in the parameter $\eta = (3\hbar\omega - E_g)/\hbar\omega \ll 1$ (E_g is the forbidden-band width) with allowance for only the allowed-allowed-allowed (*A-A-A*) transitions. In the indicated approximation Λ is frequency independent. Experiment[1] has, however, shown that a small increase in the crystal temperature (299–307 K) leads to a sharp decrease in the magnitude of Λ_{exp}. It is natural to assume that such a character of the dependence $\Lambda(T)$ is connected with the temperature variation of $E_g(T)$ and the strong dependence of the LCD near the TPA edge on the excess energy $(3\hbar\omega - E_g)$. In view of this, it was of interest to calculate the quantities $W_l^{(3)}$ and $W_c^{(3)}$ in the case of an arbitrary value of η, experimentally investigate the frequency dependences of the TPA probability and the magnitude of the LCD in InAs, and, comparing the theoretical and experimental results, analyze the relative contributions of the various types of transitions involved in TPA.

2. In the two-band model three-photon transitions are forbidden in A_3B_5 crystals for circularly polarized light in the case when $3\hbar\omega \approx E_g$.[2] This is due to the impossibility of changing by more than two units a component of the angular momentum of an electron in an interband transition in the vicinity of the $k = 0$ point. If we do not restrict ourselves to the two-band model, and take into consideration transitions with intermediate states in other bands, then the indicated forbiddenness is, generally speaking, lifted. In the three-band model, in which, besides the conduction band c and three valence subbands v_i $(i = 1, 2, 3)$, the higher-lying conduction band $\bar{c}$ is taken into consideration for *A-A-A* transitions, i.e., in the lowest approximation in the parameter $\eta \ll 1$, the expression for the TPA probability has the following form:1) [3]

$$W^{(3)}(\mathbf{e},\omega) = W^{(1)}(3\omega)\frac{\sigma^3(\omega)}{\sigma(3\omega)}\sum_\alpha\left[|a_1(\mathbf{ee})e_\alpha + a_2 e_\alpha^3|^2 + {}^3/_4|a_3 e_\alpha(e_{\alpha+1}^2 - e_{\alpha+2}^2)|^2\right], \tag{1}$$

where

$$a_1 = \frac{1}{2\hbar\omega}\left[-\frac{P_{cv}^2}{E_g - \hbar\omega}\frac{\Delta + 4\hbar\omega}{2\hbar\omega(\Delta + 2\hbar\omega)} + \frac{P_{\bar{c}c}^2}{E_{\bar{c}c} - 2\hbar\omega}\right] - a_2 - \frac{1}{2}a_3,$$

$$a_2 = \frac{P_{\bar{c}v}^2}{E_{\bar{c}v} - \hbar\omega}\frac{\Delta + 4\hbar\omega}{2\hbar\omega(\Delta + 2\hbar\omega)},$$

$$a_3 = \frac{P_{\bar{c}v}^2}{E_{\bar{c}v} - \hbar\omega}\frac{\Delta}{3\hbar\omega(\Delta + 2\hbar\omega)},$$

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