Theory of resonant optical orientation of tunnel centers

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A kinetic equation is obtained for strongly and weakly coupled tunnel centers. In the case of weak coupling the equation is valid at any ratio of the tunnel splitting to the relaxation level width, and also in any resonant field (the field frequency is close to the frequency of the local oscillation or of the electron transition). The repopulation of the wells in the field is considered. If the time of the tunneling into the excited state is shorter than the time of tunneling to the ground state then the optical orientation takes place in relatively weak fields. The repopulation kinetics of centers with orientations [100] and [111] in cubic crystals is described in this case by the presented equations. The self-induced rotation of the plane of polarization of resonant radiation in cubic crystals on account of the optical orientation of the impurities is analyzed.

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1. INTRODUCTION

Impurity molecules and noncentral ions in cubic crystals usually have several equivalent equilibrium positions in the unit cells, and resonant tunneling can take place between these positions.[1,2] The tunneling leads to a splitting of the energy levels of the intrawell local or quasilocal oscillations, as well as of the electron levels. If the potential barriers between the equilibrium positions are large enough, then the splitting for several lowest excited states is small in comparison with the distance between the levels. The dynamics of the tunnel center is determined in this case by the relation between the characteristic relaxation time τ and the tunnel splitting $\Delta\epsilon$. In the case $\tau\Delta\epsilon \gg \hbar$ the below-the-barrier states are essentially of the tunnel type, and their wave functions are transformed in accord with the representations of the cubic group (see e.g.,[3]), while the role of the relaxation reduces to a small broadening of the levels. In the opposite case $\tau\Delta\epsilon \ll \hbar$, the relaxation destroys the coherence of the equal-energy states of the impurities in different wells, i.e., it makes the tunneling in fact nonresonant. As a result, the center becomes effectively localized, the reorientation process acquires a hopping character, and the time of the tunnel transition turns out to be of the order of $\hbar^2/[\tau(\Delta\epsilon)^2]$.

The peak of the impurity absorption of light near the frequency of the intrawell transition at $\tau\Delta\epsilon \gg \hbar$ has as a result of the tunnel splitting a fine structure, and at $\tau\Delta\epsilon \ll \hbar$ it is smooth and, generally speaking, asymmetrical. In the case of absorption by local or quasilocal oscillations, the fine structure and the asymmetry of the spectrum can be connected also with the internal anharmonicity of these oscillations.[4] If the nonequidistant character of the level due to the oscillations exceeds the level width, and the temperature is low enough $\exp(\hbar\omega_0/T) \gg 1$ ($\hbar\omega_0$ is the energy of the first excited level), then the interaction of the local oscillation with the resonant raidation ($\omega \approx \omega_0$) can be described within the framework of the two-level model with tunneling between the wells.

We consider hereafter the case when the ground state of the tunnel center is quasilocalized, $\tau_0\Delta\epsilon_0 \ll \hbar$. It is clear from general considerations that the overlap integrals of the intrawell wave functions should be larger for the excited level than for the ground level, i.e., $\Delta\epsilon_{exc} > \Delta\epsilon_0$. Consequently a relatively weak resonant radiation can lead to a substantially anisotropic distribution of the tunnel centers over the equivalent minima. Resonant optical orientation (ROO) can be easily understood in the case of two-well potential if the dipole absorption in the wells occurs at mutually perpendicular polarizations of the radiation (E_x and E_y). Let $E_y = 0$. Owing to the tunneling through the excited level, an impurity initially in the ground state in the well x can, after absorbing light and relaxing, turn out to be in the ground state in the well y. If the tunneling time in the ground state is larger than in the excited state, then pumping by the field E_x will make the population in the well y larger than in the well x. The intensity of the resonant radiation needed to produce a substantial population difference, is determined by the ratio of the tunneling times in the ground and excited states; this ratio can be very large at $\Delta\epsilon_{exc} \gg \Delta\epsilon_0$ (if $\tau_{exc}\Delta\epsilon_{exc} \gg \hbar$, then the excited center is situated in the different well with practically equal probability and the tunneling time of the transition is close to zero).

In the case of strongly coupled tunnel centers, owing to the increase of the overlap integral, the activation energy of the hops between the wells may turn out to be much smaller in the excited state than in the ground state, and the tunneling time is correspondingly exponentially smaller. Optical orientation was experimentally observed for several types of strongly coupled centers.[5,6] The ROO theory for strong coupling is given in the Appendix.

To investigate the ROO it is convenient to use the kinetic equation. It is derived in Sec. 2 for a many-well tunnel center with weakly coupled dipole transition. In Sec. 3 we develop the quasistationary approximation, derive the balance equation for the well populations at $\tau\Delta\epsilon \ll \hbar$, and analyze the weak-field ROO. Owing to the reorientation of the resonant radiation, a cubic crystal with tunnel centers becomes anisotropic. The resultant

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self-induced rotation of the polarization plane of the resonant radiation is investigated in Sec. 4 for various centers. It is shown that the number of independent component of the nonlinear polarizability increases in the resonance region.

2. MODEL OF TUNNEL CENTER WITH WEAKLY COUPLED DIPOLE TRANSITION. KINETIC EQUATION

We consider a two-level tunnel center in the case when the intrawell wave functions are strongly localized. The Hamiltonian of the system in a resonant external field can be written in the form

$$H=H_0+H_1+H_1, \quad H_0=\omega_0\sum_i a_{i1}^+a_{i1}+\sum_{i\neq j}\sum_{\alpha=0,1}V_{ij\alpha}a_{i\alpha}^+a_{j\alpha}, \quad \hbar=1; \tag{1}$$

$$H_1=-\sum_i f_i(t)a_{i1}^+a_{i0}+\text{h.c.} \tag{2}$$

Here H_0 is the Hamiltonian of the tunnel center proper (the energy is reckoned from the ground level in the well); $a_{i\alpha}^+$ and $a_{i\alpha}$ are the operators of creation and annihilation of a center in the well i in the ground ($\alpha=0$) or excited ($\alpha=1$) states, satisfying the usual relations:

$$[a_{i\alpha}, a_{j\beta}^+]=\delta_{ij}\delta_{\alpha\beta}.$$

The parameters V determine the resonant tunneling, $|V|\ll\omega_0$. In the Hamiltonian (2) of the interaction with the field we took into account only the resonant intrawell terms ($f_n\sim e^{i\omega t}, \omega\approx\omega_0$) under the assumption $|f_n|\ll\omega_0; f_n(t)=(\mathbf{d}_n\mathbf{E}(t)$, $\mathbf{d}_n$ is the dipole moment of the intrawell transition, and $\mathbf{E}$ is the field intensity. It will be assumed in the future for concreteness that the direction of the dipole moment $\mathbf{d}_n$ coincides with the symmetry axis of the intrawell potential and, consequently, the excited level is nondegenerate within the limits of one well.

The Hamiltonian H_1 describes the oscillations of the continuous spectrum in a crystal with a defect and their interaction with the tunnel center:

$$H_1=H_{ph}+H_{int}^{(1)}+H_{int}^{(2)}; \quad H_{ph}=\sum_q\omega_q b_q^+b_q; \tag{3}$$

b_q and b_q^+ are the operators of annihilation and production of the oscillations of the continuous spectrum q; ω_q is its frequency. The term

$$H_{int}^{(1)}=\sum_{iq}V_{iq}(b_q+b_q^+)\sum_\alpha a_{i\alpha}^+a_{i\alpha} \tag{4}$$

determines the change of the equilibrium positions of the oscillations of the continuous spectrum upon reorientation of the impurity. This interaction is quite substantial for tunnel centers[7,8] and will henceforth not be regarded as weak.

The intrawell dipole transition is assumed to be weakly coupled with the phonons, so that the width Γ_0 of the light absorption line and the corresponding shift are small, $\Gamma_0\ll\omega_0, \omega_m$ (where ω_m is the maximal frequency of the continuum oscillations that are significant for the relaxation). The weak-interaction Hamiltonian takes the form

$$H_{int}^{(2)}=\sum_{i\alpha qq'}V_{i\alpha qq'}a_{i\alpha}^+a_{i\alpha}b_q^+b_{q'}+\sum_i[a_{i1}^+a_{i0}U_i\{b_q; b_q^+\}+\text{h.c.}]. \tag{5}$$

The first term in (5) corresponds to the change of the phonon frequencies both following the reorientation and following the transition of the impurity to the excited level, while the second term is nonadiabatic and, generally speaking, anharmonic. In second-order perturbation theory it describes the decay of the excited state with emission of one or several phonons and the level shift. No account is taken in (4) of the dependence of V_{iq} on α, which in the case of a weakly coupled dipole transition leads to a small level shift. In addition, assuming a weak overlap of the wave functions of the different wells, we have left out of H_{int} the terms proportional to $a_{i\alpha}^+a_{j\beta}(1-\delta_{ij})$.

In second order in $H_{int}^{(2)}$, the half-width of the light-absorption peak at $e^{\omega_0/T}\gg 1$ in the absence of tunneling is (cf.[9]):

$$\Gamma_0=\Gamma_m+\Gamma, \quad \Gamma_m\equiv\gamma_{ii}^{01}=\pi\sum_{qq'}|V_{i0qq'}-V_{i1qq'}|^2\bar{n}_q(\bar{n}_q+1)\delta(\omega_0-\omega_{q'}),$$
$$\Gamma=\frac{1}{2}\int_{-\infty}^{\infty}dt\exp(i\omega_0 t)\langle U_i(t)U_i^+(0)\rangle_{ph}, \quad \bar{n}_q=[\exp(\omega_q/T)-1]^{-1}, \tag{6}$$

Γ_m and Γ are the parameters of the modulation and decay broadenings, $\langle\ldots\rangle_{ph}$ denotes averaging over the phonons with Hamiltonian H_{ph}. The broadening of Γ_m is due to the relaxation of the phase difference of the wave functions of the ground and excited levels due to the quasielastic scattering of the phonons by the impurity.

When tunnel transitions between wells are considered, the analog of modulation broadening is obviously

$$\gamma_{ij}^{\alpha\alpha}=\pi\sum_{qq'}|V_{i\alpha qq'}-V_{j\alpha qq'}|^2\bar{n}_q(\bar{n}_q+1)\delta(\omega_q-\omega_{q'}). \tag{7}$$

The dynamics of the tunnel center depends on the ratio of the time of the interwell transition $\tau_{ij\alpha}$ to $\gamma_{ij}^{\alpha\alpha}$, and the value of $\tau_{ij\alpha}$ depends in turn on the interaction $H_{int}^{(1)}$. If this interaction is very strong,

$$\sum_q|V_{iq}-V_{jq}|^2/\omega_q^2\gg 1,$$

the problem of the tunnel center is close to the problem of the small-radius polaron, see the Appendix. In this section we consider the case of not too strong a coupling[1)] and low temperatures, so that

$$|\mathcal{V}_{ij\alpha}|>\Gamma_{pol}, \quad \mathcal{V}_{ij\alpha}=V_{ij\alpha}\exp\Big[-\sum_q|V_{iq}-V_{jq}|^2(2\bar{n}_q+1)/2\omega_q^2\Big], \tag{8}$$

where Γ_{pol} is the damping due to $H_{int}^{(1)}$; it is proportional at least to $(V/\omega_m)^2$ and is analogous to the damping of the small-radius polaron.[10]

To analyze the ROO and the nonlinear polarizability of tunnel centers it is convenient to eliminate $H_{int}^{(1)}$ with the aid of the standard canonical transformation

$$S=\exp\Big[-\sum_{iaq} V_{iq}a_{ia}^{+}a_{ia}(b_q-b_q^{+})/\omega_q\Big].$$

The operator of the intrawell dipole transition remains unchanged in this case, $Sa_{i1}^{+}a_{i0}S^{+}=a_{i1}^{+}a_{i0}$; the transformation of $H_{int}^{(2)}$ gives rise to terms that reduce in second order in $H_{int}^{(2)}$ to a renormalization of ω_0 and G, and in the approximation (8) V_{ija} is replaced by $\tilde{V}_{ija}$. In second order in $H_{int}^{(2)}$ and in first order in H_f and $\tilde{V}_{ija}$, the kinetic equation for the density matrix

$$\tilde{\rho}(t)=\exp\Big(i\omega_0 t\sum_n a_{n1}^{+}a_{n1}\Big)\,\mathrm{Sp}_{ph}[S\rho(t)S^{+}]\exp\Big(-i\omega_0 t\sum_n a_{n1}^{+}a_{n1}\Big)$$

can be derived in the region of long times $t\gg\omega_0^{-1},\omega_m^{-1}$ by the method of the integral operator equation,[11] as was done in [12-14] for other systems with several resonant dipole transitions:

$$\frac{\partial\tilde{\rho}}{\partial t}=-\Gamma\sum_n(a_{n1}^{+}a_{n1}\tilde{\rho}+\tilde{\rho}a_{n1}^{+}a_{n1})+2\sum_{mn}\Gamma_{mn}a_{m0}^{+}a_{m1}\tilde{\rho}a_{n1}^{+}a_{n0}$$
$$-\sum_{mn\alpha\beta}(1-\delta_{mn}\delta_{\alpha\beta})\gamma_{mn}^{\alpha\beta}a_{m\alpha}^{+}a_{m\alpha}\tilde{\rho}a_{n\beta}^{+}a_{n\beta}-i\sum_{m\neq n}\sum_{\alpha}\tilde{V}_{mn\alpha}[a_{m\alpha}^{+}a_{n\alpha},\tilde{\rho}]$$
$$+i\sum_m[f_m(t)e^{i\omega_0 t}a_{m1}^{+}a_{m0}+\text{h.c.},\tilde{\rho}],\quad \mathrm{Sp}\,\tilde{\rho}=1. \tag{9}$$

The expressions for the parameters of the modulation broadening $\gamma_{mn}^{\alpha\beta}$ in (9) are obtained from (6) and (7) with the aid of the obvious replacement of the indices (inasmuch as the modulation broadening is due to phase relaxation, it contributes to the damping of only the off-diagonal matrix elements of $\tilde{\rho}$);

$$\Gamma_{mn}=\frac{1}{2}\int_{-\infty}^{\infty}e^{i\omega_0 t}dt\Big\langle U_n\Big\{b_q(t)-\frac{V_{nq}}{\omega_q},b_q^{+}(t)-\frac{V_{nq}}{\omega_q}\Big\}$$
$$\times U_m^{+}\Big\{b_q(0)-\frac{V_{mq}}{\omega_q},b_q^{+}(0)-\frac{V_{mq}}{\omega_q}\Big\}\Big\rangle_{ph},\quad \Gamma=\Gamma_{mm}\geqslant|\Gamma_{mn}|.$$

The renormalization of ω_0 on account of the phonons is assumed carried out in (9), and the corrections $\sim|\tilde{V}|/\omega_0,\Gamma/\omega_0,|f|/\omega_0$ and γ/ω_0 are assumed discarded. In addition, it is assumed that $T>\Gamma,\gamma,|\tilde{V}|$ (but $\exp(\omega_0/T)\gg1$ and Eq. (8) is satisfied).

Equation (9) is valid at arbitrary $|\tilde{V}|/\Gamma_0$ and $|f|/\Gamma_0$, and therefore allows us to consider both the fine structure of the spectrum of absorption by a tunnel center (at $|\tilde{V}|\gg\Gamma_0$), as well as a smooth spectrum, and also the weak-field and strong-field (absorption saturation) cases. It can be shown, in particular, that when account is taken of the quadratic corrections in $|\tilde{V}|/\Gamma_0<1$ the absorption spectrum becomes non-Lorentzian, but remains symmetrical.

3. QUASISTATIONARY APPROXIMATION. OPTICAL ORIENTATION IN WEAK FIELDS

The operator equation (9) for an N-well center is equivalent to a system of $N(2N+1)$ linear equations for the matrix elements $\rho_{\alpha\beta}^{mn}=\mathrm{Sp}(a_{n\beta}^{+}a_{m\alpha}\tilde{\rho})$, which in the general case can be solved with the aid of a computer. However, if the tunnel splitting is small compared with the reciprocal relaxation time (this is apparently frequently the case for under-the-barrier excited levels[15]) and the impurity is localized in wells, Eq. (9) can be solved analytically. The criterion for the localization is slowness of the change of the well population

$$\rho_n=\sum_\alpha\rho_{\alpha\alpha}^{nn} \tag{10}$$

during the time $|\tilde{V}_{ija}|^{-1}$ of the resonant tunnel transition. Since, as seen from (9),

$$\frac{d\rho_n}{dt}=-2\,\mathrm{Im}\sum_{m(\neq n)}\sum_\alpha\tilde{V}_{mn\alpha}\rho_{\alpha\alpha}^{nm}, \tag{11}$$

localization calls for smallness of the off-diagonal elements $|\rho_{\alpha\alpha}^{nm}|\ll\rho_n$.

The structure of Eq. (9) is such that the elements of $\tilde{\rho}$ diagonal and off-diagonal in the upper indices (intra- and interwell) relax and interact with the field independently of one another, and the connection between them is due only to tunneling. If the field amplitude varies slowly:

$$f_n(t)=\tilde{f}_n(t)\exp(-i\omega t),\quad \frac{d\ln\tilde{f}_n}{dt}\ll\Gamma,\gamma, \tag{12}$$

then, in the case of small tunnel splitting

$$|\Gamma_{ik0}|\ll\Gamma_{ik}^{00}\equiv\gamma_{ik}^{00},\quad |\Gamma_{ik1}|\ll\Gamma_{ik}^{11}\equiv2\Gamma+\gamma_{ik}^{11},\quad \sum_\alpha|\Gamma_{ik\alpha}|\ll\Gamma_{ik}^{01}\equiv\Gamma+\gamma_{ik}^{01}, \tag{13}$$

the system relaxes in the following manner: after times $\sim\max(\Gamma^{-1},\gamma^{-1})$ equilibrium is established within each well [two-level system in a monochromatic field with summary level population $\rho_n(t)\approx$ const as a result (11) and (13)]; the interwell elements of $\tilde{\rho}$ likewise reach their quasistationary values in this case. The latter can be determined in first order in $\tilde{V}/\gamma$ from the system of equations

$$[\Gamma_{mn}^{\alpha\beta}+i\Omega(\delta_{\alpha0}-\delta_{\beta0})]\tilde{\rho}_{\alpha\beta}^{mn}-2\Gamma_{mn}\delta_{\alpha0}\delta_{\beta0}\tilde{\rho}_{11}^{mn}-i[\tilde{f}_m\delta_{\alpha1}+\tilde{f}_m^{*}\delta_{\alpha0}]\tilde{\rho}_{1-\alpha\beta}^{mn}$$
$$+i[\tilde{f}_n^{*}\delta_{\beta1}+\tilde{f}_n\delta_{\beta0}]\tilde{\rho}_{\alpha1-\beta}^{mn}=-i[\tilde{\Gamma}_{mn\alpha}\tilde{\rho}_{\alpha\beta}^{nn}-\tilde{\Gamma}_{mn\beta}\tilde{\rho}_{\alpha\beta}^{mm}],\quad m\neq n,\quad t\gg\Gamma^{-1},\gamma^{-1}, \tag{14}$$

$\tilde{\rho}_{\alpha\beta}^{mn}=\rho_{\alpha\beta}^{mn}\exp[-i\Omega t(\delta_{\alpha0}-\delta_{\beta0})]$, $\Omega=\omega-\omega_0$, with $\tilde{f}$ and $\rho_{\alpha\beta}^{mn}$ dependent on the time as a parameter:

$$\rho_{00}^{nn}=\rho_n(t)-\rho_{11}^{nn},\quad \rho_{01}^{nn}=-i\tilde{f}_n^{*}(t)\rho_n(t)\frac{\Gamma(\Gamma_0-i\Omega)\exp(i\Omega t)}{\Gamma(\Gamma_0^2+\Omega^2)+2\Gamma_0|f_n|^2},$$
$$\rho_{11}^{nn}=\rho_n(t)\frac{\Gamma_0|f_n|^2}{\Gamma(\Gamma_0^2+\Omega^2)+2\Gamma_0|f_n|^2}. \tag{15}$$

For each (m,n) pair, (14) constitutes a system of four equations that can be readily solved. Substituting the solution in (11), we obtain

$$\frac{d\rho_n}{dt}=-\sum_m[C_{nm}(t)\rho_n(t)-C_{mn}(t)\rho_m(t)],\quad \sum_{n=1}^{N}\rho_n=1. \tag{16}$$

Equations (16) take the form of balance equations and describe the slow variation of the well populations. The explicit expression for C_{nm} is in the general case quite complicated and is not given here, but it is seen from (13) and (14) that $C_{nm}\sim|\tilde{V}|^2/\Gamma_{mn}^{\alpha\beta}\ll|\tilde{V}|\ll\Gamma,\gamma$, i.e., both the localization criterion and the quasistationarity condition are satisfied (with large margin). The need for sufficiently rapid relaxation for reorientation of the hopping type was noted already in[8], but it was not made clear there which are the quantities that relax,

what causes the relaxation, and how the reorientation process is to be described.

If the multiwell potential has an inversion center, then it is possible to obtain from (16) an analogous system of $N/2$ equations for $\tilde{\rho}_n = \rho_n + \rho_{\bar{n}}$ ($\bar{n}$ is the number of the well that is the reflection of the well n), with $\tilde{C}_{nm} = C_{nm} + C_{n\bar{m}}$, $C_{nm} = C_{\bar{n}\bar{m}}$. The last equality is due to the fact that the inversion changes only the sign of the field in the coefficients C_{nm}, but this change is equivalent, by virtue of (12), to a time shift $\Delta t = \pi/\omega \ll |\tilde{V}|^{-1}, \Gamma^{-1}$, which cannot be reflected in C_{nm}. Consequently, C_{nm} contains only even powers of the field.

Greatest interest attaches to consideration of the ROO in the case when the tunnel splitting for the excited level is much larger than for the ground level, and orientation is possible even in fields that are weak compared with those that lead to saturation.

If the inequalities

$$|V_{nm1}| \gg |V_{nm0}|, \quad \gamma_{nm}^{00}/\Gamma \gg |V_{nm0}/V_{nm1}|, \tag{17}$$

are satisfied in addition to (13), then in non-saturating fields we obtain from (14)–(16)

$$C_{nm} = A_{nm} + B_{nm}|f_n|^2, \quad A_{nm} = 2V_{nm0}^2/\Gamma_{nm}^{00},$$
$$B_{nm} = 2V_{nm1}^2 \frac{\Gamma_0\Gamma_{mn}^{01}(\Gamma+\Gamma_{mn}^{01}) + \Omega^2(\Gamma_0-\Gamma)}{\Gamma_{mn}^{11}\Gamma(\Gamma_0^2+\Omega^2)[(\Gamma_{mn}^{01})^2+\Omega^2]}, \quad \Gamma_0|f|^2/(\Gamma_0^2+\Omega^2) \ll \Gamma, \Gamma_{nm}^{00}. \tag{18}$$

In the case of a weakly coupled dipole transition the resonant field leads to a mixing of the wave functions of the ground and excited intrawell states (as resonant tunneling in the case of equal-energy level). Consequently the parameter C_{nm} does not reduce to the sum of the probabilities of the hops in the ground and excited states, but if (17) is satisfied C_{nm} contains only the squares of the moduli of the fields that act in the wells. The relation between the field-free rate of reorientation A_{nm} and $B_{nm}|f_n|^2$ in (18) is arbitrary.

To illustrate the repopulation of the wells, we consider a two-well system at constant $|f|^2$. The solution of (16) with allowance for (18) takes in this case the form

$$\rho_1(t) = \left[\rho_1(0) - \frac{1+|f_2|^2B_{12}/A_{12}}{2+|f|^2B_{12}/A_{12}}\right]\exp[-(2A_{12}+B_{12}|f|^2)t] + \frac{1+|f_2|^2B_{12}/A_{12}}{2+|f|^2B_{12}/A_{12}},$$
$$\rho_2(t) = 1-\rho_1(t), \quad \frac{\rho_2(\infty)}{\rho_1(\infty)} = \frac{1+|f_1|^2B_{12}/A_{12}}{1+|f_2|^2B_{12}/A_{12}}, \quad |f|^2 = |f_1|^2+|f_2|^2. \tag{19}$$

According to (19), the change of the well populations from the initial value to the final stationary value is monotonic. The maximum repopulation takes place if the field excites transitions in only one of the wells,

$$\Gamma\Gamma_0 \gg |f_1|^2 \gg A_{12}/B_{12} \gg |f_2|^2.$$

In this case

$$\rho_2(\infty)/\rho_1(\infty) = |f_1|^2B_{12}/A_{12} \gg 1.$$

i.e., in a relatively weak field, when (17) is satisfied, practically total depletion of one of the two equivalent wells is possible.

The system (16) and (18) can be easily solved also for a larger number of wells in the case of tunnel centers in cubic crystals, if the symmetry group of the multiwell potential is O_h, and the minima are oriented in the directions [100] and [110]. For the total population of the wells that are equivalent with respect to inversion, $\tilde{\rho}_n$, Eq. (16) takes the form

$$\frac{d\tilde{\rho}_n}{dt} = -\frac{N}{2}(A+B|f_n|^2)\tilde{\rho}_n + \sum_{m=1}^{N/2}(A+B|f_m|^2)\tilde{\rho}_m, \quad \sum_{n=1}^{N/2}\tilde{\rho}_n = 1. \tag{20}$$

The parameter A for the centers [100] and [111] is respectively equal to $2A_{[100][010]}$ and $A_{[111][1\bar{1}1]} + A_{[111][\bar{1}\bar{1}\bar{1}]}$ (the expressions for B are analogous), while $N = 6$ and 8. Equation (20) can be represented in a reduced form by going over to the dimensionless time At and to the field $(B/A)^{1/2}f$. Therefore the kinetics of the optical orientation for the different tunnel centers with wells in the direction [100] or [111] is the same (for centers with minima along [110] there are no such minima; an equation such as (20) contains the dimensionless parameters of the concrete center).

The stationary solution of (20) in a monochromatic field is of the form

$$\tilde{\rho}_n(\infty) = \prod_{\substack{m=1\\(m\neq n)}}^{N/2}\left(1+\frac{B}{A}|f_m|^2\right)\Big[\sum_{s=1}^{N/2}\prod_{\substack{r=1\\(r\neq s)}}^{N/2}\left(1+\frac{B}{A}|f_r|^2\right)\Big]^{-1}. \tag{21}$$

If the field is relatively strong for all wells, $B|f_n|^2/A \gg 1$, then the population distribution (this takes place also in the case of two wells) does not depend on the field,

$$\tilde{\rho}_n = \Big[\sum_m |f_n/f_m|^2\Big]^{-1}.$$

If the field is strong for only wall well and for its reflection (this is possible only for [100] centers), $|f_1|^2 \gg A/B \gg |f_{2,3}|^2$, the well is depleted. If, on the contrary, the field is weak for only one well, then practically all the centers go over into this well. In contrast to the case of two wells, the transition from the field-free distribution $\tilde{\rho}_n(0)$ to (21) takes place, generally speaking, in nonmonotonic fashion, since the characteristic equation corresponding to (20) has several roots.

It is of interest also to consider the optical orientation in the case when the tunnel splitting of the excited level exceeds the width of the absorption line $|\tilde{V}_{mn1}| \gg \Gamma_0, \Gamma_{mn}^{11} \gg |\tilde{V}_{mn0}|$. If a transition to a nondegenerate (for simplicity) tunnel level takes place under the influence of resonant light, then immediately after the absorption the center can be with equal probability in any of the cells. Starting from (9), we can show that in weak fields the kinetics of the ROO is described by formulas (16) and (18), where

$$A_{nm} = 2|\tilde{V}_{nm0}|^2/\gamma_{nm}^{00}, \quad B_{nm} = 2\Gamma_0[N(\Gamma_0^2+\Omega^2)]^{-1}, \quad \Gamma_0 = \Gamma + N^{-1}\sum\gamma_{nm}^{01},$$
$$N^{-1}|f|^2/\Gamma_0 \ll \Gamma, \ \gamma_{mn}^{00}, \ |\tilde{V}_{mn0}| \ll \gamma_{mn}^{00} \tag{18a}$$

(it is assumed that the rate of establishment of equilibrium within the tunnel multiplet is low, $\gamma_{mn}^{11} \ll \Gamma$). Formulas (16), (18), and (18a), with allowance for some renormalization of the parameters, are valid also in the case when the excited level is above the barrier (the tunnel splitting $\tilde{V}_{mn1}$ does not enter in (18a). Thus, the field dependence of the R00 in the region of weak fields is the same for large and small tunnel splitting of the excited level.

4. SELF-INDUCED ROTATION OF THE RADIATION POLARIZATION IN RESONANT OPTICAL ORIENTATION

Optical orientation of tunnel centers by linearly polarized resonant radiation makes the impurity cubic center anisotropic. This anisotropy can be revealed, for example by the absorption of the additional weak light, but it influences substantially also the propagation of the orienting radiation itself, by rotating its polarization plane. Self-induced resonant rotation of the polarization was considered in[14] in the saturation of the absorption by the impurity dipole transition $A_{1g} - T_{1u}$, and also in saturation of the absorption by two-level tunnel centers. The tunnel-center model in[14] is analogous to that of the present paper, but it was assumed there that the radiation pulse duration is short in comparison with the tunneling time, and the latter was not taken into account at all. In the case of long pulses, the principal anisotropy mechanism, as shown below, can be precisely the reorientation of the centers.

The nonlinear polarizability of an impurity crystal at low tunnel splitting (13) and at a slowly varying field amplitude can be represented by using (15), accurate to terms $\sim \tilde{V}/\Gamma^{\alpha\beta}$, in the form

$$\chi_{\varkappa\varkappa'}(\omega, E) = \frac{\varepsilon-1}{4\pi}\delta_{\varkappa\varkappa'} + ic_0 \sum_n d_{n\varkappa} d_{n\varkappa'} \Gamma(\Gamma_0 + i\Omega)\rho_n [\Gamma(\Gamma_0^2+\Omega^2) + 2\Gamma_0 |\mathbf{d}_n \mathbf{E}|^2]^{-1}, \tag{22}$$

where ϵ is the dielectric constant of the host crystal; c_0 is the impurity concentration and is assumed to be quite small; $d_{n\varkappa}$ is the projection of the dipole moment in the well n on the direction of $\varkappa$ (at $\rho_n = 1/N$ and $\Omega = 0$ Eq. (22) coincides with formula (13) of[14]). Maxwell's equations and the material equations (22) and (16) describe in self-consistent fashion the propagation of the resonant radiation in the crystal.

The anisotropy of the polarizability $\chi_{\varkappa\varkappa}$ is due to the field dependence of ρ_n and of the denominator in (22). If (17) is satisfied, then strong nonlinear effects can be observed in relatively weak fields, $|\mathbf{dE}|^2 \ll \Gamma\Gamma_0$, when the field dependence of the denominator is negligible. The nonlinearity of the polarizability sets in this case after a time $\sim (A + B|f|^2)^{-1}$. The stationary polarizability in a monochromatic field at $\omega = \omega_0$ in the case of tunnel centers with [100] orientation can be obtained from formulas (22) and (21):

$$\operatorname{Im}\chi_{\varkappa\varkappa'}(\omega_0, E) = \chi_{\varkappa\varkappa}''\delta_{\varkappa\varkappa'} = \frac{c_0 d^2}{\Gamma_0} \frac{1 + a^2(|E_{\varkappa_1}|^2 + |E_{\varkappa_2}|^2) + a^4|E_{\varkappa_1}E_{\varkappa_2}|^2}{3 + 2a^2|\mathbf{E}|^2 + a^4(|E_x E_y|^2 + |E_x E_z|^2 + |E_y E_z|^2)}, \tag{23}$$

$$\varkappa_{1,2} \neq \varkappa, \quad a^2 = d^2 B/A.$$

Expression (23) is valid for arbitrary values of $a^2|\mathbf{E}|^2$, but formally (23) can be regarded as an expansion of the susceptibility up to fourth order in the field, with the nonlinear and linear components of the polarizability tensor dependent on the field via the invariants of the group O_h [the denominator in (23)].[2)] It is of interest to note that $\chi_{\varkappa\varkappa}''$ depends only on the squares of the moduli of the field components. This means that, for example, the expression for the absorped power averaged over the period contains the invariant

$$\sum_{\varkappa\neq\varkappa'} |E_\varkappa|^2 |E_{\varkappa'}|^2,$$

but not the invariant

$$\sum_{\varkappa\neq\varkappa'} E_\varkappa^2 (E_{\varkappa'})^2.$$

In the phenomenological approach with expansion of the polarization in the real field, the absorbed power would contain both invariants, and the coefficient of the first, without allowance for the time dispersion, would by twice as large as the coefficient of the second. The increase in the number of the independent components of the polarizability tensor is due in this case to the resonant character of the absorption.

It is seen from (23) that the optical orientation leads to bleaching of the impurity crystal. If, e.g., $E_{y,z} = 0$, then

$$\chi_{\varkappa\varkappa}'' = c_0 d^2/[3\Gamma_0(1 + 2a^2|\mathbf{E}|^2/3)].$$

The field dependence of the absorption coincides in this case with $\chi''(E)$ dependence upon saturation of the absorption by a nondegenerate two-level system, but the character of the bleaching is different: the tunnel centers leave those wells in which they are excited by the field, and cease to absorb the latter. If several field components differ from zero, then the components that are more weakly absorbed will be those whose amplitude is larger. This will change the propagation direction and the plane of polarization of the light.

In the case when the radiation propagates along the z axis (this propagation direction is stable) and is linearly polarized, Maxwell's equations with (23) taken into account reduce to the system

$$\frac{d|E_\varkappa|^2}{dz} = -\frac{3|E_\varkappa|^2}{l_0} \frac{1 + a^2|E_{\varkappa_1}|^2}{3 + 2a^2|\mathbf{E}|^2 + a^4|E_x E_y|^2}, \qquad l^{-1} = \frac{4\pi\omega_0}{c\varepsilon^{1/2}} \frac{c_0 d^2}{\Gamma_0},$$
$$a^2 = d^2 B_{[100]}/A_{[100]}, \quad \varkappa, \varkappa_1 = x, y, \quad \varkappa_1 \neq \varkappa, \tag{24}$$

which has one simple integral

$$E_y(z)/E_x(z) = [E_y(0)/E_x(0)] \exp\{-a^2[|E_y(z)|^2 - |E_x(z)|^2 - (|E_y(0)|^2 - |E_x(0)|^2)]/2\}. \tag{24a}$$

It is seen from (24a) that if $E_y(0)/E_x(0) > 1$ then the ratio $E_y(z)/E_x(z)$ increases with crystal thickness, but if $E_y(0)/E_x(0) < 1$, this ratio decreases. Thus, the plane of polarization rotates towards the nearest of the type [100] axes. This is understandable, since the larger field component depletes its wells more strongly and is absorbed less. The limiting direction of the polarization depends exponentially on the field intensity:

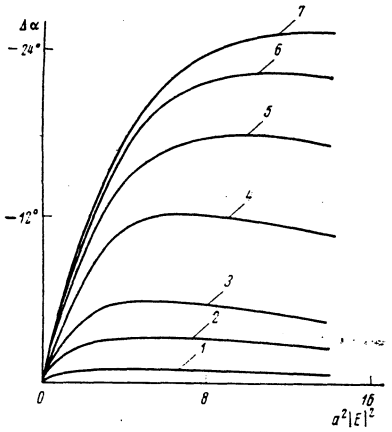

FIG. 1. Dependence of the angle of the self-induced rotation of the polarization plane on the intensity of the incident resonant radiation for tunnel centers with orientation [100]. The radiation propagates along the [001] axis, and the initial angle between the vector E and the [100] axis is 30°. Curves 1–7 correspond to crystal thicknesses $l/l_0 = 0.2, 0.6, 1, 2, 3, 4, 5$ (l_0 is the weak-field absorption line).

$$E_y(\infty)/E_x(\infty) = [E_y(0)/E_x(0)]\exp\{-a^2(|E_y(0)|^2 - |E_x(0)|^2)/2\}.$$

The field dependence of the angle of rotation of the polarization has a peculiar character for crystals of finite thickness. This dependence is shown in Fig. 1 [the multiple reflection was not taken into account in the course of the solution of (24)]. The maxima on the $\Delta\alpha(E)$ curves is due to the bleaching of the crystal in the strong fields. It can be clearly observed if the direction of propagation of the radiation coincides with the orientation of one of the wells (in this case, with the [001] well). Owing to the preferred filling of this well, a decrease takes place in both the absorption coefficients of the individual field components and in their difference, which in fact causes the rotation of the polarization,

$$\frac{d}{dz}\ln\frac{E_x}{E_y} \sim a^{-2}|\mathbf{E}|^{-2}$$

at $a^2|\mathbf{E}|^2 \gg 1$. The position of the maximum $\Delta\alpha(E)$ shifts towards larger fields with increasing crystal thickness l; for thin plates ($l/l_0 \ll 1$) the value of $\Delta\alpha$ is maximal at $a^2|E_x|^2 = 3^{1/2}|E_x/E_y|$.

In the case of tunnel centers with [111] orientation and resonant radiation propagating along z ([001]) the wells $[(-1)^\alpha(-1)^\beta \pm 1]$ are equivalent at $\alpha = \beta$ or $\alpha \neq \beta$ ($\alpha, \beta = 0.1$), and the system reduces to a two-well one with effective-well orientations [110] and [1$\bar{1}$0]. Solving at $\Omega = 0$ the system of equations similar to (24)

$$l_0\frac{d|E_\varkappa|^2}{dz} = -\frac{(1+\tilde{a}^2|E_{\varkappa_1}|^2)|E_\varkappa|^2}{1+\tilde{a}^2|\mathbf{E}|^2/2}, \quad \tilde{a}^2 = \frac{d^2}{3}\frac{B_{[111]}}{A_{[111]}}$$
$$E_\varkappa = \frac{1}{\sqrt{2}}[E_x - (-1)^\varkappa E_y], \quad \varkappa, \varkappa_1 = 1, 2, \quad \varkappa \neq \varkappa_1. \tag{25}$$

we obtain

$$E_1(z)E_2(z) = E_1(0)E_2(0)e^{-z/l_0},$$
$$v = v_0\exp\{\tilde{a}^2|E_1(0)|^2[v_0 - 1 + (1-v)(v_0/v)^{1/2}e^{-z/l_0}]\}. \tag{25a}$$
$$r \equiv r(z) = |E_2(z)/E_1(z)|^2, \quad v_0 = v(0).$$

The function v determines the rotation of the plane of polarization of the radiation. It is seen from (25) and (25a) that as it propagates in the crystal the vector $\mathbf{E}$ rotates towards the nearest of the directions [110] or [1$\bar{1}$0]:

$$v(\infty) = v_0\exp(-2\tilde{a}^2E_x(0)E_y^*(0)).$$

The angle of rotation of the polarization depends monotonically both on the crystal thickness and on the intensity of the incident radiation.

In contrast to the centers [100] and [111], for centers with orientation [110] not only the velocity but also the direction of the rotation of the polarization plane, propagating along the [001] axis, depend on the parameters of the concrete center and on the field intensity. Therefore in a sufficiently thick crystal, owing to the decrease of the radiation intensity with thickness, the direction of rotation can be different in different sections.

5. CONCLUSION

From (16), (18), (18a) and the Appendix it is seen that the kinetics of the weak-field ROO for both strong and weak coupling is described by the balance equations. This is evidence that the duration of the reorientation act is short, $\tau_{mn\alpha} \ll C_{mn}^{-1}$. The last inequality is the criterion for the localization of the tunnel center in the wells. In the nonequilibrium case it is equivalent to the inequality $|\rho_{\alpha\beta}^{mn}| \ll 1$ at $m \neq n$. In the considered models we have $\tau_{mn0} \sim |V_{mn0}|^{-1}$. We note that in the case of strong coupling we have $|\tilde{V}_{mn0}|^{-1} \gg C_{nm}^{-1}$ and for weak coupling $|\tilde{V}_{mn0}|^{-1} \sim \tau_{mn0} \ll C_{mn}^{-1}$.

The most stringent requirement that limits the applicability of the results at low temperatures in the case of weak coupling (for strong coupling we have considered only the region of relatively high temperatures) can be the condition $|\tilde{V}_{mn0}| \ll \Gamma_{mn}^{00}$ in (13), since $\Gamma_{mn}^{00} \sim (T/\omega_m)^7$ at $T \ll \omega_m$. When (13) is violated, the localization can be due to random fields if the mean squared level splitting is $\overline{\Delta^2} \gg |\tilde{V}_0|^2$. Since the parameter V_0 is usually small,[16] weak random fields are sufficient for the localization. At $|\tilde{V}_{mn\alpha}| \ll |\Gamma_{mn}^{\alpha\alpha} + i(\Delta_{m\alpha} - \Delta_{n\alpha})|$ ($\Delta_{m\alpha}$ is the shift of the level α in the well m) the quasistationary approximation is applicable, and in (15) it is necessary to replace Ω by $\Omega_m = \Omega + \Delta_{m0} - \Delta_{m1}$, while in (14) $\Gamma_{mn}^{\alpha\beta}$ must be replaced by $\Gamma_{mn}^{\alpha\beta} + i(\Delta_{m\alpha} - \Delta_{n\beta})$ (the relaxation due to transitions between wells with participation of a phonon of frequency $\omega_k = \Delta_{m\alpha} - \Delta_{n\alpha}$ is not taken into account). The second of the criteria (17) is replaced by an inequality of the type

$$|V_{nm0}/V_{nm1}| \ll \Gamma_{mn}^{11}[(\gamma_{mn}^{00})^2 + \overline{\Delta^2}]/[\gamma_{nm}^{00}((\Gamma_{mn}^{11})^2 + \overline{\Delta^2})],$$

which can be much less stringent at $T \ll \omega_m$. Although the general character of the optical orientation does not change, the averaging over the random field in equations (14)–(16) at arbitrary $|\overline{\Delta}|/\Gamma_{mn}^{\alpha\beta}$ is a relatively easy matter only if the number of wells is small.

In the experiments, Turner *et al.*[5] and Blume *et al.*[6] measured the quantum yield η of the ROO—the probability of reorientation of the center as a result of photon

absorption. It is seen from (16) and (18) (see also the Appendix) that this quantity is determined by the ratio $B_{nm}/\varphi(\tilde{\Omega})$ at strong coupling and by $B_{nm}(\Gamma_0^2+\Omega^2)/\Gamma_0$ at weak coupling. For strong coupling in the case $\Gamma \gg \Gamma_{mn\alpha}$ we have

$$B_{nm}/\varphi(\tilde{\Omega}) = 2\Gamma_{mn1}/\Gamma \sim \exp(-E_{mn1}/T).$$

Such an activation dependence of η was indeed observed in[5,6] for strongly coupled M and F_A centers. At $\Gamma_{mn1} \gg \Gamma \gg \Gamma_{mn0}$ the quantum yield turned out to be independent of temperature, $B_{mn}/\varphi(\tilde{\Omega}) = 2/N$ [this expression is obtained also in the case of weak coupling for a tunneling excited level, see (18a)]. If (13) and (17) are satisfied then, according to (18), η decreases with increasing both on account of the decrease of $\tilde{V}_{nm}$ and because of the increase of the relaxation parameters. We note that the stationary nonequilibrium distribution of the tunnel centers is determined by the parameter B_{nm}/A_{nm}, which does not have the same temperature dependences as η.

When holograms were recorded by the ROO method in[6], no account was taken of the self-induced rotation of the polarization plane of the resonant radiation which is quite large and limits the possibility of recording images in thick crystals. At $\Omega \neq 0$ both the ratio of the field components and the dephasing change with changing distance, and this also decreases the possibilities of using R00 for three-dimensional holography.

The authors thank V. S. Vikhnin for a discussion of the results.

APPENDIX

Consider the optical orientation of a weakly coupled tunnel center. The Hamiltonian $H_{int}^{(1)}$ in (3) is in the case of strong coupling

$$H_{int}^{(1)} = \sum_{i\alpha q} V_{i\alpha q}(b_q + b_q^+) a_{i\alpha}^+ a_{i\alpha}. \tag{A.1}$$

We shall assume that for all $\{i\alpha\} \neq \{j\beta\}$

$$\omega_q \gg \sum_q \frac{(V_{i\alpha q} - V_{j\beta q})^2}{\omega_q} \gg \omega_m, |f|, |V_{ij\alpha}|. \tag{A.2}$$

As already noted, in the absence of a field the problem of the strongly coupled tunnel center is close to the small-polaron problem. It is known (see[10,17]) that at sufficiently high temperatures the main mechanism of the polaron mobility are the hops between the localized states. It is easy to show that the probability of hopping of a tunnel center from a well i into a well j in the state α at sufficiently small $V_{ij\alpha}$ is

$$2\Gamma_{ij\alpha} = |V_{ij\alpha}|^2 \sqrt{2\pi} \left[\sum_q (V_{i\alpha q} - V_{j\alpha q})^2 / \mathrm{sh}(\omega_q/2T) \right]^{-1/2} \exp(-E_{ij\alpha}/T),$$

$$E_{ij\alpha} = T \sum_q \omega_q^{-2} (V_{i\alpha q} - V_{j\alpha q})^2 \,\mathrm{th}(\omega_q/4T), \quad \omega_m, T \gg \Gamma_{ij\alpha} \gg |V_{ij\alpha}|. \tag{A.3}$$

For strongly coupled tunnel centers in alkali-halide crystals there exists apparently a sufficiently wide temperature interval where, besides (A.3) the relation $T \ll P$ is satisfied. By virtue of the latter inequality, the Stokes shift $2P$ greatly exceeds the line width δ of the optical absorption, where (cf.[18])

$$P = \sum_q (V_{i1q} - V_{i0q})^2 \omega_q^{-1}, \quad \delta^2 = 2\sum_q (V_{i1q} - V_{i0q})^2 (2\bar{n}_q + 1), \quad P \gg \delta. \tag{A.4}$$

Therefore the resonantly absorbed radiation

$$f_m(t) = \tilde{f}_m(t) e^{-i\omega t}, \quad \Omega = \omega - \omega_0 - P, \quad |\Omega| \sim \delta, \quad d\tilde{f}_m/dt \ll \tilde{f}_m \delta,$$

causes in practice no induced transitions from the excited to the ground state. If (A.3) and (A.4) are satisfied, the kinetic equation for the density matrix $\tilde{\rho}(t)$ takes at $t \gg \omega_m^{-1}$ the form[3)]

$$\frac{\partial \tilde{\rho}}{\partial t} = -\sum_{m \neq n} \sum_{\alpha} \Gamma_{mn\alpha} (a_{m\alpha}^+ a_{m\alpha} \tilde{\rho} + \tilde{\rho} a_{m\alpha}^+ a_{m\alpha} - 2a_{m\alpha}^+ a_{n\alpha} \tilde{\rho} a_{n\alpha}^+ a_{m\alpha})$$
$$-\varphi(\tilde{\Omega}) \sum_m |f_m(t)|^2 (a_{m0}^+ a_{m0} \tilde{\rho} + \tilde{\rho} a_{m0}^+ a_{m0} - 2a_{m1}^+ a_{m0} \tilde{\rho} a_{m0}^+ a_{m1})$$
$$-\Gamma \sum_m (a_{m1}^+ a_{m1} \tilde{\rho} + \tilde{\rho} a_{m1}^+ a_{m1} - 2a_{m0}^+ a_{m1} \tilde{\rho} a_{m1}^+ a_{m0}),$$
$$\mathrm{Sp}\tilde{\rho} = 1, \quad \varphi(\tilde{\Omega}) = \pi^{1/2} \delta^{-1} \exp[-(\Omega/\delta)^2]. \tag{A.5}$$

Here 2Γ is the probability of a decay transition from the excited state to the ground state (e.g., with emission of a photon), and it is assumed that $\Gamma \ll \omega_m, d\Gamma/d\omega_0 \ll \Gamma/P$.

Equation (A.5) differs substantially from the kinetic equation (9); in the case of strong coupling the diagonal and off-diagonal elements are not entangled and the off-diagonal elements decay within a time $\sim \Gamma_{mn\alpha}^{-1}$ Γ^{-1} (if the transformation S is not performed, then the interwell elements turn out to be $\sim \Gamma_{ij\alpha}/|V_{ij\alpha}| \ll 1$); in the case of weak coupling the resonance field plays with respect to the transitions between levels the same role as resonant tunneling does with respect to interwell transitions, and in the case of strong coupling the field produces transitions only to an excited level; we note also that in the case of strong coupling there is no interference between the decay processes in the different wells, and from among the terms proportional to Γ_{mn} in (9), there remain in (A.5) only the diagonal ones.

Since $E_{ij1} < E_{ij0}$ as a result of the increase of the overlap integrals in the excited state, it is of interest to consider the case $\Gamma_{ij1} \gg \Gamma_{ij0}$, when the optical orientation of the tunnel centers proceeds in relatively weak fields $\varphi(\tilde{\Omega})|f|^2 \ll \Gamma$ (if $1 \gg \varphi(\tilde{\Omega})|f|^2/\Gamma > \exp(-P/T)$, then the impurity crystal can amplify the light at the frequency $\omega_0 - P$,[19] but Eq. (A.5) is violated only in the case when the probability of the induced transitions from the excited state to the ground state becomes comparable with the probability of the spontaneous ones).

At $\Gamma \gg \Gamma_{mn0}, \Gamma_{mn1}$ the intrawell equilibrium is established more rapidly than the interwell equilibrium, the quasistationary approximation is valid, and we can obtain from (A.5) the equation (16) for the well populations. If $\Gamma_{mn1} \gg \Gamma_{mn0}$ and $\Gamma \gg \varphi(\tilde{\Omega})|f|^2$, then the coefficients C_{nm} in this equation reduce to the sum of the probabilities of the reorientations in the ground and ex-

cited states and are described by formula (18), where $A_{nm}=2\Gamma_{nm0}$, $B_{nm}=2\Gamma_{mn1}\varphi(\tilde{\Omega})/\Gamma$. The ROO kinetics, the stationary distribution, as well as the self-induced rotation of the plane of polarization of the radiation are described under these conditions by formulas (19)–(25). In (22) the factor

$$\Gamma(\Gamma_0+i\Omega)\left[\Gamma(\Gamma_0^2+\Omega^2)+2\Gamma_0|\mathbf{d}_{nn}\mathbf{E}|^2\right]^{-1}$$

must be replaced by the coefficient

$$\varphi(\tilde{\Omega})+i\chi(\tilde{\Omega}),\quad \chi(\tilde{\Omega})=\frac{2}{\delta}\int_0^{\Omega\delta}\exp\left(x^2-\frac{\tilde{\Omega}^2}{\delta^2}\right)dx.$$

Since the time of energy relaxation at $\omega_0\gg\omega_m$ is quite large ($\Gamma\sim10^7$–10^8 sec^{-1} for the F center in KCl according to the estimates of[19]), the following relation may hold in a definite temperature interval

$$\Gamma_{mn1}\gg\Gamma\gg\Gamma_{mn0}.$$

In this case the fastest process is the equalization of the populations of the excited states in different wells, and the problem is similar to the case considered in Sec. 3, that of large tunnel splitting of the excited level. The kinetic equation for slowly varying well populations at $\Gamma\gg\varphi(\tilde{\Omega})|f|^2$ takes the form (16), (18a), where $A_{mn}=2\Gamma_{mn0}$ and $B_{mn}=2\varphi(\tilde{\Omega})/N$. We note that it does not contain the parameters of the tunneling in the excited state or the energy relaxation parameter Γ.

[1)]For many tunnel centers the change of the static dipole moment upon reorientation is $p\ll ea_0$ (a_0 is the lattice constant), and at any rate the polarization interaction is less than for a small polaron.

[2)]The authors are grateful to M. I. D'yakonov and È. I. Rashba for pointing out this property of the susceptibility. It can be shown that for centers with orientations [111] and [110] the structure of the numerator and the denominator in the expressions for the polarizability is analogous to the structure (23), but they are polymonials of sixth and tenth degree in the field, respectively.

[3)]The canonical transformation that relates $\tilde{\rho}$ with the total density matrix is analogous to that used in Sec. 2, but now

$$S=\exp\left[-\sum V_{i\alpha q}a_{i\alpha}^{+}+a_{i\alpha}(b_q-b_q^{*})/\omega_q\right].$$

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