

# Self-induced rotation of the polarization in the resonance absorption region of doped cubic crystals

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A study is made of the self-induced rotation of the radiation polarization under saturation conditions of the impurity absorption in a system with two-level impurities exhibiting three degenerate excited levels. The rotation is large when the width of the absorption line is much greater than the reciprocal of the impurity energy relaxation time. When the ratio of the line width to the reciprocal of the relaxation time is very large, the rotation of the polarization is described by a simple expression which holds in a wide range of radiation intensities. The magnitude and direction of the rotation are sensitive to the relative magnitudes of the relaxation parameters. When the broadening of the absorption line is mainly due to a random field, the nature of the random forces can be deduced from the behavior of the polarization rotation.

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Several mechanisms of the broadening of the absorption line of light by weakly bound impurities in crystals have been discussed. For two-level impurities, the decay and modulation mechanisms are the most important.<sup>1</sup> The decay broadening is due to the energy relaxation (transitions between levels accompanied by the emission of photons, phonons, or magnons), and the modulation broadening is due to the relaxation of the phase caused by the quasielastic scattering of phonons (magnons by impurities). The number of the relevant parameters describing the relaxation depends on the multiplicity of the level degeneracy. For nondegenerate levels, the relaxation is characterized by two parameters, i.e.,  $\Gamma$  and  $\Gamma_m$  (the decay and modulation broadening). These two parameters can be determined from the absorption line profile and from the saturation of the absorption which occurs when the intensity of the resonance radiation is increased.<sup>2</sup> For degenerate levels, additional methods are required to determine all the relevant relaxation parameters.

It was shown in Ref. 3 that the relaxation parameters for degenerate two-level impurities in cubic crystals can be determined from the measurements of the self-induced rotation of the polarization plane of strong resonance radiation. It follows from the estimates of Ref. 3 that the angle of rotation is sensitive to the relative magnitude of the relaxation parameters and can be quite large ( $\sim 10^\circ$ ) in fields  $\sim 10^4$ - $10^5$  V/cm, in which the nonlinearity of the crystal under study does not manifest itself. Impurities with a single threshold degenerate excited level are considered in Ref. 3. The wave functions of the excited level transform under a vector representation of the cubic group ( $|x\rangle$ ,  $|y\rangle$ ,  $|z\rangle$ ). For saturated absorption, the rotation of the plane of polarization is governed by the specific impurity relaxation mechanism. The rotation can be explained qualitatively as follows. Let us assume that the radiation travels in the  $z$  direction,  $E_z = 0$ , and also that the relaxation of degenerate state is independent ("decoupled" in phase). The occupancy of the states  $|x\rangle$  and  $|y\rangle$  is governed, respectively, by the components of the field  $E_x$  and  $E_y$ . Since the weaker component is absorbed more strongly, the electric field vector of a linearly polarized wave traveling in a crystal rotates toward

the nearest of the axes  $x$ ,  $y$ . However, when phonons are quasielastically scattered from impurities, not only the phases of degenerate states are decoupled but also an excitation transfer between states takes place. If this transfer is the fastest effect, the stronger field component is absorbed more strongly (it can even transform into the weaker component). In that case, the field vector rotates toward the nearest bisector of the angle between the axes  $x$ ,  $y$ .

The rotation of the polarization of linearly polarized radiation is studied in Ref. 3 under exact resonance conditions when the field frequency satisfies  $\omega = \omega_0$ , where  $\hbar\omega_0$  is the separation between the impurity levels. It is of interest to study the rotation of the polarization for  $|\omega - \omega_0| \sim \Gamma_0$ , where  $\Gamma_0$ , representing the half-width of the weak field absorption line, satisfied  $\Gamma_0 \ll \omega_0$  and the degree of polarization is arbitrary. It is also of interest to clarify the dependence of the angle of rotation of the degree of bleaching. We shall also consider inhomogeneous broadening of the absorption line and we shall show that such a broadening can lead to new effects, in particular, to the rotation of the polarization ellipse, which is typical of isotropic systems.<sup>4</sup>

## 1. ROTATION OF THE POLARIZATION IN THE ABSENCE OF INHOMOGENEOUS BROADENING

In a monochromatic field  $E(t) = Ee^{-i\omega t} + E^*e^{i\omega t}$  at low temperatures  $T \ll \hbar\omega_0$  the quantum transport equation under steady-state conditions reduces (see Ref. 3) to the following system of linear equations for the matrix elements of the impurity density matrix  $\rho$ :

$$\left. \begin{aligned} (\Gamma_0 + i\Omega) \rho_{0x} - i \sum_{x_1} \Delta_{xx_1} \rho_{0x_1} &= id \sum_{x_1} E_{x_1}^* \rho_{x_1x} - id E_x^* \rho_{00}, \\ 2\delta_{xx_1} (\Gamma + 3\Gamma_{m2}) \rho_{xx_1} + 2(1 - \delta_{xx_1}) \Gamma_\varphi \rho_{xx_1} + 2\Gamma_{m2} (\rho_{xx_1} - \rho_{xx_1}^*) \\ - i \sum_{x_2} (\Delta_{xx_1} \rho_{xx_2} - \Delta_{xx_2} \rho_{xx_1}) &= id E_x^* \rho_{0x_1} - id E_{x_1}^* \rho_{x_10} + 2\Gamma_{m2} \delta_{xx_1} (1 - \rho_{00}), \\ \rho_{00} + \sum_x \rho_{xx} &= 1; \quad \Gamma_\varphi = \Gamma + \Gamma_{m2} + \Gamma_{m1} - \Gamma_{m3}; \quad \Omega = \omega - \omega_0; \quad \hbar = 1. \end{aligned} \right\} \quad (1)$$

Here,  $d$  is the dipole moment of the transition between

the ground and excited states, the index  $\kappa = 1, 2, 3$  ( $x, y, z$ ) labels the excited states, 0 is the index of the ground state; the intrinsic half-width of the absorption line is given by  $\Gamma_0 = \Gamma + \Gamma_{m1} + 2\Gamma_{m2}$ . The parameters governing the decay  $\Gamma$  and modulation  $\Gamma_{m1,2,3}$  broadening are quoted explicitly in Ref. 3 ( $\Gamma_{m1} \approx |\Gamma_{m3}|$ ). The terms in Eq. (1) that are proportional to  $\Delta_{\kappa\kappa_1}$  are due to a random field. It is assumed that the random field is weak ( $|\Delta_{\kappa\kappa_1}| \ll \omega_0$ ) and the related modification of the interaction of impurities with phonons and the direct interaction between the ground and excited states are neglected.

The contribution of impurities to the crystal polarization is given by

$$P_x = n d_{x0}, \quad (2)$$

where  $n$  is the impurity concentration. It can be easily shown that, in the absence of inhomogeneous and modulation broadening ( $\Gamma \gg \Gamma_m, |\Delta_{\kappa\kappa_1}|$ ), we obtain

$$P_x = i n \left( 1 + \frac{i\Omega}{\Gamma_0} \right) \sigma(E) E_x, \quad \sigma(E) = d^2 \left[ \frac{\Gamma_0^2 + \Omega^2}{\Gamma_0} + \beta \frac{d^2 E^2}{\Gamma} \right]^{-1},$$

$$E^2 = \sum_x |E_x|^2. \quad (3)$$

where  $\Gamma_0 \approx \Gamma$  and  $\beta = 2$ . It follows from Eq. (3) that saturation of the absorption occurs in strong fields and the characteristic absorption cross section  $\sigma(E)$  decreases. However, the nonlinearity of  $\sigma(E)$  defined by Eq. (3) does not lead to any anisotropy of a crystal with impurities.

The limit  $\Gamma_m \gg \Gamma$  is more interesting. This inequality is satisfied for weakly bound "high-frequency" impurities at moderately low temperatures (see, for example, Ref. 5). For  $\Gamma_m \gg \Gamma \gg |\Delta_{\kappa\kappa_1}|$ , Eq. (1) can be solved in fields  $E^2 \ll \Gamma_m^2/d^2$  (the ratio  $d^2 E^2/\Gamma \Gamma_m$  is arbitrary). For  $\Gamma_{m2} \gg \Gamma$ , the density matrix in the zeroth approximation in  $d^2 E^2/\Gamma_m^2$  is  $\rho_{\kappa\kappa_1} \approx \delta_{\kappa\kappa_1} \times (1 - \rho_{00})/3$ , i.e., the occupation numbers of all excited states  $\kappa$  are identical and their phases are independent. In this approximation, Eq. (3) again holds for  $\beta = 4/3$ . Therefore, the bleaching of a crystal is governed by the parameter  $d^2 E^2/\Gamma \Gamma_0$ , irrespective of the ratio  $\Gamma/\Gamma_m$ . An anisotropy occurs only when the terms proportional to  $d^2 E^2/\Gamma_m^2$  are considered. In the first order in  $d^2 E^2/\Gamma_m^2$ , lengthy calculations yield

$$P_x = P_x^{(1)} + P_x^{(3)}; \quad P_x^{(1)} = A E_x;$$

$$P_x^{(3)} = \left[ B_1 E_x E^2 + B_2 E_x |E_x|^2 + B_3 E_x^* \sum_{\kappa \neq x} E_{\kappa}^2 \right];$$

$$A = i \sigma(E) \left( 1 + i \frac{\Omega}{\Gamma_0} \right) \left[ n - \frac{4}{3\Gamma} \sum_{\kappa_1} \text{Im}(E_{\kappa_1}^* P_{\kappa_1}^{(3)}) \right];$$

$$B_1 = \frac{C}{9b\Gamma_{m2}} (9\Gamma_{m2}^2 + 7\Gamma_{\varphi}\Gamma_{m2} - \Gamma_{\varphi}^2);$$

$$B_2 = -\frac{C(3\Gamma_{m2}^2 + \Gamma_{\varphi}\Gamma_{m2} - \Gamma_{\varphi}^2)}{3b\Gamma_{m2}}; \quad B_3 = \frac{\Gamma_{m2}}{b} C;$$

$$C = -i \frac{nd^2 \sigma(E)}{\Gamma_0 - i\Omega}; \quad b = \Gamma_{\varphi}(\Gamma_{\varphi} + 2\Gamma_{m2}); \quad \Gamma_{m2} \gg \Gamma. \quad (4)$$

Formally, Eq. (4) represents an expansion of the polarization of a cubic crystal up to third order in the field. However, it should be noted that the components of the susceptibility tensor of rank four (the parameters  $B$ ) depend on  $E^2$  and the linear susceptibility (the parameter  $A$ ) depends on  $E^2$  and on the fourth-order invariants in the field. The quantity  $P_{\kappa\kappa}^{(1)}$  and the first term in  $P_{\kappa\kappa}^{(3)}$  determine the isotropic part of the susceptibility; the second and third terms in  $P_{\kappa\kappa}^{(3)}$  govern its anisotropic

part; clearly,  $P_{\kappa\kappa}^{(3)}/P_{\kappa\kappa}^{(1)} \sim d^2 E^2/\Gamma_m^2 \ll 1$ . Substituting Eq. (4) in Maxwell's equations, we can determine the rotation of the radiation polarization in a crystal for arbitrary  $d^2 E^2/\Gamma \Gamma_0$  and  $\Omega/\Gamma$ . It is convenient to express the angle of rotation in terms of the increment of the complex difference between the phases of the field components  $\Psi_{\kappa\kappa_1}$ :

$$\Psi_{\kappa\kappa_1} = \Psi'_{\kappa\kappa_1} + i\Psi''_{\kappa\kappa_1} = \Psi_{\kappa} - \Psi_{\kappa_1}; \quad \Psi_{\kappa} = \ln(E_{\kappa}/E_{x0}), \quad (5)$$

where  $E_{\kappa 0}$  is the component  $\kappa$  (complex) of the strength of the incident radiation field. For  $d^2 E^2/\Gamma_m^2 \ll 1$ , the anisotropy is weak and, therefore,  $|\Psi_{\kappa\kappa_1}| \ll 1$ . Since the anisotropy is weak, we can evaluate  $\Psi_{\kappa\kappa_1}$  explicitly but the actual expressions for the field components (for  $\Psi_{\kappa}$ ) calculated in the first order in  $d^2 E^2/\Gamma_m^2$  cannot be obtained for arbitrary  $d^2 E^2/\Gamma \Gamma_m$ .

For example, for waves traveling in the direction  $z$  and  $E_z = 0$ , a small change in the angle  $\alpha$  between the major axis of the polarization ellipse and the  $x$  axis is given by

$$\delta\alpha = \frac{2 - \varepsilon^2}{2\varepsilon^2} \left[ \Psi'_{yx} \sin 2\alpha + \frac{2\sqrt{1 - \varepsilon^2}}{(2 - \varepsilon^2)} \Psi''_{yx} \cos 2\alpha \right], \quad |\delta\alpha| \ll 1, \quad (6)$$

where  $\varepsilon$  is the eccentricity of the polarization ellipse of the incident radiation. For  $\varepsilon = 1$  (linear polarization), we obtain

$$\Psi_{yx}(z) = -a \cos 2\alpha [d^2 E^2(0) - d^2 E^2(z)]/[4\Gamma_{\varphi}(\Gamma_0 + i\Omega)],$$

$$\delta\alpha = -\frac{a\Gamma_0 \sin 4\alpha}{16(\Gamma_0^2 + \Omega^2)\Gamma_{\varphi}} [d^2 E^2(0) - d^2 E^2(z)],$$

$$a = \frac{2}{3} (\Gamma_{m1} - 2\Gamma_{m2} - \Gamma_{m3})/\Gamma_{m2}. \quad (7)$$

Here, the spatial dependence of the field intensity is governed by the following equation, which is typical of saturated absorption (neglecting the rotation of the polarization plane):

$$E^2(z) = E^2(0) \exp \left\{ -kz + \frac{4}{3\Gamma} \frac{\Gamma_0 d^2}{\Gamma_0^2 + \Omega^2} [E^2(0) - E^2(z)] \right\}, \quad (8)$$

where  $k$  is the absorption coefficient of a weak field, given by  $k \sim nd^2 \Gamma_0/(\Gamma_0^2 + \Omega^2)$ .

It follows from Eqs. (7) and (8) that the rotation of the polarization is monotonic across the crystal. The angle  $\delta\alpha$  decreases with the field strength and, even for  $d^2 E^2 \ll \Gamma_m^2$ , it can be of the order of  $1^\circ$ . For strong fields ( $d^2 E^2 > \Gamma_0^2$ ), the quantities  $\alpha$  and  $\varepsilon$  depend nonmonotonically on the thickness of the crystal and on the field provided  $\Omega \neq 0$ . This is due to the fact that the components  $E_x$  and  $E_y$  acquire a large phase difference. All the parameters characterizing the impurity center relaxation can be deduced from the aforementioned dependences. In fact, we can obtain  $\Gamma_0$  from the half-width of the absorption line,  $\Gamma$  from the saturation of the absorption defined by Eq. (8), and  $a$  from the rotation of the polarization of linearly polarized radiation defined by Eq. (7). An additional relationship between the relaxation parameters can be obtained from the dependence of  $\delta\alpha$  on  $\varepsilon$ , which can be easily calculated analytically from Eqs.

(4)-(6). In particular, it follows from Eq. (7) that the rotation of the polarization is very sensitive to the relative magnitude of the modulation broadening parameters (it is sensitive to the sign of  $\alpha$ ). In fact, for  $\alpha > 0$ , the vector  $E$  rotates toward the nearest axis [100] and, for  $\alpha < 0$ , it rotates to the axis [110]. The type of representation under which the phonons, whose interaction results in the modulation broadening, transform determines the relationship between the parameters  $\Gamma_{m1,2,3}$ .

## 2. EFFECT OF INHOMOGENEOUS BROADENING ON THE POLARIZATION ROTATION

We shall study the effect of inhomogeneous broadening on the nonlinear impurity absorption in the most important special case, when the modulation broadening is large, i.e.,  $\Gamma_{m2} \gg \Gamma_0$ . We shall assume that the fields involved are weak,  $d^2 E^2 < \Gamma(\Delta + \Gamma_0)$ , where  $\Delta$  is the inhomogeneous line broadening due to a shift of levels. The effect of a random field is to shift the levels and also remove the degeneracy. Therefore, the impurities have three excited levels  $|k\rangle$  ( $k=1, 2, 3$ ) with energies  $\hbar(\omega_0 + \Delta_k)$ ;  $|k\rangle = \sum_i \alpha_{ki} |i\rangle$ ,  $\sum_i \Delta_{xi} \alpha_{ki} \alpha_{ki} = \Delta_k \delta_{kk}$ , and  $|\Delta_k| \ll \omega_0$ . The unitary matrix  $\|\alpha\|$  is a rotation matrix from the orientations  $x, y, z$  to the orientations  $k=1, 2, 3$ ; without loss of generality, we can assume that the unitary matrix in question is real.

As already discussed, in the first approximation in  $d^2 E^2 / \Gamma(\Gamma_0 + \Delta)$  and in the zeroth approximation in  $d^2 E^2 \cdot (\Gamma_0^2 + \Delta^2)^{-1}$ , Eq. (1) yields  $\rho_{\chi\chi} = [(1 - \rho_{00})/3] \delta_{\chi\chi}$ . Combining this result with Eq. (2), we obtain

$$\left. \begin{aligned} P_x &= \sum_i \lambda_{xi} E_i + \sum_{i,j,k} \Lambda_{xixj} E_i E_j^* E_k; \quad \lambda_{xi} = i n d^2 \left\langle \sum_k \frac{\alpha_{ki} \alpha_{ki}}{\Gamma_0 - i \Omega_k} \right\rangle, \\ \Lambda_{xixj} &= -i n d^4 \frac{4 \Gamma_0}{3 \Gamma} \left\langle \sum_{k,k'} \frac{\alpha_{ki} \alpha_{xj} \alpha_{k'i} \alpha_{k'i}}{(\Gamma_0^2 + \Omega_k^2)(\Gamma_0^2 + \Omega_{k'}^2)} \right\rangle, \\ \Omega_k &= \omega - \omega_0 - \Delta_k, \quad \Delta^2 = \langle (\Delta_k - \langle \Delta_k \rangle)^2 \rangle; \quad d^2 E^2 \ll \Gamma_0^2. \end{aligned} \right\} \quad (9)$$

Here,  $\langle \dots \rangle$  indicates averaging over the random field. The averaging includes averaging over orientations that are defined by the functions  $|k\rangle$  and averaging over the magnitude of the shift  $\Delta_k$ ; in general, these two types of averaging need not be independent. It is assumed that the inhomogeneous broadening is due to the interaction of resonant absorbing impurity centers with other imperfections rather than due to their mutual interactions and, therefore, we obtain  $P_{\chi\chi} \propto n$ . For a uniform distribution of imperfections over the crystal, i.e., when the scale of microscopic inhomogeneity is small compared with the wavelength of light, the polarizability tensor of rank two is diagonal,  $\lambda_{\chi\chi} = \lambda \delta_{\chi\chi}$  and the nonlinear polarizability tensor has three independent components. For an arbitrary ratio  $\Delta/\Gamma_0$ , it is not possible to estimate the relative magnitudes of these components. For weak inhomogeneous broadening  $\Delta \ll \Gamma_0$ , we obtain  $\Delta \ll \Gamma_0$ ,  $\Lambda_{xxxx} = 2\Lambda_{xx'xx'} = 2\Lambda_{xx'xx}$ ,  $\Lambda_{xx'xx'} = 0$ , and in the approximation defined by Eq. (9), no self-induced anisotropy occurs.

For a strong inhomogeneous broadening, we obtain  $\Delta \gg \Gamma_0$ . It is natural to assume that the splitting of levels in a random field is of the same order as the shift and  $\langle (\Delta_k - \Delta_{k'})^2 \rangle \sim \Delta^2 \gg \Gamma_0^2$ ,  $k \neq k'$ . In this limit, the sum-

mation over  $k$  and  $k'$  of the quantity  $\Lambda_{xixj}$  in Eq. (9) can be limited to the term  $k = k'$  because all the other terms are proportional to higher powers of the small parameter  $\Gamma_0/\Delta$ . As a result, we obtain

$$\left. \begin{aligned} \Lambda_{xixj} &\approx -i n d^4 \frac{4 \Gamma_0}{3 \Gamma} \left\langle \sum_k \frac{\alpha_{ki} \alpha_{xj} \alpha_{ki} \alpha_{ki}}{(\Gamma_0^2 + \Omega_k^2)^2} \right\rangle, \\ \Lambda_{xxxx} &= \Lambda_{xx'xx'} = \Lambda_{xx'xx}; \quad \Delta^2 \sim \langle (\Delta_k - \Delta_{k'})^2 \rangle \gg \Gamma_0^2. \end{aligned} \right\} \quad (10)$$

In the limit  $\langle (\Delta_k - \Delta_{k'})^2 \rangle \gg \Gamma_0^2$ , only transitions to one of the three split impurity levels can be regarded as resonance transitions and the contribution of all the other states (and also the contributions of impurities which do not exhibit a suitable resonance level) to the nonlinear polarizability is small. Therefore, it follows from Eq. (10) that a self-induced anisotropy appears in the impurity absorption band even for  $d^2 E^2 \ll \Delta^2$ .

The type of anisotropy depends on the nature of the inhomogeneous broadening. When the broadening is due to imperfections that lie far from the absorbing center (for example, the broadening due to long-range dislocation fields), the angular distribution of the orientations  $|k\rangle$  is essentially uniform. Moreover, if the dependence of  $\Delta_k$  on the orientation  $|k\rangle$  can be neglected, the averaging over the orientations  $|k\rangle$  and over the magnitude of  $\Delta_k$  are independent and Eq. (10) yields

$$\Lambda_{xxxx} = 3\Lambda_{xx'xx'} = 3\Lambda_{xx'xx} = 3\Lambda_{xx'xx'}, \quad (x' \neq x). \quad (11)$$

Equations (9) and (11) describe the nonlinear polarizability of an isotropic medium.<sup>1)</sup> It is well known<sup>4</sup> that self-induced rotation of the polarization in such a medium can occur only for elliptically polarized radiation. In contrast to Ref. 4, the angle of rotation of the polarization plane in an absorbing medium reaches saturation as a function of the crystal thickness  $\delta\alpha \sim \frac{\text{Re } \Lambda}{\text{Im } \lambda} E^2 \sqrt{1 - \epsilon^2/(2 - \epsilon^2)}$ .

Moreover, the rotation is accompanied by a change in the ellipticity.

For a short-range interaction of the absorbing impurity center with imperfections, considerable splitting and shift of the levels occur when the impurities are located in the same or neighboring unit cells. An impurity complex (see, for example, Ref. 6) elongated in the direction of one of the crystallographic axes is formed and exhibits several equivalent orientations. The averaging over a random field reduces to the averaging over the equivalent orientations  $|k\rangle$  for fixed shifts  $\Delta_k$ . The self-induced anisotropy occurs provided  $|\Delta_k - \Delta_{k'}| \gg \Gamma_0$  ( $k \neq k'$ ), i.e., the absorption line exhibits a fine structure. In the zeroth approximation in  $\Gamma_0/|\Delta_k - \Delta_{k'}| \ll 1$ , nonlinear effects were studied in Ref. 3 for arbitrary  $d^2 E^2 / \Gamma_0$ . It was shown in Ref. 3 that the orientation of complexes can be determined from the polarization rotation.

## 3. CONCLUSIONS

We have demonstrated that study of the polarization rotation under saturation conditions of the resonance impurity absorption can be used to clarify the nature of the broadening and also to determine the impurity relaxation parameters. For homogeneous broadening, the po-

polarization rotation occurs in stronger fields [ $E^2 \sim \hbar^2(\delta\omega)^2/d^2$ ,  $\delta\omega$  is the linewidth] than the saturation of the absorption ( $E^2 \sim \hbar^2\Gamma\Gamma_0/d^2$ ). Therefore, in a wide range of fields, the saturation of the absorption is described by the dependence (8) neglecting the polarization rotation [the dependence (8) was observed in Ref. 7 for threefold degenerate anharmonic local vibrations in  $\text{CaF}_2$  doped with  $\text{H}^-$ ]. For inhomogeneous broadening, the saturation of the absorption and the polarization rotation are governed by the same parameter  $\Delta E^2/\lambda \sim d^2E^2/\hbar^2\Gamma\Gamma_0$ ,  $\delta\omega \gg \Gamma_0 \gg \Gamma$ . In this case, the anisotropy of the interaction resulting in inhomogeneous broadening can be determined from the measured dependence of the polarization rotation angle on the ellipticity of the incident radiation. The dependence of the self-induced anisotropy on the radiation frequency is also of interest since, by varying the frequency, we could select an effective average distance between the absorbing center and the imperfection resulting in the splitting of levels.

<sup>1</sup>)Equation (11) is approximately satisfied for transparent isotropic media in the nonresonance region. However, Eq. (11) can be also obtained in

the resonance absorption region in the special case when the absorption is due to impurity molecules with nondegenerate levels and all the orientations of the transition dipole moment (the orientations of the molecule) are equally probable.

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