

# Resonant magneto-optical spin transitions in zinc-blende and wurtzite semiconductors

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A theoretical study of the resonant magneto-optical spin-flip transitions due to electrons bound to shallow donors is made. We present a general formalism of the integrated absorption coefficient for semiconductors having an arbitrary crystal structure. The model is based on the parity-violating spin-orbit interaction responsible for the active electric-dipole transition. Simple analytic expressions of the integrated absorption coefficient are obtained for the Faraday and Voigt field configurations and for any crystals having zinc-blende and wurtzite structure, respectively. A comparison between theory and far-infrared magneto-optical transmission measurements in InSb and  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$  yields to the strength of the spin-orbit interaction and effective Bohr radius of the donors involved.

## I. INTRODUCTION

Far-infrared laser spectroscopy is a useful tool to probe the natures of shallow donors and the band-structure parameters in III-V and II-VI compound semiconductors. Here we concentrate on the study of resonant magneto-optical spin-flip transitions. Far-infrared observations of electric-dipole-induced electron-spin resonance (EDSR) were reported for the first time more than 20 years ago.<sup>1,2</sup> This pioneering work has given rise to a great deal of EDSR measurements involving either conduction electrons or donor electrons both in the cubic (zinc-blende) structure<sup>3-9</sup> and in the hexagonal (wurtzite) structure.<sup>10,11</sup> The observed spin resonance is a magneto-optical spin-flip transition between two spin states belonging to the same Landau subband of the conduction electron, or the same orbital ground state of the donor electron. Surprisingly, the intensity of the spin-resonance line is up to two orders of magnitude stronger than the symmetry-allowed magnetic-dipole interaction due to the intrinsic magnetic moment of the electron. Furthermore, its intensity is anisotropic, and changes when the applied magnetic field is reversed or the direction of the incidence radiation is modified.<sup>6,7</sup>

The above experimental facts have been attributed to the combined action of the magnetic-dipole amplitude and the electric-dipole contribution arising from a parity-violating coupling process which mixes states with opposite spin orientations and parities.<sup>12</sup> Different coupling mechanisms have been proposed by theoretical investigations: the effect of spin-orbit coupling in crystals lacking centers of inversion symmetry,<sup>10,11,13-20</sup> the nonparabolicity of the conduction band,<sup>21-24</sup> the warping of constant energy,<sup>25</sup> inhomogeneities that lower the symmetry of the crystal,<sup>26,27</sup> and the effect of exchange interaction in semimagnetic semiconductors.<sup>28-30</sup>

A critical analysis of the previously mentioned experimental and theoretical studies show that the predominant contribution to the coupling mechanism arises from spin-orbit interaction with inversion asymmetry. From group-theoretical arguments, this interaction is linear in the electron

quasimomentum  $k$  for the materials having a wurtzite structure, and proportional to  $k^3$  in materials having a zinc-blende structure, respectively.

The present work attempts to develop a theory of EDSR transitions based on the spin-orbit coupling mechanism but avoiding the knowledge of the crystal symmetry up to the last step of the calculations of the integrated absorption coefficient.

Section II is devoted to a general formalism based on Dalgarno's "operator technique" in perturbation theory. Simple analytic expressions of the electric-dipole operator are obtained for intermediate and weak magnetic-field intensities. In Sec. III we apply the results of Sec. II to the determination of the integrated absorption coefficient in cubic ( $T_d$ ) and hexagonal ( $C_{6v}$ ) crystals. In Sec. IV we analyze the far-infrared magneto-optical transmission data in InSb (Ref. 8) and  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$  (Refs. 10 and 11) on the basis of previously obtained results.

## II. GENERAL FORMALISM

We consider the absorption of an electromagnetic wave of frequency  $\omega$  and intensity  $I_0$  by bound electrons in the presence of a uniform magnetic field  $\mathbf{B}_0 = \nabla \times \mathbf{A}_0$  corresponding to the vector potential  $\mathbf{A}_0$ . Let  $\mathbf{A}$  be the vector potential of the electromagnetic wave with a wave vector  $\mathbf{q}$  propagating along the direction of the unit vector  $\mathbf{q} = n(\omega/c)\hat{\mathbf{q}}$  in a material of index of refraction  $n$  and polarized along the  $\hat{\mathbf{e}}$  direction. Since we are only interested in the one-photon process, the electron-photon interaction is linear in the vector potential  $\mathbf{A}$  and the alternating field of the incident radiation  $\mathbf{B}$ .

As mentioned in Sec. I, we concentrate only on resonant transitions. At the resonant frequency  $\omega = \omega_{fi}$ , the integrated absorption coefficient  $\alpha_I = \alpha \Gamma_0$  is given by<sup>20,31</sup>

$$\alpha_I = 2\pi \frac{n_e}{n} \omega \alpha_0 |\langle \psi_f | M | \psi_i \rangle|^2, \quad (2.1a)$$

with

$$M = \frac{1}{\hbar\omega} \{[\hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}, H], e^{i\mathbf{q} \cdot \mathbf{r}}\} - \frac{n}{e} \left\{ (\hat{\mathbf{q}} \times \hat{\boldsymbol{\varepsilon}}) \frac{\partial H}{\partial \mathbf{B}_0}, e^{i\mathbf{q} \cdot \mathbf{r}} \right\}. \quad (2.1b)$$

$H$  is the effective-mass Hamiltonian of the electron bound to the donor and in the presence of a uniform magnetic field,  $\alpha_0 = e^2/\hbar c$  is the fine-structure constant,  $n_e$  is the number of electrons per unit volume involved in the absorption process, and  $\Gamma_0$  is the full width at half-height of the absorption line. Here, and in what follows,  $[\mu, \nu]$  and  $\{\mu, \nu\}$ , respectively, denote the commutator and anticommutator of the operators  $\mu$  and  $\nu$ .

We describe the states  $|\psi\rangle$  in Eq. (2.1) within the theory of shallow donors.<sup>32</sup> We choose a Cartesian coordinate system  $(\hat{\xi}, \hat{\eta}, \hat{\zeta})$  whose  $\hat{\zeta}$  axis is parallel to  $\mathbf{B}_0$ . Notice that this coordinate system is in general distinct from the cubic axis  $\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}$  for a given experimental field configuration. Using the symmetric configuration gauge, for the effective-mass Hamiltonian  $H$  we obtain

$$H = H_0 + H_\sigma + H_{SO}, \quad (2.2a)$$

$$H_0 = \frac{p^2}{2m^*} - \frac{e^2}{\varepsilon_0 r} + \mu_B \frac{m_0}{m^*} B_0 L_\zeta + \frac{1}{8} \frac{e^2 B_0^2}{m^* c} (\xi^2 + \eta^2) \quad (2.2b)$$

$$H_\sigma = \frac{1}{2} g \mu_B B_0 \sigma_\zeta, \quad (2.2c)$$

$H_{SO}$  is the spin-orbit coupling. Here  $m^*$  and  $m_0$  respectively, are the effective mass of the electron and the free-electron mass;  $\varepsilon_0$  is the static dielectric constant;  $\mu_B$  is the Bohr magneton; and  $g$  is the effective Landé  $g$  factor of the bound electron.

Of course for a given crystal symmetry, the Hamiltonian operator Eq. (2.2) contains, in addition, higher-order terms in the electron wave vector  $\mathbf{k} = \mathbf{p}/\hbar + \mathbf{A}_0/\hbar c$ . In the present paper we neglect these contributions, as well as the weak anisotropy of the effective Landé  $g$  factor.<sup>8,19,33</sup> Nevertheless, we take into account the inversion-asymmetric form of the spin-orbit Hamiltonian operator  $H_{SO}$ , as demonstrated in Sec. III.

Let  $H_{SO}$  be a small perturbation with respect to  $H_0 + H_\sigma$ , and suppose that the state vectors  $\{|\nu, s\rangle\}$  with orbital quantum numbers denoted collectively by  $\nu$  and spin quantum numbers  $s$ , are solutions of

$$(H_0 + H_\sigma)|\nu, s\rangle = (E_\nu + g\mu_B B_0 s)|\nu, s\rangle. \quad (2.3)$$

However, the transitions involved in expression (2.1) are related to the states  $|\psi_i\rangle$  and  $|\psi_f\rangle$  of the total Hamiltonian operator  $H$ . It is convenient to describe these states by means of the unitary transformation  $e^{iS}$  acting on the unperturbed states  $|\nu, s\rangle$ :  $|\psi_i\rangle = e^{iS}|\nu, s\rangle$  and  $|\psi_f\rangle = e^{iS}|\nu', s'\rangle$ . The Hermitian operator  $S$  is then determined so that the quantity  $e^{-iS} H e^{iS}$  is diagonal in the representation  $\{|\nu, s\rangle\}$ . As a result,  $S$  is given by the condition

$$H_{SO} + i[H_0 + H_\sigma, S] = 0 \quad (2.4)$$

to first order in  $H_{SO}$ . Condition (2.4) clearly shows that at the first order in  $S$ , the integrated absorption coefficient obtained from Eq. (2.1)

$$\alpha_I = 2\pi \frac{n_e}{n} \omega \alpha_0 |\langle \nu', s' | M + i[M, S] | \nu, s \rangle|^2, \quad (2.5)$$

leads to an additional contribution ( $[M, S]$ ) due to spin-orbit coupling.<sup>34,35</sup>

Here we limit ourselves to electric-dipole and magnetic-dipole transitions between the Zeeman sublevels  $|\pm \frac{1}{2}\rangle$  associated with the donor ground state  $|0\rangle$ . Since the transition takes place in the donor ground state, only the part  $(M + i[M, S])_g$ , which is even under inversion, has to be considered in Eq. (2.5). Using relation (2.4) and the Jacobi identity, for the even-parity transition operator we obtain

$$(M + i[M, S])_g = \frac{2i}{\hbar\omega} \{H_0 + H_\sigma, [S, \hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}]\} - \frac{n}{2} g \lambda (\hat{\mathbf{q}} \times \hat{\boldsymbol{\varepsilon}}) \cdot \boldsymbol{\sigma}. \quad (2.6)$$

Here  $\lambda$  is the Compton wavelength.

The first contribution on the right-hand side of Eq. (2.6) is responsible for the parity-violating electric-dipole transition, whereas the second term in the right-hand side of Eq. (2.6) leads to the well-known paramagnetic resonance. We now exploit the fact that we are interested in spin-flip transitions which connect the two states  $|0, \frac{1}{2}\rangle$  and  $|0, -\frac{1}{2}\rangle$ . Therefore, it is useful to express the operators  $S$  and  $H_{SO}$  in terms of the Pauli matrices  $\sigma_\xi$ ,  $\sigma_\eta$ , and  $\sigma_\zeta$  and their linear combinations

$$\sigma_\pm = \sigma_\xi \pm i\sigma_\eta \quad (2.7)$$

defining

$$S = S_\zeta \sigma_\zeta + \frac{1}{2} S_+ \sigma_- + \frac{1}{2} S_- \sigma_+ \quad (2.8a)$$

and

$$H_{SO} = H_\zeta \sigma_\zeta + \frac{1}{2} H_+ \sigma_- + \frac{1}{2} H_- \sigma_+. \quad (2.8b)$$

Condition (2.4) requires

$$H_\zeta + i[H_0, S_\zeta] = 0, \quad (2.9a)$$

$$H_\pm + i[H_0, S_\pm] \mp i g \mu_B B_0 S_\pm = 0. \quad (2.9b)$$

Finally, inserting Eq. (2.9) into Eq. (2.6), the spin-flip matrix elements become

$$\begin{aligned} \langle 0, \mp \frac{1}{2} | (M + i[M, S]) | 0, \pm \frac{1}{2} \rangle = & i \left\{ \mp \langle 0 | [S_\pm, \hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}] | 0 \rangle \right. \\ & \left. - i \frac{n}{4} g \lambda (\hat{\mathbf{q}} \times \hat{\boldsymbol{\varepsilon}})_\pm \right\} \end{aligned} \quad (2.10)$$

at the resonant condition  $\hbar\omega = g\mu_B B_0$ .

The absorption process associated with a transition from state  $|0, \frac{1}{2}\rangle$  to state  $|0, -\frac{1}{2}\rangle$  [upper signs in Eq. (2.10)] occurs in crystals with a negative Landé  $g$  factor, while for crystals with a positive Landé  $g$  factor this process takes place from the lower state  $|0, -\frac{1}{2}\rangle$  to the upper state  $|0, \frac{1}{2}\rangle$ .

We notice that the paramagnetic contribution in Eq. (2.10) is obtained in a straightforward manner for a given field configuration. In order to carry out the expectation value over the orbital ground state  $|0\rangle$  involved in the electric-dipole contribution of Eq. (2.10), we introduce a complete orthogo-

nal set of eigenvectors of  $H_0$  with the eigenvalues  $E_\nu$ . Using Eq. (2.9b), this allows us to write

$$\langle 0|[S_\pm, \hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}]|0\rangle = -i \sum_\nu \left( \frac{\langle 0|H_\pm|\nu\rangle \langle \nu|\hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}|0\rangle}{E_\nu - E_0 \pm g\mu_B B_0} + \frac{\langle 0|\hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}|\nu\rangle \langle \nu|H_\pm|0\rangle}{E_\nu - E_0 \mp g\mu_B B_0} \right), \quad (2.11)$$

From expression (2.11) it is evident that the spin-orbit coupling mixes orbital states  $|\nu\rangle$  of different parity, and that the predominant contributions to the electric-dipole transition intensity arises from the terms in the sum over  $\nu$  having small denominators. Therefore, crystals with shallow donors and large Landé  $g$  factors seem to be the best candidates for observing such transitions.

We know that an exact solution for the states  $|\nu\rangle$  is not available. Previous calculations<sup>11,15</sup> have limited themselves to one or two terms in the sum over  $\nu$  and at weak magnetic-field intensities.<sup>18,19</sup> Here we shall eliminate the summation over the intermediate states  $|\nu\rangle$  (including the continuum states) by means of the so-called ‘‘operator technique’’ first used by Dalgarno and Lewis.<sup>36</sup> Since then, this technique has been successfully applied to a great number of problems<sup>19,37</sup> in the framework of perturbation theory.

We define the dipole operators satisfying the conditions

$$([\widehat{\mathbf{D}}_\pm, \widehat{\mathbf{H}}_0] \mp g\mu_B B_0 \widehat{\mathbf{D}}_\pm)|0\rangle = \hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}|0\rangle. \quad (2.12)$$

Inserting Eq. (2.12) into Eq. (2.11) leads directly to the simple form

$$\langle 0|[S_\pm, \hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}]|0\rangle = i \langle 0|H_\pm \widehat{\mathbf{D}}_\pm + \widehat{\mathbf{D}}_\mp^\dagger H_\pm|0\rangle \quad (2.13)$$

for the electric-dipole contribution.

Finally taking into account Eqs. (2.5), (2.6), (2.10), and (2.13), the integrated absorption coefficient is given by

$$\alpha_I = 2\pi \frac{n_e}{n} \omega \alpha_0 |\langle 0, \mp \frac{1}{2} | (M + i[M, S])_g | 0, \pm \frac{1}{2} \rangle|^2, \quad (2.14a)$$

with

$$\langle 0, \mp \frac{1}{2} | (M + i[M, S])_g | 0, \pm \frac{1}{2} \rangle = \pm \langle 0|H_\pm D_\pm + D_\mp^\dagger H_\pm|0\rangle + \frac{n}{4} g \chi (\hat{\mathbf{q}} \times \hat{\boldsymbol{\varepsilon}})_\pm. \quad (2.14b)$$

It is worthwhile to notice that this approach enables us to determine the integrated absorption coefficient for any kind of crystal structure, i.e., for any form of the spin-orbit interaction provided. We know the expressions for the electric-dipole operators  $D_\pm$  from Eq. (2.12). This means that an accurate description of only the orbital ground state is required in order to find solutions for the operators  $D_\pm$ , which we assume to be functions of positions alone. For the ground-state wave function  $\langle \mathbf{r}|0\rangle$  we choose the following trial envelope function:

$$\langle \mathbf{r}|0\rangle = \frac{1}{\sqrt{(2\pi)^{3/2} a_t^* a_l^*}} \exp\left(-\frac{\xi^2 + \eta^2}{4a_t^{*2}} - \frac{\zeta^2}{4a_l^{*2}}\right) \quad (2.15)$$

where  $a_t^* = a^* a_t$  and  $a_l^* = a^* a_l$  ( $a^* = \varepsilon_0 \hbar^2 / m^* e^2$  is the effective Bohr radius) are physical parameters describing the electron orbit respectively in the plane perpendicular to the direction of the magnetic field  $\mathbf{B}_0$  and parallel to the direction of  $\mathbf{B}_0$ .

This is the well-known trial function proposed by Yafet, Keyes, and Adams<sup>38</sup> for the problem described by  $H_0$  [see Eq. (2.2b)]. The variational parameters  $a_t$  and  $a_l$  are then obtained, minimizing the expression for the energy:

$$E(a_t, a_l) = R^* \left[ \frac{1}{2a_t^2} \left( 1 + \frac{\varepsilon^2}{2} \right) + \frac{\gamma^2 a_t^2}{2} - \left( \frac{2}{\pi} \right)^{1/2} \varepsilon (1 - \varepsilon^2)^{1/2} \times \ln \frac{1 - (1 - \varepsilon^2)^{1/2}}{1 + (1 - \varepsilon^2)^{1/2}} \right], \quad (2.16a)$$

with

$$\varepsilon = \frac{a_t}{a_l}, \quad (2.16b)$$

Here we have used effective units:  $2R^* = m^* e^4 / \varepsilon_0^2 \hbar^2$  is the unit of energy, and  $a^*$  is the unit of length.  $\gamma = \mu_B (m_0 / m^*) (B_0 / R^*)$ , and  $R_0 = \sqrt{\hbar c / e B_0}$  is the Landau length.

We know that this trial function is an excellent description of a donor in the presence of a magnetic field for  $\gamma > 1$ , and is even satisfactory for  $\gamma < 1$ . Moreover, it has the advantage of providing exact solutions to equations (2.12), as we shall see later. Finally, we shall show in Sec. III that resonant spin-flip transition intensities associated with conduction electrons are recovered, taking the limits  $a_t = 1/\sqrt{\gamma}$  and  $a_l = \infty$ .

Substituting Eq. (2.15) into Eq. (2.12), we obtain the following differential equation for  $D_\pm(\rho, \theta, \varphi)$ :

$$a^{*2} \nabla^2 \widehat{\mathbf{D}}_\pm + \frac{2a^{*2}}{\psi_0} \nabla \widehat{\mathbf{D}}_\pm \cdot \nabla \psi_0 + i\gamma \frac{\partial \widehat{\mathbf{D}}_\pm}{\partial \varphi} \mp \gamma g \frac{m^*}{m_0} \widehat{\mathbf{D}}_\pm = 2 \frac{m^* a^{*2}}{\hbar^2} \hat{\boldsymbol{\varepsilon}} \cdot \mathbf{r}, \quad (2.17)$$

where  $\theta$  and  $\varphi$  are the polar angles of  $\mathbf{r}$  with  $\boldsymbol{\zeta}$  as a polar axis. Knowing the structure of  $\boldsymbol{\varepsilon} \cdot \mathbf{r}$ , the right-hand side of equation Eq. (2.17), we search for a solution of  $D_\pm$  in terms of spherical harmonics  $Y_1^{m_l}(\theta, \varphi)$ . It is easily seen that due to the orthogonality of the spherical harmonics, this solution is restricted to the following form:

$$\widehat{\mathbf{D}}_\pm(r, \theta, \varphi) = \frac{2m^* a^{*3}}{\hbar^2} [\hat{\boldsymbol{\varepsilon}}_\zeta D_\pm^0(r) Y_1^0(\theta, \varphi) + \hat{\boldsymbol{\varepsilon}}_- D_\pm^1(r) Y_1^1(\theta, \varphi) + \hat{\boldsymbol{\varepsilon}}_+ D_\pm^{-1}(r) Y_1^{-1}(\theta, \varphi)] \quad (2.18)$$

Here  $\boldsymbol{\varepsilon}_\zeta$  corresponds to the linear polarization of the electric-field vector along the direction of the unit vector  $\hat{\boldsymbol{\zeta}}$  and  $\hat{\boldsymbol{\varepsilon}}_\pm = (\hat{\boldsymbol{\zeta}} \pm i \hat{\boldsymbol{\eta}}) / \sqrt{2}$  are, respectively, related to circularly polarized light with positive and negative helicity.

Using the reduced variable  $R = r/a^*$ , the coefficients  $D_\pm^{m_l}(m_l: 0, \pm 1)$  in Eq. (2.18) satisfy the ‘‘radial’’ differential equations

$$\frac{d^2 D_{\pm}^{m_l}}{dR^2} + 2 \left( \frac{1}{R} - u^{m_l} R \right) \frac{dD_{\pm}^{m_l}}{dR} - \frac{2}{R^2} D_{\pm}^{m_l} - W_{\pm}^{m_l} D_{\pm}^{m_l} = R, \quad (2.19a)$$

with

$$W_{\pm}^{m_l} = v^{m_l} + \Gamma_{\pm}^{m_l} \quad (2.19b)$$

$$\Gamma_{\pm}^{m_l} = \gamma \left( m_l \pm g \frac{m^*}{m_0} \right). \quad (2.19c)$$

For  $m_l=0$ , we have

$$u^0 = \frac{1}{2} \left( \frac{2}{5} \frac{1}{a_t^2} + \frac{3}{5} \frac{1}{a_l^2} \right), \quad (2.19d)$$

$$v^0 = -\frac{2}{5} \left( \frac{1}{a_t^2} - \frac{1}{a_l^2} \right), \quad (2.19e)$$

and, for  $m_l=1, -1$ ,

$$u^1 = u^{-1} = \frac{1}{2} \left( \frac{4}{5} \frac{1}{a_t^2} + \frac{1}{5} \frac{1}{a_l^2} \right), \quad (2.19f)$$

$$v^1 = v^{-1} = \frac{1}{5} \left( \frac{1}{a_t^2} - \frac{1}{a_l^2} \right). \quad (2.19g)$$

A particular solution for  $D_{\pm}^{m_l}$  is adequate as long as it represents a physical acceptable solution of Eq. (2.19a). Therefore we attempt to find nonsingular solutions in the form of a power series in  $R$ :

$$D_{\pm}^{m_l} = \sum_{n=0}^{\infty} \alpha_{n,m} R^n. \quad (2.20)$$

Substituting  $D_{\pm}^{m_l}$  in Eq. (2.19), and equating equal powers of  $R$ , yields

$$\alpha_{0,m}^{\pm} = \alpha_{2,m}^{\pm} = 0, \quad (2.21a)$$

$$10\alpha_{3,m}^{\pm} - (2u^m + W_{\pm}^m) \alpha_{1,m}^{\pm} = 1, \quad (2.21b)$$

$$18\alpha_{4,m}^{\pm} - (4u^m + W_{\pm}^m) \alpha_{2,m}^{\pm} = 0, \quad (2.21c)$$

and, for  $\alpha_{n,m}^{\pm}$ ,

$$(n-1)(n+2)\alpha_{n,m}^{\pm} - [2(n-2)u^m + W_{\pm}^m] \alpha_{n-2,m}^{\pm} = 1. \quad (2.21d)$$

As a consequence, there are no even powers in  $R$ , and the asymptotic solutions of Eq. (2.19), as  $R$  tends to infinity, are singular. However, there exists a polynomial solution satisfying the condition of Eq. (2.21b) with  $\alpha_{3,m}^{\pm} = 0$ . This leads to the simple form

$$D_{\pm}^{m_l} = -\frac{1}{(2u^{m_l} + v^{m_l}) + \Gamma_{\pm}^{m_l}} R. \quad (2.22)$$

Taking Eqs. (2.19) and Eq. (2.22) into account shows that circular polarized waves affect the transverse electron orbit of radius  $a_t$ , while waves linearly polarized along the mag-

netic field  $\mathbf{B}_0$  affect the longitudinal electron motion characterized by the orbital radius  $a_l$ .

In a similar manner, we can obtain an analytic expression for the electric-dipole operator valid in the limit  $\gamma \ll 1$ . In this limit, and in order to take into account the electronic lengthening of the wave function along the magnetic field, it is evident to choose a cylindrical (hydrogenic) for the envelope function  $\langle \mathbf{r} | 0 \rangle$ . Again, the solution of Eq. (2.12) can be written in the form of Eq. (2.19). However, as shown in the Appendix, the radial coefficients are now expressed by a series expression.

In order to obtain a more precise solution for the electric dipole at all magnetic-field domains, we may consider the following linear combination:

$$\psi_{\text{com}} = A \psi_1 + B \psi_2. \quad (2.23)$$

$A$  and  $B$  are variational parameters, and  $\psi_1$  ( $\psi_2$ ) is the CYL (YKA) wave function. Calculations with this function are in progress. Dexter<sup>39</sup> has proposed a wave function with three variational parameters for studying the hydrogenic impurity polarizability. The com wave function has the advantage of possessing only two variational parameters. On the other hand, the Dexter wave function must be rejected for the weak magnetic fields because the Bohr radius thus obtained are greater than the unit and have no physical meaning. In spite of this error the Dexter wave function is widely used in the literature.

Knowing the explicit form of the electric-dipole operator, we are now in a position to determine the integrated absorption coefficient for a given field configuration. This can be easily achieved by carrying out the expectation values over the orbital ground state [cf. Eq. (2.14)] for a given form of the spin-orbit interaction. In Sec. III we apply the present formalism to materials of different crystal structure.

### III. INTEGRATED ABSORPTION COEFFICIENT IN HEXAGONAL AND CUBIC SEMICONDUCTORS

The purpose of the present section is to illustrate the general formalism developed in Sec. II by considering absorption in the hexagonal (wurtzite) structure with point-group symmetry  $C_{6v}$  and in the cubic (zinc-blende) structure with point-group symmetry  $T_d$ , respectively. We calculate the intensity of the electric-dipole spin resonance for different field configurations.

#### A. Wurtzite structure

As mentioned above, the determination of the electric-dipole spin-resonance contributions implies the knowledge of the spin-orbit interaction  $H_{\text{SO}}$  for a given crystal structure. This information can be obtained in an elegant manner by group-theoretical arguments based on the theory of invariants. We search of a form of the spin-orbit interaction which leaves the Hamiltonian  $H_{\text{SO}}$  invariant under the symmetry operations of the point group  $C_{6v}$ , as well as under the operation of time reversal. We construct this invariant with products of the kind  $\sigma_i k_j^n$  formed by the components of the Pauli matrices  $\sigma_x$ ,  $\sigma_y$ , and  $\sigma_z$ , and the electron wave number

components  $k_x$ ,  $k_y$ , and  $k_z$ . Using the character table and basic functions associated with the point group  $C_{6v}$ , we obtain

$$H_{SO} = \frac{\lambda}{2} (\hat{\sigma} \times \mathbf{k}) \cdot \mathbf{z} + \frac{\lambda_l}{2} \{k_z^2, (\hat{\sigma} \times \mathbf{k}) \cdot \mathbf{z}\} + \frac{\lambda_t}{2} \{(k_x^2 + k_y^2), (\hat{\sigma} \times \mathbf{k}) \cdot \mathbf{z}\} \quad (3.1)$$

if we restrict ourselves up to  $n=3$ . Here  $\lambda$ ,  $\lambda_l$ , and  $\lambda_t$  are phenomenological parameters characterizing the strength of the spin-orbit coupling. For the purpose of the present work we neglect the high-order  $O(k^3)$  term<sup>15</sup> in Eq. (3.1).

In the usual experimental geometry for hexagonal crystals, the  $\hat{\mathbf{c}}$ -crystal axis is taken along the cubic  $\hat{\mathbf{z}}$  axis, forming an angle  $\beta$  with the magnetic field in the plane  $(\hat{\mathbf{z}}, \hat{\mathbf{x}})$ . The crystal or the magnetic field is rotated about the  $\hat{\mathbf{y}}$  axis. This leads to the following expression for  $H_{SO}$  in the coordinate system  $(\hat{\xi}, \hat{\eta}, \hat{\zeta})$ , with the  $\hat{\zeta}$  axis along  $\mathbf{B}_0$ :

$$H_{SO} = \frac{\lambda}{2} (\hat{\mathbf{c}} \times \boldsymbol{\sigma}) \cdot \mathbf{k}, \quad (3.2a)$$

with

$$H_{\xi} = \frac{\lambda}{2} \sin \beta k_{\eta} \quad (3.2b)$$

and

$$H_{\pm} = \mp i \frac{\lambda}{2} [\sin \beta k_{\xi} + \cos \beta (k_{\xi} \pm i k_{\eta})]. \quad (3.2c)$$

Using the envelope function Eq. (2.15) and the corresponding electric-dipole operators Eqs. (2.18) and (2.22), the expectation value involved in the electric-dipole contribution (2.13) can be obtained by straightforward integration. This leads to the following analytic expressions for the integrated absorption coefficient Eqs. (2.14).

(i) *Faraday configuration* ( $\mathbf{B}_0 \parallel \hat{\zeta} \parallel \hat{\mathbf{n}}$  and  $g > 0$ ). We know that only the polarization with positive helicity  $\hat{\mathbf{e}}_+ = (\hat{\xi} + i \hat{\eta})/\sqrt{2}$  contributes [also called cyclotron resonance active (CRA)]. We find

$$\alpha_I = n_e \frac{g^2 n^2}{2c} \alpha_0 \chi^2 \omega \left\{ 1 + \frac{1}{g^2 n^2} \left( \frac{\lambda}{R^* \lambda} \right)^2 \left[ \frac{4\mu\gamma}{a_i^{-4} - \Gamma^2} \right]^2 \right\}, \quad (3.3)$$

$$\Gamma = \gamma(1 - 2\mu), \quad (3.4)$$

and

$$\mu = \frac{1}{2} |g| \frac{m^*}{m_0}, \quad (3.5)$$

and for the free-electron case ( $a_i = 1/\sqrt{\gamma}$ ) we obtain an expression in agreement with previous work.<sup>18</sup>

(ii) *Parallel Voigt geometry* ( $\mathbf{B}_0 \parallel \hat{\zeta} \parallel \hat{\mathbf{e}}$ , with  $\hat{\mathbf{n}} \perp \hat{\mathbf{c}}$  and  $\mathbf{B}_0 \parallel \hat{\mathbf{c}} \parallel \hat{\xi}$ ):

$$\alpha_I = n_e \frac{g^2 n^2}{4c} \alpha_0 \chi^2 \omega \left\{ 1 + \frac{1}{g^2 n^2} \left( \frac{\lambda}{R^* \lambda} \right)^2 \left[ \frac{2\Gamma}{\Gamma^2 - \frac{1}{a_i^2}} \right]^2 \right\} \quad (3.6)$$

taking the limiting case ( $a_i \rightarrow \infty$ ), we obtain an expression in agreement with the result obtained by Gopalan *et al.*<sup>18</sup> for band electrons.

## B. Zinc-blende structure

For this structure, one can show that previously developed symmetry arguments lead to the following<sup>13,20</sup> spin-orbit operator  $H_{SO}$ :

$$H_{SO} = \delta_0 \boldsymbol{\sigma} \cdot \boldsymbol{\kappa}, \quad (3.7)$$

$$H_{SO} = \delta_0 [\sigma_x \cdot \kappa_x + \sigma_y \cdot \kappa_y + \sigma_z \cdot \kappa_z], \quad (3.8a)$$

with

$$\kappa_x = k_y k_x k_y - k_z k_x k_z \quad (3.8b)$$

$$\kappa_y = k_z k_y k_z - k_x k_y k_x \quad (3.8c)$$

$$\kappa_z = k_x k_z k_x - k_y k_z k_y. \quad (3.8d)$$

To obtain the form of the spin-orbit operator according to Eq. (2.8), we use the Euler angles<sup>40</sup>  $\alpha$ ,  $\beta$ , and  $\gamma$  of the coordinate system  $(\hat{\xi}, \hat{\eta}, \hat{\zeta})$  with respect to the cubic axis  $(\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}})$ .<sup>20</sup>

Proceeding in the same manner in Sec. III A we find the following results for different field configurations.

(i) *Faraday configuration* ( $\mathbf{B}_0 \parallel \hat{\zeta} \parallel \hat{\xi}$ ). We find

$$\alpha_I = n_e \pi g^2 n \alpha_0 \chi^2 \omega \left| \left\{ 1 + \frac{16}{3} i A_{\text{CRI}}^b F_0 \right\} \right|^2, \quad (3.9a)$$

with

$$A_{\text{CRI}}^b = - \frac{3 \delta_0 \sqrt{\varepsilon_0}}{4 \alpha_0 \mu} [P(\gamma) - P(-\gamma)] \quad (3.9b)$$

and

$$P(\gamma) = \frac{1 + a_i^2 \gamma}{1 + a_i^2 \gamma (1 - 2\mu)} [1 - \varepsilon^2 + a_i^4 \gamma^2], \quad (3.9c)$$

$$F_0 = - \frac{3}{16} i \sin 2\alpha \sin 2\beta \sin \beta \quad (3.10)$$

for donor electrons.

Here we have taken the polarization of negative helicity  $\hat{\mathbf{e}}_- = (\hat{\xi} - i \hat{\eta})/\sqrt{2}$  [also called cyclotron resonance inactive (CRI) polarization]. This situation corresponds, for instance, to the case of InSb, whose effective Landé  $g$  factor is negative ( $g < 0$ ).

In the free-electron case ( $a_i = 1/\sqrt{\gamma}$ ,  $a_i \rightarrow \infty$ ), we recover the value for  $A_{\text{CRI}}$ .<sup>17</sup> Thus

$$\frac{A_{\text{C}}^b}{A_{\text{C}}} = (\mu - 1) [P(-\gamma) - P(\gamma)]. \quad (3.11)$$

(ii) *Parallel Voigt geometry* ( $\mathbf{B}_0 \parallel \hat{\zeta}$ ), with  $\hat{\mathbf{n}} = (0, 1, 0)$  and  $\hat{\mathbf{e}} = (0, 0, 1)$ . We obtain

$$\alpha_I = n_e \frac{\pi}{2} g^2 n \alpha_0 \chi^2 \omega \left| \{ 1 - 16 i A_{\text{OV}}^b F_1 \} \right|^2, \quad (3.12a)$$

with

$$A_{OV}^b = \frac{3\delta_0\sqrt{\varepsilon_0}}{\alpha_0} \frac{a_t^2\gamma}{(2\mu a_t^2\gamma - \varepsilon^2)(2\mu a_t^2\gamma + \varepsilon^2)} \times [2 - \varepsilon^2 - (1 - \gamma^2 a_t^4)], \quad (3.12b)$$

$$F_1 = \frac{1}{16} e^{i\gamma} [\cos 2\alpha \sin 2\beta + i \sin 2\alpha \sin \beta (3 \cos^2 \beta - 1)] \quad (3.13)$$

for donor-bound electrons. For band electrons we recover the value for  $A_{OV}$ ,<sup>17</sup> which yields

$$\frac{A_{OV}^b}{A_{OV}} = \frac{2\mu a_t^2\gamma(1 + \mu a_t^4\gamma^2 - \varepsilon^2)}{[(2\mu a_t^2\gamma)^2 - \varepsilon^4]}. \quad (3.14)$$

#### IV. COMPARISON WITH EXPERIMENT

The object of the present section is to compare the theory developed above to far-infrared magneto-optical transmission data respectively in InSb (Ref. 8) and  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$ .<sup>10,11</sup> Physical parameters characterizing the shallows donor in this materials will be obtained from the best fit between the theoretical model and measurements.

##### A. Parallel Voigt configuration in *n*-type InSb

Barticevic *et al.*<sup>8</sup> measured the spin-resonance absorption coefficient for donor-bound electrons of InSb observed at  $118.8 \mu\text{m}$  in the parallel Voigt geometry as a function of orientation of  $\mathbf{B}_0$  in the  $(1, \bar{1}, 1)$  plane. The measurements were carried out at low temperatures ( $T=4.5 \text{ K}$ ) with a value of  $\mathbf{B}_0=41.14 \text{ kG}$  at the resonance.

In order to compare theory with experiment, we first have to find the Euler angles corresponding to the experimental situation.<sup>8</sup> Therefore we take two unit vectors  $\hat{u}=1/\sqrt{2}[1,1,0]$  and  $v=1/\sqrt{6}[1,1,2]$  in the plane  $(1, \bar{1}, 1)$  and let  $\theta$  be the angle between  $\mathbf{B}_0$  and  $\hat{u}$ .

Thus  $\hat{\xi}$ ,  $\hat{\eta}$ , and  $\hat{\zeta}$  are known for a given angle  $\theta$ , and from the definition of the Euler angles<sup>40</sup> we obtain the relation

$$\tan \alpha = \frac{\sqrt{3} \cos \theta - \sin \theta}{\sqrt{3} \cos \theta + \sin \theta}, \quad (4.1a)$$

$$\cos \beta = -\sqrt{\frac{2}{3}} \sin \theta, \quad (4.1b)$$

$$\tan \gamma = -\frac{1}{\sqrt{2} \cos \theta}. \quad (4.1c)$$

Substituting Eq. (4.1) into Eq. (3.12a) leads to the following expression of the integrated absorption coefficient

$$\alpha_I = n_e \frac{\pi}{2} g^2 n \alpha_0 \lambda^2 \omega \{ [1 + f_1(x) A_{OV}^b + f_2(x) (A_{OV}^b)^2] \}^2, \quad (4.2)$$

with

$$f_1(x) = \frac{4}{\sqrt{6}} x(3 - 4x^2) \quad (4.3a)$$

and

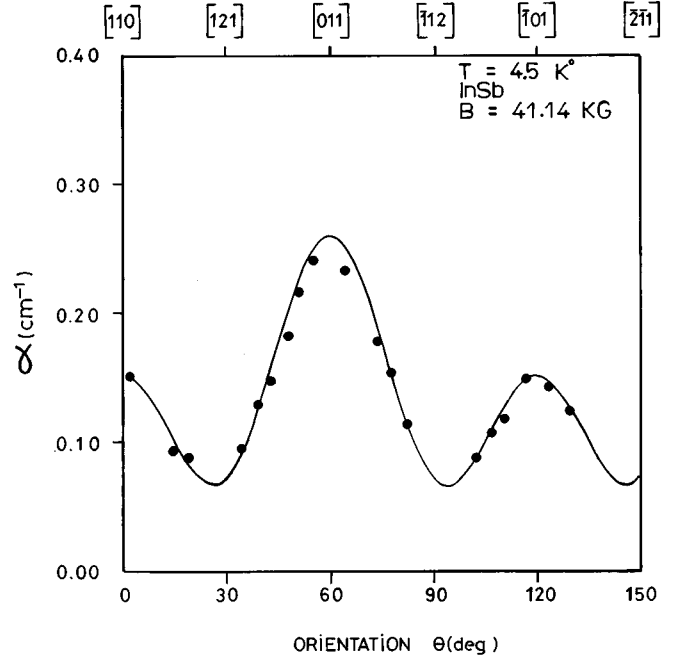


FIG. 1. Integrated absorption coefficient of a donor-bound electron of InSb observed at  $118.8 \mu\text{m}$  in the parallel Voigt geometry as a function of orientation of  $\mathbf{B}_0$  in the  $(1, \bar{1}, 1)$  plane. The measurements, taken from Ref. 8, were carried out at low temperatures ( $T=4.5 \text{ K}$ ). The solid line is the theoretical curve Eqs. (4.2) adjusted to the experiment, the best fit is obtained for  $A_{OV}^b = 6.2 \pm 0.4$ .

$$f_2(x) = \frac{1}{3} (1 + 18x^2 - 48x^4 + 32x^6), \quad (4.3b)$$

$$x = \cos \theta. \quad (4.3c)$$

The solid circles in Fig. 1 correspond to the measurements taken from Ref. 8. The solid line is the theoretical curve Eqs. (4.2) adjusted to the experiment. The best fit is obtained for a value of the quantity  $A_{OV}^b = 6.2 \pm 0.4$ . From the analytic form of Eqs. (4.2), we know that the maxima of the oscillations in Fig. 1 are given by  $x = \pm 0.5$  and  $x = \pm 1$ . It is interesting to note that the quantity  $A_{OV}^b$  determines the minima of these oscillations by the condition

$$A_{OV}^b (4x^2 - 3)x = \frac{3}{\sqrt{6}}. \quad (4.4)$$

This difference in amplitude between the  $[0,1,1]$  direction and the equivalent  $[1,1,0]$  and  $[1,0,1]$  directions is related to the effect of electric-dipole-magnetic-dipole interference, first discovered by Chen *et al.*<sup>7</sup> and Gopalan, Furdyna, and Rodriguez<sup>17</sup> for the free-electron-spin resonance.

Knowing the quantity  $A_{OV}^b$  we can now deduce a value for the spin-orbit coupling parameter  $\delta_0$  from Eq. (3.12b). We use the following material parameters for InSb:  $m^* = 0.013m_0$  and  $\varepsilon_0 = 17.9$ .<sup>8</sup> This leads a value of  $\gamma = 33.18$  at  $B_0 = 41.14 \text{ kG}$  and values  $a_t = 0.17$  and  $a_l = 0.39$  for the variational parameters deduced from Eqs. (2.16). At the resonance condition  $\hbar\omega = g\mu_B B_0$ , the effective Landé  $g$  factor is given by  $|g| = 43.82$ , which is in good agreement with previous work.<sup>8</sup> Inserting the above physical parameters into expression (3.12) yields  $\delta_0 = 56.23 \pm 3 \text{ u a} = (2.2 \pm 0.2) \times 10^{-22}$

eV cm. Within the experimental errors we find a value for  $\delta_0$  which is almost the same as the spin-orbit coupling parameter obtained for conduction electrons. This is very reasonable, because additional contributions to  $\delta_0$  due to the impurity potential are negligible with respect to the crystal potential.

### B. Faraday geometry in $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$

Dobrowolska *et al.*<sup>11</sup> measured the frequency dependence of the spin-resonance-integrated absorption coefficient of  $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$  in the Faraday configuration, and for several temperatures. The experiment was carried out at wavelengths varying from 251  $\mu\text{m}$  down to 95.5  $\mu\text{m}$ , and the resonant condition  $\hbar\omega = g\mu_B B_0$  was obtained for magnetic-field intensities varying from 0.5 to 8 T. The material parameters  $m^* = 0.12m_0$  and  $\varepsilon_0 = 10.02$  (Ref. 42) related to CdSe yield values of the physical parameter  $\gamma = 0.296$  for a magnetic field equal to 10 T. Therefore we shall compare the experimental data with theory using the cylindrical trial function in order to describe the donor ground state  $|0\rangle$  in the presence of a magnetic field. Inserting the expression for the dipole operator Eqs. (2.18) and (A8) (see the Appendix) into Eq. (2.14), for the Faraday geometry we find

$$\alpha_l = \frac{n_e}{2c} \alpha_0 \lambda^2 \omega \{ \varepsilon_0 g^2 + P^2(\omega) \}, \quad (4.5a)$$

with

$$P(\omega) = \frac{2a_t^2 \lambda \cos\beta}{\lambda R^*} a_l^{-2\gamma_1} \sum_{l=0}^{\infty} \delta_l \Delta_l^{2l+1} \quad (4.5b)$$

$$\Delta_l = \frac{\hbar\omega}{R^*} a_l^2 \left( 1 - \frac{m_0}{gm^*} \right), \quad (4.5c)$$

$$\delta_l = \left( \frac{10^4}{9 + \varepsilon^2} \right)^{4l} \left[ u_l + v_l \frac{(9 + \varepsilon^2)^2}{10} \right], \quad (4.5d)$$

$$u_l = 10 \left\{ \frac{4}{3} S(2l+1) + \left[ \frac{8}{15} S(2l+2) - \frac{2}{15} S(2l+1) \right] (\varepsilon^2 - 1) \right\}, \quad (4.5e)$$

$$v_l = \frac{1}{(1 - gm^*/m_0)} \left\{ \frac{4}{3} S(2l+1) \left[ \frac{16}{15} S(2l+2) + \frac{8}{15} S(2l+1) \right] (\varepsilon^2 - 1) \right\} \quad (4.5f)$$

$$S(l) = \sum_{k=0}^{l+2} c_{k,l}^{\pm 1} \frac{(2+k)!}{2^{2+k}} \quad (4.5g)$$

in the limit  $\gamma \ll 1$ , where only circularly polarized light with positive helicity contributes to  $\alpha_l$  since the effective Landé  $g$  factor is positive for  $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ , as we shall see. The coefficient  $c_{k,l}^{m_l}$  is given by the recursion relations shown in the Appendix. From Eqs. (4.5) we remark that  $\alpha_l(\omega)$  is expressed in the form of an expansion in powers of the angular frequency  $\omega$  of the incident light. The linear contribution is essentially imposed by the effective Landé  $g$  factor. Moreover, it can be shown that the sum over  $k > 0$  depends strongly on the parameters  $\lambda$ ,  $a_t$ , and  $a_l$ . In the limit  $\varepsilon = 1$  (the isotope case  $a_t = a_l = a$ ) Eqs. (4.5) agree with the result

obtained by Zorkani, Kartheuser, and Rodriguez,<sup>42</sup> and in the limit  $a_t = a_l = 1$ ,  $\beta = 0$ , and  $l = 0, 1$ , this equations agree with previous calculations.<sup>18,19,41-44</sup>

We now compare the theoretical model with the experimental data taken at 4.7 K for a magnetic field parallel to the  $\hat{c}$ -crystal axis ( $\beta = 0$ ) of the crystal and a donor concentration  $n_e = 2 \times 10^{16} \text{ cm}^{-3}$ . In addition to the effective mass  $m^*$ , the static dielectric constant  $\varepsilon_0$ , and the index of refraction  $n = \sqrt{\varepsilon_\infty}$ , there remain four other material parameters to be determined: the effective Landé  $g$  factor, the spin-orbit coupling parameter  $\lambda$ , the orbital radius described by  $a_l$ , and the anisotropy parameter  $\varepsilon$ .<sup>43</sup>

In the case of a semimagnetic semiconductor such as  $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ , the effective Landé  $g$  factor depends substantially on the temperature and applied magnetic field due to exchange interaction between the localized  $3d^5$  electrons of  $\text{Mn}^{2+}$  ions and the itinerant electrons.<sup>45</sup> The energies  $E_+$  of the spin-up state  $|0, +\frac{1}{2}\rangle$  and  $E_-$  of the spin-down state  $|0, -\frac{1}{2}\rangle$  associated with the donor ground state  $E_0$  can then be written in the form

$$E_{\pm} = E_0 \pm \frac{1}{2} (g^* \mu_B B_0 + N_s \alpha_e \langle S_z \rangle), \quad (4.6a)$$

where  $N_s$  is the effective  $\text{Mn}^{2+}$ -ion density,  $\alpha_e$  is the exchange integral for the  $s$ -like  $\Gamma_6$  electrons, and  $\langle S_z \rangle$  is the thermal average of the manganese spin in the magnetic-field direction. Hence

$$E_{\pm} = E_0 \pm \frac{1}{2} g \mu_B B_0, \quad (4.6b)$$

$$g = g^* + \frac{N_s \alpha_e \langle S_z \rangle}{\mu_B B_0}, \quad (4.6c)$$

Here  $g$  is an effective Landé  $g$  factor made up of two parts: the intrinsic contribution and the magnetic contribution. The intrinsic contribution  $g^*$  is essentially related to band effects. Here we take the value  $g^* = 0.5 \pm 0.1$  for CdSe, as obtained from magneto-optical studies of exciton states.<sup>46</sup> In large-gap semimagnetic semiconductors, the magnetic contribution resulting from the exchange energy  $N_s \alpha_e \langle S_z \rangle$  is the predominant contribution to  $g$ .

Expressing the exchange energy within the framework of the molecular-field approximation, the effective Landé  $g$  factor is given by

$$g = g^* + \frac{5}{2} \frac{N_s \alpha_e}{\mu_B B_0} B_{5/2}(y), \quad (4.7a)$$

$$y = \frac{5}{2} \frac{g_{\text{Mn}} \mu_B B_0}{k_B (T + T_{\text{AF}})}, \quad (4.7b)$$

where  $B_{5/2}(y)$  is the modified Brillouin function,  $g_{\text{Mn}} = 2$  is the Landé  $g$  factor of the ion, and  $T_{\text{AF}}$  is an effective temperature related to the antiferromagnetic interaction between  $\text{Mn}^{2+}$  ions. In the experimental range of excitation energies taken at the resonance condition  $\hbar\omega = g\mu_B B_0$ , we can neglect the magnetic-field dependence of  $g$ . Equations (4.7) then reduce to

$$g = g^* + \frac{35}{12} \frac{|N_s \alpha_e|}{k_B (T + T_{\text{AF}})} g_{\text{Mn}}. \quad (4.8)$$

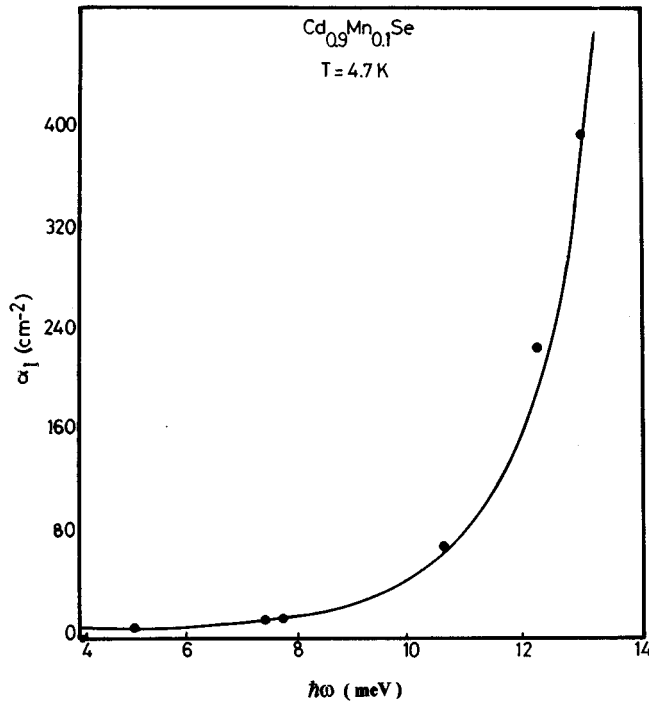


FIG. 2. Integrated absorption coefficient of a donor-bound electron of  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$  as a function of excitation energy. The solid circles correspond to the measurements in the Faraday configuration, taken from Ref. 11. The solid line is the theoretical curve Eqs. (4.5) adjusted to the experiment, the best fit is obtained for  $\lambda = (2.40 \pm 0.2) \times 10^{-10}$  eV cm,  $a_t = 43.5$  Å, and  $\varepsilon = 0.998$ .

Adjusting expression (4.8) to spin-flip Raman-scattering data<sup>47,48</sup> performed on the same ingot of  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$  yields  $|\alpha_e N_s| = 11.9$  and  $T_{\text{AF}} = 2.7$  K.

In Fig. 2 we have represented the integrated absorption coefficient as a function of excitation energy. The solid circles correspond to the measurements taken from Ref. 11. The solid line is the theoretical curve Eqs. (4.5) adjusted to the experiment with  $g = 120$  for  $T = 4.5$  K; the best fit is obtained for  $\lambda = (2.40 \pm 0.2) \times 10^{-10}$  eV cm,  $a_t = 43.5$  Å, and the anisotropy parameter  $\varepsilon = 0.998$ . The present value of  $\lambda$  is smaller than the  $(6.5 \pm 0.5) \times 10^{-10}$  eV cm reported in previous work,<sup>7,10,11</sup> but comparable to  $\lambda = 2.45 \times 10^{-10}$  eV cm reported in previous calculations,<sup>49</sup> to  $\lambda = 2.7 \times 10^{-10}$  eV cm reported by Zorkani, Kartheuser, and Rodriguez,<sup>42</sup> and to  $\lambda = 1.6 \times 10^{-10}$  eV cm obtained from spin-flip Raman scattering in CdS by Romestain, Geschwind, and Delvin.<sup>50</sup>

Finally, Fig. 3 shows the angular dependence of the resonant integrated absorption coefficient in  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$  when the crystal is rotated about an axis  $\hat{c}$  perpendicular to the plane formed by the  $\hat{c}$  axis and the field direction  $\mathbf{B}_0$  fixed.

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#### APPENDIX

We derive the solution of the electric-dipole operator defined in Eq. (2.17) in the limiting case  $\gamma \ll 1$ . Here, for the

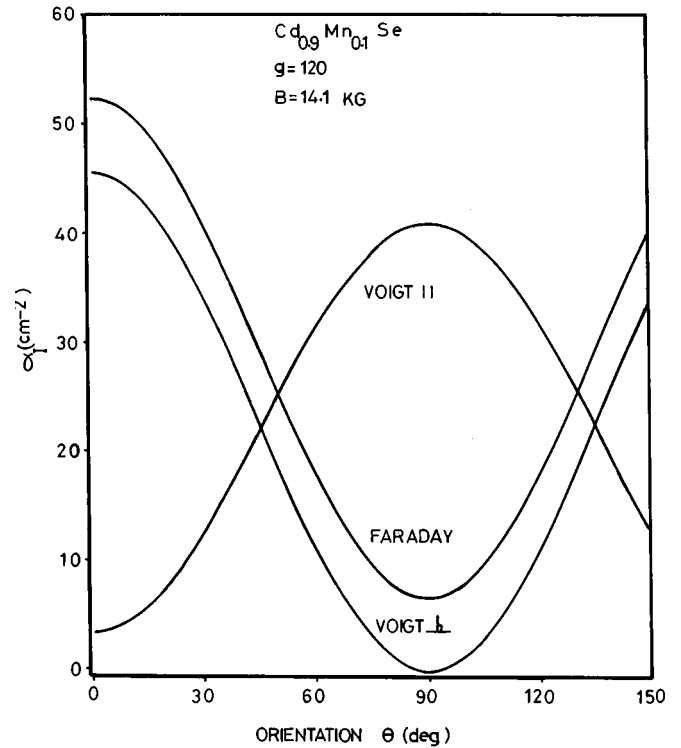


FIG. 3. The angular dependence of the integrated absorption coefficient of a donor-bound electron in  $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$  as a function of orientation of  $\mathbf{B}_0$ , when the crystal is rotated about an axis  $\hat{c}$  perpendicular to the plane formed by the  $\hat{c}$  axis and the field direction  $\mathbf{B}_0$  fixed. For a different field geometry and at a fixed excitation energy  $\hbar\omega = g\mu_B B_0$ , which  $B_0 = 1.41$  T and  $g = 120$ . [Faraday, parallel Voigt (VOIGT ||), and perpendicular Voigt (VOIGT  $\perp$ )].

envelope function of the donor ground state, we choose the trial wave function

$$\langle \mathbf{r} | 0 \rangle = \frac{1}{\sqrt{\pi a_t^* a_l^*}} \exp\left(-\left(\frac{\xi^2 + \eta^2}{a_t^*} + \frac{\zeta^2}{a_l^*}\right)^{1/2}\right). \quad (\text{A1})$$

Requiring the form of the electric-dipole operator given by Eqs. (2.18), and using the orthogonality relations of the spherical harmonics we find from Eqs. (2.17) the following differential equation (in unit  $R = r/a^*$ ):

$$\frac{d^2 D_{\pm}^{m_l}}{dR^2} + 2\left(\frac{1}{R} - a_{m_l}\right) \frac{dD_{\pm}^{m_l}}{dR} - \frac{2}{R^2} D_{\pm}^{m_l} - \Gamma_{\pm}^{m_l} D_{\pm}^{m_l} + \frac{b_{m_l}}{R} D_{\pm}^{m_l} = R. \quad (\text{A2})$$

For  $m_l = 0$ ,

$$a_0 = \frac{1}{a_t} \left( \frac{9}{10} + \frac{1}{10} \varepsilon^2 \right), \quad (\text{A3a})$$

$$b_0 = a_0 \gamma_0, \quad (\text{A3b})$$

with

$$\gamma_0 = 8 \frac{1 - \varepsilon^2}{9 + \varepsilon^2}. \quad (\text{A3c})$$

And, for  $m_l = \pm 1$ ,



$$a_{\pm 1} = \frac{1}{a_t} \left( \frac{7}{10} + \frac{3}{10} \varepsilon^2 \right), \quad (\text{A4a})$$

$$b_{\pm 1} = a_{\pm 1} \gamma_{\pm 1}, \quad (\text{A4b})$$

with

$$\gamma_{\pm 1} = 4 \frac{1 - \varepsilon^2}{9 + \varepsilon^2}. \quad (\text{A4c})$$

We search for nonsingular solutions of Eq. (A2) of a power of series in  $\Gamma_{\pm}^{m_l}$ ,

$$D_{\pm}^{m_l} = \sum_{l=0}^{\infty} Q_l^{m_l}(R) \Gamma_{\pm}^l, \quad (\text{A5})$$

with a polynomial solution of  $Q_l^{m_l}(R)$ . Substituting  $D_{\pm}^{m_l}$  into Eq. (A2), and equating equal powers of  $\Gamma_{\pm}^{m_l} = \Gamma$ , we find

$$\frac{d^2 Q_0^{m_l}}{dR^2} + 2 \left( \frac{1}{R} - a_{m_l} \right) \frac{dQ_0^{m_l}}{dR} - \frac{2}{R^2} Q_0^{m_l} + \frac{b_{m_l}}{R} Q_0^{m_l} = R \quad (\text{A6})$$

and

$$\frac{d^2 Q_l^{m_l}}{dR^2} + 2 \left( \frac{1}{R} - a_{m_l} \right) \frac{dQ_l^{m_l}}{dR} - \frac{2}{R^2} Q_l^{m_l} + \frac{b_{m_l}}{R} Q_l^{m_l} - Q_{l-1}^{m_l} = 0 \quad (\text{A7})$$

for  $l \geq 1$ .  $Q_0(R)$  is the polynomial solution of Eq. (A6) corresponding to the limit  $\Gamma_{\pm}^{m_l} = 0$ . Since  $Q_0(R)$  is of degree 2,  $Q_l^{m_l}(R)$  must be of degree  $l+2$ :

$$Q_l^{m_l}(R) = \sum_{k=0}^{l+2} c_{k,l}^{m_l} a_{m_l}^{-(2l+\gamma_{m_l}+3-k)} R^k. \quad (\text{A8})$$

Thus Eq. (A6) implies

$$c_{1,0}^{m_l} = \frac{4a_{m_l}^{\gamma_{m_l}}}{(2-\gamma_{m_l})(\gamma_{m_l}-4)}, \quad (\text{A9a})$$

$$c_{2,0}^{m_l} = \frac{a_{m_l}^{\gamma_{m_l}}}{(\gamma_{m_l}-4)}, \quad (\text{A9b})$$

$$c_{k,0}^{m_l} = 0 \quad \text{for } k > 2. \quad (\text{A9c})$$

Substitution of Eq. (A8) into Eq. (A7) leads to a polynomial solution for  $Q_l^{m_l}(R)$ , provided the coefficients  $c_{k,l}$  satisfy additional conditions.

For  $l \geq 1$ ,

$$c_{l+2,l}^{m_l} = - \frac{1}{2(l+2) + \gamma_{m_l}} c_{l+1,l-1}^{m_l}. \quad (\text{A10})$$

For  $k \geq 2$  and  $l \geq 1$ ,

$$c_{k-1,l}^{m_l} = \frac{(k+2)(k-1)c_{k,l}^{m_l} - c_{k-2,l-1}^{m_l}}{2(k-1) + \gamma_{m_l}}, \quad (\text{A11})$$

with  $c_{0,l}^{m_l} = 0$  for all values of  $l$ , and  $c_{k,l}^{m_l} = 0$  for  $k > l+2$ .

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