Spin transitions of a bound electron in a semimagnetic semiconductor

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An analysis is made of the absorption of light due to spin flip of an electron bound to a shallow donor in a semimagnetic semiconductor. It is shown that the exchange interaction of a carrier with a magnetic impurity gives rise not only to a giant spin splitting of the electron levels in an external magnetic field, but also to additional (apart from the usual spin–orbit) electric dipole transitions that alter the spin states of the carrier and magnetic impurity. The intensity of magnetic dipole transitions is low. Calculations are made of the energy positions and profiles of the absorption lines when the latter are governed by magnetic impurity fluctuations.

1. INTRODUCTION

Optical properties of semimagnetic semiconductors, representing solid solutions containing up to 10 % of the magnetic impurity ions in the paramagnetic phase, are continuing to attract much interest. The characteristic features of the behavior of carriers in such materials are governed largely by their exchange interaction with spins of this impurity.[1-4] In particular, this interaction is responsible for the "giant" spin splitting $\hbar\omega_s$ of the levels of a carrier in a magnetic field (characterized by a large effective g factor of ~ 100 in the linear magnetization region). A consequence of the giant splitting is that spin transitions in moderate magnetic fields $\mathscr{H} \sim 1$–10 T lie within the infrared range; such transitions have been observed on several occasions [5-7]

We shall show that the strongest spin transitions are of the electric dipole type. At first sight one might expect the large value of the effective g factor of a carrier in a semimagnetic semiconductor to ensure that the intensity of the magnetic dipole transitions (paramagnetic resonance) is much higher than in an ordinary semiconductor. However, it is found that the change in the ESR intensity is slight because the g factor governing this intensity is small, in contrast to the effective g factor which occurs in the transition energy.

A theory of the electric-dipole spin (combined) resonance (EDSR) was first tackled by É.I. Rashba; it was shown that the resonance may be due to the spin–orbit interaction in the Hamiltonian used in the effective-mass method,[8] to the nonparabolicity of the energy band spectrum,[9] and to inhomogeneities that lower the symmetry of a crystal.[10,11] The optical spin transitions classified as the EDSR and observed in a semimagnetic semiconductor have always been interpreted[12,13] on the basis of a traditional model of the spin–orbit interaction.[14]

In an earlier paper[15] we drew attention to the fact that in the case of a semimagnetic semiconductor there may be also a different EDSR mechanism, which is the exchange interaction that not only can account for the giant spin splitting but may induce on a par with the traditional spin–orbit mechanism specific spin transitions (spin flip of a carrier may be accompanied by a change in the spin of a paramagnetic impurity, known as the flip-flop process).

In the present paper we shall give the results of calculations carried out for spin transitions of a carrier bound to a shallow center in a wide-gap semiconductor. This case corresponds to narrow isolated EDSR lines and it has been investigated experimentally on several occasions.[4,5]

Since the magnetic-impurity concentration n in a semimagnetic solid solution is high, the wave function of a carrier at a shallow center extends over a large number of paramagnetic ions. Therefore, we encounter a small parameter $(na^3)^{-1/2}$, where a is the characteristic size of the wave function of a carrier (in the case of a hydrogen-like center this is the effective Bohr radius). The existence of such a small parameter results, firstly, in the validity of the description of the spin-independent interaction of a carrier with a magnetic impurity by band parameters representing a virtual crystal with the same composition as the real crystal. Secondly, the spin splitting energy $\hbar\omega_s$ becomes large—even in weak magnetic fields—compared with its fluctuation associated with the different numbers, positions, and spin states of the magnetic impurity in the vicinity of different centers. The absorption lines are narrow and, moreover, we can ignore the magnetic polaron effect, i.e., the change in the spectrum of impurities due to a carrier. Finally, the existence of such a parameter makes it possible to consider on the basis of perturbation theory the off-diagonal (in respect of the spin states of a carrier) part of the exchange interaction, which is responsible for the spin flip transitions.

This new exchange mechanism allows the absorption of light due to transitions of a carrier accompanied by spin flip and occurring between any levels of an impurity center, i.e., it provides a richer spectrum compared with the traditional spin–orbit EDSR. In the case of a transition between the spin sublevels of the lowest donor level this mechanism results in an additional absorption peak, which is shifted relative to the spin–orbit EDSR line by an amount equal to the Zeeman energy of the magnetic impurity. Calculations carried out for the exchange in spin–orbit EDSR mechanisms show that their intensities are comparable even for hexagonal crystals in which the spin–orbit EDSR mechanism is strongest; the absorption lines corresponding to each of these mechanisms may be identified because of the different angular and polarization dependences. In the case of cubic semimagnetic semiconductors the exchange mechanism should be the principal reason for the spin absorption of light.

We shall allow for fluctuations in the system of magnetic impurities, which broaden the absorption lines. The calculated line profile is nearly Gaussian; non-Gaussian corrections are different for different spin absorption mechanisms.

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2. GENERAL EXPRESSION FOR THE ABSORPTION COEFFICIENT

The Hamiltonian of an electron in a semimagnetic semiconductor is of the form

$$H=H_e+\hbar\omega_e s_z+\hbar\omega_M\sum_j S_{jz}+H_{ex}+H_{so},$$
$$\hbar\omega_e=g_e\beta_0\mathscr{H},\quad \hbar\omega_M=g_M\beta_0\mathscr{H}. \tag{1}$$

The first two terms in the first of the above expressions represent the Hamiltonian of an electron with a spin in a constant magnetic field $\vec{\mathscr{H}}\|\mathbf{z}$; the third term is the Zeeman Hamiltonian of the magnetic impurity; g_M is the g factor of the magnetic impurity and g_e is the g factor of an electron, which is not yet renormalized by the exchange interaction; β_0 is the Bohr magneton. The operator representing the exchange interaction of an electron with the impurities is[16,17]

$$H_{ex}=-J\sum_j \mathbf{s}\mathbf{S}_j\delta(\mathbf{r}-\mathbf{R}_j), \tag{2}$$

where $\mathbf{s}$ and $\mathbf{S}_j$ are the spin operators of an electron and of a paramagnetic impurity located at a point $\mathbf{R}_j$; J is the exchange constant. In the standard case when the magnetic impurity is in the form of Mn^{2+} ions, which in the ground state have a half-filled d shell with the orbital momentum $L_M=0$, the spin moment is $S=5/2$. The operator H_{so} represents the spin–orbit interaction and its form depends, in contrast to H_{ex} on the symmetry of a crystal.

The absorption coefficient of an electromagnetic wave with a wave vector $\mathbf{q}$ and of frequency ω obtained in the dipole approximation is

$$\alpha(\omega)=\frac{4\pi e^2\omega n_e}{c\varkappa^{1/2}}\operatorname{Re}\int_0^\infty dt\, e^{-i\hbar\omega t}\langle\operatorname{Tr}\{[\rho,F^+]e^{iHt}Fe^{-iht}\}\rangle, \tag{3}$$

where

$$F=\mathbf{er}+\mathbf{hm},$$
$$\mathbf{m}=\frac{\varkappa^{1/2}}{e}\beta_0\Big(g_e\mathbf{s}+g_M\sum_j\mathbf{S}_j\Big),\quad \mathbf{h}=[\mathbf{qe}]/|\mathbf{q}|, \tag{4}$$

$\varkappa$ is the static permittivity; n_e is the density of the donor electrons; $\mathbf{e}$ is the polarization vector of the electromagnetic wave; ρ is the equilibrium density matrix. The first term in F represents the interaction of a carrier with an electric field and the second—the interaction of a carrier and of paramagnetic impurities with the magnetic field of the electromagnetic wave. The term corresponding to the electric dipole interaction of light with the magnetic impurity ions is omitted because the excited states of the manganese ion with $L_M=1$ are separated from the ground state by much larger energies than the photon energies under discussion. The angular brackets in Eq. (3) denote averaging over the random positions of the magnetic impurity:

$$\langle A(\{\mathbf{R}\})\rangle=\int A(\{\mathbf{R}\})\prod_j\frac{d\mathbf{R}_j}{V}. \tag{5}$$

In the case of transitions inside the local center in a semimagnetic semiconductor the most important case is that when the ionization energy of a center and $\hbar\omega_s$ are large compared with the absolute temperature T. Then, only the lowest spin level of the center is filled and averaging with the density matrix can be carried out only over this specific electron state. This corresponds to the case when the commutator $[\rho,F^+]$ in Eq. (3) reduces simply to ρF^+, while the averaging over the magnetic impurity states is retained because generally $\hbar\omega_M$ is comparable with T.

The transitions accompanied by a change in the spin state of an electron may be initiated not only by the second term in F (representing ESR), but also by the first term, owing to the off-diagonal (in respect of the spin states of the carrier) terms in Eq. (1), namely H_{so} and the off-diagonal part of H_{ex} which occur in the Dirac brackets [F in Eq. (3)]. We shall follow the usual procedure,[12,14,18] and allow for H_{so} using perturbation theory. The diagonal part of H_{ex}

$$H_d=-J\sum_{j,\nu}s_zS_{jz}|\nu\rangle\langle\nu|\delta(\mathbf{r}-\mathbf{R}_j)|\nu\rangle\langle\nu| \tag{6}$$

($H_e|\nu\rangle=E_\nu|\nu\rangle$) is known to result in a giant spin splitting and is not small. The off-diagonal part of H_{ex}, which is usually ignored, contains an operator

$$H_{nd}=-J\sum_j(s_1S_{j\bar{1}}+s_{\bar{1}}S_{j1})\delta(\mathbf{r}-\mathbf{R}_j), \tag{7}$$

where

$$s_1=s_{\bar{1}}^{+}=(s_x+is_y)/\sqrt{2}$$

(and this applies also to $\mathbf{S}_j$), which mixes the states with different spin projections. This operator may give rise to spin transitions and in most cases, if $na^3\gg1$, it can be simply included in the lowest order of perturbation theory (see the Appendix A). This can be done conveniently by performing, in the trace of Eq. (3), a unitary transformation $\exp(W)H\cdot\exp(-W)$ (Ref. 18), which annihilates H_{nd} and H_{so} in the Hamiltonian of Eq. (1):

$$e^WHe^{-W}=e^W(H_0+H_{nd}+H_{so})e^{-W}=H_0,$$
$$H_0=H_e+H_d+\hbar\omega_e s_z+\hbar\omega_M\sum_j S_{jz}. \tag{8}$$

The operator W, which satisfies this equation in the first order in respect of the perturbation, is

$$W=-i\int_0^\infty e^{iH_0\tau}(H_{nd}+H_{so})e^{-iH_0\tau}d\tau\equiv W_{ex}+W_{so}, \tag{9}$$

so that in place of H and F Eq. (3) contains now H_0 and

$$F'=e^WFe^{-W}\approx F+[W,F]. \tag{10}$$

3. ELECTRIC-DIPOLE SPIN TRANSITIONS

Such transitions are initiated by the terms $[W,\mathbf{e}\cdot\mathbf{r}]$ of the operator F' in Eq. (10). The operators $F_{ex}=[W_{ex},\mathbf{e}\cdot\mathbf{r}]$ differ from $F_{so}=[W_{so},\mathbf{e}\cdot\mathbf{r}]$ because simultaneously with reversal of the electron spin they also change the spin of the magnetic impurity, so that the term with the correlation function of F_{ex} and F_{so} in Eq. (3) is off-diagonal in respect of the impurity spins and is annihilated in the trace, and the transitions do not interfere. As a result, we have

$$\alpha_{\mathrm{EDSR}}(\omega)=\alpha_{ex}(\omega)+\alpha_{so}(\omega). \tag{11}$$

a. Exchange mechanism of electric dipole spin resonance

The expression for $\alpha_{\rm ex}(\omega)$ includes the operator

$$F_{ex}(t)=e^{iH_0t}F_{ex}e^{-iH_0t}, \tag{12}$$

the matrix elements of which for the electron functions

$$|\nu\xi\rangle=|\nu\rangle|\xi\rangle \quad (s_z|\xi\rangle={}^1/_2\xi|\xi\rangle, \quad \xi=\pm1, \ \bar\xi=-\xi)$$

yield

$$F_{ex}{}^{\xi}(t)_{\nu\nu'}\equiv\langle\nu\bar\xi|F_{ex}(t)|\nu'\xi\rangle=i\frac{J}{\sqrt2}\sum_{j,\nu''}S_{j\xi}\exp(it\mathscr{E}^{\xi}_{\nu\nu'}) \\ \times\int_0^\infty\{\exp(i\tau\mathscr{E}^{\xi}_{\nu\nu''})\langle\nu|\delta(\mathbf{r}-\mathbf{R}_j)|\nu''\rangle\langle\nu''|e\mathbf{r}|\nu'\rangle \\ -\exp(i\tau\mathscr{E}^{\bar\xi}_{\nu''\nu'})\langle\nu|e\mathbf{r}|\nu''\rangle\langle\nu''|\delta(\mathbf{r}-\mathbf{R}_j)|\nu'\rangle\}d\tau, \tag{13}$$

where

$$\mathscr{E}^{\xi}_{\nu\nu'}=E_\nu-E_{\nu'}-\xi[{}^1/_2(\hbar\Omega_\nu+\hbar\Omega_{\nu'})-\hbar\omega_M]+{}^1/_2J|\psi_\nu(\mathbf{R}_j)|^2, \tag{14}$$

$$\hbar\Omega_\nu=\hbar\omega_e-J\sum_{j'}S_{j'z}|\psi_\nu(\mathbf{R}_{j'})|^2. \tag{15}$$

The quantity $\hbar\Omega_\nu$ represents the energy of the spin splitting of the νth level of a carrier obtained for fixed positions and spin states of the paramagnetic impurity. The absolute value of the average spin splitting is

$$(\hbar\Omega\nu)_{\rm av}\equiv\langle\overline{\hbar\Omega_\nu}\rangle=\langle{\rm Tr}_M(\hbar\Omega_\nu\rho_M)\rangle, \tag{16}$$

$$\rho_M=\exp\Bigl(-\lambda\sum_j S_{jz}\Bigr)\Big/Z_M \tag{17}$$

and it is independent of ν and equal to the spin splitting energy

$$\hbar\omega_s=|\hbar\omega_e+JnSB_S(\lambda S)|, \tag{18}$$

which is obtained for semimagnetic semiconductors in Ref. 4; here, B_S is the Brillouin function and $\lambda=\hbar\omega_M/T$. The magnetic polaron contribution

$${}^1/_2J|\psi_\nu(\mathbf{R}_j)|^2\lesssim J/a^3,$$

which is small for weak magnetic fields far from the limit, when $\hbar\omega_M\gg J/a^3$, is ignored in Eq. (17). This does not impose any additional restrictions (this point is discussed at the end of the present section and also in the Appendix B).

The absorption coefficient $\alpha_{\rm ex}(\omega)$ is described by

$$G_{ex}(t)=e^{-i\hbar\omega t}\sum_\nu(F_{ex}{}^{+\sigma}(0)_{0\nu}F_{ex}{}^{\bar\sigma}(t)_{\nu0})_{\rm av}\,. \tag{19}$$

Here, $\sigma=\text{sign}\,[(\hbar\Omega_0)_{\rm av}]$ depends on the direction of the electron spin in the initial state and is the same as the sign of the effective g factor. The multiplier that has to be averaged in Eq. (19) has the following structure:

$$\mathscr{G}=\sum_j\Bigl\{f(\mathbf{R}_j)\exp[i\tilde A(\mathbf{R}_j)S_{jz}]S_{j\sigma}S_{j\bar\sigma}\prod_{j'\neq j}\exp[iA(\mathbf{R}_{j'})S_{j'z}]\Bigr\}_{\rm av} \tag{20}$$

(a single sum over j appears because $S_{j_1\sigma}S_{j_2\bar\sigma}$ is off-diagonal if $j_1\neq j_2$). Here, $A(\mathbf{R})$ [and $\tilde A(\mathbf{R})$] is given by $A(\mathbf{R})=\varepsilon t+\varepsilon_1\tau_1+\varepsilon_2\tau_2$, where in all cases we have $\varepsilon\propto J|\psi_\nu(\mathbf{R})|^2\propto J/a^3$ [two auxiliary times τ originate from the presence of two F's in Eq. (19)]. The average value of $\mathscr{G}$ can be found in the Appendix B [Eq. (B10)]. However, in our case when $na^3\gg1$ the multiplier $\exp[i\tilde A(\mathbf{R}_j)S_{jz}]$ can be ignored against the background of na^3 actual similar multipliers. We then have

$$\mathscr{G}=n\overline{S_\sigma S_{\bar\sigma}}\int f(\mathbf{R})\,d\mathbf{R}\exp\Bigl\{n\int\Bigl[\frac{Z(\lambda-iA(\mathbf{R}'))}{Z(\lambda)}-1\Bigr]d\mathbf{R}'\Bigr\}, \tag{21}$$

where

$$\overline{S_\sigma S_{\bar\sigma}}={}^1/_2[S(S+1)-\overline{S_z{}^2}+\sigma\bar S_z], \tag{22}$$

$$\overline{S_z{}^p}=\frac{1}{Z}\Bigl(-\frac{\partial}{\partial\lambda}\Bigr)^pZ, \tag{23}$$

are identical for all the impurities and the index j is omitted, and

$$Z(\lambda)={\rm Tr}e^{-\lambda S_z}=\frac{{\rm sh}[(S+{}^1/_2)\lambda]}{{\rm sh}(\lambda/2)}. \tag{24}$$

In the calculation of the integrals with respect to τ_1 and τ_2 from Eq. (13) in the expression (19) averaged in this way, we expand $Z[\lambda-iA(\mathbf{R}')]$ as a series in A. After integration in the argument of the exponential function containing $\mathbf{R}'$, we obtain a coefficient of the order of $\hbar\omega_s\propto|J|n$ in the first order in front of t, τ_1 and τ_2; the quadratic terms determine the characteristic times $1/\gamma$ involved in the procedure of integration $[\gamma\propto Jn(na^3)^{-1/2}]$; the higher terms of the expansion decrease in accordance with the parameter $(na^3)^{-1/2}$ [in particular, the third term is of the order of the argument of the exponential function ignored in the derivation of Eq. (21)], so that they can be ignored. Since the complete [obtained allowing for the exponential functions in Eq. (13) which do not need averaging] coefficient in front of $\tau_{1,2}$ taken in the first degree is large compared with γ (see below), the integrals are calculated asymptotically[19] and they have large energy denominators. As a result, we obtain

$$G_{ex}(t)=\sum_\nu\exp\Bigl[i(E_\nu-E_0+\hbar\omega_s{}'-\hbar\omega)t \\ -\frac{\gamma_\nu{}^2t^2}{2}\Bigr]\overline{S_\sigma S_{\bar\sigma}}n\int|M_{ex}{}^\nu(\mathbf{R})|^2d\mathbf{R}, \tag{25}$$

$$M_{ex}{}^\nu(\mathbf{R})=\frac{J}{\sqrt2}\sum_{\nu'}\Bigl\{\frac{\langle\nu|e\mathbf{r}|\nu'\rangle\langle\nu'|\delta(\mathbf{r}-\mathbf{R})|0\rangle}{E_{\nu'}-E_0+\hbar\omega_s{}'} \\ +\frac{\langle\nu|\delta(\mathbf{r}-\mathbf{R})|\nu'\rangle\langle\nu'|e\mathbf{r}|0\rangle}{E_{\nu'}-E_\nu-\hbar\omega_s{}'}\Bigr\}, \tag{26}$$

$$\hbar\omega_s{}'=\hbar\omega_s-\sigma\hbar\omega_M, \tag{27}$$

$$\gamma_\nu\equiv\gamma_{\nu0}, \qquad \gamma^2_{\nu\nu'}=\frac{nJ^2}{4}\overline{S_z{}^2}\int[|\psi_\nu(\mathbf{R})|^2+|\psi_{\nu'}(\mathbf{R})|^2]^2d\mathbf{R}. \tag{28}$$

This averaging algorithm corresponds to neglect of fluctuations in the energy denominators $M_{\rm ex}$ against the background of the average values, which is justified if

$$|E_0-E_{\nu'}-\hbar\omega_s{}'|\gg\gamma_{\nu'0}, \ |E_\nu-E_{\nu'}+\hbar\omega_s{}'|\gg\gamma_{\nu\nu'}.$$

However, this is not a new restriction and it is in agreement with the condition of validity of perturbation theory (Appendix A). Allowance for fluctuations of the denominators, the values of which are of the order of the perturbation-theory corrections to the energies, represents redundant precision, because Eq. (10) is simplified by dropping higher terms of the same order of magnitude.

It is clear from Eq. (25) that the exchange EDSR mechanism allows absorption accompanied by spin flip as a result of transitions both between the spin sublevels of the lowest donor level and at combination frequencies with any change in the electron state. However, in view of the finite width of the absorption lines and the denser distribution of the donor levels, these transitions are masked strongly by the usual optical absorption. Therefore, the transition at a pure spin frequency between the spin sublevels of the ground state of a carrier is manifested most strongly. It corresponds to the term with $\nu = 0$ in Eq. (25), and to

$$\alpha_{ex}(\omega)=\frac{4\pi^2e^2\omega n_e}{c\varkappa^{1/2}}\overline{S_\sigma S_{\bar\sigma}}n\int|M_{ex}{}^0(\mathbf{R})|^2\,d\mathbf{R}\,\Delta(\hbar\omega-\hbar\omega_s{}'),\tag{29}$$

$$\Delta(\hbar\omega-\hbar\omega_s{}')=\frac{1}{(2\pi)^{1/2}\gamma_0}\exp\left\{-\frac{(\hbar\omega-\hbar\omega_s{}')^2}{2\gamma_0{}^2}\right\}.\tag{30}$$

We can see that in a semimagnetic semiconductor in the case of the exchange EDSR mechanism the transition energy is $\hbar\omega_s'$ of Eq. (27), which differs slightly from $\hbar\omega_s$. It should be pointed out that it is comparable with the separation between the levels of a Coulomb center in such magnetic fields that the cyclotron energy $\hbar\omega_c$ (much smaller than $\hbar\omega_s$) is low compared with the characteristic energy $R' = m^*e^4/\hbar^2\varkappa^2$ (double the Rydberg energy). Therefore, in view of $(\hbar\omega_c/R^*)^2 \ll 1$, we can ignore the term H_e which is quadratic in $\mathscr{H}$; as a result, we find that

$$H_e=-\frac{\hbar^2}{2m^*}\Delta-\frac{e^2}{\varkappa r}+\frac{\hbar\omega_c}{2}L_z,\tag{31}$$

where L_z is the projection of the orbital momentum. This approximation is standard for semimagnetic semiconductors,[12] although the results can be obtained without this approximation (Appendix D). When the Hamiltonian of Eq. (31) is used, the absorption line width becomes

$$\gamma_0=|J|n(\overline{S_z{}^2}/8\pi na^3)^{1/2},\tag{32}$$

where $a = \hbar^2\varkappa/m^*e^2$ is the effective Bohr radius and the task of calculation of M_{ex} reduces to a calculation of the sums for the hydrogen states, whereas the matrix elements of the transitions are expressed in terms of integrals with the Coulomb Green function. An alternative approach, more convenient in this task, is given in the Appendix C.

The scalar product $\mathbf{e}\cdot\mathbf{r}$ in Eq. (26) can be written conveniently in the form

$$\mathbf{er}=\sum_\mu e_\mu r_{\bar\mu},\qquad \mu=\bar1,0,1.\tag{33}$$

Here, the polarization $\mathbf{e} = \{e_x, e_y, e_z\}$ (and also $\mathbf{r}$) are written in the circular basis $\mathbf{e} = (e_{\bar1}, e_0, e_1)$, so that in the case of the Voigt polarization we have

$$\mathbf{e}=\{0,0,1\}=(0,1,0),$$

whereas for the cyclotron-active Faraday polarization, we obtain

$$\mathbf{e}=\{1,i,0\}/\sqrt{2}=(1,0,0),$$

and in the case of the cyclotron-passive polarization, we find that

$$\mathbf{e}=\{1,-i,0\}/\sqrt{2}=(0,0,1).$$

The matrix element of Eq. (26) thus splits into three terms. In each of them the denominators differ, because of L_z in Eq. (31), by an amount $\mu\hbar\omega_c/2$ which is small compared with $\hbar\omega_s'$. We can see from Eqs. (29) and (26) that the EDSR intensity rises strongly when $\hbar\omega_s'$ approaches $E_{np} - E_{1s}$ [naturally, subject to the condition of Eq. (A3)], so that in the most interesting case when $\hbar\omega_s' < E_{2p} - E_{1s}$ and the EDSR peak is clearly separate, it is convenient to represent the results of a calculation by writing down explicitly the resonance denominator, i.e.,

$$n\int|M_{ex}{}^0(\mathbf{R})|^2\,d\mathbf{R}=\frac{a^2}{na^3}\left(\frac{Jn}{R^*}\right)^2\sum_\mu\frac{\Phi_{ex}(\varepsilon_\mu{}')}{({}^3/_8-\varepsilon_\mu{}')^2}|e_\mu|^2,\tag{34}$$

where $\varepsilon_\mu' (\hbar\omega_s' + \frac{1}{2}\mu\hbar\omega_c)/R^*$ and the smooth function $\Phi_{ex}(\varepsilon)$ of Eq. (C15) is shown in Fig. 1. At low frequencies all the quantities ε are small and $\Phi_{ex}(0) = 783/2^{15}\pi$, whereas in the parametric-resonance region we have $\varepsilon \to 3/8$ and $\Phi_{ex}(3/8) = (2/3)^{14}/3\pi$. It is interesting to note that $\alpha_{ex}(\omega)$ for an arbitrary polarization of light represents a sum of the partial absorption coefficients for each of the circular components e_μ. This absence of interference is due to the fact that the cross terms with $e_\mu e_{\mu'}{}^*$ $(\mu \neq \mu')$ are annihilated as a result of averaging over the positions of the impurities—as discussed immediately after Eq. (C14).

At high temperatures $T \gg \hbar\omega_M$ these expressions simplify to

$$\hbar\omega_s=|\tilde g|\beta_0\mathscr{H},\tag{35}$$

where the effective g factor is[4]

$$\tilde g=g_e+g_M JnS(S+1)/3T,\tag{36}$$

$$\overline{S_\sigma S_{\bar\sigma}}={}^1/_3S(S+1),\tag{37}$$

and the width

$$\gamma_0=|J|n[S(S+1)/24\pi na^3]^{1/2}\tag{38}$$

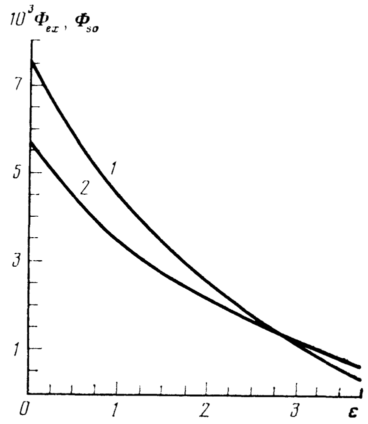

FIG. 1. Functions $\Phi_{ex}(\varepsilon)$ (curve 1) and $\Phi_{so}(\varepsilon)$ (curve 2).

is identical for $S = 5/2$ with the width calculated by a different method in Ref. 17.

b. Spin–orbit mechanism of electric dipole spin resonance

The influence of the spin–orbit interaction is strongest in hexagonal crystals, because in this case $H_{\rm so}$ is a linear function of the quasimomentum **k**, whereas in cubic crystals, for which $H_{\rm so} \propto k^3$ or k^4, the contribution of this interaction is less.[8,9,18,20] We shall therefore consider only the case where

$$H_{so}=2\alpha_0[\mathbf{sk}]\mathbf{c}, \tag{39}$$

which is typical of hexagonal crystals (α_0 is the spin–orbit interaction constant and **c** is a unit vector along the hexagonal axis). The absorption coefficient is described by relationships similar to Eqs. (19) and (12) provided we replace the subscript ex with so. Calculations similar to those in the preceding case yield

$$G_{so}(t)=\sum_{\nu}\exp[i(E_\nu-E_0+\sigma\hbar\omega_e-\hbar\omega)t]\,|M_{so}{}^{\nu}|^2D_\nu(t), \tag{40}$$

$$D_\nu(t)=\exp\Big\{n\int d\mathbf{R}\times\Big[\frac{Z(\lambda+{}^1/{}_2 i\sigma J(|\psi_0(\mathbf{R})|^2+|\psi_\nu(\mathbf{R})|^2)t)}{Z(\lambda)}-1\Big]\Big\}, \tag{41}$$

$$M_{so}{}^{\nu}=2^{1/2}\alpha_0\sum_{\nu'}\Big\{\frac{\langle\nu|e\mathbf{r}|\nu'\rangle\langle\nu'|[\mathbf{kc}]_{\bar\sigma}|0\rangle}{E_{\nu'}-E_0+\hbar\omega_s}+\frac{\langle\nu|[\mathbf{kc}]_{\bar\sigma}|\nu'\rangle\langle\nu'|e\mathbf{r}|0\rangle}{E_{\nu'}-E_\nu-\hbar\omega_s}\Big\}. \tag{42}$$

The matrix element $M^0_{\rm so}$ (pure spin transition) for ordinary nonmagnetic semiconductors had been found earlier.[14] The specific nature of a similar magnetic semiconductor is manifested by the need to average over fluctuations of the spin frequency. Averaging is applied here to a quantity of the $\exp\{i\Sigma_j A(\mathbf{R}_j)S_{jz}\}$ type and the procedure is now simpler than in the case of Eq. (20). Using Appendix B and carrying out an asymptotic calculation of the integrals with respect to τ, similar to that in the preceding case, we obtain Eq. (40). If, as before, we limit the expansion of the argument of the exponential function $D_\nu(t)$ to the term quadratic and t, then $G_{\rm so}(t)$ exhibits the same time dependence as in Eq. (25) provided we replace $\hbar\omega_s'$ with $\hbar\omega_s$. The absorption lines for the exchange and spin–orbit EDSR mechanisms are in this approximation Gaussian with the same value of γ_ν [Eq. (28)]. It should be pointed out that small [in terms of the parameter $(na^3)^{-1/2}$] non-Gaussian corrections to the line profiles are different for the two mechanisms: in the case of the spin–orbit mechanism they are governed only by the next terms of the expansion of the argument of the exponential function in Eq. (41), whereas in the case of the exchange mechanism they are determined by the factor omitted in the derivation of Eq. (21).

In contrast to the exchange mechanism, the transitions from the ground state of a carrier in the spin–orbit EDSR mechanism are allowed not to any excited states of a local center, but only to states with the same parity (originating from the hydrogen-like *s*- and *d*-levels in the absence of a magnetic field). As in the preceding case, the pure spin transition is most interesting and, moreover, we can retain Eq. (31) for H_e. Using the Appendix C, we can represent the matrix element $M^0_{\rm so}$ in the form

$$M_{so}{}^0=2^{1/2}\sigma\frac{\alpha_0\hbar\omega_s}{R^{*2}}\sum_\mu(-1)^\mu e_\mu c_{\bar\mu+\bar\sigma}[I_{so}(\varepsilon_\mu)+I_{so}(-\varepsilon_\mu)], \tag{43}$$

$$I_{so}(\varepsilon_\mu)=2v^2\frac{1-2v^2}{(v^2-1)^2}+\frac{(2v)^9}{(2-v)(v-1)^2(v+1)^{10}}\times F\Big(4,2-v;3-v;\Big(\frac{v-1}{v+1}\Big)^2\Big), \tag{44}$$

where

$$\varepsilon_\mu=(\hbar\omega_s+{}^1/{}_2\mu\hbar\omega_c)/R^*,\quad v=(1-2\varepsilon_\mu)^{-1/2},$$

F is the hypergeometric function, and $c_{\bar 1}$, c_0, and c_1 are components of a vector **c** in the circular basis, and $c_{\pm 2} = 0$.

In the case of an arbitrary direction of **e** the interference effects complicate greatly the form of $\alpha_{\rm so}$ (Ref. 18), in contrast to Eq. (34). Therefore, we shall give the expression $\alpha_{\rm so}$ in the case when **e** coincides with one of the circular polarizations e_μ. Separating, as in Eq. (34), the resonance denominator, we obtain

$$\alpha_{so}(\omega)=\frac{4\pi^2e^2\omega n_e}{c\varkappa^{1/2}}|M_{so}{}^0|^2\Delta(\hbar\omega-\hbar\omega_s), \tag{45}$$

$$|M_{so}{}^0|^2=\frac{(\alpha_0\hbar\omega_s)^2}{R^{*4}}\frac{\Phi_{so}(\varepsilon_\mu)}{({}^3/{}_8-\varepsilon_\mu)^2}|c_{\mu+\sigma}|^2. \tag{46}$$

The function $\Phi_{\rm so}$ is shown in Fig. (1); here, $\Phi_{\rm so}(0) = 3^6/2^7$, $\Phi_{\rm so}(3/8) = 2^{31}/3^{20}$.

A comparison of the absorption coefficients for the two EDSR mechanisms shows that the energy of these spin transitions differs by $\hbar\omega_M$ and both rise on approach of $\hbar\omega_s$ (or $\hbar\omega_s'$) to $E_{np} - E_{1s}$. The difference in the transition energy is due to the fact that in the exchange EDSR process there is an additional change in the spin of the paramagnetic impurity. In other respects these two mechanisms are very different: the EDSR spectra consist of different numbers of lines; the absorption due to the exchange EDSR mechanism occurs for any polarization of light and the value of $\alpha_{\rm ex}$ is independent of the magnetic field orientation relative to the crystal axes (the spin–orbit EDSR depends on the angle ϑ between **c** and $\vec{\mathscr{H}}$ and is allowed only in two polarizations $e_{\bar\sigma}$ and e_0 with the angular dependences $\cos^2\vartheta$ and $\sin^2\vartheta$, respectively, see Ref. 14). Finally, at low frequencies $\hbar\omega \ll R^*$ we have $\alpha_{\rm ex}(\omega_s') \propto \omega_s'$ ($M_{\rm ex} \to$ const) and also $\alpha_{\rm so}(\omega_s) \propto \omega_s^3$ ($M_{\rm so} \propto \omega_s$). Since, in contrast to an ordinary semiconductor, the EDSR transitions in a semimagnetic semiconductor do not occur between the Kramers levels of the system comprising a carrier and magnetic impurities (the Kramers levels correspond to spin flip not only of the carrier but also of all the magnetic impurities), these results are not surprising: $M_{\rm ex}$ (and if we allow for the magnetic Polaron effect, this applies also to $M_{\rm so}$) do not vanish for $\mathscr{H} = 0$.

We are ignoring the magnetic polaron corrections to the density matrix of Eq. (17), as discussed immediately after Eq. (18). The inclusion of these corrections makes a contribution to $\hbar\omega_s$ which is of the order of γ_0. They are undoubtedly important in weak magnetic fields where $\hbar\omega_s \sim \gamma_0$ and, in particular, they are responsible for the nonzero spin splitting of the carrier levels when $\mathscr{H} = 0$ (Ref. 4), but in our case they can be ignored because $\hbar\omega_s \gg \gamma_0$ [see Eq. (A4)].

Neglect of the diamagnetic term in H_e may be too rough in the case of, for example, narrow-gap semiconductors when because of the small effective mass the value of $\hbar\omega_c$ is large and may be comparable with R^*. In this case we can carry out calculations using the exact H_e on the basis of a variational method[14] which makes it possible to calculate approximately (see the Appendix D) the matrix elements. It is shown in Ref. 14 that the variational method in extremely high magnetic fields gives the correct results for $I_{\rm so}$ [see Eq. (C19). In our case, when $\hbar\omega_c \ll R^*$, the variational calculations give results which are in excellent agreement with the exact values (see the Appendix C). We can consequently expect the variational method to be effective also in the intermediate case and moreover, when complex (many-band, anisotropic, etc.) models of a center are employed.

4. MAGNETIC DIPOLE SPIN TRANSITIONS

These transitions are initiated by the term $\mathbf{h}\cdot\mathbf{m}$ in Eq. (4), which may occur in Eq. (3) for the absorption coefficient in the form of a correlation function between $\mathbf{h}\cdot\mathbf{m}$ and $\mathbf{h}\cdot\mathbf{m}$(ESR), exactly as in the case of the correlation function of $\mathbf{h}\cdot\mathbf{m}$ with $\mathbf{e}\cdot\mathbf{r}$ (interference of magnetic and electric dipole transitions). In particular, the absorption coefficient $\alpha_{\rm ESR}$ is governed by an expression similar to Eq. (19), where instead of $F_{\rm ex}$ we now have

$$F'_{\rm ESR} = \mathbf{hm} + [W_{ex}, \mathbf{hm}]. \qquad (47)$$

The second term in Eq. (47) not only renormalizes the absorption because of the first term in the polarization $h_{\bar\sigma}$, but also has the effect that a magnetic field of an electrodynamic wave initiates spin transitions of a carrier also in other polarizations (at frequencies $\omega_s - \sigma\omega_M$, $\omega_s - 2\sigma\omega_M$), and also between different levels of a donor. In our case, we have $\hbar\omega_s \gg \gamma_0$, and the strongest is the absorption in the polarization $h_{\bar\sigma}$ at a frequency ω_s. The corresponding absorption coefficient is calculated as before, and it is equal to

$$\alpha_{\rm ESR} = 2\pi^2\beta_0{}^2 q n_e (g_e - g_M)^2 (\omega_s/\omega_s')^2 \Delta(\hbar\omega - \hbar\omega_s). \qquad (48)$$

An interesting feature is the proportionality of $\alpha_{\rm ESR}$ not to the square of the effective g factor of Eq. (36), but to the square of the much smaller difference between g_e and g_M. In view of this and in spite of the giant enhancement of the energy of a spin transition in semimagnetic semiconductors, we cannot expect an increase in the ESR intensity compared with ordinary semiconductors and, as in the case of ordinary semiconductors,[18] the ESR intensity is low compared with the EDSR intensity. The contribution of the interference terms is intermediate, although it is much less important (because of the large difference between the EDSR and ESR intensities) than for example in the case of n-type InSb (Ref. 21) for which this interference had been observed experimentally.[20,22]

5. DISCUSSION OF RESULTS

Our conclusion about the ESR intensity is in contrast with the concept put forward in Ref. 12, where phenomenological considerations showed that the interaction of an electron spin in a semimagnetic semiconductor with a magnetic field of an optical wave is expressed in terms of the effective giant g factor $\tilde g$. This gives rise to large values of $\alpha_{\rm ESR}$, proportional to $\tilde g^2$ and comparable with $\alpha_{\rm so}$ in a wide range of $\hbar\omega_s$, with the exception of the paramagnetic resonance region. Our more consistent theory yields a much smaller {ratio $[(g_e - g_M)/\tilde g]^2$} value of $\alpha_{\rm ESR}$ of Eq. (48), which is negligibly small like $\alpha_{\rm so}$.

The experimental results on EDSR in a semimagnetic compound $\rm Cd_{0.9}Mn_{0.1}Se$ are given in Ref. 5. The experimental points and the continuous theoretical curve from Ref. 12 are plotted in Fig. 2 for an absorption peak associated with the spin–orbit mechanism. Allowance for a large value of $\alpha_{\rm ESR}$ in the theoretical curve of Ref. 12 creates a discrepancy between this curve and the experimental points on our results (dashed curve) at low frequencies.

An analysis of the experimental results in Refs. 5 and 12 was made on the assumption of a Lorentzian profile of an absorption peak and gave values of the spin–orbit interaction constant $\alpha_0 = 2.1\times 10^{-3}e^2$ (Ref. 5) and $\alpha_0 = 2.2\cdot 10^{-3}e^2$ (Ref. 12). As shown in the present paper, the peak profile is Gaussian and an analysis gives $\alpha_0 = 1.7\cdot 10^{-3}e^2$ ($2\alpha_0 = 4.9\times 10^{-10}$ eV·cm).[23] We must bear in mind that the experimental points correspond to $\alpha_{\rm so}(\omega_s)\Gamma$, where γ is the width of an absorption peak at midamplitude[5] related to γ_0 of Eq. (32) by the expression $\Gamma = \gamma_0(2\ln 2)^{1/2}/\pi c\hbar$.

We shall now compare the intensities of the spin–orbit and the exchange EDSR lines. This can be done conveniently near a parametric resonance ($\hbar\omega_s \to E_{2p} - E_{1s}$). In this case it follows from Eqs. (29), (34) and (45), (46) that for the maximum value of $\alpha_{\rm so}$, we obtain

$$\frac{\alpha_{ex}(\omega_s')}{\alpha_{so}(\omega_s)} = \frac{27}{2^{11}\pi}\frac{\overline{S_\sigma S_{\bar\sigma}}}{na^3}\left(\frac{Jna}{\alpha_0}\right)^2 = 0.68\bar x\,\overline{S_\sigma S_{\bar\sigma}} \qquad (49)$$

for the Faraday polarization and $\vec{\mathscr{H}}\|\mathbf{c}$; however, in the case of a plane-polarized wave this ratio is twice as large,[15] since the spin–orbit EDSR mechanism results in the absorption of just one ofthe circular Faraday components. We found the

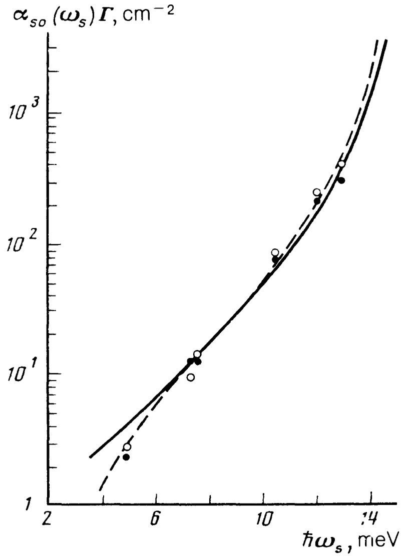

FIG. 2. Dependence of the absorption coefficient in the Faraday polarization for the spin–orbit EDSR in $\rm Cd_{0.9}Mn_{0.1}Se$ on the energy $\hbar\omega_s$: the points are the experimental results obtained for two close temperatures,[5] the continuous curve is calculated,[12] and the dashed curve is also calculated on the basis of our Eqs. (45) and (46).

numerical values employing the parameters of a $Cd_{1-x}Mn_x$ Se crystal: $a = 38$ Å, $JN_0 = 260$ meV, $N_0 = 1.83 \times 10^{22}$ cm^{-3} is the concentration of the cation sites, and the magnetic impurity concentration is written in the form $n = N_0 \bar{x}$ ($\bar{x}$ is the effective relative atomic fraction of the noninteracting magnetic ions),[17] and our value of α_0. Moreover, at these frequencies the exchange EDSR mechanism is approximately an order of magnitude weaker than the spin-orbit contribution. However, the farther away from a parametric resonance in the direction of lower frequencies the more rapid reduction in α_{so} [see Eqs. (34) and (46) and Fig. 1] the role of the exchange mechanism rises. In the case of cubic semiconductors, when the spin-orbit interaction is considerably less than in the case of hexagonal crystals and J has approximately the same value,[4] the exchange mechanism becomes dominant.

A mechanism similar to the proposed exchange EDSR, but based on allowance for fluctuations of the classical spin vector of the magnetic impurities, was considered in Ref. 24 where only an estimate was obtained of the probabilities of a spin-flip transition for just one virtual $2p$ state of a carrier. This in fact corresponds to our expression (49), but an estimate obtained in Ref. 24 is considerably smaller than ours.

Under certain experimental conditions, in addition to the spin-orbit absorption there can be a second absorption peak[5] shifted by an amount of the order of $\hbar\omega_M$ in the direction of higher energies. The profiles of both lines and their magnetic field dependences are identical, so that the second peak can be attributed again to a spin transition of a localized carrier. The present theory predicts a shift of the frequency of the transition (27) in the case of the exchange EDSR in the direction of red wavelengths (since $\tilde{g} > 0$—see Ref. 5). Such a shift is associated with the fact that in the adopted model of the exchange interaction of a carrier with isolated magnetic impurities when g_M and $\tilde{g} > 0$, the absorption of an energy $\hbar\omega_s$ by an electron in a spin-flip transition is accompanied by a reduction in the energy of one of the magnetic impurities by an amount $\hbar\omega_M$. This model is satisfactory at low concentrations of magnetic impurities; the concentration $x = 0.1$ is no longer so small as to satisfy this approximation properly, and one should bear in mind the interaction of a carrier with groups of magnetic impurities. Unfortunately, detailed experiments on samples with lower impurity concentrations, when this model would be known to be valid, have not yet been carried out (to our knowledge) and the absence of a description of the characteristics of the additional peak in Ref. 5 prevents us from a reliable attribution of this peak to the exchange EDSR mechanism. The authors are deeply grateful to É.I. Rashba for his constant interest in this investigation.

APPENDIX A. CONDITION OF VALIDITY OF PERTURBATION THEORY

The matrix element H_{nd} for the electron wave functions with the opposite spins is

$$M = \langle \psi_\nu \uparrow | H_{nd} | \psi_{\nu'} \downarrow \rangle = -\frac{J}{\sqrt{2}} \sum_j S_{j\bar{1}} \psi_\nu^* (\mathbf{R}_j) \psi_{\nu'} (\mathbf{R}_j). \tag{A1}$$

An estimate of its quantity from the norm gives

$$\langle \mathrm{Tr}_M (|M|^2 \rho_M) \rangle = {}^1/_2 J^2 n \overline{S_{j1} S_{j\bar{1}}} \int d\mathbf{R} |\psi_\nu(\mathbf{R})|^2 |\psi_{\nu'}(\mathbf{R})|^2 \sim \gamma_{\nu\nu'}^2,$$

$$\gamma_{\nu\nu'}^2 \leqslant \gamma_0{}^2 = J^2 n \overline{S_z{}^2} \int d\mathbf{R} |\psi_0(\mathbf{R})|^4. \tag{A2}$$

Here, ρ_M is the density matrix of the magnetic impurities [see Eqs. (16) and (17)], $\gamma_{\nu\nu'}$ [Eq. (28)] is the characteristic fluctuation of the separation between the donor levels, whereas γ_0 of Eqs. (28) and (32) represents broadening of the energy of the ground state. Therefore, the condition of validity of perturbation theory simply demands narrow levels compared with the separations between them, whereas the expressions for the absorption coefficients given by Eqs. (29), (45), and (48) are valid if

$$|E_\nu - E_0 - \hbar\omega_s'|, \quad |E_\nu - E_0 - \hbar\omega_s| \gg \gamma_0, \tag{A3}$$

$$\hbar\omega_s \gg \gamma_0. \tag{A4}$$

In fact, these conditions are satisfied in a wide range of $\hbar\omega_s$ because of the inequality $na^3 \gg 1$.

APPENDIX B. AVERAGING OVER MAGNETIC IMPURITIES

We are interested in the averages (16) and (5) of the characteristic operator [see Eqs. (20) and (40)]

$$\mathscr{T} = \exp\Big[i \sum_j A(\mathbf{R}_j) S_{jz} \Big], \tag{B1}$$

$$\mathscr{T}_{av} = \langle \mathrm{Tr} \tilde{\rho}_M \mathscr{T} \rangle \tag{B2}$$

$$= \prod_{j=1}^{N} \int \frac{d\mathbf{R}_j}{V} \frac{\mathrm{Tr}_M \exp\{-[\Lambda(\mathbf{R}_j) - iA(\mathbf{R}_j)] S_{jz}\}}{Z(\Lambda(\mathbf{R}_j))}.$$

Here the averaging is expressed in terms of a more general density matrix than that used in Eq. (17):

$$\tilde{\rho}_M = \exp\Big\{ -\sum_j \Lambda(\mathbf{R}_j) S_{jz} \Big\} \Big/ \tilde{Z}_M, \tag{B3}$$

$$\tilde{Z}_M = \mathrm{Tr}_M \exp\Big\{ -\sum_j \Lambda(\mathbf{R}_j) S_{jz} \Big\}, \quad \Lambda(\mathbf{R}_j) = \lambda + \frac{J}{2T} |\psi_0(\mathbf{R}_j)|^2, \tag{B4}$$

which allows for the magnetic polaron contribution (when $J > 0$). In Eq. (B2) the quantity $Z(\Lambda)$ is described by an expression similar to Eq. (24).

Equation (B2) is easily transformed to

$$\mathscr{T}_{av} = \prod_j \int \frac{d\mathbf{R}_j}{V} \frac{Z(\Lambda(\mathbf{R}_j) - iA(\mathbf{R}_j))}{Z(\Lambda(\mathbf{R}_j))}$$

$$= \Big\{ 1 + \int \frac{d\mathbf{R}}{V} \Big[\frac{Z(\Lambda(\mathbf{R}) - iA(\mathbf{R}))}{Z(\Lambda(\mathbf{R}))} - 1 \Big] \Big\}^N$$

$$= \exp\Big\{ n \int d\mathbf{R} \Big[\frac{Z(\Lambda(\mathbf{R}) - iA(\mathbf{R}))}{Z(\Lambda(\mathbf{R}))} - 1 \Big] \Big\}. \tag{B5}$$

Here, N is the number of impurities in the volume V of a crystal: $N \to \infty$, $V \to \infty$, and $N/V = n$. If we ignore the polaron shift in Eq. (B4), we obtain directly

$$\mathscr{T}_{av} = \exp\Big\{ n \int d\mathbf{R} \Big[\frac{Z(\lambda - iA(\mathbf{R}))}{Z(\lambda)} - 1 \Big] \Big\}, \tag{B6}$$

and $Z(\lambda)$ is described by Eq. (24).

Equations (B5) and (B6) are valid for any value of S.

In particular, if $S \gg 1$ we can use Eq. (24) to obtain

$$\mathscr{T}_{\text{av}}=\exp\left\{n\int d\mathbf{R}\,(e^{-iSA(\mathbf{R})}-1)\right\}. \tag{B7}$$

This expression is taken from Ref. 25 and it describes the averaging, over the coordinates of zero-spin impurities, of the functional depending on the potential of the impurities $W(\mathbf{R}_j) = -iSA(\mathbf{R}_j)$.

In the calculation of $Z(\Lambda - iA)$ in Eqs. (B5) and (B6) we can employ both Eq. (24) and the expression for the sine of the difference, but at low values of A such as those in our experiments it is sufficient to take a finite number of the terms of the expansion $Z(\Lambda - iA)$ in powers of A. Then, using Eq. (23), we find that

$$\mathscr{T}_{\text{av}}=\exp\left\{n\int d\mathbf{R}\sum_{p=1}^{\infty}\overline{S_z{}^p}\,\frac{i^pA^p(\mathbf{R})}{p!}\right\}. \tag{B8}$$

It is also easy to average a more complex operator

$$\tilde{\mathscr{J}} = \sum_j S_{jz}{}^p f(\mathbf{R}_j)\exp[i\tilde{A}(\mathbf{R}_j)S_{jz}]\exp\left[\sum_{j'\neq j}A(\mathbf{R}_{j'})S_{j'z}\right], \tag{B9}$$

which appears in the theory of the exchange EDSR [Eqs. (20) and (22)] without approximations used in the derivation of Eq. (21). By analogy with the preceding procedure, we obtain

$$\tilde{\mathscr{T}}_{\text{av}}=n\int\frac{(-1)^pZ^{(p)}(\Lambda(\mathbf{R})-i\tilde{A}(\mathbf{R}))}{Z(\Lambda(\mathbf{R}))}f(\mathbf{R})\,d\mathbf{R}$$

$$\times\exp\left\{n\int d\mathbf{R}'\left[\frac{Z(\Lambda(\mathbf{R}')-iA(\mathbf{R}'))}{Z(\Lambda(\mathbf{R}'))}-1\right]\right\}. \tag{B10}$$

APPENDIX C. CALCULATION OF SUMS AND MATRIX ELEMENTS

The matrix elements of the EDSR of Eqs. (26) and (42) can be expressed in terms of a sum of the form

$$I=\sum_{\nu}\frac{\mathscr{A}_{0\nu}(r_{\bar{\mu}})_{\nu 0}}{E_\nu-E_0-E}. \tag{C1}$$

Here, $r_{\bar{\mu}}$ is defined in accordance with Eq. (33) and $\delta(\mathbf{r} - \mathbf{R})$ applies to the exchange EDSR mechanism and r_n applies to the spin–orbit mechanism; E_ν and Ψ_ν are the energies and eigenfunctions of the Hamiltonian of Eq. (31). In the calculation of the sums over the whole spectrum (discrete and continuous) of the Hamiltonian H_e one can use the method of Ref. 26.

In units of a and R^*, dropping the dimensional factor, we can write I in the form

$$I=\langle 0|\mathscr{A}\frac{1}{H+{}^{1}/\ -\varepsilon}r_{\bar{\mu}}|0\rangle\equiv\langle 0|\mathscr{A}|f_{\bar{\mu}}\rangle, \tag{C2}$$

where

$$H=-{}^{1}/{}_{2}\Delta-1/r, \tag{C3}$$

$$\varepsilon_\mu=(E+{}^{1}/{}_{2}\mu\hbar\omega_c)/R^*. \tag{C4}$$

According to Eq. (C2), $f_{\bar{\mu}}$ satisfies the equation

$$(H+{}^{1}/{}_{2}-\varepsilon_\mu)f_{\bar{\mu}}=r_{\bar{\mu}}\psi_0. \tag{C5}$$

If we seek this solution in the form

$$f_{\bar{\mu}}(\mathbf{r})=r_{\bar{\mu}}\exp(-r/v)\,u(r) \tag{C6}$$

with $v = (1 - 2\varepsilon_\mu)^{-1/2}$ which allows for the behavior of $f_{\bar{\mu}}$ at singular points, then u is described by

$$ru''+(4-2r/v)u'+(2-4/v)u=-2\pi^{-1/2}r\exp[-r(1-1/v)], \tag{C7}$$

the solution of which can be represented as a series of the generalized Laguerre polynomials $L_n^{(3)}$ (Ref. 27). As a result, we then have

$$f_{\bar{\mu}}(\mathbf{r})=r_{\bar{\mu}}\exp\left(-\frac{r}{v}\right)\sum_{n=0}^{\infty}C_nL_n^{(3)}\left(\frac{2r}{v}\right), \tag{C8}$$

where

$$C_n=\frac{32}{\pi^{1/2}}\frac{v^2}{(v+1)^6}\frac{2(v-1)-n}{n+2-v}\varkappa^{n-1},\qquad \varkappa=\frac{v-1}{v+1}. \tag{C9}$$

In particular, if $v = 1$ ($\varepsilon_\mu = 0$), we expect

$$f_{\bar{\mu}}(\mathbf{r})=(1/2\sqrt{\pi})\,r_{\bar{\mu}}(2+r)e^{-r}. \tag{C10}$$

In the case of the exchange EDSR mechanism it follows from Eq. (C2) that

$$I_{ex}{}^{\mu}(\varepsilon_\mu,\mathbf{R})=f_{\bar{\mu}}(\mathbf{R})\,\psi_0(R)=\pi^{-1/2}f_{\bar{\mu}}(\mathbf{R})e^{-R}. \tag{C11}$$

The summation over n in $f_{\bar{\mu}}$ of Eq. (C8) is carried out using the relationship

$$\frac{1}{n+2-v}=\frac{1}{2-v}\int_0^1 t^{n/(2-v)}\,dt \tag{C12}$$

and the generating function for the Laguerre polynomials. As a result, we obtain

$$I_{ex}{}^{\mu}(\varepsilon_\mu,\mathbf{R})=R_{\bar{\mu}}\frac{32}{\pi}\frac{v^2}{(v+1)^5(v-1)}e^{-R(1+1/v)} \tag{C13}$$

$$\times\left[\frac{v}{2-v}\int_0^1 p(t)\,dt-p(1)\right],$$

$$p(t)=\frac{\exp\{-R\varkappa(t)/[1-\varkappa(t)]\}}{[1-\varkappa(t)]^4},\qquad \varkappa(t)=\varkappa t^{1/(2-v)}. \tag{C14}$$

The quantity

$$\int\left|\sum_{\mu}e_\mu[I_{ex}{}^{\mu}(\varepsilon_\mu,\mathbf{R})+I_{ex}{}^{\mu}(-\varepsilon_\mu,\mathbf{R})]\right|^2 d\mathbf{R},$$

which occurs in the absorption coefficient, does not contain the interference terms for different polarizations e_μ, because $I_{\text{ex}}^{\mu} \propto R_{\bar{\mu}}$. Finally, we find that $\Phi_{\text{ex}}(\varepsilon)$, occurring in Eq. (34) is given by

$$\Phi_{ex}(\varepsilon)={}^{1}/{}_{2}({}^{3}/{}_{8}-\varepsilon)^2\int[I_{ex}{}^{\mu}(\varepsilon,\mathbf{R})+I_{ex}{}^{\mu}(-\varepsilon,\mathbf{R})]^2\,d\mathbf{R} \tag{C15}$$

and is independent of μ. Integration with respect of $\mathbf{R}$ is elementary and the remaining integrals with respect to t are readily found numerically.

In the case of $I_{\text{so}}(\varepsilon)$ we find from Eq. (C7) that

$$I_{so}(\varepsilon)=\int\psi_0r_\mu f_{\bar{\mu}}(\mathbf{r})\,d\mathbf{r}=2v^2\frac{1-2v^2}{(v^2-1)^2}$$

$$+\frac{256}{3}\frac{v^9}{(v-1)^2(v+1)^{10}}\sum_{n=0}^{\infty}\frac{(n+1)(n+2)(n+3)}{n+2-v}\varkappa^{2n}, \tag{C16}$$

which can be expressed in terms of the hypergeometric function [see Eq. (44)].

Another method for calculation of a quantity analogous to I_{so} by an integral representation of the Green Coulomb function was proposed in Refs. 28 and 29 in connection with the scattering of light by the hydrogen atoms. In particular, the result obtained in Ref. 28 is expressed in terms of a sum of two hypergeometric functions. A similar calculation attempt was made in Ref. 12, but a closed expression was not obtained.

APPENDIX D. VARIATIONAL METHOD FOR THE CALCULATION OF SUMS

Equation (C5) can be derived by variation with respect to f^* of the functional

$$\mathscr{F}=\langle f|H-\varepsilon'|f\rangle-\langle f|r_{\bar{\mu}}|\psi_0\rangle-\langle\psi_0|r_\mu|f\rangle. \tag{D1}$$

Let us assume that $f=C\psi$ and then after variation with respect to C (or C^*) we find that

$$\mathscr{F}=-|\langle\psi|r_{\bar{\mu}}|\psi_0\rangle|^2/\langle\psi|H-\varepsilon'|\psi\rangle. \tag{D2}$$

This functional was obtained in Ref. 14 where it is shown that $\mathscr{F}=-I_{so}$ for ordinary semiconductors, when the essentially diamagnetic term in H_c and the approximation described by Eq. (31) are invalid. Minimization of $\mathscr{F}$ for a deformed functional of a p state (anisotropic Gaussian, multiplied by the coordinate) gives the exact result for high magnetic fields $\mathscr{H}$ and is satisfactory when $\mathscr{H}=0$. In the present case, when the Zeeman term is more important than the diamagnetic one, a more suitable variational function is

$$\psi=r_{\bar{\mu}}\sum_{i=1}^{l}C_ie^{-\alpha_ir}, \tag{D3}$$

which is characterized by

$$\langle\psi|r_{\bar{\mu}}|\psi_0\rangle=\frac{1}{\pi^{1/2}}\sum_i\frac{C_i}{(\alpha_i+1)^5}, \tag{D4}$$

$$\langle\psi|H-\varepsilon'|\psi\rangle=\frac{1}{32\pi}\sum_{i,j}\frac{C_iC_j}{(\alpha_i+\alpha_j)^5}\left(\frac{\alpha_i\alpha_j}{2}-\frac{\alpha_i+\alpha_j}{4}-\varepsilon'\right). \tag{D5}$$

In the case of minimization with respect to α_i and C_i it is found that already for $l=2$ the agreement with the calculations based on Eq. (C16) is accurate to within four significant figures (six significant figures for $l=4$), whereas I_{ex} for the function (D3) is identical with that calculated from Eq. (C13) to three and four significant figures, respectively. This discrepancy is to be expected, because in contrast to $-I_{so}$, which is in the form of a correction in the second order of perturbation theory in the Stark effect, I_{ex} is not an extremum of $\mathscr{F}$.

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