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Exchange-induced optical spin transitions in semimagnetic semiconductors

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Electron-spin flip by electric dipole light absorption in semimagnetic semiconductors (SMSC) is allowed not only by the generally invoked spin-orbit coupling but also due to a possibility of exchange scattering of an electron by a magnetic impurity. The theory of such exchange-induced electric dipole spin resonance (EDSR) in Mn- and Fe-based SMSC is presented. Since exchange scattering changes the electron spin, as well as the impurity state, the bound-electron absorption occurs at the photon energy $\hbar\omega=\hbar\omega_s+\varepsilon_M$, where $\hbar\omega_s$ is the electron giant spin splitting and ε_M is an excitation energy of the magnetic impurity. In the case of Mn-based SMSC, ε_M is simply the Zeeman energy of a Mn ion, while, since an Fe ion may be excited to a set of states, several absorption lines exist in Fe-based SMSC. Exchange-induced EDSR differs from the conventional spin-orbit EDSR in the polarization and magnetic-field dependences of absorption. For free carriers exchange-induced spin-flip absorption has nonresonant character and leads to a rather broad absorption band.

I. INTRODUCTION

Among the large number of different semimagnetic semiconductors (diluted magnetic semiconductors) which have been intensively examined during the past decade, wide-gap $A^2{}_{1-x}M_xB^6$ compositions (M is ordinarily Mn or Fe) hold a significant place. Many nontrivial physical phenomena caused by strong exchange interactions between the carriers and the magnetic ions have been discovered in these materials. This interaction is responsible for the striking features, e.g., giant spin splitting, bound magnetic polaron, etc.[1-3] These effects have been observed with different optical techniques in an applied magnetic field: exciton absorption, spin-flip Raman scattering, Faraday rotation.[4-8] Deep insight into the nature of the states in such a complicated system (carriers interacting with magnetic impurities) has been gained by the investigation of electric dipole spin resonance (EDSR),[9-11] which occurs in these materials in the far-infrared optical region. Optical absorption at EDSR arises due to carrier transitions, induced by the electric field of the incident light, between the states with opposite spin directions. Such transition may become allowed, for instance, by spin-orbit coupling, as was predicted by Rashba[12] (see also the review article in Ref. 13 and references therein).

In semimagnetic semiconductors, however, there exists an alternative mechanism for EDSR,[14,15] which will be described in the present paper. The same exchange interaction that strongly renormalizes the carrier spectrum allows also flip-flop scattering of the carrier at one of the magnetic ions. The electric dipole electron transition accompanied by this kind of scattering is the specific EDSR in semimagnetic semiconductors.

The term "semimagnetic semiconductor" in its narrow meaning implies that the concentration of magnetic impurities is not large enough to produce any long-range magnetic ordering, i.e., a system of magnetic ions can be considered as being paramagnetic. Usually it holds true up to $x \leq 0.1$. At such low concentrations the magnetic ions may be considered as being single, paired, etc., with the fraction of pairs and other clusters small compared to the number of singles. That is why we shall restrict ourselves to the interaction of a carrier with only the isolated single ions.

In Sec. II we consider exchange-induced spin-flip transitions for a bound electron in both Mn- and Fe-based semiconductors. As the exchange scattering changes not only the electron but also the impurity states, the transition energy of exchange EDSR differs from that of conventional spin-orbit EDSR by the excitation energy of a magnetic impurity (one can achieve the intensities of both EDSR's to be comparable; the experiment in this region of parameters may exhibit two peaks of absorption). Since the polarizational dependences of these two kinds of resonances are different, it is rather easy to distinguish between the exchange and spin-orbit absorption experimentally.

In Sec. III we extend our theory for the case of free carriers, where the electric dipole absorption exhibits additional polarization-dependent features.

In Sec. IV the results obtained are discussed and compared with the experimental data[9,10] and previous calculations[11] for spin-orbit EDSR. It contains also a brief discussion of magnetic dipole transitions in such semiconductors.

II. EXCHANGE EDSR OF AN ELECTRON BOUND ON A SHALLOW DONOR

The system under consideration may be described by the following Hamiltonian:

$$\mathcal{H}=\mathcal{H}_e+g_e\mu_B(\mathbf{s}\cdot\mathbf{H})+\sum_j\mathcal{H}_{M,j}+\mathcal{H}_{\mathrm{ex}}\ , \tag{1}$$

where the first two terms correspond to the bound electron in an applied magnetic field $\mathbf{H}$ (g_e is the host g factor of the electron, μ_B is the Bohr magneton, and $\mathbf{s}$ is the electron spin). $\mathcal{H}_{M,j}$ is the Hamiltonian of the jth impurity, and the Hamiltonian of the exchange interaction between the electron and the subsystem of magnetic ions is chosen in the conventional form:[6]

$$\mathcal{H}_{\mathrm{ex}}=-J\sum_j(\mathbf{s}\cdot\mathbf{S}_j)\delta(\mathbf{r}-\mathbf{R}_j)\ . \tag{2}$$

Here $\mathbf{r}$ is the electron coordinate, $\mathbf{R}_j$ and $\mathbf{S}_j$ are the position and the spin of jth impurity, and J is an exchange constant.

The wave function of the shallow donor electron $\psi(\mathbf{r})$ spans many interatomic distances. So, its volume in the relevant range of impurity concentration contains a large number of magnetic impurities, i.e., the strong inequality $n_M a^3 \gg 1$ holds [a is the bound electron Bohr radius; n_M is the concentration of magnetic ions (singles) $n_M=N_0\bar{x}$, where N_0 is the number of cations per unit volume and $\bar{x}<x$ is the content of the singles in the solid solution]. Since the mean interaction energy between the electron and one of the magnetic ions is of the order of $J|\psi(\mathbf{R}_j)|^2 \sim J/a^3$, then in the case when

$$J/a^3 \ll \varepsilon_M\ , \tag{3}$$

where ε_M is an energy spacing between impurity levels, one can neglect the influence of the electron on the magnetic impurities. On the other hand, as the electron is placed in the field of a large number $n_M a^3$ of impurities, their contribution to the electron spin splitting is of the order of $J\sum_j|\psi(\mathbf{R}_j)|^2 \simeq Jn_M$ and is not small. That is the reason why it is convenient to divide $\mathcal{H}_{\mathrm{ex}}$ into the large mean value $\langle\mathcal{H}_{\mathrm{ex}}\rangle$ and the fluctuating part V:

$$\mathcal{H}_{\mathrm{ex}}=\langle\mathcal{H}_{\mathrm{ex}}\rangle+V\ ,\quad \langle\mathcal{H}_{\mathrm{ex}}\rangle=-Jn_M(\mathbf{s}\cdot\langle\mathbf{S}\rangle)\ . \tag{4}$$

In such an approximation the average spin is the same for all impurities ($\langle\mathbf{S}_j\rangle=\langle\mathbf{S}\rangle$) and is parallel to $\mathbf{H}$. The mean value $\langle\mathcal{H}_{\mathrm{ex}}\rangle$ leads to the electron giant spin splitting[1]

$$\hbar\omega_s=g_e\mu_B H-Jn_M\langle S_z\rangle\quad (\mathbf{H}\|z)\ . \tag{5}$$

It is also convenient to introduce the effective electron g factor $g^*=\hbar\omega_s/\mu_B H$. In wide-gap semiconductors, as a rule $g^*>0$, and in the following we shall assume that the effective g factor is positive. There are no difficulties in generalizing the results for the case $g^*<0$.

As Mn- and Fe-based semimagnetic semiconductors possess several different features, we shall examine them separately.

A. EDSR of a bound electron in Mn-based semimagnetic semiconductors

Here we shall consider in more detail the case of Mn-based semimagnetic semiconductors. Five electrons in the half-filled d shell of manganese, according to Hund's rule, constitute the total angular momentum $L=0$ and the spin $S=\frac{5}{2}$. So

$$\mathcal{H}_{M,j}=\hbar\omega_M S_{jz}\ ,\quad \hbar\omega_M=g_M\mu_B H\quad (g_M\simeq 2)\ . \tag{6}$$

Hence the energy scheme of the magnetic ion subsystem within an electron Bohr orbit represents a set of equidistant levels separated by $\hbar\omega_M$ [Fig. 1(a)]. Each of the levels is highly degenerate because a given value M of the total spin momentum of the impurity subsystem may be constructed from the spin momenta of the individual impurities in numerous ways. To obtain the energy spectrum of the whole system (electron plus impurities) one has to allow for the exchange interaction that lifts this degeneracy. As a result, the spectrum of any single donor electron and impurities consists of a set of bands [Fig. 1(b)] separated by the energy $\hbar\omega_M$. Each of the bands has a fine structure. Every state of this structure can be obtained from the adjacent one by an increase of the spin momentum for the jth impurity and the simultaneous decrease of that for the j'th impurity by one. Respectively, the energy spacing of levels in the fine structure is $J[|\psi(\mathbf{R}_j)|^2-|\psi(\mathbf{R}_{j'})|^2]/2 \sim J/a^3 \ll \hbar\omega_M$. However, since the crystal contains many donors, the different configurations of impurities near the different donors wash out this fine structure, and the bands may be treated as quasicontinuous. It leads to an additional inhomogeneous broadening. As the states are broadened by the fluctuations of independent individual magnetic momenta of numerous impurities and by their random positions, the total picture of the density of states will have the form of the set of Gaussian bands. The width of each Gaussian band is of the order of $Jn_M/(n_M a^3)^{1/2}$.

Now we shall consider the optical spin transitions in such a system.

It is important to note that the fluctuating part V of the exchange interaction [see Eq. (4)] not only forms the

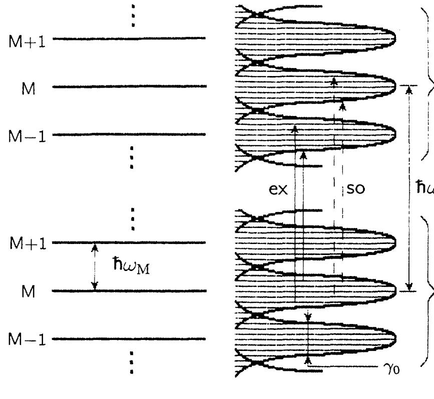

FIG. 1. Pattern of the energy spectrum of a system containing a donor electron in its ground spin doublet and magnetic impurities (Mn): (a) in the absence of an interaction between the electron and impurities; (b) with an account for this interaction (solid arrows indicate exchange-induced electron transitions, dashed indicate spin-orbit-induced transitions).

bands, but also mixes different electron orbital states (s, p, d, etc.) due to the δ functions in (2), and also the states with opposite spins due to the operator $s_+S_{j,-}+s_-S_{j,+}$ $[s_\pm=(s_x\pm is_y)/\sqrt{2}]$. Therefore, the operator $\mathbf{e}\cdot\mathbf{r}$ ($\mathbf{e}$ is a vector of polarization of incident light) of the electric dipole interaction allows the spin-flip transitions of the electron.

The magnitude of the fluctuating part V is of the order of the bandwidth, i.e., $Jn_M/(n_Ma^3)^{1/2}$. If it is small compared to the energy spacing between the donor-electron-energy levels and to its spin splitting $\hbar\omega_s$, one may treat V as a perturbation. In such an approximation the most prominent transition occurring between spin components of the ground-state spin doublet $|0\rangle\downarrow\rightarrow|0\rangle\uparrow$ may be considered as a photon-induced virtual transition $|0\rangle\downarrow\rightarrow|\nu\rangle\downarrow$ and subsequent exchange scattering to $|0\rangle\uparrow$ (or vice versa) (Fig. 2). Note that the electron spin flip is accompanied by a simultaneous change of the spin for one of the magnetic impurities.

As a resume of all that was said above, we can write (after averaging over magnetic impurities) the coefficient of light absorption as

$$\alpha_{\text{ex}}(\omega)=\frac{4\pi^2e^2\omega n_e}{\sqrt{\kappa}c}\langle S_+S_-\rangle n_M \times\int|M_{\text{ex}}(R)|^2dR\,\Delta(\hbar\omega-\hbar\omega_s')\ , \tag{7}$$

$$\hbar\omega_s'=\hbar\omega_s-\hbar\omega_M\ ,\qquad \Delta(\hbar\omega-\hbar\omega_s')=\frac{1}{(2\pi)^{1/2}\gamma_0}\exp\left[-\frac{(\hbar\omega-\hbar\omega_s')^2}{2\gamma_0^2}\right]\ , \tag{8}$$

where $\langle S_+S_-\rangle=[S(S+1)-\langle S_z^2\rangle+\langle S_z\rangle]/2$ [in the case of high temperatures $T\gg\hbar\omega_M$, $\langle S_+S_-\rangle=S(S+1)/3$]. The matrix element for the $|0\rangle\downarrow\rightarrow|0\rangle\uparrow$ electron transition is

$$M_{\text{ex}}(R)=\frac{J}{\sqrt{2}}\sum_\nu\left[\frac{\langle 0|\mathbf{e}\cdot\mathbf{r}|\nu\rangle\langle\nu|\delta(\mathbf{r}-\mathbf{R})|0\rangle}{E_\nu-E_0+\hbar\omega}+\frac{\langle 0|\delta(\mathbf{r}-\mathbf{R})|\nu\rangle\langle\nu|\mathbf{e}\cdot\mathbf{r}|0\rangle}{E_\nu-E_0-\hbar\omega}\right]\ . \tag{9}$$

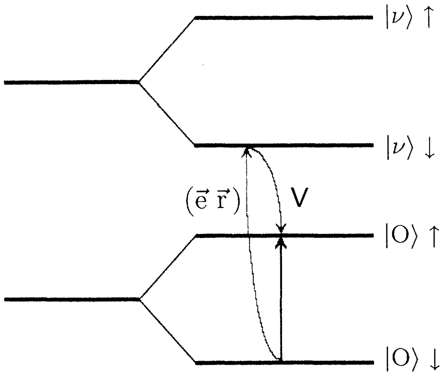

FIG. 2. Exchange-induced spin-flip transition of a bound electron (thick arrow) and the corresponding virtual transitions (thin arrows) with the operators responsible for them.

In Eqs. (7)–(9) ω is the light frequency, κ is a static dielectric constant of the semiconductor, n_e is the concentration of donors, $E_0,E_\nu(|0\rangle,|\nu\rangle)$ are the energies (wave functions) of ground and excited states of a donor electron in magnetic field (i.e., the eigenvalues and eigenfunctions of H_e).

In the spirit of the perturbation approach employed, the expressions (7) and (9) for the absorption coefficient are valid when the energy denominators in (9) are large compared to the fluctuating part $V\sim Jn_M/(n_Ma^3)^{1/2}$. So, when the initial and final states of the whole system vary each within its own band (see Fig. 1), i.e., $\hbar\omega$ varies by the value of the order of the bandwidth, the transition matrix element (9) remains almost constant [note that the initial and final orbital electron states in (9) are fixed]. In this case the shape of the absorption line is defined by the form of the density of states, i.e., it is Gaussian with the peak energy $\hbar\omega_s'=\hbar\omega_s-\hbar\omega_M$ and the width

$$\gamma_0^2=n_MJ^2\langle S_z^2\rangle\int|\psi_0(\mathbf{R})|^4dR\ . \tag{10}$$

A detailed derivation of the absorption coefficient (7), which also provides the systematic method to obtain corrections to the shape (8), has been published in Ref. 15.

To compute the transition matrix element (9), let us introduce three basic light polarizations e_μ: Voigt $e_0=e_z$ and Faraday $e_{\pm1}=(e_x\pm ie_y)/\sqrt{2}$. It is convenient to define the function

$$f_\mu(\mathbf{r};\hbar\omega)=\left\langle\mathbf{r}\left|\frac{1}{\mathcal{H}_e-E_0-\hbar\omega}r_\mu\right|\mathbf{0}\right\rangle\ . \tag{11}$$

Then

$$M_{\text{ex}}(\mathbf{R})=\frac{J}{\sqrt{2}}\sum_\mu[\mathcal{M}_\mu^*(\mathbf{R};-\hbar\omega)+\mathcal{M}_{\bar\mu}(\mathbf{R};\hbar\omega)]e_\mu\ ,\qquad \mathcal{M}_\mu(\mathbf{r};\hbar\omega)=\psi_0^*(\mathbf{r})f_\mu(\mathbf{r};\hbar\omega)\quad(\bar\mu\equiv-\mu)\ . \tag{12}$$

The function $f_\mu(\mathbf{r};\hbar\omega)$ may be computed for arbitrary form of the electron Hamiltonian and magnetic-field strength by means of a specific variational procedure.[16,15] Using this procedure $\alpha(\omega)$ may be calculated for a semiconductor with any value of the band gap, including nonparabolicity of the energy spectrum. However, for donor bound electrons the most favorable case is of a wide-gap semiconductor, where $\hbar\omega_c\ll R^*$ at all experimentally relevant magnetic fields (here $\omega_c=eH/m^*c$ is the cyclotron frequency and $R^*=m^*e^4/\hbar^2\kappa^2$ is the effective double Rydberg). In this case spin-flip transitions are not covered by the donor-cyclotron ones. Under this condition all formulas can be sufficiently simplified, as one can neglect the term proportional to H^2 in the Hamiltonian of the donor electron and write it as

$$\mathcal{H}_e=-\frac{\hbar^2}{2m^*}\Delta-\frac{e^2}{\kappa r}+\frac{1}{2}\hbar\omega_c\mathcal{L}_z \tag{13}$$

($\mathcal{L}_z$ is the z component of electron angular momentum). This form of the Hamiltonian makes it possible to solve Eq. (11) exactly, the solution being

$$f_\mu(\mathbf{r};\hbar\omega)=\frac{r_\mu}{R^*}\exp\left[-\frac{r}{av}\right]\sum_{n=0}^{\infty}C_nL_n^{(3)}\left[\frac{2r}{av}\right],$$
$$v=\left[1-\frac{2\hbar\omega-\mu\hbar\omega_c}{R^*}\right]^{-1/2}, \qquad (14)$$
$$c_n=\frac{32}{\sqrt{\pi}}\frac{v^2}{(1+v)^6}\frac{2(v-1)-n}{(n+2-v)}\left[\frac{v-1}{v+1}\right]^{n-1}$$

($L_n^{(3)}$ are the generalized Laguerre polynomials[17]). With this solution, expression (12) for $M_{\mathrm{ex}}(\mathbf{R})$ and the corresponding expression (7) for the absorption coefficient $\alpha(\omega)$ precisely take into account the virtual-electron transitions to all its intermediate states including the continuous spectrum. It is seen from Eqs. (14) and (12) that $f_\mu(\mathbf{R},\hbar\omega)$ and $M_{\mathrm{ex}}(\mathbf{R})$ depend on the direction of $\mathbf{R}$ as R_μ. Therefore, $\int|M_{\mathrm{ex}}(\mathbf{R})|^2d\mathbf{R}$ does not contain the terms proportional to $e_\mu e_{\mu'}^*$ for $\mu\neq\mu'$, and the absorption coefficient takes the form

$$\alpha(\omega)=\sum_\mu\alpha_\mu(\omega)|e_\mu|^2\ ,\quad \mu=0,\pm1\ . \qquad (15)$$

In an experimentally reasonable frequency region $0<\hbar\omega<E_{2p}-E_{1s}$, where the spin-flip transition of a bound electron is not masked with ordinary electric dipole transitions (cf. Ref. 10), the partial absorption coefficients $\alpha_\mu(\omega)$ are almost equal to each other. The slight differences are connected with the small quantity $\mu\hbar\omega_c\ll R^*$ in v (14). This means that exchange-induced electric dipole spin resonance exists at any polarization of the incident light and is nearly isotropic. The polarization dependence is most prominent in the case when the spin-flip transition energy $\hbar\omega_s'$ ($=\hbar\omega$) approaches $3R^*/8$, i.e., the distance between $|0\rangle=1s\rangle$ and $|2p\rangle$ electron states. In this case $v\rightarrow2$ in Eq. (14) and only the term with $n=0$ in $f_\mu(\mathbf{r},\hbar\omega)$ should be taken into account [it corresponds simply to allowing for one resonant virtual transition $|1s\rangle\rightarrow|2p\rangle$in (9)—just this approximation was used in Ref. 18 for spin-orbit EDSR], and from Eqs. (7), (12), (14), and (15) we obtain

$$\alpha_\mu(\omega)=\frac{2^{16}\pi}{3^{15}}\frac{e^2\omega n_e}{\sqrt{\kappa}can_M}\langle S_+S_-\rangle\times\left[\frac{Jn_M}{\frac{3}{8}R^*-\frac{\mu}{2}\hbar\omega_c-\hbar\omega_s'}\right]^2\Delta(\hbar\omega-\hbar\omega_s')\ . \qquad (16)$$

The expression (16) demonstrates the increase of absorption on approaching parametric resonance $\hbar\omega\rightarrow E_{2p}-E_{1s}$ (although, of course, it is not applicable in the case of the exact resonance, where the perturbation method fails).

It is also possible to write down the simple formula for $\alpha(\omega)$ in the limit of low magnetic fields, when $\hbar\omega_s'=\hbar\omega\ll R^*$ ($v=1$). Then, only the terms with $n=0$ and 1 in (14) are nonzero, and

$$f_\mu(\mathbf{r},\hbar\omega)=\frac{1}{2\sqrt{\pi}}\frac{r_\mu}{R^*}\left[2+\frac{r}{a}\right]\exp(-r/a)\ . \qquad (17)$$

Using (17), (7), and (12) we get

$$\alpha(\omega)=\frac{87\pi}{128}\frac{e^2\omega n_e}{\sqrt{\kappa}can_M}\left[\frac{Jn_M}{R^*}\right]^2\langle S_+S_-\rangle\Delta(\hbar\omega-\hbar\omega_s')\ . \qquad (18)$$

One interesting feature manifested by this equation is that the transition matrix element M_{ex} does not vanish as $\omega\rightarrow0$ [it is seen from a comparison of Eq. (18) with Eq. (7)], in contrast to spin-orbit EDSR.[19] For ordinary electric dipole spin transitions the vanishing of the matrix element as $\omega\rightarrow0$ (which corresponds to $H\rightarrow0$) is a consequence of the fact that the transition occurs between the states belonging to the same Kramers doublet. In our case of exchange EDSR the initial and final states differ from each other not only by the electron spin, but also by the spin of one of the magnetic impurities (see Fig. 1) and so do not constitute the Kramers doublet (the latter contains the initial state and the state with the opposite spin of the electron as well as of all magnetic impurities).

In the case of arbitrary frequency ω the calculation of $\alpha_\mu(\omega)$ from (14) and (12) is a straightforward but rather tedious procedure. The result we obtained is expressed via the sum of the hypergeometric functions and is rather clumsy to write down here.

The power of the absorption peak (integrated absorption) may be characterized by $\alpha(\omega=\omega_s')\Gamma$, where Γ is the full width of the resonance at half maximum. As the absorption peak has Gaussian form

$$\Gamma=\gamma_0(\ln4)^{1/2}/\pi c\hbar\ . \qquad (19)$$

The value of $\alpha_{\bar{1}}(\omega_s')\Gamma$ (i.e., in cyclotron-resonance-active Faraday polarization) as a function of photon energy $\hbar\omega=\hbar\omega_s'$ is plotted in Fig. 3 for $\mathrm{Cd}_{0.9}\mathrm{Mn}_{0.1}\mathrm{Se}$.

For comparison with known results we shall write an expression for the absorption coefficient $\alpha_{\mathrm{s.o.}}(\omega)$ of spin-orbit EDSR in a hexagonal crystal (CdMnSe is just such a crystal) which is caused by the term $H_{\mathrm{s.o.}}=2\alpha_0[\mathbf{s}\hat{\mathbf{k}}]\mathbf{n}$ additional to the Hamiltonian (1) (α_0 is the spin-orbit coupling constant, $\hat{\mathbf{k}}$ is the operator of electron quasimomentum in a magnetic field, and $\mathbf{n}$ is the unit vector along the hexagonal axis). After calculations the result may be expressed via the hypergeometric function

$$\alpha_{\mathrm{s.o.}}(\omega)=\frac{4\pi^2e^2\omega n_e}{c\sqrt{\kappa}}|M_{\mathrm{s.o.}}|^2\Delta(\hbar\omega-\hbar\omega_s)\ ,$$
$$M_{\mathrm{s.o.}}=\sqrt{2}\frac{\hbar\omega\alpha_0}{R^{*2}}[e_0\mathcal{J}_0(\hbar\omega)\sin\vartheta-e_{\bar{1}}J_{\bar{1}}(\hbar\omega)\cos\vartheta]\ ,$$
$$\mathcal{J}_\mu(\hbar\omega)=I_\mu(\hbar\omega)+I_\mu(-\hbar\omega)\ ,\quad \vartheta=\angle\mathbf{n},\mathbf{H}\ ,$$
$$I_\mu(\hbar\omega)=\int\psi_0^*(\mathbf{r})r_{\bar{\mu}}f_\mu(r;\hbar\omega)d\mathbf{r} \qquad (20)$$
$$=2v^2\frac{1-2v^2}{(v^2-1)^2}+\frac{(2v)^9}{(2-v)(v-1)^2(v+1)^{10}}\times F\left[4,2-v;3-v;\left[\frac{v-1}{v+1}\right]^2\right]\ .$$

Here the line shape is also defined by fluctuations in the magnetic subsystem and coincides with that [Eq. (8)] for

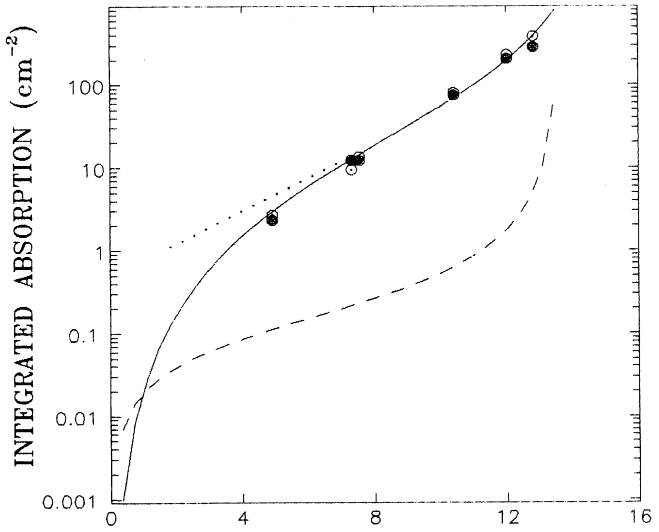

FIG. 3. Integral intensity of spin-flip absorption for donor-bound electrons in $Cd_{0.9}Mn_{0.1}Se$: points are the experimental data of Ref. 10 for spin-orbit EDSR at two close temperatures. The solid line represents $\alpha_{s.o.}(\omega=\omega_s)\Gamma$ [Eqs. (17) and (18)] as a function of photon energy $\hbar\omega$; the dotted line represents the results of Ref. 11; the results for exchange EDSR are shown with the dashed line. The following parameters were used: $N_0=1.83\times10^{22}$ cm^{-3}, $JN_0=260$ meV, $\alpha_0=2.45\times10^{-10}$ eV cm; the other parameters coincide with that of Refs. 10 and 11.

the exchange EDSR. One can see that the spin-orbit mechanism allows light absorption only in the Voigt geometry with the absorption intensity $\alpha_{s.o.}(\omega_s)\Gamma \propto \sin^2\vartheta$, and in the cyclotron-active Faraday geometry (for $g^*>0$) with $\alpha_{s.o.}(\omega_s)\Gamma \propto \cos^2\vartheta$.[20] Corresponding results for spin-orbit EDSR in $Cd_{0.9}Mn_{0.1}Se$ are presented in Fig. 3.

B. The peculiarities of exchange EDSR in Fe-based semimagnetic semiconductors

Iron has six electrons in the *d* shell and in distinction to Mn has the total spin momentum $S=2$ and the total orbital momentum $L=2$. This 25-fold degenerate state is split by both the crystal field and the spin-orbit coupling term $\lambda(\mathbf{S}\cdot\mathbf{L})$.[21] The main splitting is produced by the crystal field giving the lowest state with tenfold degeneracy, which is further lifted by the spin-orbit coupling. The resulting splitting energy ε_M between the terms in the lowest multiplet is about a few meV.[22,23] The detailed structure of this split multiplet depends on the symmetry of the crystal, but the main point is that the ground state is always a singlet. Nevertheless, it depends on magnetic field as an ordinary paramagnetic state with a nonzero spin because the Zeeman term $(\mathbf{S}\cdot\mathbf{H})$ admixes to it the other states of the multiplet (Van Vleck type of paramagnetism). In an experimentally reasonable region $T \ll \varepsilon_M$ the statistical averaging over Fe states is unnecessary (contrary to the Mn case) because each Fe ion occupies only this magnetic-field-dependent "singlet" ground state. This ground state is not the eigenstate of S_z, and the mean value

$$\langle S_{jz}\rangle=(0|S_z|0)\propto\hbar\omega_M/\varepsilon_M \quad (\text{for } \hbar\omega_M\ll\varepsilon_M) . \tag{21}$$

Here we use notation $|\;)$ rather than $|\;\rangle$ in order to distinguish impurity states from the electron states.

Analysis of exchange EDSR may be carried out in a similar way to the case of a Mn-based semiconductor. The transition process includes an exchange electron scattering on one of the Fe ions due to operator V from Eq. (4). Since $(0|S_\pm|0)=0$, the electron transition is accompanied by the impurity excitation to one of the states of the multiplet. The absorption coefficient is given by the same Eq. (7) with the replacements $\langle S_+S_-\rangle$ to $|(i|S_-|0)|^2$ and $\hbar\omega_s'$ to $\hbar\omega_s+\varepsilon_i-\varepsilon_0$. The shape of the absorption line remains Gaussian, but in this case it is defined purely by inhomogeneous broadening and, in addition, $\langle S_z^2\rangle$ should be replaced with $(0|S_z^2|0)$ in Eq. (10).

The essential feature as compared to the Mn case is that in the donor-electron spin-flip transition the Fe ion may be excited to several possible states with different energies ε_i. Due to that, the experimental absorption spectrum may contain several lines. The exact number of such lines and their relative intensities depend on the symmetry of the crystal, the strength of the magnetic field, and its orientation with respect to the crystal axes. To find the eigenstates and eigenfunctions of the matrix Hamiltonian of iron in a general case of an arbitrarily directed magnetic field such as $\hbar\omega_M\sim\varepsilon_M$, it is necessary to use numerical calculations.[24,25] However, in a simple case of a cubic crystal and magnetic field $H\parallel\langle100\rangle$ this Hamiltonian may be diagonalized analytically, and we shall sketch the results.

A crystal field with cubic symmetry splits the 25-fold degenerate term of Fe^{2+} into the orbital triplet ^5T and separated from it by the energy Δ the lower-lying orbital doublet ^5E. The wave functions of the latter are

$$\begin{aligned}
|1,m)&=Y_{2,0}|m) ,\\
|2,m)&=\frac{1}{\sqrt{2}}(Y_{2,2}+Y_{2,\bar{2}})|m) ,
\end{aligned} \tag{22}$$

where Y are the ordinary orbital (spherical) functions, and $m=0,\pm1,\pm2$ enumerates the z component of spin momentum. It was mentioned above that spin-orbit coupling $\lambda(\mathbf{S}\cdot\mathbf{L})$ is rather small $(\lambda\ll\Delta)$ and so may be treated as a perturbation. Because the matrix elements of $(\mathbf{S}\cdot\mathbf{L})$ with the functions (21) vanish, a 10×10 matrix Hamiltonian of the ^5E multiplet arises in the second order of perturbation theory

$$\begin{aligned}
\tilde{\mathcal{H}}_{nm,n'm'}=&-\frac{\lambda^2}{\Delta}(n,m|(\mathbf{S}\cdot\mathbf{L})^2|n',m')\\
&+(n,m|(L_z+2S_z)\mu_BH|n',m')\\
&\qquad (n,n'=1,2) ,
\end{aligned} \tag{23}$$

where the Zeeman term $(n,m|(L_z+2S_z)\mu_BH|n'm')=m\hbar\omega_M\delta_{m'm}\delta n'n$. This 10×10 matrix may be separated into four blocks:

$$A:\ \{|1,0),|2,\bar{2}),|2,2)\}\ ;\quad B:\ \{|1,\bar{1}),|2,1)\}\ ;$$

$$C:\ \{|1,1),|2,\bar{1})\}\ ,\quad D:\ \{|1,\bar{2}),|1,2),|2,0)\}$$

(the wave functions, which are mixed to one another, are indicated in braces). The ground state belongs to the A set; the corresponding wave function and the energy are given by the expressions

$$|0)=\frac{1}{2\eta}\left\{\frac{|2,2)}{2\xi+\eta}-\frac{|2,\bar{2})}{2\xi-\eta}+\sqrt{2}|1,0)\right\},$$

$$\varepsilon_0=-\Delta\varepsilon(3+\eta)\ ,\quad \eta=\sqrt{1+4\xi^2}\ , \tag{24}$$

$$\xi=\hbar\omega_M/\Delta\varepsilon\ ,\quad \Delta\varepsilon=6\lambda^2/\Delta\ .$$

It is directly seen from (24) that $(0|S_\pm|0)=0$, i.e., $\langle S\rangle \| H$, and[26]

$$\langle S_z\rangle=(0|S_z|0)=-4\xi/\eta=-\frac{4\hbar\omega_M}{[\Delta\varepsilon^2+(2\hbar\omega_M)^2]^{1/2}}\ . \tag{25}$$

This value defines the giant spin splitting (5) of electron levels.

The exchange-induced EDSR of a donor electron may be accompanied by excitation of an Fe ion to one of the two states belonging to the B set, because $S_-|0)$ is the superposition of $|1,\bar{1})$ and $|2,1)$. The wave functions of these states are of the form

$$|B\pm)=\frac{1}{\sqrt{2}}\left\{\left[1\mp\frac{1+2\xi}{2\zeta}\right]^{1/2}|1,\bar{1})\mp\left[1\pm\frac{1+2\xi}{2\zeta}\right]^{1/2}|2,1)\right\},$$

$$\zeta=(1+\xi^2+\xi)^{1/2}\ , \tag{26a}$$

with the energies

$$\varepsilon_{B\pm}=\Delta\varepsilon(-2\pm\zeta)\ . \tag{26b}$$

So, in this case there exist two lines of absorption with the transition energies

$$\hbar\omega_s'=\hbar\omega_s+\Delta\varepsilon(1+\eta\pm\zeta)\ . \tag{27}$$

The absorption coefficient for each of these two transitions is given by the same Eq. (7) with the changes mentioned at the beginning of this subsection. The matrix elements $\langle B\pm|S_-|0\rangle$ that define the absorption intensity may be easily found from Eqs. (24) and (26a). In particular, in the case of weak magnetic fields, such as $\hbar\omega_M \ll \Delta\varepsilon$, the intensities of these two lines are substantially different: $\langle B-|S_-|0\rangle=\sqrt{2}$, $\langle B+|S_-|0\rangle=\sqrt{3/2}(\hbar\omega_M/2\Delta\varepsilon)$, and the strongest line occurs at the photon energy $\hbar\omega=\hbar\omega_s+\Delta\varepsilon$.

III. EXCHANGE ELECTRIC DIPOLE SPIN TRANSITIONS OF FREE CARRIERS

Part V of the exchange interaction Hamiltonian (4) can also produce spin-flip transitions of free carriers. There is an important difference between the donor bound and free carrier. A free carrier possesses the continuous quantum number—the momentum along the magnetic field. Scattering of a free carrier by a magnetic impurity changes this momentum together with the other quantum numbers. Therefore, the absorption turns out nonresonant.

The simplest consequence expected is broadening of the absorption line. But as we shall see later, it also may produce a significant shift of the absorption maximum due to the contribution of indirect electron transitions.

The Hamiltonian for a free electron in a magnetic field has the conventional form

$$\mathcal{H}_e=\frac{\hbar^2\hat{k}^2}{2m^*}\quad\left[\hat{\mathbf{k}}=-i\nabla+\frac{e}{c\hbar}A\right], \tag{28}$$

with the wave functions and the energies

$$\psi_\nu=\frac{1}{l}\exp(ik_zz+iXy/\lambda^2)\cdot\chi_n(x-X)\ , \tag{29}$$

$$E_\nu=\hbar\omega_c(n+\tfrac{1}{2})+\frac{\hbar^2k_z^2}{2m^*}\ . \tag{30}$$

Here $\nu\equiv\{n,X,k_z\}$, λ is the magnetic length ($\lambda^2=c\hbar/eH$), l is a normalization length, χ_n are standard Landau functions, and we use the Landau gauge $A_y=Hx$, $A_x=A_z=0$.

In the interesting case of sufficiently large magnetic fields such as $\hbar\omega_s \gg T$, when all carriers occupy the Landau subbands with spin down only, the coefficient of exchange-induced spin-flip absorption may be represented in the form

$$\alpha(\omega)=\frac{4\pi^2e^2\omega n_e}{c\sqrt{\kappa}}\left[\frac{\hbar}{m^*\omega}\right]^2\langle S_+S_-\rangle\times n_M\int d\mathbf{R}\sum_{\nu'\nu}|M_{\nu'\nu}(\mathbf{R})|^2\rho_\nu\delta(E_{\nu'}-E_\nu+\hbar\omega_s'-\hbar\omega)\ . \tag{31}$$

Indices ν and ν' in (31) refer to the initial and final electron orbital states, respectively, ρ_ν is the electron distribution function, and $\hbar\omega_s'$ is defined in Sec. II. For free carriers it is more reasonable to write the transition matrix element in terms of the velocity operator $\hbar\hat{\mathbf{k}}/m^*$ rather than of the position $\mathbf{r}$ as was the case in (9):

$$M_{\nu'\nu}(\mathbf{R})=\frac{J}{\sqrt{2}}\sum_{\nu''}\left[\frac{\langle\nu'|\mathbf{e}\cdot\hat{\mathbf{k}}|\nu''\rangle\langle\nu''|\delta(\mathbf{r}-\mathbf{R})|\nu\rangle}{E_{\nu''}-E_\nu+\hbar\omega_s'}+\frac{\langle\nu'|\delta(\mathbf{r}-\mathbf{R})|\nu''\rangle\langle\nu''|\mathbf{e}\cdot\hat{\mathbf{k}}|\nu\rangle}{E_{\nu''}-E_{\nu'}-\hbar\omega_s'}\right] \tag{32}$$

[here the additional coefficient $\hbar/m^*\omega$ is accounted for in Eq. (31)]. This expression for the transition matrix element can be simplified if we take into account the energy conservation law and the fact that $\hat{k}_+$ ($\hat{k}_-$) acting on the function ψ_ν increases (decreases) the Landau quantum number n by unity, while $\hat{k}_z\psi_\nu=k_z\psi_\nu$. Then, for each of the three basic light polarizations e_μ (Voigt e_0 and two

Faraday $e_{\pm 1}$), the energy denominator in the first term in $M_{\nu'\nu}(R)$ may be replaced by $\hbar\omega + \mu\hbar\omega_c$, and the one in the second term by $(-\hbar\omega - \mu\hbar\omega_c)$. It is also straightforward to carry out the summation over the centers of cyclotron orbits X,X' and integration over $\mathbf{R}$ in Eq. (31), obtaining

$$n_M \int d\mathbf{R} \sum_{X,X'} |M_{\nu'\nu}(\mathbf{R})|^2 = \sum_{\mu} \Phi_\mu |e_\mu|^2 \,, \tag{33}$$

$$\Phi_\mu = \frac{l^3}{2(2\pi\lambda^2)^4} \frac{J^2 n_M}{(\hbar\omega + \mu\hbar\omega_c)^2} \begin{cases} (k_z - k_z')^2\lambda^2 \,, & \mu = 0 \\ n + n' + 1 \,, & \mu = \pm 1 \,. \end{cases}$$

It follows from (33) that Eq. (15) is valid also for the free-carrier absorption, and so formulas (31) and (33) enable us to obtain $\alpha_\mu(\omega)$ for any interrelation between ω_s and ω_c. It should be noted, however, that in wide-gap semiconductors the absorption by free carriers seems to be quite exotic due to the lack of a sufficiently high concentration of free carriers at low temperatures. Besides, since in wide-gap semiconductors $\hbar\omega_c \ll \hbar\omega_s$ (i.e., the spectrum of spin-polarized electrons represents a set of closely spaced Landau levels) and electron transitions may involve any change of Landau quantum number n [see Eq. (33)], one should expect a smooth, rather broad, and almost polarization-independent absorption band. On the other hand, in narrow-gap semiconductors, where usually $\hbar\omega_c \gg \hbar\omega_s$, it is experimentally favorable to observe pure spin-flip absorption. In this case the complicated general expression may be simplified, as the initial and final Landau quantum numbers $n = n' = 0$, the result being

$$\alpha(\omega) = \frac{1}{4\pi\sqrt{2\pi}} \frac{\omega_p^2}{\omega c\sqrt{\kappa}} \left[\frac{J n_M}{\omega + \mu\omega_c} \right]^2 \frac{1}{n_M \lambda^3} \langle S_+ S_- \rangle \times \int \frac{d\xi \exp(-\xi^2)}{[\xi^2/\beta + (\omega - \omega_s')/\omega_c]^{1/2}} \times \begin{cases} 2[2\xi^2/\beta + (\omega - \omega_s')/\omega_c] \,, & \mu = 0 \\ 1 \,, & \mu = \pm 1 \,. \end{cases} \tag{34}$$

Here $\omega_p^2 = 4\pi e^2 n_e / m^*$, $\beta = \hbar\omega_c / T$.

The formulas presented demonstrate one interesting feature of light absorption in the Voigt geometry. As one can see from Eq. (33) for the matrix element, it vanishes for direct electron transitions when $k_z = k_z'$. So, the light absorption occurs due to indirect transitions only.

On the other hand, in the Faraday geometry the direct transitions contribute to the light absorption, and, moreover, expression (34) leads to a logarithmic divergence at $\omega = \omega_s'$. This divergence is caused by singularities of the density of states at the bottoms of unperturbed Landau subbands. Of course, the developed theory is not valid in the immediate vicinity of this point, and to remove this divergence one needs to take into account some broadening of Landau levels. However, the expression (34) reproduces the general shape of the absorption peak whose maximum in the Faraday geometry is at the frequency $\omega = \omega_s'$. In the Voigt geometry such divergence does not appear at all due to the above-mentioned vanishing of the matrix element for the direct transitions. It shifts the absorption peak from $\omega = \omega_s'$ and makes a total picture of absorption rather different. Namely, after the integration over ξ in (34), which may be carried out quite simply due to the inequality $\beta \gg 1$, we get

$$\alpha(\omega) = \frac{\langle S_+ S_- \rangle}{2\sqrt{2}\pi} \frac{\omega_p^2}{\omega c\sqrt{\kappa}} \left[\frac{J n_M}{\omega + \mu\omega_c} \right]^2 \frac{1}{n_M \lambda^3} \left[\frac{\omega - \omega_s'}{\omega_c} \right]^{1/2} . \tag{35}$$

One can see from Eq. (35) that if ω_s' is proportional to H, then the absorption maximum occurs at $\omega \simeq 3\omega_s'/2$.

The peculiarities described of free-carrier spin-flip absorption with frequency-separated peaks in Voigt and Faraday geometries have been confirmed experimentally.[27]

From the macroscopic point of view the action of the magnetic impurities on the electron may be replaced by the action of slightly inhomogeneous internal magnetic field. This approach may account for some general features of the exchange interaction. It is usual for the description of magnetics, where free-carrier EDSR have been considered in Ref. 28. However, in contrast with our results, in Ref. 28 the electron quasimomentum k_z was conserved in spin-flip transitions.

IV. DISCUSSION OF RESULTS

First of all it is interesting to compare the intensities of the exchange-induced EDSR and conventional spin-orbit resonance. In Sec. II A we invoked the strongest in wide-gap semiconductors linear in $\hat{\mathbf{k}}$ spin-orbit interaction inherent to hexagonal crystals.[13] This interaction explains the origin of one of the peaks observed in $Cd_{0.9}Mn_{0.1}Se$ of bound electron magnetoabsorption.[9,10] The integrated absorption coefficients for these two resonances $\alpha(\omega = \omega_s)\Gamma$ and $\alpha_{s.o.}(\omega = \omega_s)\Gamma$ calculated in the present paper are plotted in Fig. 3 versus photon energy $\hbar\omega$ together with experimental points[10] for the cyclotron-resonance-active Faraday polarization (parameters are listed in the caption). The theoretical curve of Ref. 11 for spin-orbit EDSR is also shown in Fig. 3. The spin-orbit coupling constant α_0 was found from the best theoretical fit with the experimental points. It should be noted that our value $\alpha_0 = 1.7 \times 10^{-3} e^2$ ($2\alpha_0 = 4.9 \times 10^{-10}$ eV cm) differs from $\alpha_0 = 2.1 \times 10^{-3} e^2$ of Ref. 10 and $\alpha_0 = 2.2 \times 10^{-3} e^2$ of Ref. 11. This difference occurs mainly because a Lorentzian shape of the absorption line was assumed in Refs. 10 and 11.

Comparison of the exchange-induced and spin-orbit EDSR shows that the exchange EDSR is weaker than the spin-orbit one in this hexagonal semiconductor. However, in the region of small $\hbar\omega$ (that is, small magnetic fields) both EDSR's are of the same order of magnitude. Of course, one must keep in mind that the exchange-induced and spin-orbit absorption peaks are separated by the Zeeman energy $\hbar\omega_M$, so a double-peaked picture of absorption may be observed in experiment. In the case of wide-gap semiconductors with cubic symmetry, where spin-orbit coupling is much less than that in a hexagonal crystal, the exchange mechanism of EDSR should be the main reason for spin-flip absorption.

To fit the experimental data,[10] the authors of Ref. 11 have taken into account together with the electric dipole also the magnetic dipole absorption. They supposed that the intensity of the latter is governed by the same large value g^* that produces giant spin splitting. However, the g factor g_p entering the expression for the paramagnetic resonance intensity in Mn-based semimagnetic semiconductors is much less than g^*,[29,15] so the magnetic dipole absorption is negligible. The overestimation of the contribution of the magnetic dipole absorption in Ref. 11 is the cause of the difference between the theoretical curves in Fig. 3 for this resonance in the region of small photon energies.

From the physical point of view, the small value of g_p arises due to the small Zeeman frequency ω_M in comparison with $\omega=\omega_s$. In this case the magnetization of the magnetic ion subsystem cannot follow the high-frequency magnetic field of light $\tilde{\mathbf{H}}$, and it may be shown that $g_p=(\omega_s/\omega_s')(g_e-g_M)$.[15] The vanishing of g_p at $g_e=g_M$ may be easily understood if we consider the spin Hamiltonian of the system

$$\mathcal{H}_s=-J\sum_j(\mathbf{s}\cdot\mathbf{S}_j)|\psi_0(\mathbf{R}_j)|^2+\mu_B H\left[g_e s_z+g_M\sum_j S_{jz}\right] . \tag{36}$$

If $g_e=g_M$, the operator of the magnetic dipole interaction $g_M\mu_B(\mathbf{s}+\sum_j\mathbf{S}_j)\cdot\tilde{\mathbf{H}}$ commutes with the first term in (36) and the absorption can occur only at the Zeeman frequency ω_M.

In contrast to Mn-based semimagnetic semiconductors, in the Fe-based ones, where the Fe interlevel spacing ε_M may be comparable with the photon energy, the iron spins can follow the high-frequency magnetic field of light. The oscillating part of Fe magnetization, acting on the electron, contributes to a large value of g_p, which can even resonantly increase when $\hbar\omega_s\rightarrow\varepsilon_M$.[30] So in Fe-based semiconductors one may observe strong paramagnetic absorption compared to both EDSR's. In this case another interesting effect may be observed—the interference of electric dipole and magnetic dipole spin transitions, similar to that in InSb.[31–33]

V. CONCLUSION

The theory presented shows that in semimagnetic semiconductors there exists a specific mechanism of electric dipole spin transitions—exchange interaction between spins of a carrier and the magnetic impurities. The absorption intensity produced by this mechanism may be comparable to that produced by spin-orbit interaction in hexagonal crystals and exceeds it in the wide-gap cubic semiconductors. Magnetic field, frequency, polarization, temperature, and angular dependences of exchange and spin-orbit EDSR's are essentially different, which allows one easily to discriminate between them experimentally.

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