

Exchange mechanism for the electric-dipole spin resonance in semimagnetic semiconductors

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A mechanism for electric-dipole processes involving a spin flip in a semimagnetic semiconductor is proposed. The mechanism involves an exchange interaction of a carrier with a magnetic impurity. Its contribution to the absorption is comparable to that from the spin-orbit interaction, but it has several distinctive features.

An electric-dipole spin (combined) resonance (EDSR) has been predicted theoretically by Rashba.^{1,2} This resonance would be caused by inhomogeneities which lower the symmetry of a crystal^{3,4} a deviation of the band energy from a parabolic nature,⁵ or a spin-orbit term in the Hamiltonian,² which takes its simplest form,

$H_{so} = \alpha(\vec{\sigma} \times \mathbf{k})\mathbf{c}$, in hexagonal semiconductors.⁶ Here α is the spin-orbit coupling constant, the operators $\vec{\sigma}$ and \mathbf{k} represent the spin (the Pauli matrices) and momentum of the electron, and \mathbf{c} is a unit hexagonal axis. In recent years, an electric-dipole spin resonance has been observed experimentally⁷ in a crystal of this structure for an electron bound at a shallow donor in $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$, $x \sim 0.1$, which is a semimagnetic semiconductor that contains paramagnetic Mn^{2+} ions.^{8,9} This resonance was interpreted on the basis of the Hamiltonian H_{so} which we wrote above. However, we wish to call attention to the circumstance that another mechanism might also be responsible for electric-dipole transitions accompanied by a spin flip in semimagnetic semiconductors. This other mechanism would involve an exchange interaction of the spin of the carrier with spins of a paramagnetic impurity. The Hamiltonian for such an interaction is^{8,10}

$$H_{ex} = -J \sum_j \left(\frac{\vec{\sigma}}{2} \cdot \mathbf{S}_j \right) \delta(\mathbf{r} - \mathbf{R}_j) = H_d + H_{nd}, \quad (1)$$

$$H_{nd} = -\frac{1}{2} J \sum_j (\sigma_+ S_{j-} + \sigma_- S_{j+}) \delta(\mathbf{r} - \mathbf{R}_j),$$

where J is the exchange constant, the operator \mathbf{S}_j represents the spin of an impurity at point \mathbf{R}_j , $\sigma_{\pm} = (\sigma_x \pm i\sigma_y)/\sqrt{2}$, and there is a corresponding expression for $S_{j\pm}$. The diagonal part, H_d , which is usually taken into consideration in the functions of the null Hamiltonian, is known (Ref. 10, for example) to be responsible for the "giant" spin splitting $\hbar\omega_s$ (a large g -factor, $\sim 10^2$, in the linear-magnetization region) of the levels of a carrier in a magnetic field $\vec{\mathcal{H}} \parallel z$. The nondiagonal part, H_{nd} , like H_{so} , mixes the wave functions with different spin directions and may be the cause of the electric-dipole spin resonance. Under the inequality $na^3 \gg 1$, where $n = x/\Omega$ is the concentration of the paramagnetic impurities (Ω is the volume of the unit cell), and a is a typical radius (the first Bohr radius for a Coulomb center) of the wave function of carrier, H_{nd} is small and can be dealt with by perturbation theory.

For a carrier which is localized at a shallow hydrogen-like center, if we ignore its effect on the state of the magnetic impurity, i.e., if we ignore magnetopolaron effects ($|J|a^{-3} \ll \hbar\omega_m$), the matrix element of the electric-dipole transition is

$$M_{ex} = \sum_{\nu} \left[\frac{\langle f | \mathbf{e} \cdot \mathbf{r} | \nu \rangle \langle \nu | H_{nd} | i \rangle}{E_i - E_{\nu} - \hbar\omega'_s} + \frac{\langle f | H_{nd} | \nu \rangle \langle \nu | \mathbf{e} \cdot \mathbf{r} | i \rangle}{E_f - E_{\nu} + \hbar\omega'_s} \right], \quad (2)$$

where the eigenfunctions correspond to the operator $H_0 = H_e + H_d + H_m$, where H_e and $H_0 = H_e + H_d + H_m$, H_e and $H_m = \hbar\omega_m \sum_j S_{jz}$ are the Hamiltonians of a carrier with a spin and of noninteracting paramagnetic impurities in a magnetic field, E_{ν} are the hydrogen-like energy levels, and $\hbar\omega'_s = \hbar\omega_s \mp \hbar\omega_m$ [here and in Eq. (3) below, the upper sign is to be used if the g -factor of the carrier is positive, and the lower sign if negative]. Ignoring the interimpurity interaction is justified when the concentration n is not high or the temperature is not too low. In a semimagnetic semiconductor the condition $R^* > \hbar\omega_s \gg \hbar\omega_c$ holds (R^* is the effective Rydberg, and ω_c is the cyclotron frequency) in transitions between states of the discrete spectrum because of the large

g-factor. It thus becomes possible to simplify the calculations by omitting the term in H_e which is quadratic in $\vec{\mathcal{H}}$ (Ref. 11). In this case, M_{ex} is an integral with a Coulomb Green's function and can be evaluated.

The absorption intensity I_{ex} is determined by $\langle |M_{ex}|^2 \rangle$, which is understood as a statistical average both over the states of the magnetic impurity (a temperature $T \approx \hbar\omega_m$) and over \mathbf{R}_j . The intensity I_{ex} increases resonantly, in the same fashion as the intensity (I_{so}) of the spin-orbit electric-dipole spin resonance.^{6,11} As the resonance region is approached (e.g., for transitions between the lowest-lying spin sublevels of a carrier as $\hbar\omega'_s \rightarrow E_{2p} - E_{1s}$), the intensity ratio can be estimated as the ratio of the largest terms in (2) and in the corresponding expression for I_{so} . At the maximum value of the anisotropic I_{so} , this procedure yields

$$\frac{I_{ex}}{I_{so}} = \frac{\langle |M_{ex}|^2 \rangle}{\langle |M_{so}|^2 \rangle} = \frac{27}{2^{10} \pi} \frac{\langle S_{\pm} S_{\mp} \rangle}{na^3} \left(\frac{Jn}{\alpha a^{-1}} \right)^2, \quad (3)$$

where

$$\langle S_{\pm} S_{\mp} \rangle = \frac{\text{Sp} \{ S_{j\pm} S_{j\mp} \exp(-H_m/T) \}}{\text{Sp} \{ \exp(-H_m/T) \}} \quad (4)$$

is the same for all impurities. In the case of an electric-dipole spin resonance involving a donor in $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Se}$, with the parameter values from Refs. 10 and 11, ratio (3) is on the order of unity.

There are some distinctions between the mechanism proposed here for an electric-dipole spin resonance and the spin-orbit mechanism:

1. The intensity is independent of the orientation of $\vec{\mathcal{H}}$ with respect to the crystal axes (isotropy).
2. The transition energy for the exchange mechanism is, within quantities $\sim J/a^3$, equal to $\hbar\omega'_s$, differing slightly from the energy ($\hbar\omega_s$) of the spin-orbit electric-dipole spin resonance. It is possible that this circumstance may explain the observation of two closely spaced peaks in the experiments of Ref. 7.
3. Under the conditions $\hbar\omega_s, \hbar\omega'_s \ll E_{2p} - E_{1s}$, we have $I_{so} \sim \omega_s^3$ and $I_{ex} \sim \omega'_s$. The reason for the deviation from the corollary of Kramer's theorem⁶ in the last relation ($M_{ex} \neq 0$ as $\hbar\omega_s \rightarrow 0$) is that the transition in the system consisting of the electron and the paramagnetic impurity does not occur between Kramer's levels.
4. As can be seen from the form of H_{so} , the spin-orbit mechanism allows transitions between donor levels of the same parity (e.g., $s \rightarrow s, s \rightarrow d$). The mechanism proposed here is free of this restriction, allowing other transitions also (e.g., $s \rightarrow p$). Furthermore, it allows transitions for any polarization of the light.

Since it is difficult to experimentally identify each mechanism on the basis of the transition energy because of the small value of $\hbar\omega_m$, it might be possible to separate

mechanisms through a study of the polarization, frequency, or angular dependence of the intensity of the electric-dipole spin resonance as the angle between \vec{H} and \mathbf{c} is varied.

Another point which should be noted is that the exchange mechanism proposed here should lead to electric-dipole transitions with spin flip even in the case of free carriers not bound to impurity centers. In this case (in contrast with the spin-orbit mechanism), however, the transitions would not be resonant, since in an interaction with a magnetic impurity the carrier not only flips the spin but also, being scattered, changes momentum, so that the resonant peak is spread out into a broad band.

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