Scattering by atomic spins and magnetoresistance in dilute magnetic semiconductors

M. Foygel and A. G. Petukhov

Physics Department, South Dakota School of Mines and Technology, Rapid City, South Dakota 57701, USA (Received 20 February 2007; revised manuscript received 3 July 2007; published 6 November 2007)

We studied electrical transport in magnetic semiconductors, which is determined by scattering of free carriers off localized magnetic moments. We calculated the scattering time and the mobility of the majorityand minority-spin carriers with both the effects of thermal spin fluctuations and of spatial disorder of magnetic atoms taken into account. We discuss the role of the above effects on magnetoresistance of nondegenerate semiconductors where magnetic impurities are electrically active or neutral. The application of the external magnetic field suppresses the thermodynamic spin fluctuations thus promoting negative magnetoresistance. Simultaneously, scattering off the built-in spatial fluctuations of the atomic spin concentrations may increase with the magnetic field. The latter effect is due to the growth of the magnitude of random local Zeeman splittings with the magnetic field. It promotes positive magnetoresistance. The enhancement of spin-dependent scattering by the external magnetic field is especially large in the materials with electrically active magnetic impurities.

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I. INTRODUCTION

Dilute magnetic semiconductors (DMS) are materials of great promise in modern technology because they combine semiconductor transport and magnetic properties allowing reach and physically meaningful interplay between them.^{1,2} Basically, there are two types of wide bandgap semiconductor alloys in which cations are substituted by randomly distributed magnetic atoms, such as Mn.^{1,3} In some of them, such as (II,Mn)VI (Cd_{1-x}Mn_xTe, for instance) the substituting magnetic impurity Mn is isoelectronic. We will call these materials type-I DMS. Here, the magnetic impurities are not electrically active. However, they can effectively modify electronic transport⁴⁻⁶ and magnetism^{7,8} due to exchange coupling of the free carrier spins to the spins of magnetic atoms. As a result, scattering of the free carriers by the localized magnetic moments determines their mobility that is substantially spin and, therefore, magnetic-field dependent. It leads to giant magnetoresistance (MR), positive or negative.^{4,5,9} (If carriers are localized, the above coupling causes spin-polaron effects that substantially affect the magnetotransport properties of type-I DMS.^{10,11})

A more complex situation occurs in type-II DMS, such as (III,Mn)V or (IV,Mn) alloys where Mn atoms serve as acceptors. Examples are $Ga_{1-x}Mn_xAs$ (Refs. 12 and 13) and $Ge_{1-x}Mn_x$ (Refs. 14–17) magnetic semiconductors. Here, contrary to the type-I DMS, atomic-spin scattering essentially involves charged impurities, a substantial fraction of which are magnetic interstitials. In these materials, both the effects of the atomic-spin scattering and the scattering off the charged impurities are related to each other. They cannot be treated by means of a simple Matthiessen's rule.¹⁸ In addition, the magnetic and transport properties of DMS, such as Ge:Mn, strongly depend on disorder in the spatial distribution of magnetic impurities.^{15,17,19,20}

In this paper we will concentrate mainly on spindependent scattering which determines mobility of free carriers in DMS taking into account the spin-disorder effects that are intrinsic for these materials. There are two sources of the spin-disorder effects in question: (a) the thermodynamic fluctuations of atomic spins,^{21–23} which are present even in the ordered type-I magnetic materials with x=1,^{21,22} and (b) the built-in spatial fluctuations of local concentrations of the magnetic impurities,²⁴ which are substantial for type-II DMS even in the absence of magnetic field due to the long-range nature of Coulomb interaction.

The theory of spin-disorder scattering off the thermodynamic fluctuations of the local magnetization due to atomic moments of the magnetic atoms has been developed by de Gennes and Friedel for magnetic metals²¹ and for ordered magnetic semiconductors by Haas.²² In particular, they showed the application of an external magnetic field freezes out the above fluctuations thus leading to negative MR. Michel et al.²⁴ ignored such effects but took into account scattering off the built-in fluctuations of the local concentration of magnetic atoms in type-I DMS. They demonstrated that the field-induced decrease in the mobility associated with these inhomogeneities can be partially responsible for positive MR in these DMS. It is evident, however, that both the above scattering effects should be treated on the equal footing because they have a common source-magnetic atoms. Such a treatment is especially important because, first, as has been just mentioned, the spin-disorder effects in question usually give competing contributions to MR of DMS and, secondly, the very same magnetic atoms in type-II DMS, due to their charge, serve as a very powerful source of spin-independent scattering that actually defines transport properties in zero magnetic field. In this paper we develop a simple approach that allows us to consistently tackle a nontrivial problem of the mobility of the majority and minority spin carriers by taking into account the exchange, Coulomb, and deformation effects in scattering by the very same magnetic atoms.

In Sec. II we will calculate the relaxation time due to scattering by magnetic impurities, which determines the mobility of free carriers to be analyzed in Sec. III. In Sec. IV we will calculate the MR of different types DMS, which is in a substantial part defined by the spin-disorder effects associated with scattering of the free carriers off magnetic impurities. Here, in particular, we will compare the results of our calculations with the recent experimental data on MR (Ref. 25) in Ge:Mn in order to clarify the nature of giant positive MR in these compounds.

II. RELAXATION TIME

Let us consider a charge carrier in an extended state $|\phi_{\mathbf{k}}(\mathbf{r})X\alpha\rangle$, where $\phi_{\mathbf{k}}(\mathbf{r})=|\mathbf{k}\rangle$ is the Bloch function of the band state of energy $\epsilon_{\mathbf{k}}$ and of wave vector \mathbf{k} , X^{\pm} is the electron spin up (+) or down (-) function, and $|\alpha\rangle$ is an eigenstate of the Hamiltonian of the atomic spins with a temperature dependent probability w_{α} for the state $|\alpha\rangle$ to occur. (For the sake of simplicity we will ignore a complex nature of the angular momentum structure of the energy spectrum of such free carriers, such as holes^{7,18} in semiconductors, just assuming that the carriers possess spin $s = \pm 1/2$.) For magnetic impurities randomly located at points \mathbf{R}_{i} , the probability of their given configuration

$$dF(\mathbf{R}_1,\ldots,\mathbf{R}_M) = \prod_{i=1}^{M} \frac{d\mathbf{R}_i}{\Omega},$$
 (1)

where Ω is the volume of the system. At a given temperature, the magnetization of the system

$$M = xNg\mu_B \overline{\langle J_z \rangle} = M_{\text{sat}} \overline{\langle J_z \rangle} / J$$
 (2)

is expressed in terms of the average projection of the component of the atomic spin \mathbf{J}_i , located at point \mathbf{R}_i , along the direction *z* of the magnetization. Here *N* is the concentration of the sites in the (sub)lattice that contains magnetic atoms of the fractional concentration $x=N_m/N \leq 1$, *g* is the Lande factor of the magnetic-atom spin, μ_B is the Bohr magneton, $M_{\text{sat}}=xNg\mu_B J$ is the saturation magnetization; the brackets represent thermal averaging while the bar represents the averaging over the spatial configurations of magnetic atoms

$$\overline{\langle J_z \rangle} = \int dF(\mathbf{R}_1, \dots, \mathbf{R}_M) \sum_{\alpha} w_{\alpha} \langle \alpha | J_{iz} | \alpha \rangle = JB_J(y).$$
(3)

Here $B_J(y)$ is the Brillouin function¹⁸ of the atomic spin *J* and of the argument *y* to be found from the mean-field equation,^{1,2} which depends on the absolute temperature *T* and external magnetic field *H*. (For the DMS well into paramagnetic phase $y=g\mu_BHJ/T$.) Throughout this paper, the temperature *T* is measured in the energy units $(k_B=1)$.

We will start our consideration of a scattering time from the simplest case of a free carrier coupled to randomly distributed nonmagnetic impurities by means of the nonexchange interaction

$$U_{NM}(\mathbf{r}) = -\sum_{i} V_{NM}(\mathbf{r} - \mathbf{R}_{i}).$$
(4)

The probability, per unit time, of a free-carrier transition from a state with the wave vector \mathbf{k} to a state \mathbf{k}' , which is averaged over all possible configurations of impurities, is given by

$$\overline{P_{NM}(\mathbf{k},\mathbf{k}')} = \frac{2\pi}{\hbar} \overline{|\langle \mathbf{k} | U_{NM} | \mathbf{k}' \rangle|^2} \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}).$$
(5)

Assuming the isotropy of the dispersion law ϵ_k one can express the corresponding relaxation time that appears in the Boltzmann transport equation in terms of the above transition probability (5) as follows:¹⁸

$$\frac{1}{\tau_{\mathbf{k}}^{NM}} = \frac{\Omega}{(2\pi)^3} \int d\mathbf{k}' (1 - \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') \overline{P_{NM}(\mathbf{k}, \mathbf{k}')}.$$
 (6)

Then ignoring a spatial dependence of the periodic parts of the Bloch functions, it is easy to show that²⁶

$$\frac{1}{\tau_{\mathbf{k}}^{NM}} = \frac{1}{(2\pi)^2 \hbar} \int d\mathbf{k}' (1 - \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') \Psi_{NM}(|\mathbf{k} - \mathbf{k}'|) \,\delta(\boldsymbol{\epsilon}_{\mathbf{k}} - \boldsymbol{\epsilon}_{\mathbf{k}'}),$$
(7)

where

$$\Psi_{NM}(\mathbf{k}) = \int d\mathbf{r} \exp(-i\mathbf{k} \cdot \mathbf{r}) \overline{U_{NM}(\mathbf{r}) U_{NM}(\mathbf{0})}$$
(8)

is the Fourier transform of a pair correlation function of the nonexchange part (4) of the random impurity potential. For a simple isotropic dispersion law $\epsilon_{\mathbf{k}} = \hbar^2 k^2 / 2m$ with an effective mass *m*, Eq. (7) yields

$$\frac{1}{\tau_k^{NM}} = \frac{m}{4\pi(\hbar k)^3} \int_0^{2k} dz z^3 \Psi_{NM}(z) \,. \tag{9}$$

In magnetic semiconductor, the free carrier is coupled to the randomly distributed magnetic atoms by the following interaction:

$$U_m(\mathbf{r}) = -\sum_i \left[\mathbf{s} \cdot \mathbf{J}_i U_{\text{ex}}(\mathbf{r} - \mathbf{R}_i) + V(\mathbf{r} - \mathbf{R}_i) \right], \quad (10)$$

where $U_{\text{ex}}(\mathbf{r}) \approx \beta_{\text{ex}} \delta(\mathbf{r})$ is the exchange coupling potential strongly localized within the unit cell containing a magnetic atom ($\beta_{\text{ex}} N \approx 1 \text{ eV}$),^{2,7} **s** is the electron spin; $V(\mathbf{r})$ is a nonexchange part of the magnetic-impurity potential of the Coulomb and/or deformation nature. The probability per unit time for an electron from a state with the wave vector **k** and with spin up (+) or down (-) to get transferred to a state with **k**' and with spin up (+) or down (-) while the state of the atomic spins undergoes transition from α to α' is²²

$$P_{m}(\mathbf{k}, \pm, \alpha; \mathbf{k}', \pm, \alpha')$$

$$= \frac{2\pi}{\hbar} |\langle \phi_{\mathbf{k}}(\mathbf{r}) X^{\pm} \alpha | U_{m} | \phi_{\mathbf{k}'}(\mathbf{r}) X^{\pm} \alpha' \rangle|^{2}$$

$$\times \delta(\epsilon_{\mathbf{k}}^{\pm} + \epsilon_{\alpha} - \epsilon_{\mathbf{k}'}^{\pm} - \epsilon_{\alpha'}). \qquad (11)$$

Here for the simple isotropic conduction band

$$\boldsymbol{\epsilon}_{k}^{\pm} = \boldsymbol{\epsilon}_{0}^{\pm} + \frac{\hbar^{2}k^{2}}{2m} \tag{12}$$

and $\Delta = \overline{\epsilon_0} - \overline{\epsilon_0}^+$ is the Zeeman splitting of the electron spinsplit conduction subbands.

Then, after thermal averaging over the initial spin states, summation over the final spin states, and averaging over the PHYSICAL REVIEW B 76, 205202 (2007)

impurity configurations, one can calculate, in the first Born approximation, the inverse relaxation time for an electron with the wave vector \mathbf{k} and the spin up (+) or down (-) as follows:

$$\frac{1}{\tau_{\mathbf{k}}^{\pm}} = \frac{\Omega}{(2\pi)^{3}} \int d\mathbf{k}' (1 - \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') \sum_{\alpha'} \left[\overline{\langle P_{m}(\mathbf{k}, \pm, \alpha; \mathbf{k}', +, \alpha') \rangle} + \overline{\langle P_{m}(\mathbf{k}, \pm, \alpha; \mathbf{k}', -, \alpha') \rangle} \right] \\
= \frac{1}{(2\pi)^{2} \hbar \Omega} \int d\mathbf{k}' (1 - \hat{\mathbf{k}} \cdot \hat{\mathbf{k}}') \sum_{ij} \overline{e^{i(\mathbf{k}' - \mathbf{k})(\mathbf{R}_{i} - \mathbf{R}_{j})}} \left\{ \left[\frac{1}{4} |U_{ex}(\mathbf{k} - \mathbf{k}')|^{2} (\langle J_{iz}J_{jz} \rangle - \langle J_{iz} \rangle \langle J_{jz} \rangle) \right] \\
+ \overline{\left[V(\mathbf{k} - \mathbf{k}') \pm \frac{1}{2} U_{ex}(\mathbf{k} - \mathbf{k}') \overline{\langle J_{z} \rangle} \right]^{2}} \left] \delta(\boldsymbol{\epsilon}_{\mathbf{k}}^{\pm} - \boldsymbol{\epsilon}_{\mathbf{k}'}^{\pm}) + \overline{\frac{1}{4} |U_{ex}(\mathbf{k} - \mathbf{k}')|^{2} (\langle J_{ix}J_{jx} \rangle + \langle J_{iy}J_{jy} \rangle)} \right] \delta(\boldsymbol{\epsilon}_{\mathbf{k}}^{\pm} - \boldsymbol{\epsilon}_{\mathbf{k}'}^{\pm}) \right\}.$$
(13)

where $U_{\text{ex}}(\mathbf{k})$ and $V(\mathbf{k})$ are the Fourier transforms of the exchange and nonexchange parts of the magnetic impurity potential. Here the terms on the right-hand side, which involve the atomic spins correlation functions, are responsible for scattering off the thermodynamic fluctuations of atomic spins. By virtue of the fluctuation-dissipation theorem²⁷ the first of them can be expressed in terms of the static value of the *z* component (longitudinal component) of the magnetic susceptibility $\chi_z = \chi_{\parallel}$:

$$\Psi_{\parallel}(\mathbf{k} - \mathbf{k}') = \frac{1}{4\Omega} \sum_{i,j} \\ \times \overline{e^{i(\mathbf{k}' - \mathbf{k})(\mathbf{R}_i - \mathbf{R}_j)}} |U_{\text{ex}}(\mathbf{k} - \mathbf{k}')|^2 \langle \langle J_{iz} J_{jz} \rangle - \langle J_{iz} \rangle \langle J_{jz} \rangle) \\ = \frac{T |U_{\text{ex}}(\mathbf{k} - \mathbf{k}')|^2 \chi_{\parallel}(\mathbf{k} - \mathbf{k}')}{4(g\mu_B)^2}.$$
(14)

Similarly,

$$\Psi_{\perp}(\mathbf{k} - \mathbf{k}') = \frac{1}{4\Omega} \sum_{i,j} \overline{e^{i(\mathbf{k}' - \mathbf{k})(\mathbf{R}_i - \mathbf{R}_j)} |U_{\text{ex}}(\mathbf{k} - \mathbf{k}')|^2 \langle J_{ix} J_{jx} \rangle}$$
$$= \frac{1}{4\Omega} \sum_{i,j} \overline{e^{i(\mathbf{k}' - \mathbf{k})(\mathbf{R}_i - \mathbf{R}_j)} |U_{\text{ex}}(\mathbf{k} - \mathbf{k}')|^2 \langle J_{iy} J_{jy} \rangle}$$
$$= \frac{T\chi_{\perp}(\mathbf{k} - \mathbf{k}') |U_{\text{ex}}(\mathbf{k} - \mathbf{k}')|^2}{4(g\mu_B)^2}, \qquad (15)$$

where $\chi_{\perp} = \chi_x = \chi_y$ are transversal components of the magnetic susceptibility in a direction perpendicular to magnetic field. The above expressions (14) and (15) for the correlation functions defining the transport properties of DMS can be justified for the materials in paramegnetic phase far from the ferromagnetic phase transition region.^{28,29}

The second term on the right-hand side of Eq. (13) represents scattering off a random built-in potential of magnetic atoms due to the spatial fluctuations of their local concentrations. Similar to Eqs. (7) and (8) describing scattering by nonmagnetic atoms, it can be expressed in terms of the Fourier transform of the corresponding correlation function

$$\Psi_{m}(|\mathbf{k} - \mathbf{k}'|) = \frac{1}{\Omega} \sum_{i,j} \overline{e^{i(\mathbf{k}' - \mathbf{k})(\mathbf{R}_{i} - \mathbf{R}_{j})}} |V(\mathbf{k} - \mathbf{k}') \pm \frac{1}{2} U_{\text{ex}}(\mathbf{k} - \mathbf{k}') \overline{\langle J_{z} \rangle}|^{2}.$$
(16)

It should be mentioned here that in the case of nondegenerate semiconductors, where the typical wave numbers of scattered free carriers are small, one can usually ignore the dispersion of the Fourier transforms of the correlation functions described by Eqs. (8) and (14)–(16). The exclusion is to be made for the charged impurities, magnetic and/or nonmagnetic, when the corresponding Fourier transforms $V_{NM}(\mathbf{k})$ and $V(\mathbf{k})$ involved diverge at small k.

A. Type-I DMS

We can further simplify expression (13) for the relaxation time in the type-I magnetic semiconductor compounds where the magnetic centers are isoelectronic. In this case, the potential of such a center consists of the exchange and deformation components, both short-range ones

$$U_m^{(i)}(\mathbf{r}) = -\left[\mathbf{s} \cdot \mathbf{J}_i U_{\text{ex}}(\mathbf{r} - \mathbf{R}_i) + V(\mathbf{r} - \mathbf{R}_i)\right]$$
$$= -\left[\mathbf{s} \cdot \mathbf{J}_i \beta_{\text{ex}} + \beta_{\text{def}}\right] \delta(\mathbf{r} - \mathbf{R}_i), \qquad (17)$$

where β_{ex} is the exchange coupling constant and, for ternary solid solutions, $\beta_{def} = N^{-1} dE_C / dx$ is the deformation potential constant of the relevant band edge E_C .^{24,26} So, exactly as for any mixed semiconductor compound (see Ref. 26), the Fourier transform of the potential correlation function

$$\Psi_m(k) = Nx(1-x) \left| \beta_{\text{def}} \pm \frac{1}{2} \beta_{\text{ex}} \overline{\langle J_z \rangle} \right|^2, \quad (18)$$

where $\overline{\langle J_z \rangle}$ is given by Eq. (3). The magnetic-field dependence $\overline{\langle J_z \rangle}$ follows that of the magnetization (2). As a result,



 $a) \mathbf{H} = \mathbf{0} \qquad b) \mathbf{H} \neq \mathbf{0}$

FIG. 1. (Color online) Built-in magnetic-impurity potential in DMS in the absence (a) and in the presence (b) of external magnetic field for the case when $b = \beta_{def} / \beta_{ex} > 1$.

using simple isotropic dispersion law (12) and relations similar to Eq. (9), Eqs. (3) and (14)-(17) yields

$$\frac{1}{\tau_l^{\pm}(k)} = \frac{m\beta_{\text{ex}}^2 k}{\pi\hbar^3} \left\{ \frac{T}{(2g\mu_B)^2} [\chi_{\parallel} + 2\chi_{\perp}F_k^{\pm}] + Nx(1-x) \right. \\ \left. \times \left| \frac{\beta_{\text{def}}}{\beta_{\text{ex}}} \pm \frac{JM}{2M_{\text{sat}}} \right|^2 \right\}.$$
(19)

Here

$$F_{k}^{\pm} = \frac{\rho(\boldsymbol{\epsilon}_{k}^{\pm} - \boldsymbol{\epsilon}_{0}^{\mp})}{\rho(\boldsymbol{\epsilon}_{k}^{\pm} - \boldsymbol{\epsilon}_{0}^{\pm})} \theta(\boldsymbol{\epsilon}_{k}^{\pm} - \boldsymbol{\epsilon}_{0}^{\mp}), \qquad (20)$$

where $\theta(x)$ is the Heaviside unit step function such that $\theta(x)=1$ if $x \ge 1$ and $\theta(x)=0$ if x < 0;

$$\epsilon_0^- - \epsilon_0^+ = \overline{\Delta}(H) = xN\beta_{\text{ex}}JB_J(y) + g^*\mu_B H = \frac{\beta_{\text{ex}}M(H)}{g\mu_B} + g^*\mu_B H$$
(21)

is the average Zeeman band splitting¹⁰ and $\rho(\epsilon)$ is the density of states (DOS). For the simple parabolic band (12), $\rho(\epsilon) \propto \epsilon^{1/2}$ so that

$$F^{\pm} = F(z_{\pm}) = z_{\pm}^{1/2} \,\theta(z_{\pm}^{1/2}), \qquad (22)$$

where $z_{\pm} = 1 \pm 2m\bar{\Delta}/\hbar^2 k^2$. [In Eq. (21) g^* is the electron g factor.]

In the above expression (19) for the relaxation time of the type-I DMS, the term in the square brackets is responsible for scattering off the thermal fluctuations of atomic spins. In particular, the term with χ_{\parallel} describes the scattering processes that go without spin flip. The term with χ_{\perp} takes into account the scattering accompanied by the double spin-flip processes. For the majority-spin carriers (+), the latter processes (+) \rightarrow (-) gradually disappear with the increase of the applied magnetic field because the energies of the final-state subband progressively exceed that of the initial-state subband. As a result, the corresponding transitions become energetically less favorable.

The last term on the right-hand side of Eq. (19) describes scattering off the spatial fluctuations of the local concentrations of magnetic atoms (see Fig. 1). For the ordered magnetic semiconductors, such as EuSe or ErAs, where magnetic atoms form a regular sublattice, x=1, this term disappears. Then Eq. (19) coincides with one obtained by Haas.²² Meanwhile, in the limit of the saturation magnetic fields, when the thermal fluctuations are frozen out because both $\chi_{\parallel}(H) = \partial M / \partial H$ and $\chi_{\perp}(H)$ (Refs. 10 and 22) tend to zero, for $|\beta_{def}/\beta_{ex}| \ge J$ we recover the expression for the scattering time in disordered nonmagnetic alloys.²⁶

B. Type-II DMS

In these materials, both the magnetic impurities and the compensating nonmagnetic centers are charged, so that the spin-independent components of their potentials are of the screened Coulomb form $V_C(r)$ for which the Fourier transform¹⁸

$$V_C(k) = -\frac{4\pi Z e^2}{\kappa (k^2 + r_0^{-2})}.$$
(23)

Here Ze is the charge of a center, κ is the dielectric constant and r_0 is the screening length. We will use this dependence when calculating the Fourier transforms of the relevant correlation functions (16) and (8) due to magnetic and nonmagnetic impurities, respectively,

$$\Psi_{NM}(k) = N_{NM} \left[\frac{4\pi Z_{NM} e^2}{\kappa (k^2 + r_0^{-2})} \right]^2,$$
(24)

$$\Psi_m(k) = N_m \left[\frac{4\pi Z_m e^2}{\kappa (k^2 + r_0^{-2})} \pm \frac{\beta_{\rm ex} JM}{2M_{\rm sat}} \right]^2.$$
(25)

Here $Z_i e$ and N_i are the charge and concentration of the magnetic (m) and nonmagnetic (NM) impurities. It is easy to obtain the following expression for the relaxation time in the type-II DMS with large enough screening length $(kr_0 \ge 1)$:

$$\frac{1}{\tau_{II}^{\pm}(k)} = \frac{1}{\tau_{k}^{\pm}} + \frac{1}{\tau_{k}^{(NM)}} = \frac{m\beta_{ex}^{2}k}{\pi\hbar^{3}} \left\{ \frac{T}{(2g\mu_{B})^{2}} [\chi_{\parallel} + 2\chi_{\perp}F_{k}^{\pm}] + \frac{N_{m}JM}{2M_{sat}} \left(\frac{JM}{2M_{sat}} \pm \frac{4\pi Z_{m}e^{2}}{\beta_{ex}\kappa k^{2}} \right) + 2\pi N^{*} \left(\frac{e^{2}}{\beta_{ex}\kappa k^{2}} \right)^{2} \ln(2kr_{0}) \right\},$$
(26)

where $N^* = N_m Z_m^2 + N_{NM} Z_{NM}^2$ is the effective concentration of the charged impurities and F_k^{\pm} is given by Eq. (20). Here, as in Eq. (19), the first term in the braces is responsible for scattering off the thermodynamic fluctuations of the atomic spins. The second and the third terms describe the input from the built-in fluctuations of the local impurity potential. In particular, the last term is due to scattering off charged impurities, magnetic and nonmagnetic. (For these materials we obviously have ignored the short-range deformation potential.) It should be noted that the presence of an "interference" term in the second term in the braces violates empirical Matthiesen's rule¹⁸ because the impurity scattering processes involve the Coulomb and magnetic forces that originate from the same atoms. It can be seen that these processes are not independent even within the first Born approximation.

III. CALCULATING MOBILITY

If the relaxation time is known, one can use the standard approach to calculate the mobilities of the majority (+) and minority (-) spin carriers²²

$$\mu^{\pm} = \frac{q\hbar}{3m^2} \frac{\sum_{k} k^2 (\partial f_k^{\pm} / \partial \epsilon_k^{\pm}) \tau_k^{\pm}}{\sum_{k} f_k^{\pm}}, \qquad (27)$$

where f_k^{\pm} are the Fermi distribution functions for the spinsplit subbands and $q=\pm e$ is the carrier charge. By means of Eq. (19) we find that for type-I nondegenerate DMS

$$\mu_{I}^{\pm} = \mu_{0}^{(I)} \int_{0}^{\infty} dt \exp(-t) t \left\{ \frac{T}{(2g\mu_{B})^{2}N} [\chi_{\parallel} + 2\chi_{\perp}F(1 \mp \delta/t)] + x(1-x) \left| \frac{\beta_{\text{def}}}{\beta_{\text{ex}}} \pm \frac{JM}{2M_{\text{sat}}} \right|^{2} \right\}^{-1},$$
(28)

where

$$\mu_0^{(I)}(T) = \frac{2(2\pi)^{1/2}q\hbar^4}{3m^{5/2}N\beta_{\rm ex}^2 T^{1/2}}.$$
(29)

Here $\delta = (\epsilon_0^- - \epsilon_0^+)/T = \overline{\Delta}/T$ is the dimensionless average Zeeman band splitting and F(z) is given by Eq. (22). With the well known from the Hall effect theory coefficient $\gamma_H = \mu_H/\mu = 3\pi/8$ for the scattering by isoelectronic impurities,³⁰ in the limit of the saturating magnetic fields and for $|\beta_{def}/\beta_{ex}| \ge J$, one can easily obtain by means of Eqs. (28) and (29) the expression for the Hall mobility μ_H in mixed nonmagnetic alloys.²⁶ And again, in the limiting case of the ordered magnetic semiconductors (x=1) we retrieve the expression for the mobility μ_I^+ obtained by Haas.²²

Figure 2 shows the mobilities of spin-up and spin-down carriers in the type-I DMS calculated by means of Eq. (28) for different values of parameter $b = \beta_{def}/\beta_{ex}$. We used $N|\beta_{ex}|=1 \text{ eV},^{7,8} J=5/2, x=0.02$, and T=50 K in our calculations. It can be seen that, as a rule, the mobility of minority-spin carriers drops while that of the majority-spin carriers rises with the application of the magnetic field. The mobilities at low magnetic fields are dominated by scattering off the thermodynamic fluctuations of the atomic spins, that are getting suppressed by magnetic field. This suppression is especially large for the majority-spin carriers because for them the input from the double spin-flip $(+) \Rightarrow (-)$ processes of scattering by the thermal fluctuations are substantially reduced with the application of the external magnetic field when these processes are accompanied by the absorption of the increasingly greater amounts of thermal energy. Contrary to that, the spin-up mobility drops and spin-down mobility rises with magnetic field only if the deformation potential constant β_{def} is large and have the same sign as the exchange coupling constant β_{ex} . In this case the zero-magnetic-field mobility is governed by scattering off the build-in fluctua-



FIG. 2. (Color online) Mobility of the minority (a) and majority (b) spin carriers as a function of magnetic field for different values of the parameter $b = \beta_{def} / \beta_{ex}$.

tions of the deformation potential of magnetic impurities [Fig. 1(a)]. The application of the magnetic field increases the amplitude of the impurity potential fluctuations for the majority-spin carriers and decreases the above amplitude for the minority-spin carriers [Fig. 1(b)], which explains the calculated dependencies.

For the type-II nondegenerate DMS with large enough screening length $(8mTr_0^2/\hbar^2 \ge 1)$, Eqs. (26) and (27) yield

$$\mu_{\rm II}^{\pm} = \mu_0^{(\rm II)} \int_0^\infty dt \, \exp(-t) t \Biggl\{ \frac{T}{(2g\mu_B)^2 N_m} [\chi_{\parallel} + 2\chi_{\perp}] + \frac{JM}{2M_{\rm sat}} \Biggl(\frac{JM}{2M_{\rm sat}} \pm \frac{Z_m T_0}{Tt} \Biggr) + \frac{N^*}{2\pi N_m} \Biggl(\frac{T_0}{2Tt} \Biggr)^2 \ln(8mTtr_0^2/\hbar^2) \Biggr\}^{-1}.$$
(30)

Here $\mu_0^{(II)} \sim T^{-1/2}$ is given by Eq. (29) where *N* is to be changed for the concentration of magnetic impurities N_m . In Eq. (30), we introduced parameter $T_0 = 2\pi e^2 \hbar^2 / (\beta_{ex} \kappa m)$, which is of the order of 1 eV, so that at practically all temperatures $T \ll T_0$. Therefore, for not very large atomic spins $J \simeq 1$ the last term in the expression in the braces on the right-hand side of Eq. (30) is much larger than the previous two. As a result, for $J \simeq 1$ the mobility here is dominated by scattering off charged impurities and is approximately described by^{30,31}

$$\mu_{\rm II}^{\pm} \simeq \mu_{\rm CW}(T) = \frac{2^{7/2} \kappa^2 T^{3/2}}{N^* q^3 m^{1/2} \ln(24mTr_0^2/\hbar^2)}.$$
 (31)

Equation (30) allows one to easily calculate small magnetic-field-dependent corrections to the Conwell-Weisscopf expression (31).

In deriving Eq. (30) we took into account that in the type-II DMS the DOS $\rho(\epsilon)$ does not vanish as $(\epsilon - \epsilon_0^{\pm})^{1/2}$ in the vicinity of the percolation thresholds $\epsilon_0^{\pm,32}$ Due to the

presence of random Coulomb fields generated by magnetic and nonmagnetic charged impurities the tails of DOS appear in the forbidden gap of such a material. In other words, here $\rho(\epsilon)$ is smeared out in the vicinity of the percolation thresholds and can be approximately treated as a constant. It means that the defined by Eq. (20) function F_k^{\pm} should be set equal to unity in Eq. (26).

We calculated the mobilities in the type-II DMS for different values of parameter $T_0 = 2\pi e^2 \hbar^2 / (\beta_{ex} \kappa m)$ by using Eq. (26) with $N^*/N_m = 2$, $Z_m = 2$, $\ln(8mTr_0^2/\hbar^2) = 3$ and the same set of parameters by means of which we calculated the mobilities shown in Fig. 2. We have found them similar to those in type-I DMS with large positive values of parameter $b = \beta_{def}/\beta_{ex}$. Here too the application of magnetic field (a) enhances the amplitude of the built-in fluctuations of the magnetic-impurity potential for the majority-spin carriers thus decreasing their mobility and (b) damps out the above amplitude for the minority-spin carriers thus increasing their mobility.

IV. APPLICATION TO GMR

An applied magnetic field changes the conductivity of DMS

$$\sigma = q(\mu^+ n^+ + \mu^- n^-) \tag{32}$$

because it affects both the mobility μ^{\pm} and the concentration of the majority and minority spin carriers, which for nondegenerated DMS is equal to

$$n^{\pm} = \frac{1}{2} N_c \exp\left(-\frac{E_C - \Gamma^{\pm} - F \mp \overline{\Delta}/2}{T}\right).$$
(33)

Here F is the Fermi energy, N_c is the density of the conduction band edge E_c states, and

$$\Gamma^{\pm} = \frac{x(1-x)m}{2\pi N\hbar^2 a} \left(\beta_{\text{def}} \pm \beta_{\text{ex}} \frac{JM}{2M_{\text{sat}}}\right)^2 \tag{34}$$

is the shift of the percolation threshold of the spin-split subbands, which in mixed ternary compounds including type-I DMS is caused by the renormalization of the energy spectrum by the fluctuating short-range electronic potential and is given by Eq. (34).^{24,26} Here *a* is the lattice constant.

Let us, for the sake of simplicity, assume that the total concentration of the free carriers, $n=n^++n^-$, does not depend on magnetic field. It is usually correct when the concentration is determined by shallow nonmagnetic impurities for which the ionization energy does not depend on magnetic field.²² Then the MR can be calculated as

$$\frac{\Delta\rho(H)}{\rho(0)} = \frac{\sigma(0)}{\sigma(H)} - 1 = \left[\frac{\mu^{+}(H)/\mu(0)}{1 + \exp\{-\left[\bar{\Delta}(H) + \gamma(H)\right]/T\}} + \frac{\mu^{-}(H)/\mu(0)}{1 + \exp\{\left[\bar{\Delta}(H) + \gamma(H)\right]/T\}}\right]^{-1} - 1, \quad (35)$$

where $\mu^{\pm}(H)$ is given by Eq. (28), $\overline{\Delta}(H)$ is given by Eq. (21), and $\gamma(H) = \Gamma^+(H) - \Gamma^-(H)$.



FIG. 3. (Color online) Magnetoresistance of DMR for different values of the parameter $b = \beta_{def} / \beta_{ex}$ with (b) and without (a) bandedge shift taken into account.

Figure 3 shows the magnetoresistance of type-I DMS calculated by means of Eq. (35) with the same set of parameters we used to generate Fig. 2. The first set of graphs (a) represents the MR calculated without the differences in the shifts (34) of the spin-split band edges taken into account [$\gamma(H)$ =0], whereas the second set (b) takes this effect into consideration. [Here we used $m=0.1m_e$ and N=4/a3 (Refs. 7 and 8) in our calculations]. One can see that MR is predominantly governed by the magnetic-field dependence of the mobility of the majority carriers, which itself is dominated by scattering off the thermodynamic fluctuations of atomic spins. As a result, the MR is usually negative because both the magnitude of these fluctuations and related scattering decrease with the application of magnetic field. The negative variation of MR becomes less pronounced for materials with greater spins J of magnetic atoms. For the model under discussion the MR becomes positive only if the deformation potential constant β_{def} is large and has the same sign as the exchange coupling constant β_{ex} ($b = \beta_{def} / \beta_{ex} = 5$). In that case, the zero-magnetic field mobility is dominated by scattering off the large build-in fluctuations of the deformation potential of magnetic impurities. The application of the magnetic field increases the amplitude of these fluctuations for the majority-spin carriers thus leading to positive MR. Taking into account the renormalization of the band edges does not substantially change the MR with an insignificant exclusion for the case of $b = \beta_{def} / \beta_{ex} = -5$ when the MR becomes slightly less negative.

In the recent publication²⁵ the authors observed giant positive MR in Mn:Ge DMS in a wide temperature range between 50 and 200 K where the zero-magnetic-field resistivity does not depend on temperature.¹⁵ (Similar results on MR in Ge:Mn were observed in Ref. 17.) It has been demonstrated²⁵ that this phenomenon is related to superparamagnetic nature of magnetic clusters with enhanced concentration of Mn atoms with an average number of magnetic atoms of the order of a hundred. We think that the above



FIG. 4. (Color online) Magnetoresistance of $Mn_{0.05}Ge_{0.95}$ for different values of *J* calculated for T=50 K according to Eqs. (30) and (35).

described model of giant MR in the type-II DMS, where the charged randomly distributed Mn atoms and/or their nanoclusters fully define the scattering time of the free carriers, naturally applies for explaining the experimental data observed in Ref. 25. Figure 4 shows MR of $Mn_{0.05}Ge_{0.95}$ at T = 50 K calculated for different effective spins J of nanoclusters, where we used $\gamma(H)=0$, $N^*/N_m=2$, $Z_m=2$, $\ln(8mTr_0^2/\hbar^2) = 3$, $N|\beta_{ex}| = 4|\beta_{ex}|/a^3 = 1.2 \text{ eV}$, a = 5.66 Å, 7.8and $T_0 = 2\pi e^2 \hbar^2 / (\beta_{\text{ex}} \kappa m) = 2 \text{ eV}$. (The last figure corresponds to $\kappa = 16$ and $m = 0.4 m_e$.) It can be seen that it is very small but positive for small values of $J \sim 1$ due to the almost full mutual compensation of the inputs from the thermodynamic spin fluctuations and from the built-in spatial fluctuations of the random potential of the magnetic impurities. However, the MR remains positive and substantially increases with the increase in J thus reaching hundreds of percent for $J \sim 100$. Figure 5 demonstrates the experimental data on the giant MR of as-grown Mn_{0.05}Ge_{0.95} at different temperatures compared with our calculations based on Eqs. (30)and (35). Here we used the same set of parameters as in Fig. 4 and took an effective spin of nanoclusters J=100 which corresponds to an average number of the Mn atoms in a cluster of the order of 40.15 In addition, we took into account a possible antiferromagnetic coupling between Mn atoms in Ge by replacing the argument in the Brillouin function in Eqs. (2) and (3) with $g\mu_B HJ/(T+T_{AF})$ with $T_{AF}=30$ K. The predictions of our model fits very well experimental data



FIG. 5. (Color online) Magnetoresistance of as-grown $Mn_{0.05}Ge_{0.95}$ at different temperatures (Ref. 25). The solid lines represent theoretical fits according to Eqs. (30) and (35).

considering the fact that we ignored detailed information on the complex structure of the valence bands in Ge. (It should be mentioned here that any attempts failed that were aimed at quantitative explanation of the discussed experimental data in the framework of the bound magnetic polaron model^{10,11} because here the conductivity is due to the carriers in the extended states.)

In summary, we analyzed spin-dependent electrical conductivity in DMS where free carriers are scattered off randomly distributed magnetic impurities. The mobility of the minority and majority spin carriers is shown to be governed by competing impurity spin-disorder effects caused by (a) thermodynamic fluctuations of the atomic spins and (b) random impurity potential generated by the spacially fluctuating concentration of the magnetic atoms. The former effect usually dominates the mobility of the majority-spin carriers in the type-I DMS. It is quenched by external magnetic field leading to giant negative magnetoresistance. In the type-II DMS, where magnetic impurities are charged, the spindependent scattering rate is dominated by the random Coulomb impurity potential. It is shown to be greatly enhanced by the external magnetic field due to the growing fluctuations of local Zeeman splittings of the expanded electronic states leading to giant positive magnetoresistance in such DMS.

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