Scaling approach to hopping magnetoresistivity in dilute magnetic semiconductors

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We suggest a mechanism causing large positive magnetoresistance (MR) in dilute magnetic semiconductors when hopping via nonmagnetic donor impurities dominates the conductivity. The effect is due to the increase in the characteristic width σ of the donor energy distribution with increasing magnetic field B, caused by exchange interactions between magnetic Mn atoms and the electrons localized on nonmagnetic Cl donor impurities. Using general scaling arguments based solely on the dependencies of hopping rates on temperature and on the energies of hopping sites we show that this mechanism accounts quantitatively for our experimental data on MR in n-type $Zn_{1-x}Mn_xSe$:Cl. We suggest a method for extracting the dependence of σ on magnetic field from the MR data. The mechanism explains the experimentally observed universal dependence of the MR effect on the ratio B/T at different temperatures T under the premise that transport is due to the nearest-neighbor or Mott hopping mechanism.

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I. INTRODUCTION

Magnetoresistance (MR) effects in dilute magnetic semiconductors (DMSs) attract much attention due to exciting fundamental physics related to such effects and due to the potential use of DMSs in spintronic devices.[1,2] In this paper we focus on the MR effects in n-doped DMSs, such as $Zn_{1-x}Mn_xSe$:Cl, where charge transport at low temperatures is due to incoherent tunneling (hopping) of electrons via nonmagnetic Cl donors and Mn is isovalently incorporated into the host lattice. The existence of the hopping transport regime in DMS systems has been proven for decades.[3] However, the variety of MR effects in such DMSs is so rich that one hardly can claim a consensus among researchers about the physical mechanisms responsible for the effects. Typically, the resistivity in DMSs at low T increases with magnetic field B at small fields, reaches its maximum at $B \simeq 1$–10 T, and then decreases.

In attempts to account for the MR effects one should clearly distinguish between mechanisms related to the presence of magnetic atoms and those general for hopping transport also in nonmagnetic semiconductors. Among the latter effects are a positive MR due to shrinkage of the wave functions of localized states[4] and a negative MR due to the suppression of self-interference in transport paths,[5–8] due to spin-blockade effects in systems that allow double occupation of localized states,[9–19] and due to scattering on localized magnetic moments.[20] Among the former effects specific for DMSs one should highlight the effect of hopping assisted by thermal fluctuations of the spins of magnetic impurities that leads to a positive MR,[21] and the suppression of bound magnetic polarons in high fields that causes a negative MR.[21,22] The two latter effects are successfully used for the theoretical analysis of MR in compounds such as p-type $Cd_{1-x}Mn_xSe$, where negative magnetoresistance is the largest effect.[22] However, the effect of *compositional disorder* (local fluctuations of Mn content x) on the hopping transport process was not studied in those earlier papers, while this effect is decisive for the MR mechanism suggested in this paper. Although known in the literature[23] the effect of compositional disorder for hopping transport has not yet been applied to account for the positive MR.

In $Zn_{1-x}Mn_xSe$:Cl, the exchange interaction between electrons localized on Cl donors with randomly distributed Mn atoms leads to a broadening of the donor energy distribution when the Mn spins are aligned by an external magnetic field[3,24] as illustrated in Fig. 1. A broadening of the donor energy distribution leads to an increase in the resistivity of the material, i.e., magnetic-field tuning of the compositional disorder results in a large *positive* MR effect.

We propose a scaling relation for the resistivity as a function of the temperature T and of the width σ of the energy distribution of donor levels. Comparing this theory to experimental results obtained in n-type $Zn_{1-x}Mn_xSe$ explains the observation of a large positive MR saturating at high magnetic fields. The proposed scaling relation presumes that σ and kT are the only essential energy scales in the considered problem. This is justified provided that charge transport is due to nearest-neighbor hopping or due to Mott variable-range hopping (VRH). In the case of Efros-Shklovskii VRH[4] another energy scale, namely the width of the Coulomb gap, comes into play.

II. EXPERIMENTAL DATA

The layer structure grown by molecular-beam epitaxy on (100) GaAs consists of an undoped ZnSe buffer followed by the n-type $Zn_{0.94}Mn_{0.06}Se$:Cl layer.[25,26] The transport measurements were performed in van-der-Pauw geometry. In Fig. 2 the measured MR data are shown for low temperatures, $T \leqslant 10$ K, at which activation of electrons into the conduction

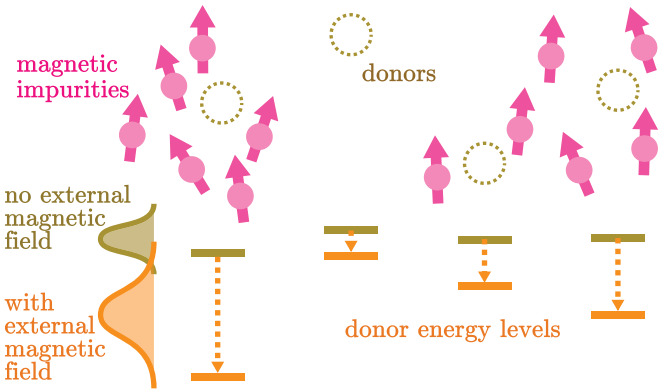

FIG. 1. (Color online) Illustration of the donor band broadening mechanism. The more magnetic impurities are near a donor, the larger is its energy shift due to the exchange interaction when the spins of the magnetic impurities are aligned by the external magnetic field. This increases the width σ of the donor energy distribution; see Eq. (18).

band can be neglected and the conductivity is due to hopping of electrons via donor impurities.

Cl impurities form shallow donors in ZnSe with binding energy $\varepsilon_{\text{bind}} \approx 26$ meV,[27] and Bohr radius $a_{\text{B}} \approx 28$ Å.[28] The donor concentration N_D is a decisive parameter for the low-temperature conductivity mechanism. At $N_D^{1/3} a_{\text{B}} < 0.25$ charge transport at low temperatures occurs by hopping via the donor sites. In the opposite case, metallic behavior is expected.[28] We focus on a sample with chlorine concentration $N_D = 4.5 \times 10^{17}$ cm^{-3} estimated from room-temperature Hall measurements, yielding $N_D^{1/3} a_{\text{B}} \approx 0.2$.

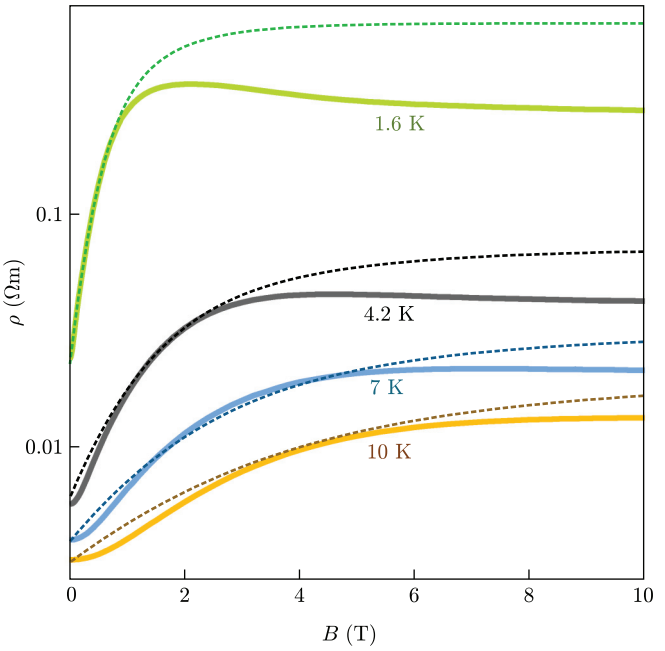

FIG. 2. (Color online) Resistivity dependent on magnetic field in n-type $Zn_{0.94}Mn_{0.06}Se$:Cl with donor concentration 4.5×10^{17} cm^{-3}. Solid lines: experimental data; dashed lines: calculated via Eqs. (8) and (19) using $g = 2$, $\gamma_1 = 1$, $\gamma_2 = 1.8$, $J = 5/2$.

III. PHENOMENOLOGICAL DESCRIPTION

We analyze the MR effects in the framework of the Gaussian disorder model, in which electrons hop between sites randomly distributed in space with a Gaussian distribution of energies. No correlations between energies and positions of sites are taken into account. The hopping rates Γ_{ij} are described by the Miller-Abrahams expression[4]

$$\Gamma_{ij} = \nu_0 \, e^{-2(r_{ij}/a_{\text{B}})} \begin{cases} e^{-\Delta\varepsilon_{ij}/kT} & \text{when} \quad \Delta\varepsilon_{ij} > 0, \\ 1 & \text{when} \quad \Delta\varepsilon_{ij} \leqslant 0, \end{cases} \tag{1}$$

where a_{B} is the electron localization length, ν_0 is the attempt-to-escape frequency, $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the distance between the two sites, and $\Delta\varepsilon_{ij}$ is the difference between their energies,

$$\Delta\varepsilon_{ij} = \varepsilon_j - \varepsilon_i + e\mathbf{E} \cdot (\mathbf{r}_i - \mathbf{r}_j), \tag{2}$$

$\mathbf{E}$ being the external electric field.

The resistivity ρ depends on ν_0, a_{B}, on the concentrations of donors and electrons, on the width σ of the distribution of donor energies, and on the temperature T. All these parameters, except σ and T, are constant for a given sample. Therefore the MR at a given T is due to a magnetic-field-induced change of the width σ: $\rho = \rho(\sigma, T)$. A possible cause of changing σ in magnetic field is due to the exchange interaction between the electrons bound on donors and magnetic impurities controlled by magnetic field.

The expression for hopping rates (1) possesses a remarkable feature: when all energies, temperature, and electric field $\mathbf{E}$ are scaled by some factor λ, the rates and concomitantly the current density $\mathbf{j}$ remain unchanged:

$$\mathbf{j}(\lambda\sigma, \lambda T, \lambda\mathbf{E}) = \mathbf{j}(\sigma, T, \mathbf{E}). \tag{3}$$

Expressing the current density $\mathbf{j}$ via the resistivity ρ,

$$\mathbf{j}(\sigma, T, E) = \mathbf{E}/\rho(\sigma, T), \tag{4}$$

one obtains from Eq. (3) that the ratio ρ/T remains unchanged when σ and T are scaled simultaneously:

$$\frac{\rho(\lambda\sigma, \lambda T)}{\lambda T} = \frac{\rho(\sigma, T)}{T}. \tag{5}$$

This means that ρ/T is a function of one variable only, namely, the ratio σ/T. Let us denote this function as $\tilde{f}$:

$$\frac{\rho(\sigma, T)}{T} = \tilde{f}\left(\frac{\sigma}{T}\right). \tag{6}$$

A similar scaling relation has been suggested by Ambegaokar *et al.*,[29] though for the case of small temperatures, when kT is much less than the other energy parameters in the problem. On the contrary, Eq. (6) relies solely on the form of the Miller-Abrahams transition rates.

For studying the magnetoresistance, it is convenient to define a new function f as

$$f(b/\sigma_0) = \tilde{f}(b), \tag{7}$$

where σ_0 is the width of the energy-level distribution at zero magnetic field. A combination of Eqs. (6) and (7) yields

$$\frac{\rho}{T} = f\left(\frac{\sigma}{\sigma_0 T}\right). \tag{8}$$

Equation (8) is the key relation of our scaling approach. It will be used below to extract the magnetic-field dependence

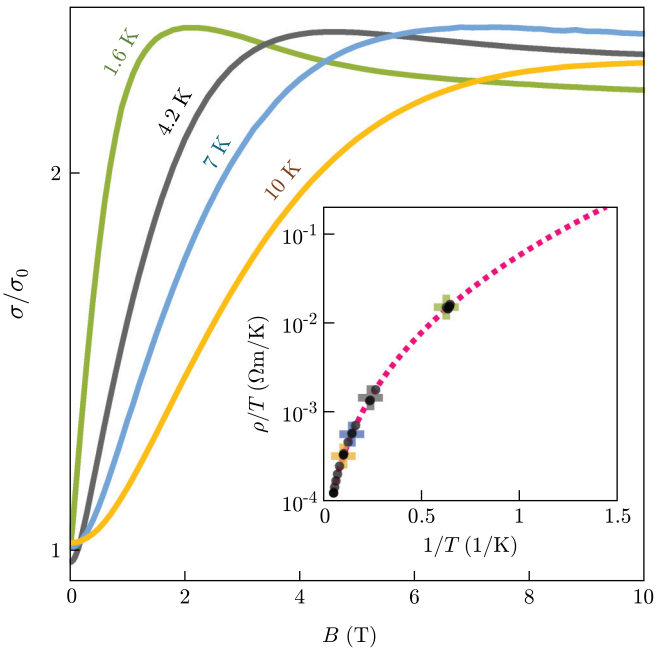

FIG. 3. (Color online) Magnetic-field dependence of the width σ of the energy-level distribution derived from MR data in Fig. 2. Inset: function f derived from zero-field data using Eq. (9) (symbols: experimental data; line: interpolation). Crosses correspond to the temperatures of the shown MR curves.

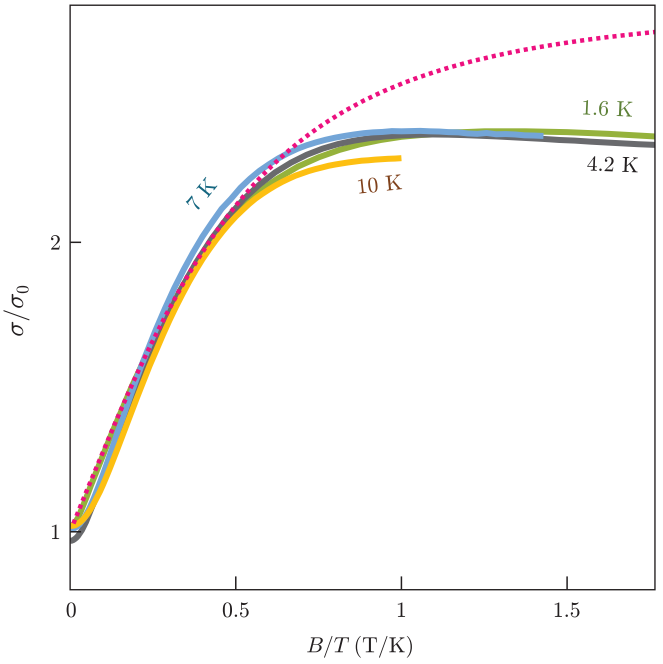

FIG. 4. (Color online) Dependence of the width σ of the energy-level distribution on the ratio B/T. Solid lines: experimental data; dotted line: fitting by Eq. (19).

of the width σ from the experimental data. This will be done in two steps: (i) extracting the function f, and (ii) determining the ratio σ/σ_0 as a function of magnetic field B and temperature T.

The function f can be derived from the temperature dependence of the resistivity. Since $\sigma = \sigma_0$ at $B = 0$, one obtains from Eq. (8)

$$\frac{\rho}{T} = f\left(\frac{1}{T}\right) \quad \text{at} \quad B = 0. \tag{9}$$

Plotting ρ/T versus $1/T$, one obtains the function f (see inset in Fig. 3).[30] With known f and using Eq. (8), we can restore the ratio $\sigma(B,T)/\sigma_0$ from experimental data for $\rho(B,T)$,

$$\frac{\sigma(B,T)}{\sigma_0} = T\, f^{-1}\left(\frac{\rho(B,T)}{T}\right). \tag{10}$$

Figure 3 shows the magnetic-field dependence of σ/σ_0 for different temperatures derived from the data in Fig. 2.

The most remarkable feature of Fig. 3 is the similarity of the $\sigma(B)$ dependencies at different T values. In order to make this more evident, we plot in Fig. 4 the same data as functions of the ratio B/T. One can see that the dependencies $\sigma(B/T)$ for all temperatures are almost identical.

IV. MECHANISM OF MAGNETORESISTANCE

The dependence of σ on B/T shown in Fig. 3 can be naturally related to the magnetization of Mn atoms in $Zn_{1-x}Mn_xSe$ by the external magnetic field. In a diluted system one can assume that the Mn atoms can be considered as isolated from each other and that the antiferromagnetic coupling between adjacent Mn ions is negligible. In this case, the average z component $\langle J_z\rangle$ of the d-electron angular momentum of a Mn ion is[31]

$$\langle J_z\rangle = J\, B_J(g\mu_B B/kT), \tag{11}$$

where z is the direction of the magnetic field $\mathbf{B}$, μ_B is the Bohr magneton, k is the Boltzmann constant, $J = 5/2$ and $g = 2$ are the angular momentum and the g factor, respectively, of the $3d$ shell of Mn^{2+}, and B_J is the Brillouin function defined as

$$B_J(\xi) = \frac{1}{2J}\left[(2J+1)\coth\left(\frac{(2J+1)\xi}{2}\right) - \coth\left(\frac{\xi}{2}\right)\right]. \tag{12}$$

The fact that both the width σ of the donor level distribution and the magnetization $\langle J_z\rangle$ of the Mn atoms depend only on the ratio B/T makes it highly plausible that there is a causal relationship between these quantities. Therefore we propose that the broadening of the donor-level distribution in a magnetic field is related to the magnetization of the Mn spins.

The suggested MR mechanism can be analyzed in more detail starting from the standard s-d spin Hamiltonian.[32] We consider the exchange interaction $\hat{V}_{\text{ex,in}}$ between the spin $\hat{\mathbf{s}}_i$ ($s_{z,i} = s_z = \pm 1/2$) of the s-like electron on the ith donor and the angular momentum $\hat{\mathbf{J}}_n$ of the d shell of the nth Mn atom:

$$\hat{V}_{\text{ex,in}} = \alpha|\phi_i(\mathbf{r}_n)|^2\hat{\mathbf{s}}_i\cdot\hat{\mathbf{J}}_n, \tag{13}$$

where ϕ_i is the wave function of the ith donor, $\mathbf{r}_n$ is the position of the nth Mn atom, and α is a material-dependent exchange constant. Summation over n and averaging over orientations of Mn angular momenta results in a spin-dependent correction

$\varepsilon_{\mathrm{ex},i}$ to every donor energy level where

$$\varepsilon_{\mathrm{ex},i}(s_z,B,T) = \alpha\, s_z\, \langle J_z\rangle \sum_n |\phi_i(\mathbf{r}_n)|^2. \tag{14}$$

Neglecting the normal Zeeman splitting energy $s_z\, g_c\, \mu_B\, B$ which is about an order of magnitude smaller than the Giant Zeeman splitting at $T < 10$ K and $B < 10$ T in these samples, we obtain for the donor energy ε_i in the presence of a magnetic field

$$\varepsilon_i = \varepsilon_i^{(0)} + \varepsilon_{\mathrm{ex},i}(s_z,B,T), \tag{15}$$

where $\varepsilon_i^{(0)}$ is the donor energy at zero magnetic field.

The exchange energies $\varepsilon_{\mathrm{ex},i}$ are different for different donors because of spatial fluctuations in the Mn content. For simplicity, we consider only the lower spin states ($s_z = -1/2$) as being involved in the transport process. This is reasonable for large enough magnetic fields when $|\varepsilon_{\mathrm{ex},i}(+1/2,B,T) - \varepsilon_{\mathrm{ex},i}(-1/2,B,T)| \gtrsim kT$. Our estimates show that the inequality holds for $B/T \gtrsim 0.02$ T/K at $T \simeq 10$ K.

Donor energies in zero magnetic field, $\varepsilon_i^{(0)}$, can correlate with the local Mn content. Let us denote by $\tilde{\varepsilon}_i^{(0)}$ the energy of the donors in the binary host material, i.e., ZnSe. It is natural to assume that the Mn-related shift $\varepsilon_i^{(0)} - \tilde{\varepsilon}_i^{(0)}$ of the donor level is proportional to the local Mn content in the vicinity of the donor.[24] Since the local Mn content can be measured by the sum in the right-hand side of Eq. (14),

$$\varepsilon_i^{(0)} - \tilde{\varepsilon}_i^{(0)} = y \sum_n |\phi_i(\mathbf{r}_n)|^2, \tag{16}$$

with some coefficient y. Combining this with Eqs. (14) and (15) one obtains for $s_z = -1/2$ the donor energy

$$\varepsilon_i = \tilde{\varepsilon}_i^{(0)} + \left(y - \frac{1}{2}\alpha\, \langle J_z\rangle\right) \sum_n |\phi_i(\mathbf{r}_n)|^2. \tag{17}$$

Herewith one obtains the variance σ^2 of donor energies in magnetic field as

$$\sigma^2 = \mathrm{Var}\left(\tilde{\varepsilon}_i^{(0)}\right) + \left(y - \frac{\alpha}{2}\, \langle J_z\rangle\right)^2 \mathrm{Var}\,(\Sigma), \tag{18}$$

with $\Sigma = \sum_n |\phi_i(\mathbf{r}_n)|^2$. This means that the variance σ^2 is a *quadratic* function of the magnetization[24] $\langle J_z\rangle = J B_J(g\mu_B B/kT)$.

Hence, the ratio σ/σ_0 is the following function of the magnetic field:

$$\frac{\sigma(B)}{\sigma_0} = \sqrt{1 - \gamma_1^2 + \left[\gamma_1 + \gamma_2\, B_J\left(\frac{g\mu_B B}{kT}\right)\right]^2}, \tag{19}$$

where γ_1 and γ_2 are dimensionless parameters defined as

$$\gamma_1 = \frac{-y\sqrt{\mathrm{Var}\,(\Sigma)}}{\sqrt{\mathrm{Var}\left(\tilde{\varepsilon}_i^{(0)}\right) + y^2 \mathrm{Var}\,(\Sigma)}}, \tag{20}$$

$$\gamma_2 = \frac{\frac{1}{2}\alpha J\sqrt{\mathrm{Var}\,(\Sigma)}}{\sqrt{\mathrm{Var}\left(\tilde{\varepsilon}_i^{(0)}\right) + y^2 \mathrm{Var}\,(\Sigma)}}. \tag{21}$$

Let us use Eq. (19) to fit the experimental data in Fig. 4, with fitting parameters γ_1 and γ_2. The best fit ($\gamma_1 = 1, \gamma_2 = 1.8$) is shown in Fig. 4 by a dotted line. Assigning the value 1 to γ_1 means that the disorder in donor energies is solely due to the Mn impurities. The agreement between experiment and theory is good for positive MR. This indicates that the basic understanding of the mechanism of the positive MR effect is correct. To further corroborate this statement, we also calculated the MR curves using Eqs. (8) and (19) (dashed lines in Fig. 2) and compared them with experiment (solid lines in Fig. 2). The calculated MR appears in good agreement with the measurements in the region of positive MR.

At high magnetic fields the experimental MR curves in Fig. 2 saturate at lower magnetic fields than the calculated ones. For the lowest temperatures a region with negative MR is seen. This could be due to the effect of magnetic polarons. Hopping transport of bound magnetic polarons in DMS materials has been shown to yield a negative MR, since the polaron binding energy decreases when the system is magnetized.[21,22]

V. EFFECTS OF ELECTRON-ELECTRON INTERACTION

The scaling description suggested above presumes that the system has only one energy scale, namely the one given by the width σ of the site energy distribution so that the transport problem under consideration contains two energy scales, σ and kT in the dimensionless combination σ/kT. It is correct as long as charge transport is due to the nearest-neighbor hopping mechanism or due to Mott VRH mechanism, for which long-range Coulomb interactions do not play an essential role. If, however, Coulomb correlations are significant, charge transport at low temperatures should be described by Efros-Shklovskii (ES) VRH theory.[4] In the latter case, the scaling approach would have to be modified in order to take into account another energy scale, namely the width of the Coulomb gap given by

$$\Delta_C \approx e^3 g_0^{1/2}/\kappa^{3/2}, \tag{22}$$

where e is the elementary charge, κ is the dielectric constant, and g_0 is the density of states just outside the Coulomb gap in the vicinity of Fermi level ε_F.[4]

Temperature dependence of hopping resistivity is known to be indicative for the underlying transport mechanism.[4] For a noninteracting electron system this dependence is given by Mott VRH law

$$\rho \propto \exp\,(T_{0M}/T)^{1/4} \tag{23}$$

with a characteristic temperature[4]

$$T_{0M} = 21.2\frac{1}{kg(\varepsilon_F)\xi^3}, \tag{24}$$

where ξ is the localization length.

If Coulomb correlations are decisive for hopping transport, the resistivity is determined by the Coulomb gap and is expected to follow the ES VRH law,[4]

$$\rho \propto \exp\,(T_{0ES}/T)^{1/2} \tag{25}$$

with a characteristic temperature[4]

$$T_{0ES} = 2.8\frac{e^2}{k\kappa\xi}. \tag{26}$$

In order to estimate the importance of Coulomb correlations in our sample, we therefore examine in Fig. 5 the temperature

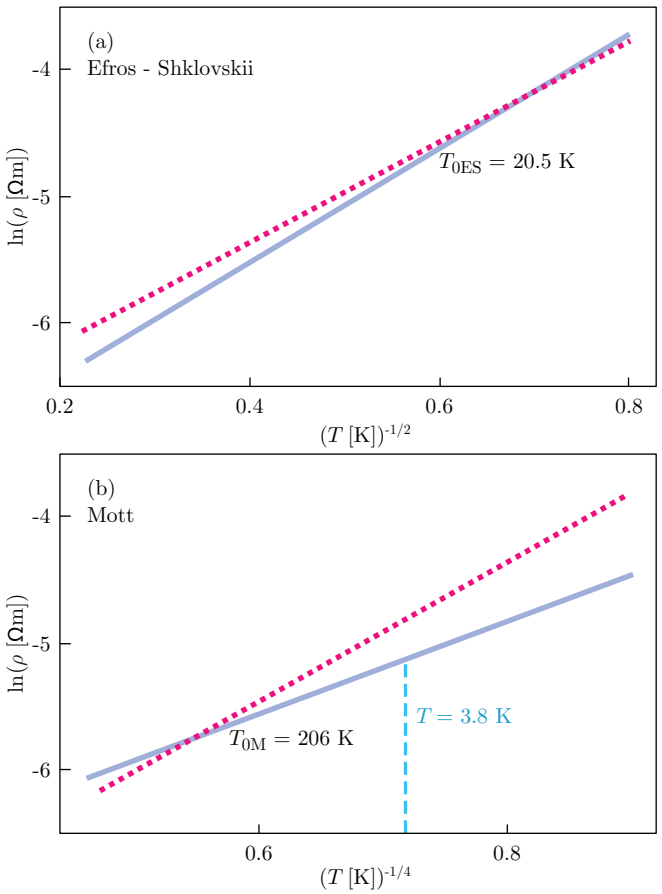

FIG. 5. (Color online) The temperature dependence of the resistivity, compared to the Efros-Shklovskii law (25) and the Mott law (23).

dependence of the resistivity at zero magnetic field. If one would demand the same transport mechanism to be valid in the whole studied temperature range, one should conclude, looking at Fig. 5, that ES law shown in Fig. 5(a) provides the best fit over the full temperature range as shown by the dashed line in this figure. However, such a conclusion would decisively rely on the ρ value at the lowest temperature, $T = 1.6$ K. Furthermore, it is clearly seen in Fig. 5(a) that at $T \geqslant 3.8$ K the dependence $\rho(T)$ is weaker than at lower temperatures, $T < 3.8$ K. Such a behavior of $\rho(T)$ is not at all surprising. Similar data are known for numerous systems with the VRH transport mechanism, such as n-doped CdSe,[33,34] uncompensated ion-implanted Si:As,[35] uncompensated Si:P,[36] and many others. Indeed it is a common situation that ES VRH valid at low T converts at some temperature T_{cross} into Mott VRH.[33–36] For our data T_{cross} is apparently close to 3.8 K. The clear ability of Mott law to describe our experimental data at $T > 3.8$ K is shown by the solid line in Fig. 5(b). Theoretical estimates for T_{cross} via the relation $T_{\text{cross}} \approx T_{0ES}^6/[T_{0M}^5(0.24)^4]$ suggested in the literature give usually much lower values for T_{cross} than those observed experimentally.[33,35] We have a similar situation. Using the values $T_{0ES} = 20.5$ K and $T_{0M} = 206$ K as obtained from the slopes of solid lines in Figs. 5(a) and 5(b), respectively, one would come to the estimate $T_{\text{cross}} \approx 0.06$ K, which is essentially lower than the observed magnitude $T_{\text{cross}} \approx 3.8$ K. Apparently our experimental data are in perfect agreement with the data usually obtained in low-compensated doped semiconductors in the vicinity of metal-insulator (MI) transition. The fact that Coulomb correlations do not determine the behavior of the system at $T > 3.8$ K may be due to the divergence of the dielectric constant κ in the vicinity of the MI transition[33–36] or due to dynamical screening of the interactions at rising temperature.[37] One can conclude that at $T > 3.8$ K the hopping transport mechanism in our samples is not dominated by Coulomb correlations. Since the most part of our scaling analysis was carried out at $T > 3.8$ K, one can conclude that our scaling description is justified at such temperatures. Further work is needed to extend the scaling approach for the Efros-Shklovskii conduction regime.

The deviation from the Mott law given by Eq. (23) observed in our experiments at the lowest temperature $T = 1.6$ K could be also due to the effect of magnetic polarons, which could be also responsible for the negative MR at high B seen in Fig. 2 at $T = 1.6$ K. As shown by Foygel *et al.*,[38] magnetic polarons lead to the modification of the exponent in Eq. (23) from 1/4 to 2/5 provided hopping transport is not dominated by Coulomb correlations.

VI. CONCLUSIONS

A mechanism is suggested for large positive magnetoresistance in diluted magnetic semiconductors in the hopping transport regime. The mechanism is based on the effect of compositional disorder on hopping transport.[23] The width of the energy-level distribution due to compositional disorder is controlled by the exchange interaction between electrons and magnetic impurities, while the magnetization of magnetic ions is controlled by the external magnetic field. The broadening of the energy distribution with magnetization takes place independently of the hopping conduction mode, i.e., the suggested mechanism is valid for the nearest-neighbor hopping as well as for the Mott variable-range hopping and for the Efros-Shklovskii regime.

A scaling ansatz is suggested to describe our experimental data in n-type $Zn_{1-x}Mn_xSe$:Cl in the vicinity of the metal-insulator transition. The scaling approach allows one to estimate quantitatively the broadening of the energy spectrum due to compositional disorder directly from experimental data. Furthermore, it is shown that the width of the energy-level distribution depends solely on the ratio B/T.

The scaling theory is applicable if the effects of Coulomb correlations can be neglected, for instance for the nearest-neighbor hopping, or in the Mott variable-range-hopping regime, which we show to be valid in our samples at not too low temperatures.

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