Variable-Range Hopping of Spin Polarons: Magnetoresponse in a Modified Mott Regime

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Abstract

We analyze electrical conductivity controlled by hopping of bound spin polarons in disordered solids with wide distributions of electron energies and polaron shifts (barriers). By means of percolation theory and Monte Carlo simulations we have shown that in such materials at low temperatures, when hopping occurs in the vicinity of the Fermi level, a hard polaron gap does not manifest itself in the transport properties. This happens because as temperature decreases the hopping polaron trades the decreasing electron and polaron barriers for increasing hopping distance. As a result, in the absence of the Coulomb correlation effects, in this variable-range variable-barrier hopping regime, the electrical resistivity, $\rho$, as a function of temperature, $T$, obeys a non-activation law: $\ln(\rho/\rho_0) = \left(\frac{T}{T'}\right)^p$ with $p = 2/(d+2)$, where $d$ is the dimensionality of the system. It differs from the standard Mott law for which $p = 1/(d+1)$. Also, we studied the effects of upper and lower boundaries in the polaron shift distribution on hopping conduction, which may result in a partial re-entrance of the hard polaron gap. We discuss possible applications to the problem of giant negative magnetoresistance in dilute magnetic semiconductors and nanocomposites where for paramagnetic materials $p = 3/(d+2)$.

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I. INTRODUCTION

Electrical conductivity due to variable-range hopping (VRH) in doped crystalline and amorphous semiconductors has been a subject of intensive experimental and theoretical studies for more than three decades [1, 2, 3, 4]. The temperature dependence of the VRH conductivity is described by stretched exponents $\sigma \propto \exp \left(-T/T_0\right)^p$ and depending on the value of $p$ can be attributed to one of the two major regimes such as the Mott ($p = 1/4$) and the Shklovskii-Efros ($p = 1/2$) regimes [1, 2]. Several observations of giant and colossal negative VRH magnetoresistance in systems with strong spin disorder such as dilute magnetic semiconductors (DMS) have been reported recently [5, 6, 7]. It seems rather natural to attribute VRH magnetoresistance to spin polarons that are believed to be a major cause of negative magnetoresistance in DMS [8, 9, 10]. However, a theory of VRH magnetoresistance has never been addressed in the literature, partially, because of the belief that spin-polaron hopping may lead only to a simple activation dependence of the conductivity which mimics nearest-neighbor hopping [11]. We will demonstrate that in the systems with significant disorder of the magnetic energies spin-polaron hopping is described by a stretched exponent similar to that of the Mott behavior.

The study of electrical conductivity in solids, which is controlled by polaron hopping, has centered so far on materials where the polaronic part of the activation energy is fixed [1, 3, 10, 11, 12, 13, 14]. Indeed, the lowering of the free energy of the system due to atomic spin alignment [9] (or lattice polarization [13]) in the vicinity of the trapped carrier, the so-called polaron shift, and its associated hopping barrier are generally determined by the spatial extent of the localized states between which the hopping occurs. This is usually true no matter how strong the coupling between a localized carrier and magnons (or phonons) and how many of them are emitted or absorbed during the hopping event [15]. (Sometimes, non-linear polaron effects may lead to an additional self-localization of the bound state resulting in the actual collapse of its wave function [9]. These situations are out of the scope of the present paper for they would hardly be favorable for the hopping process.) That is why in standard lightly doped semiconductors the polaron hopping barrier can be considered fixed [9]. (This is also correct in heavily doped and compensated nonmagnetic semiconductors, where at low temperatures conductivity is controlled by electron hopping between aggregations of impurities of a fixed size close to the screening length [2].) In all
these cases, the bound polaron spectrum has a hard gap resulting in a predominantly activation temperature dependence of conductivity even in the variable-range hopping (VRH) regime [3, 11].

Recent technological developments in producing novel dilute magnetic semiconductors, especially GaAs : Mn, and magnetic nanostructures, such as GaAs/ErAs, made it possible to observe electrical conductivity due to hopping via magnetic impurities [3] and nano-islands [10] thus revealing a rich interplay of magnetic and electrical transport properties. For instance, Mn atoms play a dual role as acceptor sites and magnetic ions in GaAs resulting in the very unusual magnetic properties of this material [3, 17]. Giant negative magnetoresistance has been observed [3, 5, 6] below the metal-insulator transition point of GaAs : Mn, which clearly displays the VRH behavior with no sign of the hard magneto-polaron gap. The absence of this gap in the electrical conduction experiments can be understood by taking into account local fluctuations of the bound polaron shifts. These, in turn, may be related to the distribution of exchange couplings between Mn ions and holes as was suggested in Ref. [17]. In addition, it is quite possible that the fluctuations of the local spin polaron shifts are not correlated with those of the "bare" electron energies. In heavily doped and compensated samples [2] of GaAs : Mn, the former can be determined by the total number of magnetic impurities in the localization volume while the latter may depend on the electrically active ones. For ErAs nanoislands embedded in GaAs matrix [10], the distribution of polaron shifts follows the distribution of the islands’ volumes. Meanwhile, the "bare" electron spectrum of the islands is apparently determined by the distribution of the islands’ sizes and shapes. We will show that fluctuations of the local (lattice and spin) polaron shifts in certain circumstances may wash out the hard polaron gap, leading to a substantial modification of the standard Mott law for the electrical conductivity [20] even in the absence of Coulomb correlation effects.

Bound spin (magnetic) polarons, in contrast to their lattice counterparts, possess polaron shifts that, as well as their distribution, can be tuned by applied magnetic field thus resulting in giant or even colossal magnetoresistance [10, 11, 13, 19]. Externally controlled switching off and on of the magneto-polaron effect also allows one to better identify the hopping mechanism in magnetic materials.

The main goal of the present paper is to develop a consistent theoretical approach to the description of variable-range hopping of polarons in materials where disorder gives rise not
only to the electron-energy distribution but, also, to the distribution of local polaron shifts. This approach is based on percolation theory and Monte Carlo simulations. In Section II, we will present simple qualitative arguments concerning the main results of the present paper. A rigorous theoretical derivation of these results is given in Section III. Section IV deals with the Monte Carlo simulations of the polaron hopping problem aimed at finding the universal constants and functions that appear in the applications of the percolation theory. Effects of the low boundary in the distribution function of the polaron barriers will be considered in Section V. In Section VI we briefly discuss giant negative magnetoresistance in magnetic semiconductors and nanostructures, a phenomenon that in some materials can be controlled by variable-range variable barrier hopping of the spin polarons.

II. QUALITATIVE CONSIDERATION

We will start from a qualitative description of the low-temperature electrical resistivity in a system where localized states have electronic energies randomly distributed in the vicinity of the Fermi energy $E_F$. The derivation of the original Mott’s law is based on the assumption that the density of states in such a system near $E_F$ is constant [1, 2, 20]. In addition to this assumption, we will suppose that the states in question possess randomly distributed polaron barriers as well. Our goal is to find out how this factor would modify the standard Mott’s law for DC hopping conductivity.

In the original Mott’s derivation, it is assumed that at a given temperature $T$ most of the hopping events involve random sites with one-electron energies belonging to a strip of width $\epsilon$ around the Fermi level. In our case, we have to account for an additional random distribution of the polaron barriers. Thus we generalize the Mott conjecture and introduce a hyperstrip of width $\epsilon$ and height $W$, where $W$ is a scatter of the polaron barriers. The probability of a typical hopping event for two sites belonging to this hyperstrip reads:

$$w(\epsilon, W) \propto \exp \left( -\frac{2r}{a} - \frac{\epsilon + W/2}{T} \right),$$  

(1)

where $a$ is the localization radius of the electron wave function, $T$ is measured in the energy units, and $r$ is the average distance between the sites of the hyperstrip:

$$r \simeq N^{-1/d} = (G\epsilon W)^{-1/d}. \quad (2)$$
Here $N$ is the concentration of the sites in the hyperstrip, $d = 2$ or 3 is the spatial dimension of the system and $G$ is a joint density of (polaronic) states (DOS), such that $G(\epsilon_0, W_0) \, d\epsilon \, dW$ is a probability, per unit volume, to find a site with $\epsilon$ in the interval $(\epsilon_0, \epsilon_0 + d\epsilon)$ and $W$ in the interval $(W_0, W_0 + dW)$. (As in the original Mott’s derivation [1, 20], we assume that $G$ is constant. However, this is not true if the Coulomb correlations are taken into account [2].)

After substituting (2) in (1), it is easy to find that the probability (1) as a function of $\epsilon$ and $W$ will reach its sharp maximum at

$$\epsilon_{opt} = W_{opt}/2 = \left( \frac{2T}{d a G^{1/d}} \right)^{d/(d+2)} \approx T \left( \frac{\tilde{T}_0^{(d)}}{T} \right)^{2/(d+2)}. \quad (3)$$

The hopping conductivity, $\sigma$, is proportional to $w(\epsilon_{opt}, W_{opt})$. Therefore, substituting the optimal values (3) and (2) in (1) yields the following expression for the hopping resistivity:

$$\rho = 1/\sigma = \rho_0 \exp \left[ \left( \frac{\tilde{T}_0^{(d)}}{T} \right)^{2/(d+2)} \right], \quad (4)$$

where

$$\tilde{T}_0^{(d)} = \left( \frac{\beta_0^{(d)}}{G a^d} \right)^{1/2} \quad (5)$$

and $\beta_0^{(d)}$ is a numerical factor of the order of unity. Expression (4) constitutes the main result of the present paper. (In the next section, we will obtain this result by means of a more rigorous percolation theory approach. Also, by using the Monte Carlo simulation described in Section 4 we will be able to find the numerical factor $\beta_0^{(d)}$ in Eq. (5).) It can be seen that, in accordance with the above derived formula, in the 3D (2D) case the well known Mott’s exponent $1/4$ ($1/3$) (see [2, 20]) should be substituted with a somewhat larger exponent of $2/5$ ($1/2$).

As the temperature goes up both the width and the height, (3), of the optimal hyperstrip increase while the optimal hopping distance (2) decreases. It means that with increasing temperature the polaron chooses to jump over increasingly smaller distances at the cost of overcoming the increasingly higher and higher polaron (and electron energy) barriers. That is why we call this regime a variable-range, variable-barrier hopping (VRVBH). In this regime, in contrast to the nearest-neighbor hopping (NNH) of polarons, both the lattice
or the magnetic ones, the polaron hard gap is washed out by the fluctuations of the polaron shifts (barriers).

If, however, there is a maximal polaron shift in a system, $W_{\text{max}}$, then at high enough temperatures, such that $T > T_{1}^{(d)}$, where

$$T_{1}^{(d)} = \left( \left( t_{0}^{(d)} W_{\text{max}} / 4 \right)^{d+1} / T_{M}^{(d)} \right)^{1/d},$$  \hspace{1cm} (6)$$

the optimal polaron shift becomes $W_{\text{max}}$. (Here $t_{0}^{(d)}$ is a numerical factor of the order of unity, which will be determined later by Monte Carlo simulation.) In this case, the combined DOS, $G$, should be substituted with an ordinary electron DOS, $g$, and the VRVBH law (4) with a slightly modified Mott’s law for polaron VRH

$$\rho = \rho_{0} \exp \left[ \left( T_{M}^{(d)} / T \right)^{1/(d+1)} + \tau_{d} W_{\text{max}} / T \right],$$  \hspace{1cm} (7)$$

where the second (hard-gap activation) term under the exponent sign is small compared to the first (Mott’s) term. Here

$$T_{M}^{(d)} = \frac{\beta_{M}^{(d)}}{g a^{d}},$$  \hspace{1cm} (8)$$

where $\tau_{d}$ and $\beta_{M}^{(d)}$ are numerical factors of the order of unity, which will be established later.

At low temperatures, both the electron, $g$, and the combined, $G$, DOS cannot anymore be treated as constant in the vicinity of the Fermi level due to the appearance of the soft Coulomb gap in the electron spectrum. The latter is related to the long-range Coulomb correlation effects in the spatial distribution of electrons above and holes below the Fermi energy. The maximal width of this gap can be estimated as

$$\epsilon_{C} \simeq \left[ \left( e^{2} / \kappa \right)^{d} g \right]^{1/(d-1)} \simeq \left[ \left( e^{2} / \kappa a \right)^{d} / T_{M}^{(d)} \right]^{1/(d-1)},$$  \hspace{1cm} (9)$$

where $\kappa$ is the dielectric constant of the material and $e$ is the electron charge. This gap will modify the VRVBH law at low temperatures when its width exceeds the width of the optimal strip: $\epsilon_{C} > \epsilon_{\text{opt}}$, i.e. when $T < T_{2}^{(d)}$ with

$$T_{2}^{(d)} \simeq \epsilon_{C} \left( \epsilon_{C} / \tilde{T}_{0}^{(d)} \right)^{2/d},$$  \hspace{1cm} (10)$$

6
where $\tilde{T}_0^{(d)}$ is given by expression (3). It can be seen that the temperature interval ($T_2^{(d)} < T < T_1^{(d)}$), where the VRVBH regime (4) exists, may be sufficiently large if the polaronic effect is strong enough. Based on the evaluations of $G$, presented in Section 6, it is possible to show that the required condition $T_2^{(d)} \ll T_1^{(d)}$ is equivalent to $W_{\text{max}} \gg \epsilon_C$.

III. CALCULATION OF RESISTIVITY. A PERCOLATION THEORY APPROACH

Let us consider two sites (quantum dots, impurity centers or their aggregations, etc.) $i$ and $j$ separated by a distance $r_{ij}$. To find the bound polaron hopping rate in the so-called nonadiabatic limit [12, 13] one can employ the semiclassical approach developed in [12, 13] for the lattice small polarons (SP) or in [10] for the bound magnetic polarons (BMP). In both cases, however, is possible to show (see Appendix) that if $i \rightarrow j$ tunneling transition is governed either by strong multiphonon coupling (for the lattice SP) or by thermodynamic fluctuations of the local magnetization (for the BMP), when $W_i + W_j \geq |\tilde{\epsilon}_j - \tilde{\epsilon}_i|$, the hopping rate

$$\gamma_{ij} = \gamma_{ij}^0 \exp \left[ -\frac{2r_{ij}}{a} - \frac{(W_i + W_j + \tilde{\epsilon}_j - \tilde{\epsilon}_i)^2}{4(W_i + W_j)T} \right], \quad (11)$$

where $a$ is the spatial-decay length of the localized wave function into the semiconductor matrix, $\tilde{\epsilon}_l = \epsilon_l(0) - W_l$ is an equilibrium energy of the $l$-th site with the localized carrier; $\epsilon_l(0)$ is the “bare” electron energy of the carrier-free state and $W_l$ is the bound polaron shift. The latter describes the lowering of the free energy of the system due to either lattice polarization or to atomic spin alignment in the vicinity of the trapped carrier. (Usually $W_l$ is inversely proportional to the number of polar or magnetic atoms in the localization volume [10, 13].)

If, however, electron-phonon (electron-magnon) coupling is not strong, i.e. if $W_i + W_j < |\tilde{\epsilon}_j - \tilde{\epsilon}_i|$, the single-acoustic-phonon assisted tunneling of Miller-Abrahams type [2] prevails over the resonant one described by Eq. (11). Then (see [2, 10] for details)

$$\gamma_{ij} \propto (N_P + 1/2) \pm 1/2, \quad (12)$$

where
\[ N_P = \left[ \exp \left( \frac{\tilde{\epsilon}_i - \tilde{\epsilon}_j}{T} \right) - 1 \right]^{-1} \]  

(13)
is Planck’s distribution function. Here, in Eq. (11), “+” and “−” correspond to the cases of phonon emission (\(\tilde{\epsilon}_j < \tilde{\epsilon}_i\)) and absorption (\(\tilde{\epsilon}_j > \tilde{\epsilon}_i\)), respectively.

The effective impedance between the localized sites in question \[ Z_{ij} = \frac{T}{e^{2\gamma_{ij}f_i (1 - f_j)}} \]  

(14)
where \(f_i = \{1 + \exp[(\tilde{\epsilon}_i - E_F)/T]\}^{-1}\) is Fermi’s distribution function of the bound polaron with Fermi level \(E_F\). From (11) - (14) it follows that

\[ Z_{ij} = Z_{ji} \propto \exp \left( \frac{2r_{ij}}{a} + \frac{\epsilon_{ij} + \Lambda_{ij}}{T} \right), \]  

(15)
where

\[ \epsilon_{ij} = \frac{1}{2} (|\tilde{\epsilon}_i - E_F| + |\tilde{\epsilon}_j - E_F| + |\tilde{\epsilon}_i - \tilde{\epsilon}_j|) \]  

(16)
and

\[ \Lambda_{ij} = \begin{cases} 0, & |\tilde{\epsilon}_i - \tilde{\epsilon}_j| > W_i + W_j \\ \frac{1}{4} (W_i + W_j)^2 / (W_i + W_j), & |\tilde{\epsilon}_i - \tilde{\epsilon}_j| \leq W_i + W_j \end{cases} . \]  

(17)
It should be mentioned that Eqs (15) - (17), though resembling those previously obtained in [10, 11], differ from them in one important point. Here the modified BMP energies \(\tilde{\epsilon}_i = \epsilon_i (0) - W_i\) play role of the “bare” electron energies \(\epsilon_i (0)\) used in [10, 11]. In the absence of fluctuations of polaron shifts (\(W_i = W_j = W\)), this substitution is irrelevant because the same constant polaron shift \(W\) is incorporated into \(E_F\) [9]. It is, however, substantial if these fluctuations play a significant role as they do in the case of variable-range hopping of the bound polarons.

Now we are in a position to calculate the electrical resistivity of the system by using the percolation theory approach [2]. We start from the following bonding criterion (see Eq. (15))

\[ \xi_{ij} = \frac{2r_{ij}}{a} + \frac{\epsilon_{ij} + \Lambda_{ij}}{T} \leq \xi. \]  

(18)
Let us introduce the maximum values of \(\tilde{\epsilon}, W,\) and \(r,\) compatible with this criterion,
\[ \tilde{\epsilon}_{\text{opt}} = T\xi, \quad W_{\text{opt}} = 4T\xi, \quad r_{\text{opt}} = a\xi/2, \] (19)
as well as the following dimensionless variables:

\[ s_{ij} = |\vec{s}_{i} - \vec{s}_{j}|, \quad \delta_{ij} = \frac{1}{2}(|\delta_{i}| + |\delta_{j}| + |\delta_{i} - \delta_{j}|), \] (23)

\[ \lambda_{ij}(\varphi) = \begin{cases} 0, & \varphi |\delta_{i} - \delta_{j}| \geq 4(w_{i} + w_{j}) \\ \frac{4(w_{i} + w_{j}) - \varphi |\delta_{i} - \delta_{j}|^{2}}{16\varphi(w_{i} + w_{j})}, & \varphi |\delta_{i} - \delta_{j}| < 4(w_{i} + w_{j}) \end{cases}. \] (24)

In terms of these variables the bonding criterion (18) can be rewritten as

\[ s_{ij} + \delta_{ij} + \lambda_{ij}(\varphi) \leq 1, \] (21)

where

\[ \varphi \] accounts for a possible existence of an upper boundary, \( W_{\text{max}} \), in the distribution of the polaron shifts \( W_{l} \). If, however, the optimal polaron shift \( W_{\text{opt}} = 4T\xi < W_{\text{max}} \), this boundary does not play any role, and one should put \( \varphi = 1 \) in (24). This is true at low temperatures, \( T < T_{1}(d) \) (3), otherwise \( \varphi > 1 \).

Now the following percolation problem can be formulated in the \( d+2 \) dimensional space. There are randomly distributed sites in this space with the dimensionless concentration

\[ \nu_{0}^{(d)}(\xi) = G \cdot 2\tilde{\epsilon}_{\text{opt}} \cdot W_{\text{opt}} \cdot r_{\text{opt}}^{d} = 2^{3-d}Ga^{d}T^{2}\xi^{d+2}, \] (25)
such that each of these sites, \( l \), has two random parameters, \( \delta_{l} \) and \( w_{l} \), uniformly distributed inside the hyperstrip with \( |\delta_{l}| < 1 \) and \( 0 < w_{l} < 1 \). Given the bonding criterion (21) with \( \varphi = 1 \), one should numerically find a minimal (threshold) dimensionless concentration, \( \nu_{c0}^{(d)} = \nu_{c0}^{(d)}(\xi_{c0}^{(d)}) \), at which the percolation onset takes place. If such a concentration is found then, in accordance with Eq. (25), the critical exponent
\[ \xi_{c0}^{(d)} = \left( \frac{\nu_{c0}^{(d)}}{2^{3-d}Ga^dT^2} \right)^{1/(d+2)} \]  

(26)

and therefore the resistivity

\[ \rho = \rho_0 \exp \left( \frac{\xi_{c0}^{(d)}}{\nu_{c0}^{(d)}} \right) \]  

(27)

is given by the previously obtained Eqs (4) and (5), where the dimensionless factor

\[ \beta_0^{(d)} = 2^{d-3} \nu_{c0}^{(d)}. \]  

(28)

If there is a maximal polaron shift \( W_{\text{max}} \) then at high temperatures \( T > T_1^{(d)} \), such that \( W_{\text{opt}} = 4T \xi_{c0}^{(d)} \geq W_{\text{max}} \), all the polaron hopping barriers are accessible. In this case, \( GW_{\text{opt}} = g \) and the dimensionless concentration (25) should be substituted by

\[ \nu^{(d)}_1 (\xi) = g \cdot 2\xi_{\text{opt}} \cdot r_{\text{opt}}^{d} = 2^{1-d} g a^d T \xi^{d+1}. \]  

(29)

Then given the bonding criterion (21) one should find the threshold critical concentration \( \nu_{c1}^{(d)} (\varphi) = \nu_{c1}^{(d)} (\xi_{c1}^{(d)}) \) in the (d+1)-dimension space for any dimensionless temperature \( \varphi = 4T \xi_{c1}^{(d)}/W_{\text{max}} > 1. \)

As far as this concentration is found, the critical exponent can be expressed as

\[ \xi_{c1}^{(d)} (\varphi) = \left[ \frac{2^{d-1} \nu_{c1}^{(d)} (\varphi)}{g a^d T} \right]^{1/(d+1)}. \]  

(30)

So, the parameter \( \varphi > 1 \) can be found as a solution of the following transcendental equation:

\[ \varphi = t \left[ \frac{\nu_{c1}^{(d)} (\varphi)}{\nu_{c1}^{(d)} (\varphi \to \infty)} \right]^{1/(d+1)}. \]  

(31)

Here a dimensionless parameter

\[ t = 4 \left( T^d T_M^{(d)} \right)^{1/(d+1)} \geq t_0^{(d)} \]  

(32)

has been introduced where the Mott’s parameter \( T_M^{(d)} \) is given by Eq (8) with

\[ \beta_M^{(d)} = 2^{d-1} \nu_{c1}^{(d)} (\infty). \]  

(33)
(The minimal value of the parameter \( t_0^{(d)} \), which defines the transition temperature \( T_1^{(d)} \) \((3)\), can be found from Eq \((31)\) by setting there \( \varphi = 1 \).)

For the high-temperature VRH regime \((T > T_1^{(d)})\), the resistivity can be presented in the following form

\[
\rho = \rho_0 \exp \left( \xi_{c_1}^{(d)} \right) = \rho_0 \exp \left[ \left( \frac{T_1^{(d)}}{T} \right)^{\frac{1}{(d+1)}} f_1^{(d)}(t) \right],
\]

It obeys a modified VRH Mott law with

\[
f_1^{(d)}(t) = \varphi(t)/t \tag{35}
\]

being a universal function of the dimensionless parameter \((32)\), where the function \( \varphi(t) \) itself is a solution of the transcendental equation \((31)\). Function \( \varphi(t) \) can be found given the threshold concentration, \( \nu_{c_1}^{(d)} \), as a function of \( \varphi > 1 \). In particular, from \((21)\) - \((24)\) it follows that due to the fact that \( \lambda_{ij}(\varphi) > 0 \) and is a dropping function, \( \nu_{c_1}^{(d)}(\varphi)/\nu_{c_1}^{(d)}(\infty > 1 \) and is a dropping function too. As a result, \( f_1^{(d)}(t) \) is also a dropping function. At high temperatures, such that \( T >> T_1 \), parameter \( t >> 1 \). Then the solution of Eq. \((31)\) can be presented as \( \varphi(t) \simeq t \) and therefore \( f_1(t) \simeq 1 \) in this limit. In this case, the resistivity is governed by the conventional Mott’s law. (Also, from Eq \((24)\) it directly follows that \( \lambda_{ij} \rightarrow 0 \) when \( \varphi \rightarrow \infty \). In this limit, the bonding criterion \((21)\) coincides with that of the standard VRH problem \((2)\).)

It should be noted that the condition \( T = T_1^{(d)} \) \((3)\) is equivalent to \( \xi_{c_0}^{(d)} = \xi_{c_1}^{(d)} \), i.e. the transition from Eq. \((1)\) to Eq. \((34)\) takes place when the widths of the corresponding optimal energy strips, \( \tilde{\epsilon}_{opt} \) are equal to each other and, simultaneously, the optimal polaron shift \( W_{opt} = W_{max} \).

IV. POLARON HOPPING. MONTE CARLO SIMULATIONS

A Monte Carlo simulation procedure has been developed and implemented, which aimed at finding numerical factors \( \beta_0^{(d)}, \beta_M^{(d)} \) and \( \tau_d \) in Eqs \((4)\) and \((7)\), as well as the universal function \( f_1^{(d)}(t) \) and parameter \( t_0^{(d)} \) in Eqs \((34)\) and \((6)\) . This procedure can be described as follows. In a cube or square of a size \( L \), \( N \) randomly distributed sites are planted with an average concentration \( \nu_L = N/L^d \), less than the critical one. Each of these sites, \( l \), is
ascribed two random parameters, $\delta_l$ and $w_l$, such that $|\delta_l| < 1$ and $0 < w_l < 1$. Any two sites, $i$ and $j$, are considered connected (bonded) if they satisfy the pairwise bonding criterion with $\varphi \geq 1$, where $s_{ij}$ is the distance between these sites measured in the same units as $L$. Two sites are said to belong to the same cluster if there is a sequence of connections from the first site, through other sites, to the second one. (Each cluster has its own identification number.) To test for percolation, boundary layers of thickness $b$ are introduced. For a given $\varphi \geq 1$, the code checks if a percolation cluster has been established, which has at least one site belonging to each of a pair of opposite boundary layers. If it has not, new sites are added in small increments and the clusters updated until percolation is detected at a critical (threshold) concentration $\nu_{cL}(\varphi)$. Obviously, the latter depends on a given realization of the percolation cluster, so the procedure was repeated from 50 to 100 times in order to find an average critical concentration $\langle \nu_{cL}(\varphi) \rangle$.

Thus found critical concentrations for several increasing sizes $L$ have been used to determine a thermodynamic limit ($L \to \infty$) of the critical concentration, $\nu_c(\varphi)$, by using the following power-law extrapolation:

$$\langle \nu_{cL}(\varphi) \rangle = \nu_c(\varphi) + AL^{-\alpha},$$  

where $A$ and $\alpha$ are constants. (It has been found that the distribution of $\nu_{cL}(\varphi)$ is Gaussian with dispersion that tends to zero as the size of the system increases.)

Fig. clearly demonstrates that, for example, for $\varphi = 1$ and $\varphi = 1000$, the average critical concentrations as functions of the size $L$ converge to their corresponding thermodynamic limits, $\nu_c(\varphi)$, that do not depend on the thickness of the boundary layer $b$. (In our simulations, the latter has been chosen to be close to $0.2\nu_c^{-1/d}(\varphi)$ and $0.1\nu_c^{-1/d}(\varphi)$ for any given $\varphi \geq 1$. And, as has been explained in [2], for any given $L$, the thicker boundary layer corresponds to a systematically smaller critical concentration.)

In particular, we have found that the critical concentration $\nu_{c}^{(d)}(1) = \nu_{c0}^{(d)}$, that determines the critical exponent describing the conductivity in the VBVRH regime, is equal to $42.19 \pm 0.25$ and $31.25 \pm 0.18$ for 2D and 3D systems, respectively. This yields the following values of the numerical factor, which specify parameter $T_0^{(d)}$ in Eq (4): $\beta_0^{(2)} = 21.09 \pm 0.12$ and $\beta_0^{(3)} = 31.25 \pm 0.18$.

Also, the above described procedure has allowed us to establish values of the critical concentrations in the limit of large $\varphi$: $\nu_{c1}^{(2)}(\infty) = 6.88 \pm 0.08$ and $\nu_{c1}^{(3)}(\infty) = 4.24 \pm 0.03$.  

$$12$$
FIG. 1: Average critical concentrations, $\langle \nu_{cL}(\varphi) \rangle$, as functions of size, $L$, of the $2D$ and $3D$ system calculated for two different thicknesses, $b$, of the boundary layer for $\varphi = 1$ (the upper four curves) and for $\varphi = 1000$ (the lower four curves). The fitted curves are obtained by means of Eq. (36).

These, in turn, determine the numerical factors (33) in the Mott’s law parameters (8): $\beta_M^{(2)} = 13.76 \pm 0.16$ and $\beta_M^{(3)} = 16.96 \pm 0.12$. They can be compared with previously obtained values $\beta_M^{(2)} = 13.8 \pm 1.2$ and $\beta_M^{(3)} = 21.2 \pm 1.2$ [2] (Our somewhat smaller, and more accurate, value of $\beta_M^{(3)}$, can be justified by using up to $3 \times 10^5$ sites in the present simulations vs the previously used $1.5 \times 10^2$ sites [2]). It should be mentioned that our values of $\beta_M^{(d)}$ are in a surprisingly good agreement with the ones ($\beta_M^{(2)} = 13.3$ and $\tau_M^{(3)} = 16.4$) obtained by Ioselevich [19] who applied an approximate multicomponent percolation criterion to analysis of both the NNH and VRH problems.

Fig. 2 shows the calculated universal function (35) which describes a continuous transition from the VRVVBH regime of polaron conductivity (4) to the standard Mott regime. For $t \geq t_0$, it can be approximated as
FIG. 2: Universal function $f_1^{(d)}(t)$ found by means of the Monte Carlo simulations (circles). The solid curves correspond to extrapolation formula (37).

| $d$ | $a$  | $b$  | $c$   | $t_0$ | $\beta_0$ | $\beta_M$ | $\tau$ |
|-----|------|------|-------|-------|------------|------------|--------|
| 2   | 0.967| 0.756| -0.157| 0.546 | 21.1       | 13.8       | 0.189  |
| 3   | 0.982| 0.694| -0.178| 0.607 | 31.2       | 17.0       | 0.174  |

TABLE I: Parameters of the universal function $f_1^{(d)}(t)$.

\[ f_1^{(d)}(t) = a + bt^{-1} + ct^{-2}. \]  \hfill (37)

Fitting parameters for 2D and 3D cases, as well as parameters $\tau_d = b/4$ from Eq. (7) and other relevant parameters can be found in Table I.
V. LOW TEMPERATURES. MINIMUM POLARON BARRIER EFFECT

In certain disordered systems, such as doped semiconductors close to the metal-insulator transition, the distribution of polaron shifts, being broad on its high side, may be very sharp on its low side\[11\]. It means that a system may have some minimal polaron shift, $W_{\text{min}}$. In this case, the law \[\text{(4)}\] is also violated at low temperatures when the optimal polaron shift becomes smaller than the minimal one: $W_{\text{opt}} = 4T_{c_0}^{(d)} < W_{\text{min}}$, i.e. at $T < T_3$, where

$$T_3^{(d)} \simeq (W_{\text{min}}/W_{\text{max}})^{(d+1)/d}T_1^{(d)}.$$  

(38)

where $T_1^{(d)}$ is given by Eq \[\text{(3)}\]. (The regime described by Eq. \[\text{(27)}\] can be observed within a temperature interval $\text{max}(T_2^{(d)}, T_3^{(d)}) < T < T_1^{(d)}$ that clearly exists if $(W_{\text{max}} - W_{\text{min}})/\epsilon C >> 1$.) At such low temperatures, $W_{\text{min}}/2$ is the only polaron barrier accessible and the “bare”, $\epsilon_l(0)$ or the modified, $\tilde{\epsilon}_l = \epsilon_l(0) - W_{\text{min}}$, electron energy is the only fluctuating variable. The connectivity criterion \[\text{(18)}\] can be rewritten as $\xi_{ij} = \xi_{ij}^0 + \Delta \xi_{ij} < \xi + \Delta \xi_{ij}$, where $\Delta \xi_{ij} = W_{\text{min}}/2T$ and

$$\xi_{ij}^0 = \frac{2r_{ij}}{a} + \frac{\epsilon_{ij} + \Lambda_{ij}^0}{T}.$$  

(39)

Here

$$\Lambda_{ij}^0 = \begin{cases} -W_{\text{min}}/2, & |\tilde{\epsilon}_i - \tilde{\epsilon}_j| > 2W_{\text{min}} \\ (\tilde{\epsilon}_i - \tilde{\epsilon}_j)^2/8W_{\text{min}} - |\tilde{\epsilon}_i - \tilde{\epsilon}_j|/2, & |\tilde{\epsilon}_i - \tilde{\epsilon}_j| \leq 2W_{\text{min}} \end{cases}.$$  

(40)

After introducing the maximum values of $\tilde{\epsilon}_i$ and $\tilde{\mathbf{r}}_i$ (see \[\text{(19)}\]), compatible with the criterion $\xi_{ij}^0 < \xi$, as well as the dimensionless variables $\tilde{\mathbf{r}}_i$ and $\delta_i$ (Eq. \[\text{(20)}\]), the following percolation problem can be formulated (see also \[\text{(11)}\]). For any given dimensionless temperature, $\theta = 2T_{c_2}^{(d)}/W_{\text{min}}$, one should find the critical dimensionless concentration, $\nu_{c_2}^{(d)}(\theta)$, when percolation first appears given the connectivity criterion

$$s_{ij} + \delta_{ij} + \lambda_{ij}^0(\theta) \leq 1$$  

(41)

where

$$\lambda_{ij}^0(\theta) = \begin{cases} -1/\theta, & \theta |\delta_i - \delta_j| \geq 4 \\ \theta (\delta_i - \delta_j)^2/16 - |\delta_i - \delta_j|/2, & \theta |\delta_i - \delta_j| < 4 \end{cases}.$$  

(42)
and assuming that $\delta_{\ell}$ are uniformly distributed within the strip $|\delta_{\ell}| < 1$. When such critical concentration is known, then the critical exponent, $\xi^{(d)}_{e2}$, is given by Eq. (30) where $\nu^{(d)}_{e1}(\varphi)$ should be substituted by $\nu^{(d)}_{e2}(\theta)$. The resistivity can be presented in the following form

$$\rho = \rho_0 \exp \left[ \frac{W_{\text{min}}}{2T} + \xi^{(d)}_{e2}(\theta) \right] = \rho_0 \exp \left[ \frac{W_{\text{min}}}{2T} f^{(d)}_2(\eta) \right],$$

where

$$\eta = \frac{2 \left( T^d T^{(d)}_M \right)^{1/(d+1)}}{W_{\text{min}}}.\quad (44)$$

Here $T^{(d)}_M$ is given by Eq. (8) and $f^{(d)}_2(\eta) = 1 + \theta(\eta)$. Function $\theta(\eta)$ is a solution of the transcendental equation

$$\theta = \eta \left[ \frac{\nu^{(d)}_{e2}(\theta)}{\nu^{(d)}_{e2}(\theta \to \infty)} \right]^{1/(d+1)},$$

where $\nu^{(d)}_{e2}(\theta \to \infty) = \nu^{(d)}_{e1}(\varphi \to \infty)$ has been calculated in the previous section.

As it follows from Eq. (12), $\lambda_{ij}^0(\theta) < 0$ and is a rising function. As a result, $\nu^{(d)}_{e2}(\theta) / \nu^{(d)}_{e2}(\infty) < 1$ and is a rising function. Therefore, in Eq. (43), $f^{(d)}_2(\eta)$ is a rising function as well. In addition, if $\theta \to 0$, $\nu^{(d)}_{e2}(\theta) / \nu^{(d)}_{e2}(\infty) \to \varsigma_d < 1$. Then at low temperatures, when $\eta(44) \ll 1$, $f^{(d)}_2(\eta) \simeq 1 + \varsigma_d^{1/(d+1)} \eta$ which yields

$$\rho = \rho_0 \exp \left[ \frac{W_{\text{min}}}{2T} + \frac{\varsigma_d T^{(d)}_M}{T} \right]^{1/(d+1)}.$$

(46)

This almost activation type of conductivity (for the second term in the exponent is much smaller than the first one) will be observed if $T << T^{(d)}_3$.

The Monte Carlo simulations allowed us to calculate the universal function $f^{(d)}_2(\eta)$ in Eq. (43), which can be approximated as

$$f^{(d)}_2(\eta) = 1 + \eta \left[ 1 - A \exp (-B \eta) \right].$$

(47)

Fitting parameters for this formula and constant $\varsigma_d = (1 - A)^{d+1}$ from Eq. (46) are presented in Table I.

And finally, if $W_{\text{max}} - W_{\text{min}}$ is smaller than the width $\epsilon_{opt} (3)$ of the Mott strip, the VRVBH regime, described by Eq. (11) ceases to exist. The fluctuations of polaronic shifts are not important anymore ($W_{\text{max}} \simeq W_{\text{min}}$), and, if the Coulomb correlation effects are
not substantial \((W_{\text{max}} >> \epsilon_C)\), the resistivity is described by Eq. (43) at all relevant temperatures. It is interesting to mention that in the high-temperature case \((t \simeq \eta >> 1)\), when \(f_1^{(d)}(t) \simeq 1\) and \(f_2^{(d)}(\eta) \simeq \eta\), both formulas (34) and (43) lead to practically the same modified Mott law.

\section*{VI. APPLICATION TO GIANT MAGNETORESISTANCE}

In the previous sections, we described a new mechanism of polaron conductivity controlled by variable-range hopping which leads to a non-activation temperature dependence

\[ \rho = \rho_0 \exp \left[ \left( \frac{\tilde{T}}{T} \right)^p \right] \quad (48) \]

with \(p = 2/(d + 2)\). This result, which differs from the standard Mott’s \(p = 1/(d + 1)\), can be justified for both the lattice and the spin polarons if their polaron shifts (barriers) fluctuate independently from their bare electron energies. However, the case of bound spin (or magnetic) polarons is of special interest because not only the polaron shifts but their distribution can be altered under the action of the external magnetic field, \(H\). It can be shown, that, indeed, the combined density of polaron states, \(G\), which defines parameter \(\tilde{T}\) in Eq. (48) (see Eqs (4) and (5)), will decrease with magnetic field. As a result, the resistivity will sharply (exponentially) decrease with magnetic field, so that Eq. (4) also describes giant (or colossal) negative magnetoresistance observed in dilute magnetic semiconductors and nanostructures.

To demonstrate this, let us suppose that, for the sake of simplicity, the “bare” electron energies \(\epsilon\) and the polaron shifts \(W\) and are not correlated and both are distributed uniformly, each in the interval \((\bar{\epsilon} - \Delta \epsilon, \bar{\epsilon} + \Delta \epsilon)\) and \((W_{\text{min}}, W_{\text{max}})\), correspondingly. Then the density of the “bare” electron states

\begin{table}[h]
\centering
\caption{Parameters of the universal function \(f_2^{(d)}(\eta)\).}
\begin{tabular}{ccc}
\hline
\(d = 2\) & \(A\) & \(B\) & \(\zeta\) \\
\hline
\(d = 2\) & 0.137 & 0.0759 & 0.643 \\
\hline
\(d = 3\) & 0.109 & 0.0612 & 0.630 \\
\hline
\end{tabular}
\end{table}
\[ g = \begin{cases} 
0, & |\epsilon - \overline{\epsilon}| \geq \Delta \epsilon \\
 g_0 = N_t / 2 \Delta \epsilon, & |\epsilon - \overline{\epsilon}| < \Delta \epsilon 
\end{cases} \]  
(49)

while the density of the BMP shifts

\[ g_W = \begin{cases} 
0, & |W - \overline{W}| \geq \Delta W \\
 N_t / 2 \Delta W, & |W - \overline{W}| < \Delta W 
\end{cases} \]  
(50)

where \( N_t \) is the concentration of the sites and \( 2 \Delta W = W_{\text{min}} - W_{\text{max}} \), \( \overline{W} = (W_{\text{min}} + W_{\text{max}}) / 2 \). For the chosen distributions,

\[ G \simeq \frac{N_t}{2 \Delta \epsilon \cdot 2 \Delta W} = \frac{g_0}{2 \Delta W} = \frac{g_0 g_W}{N_t} \]  
(51)

given \( \Delta \overline{\epsilon} \simeq \Delta \epsilon \gg \Delta W \).

For bound spin (magnetic) polarons, their shifts, \( W \), proportional to the magnetic susceptibility, \( \chi(H, T) \) \[9, 10, 11\]. The same is true for the width, \( 2 \Delta W = W_{\text{min}} - W_{\text{max}} \), of the shift distribution (50). Therefore, by aligning the atomic spins the applied magnetic field not only reduces the polaron hopping barriers but also makes their distribution sharper. Then, from Eqs. (4) and (51) it follows that parameter \( \overline{T}_0^{(d)} \) in expression (4)

\[ \overline{T}_0^{(d)}(H) \propto \sqrt{\chi(H, T)} \]  
(52)

It substantially decreases with magnetic field when the magnetization approaches its saturation value thus leading to giant or colossal negative magnetoresistance.

If the material is in the paramagnetic phase and if the anti-ferromagnetic coupling between the magnetic atoms is suppressed, then the susceptibility in Eq. (52) obeys the Curie law: \( \chi(H, T) \propto 1/T \). In this case, the exponent \( p = 2/(d + 2) \) in expression (52) describing VRVBH regime should be substituted with a somewhat larger quantity \( p = 3/(d + 2) \). It can be seen that in this regime for 3D systems, \( 0.4 < p < 0.6 \). That kind of dependence can be easily confused with the standard \( p = 0.5 \) typical for electron VRH in the presence of Coulomb correlations \[2\]. (The effect of these correlations on VRH of the spin and lattice polarons will be considered elsewhere. In particular, it can be shown that in this case \( p = (d + 1) / (2d + 1) \) or \( p = (d + 2) / (2d + 1) \) in the paramagnetic phase.) However, at high magnetic fields (for spin polarons) the \( p = 2/(d + 2) \) or \( p = 3/(d + 2) \) VRVBH dependence
(for the spin polarons) will be replaced by the \( p = 1/(d + 1) \) Mott’s law while at the same conditions the presence of the soft Coulomb gap will result in the \( p = 0.5 \) dependence.

Giant negative magnetoresistance in the variable-range hopping region apparently has been observed in recently discovered dilute magnetic semiconductors, such as GaAs:Mn [5, 6, 7] and MnGe [21] below the metal-insulator transition point. In all these materials, magnetoresistance is described by Eq. (48) with \( p = 0.25 \div 0.56 \) and \( \tilde{T} \) significantly decreasing with magnetic field. Additional, more detailed measurements of the transport and magnetic properties in question are needed to more specifically identify and quantitatively describe the hopping mechanism in these materials. Also, the effect of the soft Coulomb gap should be taken into account when describing electrical conductivity governed by variable-range polaron hopping at very low temperatures.

VII. CONCLUSIONS

We have examined variable-range hopping of polarons in polar and/or magnetic disordered materials. In addition to electron impurity band effects, where site positions and the ”bare” electron energies are randomly scattered, we took into account that the polaron shifts may be widely distributed as well. It results in a significant modification of the law that governs the low-temperature electrical conductivity in such materials. In the absence of Coulomb correlation effects, the standard Mott dependence should be substituted by another, non-activation one, given by Eq. (4), which describes the variable-range variable-barrier hopping of lattice or spin polarons.

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APPENDIX A: POLARON HOPPING RATE

Here we will derive Eq. (11) for the polaron hopping rate. We will use a semi-classical approach [12] to describe lattice polaron hopping in the framework of a generalized molecular-
crystal model [13]. We will further generalize this model order to take into account that the initial and final states may have different polaron shifts.

In the Holstein’s occurrence probability approach, the polaron hopping rate between the initial state $i$ and the final state $j$ can be written as [13, 15]

$$
\gamma_{ij} = \int_{-\infty}^{\infty} d\hat{E} \ P(\hat{E}) \left| \hat{E} \right| \left\langle \delta(\epsilon_j - \epsilon_i) \delta\left(\epsilon_j - \epsilon_i - \hat{E}\right) \right\rangle. \quad (A1)
$$

Here

$$
\epsilon_i = \epsilon_i(0) - \sum_{n=1}^{L_i} A_n^{(i)} x_n, \quad \epsilon_j = \epsilon_j(0) - \sum_{n=1}^{L_j} A_n^{(j)} y_n \quad (A2)
$$

are the local electron energies of the initial and final states coupled to local vibrational modes $x_n$ and $y_n$ with $A_n^{(i,j)}$ being the coupling constants ($L_l$ is the total number of the vibration modes coupled to the localized carrier at $l$-th site) and

$$
\dot{E} = (\epsilon_j - \epsilon_i)_{\epsilon_j = \epsilon_i} \quad (A3)
$$

being the time rate of change of the relative electronic energies evaluated at the event when the electronic terms coincide. In Eq.(A1), the brackets indicate a thermal average over all the configurational coordinates $x_n$ and $y_n$ and the velocities $\dot{x}_n$ and $\dot{y}_n$; $P(\hat{E})$ is the probability that the carrier will perform a hop at the coincidence event.

By using the integral representation of the $\delta$-functions and the harmonic approximation for the vibrational energy (see Ref. [13] for details) one can perform averaging in Eq.(A1) and rewrite it as

$$
\gamma_{ij} = \left[ 4\pi T \sqrt{(W_i^2 + W_j^2) (W_i + W_j)} \right]^{-1} \exp \left\{ - \left[ (\Delta + 2W_i)^2 \right] / 4 (W_i + W_j) - J_{ij} \right\} / T \right\}
\times \int_{-\infty}^{\infty} d\hat{E} \ P(\hat{E}) \left| \hat{E} \right| \exp \left[ - \hat{E}^2 / 4T (W_i^2 + W_j^2) \right], \quad (A4)
$$

where $\Delta = \epsilon_j(0) - \epsilon_i(0)$ is the electron energy difference between the carrier-free states; $J_{ij} = J_0 \exp (-r_{ij}/a)$ is the hopping integral;

$$
W_l = \sum_{n=1}^{L_l} (A_n^{(l)})^2 / 2k_n^{(l)}, \quad \Omega_l^2 = W_l^{-1} \sum_{n=1}^{L_l} (\omega_n^{(l)} A_n^{(l)})^2 / 2k_n^{(l)} \quad (A5)
$$
are the polaron shift and the squared average vibrational frequency of the $l$-th site, correspondingly. Here $\omega_n^{(l)}$ and $k_n^{(l)}$ are respectively the frequency and the stiffness constant of $n$-th vibrational mode that belongs to site $l$.

In the so called non-adiabatic limit, when the hopping integral $J \to 0$, the probability
\[ P \left( \dot{E} \right) = 2\pi J_{ij}^2 / \hbar \dot{E} < < 1, \]
and therefore
\[ \gamma_{ij}^{(n-ad)} = \frac{|J_{ij}|^2}{\hbar} \sqrt{\frac{\pi}{(W_i + W_j) T}} \exp \left[ -\frac{(\Delta + 2W_i)^2}{4 (W_i + W_j) T} \right]. \]
which coincides with Eq.(A5). Otherwise, in the adiabatic limit (see [13] for details),
\[ \gamma_{ij}^{(ad)} = \frac{1}{2\pi} \sqrt{\frac{W_i \Omega_i^2 + W_j \Omega_j^2}{W_i + W_j}} \exp \left\{ -\left[ (\Delta + 2W_i)^2 / 4 (W_i + W_j) - J_{ij} \right] / T \right\} \]
When the vibrational and coupling constants of the initial and final sites are the same, $W_i = W_k = W$, $\Omega_i = \Omega_j = \Omega$, the above expressions for the hopping rate coincide with those obtained in Ref. [13]. An expression for the hopping rate, which is similar to Eq.(A7) but with a different pre-exponential factor, can be obtained for the bound magnetic polarons by using a procedure described in Ref. [11]. (This factor properly takes into account the vector nature of the order parameter (magnetization) and can be responsible for the giant positive hopping magnetoresistance that may be observed at low magnetic fields [16]. However, at high magnetic fields it coincides with that from Eq. (A7).)

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