Spin Polarons and Variable Range Hopping in Magnetically Disordered Systems

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Magnetic polarons in magnetically disordered systems (for brevity, spin-glass polarons) are in one respect very unlike other polarons: Usual polaron hopping at low T proceeds via tunneling, and these polarons do not affect the T dependence of the hopping rate, while the spin-glass polaron tunneling is suppressed by the disorder, and their hopping proceeds via activation for all T. It gives rise to the reentrance of the simple activated T dependence of variable range hopping conductivity at very low T. Recent experiments in CdMnTe are discussed.

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Recent findings [1,2] of a simple activation T dependence of conductivity σ at very low T in a few semiconductors has renewed interest in the "magnetic hard gaps," which were observed in a number of substances [3]. The essence of this phenomenon is as follows.

At low T the $\sigma(T)$ is controlled by a variable range hopping (VRH) [4], which gives

$$\sigma(T) \propto \exp[-(T_0/T)^{\alpha}], \quad (1)$$

with either $\alpha = \frac{1}{4}$ (Mott law [5]), or $\alpha = \frac{1}{2}$ [Efros-Shklovskii (ES) law [6]], depending on the shape of a density of states $N(\varepsilon)$ near the Fermi level ε_F. The Mott law corresponds to $N(\varepsilon_F) = \text{const}$, while the ES law implies the soft Coulomb gap [7]: $N(\varepsilon) \propto (\varepsilon - \varepsilon_F)^2$. Both laws were observed experimentally in a variety of materials (see [4,8]). In some cases, however, the reentrance of a usual activation law,

$$\sigma(T) \propto \exp(-E_H/T), \quad (2)$$

was observed at very low T [1–3]. This reentrance effect was removed in a strong magnetic field [1,2,9], where the VRH law (1) persisted at the lowest T studied [10].

It is tempting to relate the law (2) to the hard gap in the density of states, i.e., $N(\varepsilon) \approx 0$ for $|\varepsilon - \varepsilon_F| < \Delta_H$. The Coulomb interaction can give rise to a hard gap [11], but in our case it should be due to a coupling of electrons to magnetic degrees of freedom [9], which can be frozen by the field. However, as we will see, this simple idea is tricky, and it is rather good luck that this scenario (though in a modified form) survives in some cases.

In this Letter we consider only a semimagnetic semiconductor $Cd_{1-x}Mn_xTe$:In (with $x = 0.09$), for which a reentrance effect at low $T < 1$ K was observed in a wide range of donor concentrations [1], and which is the simplest model for the phenomenon under study. The usual explanation of data [1] lies within a frame of the above hard-gap scenario: An electron, bound to a particular In donor, polarizes the spins of Mn^{2+} ions within a radius ξ of a localized state, forming a bound magnetic polaron (BMP) (the existence of a BMP in this compound is well established; see [12]). Adding a *bare* electron to the system (or removing it, or shifting it to another donor, where the neighboring spins are not polarized) always costs the energy of spin relaxation. This provides a hard gap to the electronic density of states and an activational character to the hopping [1].

The very generality of the above picture makes it somewhat suspicious. Actually it can be as well applied to any kind of polaron: a usual lattice polaron, or so-called "electronic polaron" (see [7,13,14]). Indeed all kinds of polarons give rise to a hard gap in an electronic single-particle density of states $N_{sp}(\varepsilon)$. Nevertheless, for the case of lattice polarons it is well known that for low $T < T_c \sim \omega_{ph}$ (ω_{ph} being a characteristic phonon frequency), the polaron hopping proceeds via tunneling but not activation, and an activational exponent $\exp(-W/T)$ is replaced by a T-independent one $\exp(-cW/\omega_{ph})$ in the expression for a hopping rate Γ_{ij} [15]. The single-particle density of states N_{sp} is irrelevant for the hopping at low T, since it is the polaron that hops, not a bare electron. In addition to the theoretical arguments there are also experimental observations of a law (1) in the systems, where the existence of polarons was well established (see [4]). The same arguments were extended [7,13,14] to the case of an electronic polaron [16].

It should be noted that the self-consistent many-body theory of VRH is still lacking. The very concept of long single-electron (or polaron) hops is subject to criticism [11]. Apparently, the choice between long single-electron hops [6,7] and correlated multielectron short hops [17] cannot yet be made. Although the resolution of this controversy is generally very important, it is not essential for the problem of the magnetic hard gap. So we will basically ignore all the multielectron effects.

The case of a magnetic polaron in a regular antiferromagnet does not differ considerably from the case of a lattice polaron. The characteristic magnon frequency $\omega_{mag} \sim T_N$ plays the role of ω_{ph}. Indeed the Mott law was observed in antiferromagnetic semiconductors (where the existence of BMP was reliably established [4,18]), well below the Néel temperature T_N.

For all the systems considered above the polaronic

effect does produce a hard gap in $N_{sp}(\varepsilon)$, but this gap is not manifested in the T dependence of σ.

Fortunately an activational scenario can be saved when the magnetic subsystem is disordered. It is known that disordered magnets are characterized by an extremely slow relaxation of magnetization, both homogeneous and inhomogeneous [19]. Semimagnetic semiconductors demonstrate the spin-glass-like properties even for low concentrations of magnetic ions (see [20] and references therein). The direct measurement [21] of a bound polaron formation rate gave $\tau_R^{-1} \sim 2\times 10^{-2}$ K at $T \sim 1$-2 K. Note that it is much less than the characteristic interactions in the system. For comparison, the same rate for a lattice polaron is $\tau_R^{-1} \sim \omega_{ph} \sim 10^2$ K. The point is that the magnitude of τ_R^{-1} determines the crossover temperature T_c between activation and tunneling. Indeed the excitations with frequencies $\omega_\lambda \sim \tau_R^{-1}$ should dominate in the system. If one considers roughly these excitations as noninteracting ones, then each of them has to penetrate its own barrier W_λ and contributes a factor $\exp\{-4W_\lambda/\omega_\lambda \tanh(\omega_\lambda/4T)\}$ to Γ_{ij} (see [4]). Then it is clear that tunneling can dominate only if $T < \tau_R^{-1}$. We believe that the interaction of excitations, which certainly should be important in spin glasses, does not change the above consideration qualitatively.

So for the magnetic polarons in disordered magnets, contrary to all other kinds of polarons, the dynamic relaxation of a polaron is blocked even at low T and hence the tunneling never wins in competition with activation. It enables one to treat the hopping in such a system in a classical way (as in the case of noninteracting spins [22]).

Consider a simple model, consisting of electrons, hopping via donors embedded in a magnetic medium (spins $\mathbf{S}_n$). The Hamiltonian of such a system has a form

$$\hat{H} = \sum_i [\hat{H}_0^{(i)} + (\varepsilon_i + \hat{H}_{int}^{(i)}) a_i^\dagger a_i] + \sum_{i,j} I_{ij} (a_i^\dagger a_j + a_j^\dagger a_i) , \quad (3)$$

where $a_i^\dagger$ are the operators, creating an electron on the donor i (their spin indices are omitted; see discussion below); the double occupancy of donors is forbidden. $I_{ij} \propto \exp(-|\mathbf{r}_i - \mathbf{r}_j|/\xi)$ is a "bare" electronic hopping integral and ε_i are the random shifts of electron energies. Since we are interested only in low-T properties, we have to sum in (3) only over those rare donors which are very close to the Fermi level. It enables one to divide all the magnetic medium into the blocks ("submedia") adjacent to each donor and to neglect the cross interaction of the submedia. The Hamiltonian of the submedium of donor i is $\hat{H}_0^{(i)}$ in the absence of an electron on that donor, and $\hat{H}_{int}^{(i)}$ is an electron-submedium interaction:

$$\hat{H}_0^{(i)} = \sum_{n,n'} J_{n,n'} \mathbf{S}_n \mathbf{S}_{n'}; \quad \hat{H}_{int}^{(i)} = \sum_n \mathbf{h}_n^{(i)} \mathbf{S}_n , \quad (4)$$

where $J_{n,n'}$ are the intramedia exchange constants and $\mathbf{h}_n^{(i)}$ are the local mean fields, accounting for the interaction between $\mathbf{S}_n$ and the spin of a localized electron σ.

There are two important time scales in the problem: One is τ_{hop}—a characteristic lifetime of an electron at a given donor, and another is τ_R—a relaxation time for the spin medium. We consider only the case $\tau_R \ll \tau_{hop}$, when the spin medium has enough time to develop a partial thermodynamic equilibrium for any momentary electronic configuration. The authors of [1] have estimated $\tau_R \sim 300$ ps, $\tau_{hop} > 600$ ps for $T \sim 1$ K and the inequality may be improved still further at lower temperatures, since $\tau_{hop} \propto \exp(E_H/T)$ increases at $T \to 0$ stronger than τ_R (see [19]). Since the dynamics of spin degrees of freedom is frozen on the time scale of interest, we can treat the *local* magnetization $\mathbf{M}_n$ (but not only a total one) as conserved. We assume also that the mean fields $\mathbf{h}_n$ are small and the media can be characterized by the linear susceptibility $\chi(T,H)$. Then the rate of hops $\Gamma_{ij} \propto \exp(-\xi_{ij})$, where $\xi_{ij} = 2r_{ij}/\xi + \varepsilon_{ij}/T$ and

$$\varepsilon_{ij} = ||\varepsilon_i|| + ||-\varepsilon_j|| + \min_{\{\mathbf{M}\}} \left\{ \sum_n \left[\frac{(\mathbf{M}_n^{(i)} - \chi \mathbf{h}_n^{(i)})^2}{2\chi} + \frac{(\mathbf{M}_n^{(j)})^2}{2\chi} \right] + \left| \left| \varepsilon_j - \varepsilon_i + \sum_n (\mathbf{h}_n^{(i)} \mathbf{M}_n^{(i)} - \mathbf{h}_n^{(j)} \mathbf{M}_n^{(j)}) \right| \right| \right\}, \quad ||x|| \equiv x\theta(x) \quad (5)$$

(see [8,22]). The first two terms in (5) are the contributions of filling factors $n_i(1-n_j)$ and the third term comes from the optimal hop probability. It consists of two parts: The first part describes the probability of a given fluctuation to occur, the second one is the probability of a phonon absorption at a given fluctuation (a usual electron-phonon interaction is implied). Since $||x||$ has a kink at $x=0$, one should explore the three types of minima in (5), corresponding to the negative, positive, and zero arguments of $||x||$. The first two describe phonon-assisted hops (with emission or absorption of a phonon, respectively), and the third one corresponds to the "fluctuational hops" [22]. Simple calculations give

$$\varepsilon_{ij} = \frac{|\varepsilon_j| + |\varepsilon_i|}{2} + \begin{cases} \dfrac{|\varepsilon_j - \varepsilon_i|}{2} & (|\varepsilon_j - \varepsilon_i| > \Delta_j + \Delta_i, \text{ phonon-assisted hops}), \\ \dfrac{\Delta_j + \Delta_i}{4} + \dfrac{(\varepsilon_j - \varepsilon_i)^2}{4(\Delta_j + \Delta_i)} & (|\varepsilon_j - \varepsilon_i| < \Delta_j + \Delta_i, \text{ fluctuational hops}), \end{cases}$$

where the polaronic shift $\Delta_i(T,H) = \chi(T,H) \sum_n \mathbf{h}_n^{(i)2}/2$ was introduced. If the quenched fluctuations of Δ_i can be neglected [i.e., $\Delta_i \equiv \Delta_{pol}(T,H)$], then the problem of a network's dc conductivity can be reduced (see, e.g., [8]) to a percolation problem in 4D space ($\mathbf{r}_i$ and ε_i) with a connectivity condition $\xi_{ij} < \xi_c$.

For relatively high T such that the Mott energy strip's width is larger than a magnetic hard gap, $\Delta_{\rm Mott}(T) \sim T(T_0/T)^a \gg \Delta_{\rm pol}$, we arrive at the conventional VRH law (1). In the opposite case, when $\Delta_{\rm Mott}(T) \ll \Delta_{\rm pol}$, one has $\varepsilon_{ij} \approx (\Delta_{\rm pol} + |\varepsilon_j| + |\varepsilon_i|)/2$, which leads to

$$\sigma(T) \propto \exp[-E_H/T - (\tilde{T}_0/T)^a], \quad (6)$$

where the second term in the exponent is small compared to first one and $\tilde{T}_0 = \lambda(a) T_0$, $\lambda(a) < 1$ being a universal numerical factor. The activation energy $E_H(T,H) = \Delta_{\rm pol}/2$ corresponds to the "optimal hops" which occur together with a fluctuation of magnetization, equalizing the electronic energy levels on the initial and final donors (see [4,22]). This energy is 4 times less than that for the hops occurring in the equilibrium magnetization, $\bar{E}_H = 2\Delta_{\rm pol}$. Note that if we held to a straightforward hard-gap ideology, we would obtain just the latter overestimated value for E_H.

A general formula covering the intermediate region cannot be derived analytically. While in the limiting cases the percolation problem can be finally reduced to the determination of numerical factors, entering T_0 and $\tilde{T}_0$, in the general case one has to determine numerically an entire universal function f_a of a dimensionless parameter $\eta = (T/E_H)(T_0/T)^a$. In terms of this function the conductivity can be written as

$$\sigma(T) \propto \exp\{-[E_H(T)/T] f(\eta)\}. \quad (7)$$

The asymptotics of $f_a(\eta)$ can be extracted from (1) and (6): $f_a(\eta) \approx 1 + \lambda^a(a)\eta$ if $\eta \ll 1$ and $f_a(\eta) \approx \eta$ if $\eta \gg 1$.

The effect of magnetic field is trivial: It suppresses χ and hence E_H. If H is close to saturation, then $\chi \to 0$ and the effects of the BMP vanish altogether.

We now discuss the realization of the above model in the case of real $Cd_{0.91}Mn_{0.09}Te$:In. Magnetic ions Mn^{2+} substitute randomly Cd ions of the CdTe matrix and form a random network of antiferromagnetically interacting spins. The thresholds for the percolation through the first nearest, second nearest, and third nearest Mn are $x_{c1} \approx 0.195$, $x_{c2} \approx 0.136$, and $x_{c3} \approx 0.061$, respectively (see [20] and references therein). The corresponding exchanges $J_1 \approx 6$ K, $J_2 \approx 1$ K; J_3 is so far unknown but can be estimated as $J_3 \sim 0.3$ K. The magnetization measurements [23] show that there is a spin-glass transition (or, at least, a saturation of the susceptibility) with $T_g \sim 0.3$ K for $x \approx 0.1$. Then for the low-T susceptibility per magnetic ion we can use an estimate $\chi \sim S(S+1) \times X_{\rm eff}(x)/3T_g \sim 1\ {\rm K}^{-1}$, where $X_{\rm eff}(x)$ is a factor, excluding spins, participating in strongly coupled clusters [$X_{\rm eff}(0.09) \approx 0.2$ for the first and second nearest neighbor clusters; see [20]]. A contact interaction of an electron's spin σ with the manganese spins $\mathbf{S}_n$ is

$$\hat{H}_{\rm int} = J\Omega_0 \sum_n (\mathbf{S}_n \sigma) \delta(\mathbf{r} - \mathbf{r}_n), \quad (8)$$

where $\Omega_0 = 270$ Å^3 is a unit cell volume, $J \approx 2640$ K [12], $\mathbf{r}$ is an electron's coordinate, and $\mathbf{r}_n$ are positions of magnetic ions. Within a mean-field approximation one can reduce the Hamiltonian (8) to the form (4) with $\mathbf{h}_n^{(i)} \approx \frac{1}{2} J\Omega_0 |\psi_i(\mathbf{r}_n)|^2 \mathbf{n}_i$, where $\psi_i(\mathbf{r})$ is a wave function of an electron bound to a donor i and $\mathbf{n}_i$ is a unit vector in a direction of BMP magnetization. Since the rate of electronic spin flips is higher than τ_R^{-1} (see [12]), the coherence of electronic spin is destroyed on the time scale $\tau_{\rm hop} \gg \tau_R$. This fact has enabled us to omit the spin indices of the electronic operators in (3).

First consider the case when the concentration of active donors n is low and the system is far from the insulator-metal transition (IMT). Then $\psi_i(\mathbf{r}) = \psi_0(\mathbf{r} - \mathbf{r}_i)$ is a usual hydrogenlike ground-state wave function with the Bohr radius $a_B \approx 60$ Å $(= \xi)$, and

$$E_{H(0)} = \frac{\chi J^2 \Omega_0^2}{16} \sum_n |\psi_i(\mathbf{r}_n)|^4 \approx \frac{x\chi J^2 \Omega_0}{16} \int |\psi_0(r)|^4 d^3 r = \frac{x\chi J^2}{16} \frac{\Omega_0}{\pi a_B^3} \approx 30\ {\rm K},$$

for $T \lesssim T_g$. We have neglected the quenched fluctuations of Δ_i, since a number of spins within a Bohr orbit $N \sim x\pi a_B^3/\Omega_0 \sim 200$ and hence the relative variation of Δ_i is small $(\sim N^{-1/2})$. For $N \sim 1$ one cannot expect the low-T activation at all because a considerable fraction of donors do not exhibit any BMP effect. A large $|\mathbf{h}_n|_{\max} = (J/2)(\Omega_0/\pi a_B^3) \approx 0.5\ {\rm K} \sim T_g$ hints that a linear approximation fails; the nonlinear effects should suppress E_H.

Actually the above results cannot be compared to the experimental data [1] directly, since the estimates [1] give $\xi \gg a_B$, which implies the closeness of the IMT. To interpret these data one is tempted to just substitute ξ instead of a_B, but the situation is more complex. Indeed, near IMT an electron is not bound to one particular donor, but rather is spread over a large number of donors: $\psi_{\langle i \rangle}(\mathbf{r}) = \sum_{i'} \phi_{ii'} \psi_0(\mathbf{r} - \mathbf{r}_{i'})$. The label $\langle i \rangle$ now means something like the "central donor of a distribution." Its precise meaning is, fortunately, not very essential, since a characteristic hop length $\sim \xi(T_0/T)^a \gg \xi$. The polaronic shift for the state centered at the donor i is

$$\Delta_i = 2E_{H(0)} \Lambda_i, \quad \Lambda_i = \sum_{i'} |\phi_{ii'}|^4 < 1.$$

A factor Λ_i (an inverse "participation ratio") strongly suppresses a polaronic effect in the vicinity of the IMT. The magnitude of $|\mathbf{h}_n|$ decreases also, thus improving the linearity conditions.

It is known that $\phi_{ii'}$ has a fractal structure near the IMT of Anderson type [24]. The numerical calculations [25] have shown that Λ is by no means a self-averaging variable; the distribution $p(\Lambda)$ is very broad on its large-Λ side, though it is sharp on the low-Λ side: $p(\Lambda) \approx 0$ for $\Lambda < \Lambda_{\min}$ (i.e., there is a hard gap in Λ distribution). Hence in the very low-T limit, when $\Delta_{\rm Mott} < E_{H(\min)} \equiv \Lambda_{\min} E_{H(0)}$, the hard gap E_H in (2) is just $E_{H(\min)}$. One can expect the following scaling laws near the IMT:

$$\kappa \sim (1 - n/n_c)^{-\nu_1}, \quad \xi \sim (1 - n/n_c)^{-\nu},$$
$$E_H \propto \Lambda_{\min} \sim (1 - n/n_c)^{\gamma_0}, \quad T_0 \propto 1/\kappa\xi \sim (1 - n/n_c)^{\nu + \nu_1},$$
$$T_{c'} = E_H^2/T_0 \sim (1 - n/n_c)^{2\gamma_0 - \nu - \nu_1},$$

where $T_{c'}$ is the temperature of crossover from (1) to (2). According to the numerical calculations [25], the critical exponent for $\Lambda_{\min}$ is $\gamma_0 \approx 1.9$. The possibility to extract γ_0, ν, and ν_1 from the data [1] is very attractive, but unfortunately the experimental points are not sufficiently close to the IMT ($n < 0.62 n_c$).

It can also be shown that in a very close vicinity of IMT there is an interesting regime [intermediate between (1) and (2)] dominated by the fluctuations of Λ_i. It can be described by the percolation in 5D space, which leads to the VRH law (1) with T_0, slowly increasing upon lowering T. The details will be given elsewhere.

In general, for any system with wide distribution $p(\Delta)$ of polaronic shifts (and we believe that this is the case for the experiment [2]), one can expect the low-T simple activation if $p(\Delta)$ has a "hard gap" at low T (which seems to be likely in the vicinity of the IMT [25]).

In conclusion, we have argued that the hopping of spin polarons in magnetically disordered media (e.g., in a semimagnetic semiconductor) proceeds via activation for all temperatures. It provides an explanation for the phenomenon of the magnetic hard gap, observed in a number of substances, and resolves a puzzle, i.e., why the magnetic hard gap is manifested in the T dependence of conductivity, while the other hard gaps (due to other kinds of polarons) are not. We have found the modification of the percolation method, accounting for the spin polarons, and obtained the low-T activation energy for both the strongly localized regime and the close vicinity of the insulator-metal transition. Our approach, being close to the initial idea of the magnetic hard gap [1,9], differs from it in one respect: We do not assume a magnetic medium to be in equilibrium at the moment of the hop, but rather we choose an optimal fluctuation of this medium, which provides a maximal probability to the hop. We have discussed the concrete realization of the scenario only for CdMnTe, though we believe that it can be applied to some other systems with a large number of spins within the localization radius (e.g., the silicon close to IMT).

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