Magnetic and transport percolation in diluted magnetic semiconductors

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The ferromagnetic transition in a diluted magnetic semiconductor with localized charge carriers is inevitably a percolation transition. In this work we theoretically study the correlation between this magnetic percolation and transport properties of the sample, including the possibility of a simultaneous transport percolation. We find nontrivial signatures of the percolating magnetic clusters in the transport properties of the system, including interesting non-monotonic temperature dependence of the system resistivity.

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I. INTRODUCTION

“Diluted magnetic semiconductors” (DMS) is a common name for a wide class of semiconductor materials doped with magnetic impurities (with their relative concentration up to a few percent) which become ferromagnetic at low temperatures. Magnetic semiconductors are now actively investigated both theoretically and experimentally, due to their potential applications in new generations of semiconductor devices.

An example of a DMS material is given by Ga$_{1-x}$Mn$_x$As (GaAs doped with Mn). For the ferromagnetic transition to occur, the concentration of Mn impurities should be relatively high, $x \sim 0.01 - 0.05$. Such high concentration of impurities makes growth of magnetic Ga$_{1-x}$Mn$_x$As a difficult problem, which was solved only relatively recently. In order to prevent phase separation, magnetic Ga$_{1-x}$Mn$_x$As should be grown at low temperatures ($T \sim 200 - 300^\circ\text{C}$), which results in abundance of different types of crystal defects. These problems are not specific to Ga$_{1-x}$Mn$_x$As, in fact, all presently known magnetic semiconductors suffer from the same problems. As a result, theoretical study of magnetic semiconductors is very difficult: two factors – strong disorder and exchange interaction (which causes ferromagnetic transition) – must be taken into account non-perturbatively.

The mechanism of ferromagnetic transition is common for most (if not for all) magnetic semiconductor systems. Since the concentration of magnetic impurities is only of the order of few percent, their direct exchange interaction does not play the decisive role in determining the ground state of the system. The ferromagnetic transition occurs due to interaction between impurity spins mediated by charge carriers, which are present in appreciable quantities because of the high concentration of defects in the crystal lattice. This picture seems to be accepted more or less universally, but the detailed nature of the transition varies greatly from a system to a system. Because of the large number of different magnetic semiconductor materials, high defect concentration in them, and possible variations of growth processes, it is highly unlikely that a “universal theory of magnetic semiconductors” will ever be developed. A theory limited to a certain subclass of magnetic semiconductor materials, which share same basic properties, is more likely to provide useful information on the nature of the processes underlying the ferromagnetic transition and give some insight into experimental results.

In this paper, we study ferromagnetic semiconductors that in the limit of zero temperature become insulating. The mechanism of ferromagnetic transition for an extreme case of such a system, when the charge carriers are pinned to some point defects, was considered in our earlier paper. Here we include transport properties in our consideration, and also extend our scope to the systems where the charge carriers are localized within extended regions rather than pinned with the wave function decaying exponentially from the localization center.

A characteristic feature of DMS physics is the presence of a metal-insulator transition as a function of the magnetic impurity concentration $x$. Typically for low $x$ ($\lesssim 0.03$) the system (e.g. Ga$_{1-x}$Mn$_x$As) is an insulator with strongly localized carriers and with the resistivity increasing exponentially as $T \to 0$. For larger $x$ the system tends to be “weakly” metallic in the sense that the resistivity, although very large ($\sim 0.01 \, \Omega \cdot \text{cm}$), does not exhibit strong temperature dependence. There are reports of a reentrant insulating phase at higher Mn concentration, $x > 0.05$ (More recent experiments on carefully annealed samples do not find this reentrant insulating behavior at higher Mn concentration.) In all situations both metallic and insulating phases are found to be ferromagnetic. Other DMS systems show typically the same behavior (i.e. the existence of a metal-insulator transition with a weakly metallic state, which has a very high resistivity, for higher values of $x$) or often just an insulating ferromagnetic state without any metallic behavior whatsoever. Systems showing a metal-insulator transition tend to have higher ferromagnetic transition temperature $T_c$ in their metallic phases, and therefore understanding the insulating ferromagnetic DMS phase takes on significance, not just from the perspective of fundamental physics involving interplay of disorder and magnetism, but also from the practical technological motivation of enhancing $T_c$ in DMS materials. This is particularly true since the current zeroth-order theoretical
understanding of the optimally doped metallic DMS ferromagnetism is based on a simple continuum virtual crystal approximation Weiss mean-field theory treatment of an RKKY-Zener model that is intrinsically invalid in the insulating regime. In fact, it is currently unclear whether the observed DMS metal-insulator transition is a disorder-driven impurity band or valence band transition. It is, however, clear that disorder plays a critical role in DMS physics, both in its transport properties and in the magnetic properties. We focus in this work on understanding DMS transport properties in the localized insulating regime by applying our recently developed theory\(^9\) which has been successful in explaining the magnetic properties on the insulating side of the metal-insulator transition. We hope to elucidate the role of disorder and magnetic clustering on DMS transport properties through this theoretical study. Although the DMS systems with the highest \(T_c\) do not belong to this class, their resistivity is still very large, so disorder must play a very important role in the properties of these systems. Study of DMS systems that are insulating at \(T \to 0\), as done in this paper, can shed some light on this role, which is difficult to understand using the approach based on the picture of free charge carriers.

So the systems we consider in this paper are insulating at \(T = 0\), which means that the Fermi level is below the mobility edge in the impurity band or below the bottom of the conduction band (for electrons; for holes it would be the valence band) if there are no extended states in the impurity band. From now on, we will refer to this class of magnetic semiconductor systems as “systems with localized charge carriers.” Taking into account the high defect concentration in a typical magnetic semiconductor material, we must conclude that the charge carrier density in systems with localized charge carriers is highly inhomogeneous. Since it is charge carriers that transmit exchange interaction between magnetic impurities, this effective interaction must also be highly inhomogeneous. Thus, when the temperature is lowered, the ferromagnetic transition will first occur locally, in the regions with higher charge-carrier density, i.e. with stronger effective exchange interaction between magnetic impurities. As the temperature goes further down, these finite-size regions, which have random sizes and positions, will grow and merge, until finally the ferromagnetic correlation is established across the whole sample.\(^9\) Such a scenario, which must hold for any magnetic semiconductor with localized carriers, implies that ferromagnetic transition is inevitably a percolation transition, with clusters of ferromagnetic regions already existing at \(T > T_c\) and an infinite magnetic cluster opening up at \(T = T_c\). This percolation picture of ferromagnetism in insulating DMS systems is now well-accepted and has been verified\(^{2,12}\) via direct numerical simulations.

The foundation for the study of ferromagnetic percolation transition was laid down in the 70s for diluted ferromagnetic alloys, where magnetic atoms interact directly with each other\(^2\). Early numerical simulations of localized DMS systems\(^{10}\) already indicated indirect evidence for magnetic percolation playing a role in the magnetic transition. Our earlier paper\(^2\) extended results of Ref.\(^9\) to magnetic semiconductor systems, where the exchange interaction between magnetic atoms is not direct, but is rather mediated by charge carriers.

The word “percolation” by itself assumes some transport. Even though the mathematical percolation theory with its physical applications goes beyond this literal meaning (an example is given by diluted ferromagnetic alloys of Ref.\(^9\) where no transport is considered), one may still naturally wonder if a percolation transition in a given system means some enhancement of charge transport. In ferromagnetic semiconductors, this question is quite relevant, since charge carriers, which have some ability to move around the sample, are present, and since the ferromagnetic percolation transition is facilitated by these very carriers. In fact, some experiments on magnetic semiconductors did observe connections between ferromagnetic transition and enhancement of charge transport, up to the point of the temperature dependence of resistivity being non-monotonic with the maximum around the ferromagnetic transition temperature\(^3,4,11\). In addition, the existence of the metal-insulator transition in DMS systems makes transport percolation considerations relevant since there is an intrinsic connection between metal-insulator transition and percolation. We emphasize in this context that the magnetic percolation transition, considered in Ref.\(^2\), is purely a statement on the percolation properties of the temperature-dependent magnetic clusters in DMS materials, and has little to do (in a direct sense) with transport percolation properties (which must take into account the site-to-site hopping of the localized charge carriers, as we do in this paper) – it is, in principle, entirely possible for a system to have a magnetic percolation transition with no transport percolation altogether. In fact, these are two separate physical phenomena whose relationship is theoretically explored in this paper in the specific context of DMS systems.

The main purpose of this work is to establish connection between ferromagnetic transition in a magnetic semiconductor system with localized carriers (which, as we mentioned above, must have percolation nature) and transport properties of such a system. The outline of the paper is as follows: In Sec.\(^\text{II}\) we introduce our model. In Sec.\(^\text{III}\) we consider the case of charge carriers strongly pinned to localization centers. The ferromagnetic properties in such a system were studied in our earlier papers\(^2,12\); here we discuss its transport properties. The considerations of this section are based on the impurity positions being uncorrelated. The case of impurities forming clusters in considered in Sec.\(^\text{IV}\). Finally, in Sec.\(^\text{V}\) we qualitatively discuss the transport properties of a system, whose charge carriers are on the verge of being delocalized. For all these systems, we also discuss dependence of the resistivity on the applied magnetic field.
II. MODEL HAMILTONIAN

Charge carriers in Ga$_{1-x}$Mn$_x$As are holes, donated by Mn impurities. While other magnetic semiconductors may have electrons as the carriers mediating exchange interaction between magnetic impurities, in this paper we will refer to the charge carries as "holes," just for the sake of brevity. All our conclusions would hold no matter what the charge carriers are.

In fact, the whole model system we use is based on Ga$_{1-x}$Mn$_x$As, where Mn atoms act both as magnetic impurities and acceptors, thus providing both local moments, which order ferromagnetically at $T < T_c$, and charge carriers, which mediate ferromagnetic interaction between these local moments. We do not expect our results to be specific to this particular model as far as the qualitative results are concerned, so our conclusions should be valid for other systems carrier-mediated insulating DMS as well.

In our model, a Mn impurity in Ga$_{1-x}$Mn$_x$As is presented by two spin-degenerate levels, which we will refer to as "the deep level" and "the shallow level." The deep level, when occupied by one electron, provides the impurity’s spin. Coulomb repulsion prevents a second electron to as "the deep level" and "the shallow level." The deep level of one impurity to the shallow level of an-}

The full Hamiltonian of the system reads:

$$H_0 = \sum_m \left[ \sum_\alpha \left( -\varepsilon_d \right) a_\alpha^\dagger a_\alpha + U_d \sum_m a_\alpha^\dagger a_\alpha a_\alpha^\dagger a_\alpha \right]$$

$$+ \sum_\alpha \left( -\varepsilon_0 \right) c_\alpha^\dagger c_\alpha + U_0 c_\alpha^\dagger c_\alpha c_\alpha^\dagger c_\alpha$$

$$+ \sum_{\alpha\beta} U_{0d} c_\alpha^\dagger c_\alpha a_\beta^\dagger a_\beta$$

$$+ \sum_{mna} t_{mn}^c c_\alpha^\dagger c_\alpha a_\alpha$$

$$+ \sum_{mna} \left( t_{m(0)\alpha}^d c_\alpha^\dagger a_\alpha + \text{h.c.} \right),$$

(1)

where indices $m$ and $n$ run over magnetic impurities, $a_\alpha^\dagger$ and $c_\alpha^\dagger$ are the creation operators for an electron with spin $\alpha$ localized at the $m$th impurity at the deep/shallow level respectively. The first and the third terms in the brackets represent the Coulomb repulsion between two holes on each of these two levels, while the fifth term is for the inter-level Coulomb repulsion. The first term after the brackets accounts for hole hopping between the shallow levels of two impurities.

The last term of Eq. (1) describes hopping from the deep level of one impurity to the shallow level of another. Hopping between deep levels is neglected because of the rapid fall-off of the electron wave functions at these levels. We take the Fermi energy equal to zero, $\varepsilon_F \equiv 0$ throughout the paper.

The parameters of Hamiltonian (1) must be chosen to mimic magnetic impurities in Ga$_{1-x}$Mn$_x$As. The deep level in the ground state must be taken by only one hole, so we take $U_d > \varepsilon_d > 0$. The shallow, acceptor levels of the impurities are to donate, on average, less than one hole per impurity, so the Fermi level coincides with the energy of a hole placed onto the shallow level of an impurity, whose lower level is already taken by one hole, which means that $U_{0d} - \varepsilon_0 = \varepsilon_F \equiv 0$. The bottom of the conduction band is assumed to be separated from the impurity level $U_{0d} - \varepsilon_0$ by the energy $E_0 \gg T$, $t_{mn}$, so the (delocalized) valence band states can be safely excluded from our consideration.

We assume the wave functions of these localized levels to fall off exponentially, with the characteristic decay radii equal to $a_d$ and $a_0$ for deep and shallow levels respectively, with $a_d \ll a_0$. Then the hopping matrix elements $t_{mn}^{(0d)}$ and $t_{mn}^{(d)}$, which are proportional to the overlap of the corresponding wave functions, are given by

$$t_{mn} = t_0 \left( 1 + \frac{r_{mn}}{a_0} \right) \exp \left( -\frac{r_{mn}}{a_0} \right),$$

(2a)

$$t_{mn}^{(0d)} = t_0 \left( 1 + \frac{r_{mn}}{a_0} \right) \exp \left( -\frac{r_{mn}}{a_0} \right).$$

(2b)

With these parameters, we may make the Schrieffer-Wolff transformation for the deep levels, thus reducing Hamiltonian (1) to

$$\hat{H} = \sum_m \left[ \sum_\alpha \left( -\varepsilon_0 \right) c_\alpha^\dagger c_\alpha + U_0 c_\alpha^\dagger c_\alpha c_\alpha^\dagger c_\alpha + \sum_{mna} t_{mn}^c c_\alpha^\dagger c_\alpha a_\alpha + \sum_{mn\alpha\beta} J_{mn}^{\alpha\beta} (\mathbf{S}_l \cdot \sigma_{\alpha\beta}) c_\alpha^\dagger c_\alpha c_\alpha^\dagger c_\alpha^\dagger \right],$$

(3)

where $\sigma \equiv (\sigma_x, \sigma_y, \sigma_z)$ is the vector of Pauli matrices,

$$J_{mn}^{\alpha\beta} = \frac{1}{U_d} \left( \frac{1}{\varepsilon_d} + \frac{1}{U_d - \varepsilon_d} \right)$$

$$\left( t_0^{(0d)} \right)^2 \left( \frac{1}{\varepsilon_d} + \frac{1}{U_d - \varepsilon_d} \right) \exp \left( -\frac{r_{ml} + r_{ln}}{a_0} \right),$$

(4)

and

$$\mathbf{S}_l = \sum_{\alpha\beta} \sigma_{\alpha\beta} a_\alpha^\dagger a_\beta.$$

(5)

The other parameters of the system are the concentration of impurities $n_i$ and the concentration of holes $n_h$. The
spatial distribution of impurities is assumed to be random throughout the paper, although not necessarily uncorrelated. We also assume, as it is the case in Ga$_{1-x}$Mn$_x$As and many other DMS, that the system is very heavily compensated, $n_h \ll n_i$, with many more magnetic impurities that charge carriers present in the system.

Depending on the values of the system parameters, the system described by Hamiltonian (3) may be either insulating or metallic in the limit $T \to 0$. In this paper we will concentrate on the former case. Ferromagnetic transition in such a system inevitably has percolation nature, as we pointed out in the introduction.

### III. STRONGLY LOCALIZED CHARGE CARRIERS

In this section, we consider the case of $a_0^3 n_i \ll 1$, with the impurity positions being uncorrelated. Randomness in the impurity positions leads to randomness in the hopping matrix elements $t_{mn}$, which exponentially depend on the distances $r_{mn}$ between the corresponding impurities. This strong (by orders of magnitude, provided $a_0^3 n_i \ll 1$) variation of $t_{mn}$ creates strong variation of localization potentials for the holes, which thus will be tied to the pairs of least-separated impurities. We must note that the concrete mechanism of hole localization does not have crucial impact on the derivations and conclusions presented in this section. In order to keep our presentation straightforward we limit ourselves to the framework of Hamiltonian (3), when the holes are localized at these closest impurity pairs. Any additional disorder can be taken into account by inclusion of new terms to Eq. (3) and proper modification of the hole localization parameters.

With strongly localized holes, for study of magnetic properties of the system Hamiltonian (3) can be reduced to:

$$\hat{H} = \sum_{mj} J_{mj} \hat{S}_m \hat{S}_j, \quad (6)$$

where indices $m$ and $j$ label magnetic impurities and holes respectively, and $\hat{S}_m, \hat{S}_j$ are the impurity/hole spin operators. The expression for the matrix elements $J_{mj}$ of the impurity-hole exchange interaction depend on the hole localization mechanism. In the case of localization by the pairs of least-separated impurities, it is given by

$$J_{mj} = \frac{1}{2} \left( J_{mn_1}^{(1)} + J_{mn_2}^{(2)} \right) \quad (7)$$

for a hole localized at the impurities labelled by indices $n_1^{(1)}$ and $n_2^{(2)}$.

Since the concentration of holes is much smaller than that of impurities, $n_h \ll n_i$, one localized hole is surrounded by many magnetic impurities. Exchange interaction between the (localized) holes and magnetic impurities leads to their mutual polarization when temperature $T$ is below exchange constant $J_{mj}$. Since exchange coupling $J_{mj}$ decays with the impurity-hole distance $r_{mj}$, see Eq. (2) and (4), the first impurities to get their spins aligned with a hole’s spin as the temperature decreases are the ones most close to the hole’s localization site. At lower temperatures, more distant impurities have their spins polarized by the hole to whose domain they belong. This complex consisting of a hole and magnetic impurities polarized by it is conventionally called “bound magnetic polaron.” The characteristic radius size of a polaron grows as the temperature decreases

$$r_{pol}(T) = \frac{a_0}{2} \ln \frac{A J_0 \sqrt{a_0 n_i}}{T}, \quad (8)$$

where $A \sim 1$ and

$$J_0 \equiv \left( t_0^{(0d)} \right)^2 \left( \frac{1}{\varepsilon_d} + \frac{1}{U_d - \varepsilon_d} \right),$$

cf. Eq. (4). As polarons overlap, they form polaron clusters with all impurities belonging to a given cluster having their spins aligned in the same direction. The temperature

$$T_c \sim s S |J_0| (a_0^3 n_i)^{1/2} \sqrt{n_i/n_h} \exp \left( -\frac{0.86 \pi a_0^3 n_i}{n_h} \right), \quad (9)$$

at which the infinite cluster spanning the whole sample appears is the ferromagnetic transition temperature. The quantitative theory of this transition was recently developed and presented by us in Ref. 2 and further developed in Ref. 12. In this paper we concentrate on transport properties of such a system and their correlation with the magnetic properties.

At temperatures of the order of the ferromagnetic transition temperature, the hole transport in the system occurs by means of nearest-neighbor hopping. The resistivity $\rho$ of such a system depends on temperature exponentially,

$$\rho \propto \exp(E_{hop}/T). \quad (10)$$

The characteristic hopping activation energy $E_{hop}$ has two contributions. First, there is random level mismatch between two localization sites, produced by the disorder. The second contribution to the hopping activation energy comes from the interaction of a localized hole with neighboring magnetic impurities. This interaction leads to polarization of impurity spins by the hole spin and lowers the energy of the resulting bound magnetic polaron. The energy associated with this polarization (polaron’s “binding energy”) is given by

$$E_{pol} = 8\pi J_0 a_0^3 n_i, \quad (11)$$

provided $r_{pol} > a_0$, which is satisfied at temperatures of the order and below the ferromagnetic transition temperature $T_c$. When a hole hops to another localization site, it abandons the region it has polarized and lands among non-polarized impurities unless this new localization site
is within another bound magnetic polaron. The “binding energy” $E_{\text{pol}}$ of a polaron is much less than the characteristic energy level mismatch $E_{\text{hop}}^{(0)} \sim t_0$ at different localization sites. Therefore this “binding energy” will not have a noticeable effect on the hopping trajectory of a hole. At temperatures close to the ferromagnetic transition temperature $T_c$, while the bound magnetic polarons barely touch each other, it means that most of the hops will be to unpolarized regions, see Fig. 1. Therefore, the characteristic hopping activation energy will acquire an addition term $E_{\text{pol}}$,\vspace{-0.1cm}

$$E_{\text{hop}} = E_{\text{hop}}^{(0)} + E_{\text{pol}} \text{ at } T \lesssim T_c.$$ \hspace{1cm} (12)

At low enough temperatures, however, the infinite cluster of bound magnetic polarons will cover the whole sample, so wherever a hole hops, it will land in a region which is already polarized in a optimal way, so the impurity polarization will not have any contribution to the hopping activation energy. Our Monte-Carlo studies show that the characteristic temperature $T_{\text{cover}}$ at which the infinite cluster covers most of the sample corresponds to $\tau_{\text{pol}}(T_{\text{cover}}) \sim 2\tau_{\text{pol}}(T_c)$, which yields $T_{\text{cover}} \sim T_c \exp\left(-\frac{0.86}{(a_0^2 t_{\text{th}})^{\frac{3}{2}}}\right) \ll T_c$. Thus

$$E_{\text{hop}} = E_{\text{hop}}^{(0)} \text{ at } T \lesssim T_{\text{cover}} \ll T_c.$$ \hspace{1cm} (13)

The concrete scenario of the infinite cluster spreading over the transport paths highly depends on the sample, because even though these two random networks are not completely independent, they generally do not coincide. Since the size of a bound magnetic polaron grows logarithmically slowly as the temperature goes down, sample-dependent variations in the mutual arrangement of the transport paths and the infinite cluster lead to strong variations in the temperatures at which the transition from the high-temperature [Eq. (12)] to the low-temperature [Eq. (13)] exponent occurs and in the precise functional form of $\rho(T)$ during this transition. However, since the transition is slow, the overall temperature dependence $\rho(T)$ of the resistivity must be monotonic, because even though magnetization reduces the hopping barrier, the decrease in temperature make it harder for electrons to get activated to overcome it.

External magnetic field $B$ applied to the sample polarizes the impurities all over the sample, with the average value of the impurity polarization given by

$$\langle S_z \rangle = SB_S\left(\frac{g_{\mu_B}B}{k_B T}\right)$$ \hspace{1cm} (14)

where $\mu_B$ is the Bohr magneton,\vspace{-0.1cm}

$$B_S(x) = \frac{2s + 1}{2s} \coth \frac{2s + 1}{2s} x - \frac{1}{2s} \coth \frac{1}{2s} x$$ \hspace{1cm} (15)

is the Brillouin function, and the direction of the $z$ axis is chosen to be parallel to the magnetic field. As the result of this polarization, the impurities around the localization site a hole hops into are not entirely uncorrelated with those around the site it hops from. The “polaron binding energy” (11) is therefore reduced by

$$\Delta E_{\text{pol}}(B) = -8\pi J_0 a_0^3 n_B B_S \left(\frac{g_{\mu_B}B}{k_BT}\right),$$ \hspace{1cm} (16)

and the resulting magnetic-field dependence of the resis-
tivity is given by
\[\rho(B) = \rho|_{B=0} \exp \left[ -\frac{8\pi J_0 a_B^3}{k_B T} \mathcal{B}_2 \left( \frac{g_k \mu_B B}{k_B T} \right) \right], \tag{17}\]
as it follows from Eqs. (10), (12), and (16). One can see that the resistivity decreases as the applied magnetic field grows, in agreement with the experimental results.

IV. CHARGE CARRIERS LOCALIZED WITHIN CLUSTERS

The previous section addressed transport in magnetic semiconductors with strongly localized charge carriers. “Strongly localized” in that case meant that a charge carrier is pinned to some point (a pair of close impurities or some other defect in crystal structure), with the carrier localization radius being smaller than the characteristic distance between pinning centers. Coulomb repulsion prevents a carrier from entering a localization site already taken by another carrier. This regime takes place when concentrations \(n_i\) of magnetic impurities and \(n_h\) of charge carriers are small. For such small \(n_i\) and \(n_h\), ferromagnetic transition temperature \(T_c\) is typically of the order of few Kelvins for realistic parameters. For practical applications, however, it is desirable to have \(T_c\) higher than a few Kelvins, which implies higher \(n_i\) and \(n_h\), so one may naturally wonder what happens to the charge transport when the magnetic impurity concentration is so high that the assumptions of Sec. III are no longer applicable.

If the positions of impurities are uncorrelated, charge carriers become delocalized when parameter \(n_i a_B^3\) exceeds some critical value of the order of unity. The analytical description of this transition is very challenging, and hardly any quantitative results can be obtained. We postpone the discussion of this case until the next section. In this section we consider the case when magnetic impurities are grouped into clusters. This arrangement is not improbable at all, taking into account significant differences between the non-magnetic atoms of the host lattice and the magnetic dopants. In fact there are theoretical indications\(^\text{15}\) that the impurities should group into clusters, due to electrostatic interactions during the growth process.

For the parameter region in the vicinity of \(n_i a_B^3 \sim 1\) clustering of impurities means that a hole will be able to move freely within a cluster, but not between clusters, i.e. the characteristic size of a hole wave function will be of the order of a cluster size rather than of the size of a wave function of a hole localized at an isolated impurity or an impurity pair. Because of the hole wave function being spread over many impurities, the Coulomb energy which repels a hole from a site occupied by another hole is reduced as compared to the case of strongly localized holes considered in Sec. III. If the cluster size is large enough, several holes may enter one cluster, and the characteristic magnitude of the Coulomb energy in a cluster at the Fermi level is
\[E_{\text{Coul}} \sim U \frac{N_h}{N_i}, \tag{18}\]
where \(N_h\) and \(N_i\) are the number of impurities in the cluster and the number of holes sitting in it, respectively. Since \(n_i \gg n_h\), for a typical cluster we must have \(N_i \gg N_h\), and, therefore \(E_{\text{Coul}} \ll U\).

The activation energy \(E_{\text{hop}}\) for hole hopping between the clusters contains contributions from the Coulomb- and disorder-induced level mismatch between the clusters and from the exchange interaction between the holes hopping between the clusters and the impurities forming these clusters. The first contribution stems mainly from the Coulomb repulsion between the holes in clusters. The characteristic value of the gap between the highest occupied and the lowest unoccupied levels in a cluster is given by Eq. (18). The relative positions of these levels in different clusters are completely random, so a characteristic value of the mismatch between the highest occupied and lowest unoccupied levels in two neighboring clusters will also be of the order of \(E_{\text{Coul}}\) given by Eq. (18). The random mismatch \(E_{\text{sp}}\) between two single-particle levels in two clusters also plays some role, but the characteristic value of this mismatch
\[E_{\text{sp}} \sim \frac{t_0}{N_i^{2/3} N_c^{1/3}} \tag{19}\]
should be much less than \(E_{\text{Coul}}\) for realistic systems.

The second contribution to the hopping activation energy comes, as it was stated above, from the exchange interaction between holes and impurities. At high temperatures, spins of impurities and holes in a cluster are not polarized, and there is no contribution to the hopping activation energy due to exchange. At lower temperatures, however, exchange interaction between magnetic impurities and holes leads to ferromagnetic transition. The mechanics of the latter is as follows: first, exchange interaction between holes and impurities in clusters they occupy lead to ferromagnetic transition within clusters. The exchange interaction between the clusters is exponentially small, \(\propto \exp(-r_{cl}/a_0)\), where \(r_{cl}\) is the characteristic distance between the clusters. Therefore, as the temperature is lowered and ferromagnetic transition occurs within clusters, the magnetic moments of different clusters are still uncorrelated. At even lower temperatures, the correlation between the clusters is established, and the macroscopic ferromagnetic transition occurs. At high temperatures when the even clusters are not polarized a hole hopping from a cluster to another one experiences no change of the exchange energy. However at temperatures close to the ferromagnetic transition temperature the clusters are inevitably polarized, and this creates an additional energy barrier for a hole hopping between two uncorrelated clusters.

While the ferromagnetic transition in a cluster does not imply full polarization of all impurities in it, at the temperatures of the order of the ferromagnetic transition
FIG. 3: Charge transport in a DMS system with clustered magnetic impurities. The picture refers to the temperatures low enough for the ferromagnetic transition to occur within clusters, whose spins are shown with black arrows. Holes hop between the clusters preferring shorter gaps, so the percolation paths coincide with transport paths.

FIG. 4: Sketch of the temperature dependence of resistivity in a DMS system with clustered impurities. Since percolation and transport paths coincide, ferromagnetic transition leads to immediate decrease in the hopping energy, which may result in non-monotonic temperature behavior of the sample resistivity.

At temperatures all the impurities must be polarized, and the energy associated with the exchange interaction of holes in a cluster with all the impurities in it equals

$$E_{\text{exch}} = J_0 .$$

(20)

When two clusters are not correlated, a hole hopping from one cluster to the other one experiences the change of potential of the order of $E_{\text{exch}}$, so the expression for the characteristic hopping activation energy reads

$$E_{\text{hop}} = E_{\text{Coul}} + E_{\text{exch}} \text{ at } T_c \lesssim T .$$

(21)

At temperatures below $T_c$, the correlation between the cluster spins is established, and a hole hopping into another cluster experiences exactly the same exchange potential as in the cluster it has left. This reduction of the exchange contribution to the hopping activation energy from $J_0$ to zero (roughly speaking) occurs while temperature $T$ is still of the order of $T_c$. Therefore, the exponent in Eq. (10) may actually become smaller as the temperature goes down past $T_c$, provided the second term in Eq. (21), which vanishes when the spins of clusters become aligned, is larger than the first, temperature-independent term, that is

$$U \frac{N_h}{N_i} < J_0$$

(22)

The external magnetic field applied to a sample lowers its resistance, similarly to the case discussed in the previous section. However, now the reduction of the resistivity in the systems is due to polarization of impurity clusters as a whole, not of individual impurities, since all impurities in a cluster have their spins aligned in the same direction. Since polarization of a cluster by magnetic field of a given magnitude strongly depends on the size of the cluster, and since the sizes of impurity clusters are random with possibly wide distribution, the quantitative dependence of resistivity on the applied magnetic field is highly sample-dependent.

V. CHARGE CARRIERS ON THE VERGE OF DELOCALIZATION

At sufficiently high concentrations of impurities and holes, some of the holes are delocalized even in the limit $T \to 0$. The ferromagnetic transition in systems of this class does not have percolation nature, unless the fraction of delocalized carriers is very small. With physics of their ferromagnetic transition and charge transport being entirely different from that of the systems with localized charge carriers, these systems are beyond the scope of the current paper. In this section, we consider systems which are close to localization threshold, but still not beyond it. In the ground state of such a system, all charge carriers are localized within some finite regions, and the resistivity of the sample goes to infinity as $T \to 0$. However, because of the small separation between the Fermi level and the mobility edge, finite temperature excites some holes into delocalized states. It is hardly possible to make any quantitative statements about magnetization and resistivity of the system, but still some qualitative statements can be made with the help of intuition we have developed dealing with the case of the previous section.

Since, despite some holes being thermally excited into delocalized states, many holes are still localized, the effective exchange interaction between the magnetic impurities, which is induced by holes, is still highly inhomogeneous across the sample. There will be “puddles” filled with holes, where interimpurity exchange concentration is strong, and relatively hole-free regions, which are covered only by tails of hole wave functions decaying away from these puddles, see Fig. 5. Qualitatively, this picture
is similar to that of Sec. IV, shown in Fig. 3 with the difference that in the case considered in the present section transport paths go mostly along hole-filled regions with fewer gaps on the way. These gaps are bridged by the delocalized states, so there is no need for hopping.

Similarly to the case of previous sections, it is regions with higher hole density that undergo (local) ferromagnetic transition first, with ferromagnetic correlations across the sample established at lower temperatures. When two neighboring “hole puddles” have different spin orientation, a delocalized hole going from one puddle to the other encounters an additional barrier, which can partially reflect it back or even entirely prevent it from entering the puddle. When the spins of two puddles oriented in the same direction, as it is the case at the temperatures below the ferromagnetic transition temperature, no such problem arise, and a delocalized hole move freely from a puddle to a puddle across the sample.

We thus have two competing phenomena that occur as the temperature goes down. On one hand, at lower temperatures we have fewer holes excited to delocalized states. On the other hand ferromagnetic transition occurring as temperature goes down makes it easier for the hole to travel across the sample. The interplay of these two phenomena determines the temperature dependence of resistivity. Due to the extremely complex nature of hole wave functions close to the mobility edge, we are unable to provide any qualitative results. However, with the system under consideration being in some sense an extreme case of the system consider in the previous section, we may deduce that the non-monotonic behavior of resistivity must be possible, and that the non-monotonicity becomes more pronounced as the Fermi level of the holes approaches the mobility edge, i.e. when the concentrations of holes and impurities become higher. Similarly to the previous cases, magnetic field applied to the system would align the spins of hole puddles and enhance the transport.

VI. CONCLUSION

We have considered correlations between ferromagnetic percolation transition and charge transport in magnetic semiconductors with localized charge carriers. We found that ferromagnetic transition alone would enhance charge transport, but the temperature decrease, which is needed for the transition, may completely eliminate these gains. In a system with strongly localized charge carriers (Sec. III), decrease in the hopping rate of the charge carriers due to temperature decrease overcomes decrease in the hopping activation energy due to ferromagnetic transition, and the temperature dependence of the resistivity is always monotonic. At higher concentrations of magnetic impurities and charge carriers, the situation may be the opposite, and resistivity may experience a dip as the temperature goes down past the ferromagnetic transition temperature (Secs. IV-V). The higher the concentrations of the impurities and carriers are, the more pronounced the dip is. In all cases considered in the paper, the resistivity of the system decreases if an external magnetic field is applied.

It is important to point out that our theoretical DMS transport results are in qualitative agreement with the available experimental transport results in the localized insulating regime, where the resistivity increases strongly (exponentially) with temperature as $T \to 0$. In particular, deep into the insulating regime, far away from the metal-insulator transition point, the experimentally measured resistivity rises monotonically with decreasing temperature similar to our results (Fig. 2) presented in Sec. III for strongly localized charge carrier system. On the other hand, close to the metal-insulator transition (but still on the insulating side) transport experiments indeed observed a striking temperature-dependent non-monotonicity in the measured resistivity as we find in our theory for “not-so-strongly” localized charge carriers (Sec. IV Fig. 1). Unfortunately, the insulating DMS ferromagnetic regime has not yet been extensively studied experimentally (compared with the metallic DMS regime with optimal $T_c$ values) although our view is that this regime is as important and as interesting as the metallic regime in terms of the development of our understanding of DMS physics. We urge that more experimental work done in the insulating ferromagnetic regime (and on the metal-insulator transition itself) for us to develop a better and more quantitative understanding of transport and magnetization phenomena in DMS systems.

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