

## Polaron Percolation in Diluted Magnetic Semiconductors

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We theoretically study the development of spontaneous magnetization in diluted magnetic semiconductors as arising from a percolation of bound magnetic polarons. Within the framework of a generalized percolation theory we derive analytic expressions for the Curie temperature and the magnetization in the limit of low carrier density, obtaining excellent quantitative agreement with Monte Carlo simulation results and good qualitative agreement with experimental results.

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Diluted magnetic semiconductors (e.g.,  $\text{In}_{1-x}\text{Mn}_x\text{As}$ ,  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ ,  $\text{Ge}_{1-x}\text{Mn}_x$ ), which are materials with some fraction of the nonmagnetic lattice atoms replaced by magnetic atoms (i.e., a fraction  $x$  of Ga, In, or Ge atoms being randomly replaced by Mn atoms), have attracted a great deal of attention [1–10] following the recent discovery [11] of ferromagnetism in  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ , with a Curie temperature  $T_c \approx 100$  K in the  $x = 0.03$ – $0.07$  range. The subject is of considerable fundamental and technological interest. Technologically, a semiconductor, which is also a ferromagnet, raises the exciting potential of spintronic applications [12], where logic and memory operations could in principle be seamlessly integrated on a single device. From a fundamental perspective, understanding ferromagnetism in a novel material (which is also a semiconductor) is an important challenge. It is therefore not surprising that there has been a great deal of theoretical activity [1–10] trying to understand the ferromagnetic mechanism in GaMnAs. Although no theoretical consensus on the precise ferromagnetic mechanism has yet been reached in the literature it is now established that the interaction between the magnetic atoms, which leads to the ferromagnetic phase at low enough temperatures, is induced by charge carriers (holes in the case of GaMnAs) in the semiconductor host. Theoretical investigation of these systems is hampered by the fact that both disorder and interactions are strong and must be taken into account nonperturbatively. This problem has been approached in a number of ways, including theoretical approximations assuming charge carriers being almost free [1–4] and numerical studies in the opposite limit of strongly localized charge carriers [5–7]. However, a comprehensive understanding of the physics of diluted magnetic semiconductors has not been achieved yet.

Theoretical approaches [1–4] treating the charge carriers as free carriers in the valence band of the semiconductor have employed the simple Weiss mean field theory incorporating the band structure details (e.g., spin-orbit coupling, etc.), without taking into account effects of disorder. While this approach is claimed to provide quantitatively accurate values of  $T_c$ , it fails qualitatively in accounting for experimentally observed transport

properties [13] of GaMnAs where the resistivity is very high and seems to obey a Mott variable range hopping behavior at low temperatures reminiscent of an insulating system. In addition, the resistivity always decreases with increasing temperature above  $T_c$ , which is again typical of a localized insulating system. A recent dynamical mean field theory calculation [10] concluded that the ferromagnetic GaMnAs may actually belong to a situation where the carriers are just on the edge of being strongly localized. The approach [5–7] treating the carriers as localized carriers in a semiconductor impurity band (thus being opposite to the free-carrier mean-field approach), which has so far been explored only numerically, also leads to reasonable agreement with experiments and indicates a very strong dependence of  $T_c$  on disorder. In this paper we provide an analytical theory which takes into account both disorder and strong magnetic interaction starting with localized carriers and using the physically appealing magnetic polaron percolation picture. Our theory has a starting point similar to that in Refs. [5–7] except that ours is a completely analytical physical theory in contrast to the numerical approach used in the works [5–7]. Where applicable our analytical results agree well with the numerical results of Refs. [5–7].

In this paper we consider a system in which transition to the insulating state due to localization of the charge carriers occurs at temperatures higher than the Curie temperature  $T_c$ . The carriers will be called “holes” throughout the text of this paper since in GaMnAs the carriers are holes although the theory to be developed in this paper applies equally to the situation where the carriers are electrons. Exchange interaction of localized holes with magnetic impurities leads to the formation of bound magnetic polarons [14,15]. Since the concentration of magnetic impurities is much larger than the hole concentration [16], most likely due to compensation by As antisite defects, a bound magnetic polaron consists of one localized hole, and a large number of magnetic impurities around the hole localization center. Even though the direct exchange interaction of the localized holes is antiferromagnetic, the interaction between bound magnetic polarons may be ferromagnetic [15] at large enough concentrations of magnetic impurities.

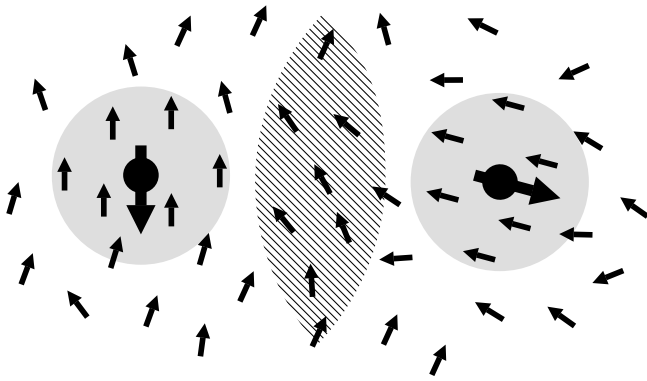


FIG. 1. Interaction of two bound magnetic polarons (after Ref. [15]). The polarons are shown with gray circles; small and large arrows show impurity and hole spins, respectively.

To understand the physics of this phenomenon, one may consider two neighboring polarons (Fig. 1). The localized holes of these polarons both act on the impurities surrounding them thus producing an effective magnetic field for these impurities. The energy minimum is reached by this system when the impurity spins are parallel to this effective field, and the magnitude of the field is maximum. The maximum of this effective magnetic field is achieved when the spins of the localized holes are parallel. Therefore at low temperatures the system should eventually reach the state where the spins of all holes point in the same direction, and all impurity spins point in the same or in the opposite direction, depending on the sign of the impurity-hole exchange interaction.

If the hole localization radius is much less than the characteristic distance between the localized holes, the disorder in the hole positions must have dramatic effect on the whole picture of the ferromagnetic transition. This notion has been confirmed by Berciu and Bhatt [6], who have shown, by means of numerical simulations, that both the Curie temperature and the shape of the magnetization curve  $M(T)$  are indeed strongly affected by disorder. It has been known that the percolation theory [17] provides many adequate tools to deal with ferromagnetism in disordered systems with strong localization of carriers [18]. In this paper we present a quantitative description of the spontaneous magnetization in magnetic semiconductors within the framework of the percolation theory.

In our model, the charge carriers are localized. The hole wave function is assumed to fall off exponentially away from localization centers, with decay length  $a_B$ . We consider the low carrier density regime in which the mean distance between the localized holes is much larger than the hole localization radius,  $a_B^3 n_h \ll 1$ . We note that, for GaMnAs,  $a_B \approx 10 \text{ \AA}$ , and therefore our theory applies in the regime  $n_h \ll 10^{21} \text{ cm}^{-3}$ , with experimental  $n_h$  values currently being around  $10^{19} \text{ cm}^{-3}$ . The localization centers are distributed randomly in the sample. Magnetic impurities are distributed within the sample with concen-

tration  $n_i \gg n_h$  randomly as well. The Hamiltonian of the system has the form

$$\hat{H} = \sum_{kj} J_{kj} \hat{S}_k \hat{s}_j, \quad (1)$$

where indices  $k$  and  $j$  label magnetic impurities and holes respectively,  $\hat{S}_k/\hat{s}_j$  are the impurity/hole spin operators. Matrix elements  $J_{kj}$  of the impurity-hole exchange interaction decay exponentially with the distance between the interacting impurity and hole,  $J_{kj} = J_0 \exp(-2|\mathbf{r}_k - \mathbf{r}_j|/a_B)$ . The direct (antiferromagnetic) exchange interaction between the magnetic impurities is neglected, since their relative concentration  $x$  in the lattice of the host semiconductor is much less than unity although this may be important for larger values of  $x$ , leading to the suppression of  $T_c$ . We will not discuss possible mechanisms of hole localization, since the properties of our model hold for any of them as long as the decay of the localized hole's wave function is exponential.

At some temperature  $T$ , magnetic impurities that are at distances  $r < R_p(T) \equiv (a_B/2) \ln(sS|J_0|/T)$  from hole localization centers have their spins strongly correlated with the spins of the corresponding holes (here  $s$  and  $S$  are the absolute values of the hole or impurity spin, respectively). The spins which do not have a localized hole within a circle of radius  $R_p(T)$  around them are essentially free. The quantity  $R_p(T)$  is the effective radius of a magnetic polaron; it grows as the temperature is lowered. Clearly at low enough temperatures neighboring magnetic polarons overlap and interact with each other via interaction with impurities between them [15]. This interaction produces alignment of the polaron spins. When the cluster of correlated polarons having the size of the sample (the infinite cluster) appears, the ferromagnetic transition occurs.

Even though the picture presented above is qualitatively correct, one needs to find the maximum characteristic temperature  $T_{2p}(r)$  at which the spins of two polarons at a given distance  $r$  from each other are still strongly correlated. The Hamiltonian of a two-polaron subsystem is given by Eq. (1), where hole index  $j$  takes only two values  $j_1$  and  $j_2$  corresponding to the two polarons under consideration. Our present goal is to find the characteristic spin-correlation temperature  $T_{2p}(r)$  at given  $r$ ,  $a_B$ ,  $J_0$ , and  $n_i$ . Since we are interested in the system behavior only at and below the percolation transition, we may limit our consideration to polaron pairs with  $r \gtrsim n_h^{-1/3} \gg a_B$ . A rough preliminary estimate, which can be obtained from straightforward dimensional analysis, reads

$$T_{2p}(r) \sim A(r, a_B, n_i) sS |J_0| \exp(-r/a_B), \quad (2)$$

where dependence of the dimensionless prefactor function  $A(r, a_B, n_i)$  on its arguments is weaker than exponential. Since at  $T \sim sS |J_0| \exp(-r/a_B)$  the two polarons *must* be correlated,  $A \geq 1$ . At temperatures of the order of the right-hand side of Eq. (2), each polaron already has a large number of impurity spins near its center strongly polarized

along the direction of the spin of its hole. Because of this coupling, the spin of the polaron's hole becomes "massive," and may be considered a classical field as far as its interaction with more remote impurities is concerned. This effective field produces polarization of the remote impurities; the characteristic value of the projection  $S_k^{(j_1)}$  of the spin of  $k$ th impurity onto the spin direction of the  $j_1$ th polaron is roughly  $S \min\{1, sJ_{kj_1}/T\}$ . The characteristic energy of interaction of this impurity with the other polaron is of the order of  $sSJ_{kj_2}$ , therefore the contribution of the  $k$ th impurity to the interaction of two polarons is of the order of  $S^2 sJ_{kj_2} \min\{1, sJ_{kj_1}/T\}$  [here we assume that  $sSJ_{kj_1}, T > sSJ_{kj_2}$  since  $T_{2p}(r) \geq sS|J_0| \exp(-r/a_B)$ ]. This estimate allows us to determine which impurities are important for the interaction of two given polarons. Namely, these impurities are the ones in the lens-shaped region between the two localization centers; see Fig. 1. The diameter of this region is of the order of  $\sqrt{ra_B}$ ; more remote impurities are interacting too weakly with both polarons to be of any importance. Finding the thickness of the lens-shaped region is less trivial. As we consider impurities away from the middle point and closer, say, to  $j_1$ , decrease of  $J_{kj_2}$  is fully compensated by the increase of  $J_{kj_1}$ , until we enter the region where the spins of the impurities are saturated due to their proximity to  $j_1$ . Thus the size of the region of interest in the direction along  $\mathbf{r}_{j_1} - \mathbf{r}_{j_2}$  depends on the temperature. Since the characteristic correlation temperature  $T_{2p}(r)$  is the quantity to find, we will proceed by iterations, starting with the value given by the right-hand side of Eq. (2) with  $A(r, a_B, n_i) = 1$  (so the thickness of the lens-shaped interaction region equals  $a_B$ ). As we will see, just one iteration yields the answer with good precision.

We will take into account only the impurities in the interaction region; the total number of these impurities equals  $N \sim a_B^2 r n_i$  in the first iteration. The coupling of each of these impurities to either hole is approximately  $J(r/2) \equiv J_0 \exp(-r/a_B)$ . It allows us to reduce the Hamiltonian to the following form:

$$\hat{H} = sJ(r/2) \cos \frac{\theta}{2} \sum_{k=1}^N \mathbf{S}_k^{(z)}. \quad (3)$$

Here the angle between the spins of the two polarons is denoted  $\theta$ , and the direction of the  $z$  axis is chosen along the direction of the vector  $s_{j_1} + s_{j_2}$ . We have neglected spatial variation of the polaron exchange field in the interaction region, since the relative magnitude of this variation does not exceed unity.

The partition function of the system described by Hamiltonian (3) can easily be found, and after straightforward algebra we arrive at the expression for the average cosine of the angle  $\theta$  between the spins of the two interacting polarons:

$$\langle \cos \theta \rangle \sim \begin{cases} N[sSJ(r/2)/T]^2, & \sqrt{N} sS|J(r/2)| \ll T \\ 1 - \frac{1}{N[sSJ(r/2)/T]^2}, & \sqrt{N} sS|J(r/2)| \gg T. \end{cases} \quad (4)$$

Therefore, the spins of the two polarons at distance  $r$  are correlated at temperatures below  $\sqrt{N} sS|J(r/2)|$ . Using the estimate  $N \approx a_B^2 r n_i$ , we finally arrive at

$$T_{2p}(r) \sim a_B \sqrt{r n_i} sS|J_0| \exp(-r/a_B). \quad (5)$$

The maximum possible distance  $r_{\text{corr}}(T)$  between two polarons with correlated spins at a given temperature  $T$  is therefore

$$r_{\text{corr}}(T) \sim a_B \left[ \ln \frac{sS|J_0|}{T} + \frac{1}{2} \ln \left( a_B^3 n_i \ln \frac{sS|J_0|}{T} \right) \right]. \quad (6)$$

Now we can use result (5) to get a better estimate for the thickness of the lens-shaped interaction region in order to perform the next iteration. It is determined by the condition  $J_{kj_2} < T$ , so the thickness is of the order of  $a_B \ln(a_B \sqrt{r n_i})$ . The resulting correction to  $T_{2p}(r)$  is just a factor of  $\ln(a_B \sqrt{r n_i})$ . Taking this correction into account is clearly beyond the accuracy limits of the approximation used above, so we neglect it, and use Eq. (5) as our final result.

Now we consider a system of randomly placed magnetic polarons. As the temperature is being lowered, the spins of neighboring polarons become aligned, and clusters of polarons with the same spin appear. At any given temperature  $T$ , the polarons separated by a distance smaller than  $r_{\text{corr}}(T)$  are joined into magnetic clusters. The lower the temperature, the more such "links" between polarons are established, and the larger the average cluster size. Finally, at low enough temperatures, a cluster having the dimensions of the sample, the so-called "infinite cluster" appears, and the magnetization of the sample acquires some finite value. The problem of finding the transition temperature is identical to the problem of finding the critical percolation radius in the problem of randomly placed overlapping spheres [17]. The latter problem has been solved numerically, and it has been demonstrated that the infinite cluster forms in the system when the link length reaches the value  $r_{\text{perc}} \approx 0.86/\sqrt[3]{n_h}$  [19]. Substituting this expression into Eq. (5), we get the expression for the ferromagnetic transition temperature

$$T_c \sim sS|J_0| (a_B^3 n_h)^{1/3} \sqrt{n_i/n_h} \exp\left(-\frac{0.86}{(a_B^3 n_h)^{1/3}}\right). \quad (7)$$

The limit of applicability of Eq. (7) is determined by the condition  $a_B^3 n_h \ll 1$ . A similar exponent was obtained in Ref. [20] for ferromagnetic transition in PdFe alloys.

It is instructive to point out that our Eq. (7) is consistent with the mean-field result derived in the literature [1–4] for the opposite limit of almost free holes,

$$T_c \sim n_i J_{\text{mf}}^2 \xi(T_c),$$

where  $J_{\text{mf}}$  is related to  $J_0$  of Eq. (1) by  $J_{\text{mf}} \sim a_B^3 J_0$ , and  $\xi(T) \sim n_h/\max\{\varepsilon_F, T\}$  is the magnetic susceptibility of holes, with  $\varepsilon_F$  being the Fermi energy. In the case of low electron density,  $T \gg \varepsilon_F$ ,

$$T_c \sim \sqrt{n_i n_h} |J_{\text{mf}}|,$$

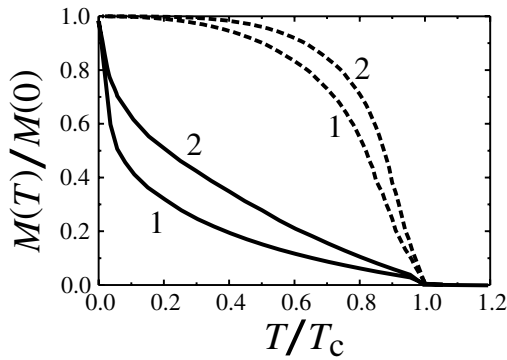


FIG. 2. The solid lines show the relative magnetization of the magnetic impurities [Eq. (9)] for  $a_B^3 n_h = 10^{-3}$  (curve 1) and  $10^{-2}$  (curve 2). The dashed lines show the relative magnetization of localized holes, whose contribution to the total sample magnetization is small.

which matches the result (7) of the bound-polaron picture at the limit of applicability of the latter,  $a_B^3 n_h \sim 1$ .

Since  $n_h \ll n_i$ , one polaron includes many magnetic impurities, and the total magnetization of the sample is that of impurities:

$$M(T) = S n_i \mathcal{V} [r_{\text{corr}}(T, n_i, n_h, a_B) \sqrt[3]{n_h}], \quad (8)$$

where  $r_{\text{corr}}$  is defined by Eq. (6), and universal function  $\mathcal{V}(y)$  is the infinite cluster's volume in the model of overlapping spheres; it depends only on the product  $y$  of the spheres' diameters and the cubic root of their concentration.

Using Eqs. (6) and (7), we cast Eq. (8) in the form

$$\frac{M(T)}{M(0)} = \mathcal{V} \left[ 0.86 + (a_B^3 n_h)^{1/3} \ln \frac{T_c}{T} \right], \quad (9)$$

with the Curie temperature  $T_c$  given by Eq. (7). One can see that the shape of the magnetization curve is determined by only one dimensionless parameter  $a_B^3 n_h$ , while the expression (7) for  $T_c$  is more complicated and depends on all parameters of the model. Figure 2 shows the temperature dependence of the magnetization at two values of  $a_B^3 n_h$ ; the curve is more concave at smaller values of this parameter. The concave shape of the curve is consistent with the experimental magnetization data in  $\text{Ge}_{1-x}\text{Mn}_x$  [21] and low- $T_c$  III-V samples [22] where our polaron percolation picture applies better due to stronger carrier localization associated with lower values of  $a_B^3 n_h$ . Our magnetization results also agree with the numerical results of Ref. [6].

To conclude we have developed an analytic polaron percolation theory for diluted magnetic semiconductor ferromagnetism in the limit of low carrier density or equivalently strong carrier localization  $a_B^3 n_h \ll 1$ . Interestingly

our polaron percolation theory reproduces the free carrier Weiss mean field theory in the limit of  $a_B^3 n_h \sim 1$ . Our analytic results are in good agreement with existing numerical results in the strongly localized limit [5–7]. The experimental systems currently have  $a_B^3 n_h \sim 10^{-1} - 10^{-3}$  which makes our theory marginally applicable to the experimental systems, and we get reasonable agreement with experimental results for  $T_c$  [23] and for  $M(T)$ .

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  - [23] Substituting the values of the parameters for  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$   $a_B = 7.8 \text{ \AA}$ ,  $J = 15 \text{ meV}$ ,  $x = 0.05$ ,  $n_h/n_i = 0.1$  [6], we get  $T_c \sim 10 \text{ K}$ , which is in reasonable agreement with the experimental result  $T_c \sim 100 \text{ K}$  taking into account only marginal applicability of our model to GaMnAs.