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Hole concentration vs. Mn fraction in a diluted (Ga,Mn)As ferromagnetic semiconductor

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ABSTRACT

The dependence of the hole density on that of Mn sites in $Ga_{1-x}Mn_xAs$ is studied within a mean-field approach to the hole-mediated ferromagnetism in III-V Mn-based semiconductor compounds. We parametrize the hole concentration, p, as a function of the fraction of Mn sites, x, in terms of the product $m^*(J_{pd})^2$ (where m^* is the hole effective mass and J_{pd} is the Kondo-like hole/local-moment coupling), and the critical temperature T_c. By fitting $m^*(J_{pd})^2$ to experimental data for $T_c(x)$, we establish the dependence of p on x, which is interpreted in terms of a reentrant metal-insulator transition taking place in the hole gas.

INTRODUCTION

The discovery in the early 1990's of ferromagnetism in III-V materials alloyed with transition elements like Mn [1,2] has increased the interest in the study of the electronic, optical and transport properties of diluted magnetic semiconductors (DMS). Ferromagnetic semiconductors bring about the possibility of controlling both spin and charge degrees of freedom. The combination of this feature with the capability of growing low-dimensional structures has opened up new perspectives for the production of spintronic devices. Applications would include non-volatile memory systems [3] and quantum computing [4].

A great deal of attention has been focused on $Ga_{1-x}Mn_xAs$ alloys, which exhibit very interesting magnetic and transport properties. When Mn atoms replace Ga in GaAs, their five 3d electrons remain localized in a core state, giving rise to S = 5/2 local moments. In addition, Mn atoms have one less electron in the 4s level than Ga, so they act as acceptors generating hole states in the material; for large enough doping, these states merge to form an impurity band. It should be pointed out that the equilibrium solubility of Mn atoms in GaAs is quite low [5], being only of the order of 10^{19} cm^{-3}. However, with help of molecular-beam epitaxy techniques at low temperatures, it has been possible to produce homogeneous samples of $Ga_{1-x}Mn_xAs$ with x as high as 0.071. Magnetization measurements in these systems show that for $0.015 \leq x \leq 0.071$ they become ferromagnetic, with doping-dependent critical temperatures $T_c(x)$ reaching a maximum of 110 K for x = 0.053 [6].

The origin of ferromagnetic order in these materials is not fully understood, but there exists consensus on the fact that it results from the exchange coupling between the localized Mn moments mediated by the holes. The strength of the of the coupling is expected to depend on the hole concentration p, which eventually determines the Curie temperature $T_c(x)$. In principle, one would expect that each Mn would provide one hole, leading to a density of holes equal to that of the magnetic ions. However, this has not been confirmed by experiment. Although an accurate determination of the hole concentration is hindered by the anomalous Hall term, experimental data indicate that p is of the order of a 15 to 30 % fraction of that of magnetic ions [6-9]. The

mechanism responsible for the discrepancy between hole and Mn densities is not clear. Matsukura et al [7] have pointed out that such discrepancy might be due to compensation of Mn acceptors by deep donors such as As antisites, which are known to be present at high concentration in low-temperature grown GaAs [10]. However, other mechanisms such as the formation of sixfold-coordinated centers with As (Mn^{6As}), which would compensate Mn atoms on substitutional Ga lattice sites [11], should not be ruled out. As a consequence, no theory relating the hole concentration and that of Mn has as yet been proposed. However, this issue is of great interest for the design of new devices. The main purpose of the present work is to present a preliminary quantitative analysis on this matter.

THEORETICAL FRAMEWORK

The appearance of ferromagnetism in $Ga_{1-x}Mn_xAs$ has been explained on the basis of the following picture. The Mn ions interact with the holes via a local antiferromagnetic Kondo-like exchange coupling J_{pd} between their moments [9,12-15], which results in the polarization of the hole subsystem; this, in turn, gives rise to an effective ferromagnetic coupling between the Mn moments. We point out that there has been some debate as far as the details of the above picture are concerned (e.g., whether or not such effective interaction is well described by an RKKY term [16,17]). Nevertheless, there is consensus on the fundamental role played by the hole-mediated mechanism. We emphasize, however, that the approach we follow here does not depend on the details of the effective Mn-Mn interaction.

We can thus think of two coupled subsystems (localized moments and holes) as described by the total Hamiltonian

$$H = H_{Mn} + H_h + J_{pd} \sum_{i,I} \mathbf{S}_I . \mathbf{s}_i \delta(\mathbf{r}_i - \mathbf{R}_I) . \qquad (1)$$

Here H_{Mn} describes the *direct* (i.e., non--hole-mediated) antiferromagnetic exchange between Mn spins, H_h describes the hole subsystem, and the last term corresponds to the aforementioned Mn-hole exchange interaction, with $\mathbf{S}_I$ and $\mathbf{s}_i$ labeling the localized Mn spins (S = 5/2) and the hole spins (s = 1/2), respectively. As a first approach, we neglect H_{Mn} and consider H_h within a parabolic-band effective-mass approximation; a more general description of H_h may include a multi-band Kohn-Luttinger treatment [9,18], effects of impurity potentials, a site energy term arising from the Mn potential, and a correlation energy representing hole-hole repulsion.

The problem defined by the Hamiltonian in Eq. (1) is indeed quite complex, but some physical insight can be gained by a mean-field approach. Assuming that the magnetization density of the hole subsystem, $M_h = < n_\uparrow - n_\downarrow >$, is uniform within the length scale of magnetic interactions, the Mn magnetization is given by

$$M = N_{Mn} g \mu_B M_I = n_{Mn} g \mu_B S \, B_S \left[\left(\frac{J_{pd} S}{2 k_B T} \right) M_h \right], \qquad (2)$$

where $n_{Mn} = x \, n_c$ is the density of Mn ions, with n_c being the density of Ga lattice sites, M_I is the magnetization density of the Mn ions, g = 2 is the Mn Landé g - factor, and B_S is the Brillouin

function. The assumption of homogeneity of the hole magnetization allows us to evaluate M_h self-consistently by considering a Fermi sea of holes with effective mass m^*, in the presence of the mean magnetic field generated by the Mn ions. We therefore find that

$$M_h \propto m^* J_{pd} \times M_I \, p^{1/3}. \qquad (3)$$

RESULTS AND DISCUSSION

The critical temperature as a function of hole density and Mn composition is obtained by linearizing the self-consistency relations given by Eqs. (2) and (3), i.e.,

$$T_c \propto \left[\left(m^*/m_e\right) J_{pd}^2\right] \times p^{1/3}, \qquad (4)$$

which leads one to write the hole concentration as

$$p = \zeta \left\{ \frac{T_c(x)}{(m^*/m_e) J_{pd}^2 x} \right\}^3, \qquad (5)$$

where $\zeta = 5.29 \times 10^{-16}$, in units such that J_{pd} is given in eV nm^3.

The sought-after relation between p and x is provided by Eq. (5). However, it depends on the parameters m^* and J_{pd}, whose values have not been determined with certainty from the experimental data. If one tentatively uses $m^* = m_e$, $J = 0.15$ eVnm^3 [14], and the experimental transport data [6] for T_c , the resulting estimates for p(x) are as given by the full circles in Fig. 1. The error bars reflect the uncertainties in the determination of T_c, as displayed in Fig. 3(c) of Ref. 6. Nevertheless, a more reliable estimate can be obtained by adopting the following strategy. Since the Hall resistance measurements [19] yield an unambiguous [20] value of $p = 3.5 \times 10^{20}$ cm^{-3} for the sample with $x = 0.053$, for which $T_c = 110$ K, one may use Eq. (5) to determine the value of the product $(m^*/m_e)(J_{pd})^2 = 2.4 \times 10^{-2}$ (eV $nm^3)^2$. Thus, by taking this estimated value along with the experimental data [6] for T_c into Eq. (5), one gets the data for p(x) shown as full squares over a wide range of x. The adequacy of the above procedure is confirmed by the fact that the calculated values for p(x) lie below the concentration of Mn ions, shown as a dotted line, in agreement with experiment. Before accepting these estimates for p(x) at face value, one should note that a closer look at the experimental data for $T_c(x)$ [6] suggests a linear behavior in the range of x of the order 0.015 - 0.035, which would imply, through Eq. (4), a constant p in that range; such constant behavior, however, should not prevail at low concentrations, $x \rightarrow 0$, where presumably one should have $p \propto x \rightarrow 0$. These considerations are incorporated in the full curves displayed in Fig.1, which lie within the corresponding error bars of the calculated hole concentrations.

The boundaries of the metal-insulator transitions (MIT's), as determined from resistivity measurements [6], are indicated in Fig. 1 by vertical dashed lines. We remark that the present theoretical estimates for p in the insulating phases are based on the assumption that the scale

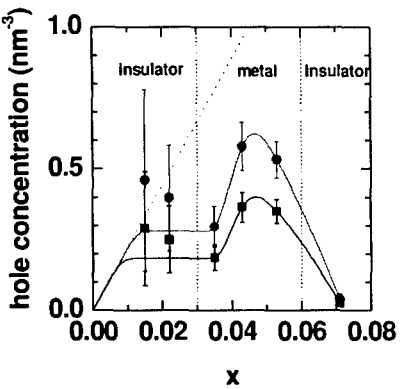

Figure 1. Theoretical results for the hole concentration as a function of the fraction of Mn sites for (Ga,Mn)As ferromagnetic alloys. The dashed line corresponds to a hole concentration equal to that of Mn sites, whereas the filled circles and squares are the present mean-field results. Full curves are obtained as reasoned in the text. I and M respectively denote insulating and metallic phases.

of localization length in insulating samples, though finite, is significantly larger than the length of magnetic interactions [9], in which case the present mean-field approach is a good starting point.

The most prominent feature of Fig. 1 is the fact that p(x) displays a maximum within the metallic phase. It means that attempts to increase T_c should be carried out for samples in the metallic phase, for Mn concentrations about 0.05. Moreover, notwithstanding the considerable uncertainties [21] in the measurements of p, the present theoretical estimates for the hole concentration exhibit trends in quite good agreement with those obtained from Hall measurements by Matsukura et al [7].

The fact that the dependence of p with x is essentially related to the occurrence of MIT's taking place in the hole subsystem can be seen from the following qualitative picture. Within our approximation, the Fermi energy tracks the behavior of p, since $\varepsilon_F \propto p^{2/3}$, while the exchange splitting $\Delta \propto x$. Figure 2(a) shows the schematic impurity bands for each spin channel, in the very-low doping regime in which the gas is supposed to be unpolarized. As x increases, the gas can sustain polarization and still be insulating, provided the Fermi energy lies below the mobility edge, as shown in Fig. 2(b). Further increase in x causes ε_F to increase and to lie within the delocalized states of the up-spin impurity band, as depicted in Fig. 2(c): The system becomes metallic. Whether or not the Fermi energy also lies within the delocalized region of the down-spin impurity band is a very interesting question, which cannot be answered by our simple model; the solution of this particular issue should have bearings on the efficiency of $Ga_{1-x}Mn_xAs$ - based devices as spin filters. Once ε_F reaches a maximum within the metallic phase, its initial decrease upon increasing x is compensated by an increase in Δ, so that the Fermi level still lies within the delocalized states. However, with continuing increase in x the exchange splitting can no longer make up for the decrease in ε_F, and the latter eventually crosses the mobility edge again, lying within localized states [Fig. 2(d)]: The system reenters an insulating phase.

The simple approach used in the present work provides a straightforward and immediate estimate of the hole concentration in $Ga_{1-x}Mn_xAs$ as a function of both x and T_c [22], which is an important ingredient to guide further experimental work on this subject. Clearly, the present

approach could be extended to include a more complete description of the acceptor states, taking into account the spin degrees of freedom, spin-orbit coupling, compressive/tensile strains, etc. Moreover, a proper treatment of disorder -- e.g., by explicitly considering a random, instead of continuous, distribution of Mn ions -- should lead to a more realistic description of the MIT. In this respect, many-body effects due to correlation among the holes should also influence p(x), especially in the insulating phase. Of course, an appropriate description of the physical mechanisms related to As antisites and Mn^{6As} centers, as far as the hole vs. Mn concentrations is concerned, is certainly a formidable task, which nonetheless deserves future theoretical attention.

In summary, we have considered a mean-field theory for the hole-mediated coupling between the localized Mn moments in $Ga_{1-x}Mn_xAs$. The hole concentration as a function of the fraction of Mn sites is obtained in terms of the product $m^*(J_{pd})^2$ and the critical temperature T_c. By using experimental data for these latter quantities, we have established that the occurrence of a reentrant MIT taking place in the hole gas can be traced back to the dependence of the hole concentration with x. Although not shown here, we have also calculated the dependence of the Mn magnetization with x, for different temperatures, and found that as T increases, the width of the composition-dependent magnetization decreases drammatically, and that the magnetization maxima also decreases, indicating the need for quality-control of Mn doping composition in DMS devices.

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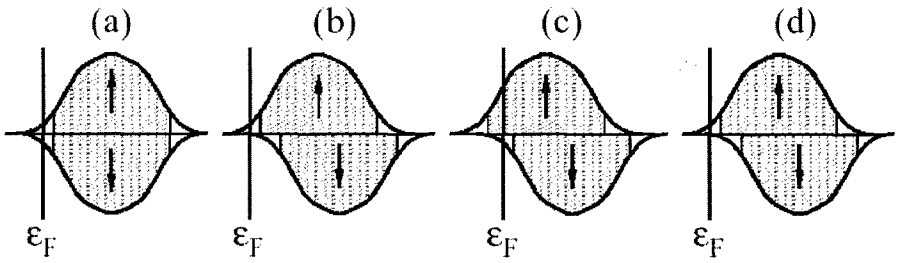

Figure 2. Schematic density of states (DOS) versus energy for the impurity band, for up- (top) and down-spins (bottom). Under each DOS curve, the hashed and empty regions correspond, respectively, to delocalized and localized states; these are separated by mobility edges. The exchange splitting is proportional to the off-set between the ↑ and ↓ bands, and the Fermi energy (ε_F) increases to the right.

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21. See, e.g., Fig. 2 of Ref. 7: for $x = 0.022$, the experimentally determined p has an error bar which covers over one decade, while for $x = 0.071$ the error bar runs over two decades.
22. For a treatment of the same model within spin-wave theory and dynamical mean-field theory, see J. Konig, H.-H. Lin, and A. H. MacDonald, *Phys. Rev. Lett.* **84**, 5628 (2000); *ibid.* **86**, 5637 (2001), and A. Chattopadhyay, S. Das Sarma, and A. J. Millis, *cond-mat*/0106455. The explicit dependence of p with x, however, is not discussed in those works.