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Physica B 312–313 (2002) 815–817

PHYSICA B

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Mean-field approach to disorder effects on ferromagnetism in $Ga_{1-x}Mn_xAs$

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Abstract

Using a numerical mean field treatment, we examine the onset of ferromagnetism and the nature of the ferromagnetic phase in $Ga_{1-x}Mn_xAs$ for charge carrier densities in the vicinity of the metal–insulator transition. Our approach, which explicitly takes into account the positional disorder of the Mn ions, shows that the ferromagnetic transition temperature is significantly enhanced by the disorder. Concurrently, a very unusual temperature dependence of the magnetization is obtained in the ferromagnetic phase. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Diluted magnetic semiconductors; Ferromagnetism

Interest in diluted magnetic semiconductors was given an impetus by the recent demonstration of a ferromagnetic critical temperature $T_C = 110$ K for $x \approx 0.05$ in GaMnAs [1]. To date, most theoretical models proposed assume that the holes occupy a Fermi sea in the valence band [2]. This implies a density of charge carriers large enough to effectively screen the Coulomb interactions between the Mn ions and the charge carriers. Then, the holes have a spatially homogeneous distribution throughout the system, and the disorder in the Mn positions plays little (or no) role. However, it is known that GaMnAs is insulating for $x < 0.03$ and shows reentrant insulating behavior for $x > 0.07$, for a review of properties of GaMnAs see Ref. [3]. Also, experimentally it was found that these alloys are heavily compensated, with a concentration of charge carriers which is only about 10% of the concentration of Mn dopants [4]. This suggests small Fermi energies and very long screening lengths of Coulomb interactions, and opens up the possibility that the charge carriers are in fact at the bottom of an impurity band whose properties may differ significantly from those of the valence band. In particular, since this impurity band is made up of shallow levels trapped about the various Mn acceptors, disorder in the positions of the Mn dopants may play a significant role in the magnetic properties of this system.

We examine this conjecture in this paper, by investigating the Hamiltonian:

$$H = \sum_{ij,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \hat{\mathbf{s}}_j. \quad (1)$$

Here, i indexes the positions $\mathbf{R}_i$ of the Mn dopants, which are randomly placed on the FCC Ga sublattice, and $c_{i\sigma}$ is the annihilation operator for a charge carrier with spin σ in the impurity level centered on the ith Mn acceptor. For simplicity, we assume that these impurity levels are described by s-type orbitals $\phi_i(\mathbf{r}) = \exp(-|\mathbf{r} - \mathbf{R}_i|/a_B)$, where the Bohr radius $a_B \approx 8$ Å for this system [5]. Charge carriers can hop from one orbital to another, with a hopping integral for s-orbitals given by $t_{ij} = 2(1 + r/a_B)\exp(-r/a_B)$, where $r = |\mathbf{R}_i - \mathbf{R}_j|$ [6]. The hopping integral is measured in Ry, where 1 Ry = 112.4 meV is the binding energy of the hole in the shallow orbital of Mn in GaAs [5]. The second term in the Hamiltonian describes the antiferromagnetic (AFM) Heisenberg exchange interaction between the spin $\frac{5}{2}\mathbf{S}_i$ of the ith Mn with the spin $\hat{\mathbf{s}}_j = \frac{1}{2} c^{\dagger}_{j\alpha} \sigma_{\alpha\beta} c_{j\beta}$ of the charge carrier in an orbital centered at the jth impurity. The AFM coupling between a Mn spin and the angular momentum $\mathbf{J}$ of its own hole ($i = j$) is $\varepsilon = 5$ meV [5]. Therefore, we choose the coupling $J_{ii} = J = 3\varepsilon$. The

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PII: S0921-4526(01)01209-1

factor of 3 is the simplest way to account for the fact that the heavy hole has a total angular momentum $j = \frac{3}{2}$ instead of a spin $\frac{1}{2}$ as described by the above model. For $i \neq j$, the exchange integral is proportional to the probability to find the hole near the Mn spin, $J_{ij} = J|\phi_j(\mathbf{R}_i)|^2$.

We treat this Hamiltonian within the mean-field approximation, factorizing the spin interaction term as $\mathbf{S}_i \cdot \hat{\mathbf{s}}_j \to \langle \mathbf{S}_i \rangle \cdot \hat{\mathbf{s}}_j + \mathbf{S}_i \cdot \langle \hat{\mathbf{s}}_j \rangle - \langle \mathbf{S}_i \rangle \cdot \langle \hat{\mathbf{s}}_j \rangle$. The charge carrier Hamiltonian is now quadratic and can be numerically diagonalized for any configuration $\langle \mathbf{S}_i \rangle$ of the Mn spins, allowing the calculation of the charge carrier spin expectation values $\langle \hat{\mathbf{s}}_j \rangle$. In turn, these allow us to compute the new expectation values for the Mn spins, $\langle \mathbf{S}_i \rangle$, and the process is repeated until self-consistency is reached. As is usually the case in mean-field approximations, the symmetry to spin rotations is spontaneously broken and the expectation values are non-zero for some direction, which we choose as the z-axis. Thus, we can compute the average Mn spin $S_{\mathrm{Mn}} = 1/N_{\mathrm{d}} \sum_i \langle S_i^z \rangle$ and the average hole spin $s_{\mathrm{h}} = 1/N_{\mathrm{h}} \sum_i \langle \hat{s}_i^z \rangle$, where N_{d} and N_{h} are the total number of Mn spins and charge carriers, respectively.

In Fig. 1 we plot the average Mn spin $S_{\mathrm{Mn}} > 0$ and the average hole spin $s_{\mathrm{h}} < 0$ as a function of temperature, for typical configurations. The opposite signs are a signature of the AFM exchange interaction. The four sets of curves correspond to systems with Mn doping $x = 0.0093$, which contain $N_{\mathrm{d}} = 512$ Mn impurities and $N_{\mathrm{h}} = 51$ holes, but which differ in the amount of disorder allowed in the Mn positions. The curve with the lowest T_{C} corresponds to a perfectly ordered Mn simple cubic supperlattice, with a lattice constant $a_{\mathrm{L}} = a/(4x)^{1/3} = 3a$, where $a = 5.65$ Å is the lattice constant of the underlying FCC Ga sublattice. In this case, there is no disorder in the system. The symmetry to superlattice translations implies that $\langle \mathbf{S}_i \rangle$ and $\langle \hat{\mathbf{s}}_i \rangle$ are independent of i. As a result, the charge carrier Hamiltonian is diagonalized with Bloch waves, and we get an impurity band of delocalized states, i.e. with a structure very similar to the conduction band. The magnetization curves we obtain in this case have a Brillouin-like shape, and are very similar to the ones obtained in models which assume that the holes occupy only conduction band states, and therefore are spatially homogeneously distributed [2]. In order of increasing T_{C}, the other three sets of curves in Fig. 1 correspond to configurations in which the Mn dopants are weakly, moderately and completely disordered, respectively. A configuration with weak disorder is obtained by randomly displacing each Mn dopant from its ordered supperlattice position to one of the 12 nearest neighbor sites from the underlying FCC Ga sublattice. The moderate disorder configuration is obtained by randomly placing the Mn on the FCC Ga sublattice, subject to the constraint $|\mathbf{R}_i - \mathbf{R}_j| > 2a$ for any $i \neq j$. Finally, a completely disordered sample is obtained by randomly placing the Mn on the Ga sublattice, with no constraints.

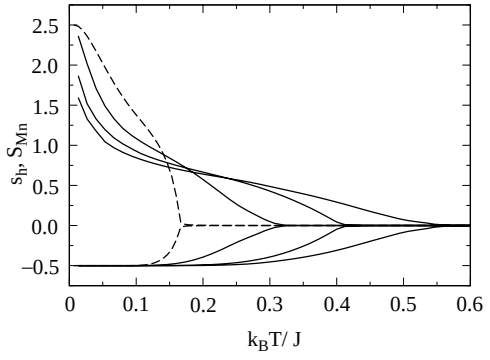

Fig. 1. The average Mn spin S_{Mn} and average spin per hole s_{h} for doping concentration $x = 0.00926$ and $N_{\mathrm{d}}/N_{\mathrm{h}} = 10$. In increasing order of T_{c}, the curves correspond to an ordered, weakly disordered, moderately disordered and completely random distributions of Mn (see text).

The most surprising observation, at first sight, is that the critical temperature T_{C} *increases significantly* with increasing disorder. In fact, there is a very simple physical explanation for this. In the ordered system, the holes are homogeneously distributed among the Mn sites, and therefore they polarize all the Mn spins in the system equally ($\langle \mathbf{S}_i \rangle$ is independent of i). Since the number of holes is only 10% of the number of Mn, the critical temperature below which an ordered magnetic state can be established is fairly low. On the other hand, in a disordered system, the holes are inhomogeneously distributed. Regions with higher local concentration of Mn have a higher local density of holes, while regions with low local Mn concentration are less likely to be visited by holes. Since the AFM interaction between a Mn spin and a charge carrier spin is proportional to the probability of having the charge carrier in the vicinity of the spin, it follows that this interaction is strongly enhanced for spins in the higher Mn concentration regions. As a result, these regions become magnetized at much higher temperatures, leading to an increased T_{C}. On the other hand, for spins in the lower Mn concentration regions the AFM exchange is significantly decreased, and as a result these spins do not order unless the temperature becomes very low. This explains the unusual concave shape of the magnetization curve, which is similar to experimental curves reported for GaMnAs [4]. We would also like to emphasize that the same effect is observed in systems with larger concentration x of Mn, although the increase in T_{C} is

comparatively smaller, due to the fact that local fluctuations in the Mn concentration are smaller as x increases.

In conclusion, we have shown that if the Coulomb attraction between Mn ions and charge carriers is not fully screened, disorder in the Mn positions can have a significant effect on the nature of the ferromagnetic phase of GaMnAs.

M.B. acknowledges a Postdoctoral Fellowship from NSERC. This project was supported by NSF DMR-9809483.

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