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Hall effect and magnetic properties of III–V based (Ga$_{1-x}$Mn$_x$)As/AlAs magnetic semiconductor superlattices
Magnetic-field effects in defect-controlled ferromagnetic Ga$_{1-x}$Mn$_x$As semiconductors

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We have studied the magnetic-field and concentration dependences of the magnetizations of the hole and Mn subsystems in diluted ferromagnetic semiconductor Ga$_{1-x}$Mn$_x$As. A mean-field approximation to the hole-mediated interaction is used, in which the hole concentration $p(x)$ is parametrized in terms of a fitting (of the hole effective mass and hole/local moment coupling) to experimental data on the $T_c$ critical temperature. The dependence of the magnetizations with $x$, for a given temperature, presents a sharply peaked structure, with maxima increasing with applied magnetic field, which indicates that application to diluted-magnetic-semiconductor devices would require quality control of the Mn-doping composition. We also compare various experimental data for $T_c(x)$ and $p(x)$ on different Ga$_{1-x}$Mn$_x$As samples and stress the need of further detailed experimental work to assure that the experimental measurements are reproducible. © 2003 American Institute of Physics. [DOI: 10.1063/1.1534622]

Diluted magnetic semiconductors (DMS) have become one of the most promising classes of materials for spintronics applications. This is mainly due to the possibility of manipulating both the charge and spin degrees of freedom of electrons or holes to process and store information in magnetic materials. The discovery of hole-induced ferromagnetism in p-type (In,Mn)As systems$^1$ was followed by the successful growth of ferromagnetic (Ga,Mn)As alloys.$^2$ Interest in the understanding of the physics in these materials has boosted due to the fact that ferromagnetic III–V alloys may be readily combined into semiconductor heterostructure systems, opening up a range of applications of optoelectronic devices through the combination of quantum and magnetic phenomena in these materials. However, several issues in relation to these systems need to be elucidated before full-scale applications can be efficiently implemented. For instance, a light-emitting device based on III–V heterostructures has been proposed,$^3$ which relies on the injection of holes from a (Ga,Mn)As layer in the presence of a magnetic field. Therefore, the magnetic response of the hole subsystem should be known to some detail, which, in turn, must reflect the dependence of the hole concentration with the Mn composition. The latter is still a challenging problem: While in principle each Mn atom should provide one hole, leading to a density of holes, $p$, equal to that of the magnetic ions, early experimental data already indicated that $p$ is only a 15% to 30% fraction of that of magnetic ions.$^4,5$ We have recently addressed this issue$^6$ through a mean-field approximation to a Hamiltonian incorporating the hole-mediated mechanism$^7$ and found that the hole concentration displays a nonmonotonic behavior with a maximum near $x = 0.05$, within the insulating phase. In spite of its simplicity, this mean-field framework should provide a qualitative description of the response to a magnetic field. With this in mind, here we obtain the magnetization of both the hole gas and of the Mn subsystem, as functions of the magnetic field and of the Mn composition.

We start with a Hamiltonian for the coupled hole and local moments subsystems in the form

$$
\mathcal{H} = \mathcal{H}_h + J_{pd} \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{s}_j \delta(\mathbf{r}_i - \mathbf{r}_j) + g_{\text{Mn}} \mu_B \mathbf{H} \cdot \sum_i \mathbf{S}_i - g_h \mu_B \mathbf{H} \cdot \sum_i \mathbf{s}_i,
$$

(1)

where the direct (i.e., non-hole-mediated) antiferromagnetic exchange between Mn spins has been neglected, $\mathcal{H}_h$ describes the hole subsystem, $\mathbf{S}_i$ and $\mathbf{s}_i$ label the localized Mn spins ($S = 5/2$) and the hole spins ($s = 1/2$), respectively; the second term corresponds to the Mn-hole exchange interaction, and the last two terms represent the coupling to the external field $\mathbf{H}$. Within the spirit of a mean field approximation, the magnetization of the Mn subsystem is then given by

$$
M = n_s g_{\text{Mn}} \mu_B x S B_S \left[ \frac{S}{2k_B T} \left( J_{pd} M_h + 2 g \mu_B H \right) \right],
$$

(2)

which must be determined self-consistently with the hole magnetization.
$M_n = A \left( \frac{4J_{pd}}{a^3} \frac{M}{n_s g \mu_B} + g \mu_B H \right) p^{1/3},$  

(3)

where $n_s$ is the density of Ga lattice sites, $g = g_h - g_{Mn} = 2$, $\mu_B$ is the Bohr magneton, $S = 5/2$ is the Mn spin, $B_s(\cdots)$ is the Brillouin function, $A = (3\pi^2)^{-2/3}(3m^*/2\hbar^2)$, and $a$ is the GaAs lattice constant; the product $m^*J_{pd}$ was determined in Ref. 6 through a fit to experimental data, and we now take $m^* = m_e$, the electronic bare mass, which is within the limits recently set by infrared spectroscopy. From Eq. (3), we see that the dependence of $p$ with $x$ influences the magnetic behavior in a fundamental way. Here we use $p(x)$ as parametrized in terms of a fitting to experimental data on the critical temperature $T_c$, and shown as the lower curve in Fig. 1; the qualitative agreement of these data with those recently obtained by Hall measurements (represented by the squares in Fig. 1) indicates that our procedure provides a reliable input to discuss the magnetic behavior in the presence of an external field.

In Fig. 2 we show the hole and Mn magnetizations—obtained from self-consistent solutions of Eqs. (2) and

FIG. 1. Hole concentration as a function of Mn composition in Ga$_{1-x}$Mn$_x$As alloys: the full curve is the theoretical result obtained in Ref. 6, the cross corresponds to the experimental datum quoted in Ref. 5, the full squares are the experimental data by Edmonds et al. (Ref. 8), the full triangles are the experimental data by Seong et al. (Ref. 9), and the dashed line corresponds to a hole concentration equal to that of the Mn sites.

FIG. 2. Magnetizations of the hole ($M_h$, full lines) and Manganese ($M/n_s g \mu_B$, dotted lines) subsystems as functions of the applied magnetic field, for a fixed temperature of 100 K, and different Mn concentrations.

FIG. 3. Magnetizations of the hole (a) and Mn (b) subsystems as functions of the Mn concentration for different applied magnetic fields, and for a fixed temperature of 100 K.

FIG. 4. Experimental $T_c(x)$ in Ga$_{1-x}$Mn$_x$As alloys: circles are data from Ohno et al. (Ref. 5), squares from Edmonds et al. (Ref. 8), triangles from Seong et al. (Ref. 9) and Potashnik et al. (Ref. 11), and diamonds from van Esch et al. (Ref. 12). All lines through data points are guides to the eye.
— as functions of the magnetic field, for a fixed temperature $T = 100 \text{ K}$, and for three different Mn compositions. For both $x = 0.043$ and $x = 0.071$, the system does not sustain spontaneous magnetic order at this temperature. Nonetheless, their magnetic responses to an applied field are quite distinct. While for $x = 0.043$ the Mn and hole subsystems display roughly the same susceptibility, for $x = 0.071$ the Mn moments (dotted lines in Fig. 2) are more susceptible to the field than the holes (full lines); this appears as a result of $p(x = 0.071) < p(x = 0.043)$, which, in turn, may be correlated with the fact that the sample with $x = 0.043$ is metallic, and the one with $x = 0.071$ is insulating.

Figure 3 displays our results for the Mn and hole magnetizations at $T = 100 \text{ K}$, for varying Mn compositions and different values of the external magnetic field. One notices a sharply peaked structure, with both maxima and widths increasing with applied magnetic field, as one would expect. Again, one finds a difference in the behavior of the Mn and hole magnetizations: While the peak position of $M_h$ does not change with the applied field, the Mn magnetization peaks move slightly towards larger compositions; it would be interesting to investigate whether this difference can be detected experimentally, or if it is a mere artifact of the present approximations. At any rate, the fact that the magnetization vs $x$ curves broaden in the presence of an external field means that the working window of compositions for spintronics applications is also broadened.

At this point it is instructive to resume the discussion of Fig. 1. The theoretical hole concentration $p(x)$ of Fig. 1, which was parametrized via a fitting of the experimental Hall-resistance measurement at $x = 0.053$, $T_c = 110 \text{ K}$, and $p = 3.5 \times 10^{20} \text{ cm}^{-3}$ (see crosses in the lower curve of Fig. 1), is in fair overall agreement with the more recent Hall experiments of Edmonds et al. By contrast, Raman scattering measurements of the hole densities, performed by Seong et al. in four Ga$_{1-x}$Mn$_x$As samples ($x = 0.038, 0.061$, and $0.083$), resulted in a monotonically increasing $p(x)$; see the triangles in Fig. 1. This monotonic behavior is also in disagreement with the early results by Matsukura et al. Seong et al. argue that Raman scattering—unlike standard Hall measurements—provides an unambiguous and reliable method of determining the hole density in Ga$_{1-x}$Mn$_x$As systems, and comment that the difference between their results and those of Ref. 4 could be attributed to differences in detailed growth conditions. Unfortunately, it seems that the details of growth conditions (even in as-grown samples) also affects the behavior of $T_c$ with Mn composition: In Fig. 4 we display the critical temperatures as obtained by several groups. The inescapable conclusion is that the measurements of both $p(x)$ and of $T_c(x)$ in Ga$_{1-x}$Mn$_x$As are strongly sample dependent (or growth-conditions dependent). This indicates the need of further detailed experimental work on Ga$_{1-x}$Mn$_x$As compounds.

Summing up, we have investigated the effects of a magnetic field on the magnetizations of the hole and Mn subsystems in Ga$_{1-x}$Mn$_x$As semiconductor compounds. Through a mean-field approach, we have established that holes are less susceptible to the magnetic field than Mn ions at larger dopings; we have also found that the dependence of the magnetizations with $x$, for a given temperature, presents a sharply peaked structure, with both maxima and widths increasing with applied magnetic field, thus indicating that diluted-magnetic-semiconductor devices would require quality control of the Mn-doping composition.

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