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.

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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 was followed by the successful growth of ferromagnetic (Ga,Mn)As alloys. 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, 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. We have recently addressed this issue through a mean-field approximation to a Hamiltonian incorporating the hole-mediated mechanism and found that the hole concentration displays a non-monotonic 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.

Figure 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. The cross corresponds to the experimental datum quoted in Ref. The full squares are the experimental data by Edmonds et al. The full triangles are the experimental data by Seong et al. and the dashed line corresponds to a hole concentration equal to that of the Mn sites.

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

$$\mathcal{H} = \mathcal{H}_h + J_{pd} \sum_{i,l} \mathbf{S}_l \cdot \mathbf{s}_i \delta (\mathbf{r}_i - \mathbf{R}_l) +$$

$$+ g_{Mn} \mu_B \mathbf{H} \cdot \sum_l \mathbf{S}_l - g_h \mu_B \mathbf{H} \cdot \sum_i \mathbf{s}_i, \quad \text{(1)}$$
that the dependence of

from infrared spectroscopy [10]. From Eq. (3), we see

the electronic bare mass, which is within the limits recently

$p$ behavior in a fundamental way. Here we use

$B$ the Mn spin, $m$ fit to experimental data, and we now take

the product

$g\mu_B$– as functions of the magnetic field, for a fixed tempera-

ture $T$. 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

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netizations at $T = 100 K$, for varying Mn compositions

from self-consistent solutions of Eqs. (2) and (3) – as functions of the magnetic field, for a fixed tempera-
ture $T = 100 K$, 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 $p(x = 0.071) \ll p(x = 0.043)$, 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 $T = 100 K$, 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 ap-

where the direct (i.e., non–hole-mediated) antiferromag-
netic exchange between Mn spins has been neglected, $\mathcal{H}_h$

describes the hole subsystem, $S_f$ and $s_i$ label the local-
ized 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 $H$. Within the spirit

of a mean field approximation, the magnetization of the

Mn subsystem is then given by

$M = n_s g\mu_B x S B g \left[ \frac{S}{2k_B T} (J_{pd} M_h + 2g\mu_B H) \right]$, \hspace{1cm} (2)

which must be determined self-consistently with the hole

magnetization,

$M_h = A \left( \frac{4J_{pd} M}{a^3 n_s g\mu_B} + g\mu_B H \right) \rho^{1/3}$, \hspace{1cm} (3)

where $n_s$ is the density of Ga lattice sites, $g = g_h =

$g_{\text{Ga}} = 2$, $\mu_B$ is the Bohr magneton, $S = 5/2$ is

the Mn spin, $B_g$, $[\ldots]$ is the Brillouin function, $A =

(3\pi^2)^{-2/3} (3m^* / 2\hbar^2)$, and $a$ is the GaAs lattice constant; the product $m^* J_{pd}^2$ 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 [10]. 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 $T_c$ [8], 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 (3) – as functions of the magnetic field, for a fixed tempera-
ture $T = 100 K$, and for three different Mn compositions.
The theoretical hole concentration $p$ of Fig. 1 is broadened.

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 [4, 5, 8, 9, 11, 12]: 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 in order to assure that data on $T_c(x)$ and $p(x)$ are reproducible.

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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[1] H. Ohno, H. Munekata, T. Penney, S. von Molnár, and L. L. Chang, Phys. Rev. Lett. 68, 2664 (1992).
[2] H. Ohno, A. Shen, F. Matsukura, A. Oiwa, A. Endo, S. Katsumoto, and Y. Iye, Appl. Phys. Lett. 69, 363 (1996).
[3] Y. Ohno, D. K. Young, B. Beschoten, F. Matsukura, H. Ohno, and D. D. Awschalom, Nature 402, 790 (1999).
[4] F. Matsukura, H. Ohno, A. Shen, Y. Sugawara, Phys. Rev. B 57, R2037 (1998).
[5] H. Ohno, J. Magn. Magn. Mater. 200, 110 (1999); T. Omiya, F. Matsukura, T. Dietl, Y. Ohno, T. Sakon, M. Motokawa and H. Ohno, Physica E 7, 976 (2000); H. Ohno and F. Matsukura, Solid State Comm. 117, 179 (2001); T. Dietl and H. Ohno, Physica E 9, 185 (2001).
[6] R. R. dos Santos, L. E. Oliveira, and J. d’Albuquerque e Castro, J. Phys.: Condens. Matt. 14, 3751 (2002).
[7] T. Dietl, A. Haury, and Y. Merle d’Aubigné, Phys. Rev. B 55, R3347 (1997).
[8] K. W. Edmonds, K. Y. Wang, R. P. Campion, A. C. Neumann, C. T. Foxon, B. L. Gallagher, and P. C. Main, cond-mat/0205517.
[9] M. J. Seong, S. H. Chun, H. M. Cheong, N. Samarth, and A. Mascarenhas, Phys. Rev. B 66, 033202 (2002).
[10] E. J. Singley, R. Kawakami, D. D. Awschalom, and D. N. Basov, Phys. Rev. Lett. 89, 097203 (2002).
[11] S. J. Potashnik, K. C. Ku, S. H. Chun, J. J. Berry, N. Samarth, and P. Schiffer, Appl. Phys. Lett. 79, 1495 (2001).
[12] A. Van Esch, L. Van Bockstal, J. De Boeck, G. Verbanck, A. S. van Steenbergen, P. J. Wellmann, B. Grietens, R. Bogaerts, F. Herlach, and G. Borghs, Phys. Rev. B 56, 13103 (1997).

Figure 4: Experimental $T_c(x)$ in Ga$_{1-x}$Mn$_x$As alloys: circles are data from Ohno et al. [4], squares from Edmonds et al. [5], triangles from Seong et al. [6], and diamonds from van Esch et al. [12]. All lines through data points are guides to the eye.