Magnetic ordering in GaAlAs:Mn double well structure

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The magnetic order in the diluted magnetic semiconductor barrier of double AlAs/GaAs: Mn quantum well structures is investigated by Monte Carlo simulations. A confinement adapted RKKY mechanism is implemented for indirect exchange between Mn ions mediated by holes. It is shown that, depending on the barrier width and the hole concentration a ferromagnetic or a spin-glass order can be established.
In Ga$_{1-x}$Mn$_x$As [1,2], a new prototype of Diluted Magnetic Semiconductors (DMS) [3,4], the Mn$^{2+}$ cations have the 3$d$ shell partially filled with five electrons, in such a way that they carry a magnetic moment with $S = 5/2$. Besides, the Mn ion binds a hole to satisfy charge neutrality, what is, in itself, a complicate impurity problem. Two Mn ions occupying nearest neighbors positions interact via an anti-ferromagnetic coupling of their magnetic moments. In the fcc alloys, these interactions are known to be frustrated (see, e.g., Ref. [5]), establishing the possibility of settling a spin glass phase at low temperature. However, an indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange provided by the sp-d interaction between the Mn$^{2+}$ spins and the spins of the Fermi gas (unbound holes, or holes in an impurity band) competes with the nearest neighbors anti-ferromagnetic interaction. At low magnetic ion concentration the indirect exchange mechanism may become the dominant interaction, leading either to a spin-glass phase or a ferromagnetic order.

Recently some groups [1–7] succeeded in producing homogeneous samples of Ga$_{1-x}$Mn$_x$As alloys with $x$ up to 7% avoiding the formation of MnAs clusters by using low temperature ($200 - 300^\circ$C) MBE techniques. Besides its practical importance, this kind of DMS introduces an interesting problem from the physical point of view: Mn in the alloy is a strong $p$ dopant, the free hole concentration reaching even $p = 10^{20-21}cm^{-3}$ [1,2]. At small Mn concentrations, the alloy is a paramagnet and an insulator. As $x$ increases it becomes ferromagnetic, going through a non-metal/metal transition for higher concentrations ($x \approx 0.03$), and keeping its ferromagnetic phase. For $x$ above 7%, the alloy becomes a ferromagnetic insulator. In the metallic phase, the ferromagnetic transition is observed in the range of $30 - 100K$, depending on the value of $x$. The ferromagnetic order in the metallic phase is understood as resulting from the indirect exchange between the Mn ions mediated by the hole gas. In quantum wells, it seems to exist a threshold for the width of the magnetic layer in order for a ferromagnetic phase to appear [6].

In this work we perform Monte Carlo calculations to study the magnetic order resulting from the indirect exchange between magnetic moments in a particular symmetric double quantum well DMS structure formed by two GaAs wells of width $L$ separated by a Ga$_{0.65}$Al$_{0.35}$As : Mn DMS barrier width $d$. In that DMS the Mn$^{2+}$ ion concentration is taken as $x = 0.05$. The Mn$^{2+}$ ions substitute the cations elements, each of them providing a hole. In addition, the DMS is assumed to be in a metallic phase, but the density of free carriers (holes) is only a fraction $r$ of the magnetic ion concentration, what is in agreement with experimental data of Ref. [6]. A confinement-adapted RKKY [11] formalism is used to obtain the indirect exchange for the double quantum well structure:

\[
H_{ex} = -\sum_{i<j} J_{ij} \vec{S}_i \cdot \vec{S}_j, \tag{1}
\]

\[
J_{ij} = \left(\frac{I}{2A}\right)^2 \sum_{n,n'} q \sum_{\vec{q}} 2\text{Re} \left[ \phi_n^*(z_i)\phi_{n'}^*(z_j)\phi_n(z_j)\phi_{n'}(z_j)e^{-iq(R_i-R_j)} \right] \chi^{n,n'}(\vec{q}). \tag{2}
\]

with $\phi_n(z)$ representing the eigenfunctions of the potential well, $I$ the sp-d interaction [12,13], and $A$ the normalization area for the otherwise free motion in the $(x, y)$ plane. The coordinates $(\vec{R}_i, z_i)$ describe the position of the impurity $i$ in the plane $(x, y)$, and in the growth direction inside the barrier. $\chi^{n,n'}(\vec{q})$ is the equivalent to the Lindhard function:

\[
\chi^{n,n'}(\vec{q}) = \sum_{\vec{k}} \frac{\theta(E_F - \epsilon_{n,\vec{k}}) - \theta(E_F - \epsilon_{n',\vec{k}+\vec{q}})}{\epsilon_{n,\vec{k}} - \epsilon_{n',\vec{k}+\vec{q}}}. \tag{3}
\]

It is worthwhile to mention that the hole system is expected to show a spin polarization in a DMS magnetically ordered phase. This effect, which is important in spin resonant tunneling experiments, does not result into a major change in the order of the Mn$^{2+}$ impurities.

The intra-subband contribution of an occupied subband $n$ to the exchange reads:

\[
J_{ij}^{(n)} = -\left(\frac{I}{2}\right)^2 \frac{m^*_t}{\pi\hbar^2} k_F^{(n)2} \int \frac{d^3k_F}{(2\pi)^3} \phi_n(z_i) \phi_n(z_j), \tag{4}
\]

\[
[J_0(k_F^{(n)} R_{ij})N_0(k_F^{(n)} R_{ij}) + J_1(k_F^{(n)} R_{ij})N_1(k_F^{(n)} R_{ij})].
\]

where $m^*_t$ is the transversal effective mass, and $k_F^{(n)}$ is the $n$-th subband Fermi wave vector.

The contribution of the inter-subband terms cannot be expressed easily in a closed form. Starting over from Eq. [4] we arrive to:

\[
J_{ij}^{(n,n')} = \left(\frac{I}{2}\right)^2 \frac{1}{\pi} \text{Re} \left[ \phi_{n'}^*(z_i)\phi_n(z_i)\phi_{n'}(z_j)\phi_n(z_j) \right] \int_0^\infty dqq F_{n,n'}(q)J_0(qR_{ij}), \tag{5}
\]
are adjusted in such a way that the total number $N$ monolayers (ML) in the barrier. Periodic boundary conditions are imposed in the $(L_x, L_y, L_z)$ ions, with a concentration $x, y, z$. The calculation is performed in a finite box whose axes are parallel to [100] directions, of dimensions $L_x = L_y$, and $L_z = N \alpha/2$, where $\alpha$ is the lattice parameter of GaAs, and $N$ the number of DMS monolayers (ML) in the barrier. Periodic boundary conditions are imposed in the $(x, y)$ plane. Lateral dimensions are adjusted in such a way that the total number $N_c$ of spins is about 4400, for all $L_z$. Their initial orientations are randomly assigned.

We present results for the eight samples described in Table 1. $L$ was chosen typically 6 nm. The DMS barrier width was varied from 4 ML to 18 ML and the density fraction $r$ was taken equal to 0.1 and 0.25. In Fig. 1 the normalized magnetization $< M >$ is plotted versus temperature. Samples # 3, # 5 and # 8 show a ferromagnetic order at $T_c \approx 35, 55$ and 15 K, respectively. The complete saturation at $T = 0$ K is not achieved, presumably due to boundary effects in the $z$-direction. It is striking that the magnetization curves in those samples are far from the canonical Brillouin function, as already experimentally remarked in Ref. [1], being instead very similar to the ones obtained by these authors. On the other hand, samples # 2 and # 7 are still paramagnetic at $T \approx 1$ K. The Edwards-Anderson order parameter $q$ given by Eq. (8) is shown in Fig. 2 as a function of temperature. The ferromagnetic samples have nearly the same $q(T)$ as $< M > (T)$, what is not surprising. However, samples # 1, # 4 and # 6 show large $q$-values at low temperature, indicating a spin-glass like magnetic order. In particular, samples # 4 and # 6 show a non negligible magnetization at low temperatures. This points to an occurrence of a canted spin phase.

The results seem to indicate that two relevant parameters compete in establishing the magnetic order in the DMS layer. On one hand, a ferromagnetic order is expected to settle as the layer width is increased, in accordance with what observed in Ref. [10]. On the other hand, depending on the hole density, and because of the oscillatory nature of RKKY interaction, in addition to the ferromagnetic couplings, antiferromagnetic couplings can be switched on. This is the ingredient which together with disorder are the origin of the spin-glass phase. This is illustrated in Fig. 3 where the RKKY interactions $J_{ij}$ are plotted versus the in-plane distance $R_{ij}$ for samples # 5, # 6 and # 8. One sees that for ferromagnetic samples (# 5 and # 8) $J_{ij}$ is essentially positive, while for sample # 6, a non negligible negative part is present, especially when considering the occurrence of pair couplings with large $R_{ij}$’s. Therefore it is not surprising that a (canted) spin-glass phase sets in.

In conclusion, we have shown that depending on the DMS barrier width of a double QW structure and the hole concentration, different magnetic phases can be obtained. For applications, favorable spin configurations can thus be designed. In particular, the possibility of having a ferromagnetic order in the DMS barrier of a double QW structure is important for spin tunneling and resonant spin tunneling in nanostructures [17].

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TABLE I. Samples characteristics: The width of each one of the two GaAs well in the structure is $L = 6\, nm$; $N$ is the number of DMS barrier monolayers (ML), $r$ is the ratio of hole density to Mn density; $T_c(K)$ is the magnetic transition temperature for F: ferromagnetic, P: paramagnetic, SG: spin-glass phases. The sign ($*$) indicates a possible canted phase. The calculation is performed with the value of $N_0\beta = -1.2\, eV$ according to ref. [13].

| Sample | $N(ML)$ | $r$  | Phase | $T_c(K)$ |
|--------|---------|------|-------|----------|
| # 1    | 4       | 0.1  | SG    | 2        |
| # 2    | 12      | 0.1  | P     | $\leq 1$ |
| # 3    | 18      | 0.1  | F     | 35       |
| # 4    | 4       | 0.25 | SG*   | 15       |
| # 5    | 18      | 0.25 | F     | 55       |
| # 6    | 12      | 0.25 | SG*   | 30       |
| # 7    | 9       | 0.1  | P     | $\leq 1$ |
| # 8    | 9       | 0.25 | F     | 15       |
FIG. 1. Normalized magnetization vs temperature for samples indicated in Table 1.

FIG. 2. Edwards-Anderson order parameter $q$ vs temperature for samples indicated in Table 1.

FIG. 3. RKKY exchange interaction $J_{ij}$ vs $R_{ij}$ for samples # 5, # 6 and # 8.
Ga$_{0.65}$Al$_{0.35}$As:Mn (5%)
Double QW L=60 ang.
Ga$_{0.65}$Al$_{0.35}$As:Mn (5%)

Double QW L=60 ang.
$R_{ij}$ (monolayers)

$L=60$ (ang.) $x=0.05$ $r=0.25x$

$N=18$ ML

$N=12$ ML

$N=9$ ML