Mean-field theory of magnetic properties of Mn$_x$III$_{1-x}$V semiconductors

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Abstract

We present a mean-field theory of carrier-induced ferromagnetism in Mn$_x$III$_{1-x}$V diluted magnetic semiconductors with a special emphasis placed on the magnetic anisotropy. The valence band holes are described using the six band Kohn-Luttinger model. We find that the magnetic anisotropy is a complicated function of sample parameters such as hole density or strain. Results of our numerical simulations are in agreement with magnetic anisotropy measurements on samples with both compressive and tensile strains.

Keywords: Diluted Magnetic Semiconductors, Ferromagnetism, Magnetic anisotropy

1. Introduction

Experiments [1,2] in Mn$_x$III$_{1-x}$V diluted magnetic semiconductors (DMS) have demonstrated that these ferromagnets have remarkably square hysteresis loops with coercivities typically $\sim 400$ Oe, and that the magnetic easy axis is dependent on epitaxial growth lattice-matching strains. The physical origin of the anisotropy energy in our model is spin-orbit coupling in the valence band. Our work is based on six band envelope function description of the valence band holes and a mean-field treatment of their exchange interactions with Mn$^{++}$ ions. Even in the mean-field theory, we find that the magnetic anisotropy physics of these materials is rich and we predict easy axis reorientations as a function of sample parameters including hole density or epitaxial growth lattice-matching strains. Similar conclusions have been presented in a closely related, independent study [3]. A formal theory of magnetic anisotropy in cubic semiconductor DMS is derived in Section 2. Numerical results for typical experimental samples are presented in Section 3.

2. Formal theory

In zero external fields the Hamiltonian for the valence band electrons interacting with localized $d$ electrons on the Mn$^{++}$ ions can be written as [4]
\[
H = H_L + J_{pd} \sum_{i,l} \mathbf{s}_I \cdot \mathbf{s}_i \delta(\mathbf{r}_i - \mathbf{R}_I),
\]

where \(i\) labels a valence band hole and \(I\) labels a magnetic ion, \(\mathbf{s}_I\) is a localized spin, \(\mathbf{s}_i\) is a hole spin, and \(H_L\) is the six-band envelope-function Hamiltonian [5] for the valence bands. In \(III_{1-x}Mn_xV\) semiconductors, the four \(j = 3/2\) bands are separated by a spin-orbit splitting \(\Delta_{so}\) from the two \(j = 1/2\) bands. In the relevant range of hole and \(Mn^{++}\) densities, no more than four bands are ever occupied. Nevertheless, mixing between \(j = 3/2\) and \(j = 1/2\) bands does occur, and it can alter the balance of delicate cancellations which often controls the net anisotropy energy. The exchange interaction of the localized moments:

\[
J = \frac{1}{2} \left( \frac{N}{V} \right) \left( \frac{\langle b^\dagger b \rangle}{\langle b^\dagger b \rangle + \langle b b^\dagger \rangle} \right)
\]

The MBE growth techniques produce \(III_{1-x}Mn_xV\) films whose lattices are locked to those of their substrates. X-ray diffraction studies [11] have established that the resulting strains are not relaxed by dislocations or other defects, even for thick films. Strains in the \(III_{1-x}Mn_xV\) film break the cubic symmetry assumed in Eq. (3). However, the influence of MBE growth lattice-matching strains on the hole bands of cubic semiconductors is well understood [12] and we can use the same formal mean-field theory to account for strain effects on magnetic anisotropy.

3. Numerical results

We turn now to a series of illustrative calculations intended to closely model the ground state of \(Ga_{0.95}Mn_{0.05}As\). For this Mn density and the smaller values of \(J_{pd}\) favored by recent estimates, \(h = J_{pd}N_{Mn}J \sim 0.01Ry\) at zero temperature. This value of \(h\) is not so much smaller than the spin-orbit splitting parameter in GaAs \((\Delta_{so} = 0.025\ \text{Ry})\), so that accurate calculations require a six band model. Even with \(x\) fixed, our calculations show that the magnetic anisotropy of \(Ga_{0.95}Mn_{0.05}As\) ferromagnets is strongly dependent on both hole density and strain. The hole density can be varied by changing growth conditions or by adding other dopants to the material, and strain in a \(Ga_{0.95}Mn_{0.05}As\) film can be altered by changing substrates. The cubic anisotropy coefficients (in units of energy per volume) for strain-free material are plotted as a function of hole density in Fig. 1. The easy axis is nearly always determined by the leading cubic anisotropy coefficient \(K_1^{ca}\), except near values of \(p\) where this coefficient vanishes. As a consequence the easy-axis in strain free samples is almost always either along one of the cube edge directions \((K_1^{ca} > 0)\), or along one of the cube diagonal directions \((K_1^{ca} < 0)\). Transitions in which the easy axis moves between these two directions occur twice over the range of hole densities studied. (Similar transitions occur as a function of \(h\), and therefore temperature, for fixed hole density.)
Near the hole density 0.01 nm$^{-3}$, both anisotropy coefficients vanish and a fine-tuned isotropy is achieved. The slopes of the anisotropy coefficient curves vary as the number of occupied bands increases from 1 to 4 with increasing hole density. This behavior is clearly seen from the correlation between oscillations of the anisotropy coefficients and onsets of higher band occupations.

Six-band model Fermi surfaces are illustrated in Figs. 2 and 3 by plotting their intersections with the $k_z = 0$ plane at $p = 0.1$nm$^{-3}$ for the cases of $\langle 100 \rangle$ and $\langle 110 \rangle$ ordered moment orientations. The dependence of quasiparticle band structure on ordered moment orientation, apparent in comparing these figures, should lead to large anisotropic magnetoresistance effects in III$_{1-x}$Mn$_x$V ferromagnets. We also note that in the case of cube edge orientations, the Fermi surfaces of different bands intersect. This property could have important implications for the decay of long-wavelength collective modes.

In Fig. 4 we present mean-field theory predictions for the strain-dependence of the anisotropy energy at $h = 0.01$Ry and hole density $p = 0.35$nm$^{-3}$. According to our calculations, the easy axes in the absence of strain are along the cube edges in this case. This calculation is thus for a hole density approximately three times smaller than the Mn density, as indicated by recent exper-
strains, the sign of the anisotropy changes emphasizing the subtlety of these effects and the latitude which exists for strain-engineering of magnetic properties.

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Fig. 4. Energy differences among ⟨001⟩, ⟨100⟩, ⟨110⟩, and ⟨111⟩ magnetization orientations vs. in-plane strain $e_0$ at $h = 0.01$ Ry and $p = 3.5$ nm$^{-3}$. For compressive strains ($e_0 < 0$) the system has an easy magnetic plane perpendicular to the growth direction. For tensile strains ($e_0 > 0$) the anisotropy is easy-axis with the preferred magnetization orientation along the growth direction. The anisotropy changes sign at large tensile strain.

The relevant value of the in-plane strain produced by the substrate-film lattice mismatch,

$$e_0 = \frac{a_s - a_f}{a_f},$$

depends on the substrate on which the epitaxial Ga$_{0.95}$Mn$_{0.05}$As film is grown. The most important conclusion from Fig. 4 is that strains as small as 1% are sufficient to completely alter the magnetic anisotropy energy landscape. For example for (Ga,Mn)As on GaAs, $e_0 = -0.0028$ at $x = 0.05$, the anisotropy has a relatively strong uniaxial contribution even for this relatively modest compressive strain, which favors in-plane moment orientations, in agreement with experiment. A relatively small (~1 kJ m$^{-3}$) residual plane-anisotropy remains which favors (110) over (100). For $x = 0.05$ (Ga,Mn)As on a $x = 0.15$ (In,Ga)As buffer the strain is tensile, $e_0 = 0.0077$, and we predict a substantial uniaxial contribution to the anisotropy energy which favors growth direction orientations, again in agreement with experiment. For the tensile case, the anisotropy energy changes more dramatically than for compressive strains due to the depopulation of higher subbands. At large tensile

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