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Limits on the Curie temperature of (III,Mn)V ferromagnetic semiconductors

John Schliemann[a] and Jürgen König
Department of Physics, Indiana University, Bloomington, Indiana 47405 and
Department of Physics, The University of Texas, Austin, Texas 78712

Hsiu-Hau Lin
Department of Physics, National Tsing-Hua University, Hsinchu 300, Taiwan, Republic of China

Allan H. MacDonald
Department of Physics, Indiana University, Bloomington, Indiana 47405 and
Department of Physics, The University of Texas, Austin, Texas 78712

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Mean-field-theory predicts that the Curie temperature T_c of a (III,Mn)V ferromagnet will be proportional to the valence band density-of-states of its host (III,V) semiconductor, suggesting a route toward room-temperature ferromagnetism in this materials class. In this letter, we use theoretical estimates of spin-wave energies and Monte Carlo simulations to demonstrate that long-wavelength collective fluctuations, neglected by mean-field theory, will limit the critical temperature in large density-of-states materials. We discuss implications for high T_c searches. [DOI: 10.1063/1.1355300]

The recent discovery[1] of ferromagnetism at relatively high temperatures ($T_c > 100$ K) in III–V compound semiconductors containing Mn has generated intense interest, mainly because of the technological roadways that would be opened by room temperature ferromagnetism in semiconductors with favorable materials properties. The search for systems in this materials class with higher critical temperatures is an important current activity that has been guided thus far by mean-field theoretical[2,3] considerations. In this letter we address the importance of collective magnetization fluctuations, neglected by mean-field theory, in limiting the critical temperature and discuss the implications of these considerations for high-T_c searches.

Our analysis is based on the kinetic-exchange model for interactions between the Mn spins and band electrons

$$H = H_0 + J_{\mathrm{pd}} \sum_I \int d^3r \mathbf{S_I} \cdot \mathbf{s}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{R}_I), \tag{1}$$

where $\mathbf{S}_I$ describes a Mn spin with spin length $S = 5/2$ at site $\mathbf{R}_I$, $\mathbf{s(r)}$ is the band-carrier density, and $J_{\mathrm{pd}} > 0$ represents the exchange integral. H_0 represents a simplified single-parabolic-band model for the host semiconductor valence bands.

The simplest treatment of this model is a mean field theory which takes the magnetizations of carriers and ion spins to be uniform in space and neglects correlations between them. A straightforward calculation yields for the critical temperature in mean field approximation[2,3]

$$T_c^{\mathrm{MF}} = \frac{\chi_P}{(g^* \mu_B/2)^2} \frac{S(S+1) N J_{\mathrm{pd}}^2}{12}, \tag{2}$$

where g^* is the g factor of the carriers and χ_P is their Pauli susceptibility, which is proportional to the effective band mass. This observation has given rise to concrete predictions for critical temperatures for several host semiconductors based on their different band masses.[2]

In mean-field theory, ferromagnetism occurs because the penalty in entropic free energy paid to polarize the Mn spins vanishes at $T = 0$. Any coupling to a band-electron system with a finite magnetic susceptibility is sufficient to yield ferromagnetism. While this mean-field theory probably captures much of the physics of (III,Mn)V ferromagnetism, it has a qualitative deficiency which will have an important quantitative impact on T_c predictions in circumstances we identify later. Mean-field theory fails to account for the small energy cost of magnetization configurations in which spin orientations vary slowly[4] in space, reducing the average magnetization but maintaining local correlations between Mn and band-electrons spin orientations. In the following paragraphs we estimate the critical temperature for the case when these collective excitations dominate thermal magnetization suppression.

Isotropic ferromagnets[5] have spin-wave Goldstone collective modes whose energies vanish at long wavelengths

$$E(k) = Dk^2 + \cdots, \tag{3}$$

where k is the wave vector of the mode. Each spin-wave excitation reduces the total spin of the ferromagnetic state by 1. The coefficient D is inversely proportional to the saturation magnetization and proportional to the exchange constant A of classical micromagnetic theory, that parameterizes the free-energy cost of spatial variations in magnetization orientation. We have previously presented a theory of spin-wave excitations in (III,Mn)V ferromagnets.[6] These collective excitations are not accounted for in the mean-field approximation. If the spin stiffness is small, they will dominate the suppression of the magnetization at all finite temperatures and limit the critical temperature. A rough upper bound[7] on the resulting critical temperature, T_c^{coll}, can be found by using the $T = 0$ stiffness value and finding the temperature

[a]Electronic mail: joschlie@physics.utexas.edu

where the number of excited spin waves per unit volume equals the spin per volume of the ground state

$$SN=\frac{1}{2\pi^2}\int_0^{k_D} dk\, k^2 n[E(k)]. \tag{4}$$

A Debye wave vector[4] $k_D=(6\pi^2 N)^{1/3}$ cuts off the sum over wave vectors at the correct number of modes, and $n(E)$ is the Bose occupation number. We find that

$$k_B T_c^{\mathrm{coll}}=\frac{2S+1}{6} D k_D^2 \tag{5}$$

for $S\geqslant 5/2$. To obtain this equation, we have assumed that the spin waves can be approximated as noninteracting Bose particles, replaced the dispersion by the long-wavelength limit Eq. (3), and noted that the critical temperature estimate is proportional to Dk_D^2, justifying the use of the classical expression for the mode occupation number $n(k)\approx k_B T/E(k)-1/2$. These considerations set an upper bound on the critical temperature which is proportional to the spin stiffness, a bound not respected by the mean-field theory. A familar example of ferromagents in which long range order is suppressed by long-wavelength collective excitations is provided by the ferromagnetic transition metals Fe, Co, and Ni. In that case, an expression similar to Eq. (5) and proportional to the micromagnetic exchange constant, predicts critical temperatures with 20% accuracy[8,9] whereas mean-field-theory overestimates T_c by factors of 5–10. In (III,Mn)V ferromagnets, we will see that both mean-field and collective regimes can occur depending on carrier density and host semiconductor band parameters.

Our theoretical results for (III,Mn)V ferromagnet collective modes lead to physically transparent results for the spin stiffness in both strong and weak exchange coupling limits. The dimensionless parameter which characterizes the strength of the exchange coupling is the ratio Δ/ϵ_F, where ϵ_F is the band-system Fermi energy and $\Delta=J_{\mathrm{pd}}NS$ is its mean-field spin splitting at $T=0$. For small Δ/ϵ_F, the RKKY regime,[2] exchange coupling is a weak perturbation on the band system. In this regime we find that $D=\delta/(12k_F^2)$ where $\delta=J_{\mathrm{pd}}n\xi/2=(3/8S)(n/N)(\Delta^2/\epsilon_F)$ is the energy cost of an uncorrelated spin reduction at a single Mn site. Note that in this regime $\delta\sim T_c^{\mathrm{MF}}$ and that

$$T_c^{\mathrm{coll,RKKY}}=T_c^{\mathrm{MF}}\frac{2S+1}{12(S+1)\sqrt[3]{2}}\left(\frac{N}{n}\right)^{2/3}. \tag{6}$$

In the weak coupling regime mean-field theory is reliable only for $n/N\ll 1$, as expected since in this case the RKKY interaction has a range which is long compared to the distance between Mn spins. In the large Δ/ϵ_F regime exchange coupling completely polarizes the band-electron system. In this case (and for $n\ll 2NS$) we find that $D=(n/2NS)\times(\epsilon_F/k_F^2)$. For a fully polarized band the energetic cost of varying the moment orientation direction is entirely due to band kinetic energy. We, thus, obtain as a third T_c bound

$$T_c^{\mathrm{coll},s}=\frac{2S+1}{12S}\epsilon_F\left(\frac{n}{N}\right)^{1/3}. \tag{7}$$

The different regimes deduced from these considerations are illustrated in Fig. 1.

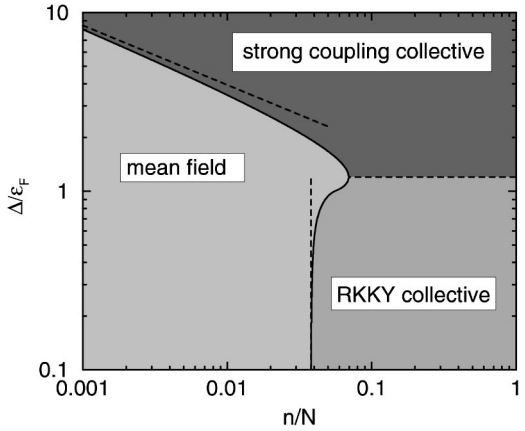

FIG. 1. Critical-temperature-limit regimes. In the mean-field regime T_c is limited by individual Mn spin fluctuations. In the collective regimes, the critical temperature is limited by long-wavelength fluctuations with a stiffness proportional to the bandwidth for weak (RKKY) exchange coupling and inversely proportional to the bandwidth for strong exchange coupling. At the solid line $T_c^{\mathrm{MF}}=T_c^{\mathrm{coll}}$. Dashed lines: expansions for large and small Δ/ϵ_F, Eqs. (6) and (7), and the crossover from the RKKY to the strong coupling collective regime.

To substantiate these qualitative considerations, we have performed hybrid-Monte-Carlo[10] simulations, treating the Mn spins as discrete classical degrees of freedom, an approximation that is justified near the critical temperature. We allow for disorder by choosing the Mn positions randomly. Microscopic $p-d$ exchange physics is modeled by allowing the interaction to have a finite range a_0.[11] We simulate this by replacing the delta function by a Gaussian distribution in Eq. (1).

An exhaustive description of our Monte Carlo approach, including a detailed account of all technical aspects such as thermalization procedures, finite-size effects, etc., will be given elsewhere.[8] Here we shall, for brevity, concentrate on the results. In the following we consider the strong coupling regime where mean-field theory is not reliable and finite-size effects in our simulations are small. One important finding is that randomness in the Mn positions can *enhance* the spin stiffness (for large J_{pd} by up to factor of two) in comparison

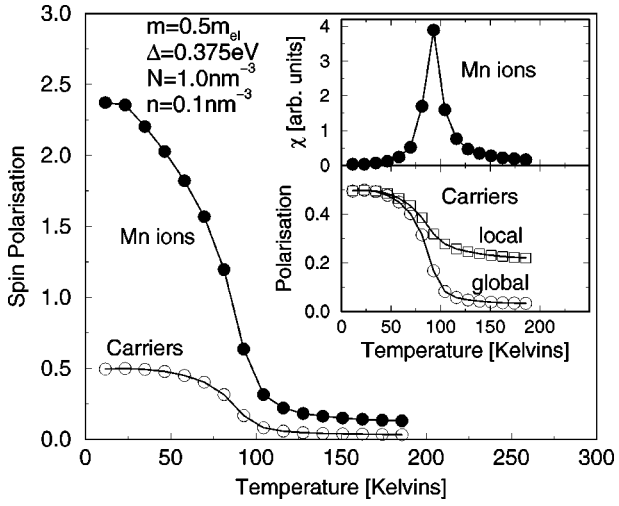

FIG. 2. Magnetization curves for Mn ions and carriers. The upper inset shows the magnetic susceptibility due to the ions, while in the lower inset the local and the global spin polarization per carrier is plotted. The data were obtained for a system of 540 Mn spins and 54 carriers.

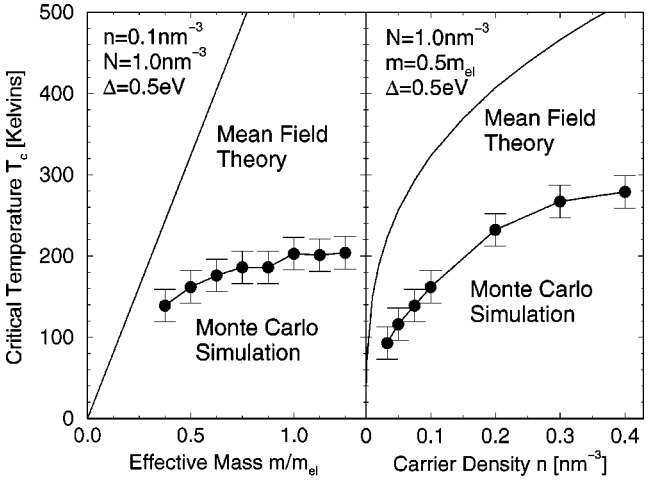

FIG. 3. The critical temperature T_c as a function of the carrier mass (left panel) and as a function of the carrier density (right panel). The results of the Monte Carlo runs are compared with the mean-field predictions.

to the estimates[6] based on course-grained Mn spins with Debye cutoffs discussed above. The importance of disorder was also emphasized by Wan and Bhatt[12] for a model of interacting magnetic polarons where both the ion and the carrier spins are treated classically, while we account for the quantum-mechanical nature of the free carriers in our simulation.

Figure 2 shows a typical magnetization curve for manganese ions and carriers magnetizations in the collective regime. The maximum of the finite-size Mn susceptibility marks the ferromagnetic transition temperature. The lower inset compares the *global* and *local* free carrier polarizations, $m_{\mathrm{loc}}=\overline{\langle|\mathbf{s}(\mathbf{r})|/n(\mathbf{r})\rangle}$. As anticipated by our qualitative discussion, the typical free carrier local band polarization remains large *above* the critical temperature; it would vanish in a mean-field picture. Ferromagnetism, and the technologically useful robust collective physics it gives rise to, disappears in this case only because of the loss of long-range spatial coherence.

The left panel of Fig. 3 shows the critical temperature as a function of the carrier effective mass. Mean-field theory predicts that T_c will grow linearly with increasing mass. The Monte Carlo results, however, are substantially lower and show a saturation of T_c at carrier masses close to the bare electron mass. For even higher masses, we expect T_c to decrease, reflecting the reduction of spin stiffness expected in this regime.

The right panel of Fig. 3 compares the mean-field prediction and the Monte Carlo results for T_c as a function of the carrier density. For higher carrier densities we expect ferromagnetism to give way to spin-glass order.

We conclude from the present work that high critical temperatures cannot be achieved simply by narrowing the free carrier band or placing its Fermi energy at a density-of-states peak in order to enhance its Pauli magnetic susceptibility χ_P. It will also be necessary to engineer a suppression of collective magnetization fluctuations.

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