The recent discovery\(^1\) of ferromagnetism at relatively high temperatures \((T_c > 100 \text{ K})\) 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 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.

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

\[
H = H_0 + J_{\text{pd}} \sum \int d^3 r S_i^z(r) \delta(r-R_i),
\]

where \(S_i^z\) describes a Mn spin with spin length \(S=5/2\) at site \(R_i\), \(\delta(r-R_i)\) is the band-carrier density, and \(J_{\text{pd}}>0\) 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\(^2,3\)

\[
T_c^{\text{MF}} = \frac{\chi_F}{(g^* \mu_B/2)^2} \frac{S(S+1)NJ_{\text{pd}}^2}{12},
\]

where \(g^*\) is the \(g\) factor of the carriers and \(\chi_F\) 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.\(^5\)

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

\[
E(k) = Dk^2 + \cdots,
\]

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, \(T_c^{\text{coll}}\), can be found by using the \(T=0\) stiffness value and finding the temperature

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

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

(4)

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

\[
k_B T_c^{\text{coll}} = \frac{2S + 1}{6} \frac{Dk_D^2}{n}\]

(5)

for \(S \geq 5/2\). 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 is proportional to \(Dk_D^2\), justifying the use of the classical expression for the mode occupation number \(n(k) = k_B T / E(k) - 1/2\). 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 familiar example of ferromagnets 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 whereas mean-field theory overestimates \(T_c\) by factors of 5–10. In \((\text{III},\text{Mn})V\) 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 \((\text{III},\text{Mn})V\) 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 \(\Delta / \epsilon_F\), where \(\epsilon_F\) is the band-system Fermi energy and \(\Delta = J_{pd}NS\) is its mean-field value at \(T=0\). For small \(\Delta / \epsilon_F\), the RKKY regime,\(^2\) exchange coupling is a weak perturbation on the band system. In this regime we find that \(D = \delta/(12S^2)\), where \(\delta = J_{pd}N\xi^2/(3/8)(n/N)(\Delta^3/\epsilon_F)\) is the cost of an uncorrelated spin reduction at a single Mn site. Note that in this regime \(\delta \approx T_c^{\text{MF}}\) and that

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

(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 \(\Delta / \epsilon_F\) regime exchange coupling completely polarizes the band-electron system. In this case (and for \(n \ll 2NS\)) we find that \(D = (n/2NS) \times (\epsilon_F / k_D^2)\). 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

\[
T_c^{\text{coll},\text{RKKY}} = \frac{2S + 1}{12S} \epsilon_F \left(\frac{n}{N}\right)^{1/3}.
\]

(7)

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

To substantiate these qualitative considerations, we have performed hybrid-Monte-Carlo 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...
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 $\chi_p$. It will also be necessary to engineer a suppression of collective magnetization fluctuations.

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4 For these qualitative considerations we replace the discrete Mn spins by a continuous distribution. This is a good approximation when the Mn density is much larger than the free-carrier density.

5 Because of spin-orbit coupling in the valence band the magnetic anisotropy energy is nonzero in (III,Mn)V ferromagnets and there is a resulting gap in the spin-wave excitation spectrum. [J. König, T. Jungwirth, and A. H. MacDonald, Phys. Rev. B (submitted)]. However, this gap is quite small and does not change the essence of the present discussion.


7 At finite temperature interactions between spin waves will reduce the spin stiffness. To model this effect a self-consistent spin-wave scheme would be required. Here, however, we use the zero-temperature spin stiffness for an upper bound.

8 J. Schliemann, J. König, and A. H. MacDonald, cond-mat/0012233.


11 A. K. Bhattacharjee and C. Benoît à la Guillaume, Solid State Commun. 113, 17 (2000). In the present Monte Carlo simulations we use $a_0 = 0.1$ nm.

12 X. Wan and R. N. Bhatt, cond-mat/0009161.