Theory of Diluted Magnetic Semiconductor Ferromagnetism

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(Received 24 January 2000)

We present a theory of carrier-induced ferromagnetism in diluted magnetic semiconductors ($III_{1-x}Mn_xV$) which allows for arbitrary itinerant-carrier spin polarization and dynamic correlations. Both ingredients are essential in identifying the system's elementary excitations and describing their properties. We find a branch of collective modes, in addition to the spin waves and Stoner continuum which occur in metallic ferromagnets, and predict that the low-temperature spin stiffness is independent of the strength of the exchange coupling between magnetic ions and itinerant carriers. We discuss the temperature dependence of the magnetization and the heat capacity.

PACS numbers: 75.30.Ds, 75.40.Gb, 75.50.Dd

Introduction. — The recent discovery of carrier-induced ferromagnetism [1,2] in diluted magnetic semiconductors (DMS) has generated intense interest, in part because it opens the prospect of developing devices which combine information processing and storage functionalities in one material [3–9]. Critical temperatures T_c exceeding 100 K have been realized [3] by using low-temperature molecular beam epitaxy growth to introduce a high concentration c of randomly distributed Mn^{2+} ions in GaAs systems with a high hole density c^*. The tendency toward ferromagnetism and trends in the observed T_c's have been explained using a picture [10–14] in which uniform itinerant-carrier spin polarization mediates a long-range ferromagnetic interaction between the Mn^{2+} ions with spin $S = 5/2$.

We present here the first theory which accounts for dynamic correlations in the ordered state and is able to describe its fundamental properties. We use a path-integral formulation and, in the RKKY spirit of the previous theory, integrate out the itinerant carriers, and expand the effective action for the impurity spins up to quadratic order. In addition to the usual Goldstone spin waves, we find itinerant-carrier dominated collective excitations analogous to the optical spin waves in a ferrimagnet. We also find that the spin stiffness is inversely proportional to the itinerant-carrier mass and independent of the strength of the exchange coupling between magnetic ions and itinerant carriers, in contrast to the mean-field critical temperature which is proportional to the itinerant-carrier mass and to the square of the exchange coupling. We extend our calculations to the critical temperature using an approximate self-consistent spin-wave scheme and address the temperature-dependent magnetization and specific heat.

Hamiltonian and effective action. — We study a model which provides an accurate [15] description of Mn based zinc blende DMS's. Magnetic ions with $S = 5/2$ at positions $\vec{R}_I$ are antiferromagnetically coupled to valence-band carriers described by envelope functions:

$$H = H_0 + J_{pd} \int d^3 r \, \vec{S}(\vec{r}) \cdot \vec{s}(\vec{r}), \qquad (1)$$

where $\vec{S}(\vec{r}) = \sum_I \vec{S}_I \delta(\vec{r} - \vec{R}_I)$ is the impurity-spin density. The itinerant-carrier spin density is expressed in terms of carrier field operators by $\vec{s}(\vec{r}) = \frac{1}{2} \sum_{\sigma\sigma'} \Psi_\sigma^\dagger(\vec{r}) \vec{\tau}_{\sigma\sigma'} \Psi_{\sigma'}(\vec{r})$. ($\vec{\tau}$ is the vector of Pauli spin matrices.) H_0 includes the valence-band envelope-function Hamiltonian [16] and, if an external magnetic field $\vec{B}$ is present, the Zeeman energy,

$$H_0 = \int d^3 r \left\{ \sum_\sigma \hat{\Psi}_\sigma^\dagger(\vec{r}) \left(-\frac{\hbar^2 \vec{\nabla}^2}{2m^*} - \mu^* \right) \hat{\Psi}_\sigma(\vec{r}) \right.$$
$$\left. - g\mu_B \vec{B} \cdot \vec{S}(\vec{r}) - g^* \mu_B \vec{B} \cdot \vec{s}(\vec{r}) \right\}. \quad (2)$$

The effective mass, chemical potential, and g factor of the itinerant carriers are labeled by m^*, μ^*, and g^*. The model we use here is, thus, related to colossal magnetoresistance materials [17] and identical to those for dense Kondo systems, which simplify when the itinerant-carrier density c^* is much smaller than the magnetic ion density c [18]. The fact that $c^*/c \ll 1$ in ferromagnetic-semiconductor materials is essential to their ferromagnetism. Similar models have been used for ferromagnetism induced by magnetic ions in nearly ferromagnetic metals such as palladium [19].

We represent the impurity spins in terms of Holstein-Primakoff (HP) bosons [20]. By coarse graining, the spin density $\vec{S}(\vec{r})$ can be replaced by a smooth function,

$$S^+(\vec{r}) = \left(\sqrt{2cS - b^\dagger(\vec{r})b(\vec{r})} \right) b(\vec{r}), \qquad (3)$$

$$S^-(\vec{r}) = b^\dagger(\vec{r}) \sqrt{2cS - b^\dagger(\vec{r})b(\vec{r})}, \qquad (4)$$

$$S^z(\vec{r}) = cS - b^\dagger(\vec{r})b(\vec{r}), \qquad (5)$$

with bosonic fields $b^\dagger(\vec{r})$, $b(\vec{r})$. The partition function as a coherent-state path integral in imaginary times reads

$$Z = \int \mathcal{D}[\bar{z}z] \mathcal{D}[\bar{\Psi}\Psi] e^{-\int_0^\beta d\tau \, L(\bar{z}z, \bar{\Psi}\Psi)}, \qquad (6)$$

with $L = \int d^3 r [\bar{z} \partial_\tau z + \sum_\sigma \bar{\Psi}_\sigma \partial_\tau \Psi_\sigma] + H(\bar{z}z, \bar{\Psi}\Psi)$. The bosonic (impurity spins) and fermionic (itinerant carriers) degrees of freedom are labeled by the complex variables $\bar{z}, z$ and the Grassmann numbers $\bar{\Psi}, \Psi$, respectively.

Since the Hamiltonian is bilinear in fermionic fields, we can integrate out the itinerant carriers and arrive at an effective description in terms of the localized spin density only, $Z = \int \mathcal{D}[\bar{z}z] \exp(-S_{\text{eff}}[\bar{z}z])$ with the action

$$S_{\text{eff}}[\bar{z}z] = \int_0^\beta d\tau \int d^3 r [\bar{z}\partial_\tau z - g\mu_B B(cS - \bar{z}z)]$$
$$- \ln \det[(G^{\text{MF}})^{-1} + \delta G^{-1}(\bar{z}z)]. \quad (7)$$

Here, we have already split the total kernel G^{-1} into a mean-field part $(G^{\text{MF}})^{-1}$ and a fluctuating part δG^{-1},

$$(G^{\text{MF}})^{-1} = \left(\partial_\tau - \frac{\hbar^2 \vec{\nabla}^2}{2m^*} - \mu^*\right)\mathbf{1} + \frac{\Delta}{2}\tau^z, \quad (8)$$

$$\delta G^{-1} = \frac{J_{pd}}{2}[(z\tau^- + \bar{z}\tau^+)\sqrt{2cS - \bar{z}z} - \bar{z}z\tau^z], \quad (9)$$

where $\Delta = cJ_{pd}S - g^*\mu_B B$ is the zero-temperature spin-splitting gap for the itinerant carriers. The physics of the itinerant carriers is embedded in the effective action of the magnetic ions. It is responsible for the retarded and nonlocal character of the interactions between magnetic ions, which is described here for the first time.

Independent spin-wave theory.—The independent spin-wave theory, which is a good approximation at low temperatures, is obtained by expanding Eq. (7) up to quadratic order in z. We find (in Fourier representation)

$$S_{\text{eff}}[\bar{z}z] = \frac{1}{\beta V} \sum_{|\vec{p}|<p_c, m} \bar{z}(\vec{p}, \nu_m) D^{-1}(\vec{p}, \nu_m) z(\vec{p}, \nu_m), \quad (10)$$

in addition to the temperature-dependent mean-field contribution. A Debye cutoff ($p_c^3 = 6\pi^2 c$) ensures that we include the correct number of magnetic ion degrees of freedom. The kernel of the quadratic action is the inverse of the spin-wave propagator,

$$D^{-1}(\vec{p}, \nu_m) = -i\nu_m + g\mu_B B + J_{pd}n^* + \frac{cJ_{pd}^2 S}{2\beta V}$$

$$\times \sum_{n,\vec{k}} G_\uparrow^{\text{MF}}(\vec{k}, \omega_n) G_\downarrow^{\text{MF}}(\vec{k} + \vec{p}, \omega_n + \nu_m), \quad (11)$$

with the mean-field itinerant carrier Green's function $G_\sigma^{\text{MF}}(\vec{k}, \omega_n) = -[i\omega_n - (\epsilon_{\vec{k}} + \sigma\Delta/2 - \mu^*)]^{-1}$. The mean-field spin density is denoted by $n^* = (n_\downarrow - n_\uparrow)/2$, and $\epsilon_{\vec{k}} = \hbar^2 k^2/(2m^*)$.

Excitations.—We obtain the spectral density of the spin-fluctuation propagator by analytical continuation, $i\nu_m \rightarrow \Omega + i0^+$ and $A(\vec{p}, \Omega) = \text{Im}D(\vec{p}, \Omega)/\pi$. We find three different types of spin excitations. In all figures we take $B = 0$ and use as typical parameters [3] $m^* = 0.5m_e$, $J_{pd} = 0.15$ eV nm^3, and $c = 1$ nm^{-3}, where m_e is the free-electron mass. For these parameters the mean-field itinerant-carrier system is fully polarized at $T = 0$.

(i) Our model has a gapless Goldstone-mode branch (see Fig. 1) reflecting the spontaneous breaking of rotational

symmetry [21]. Expansion of the $T = 0$ propagator at $\Delta > \epsilon_F$, where ϵ_F is the Fermi energy of the majority-spin band, yields for small and large momenta the dispersion of the collective modes,

$$\Omega_p^{(1)} = \frac{x}{1-x} \epsilon_p \left(1 - \frac{4\epsilon_F}{5\Delta}\right) + \mathcal{O}(p^4), \quad (12)$$

and $\Omega_p^{(1)} = x\Delta(1 - \Delta/\epsilon_p) + \mathcal{O}(1/p^4)$. At short wavelengths we obtain the mean-field result $x\Delta$, the spin splitting of a magnetic ion in the effective field produced by fully spin-polarized itinerant carriers. Note that the itinerant-carrier and magnetic-ion mean-field spin splittings differ by a factor of $x = c^*/(2cS) \ll 1$. At long wavelengths the magnon dispersion in an isotropic ferromagnet is proportional the spin stiffness ρ divided by the magnetization M. In the adiabatic limit [17,22], $\epsilon_F \ll \Delta$, our long-wavelength result reflects a spin stiffness due entirely to the increase in kinetic energy of a fully spin-polarized band when the orientation has a spatial dependence, $\rho = c^*\hbar^2/(4m^*)$, and a magnetization M which has opposing contributions from magnetic ions and itinerant carriers, $M = cS - c^*/2 = cS(1 - x)$. In this limit, the mean-field critical temperature and the spin stiffness have opposite dependences on the itinerant-carrier mass.

The low-energy mode describes spin waves in the local-impurity system. This contrasts with the case of ferromagnetism induced by local moments [19] where the low-energy mode is primarily in the free-carrier part.

(ii) We find a continuum of Stoner spin-flip particle-hole excitations. They correspond to flipping a single spin in the itinerant-carrier system and, therefore, occur at much larger energies $\sim\Delta$ (see Fig. 2). For $\Delta > \epsilon_F$ and zero temperature, all these excitations carry spin $S^z = +1$, i.e., increase the spin polarization, and therefore turn up at negative frequencies in the boson propagator we study. (When $\Delta < \epsilon_F$, excitations with both $S^z = +1$ and $S^z = -1$ contribute to the spectral function.) This continuum
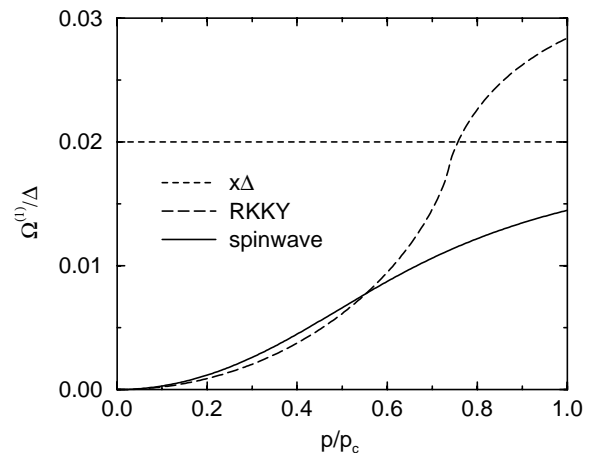

FIG. 1. Spin-wave dispersion for $c^* = 0.1$ nm^{-3}. The short wavelength limit is the mean-field result $x\Delta$. For comparison, we show also the result obtained from an RKKY picture.

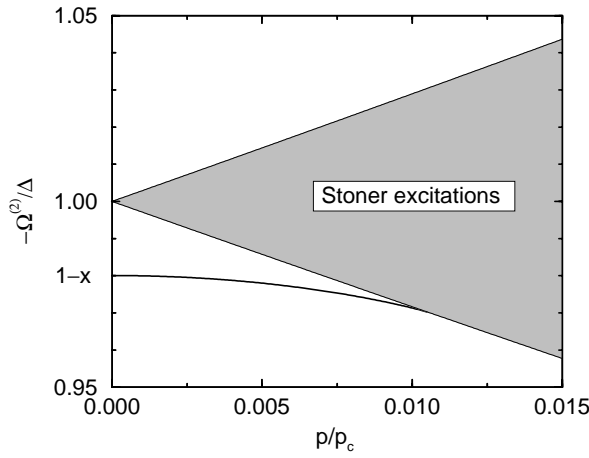FIG. 2. Stoner excitations and collective modes in the free-carrier system for $c^* = 0.1$ nm^{-3}.

lies between the curves $-\Delta - \epsilon_p \pm 2\sqrt{\epsilon_p \epsilon_F}$ and for $\Delta < \epsilon_F$ also between $-\Delta + \epsilon_p \pm 2\sqrt{\epsilon_p(\epsilon_F - \Delta)}$.

(iii) We find additional collective modes associated primarily with the itinerant-carrier system at energies below the Stoner continuum (see Fig. 2). At $T = 0$, we obtain

$$-\Omega_p^{(2)} = \Delta(1 - x) - \frac{1}{1 - x} \epsilon_p \left(\frac{4\epsilon_F}{5x\Delta} - 1 \right) + \mathcal{O}(p^4),$$
(13)

for $\Delta > \epsilon_F$. The spectral weight of these modes is $-x/(1 - x)$ at zero momentum.

The finite spectral weight at negative energies indicates that the ground state is not fully spin polarized because of quantum fluctuations.

Comparison to RKKY picture.—For comparison, we evaluate the $T = 0$ magnon dispersion assuming an RKKY interaction between magnetic ions. This approximation results from our theory if we neglect the spin polarization in the itinerant carriers and evaluate the static limit of the resulting spin-wave propagator. The Stoner excitations and magnons shown in Fig. 2 do not emerge and the spin-wave dispersion (Fig. 1) is incorrect except for the limit $\Delta \ll \epsilon_F$. While the RKKY picture does, in some circumstances, provide a realistic estimate of T_c, it completely fails as a theory of the ferromagnetic state.

Comparison to a ferrimagnet.—Some features of the excitation spectrum are, not coincidentally, similar to those of a localized spin ferrimagnet with antiferromagnetically coupled large spin S (corresponding to the magnetic ions) and small spin s (corresponding to the free carriers) subsystems on a bipartite cubic lattice. Representing the spins by HP bosons and expanding up to quadratic order, we either diagonalize the resulting Hamiltonian directly or, as in our ferromagnetic-semiconductor calculations, integrate out the smaller spins using a path-integral formulation to obtain equivalent results. We find that

$$\Omega_{\vec{p}}^{(1)/(2)} = \frac{\Delta}{2} \left[-(1 - x) \pm \sqrt{(1 - x)^2 + 4x\gamma_{\vec{p}}} \right], \quad (14)$$

with $x = s/S$ and $\Delta = 6JS$, where J is the exchange coupling, and $\gamma_{\vec{p}} = (1/3)\sum_i[1 - \cos(p_i a)]$ with lattice constant a. We recover two collective modes, the coupled spin waves of the two subsystems. One is gapless, the other one gapped with $\Delta(1 - x)$. The bandwidth is $x\Delta$, and the spectral weights at zero momentum are $1/(1 - x)$ and $-x/(1 - x)$, respectively, as in our model.

Self-consistent scheme.—Near the transition temperature T_c, the spin-wave density is of the order of cS, and expanding around a fully polarized state is not a good starting point. Instead, we adopt a scheme which accounts self-consistently for the reduction of spin density in the localized impurity and itinerant carrier subsystems [23]. Our approach is motivated by the Weiss mean-field theory, which is expected to be accurate for a model with static long-range interactions between the magnetic ions. This model can be obtained in our approach by taking the Ising limit (i.e., replacing $\vec{S} \cdot \vec{s}$ by $S^z s^z$) and letting the mean-field itinerant-carrier spin splitting, $\Delta(T) = J_{pd}\langle S^z \rangle$, reflect the thermal suppression of the impurity-spin density $\langle S^z \rangle$. In this simple model the spin-wave spectrum is independent of momentum, $\Omega_p(T) = J_{pd}n^*(T)$, allowing a unitary transformation to spin-wave eigenstates with a site label. The constraint on the number of spin bosons ($\leq 2S$) on a site is then easily applied, and we obtain

$$\langle S^z \rangle = \frac{1}{V} \sum_{|\vec{p}|<p_c} SB_S(\beta S\Omega_p)$$

$$= \frac{1}{V} \sum_{|\vec{p}|<p_c} \{S - n(\Omega_p) + (2S + 1)n[(2S + 1)\Omega_p]\},$$
(15)

where $B_S(x)$ is the Brillouin function, and $n(\omega)$ is the Bose function. The first two terms in the second form of Eq. (15) give the spin density obtained by treating spin waves as bosons, while the last term is the correction from spin kinematics which rules out unphysical states.
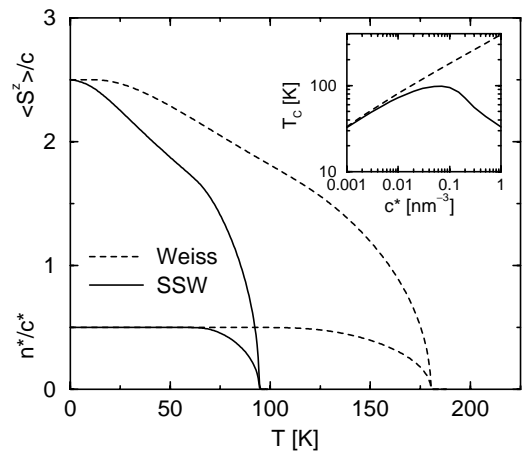

FIG. 3. Average impurity and itinerant-carrier spin as a function of temperature for $c^* = 0.1$ nm^{-3} in Weiss mean-field (Weiss) and self-consistent spin-wave theory (SSW). Inset: T_c as a function of c^*.
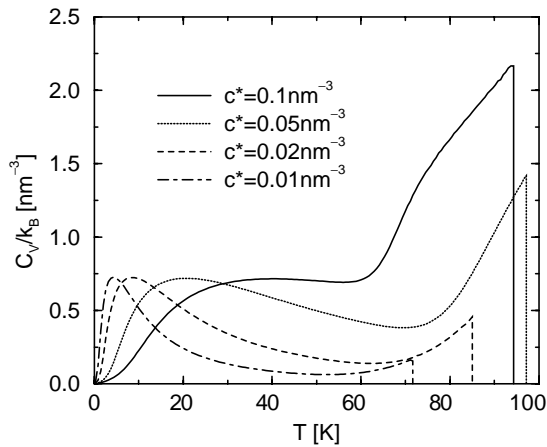
FIG. 4.   Specific heat due to spin waves.

Our self-consistent spin-wave approximation consists of using Eq. (15) when Ω_p is p dependent (see Fig. 3). Although the characteristic $T^{3/2}$ law for the localized-ions magnetization is recovered at low temperatures, the prefactor is reduced by $1 - (2S + 1)^{-1/2}$, compared to the correct [24] value of linearized spin-wave theory. That is, constraining boson populations in momentum space is too restrictive at low temperatures, although the error is not serious for $S = 5/2$. The transition temperature of the self-consistent spin-wave theory is lower than the estimate from Weiss mean-field theory.

The presence of spin waves also shows up in the specific heat C_V, which we calculate within the same scheme (see Fig. 4). Calculating the entropy S, we obtain

$$C_V = \frac{T}{V}\frac{dS}{dT} = \frac{1}{V}\sum_{|\vec{p}|<p_c}\Omega_p(T)\left(\frac{dN_p}{dT}\right), \qquad (16)$$

where $N_p = S - SB_S(\beta S\Omega_p)$ is the average number of spin bosons at each momentum. The specific heat from the kinetic energy of itinerant carriers turns out to be negligibly small. The specific heat of magnetic ions C_V is proportional to $T^{3/2}$ at low temperature, shows a Schottky-like anomaly, and has a jump at the transition temperature. We estimate that the jump is 5% of the lattice specific heat when $T_c \sim 100$ K, suggesting that this should be observable.

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