We present a theory of carrier-induced ferromagnetism in diluted magnetic semiconductors (DMS) which 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 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 dominated collective excitations analogous to the optical spin waves in a ferrimagnet.

**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 $\tilde{R}_i$ are antiferromagnetically coupled to valence-band carriers described by envelope functions:

$$H = H_0 + J_{pd} \int d^3r \tilde{S}(\tilde{r}) \cdot \tilde{\delta}(\tilde{r})$$

(1)

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

$$H_0 = \int d^3r \left( \sum_{\sigma} \Psi_{\sigma}^\dagger(\tilde{r}) \left( -\frac{\hbar^2\nabla^2}{2m^*} - \mu^* \right) \Psi_{\sigma}(\tilde{r}) \right) - g \mu_B B \cdot \tilde{S}(\tilde{r}) - g^* \mu_B B \cdot \tilde{\delta}(\tilde{r}).$$

(2)

The effective mass, chemical potential, and $g$ factor of the itinerant carriers are labeled by $m^*$, $\mu^*$, 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 is much smaller than the magnetic ion density $c$ fact that

$$H^{\text{imp}} = \int d^3r \left( \frac{\hbar^2}{2m_c^*} \nabla^2 \tilde{S}(\tilde{r}) \right)$$

(3)

$$H^{\text{imp}} = \int d^3r \left( \frac{\hbar^2}{2m^*} \nabla^2 \tilde{s}(\tilde{r}) \right)$$

(4)

$$H^{\text{imp}} = \int d^3r \left( \frac{\hbar^2}{2m^*} \nabla^2 \tilde{\delta}(\tilde{r}) \right)$$

(5)

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

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

(6)

with $L = \int d^3r \left( \dot{\xi} \delta_{\tau z} + \sum_{\sigma} \dot{\Psi}_{\sigma} \partial_{\tau} \Psi_{\sigma} \right) + H(\xi \bar{z}, \bar{\Psi} \Psi)$. The bosonic (impurity spins) and fermionic (itinerant carriers) degrees of freedom are labeled by the complex variables $\xi, \bar{z}$ 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, \( Z = \int D[\bar{z} z] \exp(-S_{\text{eff}}[\bar{z} z]) \) with the action
\[
S_{\text{eff}}[\bar{z} z] = \int_0^\beta d\tau \int d^3 r [\bar{z} \partial_\tau z - g \mu_B B(cS - \bar{z} z)] - \ln \det[(G^{\text{MF}})^{-1} + \delta G^{-1}(\bar{z} z)].
\]
Here, we have already split the total kernel \( G^{-1} \) into a mean-field part \((G^{\text{MF}})^{-1}\) and a fluctuating part \(\delta G^{-1}\),
\[
(G^{\text{MF}})^{-1} = \left( \frac{\hat{\delta} - \hbar^2 \hat{\nabla}^2}{2m^*} - \mu^* \right) \mathbf{1} + \frac{\Delta}{2} \tau^z,
\]
\[
\delta G^{-1} = \frac{J_{pd}}{2} \left[ (z \tau^- + \bar{z} \tau^+) \sqrt{2cS - \bar{z} z} - \bar{z} z \right] \tau^z,
\]
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)
\[
S_{\text{eff}}[\bar{z} z] = \frac{1}{\beta V} \sum_{|\vec{p}| < p_m, \vec{m}} \bar{z}(\vec{p}, \vec{m}) D^{-1}(\vec{p}, \vec{m}) z(\vec{p}, \vec{m}),
\]
in addition to the temperature-dependent mean-field contribution. A Debye cutoff \((\rho_c^3 = 6\pi^2 c)\) 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,
\[
D^{-1}(\vec{p}, \vec{m}) = -i \nu_m + g \mu_B B + J_{pd} n^* + \frac{cJ_{pd} S}{2\beta V} \sum_{n, \vec{k}} G^{\text{MF}}_{\vec{k}}(\vec{k}, \omega_n) G^{\text{MF}}_{\vec{k}}(\vec{k} + \vec{p}, \omega_n + \nu_m),
\]
with the mean-field itinerant carrier Green’s function \( G^{\text{MF}}_{\vec{k}}(\vec{k}, \omega_n) = -[i \omega_n - (\epsilon_k + \sigma \Delta / 2 - \mu^*)]^{-1} \). The mean-field spin density is denoted by \( n^* = (n_l - n_t)/2 \), and \( \epsilon_k = \hbar^2 k^2/(2m^*) \).

**Excitations.**— We obtain the spectral density of the spin-fluctuation propagator by analytical continuation, \( \nu_m \rightarrow \Omega + i0^+ \) 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, \]
\( J_{pd} = 0.15 \text{ eV nm}^3, \) and \( c = 1 \text{ nm}^{-3} \), 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 \( \epsilon_F \) is the Fermi energy of the majority-spin band, yields for small and large momenta the dispersion of the collective modes,
\[
\Omega^{(i)}_p = \frac{x}{1 - x} \epsilon_p \left( 1 - \frac{4\epsilon_p}{5\Delta} \right) + O(p^4),
\]
\[
\Omega^{(i)}_p = x\Delta - \frac{1}{2} \epsilon_p + O(1/p^4).
\]
Fig. 1. Spin-wave dispersion for \( c = 0.1 \text{ nm}^{-3} \). The short wavelength limit is the mean-field result \( x\Delta \). For comparison, we show also the result obtained from an RKKY picture.
obtain equivalent results. We find that out the smaller spins using a path-integral formulation to by HP bosons and expanding up to quadratic order, we represent the spins and small spin coupled large spin excitation spectrum are, not coincidentally, similar to those completely fails as a theory of the ferromagnetic state.

(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

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

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

$$\Omega_p^{(1/2)} = \frac{\Delta}{2} \left[ -(1 - x) \pm \sqrt{(1 - x)^2 + 4x\gamma_p} \right],$$

with $x = s/S$ and $\Delta = 6JS$, where $J$ is the exchange coupling, and $\gamma_p = (1/3) \sum_a [1 - \cos(p_a)]$ 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 $eS$, 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 $\hat{S} \cdot \hat{s}$ by $S^2s^2$) and letting the mean-field itinerant-carrier spin splitting, $\Delta(T) = J_{pd}(S^2)$, reflect the thermal suppression of the impurity-spin density $\langle S^2 \rangle$. 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 ($\pm 2S$) on a site is then easily applied, and we obtain

$$\langle S^2 \rangle = \frac{1}{V} \sum_{|\rho| < \rho_c} SB_S(\beta S\Omega_p)$$

$$= \frac{1}{V} \sum_{|\rho| < \rho_c} \left[ S - n(\Omega_p) + (2S + 1)n[(2S + 1)\Omega_p] \right],$$

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.

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

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Our self-consistent spin-wave approximation consists of using Eq. (15) when $\Omega_p$ 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

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

where $N_p = S - SB_S(\beta 5\Omega_p)$ 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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[16] To simplify the present discussion, we use a generic single-band model and neglect interactions between the free carriers as well as the antiferromagnetic Mn-Mn interaction. In quantitative calculations for particular materials it is necessary to account for enhanced ferromagnetism due to interactions [12], realistic heavy and light hole bands, and, at high carrier densities, also the spin-orbit split-off band [13]. These important effects will be discussed separately [J. König, H. Lin, and A. H. MacDonald (to be published)].
[20] Because of spin-orbit coupling in the valence bands, the spin-wave spectrum in realistic models will have a strain- and disorder-dependent gap; M. Abolfath, J. Brum, T. Jungwirth, and A. H. MacDonald (to be published).
[22] In the following, we neglect Stoner excitations and magnons in the itinerant carriers; i.e., we assume sharp spin-wave excitations with spectral weight one.