Transition Temperature of a Magnetic Semiconductor with Angular Momentum j

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We employ dynamical mean-field theory to identify the materials properties that optimize T_c for a generalized double-exchange model. We reach the surprising conclusion that T_c achieves a maximum when the band angular momentum j equals $3/2$ and when the masses in the $m_j = \pm 1/2$ and $\pm 3/2$ subbands are equal. However, we also find that T_c is significantly reduced as the ratio of the masses decreases from one. Consequently, the search for dilute-magnetic semiconductor materials with high T_c should proceed on two fronts. In semiconductors with p bands, such as the currently studied Mn-doped Ge and GaAs semiconductors, T_c may be optimized by tuning the band masses through strain engineering or artificial nanostructures. On the other hand, semiconductors with s or d bands with nearly equal effective masses might prove to have higher T_c's than p-band materials with disparate effective masses.

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The discovery of dilute-magnetic semiconductors (DMS) [1,2] with current transition temperatures up to 250 K [3,4] initiated an active search for the optimal material for spintronic device applications [5]. To facilitate the search for new magnetic semiconductors with high transition temperatures, we evaluate T_c for a double-exchange system with general angular momentum j using dynamical mean-field theory (DMFT). Surprisingly, T_c is found to reach a maximum for $j = 3/2$ and for equal light and heavy hole masses. However, we also find that T_c decreases significantly when the ratio of the masses is reduced.

The notion of using magnetic semiconductors in spintronic devices dates back to the 1960s, when europium chalcogenides [6] and chromium spinels [7] were extensively studied. In the eighties, II-VI [8] and IV-VI [9] DMS were developed by alloying nonmagnetic semiconductors with magnetic ions. Improvement in growth techniques [1,2] has pushed the T_c of $Ga_{1-x}Mn_xAs$ to values up to 250 K. As we discuss later, the large value of T_c in delta-doped GaAs [4] might be associated with the reduced magnetic frustration when the Mn ions are restricted to two-dimensional planes.

Our calculation employs DMFT [10,11], which is one of the most powerful many-body techniques for studying electronic models such as the Hubbard [12,13] and double-exchange (DE) [14–19] models. DMFT becomes exact in the limit of infinite dimensions and accurately captures the physics of correlated electron systems even in three dimensions. Since it becomes exact in the dilute limit, DMFT is well suited to capture the relevant physics of DMS. Recent work on DMS materials has used DMFT to study variants of the DE model [20,21] with less than one local moment per site.

In this Letter, we focus on the large coupling regime, where J_c is sufficiently large to produce a well-defined impurity band formed by the superposition of localized states around impurity sites. Although the precise role played by the impurity band in DMS is still controversial, an array of experimental probes display features characteristic of an impurity band [22–29]. For large couplings, T_c reaches a plateau as a function of J_c and the qualitative results of this model should be independent of the precise magnitude of J_c [19]. Perhaps more importantly, the behavior of $n_h < x$ holes in the impurity band of a DMS with $x < 1$ magnetic ions per site is accurately predicted by that of a DE model with filling $p = n_h/x < 1$ [21], where n_h is the concentration of holes and x the doping. Therefore, our calculation can be compared with experimental measurements by replacing our filling by the proper ratio of n_h/x [30]. A generalized DE model with one local moment per site and large J_c also has the distinct advantage that analytic results are possible for any angular momentum j of the charge carriers.

In semiconductors like GaAs, the angular momentum of the holes is obtained from the vector sum of the $s = 1/2$ spin with the $l = 1$ orbital angular momentum of the p bands. The $j = 3/2$ band lies highest in energy while the spin-orbit split $j = 1/2$ band lies an energy $\Delta_{so} \approx 340$ meV below [31]. Consequently, almost all of the holes in Mn-doped GaAs populate the $j = 3/2$ band, which in turn is split by crystal fields [32] into a $m_j = \pm 3/2$ subband with heavy holes and a $m_j = \pm 1/2$ subband with light holes. More exotic semiconductors with ($l = 2$) d bands, such as the chalcogenides [6], may contain carriers with total angular momentum $j = 5/2$.

The optimization of T_c is performed in two stages. First, we calculate T_c for a DE model where the charge carriers have angular momentum j but equal masses in all $2j + 1$ subbands. After concluding that the system with $j = 3/2$ and a half-filled lower or upper band has the highest T_c, we examine the effect of different band masses in the $m_j = \pm 1/2$ and $\pm 3/2$ subbands.

The Hamiltonian of a generalized DE model with carriers (holes or electrons) of angular momentum j and equal

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masses in all subbands is given by

$$H = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} c^{\dagger}_{\mathbf{k}\alpha} c_{\mathbf{k}\alpha} - \frac{J_c}{N} \sum_{i,\mathbf{k},\mathbf{k}'} e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i} \mathbf{S}_i \cdot c^{\dagger}_{\mathbf{k}'\alpha} \vec{J}_{\alpha\beta} c_{\mathbf{k}\beta}, \quad (1)$$

where $c^{\dagger}_{\mathbf{k}\alpha}$ and $c_{\mathbf{k}\alpha}$ are the creation and destruction operators for an electron with angular-momentum component $m_j = \alpha$ ($\alpha = -j, -j+1, \ldots, j$) and momentum $\mathbf{k}$, $\mathbf{S}_i = S\mathbf{m}_i$ is the spin of the local moment (treated classically) at site $\mathbf{R}_i$, and $\vec{J}_{\alpha\beta}$ are the $(2j+1)$-dimensional angular-momentum matrices. In DMS only a fraction x of the lattice sites contains magnetic impurities.

Within DMFT, the local effective action $S_{\text{eff}}(\mathbf{m})$ is governed by the bare Green's function $G_0(i\nu_n)$, which contains dynamical information about the hopping of electrons onto and off neighboring sites. Because $S_{\text{eff}}(\mathbf{m})$ is quadratic in the field variables, the full local Green's function $G(i\nu_n)_{\alpha\beta}$ may be readily solved by integration, with the result [14] $\underline{G}(i\nu_n) = \langle (\underline{G}_0(i\nu_n)^{-1} + \tilde{J}_c \vec{\underline{J}} \cdot \mathbf{m})^{-1} \rangle_{\mathbf{m}}$, where $\nu_n = (2n+1)\pi T$ are Matsubara frequencies and underlined quantities are matrices in $(2j+1) \times (2j+1)$ spin space; $\langle X(\mathbf{m}) \rangle_{\mathbf{m}} = \int d\Omega_{\mathbf{m}} X(\mathbf{m}) P(\mathbf{m})$ denotes an average over the orientations $\mathbf{m}$ of the local moment with a probability $P(\mathbf{m}) \propto \exp[-S_{\text{eff}}(\mathbf{m})]$, and $\tilde{J}_c = J_c S$. This relation is solved for a semicircular density of states with full bandwidth W in terms of the electron filling p ($p = 1$ corresponds to one electron per site so that $0 \le p \le 2j+1$) and the local-moment order parameter $M = \langle m_z \rangle_{\mathbf{m}}$, which becomes nonzero below T_c.

As $\tilde{J}_c$ increases, the $(2j+1)$-degenerate band splits into $2j+1$ subbands, each labeled by quantum number m_j and centered at energy $-m_j \tilde{J}_c$. Because of the effect of electronic correlations, the full bandwidth of each subband is lowered from W to $W' = W/\sqrt{2j+1}$. So prior to taking the limit of large J_c for the m_j subband, we must rewrite the chemical potential as $\mu = -m_j \tilde{J}_c + \delta\mu$ where $|\delta\mu| \le W'/2$.

Since matrix quantities like $\underline{G}(i\nu_n)$, $\underline{G}_0(i\nu_n)$, and the self-energy $\underline{\Sigma}(i\nu_n) = \underline{G}_0(i\nu_n)^{-1} - \underline{G}(i\nu_n)^{-1}$ have already been averaged over the orientations of $\mathbf{m}$, they are spin diagonal and may be expanded in powers of $\underline{J}_z$ as $\underline{A} = A_0 \underline{I} + A_1 \underline{J}_z + A_2 \underline{J}_z^2 + \cdots$, where A_n is of order M^n. So to linear order in M, only the first two terms in this expansion are retained and the bare inverse Green's function may be parametrized as $\underline{G}_0(i\nu_n)^{-1} = (z_n - m_j \tilde{J}_c + R_n)\underline{I} + Q_n \underline{J}_z$, where $z_n = i\nu_n + \delta\mu$.

For large J_c and to linear order in M, we solved self-consistent equations for R_n and Q_n. The Curie temperature satisfies the condition

$$\sum_n \frac{R_n^2}{R_n^2 - j(j+1)W'^2/16 m_j^2} = 1, \quad (2)$$

while the filling $p = T\sum_n \mathrm{Tr}\,\mathrm{Re}[\underline{G}(i\nu_n)] + j - m_j + \frac{1}{2}$. These two expressions generalize previous results [14,18] for $j = 1/2$.

For a given j, the largest T_c always occurs when the chemical potential lies in the subbands with $m_j = \pm j$ because those holes or electrons are able to most effectively take advantage of the exchange coupling that mediates the ferromagnetism between the local moments. As expected, T_c is particle-hole symmetric within each subband and is the same for a system with p electrons ($1-p$ holes) or p holes ($1-p$ electrons) per site. So the largest Curie temperature, $T_c^{\max}$, is obtained when the lowest or highest subband is half filled.

For a half-filled subband with $m_j = \pm j$, we obtain an analytic expression for $T_c^{\max}$ by converting the Matsubara sum into an integral (assuming that $T_c^{\max}/W$ is small), with the result

$$\frac{T_c^{\max}}{W} = \frac{1}{4\pi\sqrt{2j+1}} \left\{ 1 - \frac{1}{2\sqrt{j(j+1)}} \tan^{-1}[2\sqrt{j(j+1)}] \right\}. \quad (3)$$

This expression gives a slightly larger value than the exact result obtained from Eq. (2), which is used to plot $T_c^{\max}/W$ versus $(2j-1)/2$ in Fig. 1. For large j, $T_c^{\max} \approx (W'/4\pi)\{1 - \pi/4j\}$ saturates at $W'/4\pi$, as indicated by the dashed line in Fig. 1. Compared to the maximum $T_c^{\max}$ for $j = 1/2$ of $0.0219W$, $T_c^{\max}$ for $j = 3/2$ of $0.0256W$ is 16% higher. Even systems with $j = 5/2$ and $7/2$ have higher T_c's than one with $j = 1/2$. The suppression of T_c for $j = 1/2$ is due to the stronger fluctuations at the smallest angular momentum.

To interpret Fig. 1, keep in mind that in the absence of magnetic impurities, a semiconductor is characterized by the bandwidth W of the conduction band and by the angular momentum j of the charge carriers. So for a class of undoped materials with the same bandwidth, the highest transition temperature will be found in the magnetic semiconductor with $j = 3/2$. An unavoidable weakness of the present approach is that the local moments with spin S are treated classically whereas the charge carriers with angular momentum j are treated quantum mechanically. Including the fluctuations of the local moments will further suppress T_c. Therefore, the optimum DMS system will have $S \gg j$

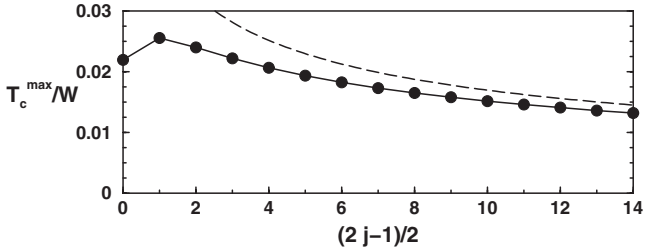

FIG. 1. Ferromagnetic transition temperature for a double-exchange model with general angular momentum j. The maximum transition temperature $T_c^{\max}/W$ (in a half-filled band with $m_j = \pm j$) vs $(2j-1)/2$ with the limiting value $1/(4\pi\sqrt{2j+1})$ plotted in the dashed curve. Large j values, although unphysical, are plotted to illustrate the asymptotic behavior.

so that fluctuations of the local moment can be neglected. This assumption is barely satisfied in Mn-doped GaAs, where $S = 5/2$ [3].

Having established that the semiconductor with $j = 3/2$ has the highest Curie temperature, we now examine the effect of different masses in the heavy $m_j = \pm 3/2$ and light $m_j = \pm 1/2$ subbands of the parent material. For GaAs, the band masses are $m_h = 0.50m$ and $m_l = 0.07m$ (m is the electron mass) with a ratio $r = m_l/m_h = 0.14$ [31]. As pointed out by Zaránd and Jankó [33], the different band masses introduce magnetic frustration because the kinetic energy $K = \sum_{\mathbf{k},\alpha\beta} \epsilon_{\mathbf{k},\alpha\beta} c^\dagger_{\mathbf{k},\alpha} c_{\mathbf{k}\beta}$ is only diagonalized when the angular momentum of the charge carriers is quantized along the momentum direction (for $r = 1$, the quantization axis is arbitrary). Because of the chirality of the holes mediating the ferromagnetism, the moments of a pair of magnetic ions at sites $\mathbf{R}_1$ and $\mathbf{R}_2$ prefer to align perpendicular to the unit vector $\mathbf{r}_{12}$ along the $\mathbf{R}_2 - \mathbf{R}_1$ direction, as sketched in Fig. 2(a).

This anisotropy is easy to understand. Most of the carriers that couple magnetic ions 1 and 2 will have momentum $\mathbf{k}$ along $\mathbf{r}_{12}$ and angular momentum $\mathbf{j}$ parallel to the magnetic moments. If the moments lie parallel to $\mathbf{r}_{12}$, only heavy holes with angular-momentum component $\mathbf{j} \cdot \mathbf{r}_{12} = 3/2$ would couple the two magnetic moments. But if the moments lie perpendicular to $\mathbf{r}_{12}$ as in Fig. 2(a), holes with angular momentum perpendicular to $\mathbf{r}_{12}$ but momentum parallel to $\mathbf{r}_{12}$ would contain all four angular-momentum components $\mathbf{j} \cdot \mathbf{r}_{12} = \pm 3/2$ (heavy) and $\pm 1/2$ (light). Because of the higher mobility of the light holes, the latter coupling is more effective. When four or more magnetic ions are distributed in space, the magnetic interactions will be frustrated because some of the pairs cannot obtain their lowest energy configuration. This is shown schematically in Fig. 2(b), where the circles (crosses) indicate pairs of moments that are (not) in their minimum energy configurations. The large value of T_c in delta-doped GaAs [4] may be related to the reduced magnetic frustration when Mn ions restricted to two-dimensional planes can order with their moments perpendicular to the plane.

The calculation of T_c is performed using two semicircular densities of states such as the ones sketched in the right-hand corner of Fig. 3(a) with bandwidths W_l and $W_h = rW_l$. As the interaction J_c is turned on, the interacting density of states separates into four identical subbands which are *not* particle-hole symmetric. In Fig. 3(a), we plot one of these subbands for a range of effective mass ratios r. For $r < 1$, the interacting density of states is always weighted towards lower energies, where the heavy holes dominate.

In Fig. 3(b), we plot T_c/W_l versus filling for the same values of r used in Fig. 3(a). Not surprisingly, T_c decreases as r decreases from 1 due to the magnetic frustration introduced by the chirality of the charge carriers. Hence, the maximum T_c for a fixed light bandwidth is obtained when the band masses are identical.

As r drops from 1 to 0, $T_c^{\max}/W_l$ plotted in Fig. 4 decreases by 50% from 0.0256 to 0.0127. For the experimental value $r = 0.14$ [31], $T_c^{\max} \approx 0.0133 W_l$ has decreased 48% from its unfrustrated value. The experimental parameters r for a variety of semiconductors with $j = 3/2$ are also indicated in Fig. 4. Based on numerical results with Mn concentration $x < 1$ [21], however, the suppression of T_c may not be quite so dramatic as suggested by this figure [30]. Notice also that a system with $j = 1/2$ or $j = 5/2$ and nearly identical band masses may

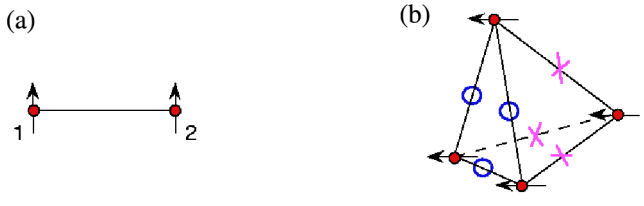

FIG. 2 (color online). Cartoon illustrating the magnetic frustration due to the chirality of the holes mediating the ferromagnetism. (a) The minimum energy configuration for a pair of magnetic moments, (b) the frustration that occurs when 4 magnetic ions lie at the corners of a tetrahedron.

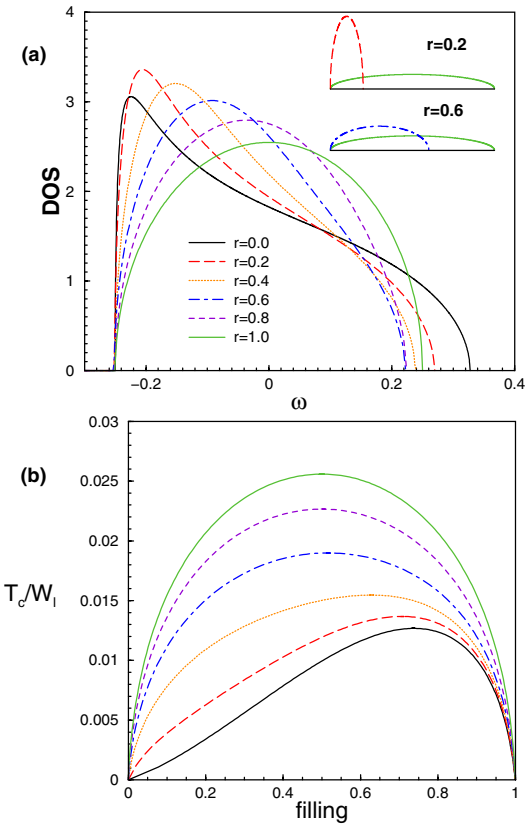

FIG. 3 (color online). Results for a double-exchange model with $j = 3/2$ and different carrier masses in the heavy and light subbands. (a) The interacting density of states for $j = 3/2$ and a range of ratios r between the effective masses; at the right-hand corner the noninteracting densities of states for $r = 0.2$ and 0.6. (b) The dependence of T_c/W_l on filling for $j = 3/2$ and the same values of r.

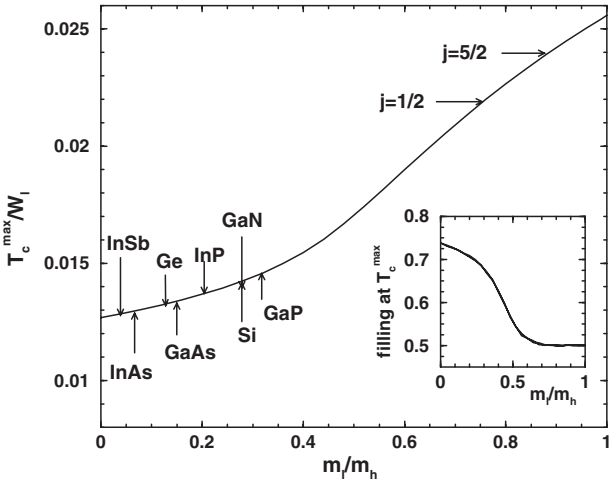

FIG. 4. Relation between $T_c^{\max}$ and the ratio of effective masses $r = m_l/m_h$. In the main panel, $T_c^{\max}/W_l$ is plotted vs $r = m_l/m_h$, with the experimental ratios denoted for several semiconductors. The horizontal arrows denote $T_c^{\max}/W_l$ for $j = 1/2$ and $j = 5/2$ (equal masses in all subbands). The inset displays the filling for which the largest T_c is obtained vs m_l/m_h.

have a higher transition temperature than one with $j = 3/2$ but very different effective carrier masses.

In the inset to Fig. 4, we plot the filling at which T_c is maximized versus the ratio of masses. This filling varies from $p = 0.5$ at $r = 1$ to roughly 0.74 at $r = 0$. So for $r < 1$, the transition temperature is no longer particle-hole symmetric within the lowest subband. This may explain why the transition temperature in $Ga_{1-x}Mn_xAs$ continues to increase for hole fillings supposedly larger than half of the Mn doping [34,35]. Based on this work, we expect that T_c reaches a maximum for a filling close to 70%.

To summarize, we have examined the general dependence of the transition temperature of a magnetic semiconductor on the angular momentum and filling of the charge carrier bands. For a fixed bandwidth of the parent compound, T_c is maximized when $j = 3/2$, the same angular momentum carried by the holes in GaAs and Ge. For $j > 3/2$, the angular momentum of the charge carriers effectively suppresses T_c due to the narrowing of the impurity band by electronic correlations. However, the suppression of T_c due to the different effective masses in p-band semiconductors is even more significant. Because of the strong reduction of T_c by magnetic frustration, s-band ($j = 1/2$) or d-band ($j = 5/2$) semiconductors with more nearly equal band masses may prove to have higher T_c's than p-band ($j = 3/2$) semiconductors with disparate effective masses. For the class of $j = 3/2$ semiconductors, T_c will be maximized when the effects of magnetic frustration are reduced as much as possible. This may be achieved either by applying strain, thereby altering the band structure, or through digital doping [4].

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