Anisotropic exchange interactions in III-V diluted magnetic semiconductors

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The RKKY interaction between substitutional Mn local moments in GaAs is both spin-direction dependent and spatially anisotropic. In this paper we address the strength of these anisotropies using a semiphenomenological tight-binding model that treats the hybridization between Mn *d*-orbitals and As *p*-orbitals perturbatively and accounts realistically for its nonlocality. We show that valence-band spin-orbit coupling, exchange nonlocality, and band-structure anisotropy all play a role in determining the strength of these effects. We use the results to estimate the degree of ground-state magnetization suppression due to frustrating interactions between randomly located Mn ions and to comment on the relationships between different models of III-V diluted magnetic semiconductor ferromagnetism.

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I. INTRODUCTION

The current interest in diluted magnetic semiconductors (DMS) is fueled by possible applications in spintronics and by basic-science issues involving the interplay between disorder, spin-orbit coupling, and magnetism. We concentrate here on the prototypical ferromagnetic III-V DMS $Ga_{1-x}Mn_xAs$, which, once interstitial Mn ions have been eliminated, exhibits robust homogeneous ferromagnetism with critical temperatures T_c above 160 K for $x \gtrsim 0.05$.[1] It is generally agreed that the substitutional Mn ions are in Mn^{2+} states with $S=5/2$, $L=0$ local moments. Exchange interactions between the Mn moments and the valence-band orbitals of their As neighbors lead to intermoment coupling. The effective exchange interaction between Mn moments is spatially anisotropic and also anisotropic in spin space. This paper is motivated primarily by theoretical interest[2–4] in the role of anisotropies in determining the character of the magnetic ground state.

The theory of (III,Mn)V ferromagnetism has been developed in several directions. A simple phenomenological approach[5–10] approximates the valence-band holes by the host-semiconductor Kohn-Luttinger envelope-function Hamiltonian, and couples them to randomly located Mn spins by a *local isotropic* exchange interaction J_{pd}. This leads to a semiquantitative description of many transport and magnetic properties, particularly in the high-carrier-density, high-T_c systems that are free of compensating Mn interstitials. However, it has led to conflicting conclusions on the importance of exchange anisotropy. The RKKY interaction obtained by Zaránd and Jankó[2] is highly anisotropic in *spin* space, i.e., it depends strongly on the orientation of two spins relative to their connecting vector, but is spatially isotropic because it relies on a *local* hole-impurity exchange interaction and a *spherical* approximation for the bands. Using a more realistic band model, Brey and Gómez-Santos[3] find that both spin and real-space anisotropies are weak. Their conclusion, however, depends in part on the momentum-space cutoff used for the exchange interaction J_{pd}, i.e., on atomic-length-scale physics not described realistically in the envelope-function approach. First-principles calculations do not have these limitations, but ususally neglect spin-orbit interactions so that the Mn spin-spin interactions are always isotropic in spin space. In addition the interactions they predict are very sensitive to their placement of Mn *d*-orbital energies relative to the valence band, a quantity that is not predicted reliably by the commonly used local spin-density approximation. In this paper we address exchange anisotropy using an approach that is partly phenomenological, but at a more microscopic level than the envelope function models. We employ a tight-binding model that combines virtues of the envelope function and *ab initio* approaches and, in particular, accounts naturally for nonlocal exchange interactions. We conclude that the bulk magnetization suppression due to frustrating interactions between impurity moments is small. We also discuss the wide variation in critical temperatures resulting from this and other approaches and suggest a possible route toward higher temperature ferromagnetism in (III,Mn)V DMS materials.

II. THEORY

Our analysis of anisotropies is based on a Slater-Koster[11] tight-binding model for the host-semiconductor band structure combined with a perturbative treatment[2,3,5,6,10,12–22] of hybridization between Mn *d*-orbitals and As *p*-orbitals. A similar model has been used previously to obtain the local density of states in the region near a Mn impurity.[23] In Slater–Koster theory, the electronic structure is specified by orbital-dependent on-site energies and hopping amplitudes that are treated as fitting parameters. Spin-orbit coupling is included as a local interaction at each lattice site.[24] Our Hamiltonian reads $H=H_c+H_d+H_{\rm hyb}$, where

$$H_c = \sum_{\mathbf{k}} \sum_{\alpha\alpha'\sigma\sigma'} \epsilon_{\alpha\sigma;\alpha'\sigma'}(\mathbf{k}) c^\dagger_{\mathbf{k}\alpha\sigma} c_{\mathbf{k}\alpha'\sigma'} \qquad (1)$$

describes perfect GaAs.[11,24] Here, $c^\dagger_{\mathbf{k}\alpha\sigma}$ creates an electron with wave vector $\mathbf{k}$ in orbital α with spin σ. The strong interactions within the open *d*-shell are parametrized by a local Hubbard repulsion U and Hund's-first-rule coupling J_H,

$H_d=(\epsilon_d+J_H-U/2)\hat{N}+1/2(U-J_H/2)\hat{N}^2-J_H\mathbf{S}\cdot\mathbf{S}$, with $\hat{N}\equiv\Sigma_{n\sigma}d^\dagger_{n\sigma}d_{n\sigma}$ and $\mathbf{S}\equiv\Sigma_{n\sigma\sigma'}d^\dagger_{n\sigma}(\boldsymbol{\sigma}_{\sigma\sigma'}/2)d_{n\sigma'}$. Here $d^\dagger_{n\sigma}$ creates an electron in d-orbital n with spin σ.[25,26] We assume[27] $U\approx 3.5$ eV and[28] $J_H\approx 0.55$ eV. $H_{\rm hyb}$ describes the hybridization between the d-orbitals and sp-bands,

$$H_{\rm hyb}\equiv H^-_{\rm hyb}+H^+_{\rm hyb}, \tag{2}$$

$$H^-_{\rm hyb}\equiv\frac{1}{\sqrt{\mathcal{N}}}\sum_{\mathbf{k}}\sum_{\alpha n}t_{\mathbf{k}\alpha n}c^\dagger_{\mathbf{k}\alpha\sigma}d_{n\sigma}, \tag{3}$$

$$H^+_{\rm hyb}\equiv\frac{1}{\mathcal{N}}\sum_{\mathbf{k}}\sum_{\alpha\sigma n}t^*_{\mathbf{k}\alpha n}d^\dagger_{n\sigma}c_{\mathbf{k}\alpha\sigma}, \tag{4}$$

where $\mathcal{N}$ is the number of unit cells. The coefficients are expressed in terms of real-space hopping matrix elements, $t_{\mathbf{k}\alpha n}=\Sigma_i e^{-i\mathbf{k}\cdot\mathbf{u}_i}t_{i\alpha n}$, where the sum runs over the nearest-neighbor As sites surrounding each Mn impurity. The symmetries of $t_{\mathbf{k}\alpha n}$ are obtained from Slater-Koster theory,[11] which expresses the matrix elements in terms of two-center integrals. We use $(pd\sigma)=1.0$ eV and $(pd\pi)=-0.46$ eV from photoemission[27] and $(sd\sigma)=1.5$ eV from a rough spin average of *ab initio* calculations for zinc-blende MnAs.[29,30] We note that our model neglects chemical shifts of the p orbitals on the Mn site and Coulomb interactions between the band electrons and Mn ions which are responsible for disorder in the valence band system. For high Mn concentration Coulomb disorder is relatively strong,[14] which may have an influence on the RKKY interactions that is not accounted for by our calculations but would likely reduce spatial anisotropies.

There are two basic approaches to solving this type of model. For *weak* interactions one can treat H_d in a mean-field approximation. The second approach, which we follow here, is appropriate in the *strong-coupling* regime of U large compared to hybridization. In this case we can integrate out d-shell charge fluctuations using canonical perturbation theory (CPT),[31,32] leaving Mn sites with spin degrees of freedom. First consider a single Mn impurity.[32] We introduce the canonically transformed Hamiltonian $\tilde{H}\equiv e^{-i\epsilon T}(H_c+H_d+\epsilon H_{\rm hyb})e^{i\epsilon T}$ and expand in ϵ. The Hermitian operator T is chosen so that the sum of terms linear in ϵ vanishes. To obtain manageable expressions we neglect the energetic spread of virtual band-electron states compared to the energy difference $\sim U$ between different Mn valence states. To be consistent we also ignore contributions from bands other than the heavy-hole, light-hole, and split-off bands. Truncating the expansion at second order and projecting onto the $N=5$, $S=5/2$ ground-state subspace, we obtain

$$\tilde{H}\cong H_c+\frac{H^+_{\rm hyb}H^-_{\rm hyb}}{E_{5,5/2}-E_{4,2}}+\frac{H^-_{\rm hyb}H^+_{\rm hyb}}{E_{5,5/2}-E_{6,2}}. \tag{5}$$

We have used that $H^\pm_{\rm hyb}$ applied to a state in the $(N,S)=(5,5/2)$ sector results in a state with sharp quantum numbers $(N,S)=(6,2)$ and (4,2), respectively. E_{NS} is the isolated-ion energy for quantum numbers (N,S). Inserting Eq. (2) and noting that $\Sigma_{\sigma\sigma'}d^\dagger_{n\sigma}(\boldsymbol{\sigma}_{\sigma\sigma'}/2)d_{n\sigma'}=\mathbf{S}/5$ in the (5,5/2) sector, we obtain a Hamiltonian that includes a microscopic hole-impurity exchange interaction,

$$\tilde{H}=H_c+(\text{charge scattering})-\frac{1}{\Delta}\frac{1}{\mathcal{N}}\sum_{\mathbf{k},\mathbf{k}'}\sum_{\alpha\alpha' n}t^*_{\mathbf{k}\alpha n}t_{\mathbf{k}'\alpha' n}\sum_{\sigma\sigma'}c^\dagger_{\mathbf{k}'\alpha'\sigma'}\frac{\boldsymbol{\sigma}_{\sigma'\sigma}}{2}c_{\mathbf{k}\alpha\sigma}\cdot\mathbf{S} \tag{6}$$

with

$$\frac{1}{\Delta}\equiv\frac{2}{5}\left(\frac{1}{\epsilon_d-4J_H+4U}+\frac{1}{-\epsilon_d-J_H-5U}\right). \tag{7}$$

The two energy denominators in $1/\Delta$ are, respectively, the isolated-ion $d^4\rightarrow d^5$ and $d^6\rightarrow d^5$ transition energies measured from the chemical potential. Both must be negative for (5, 5/2) to be the isolated-ion (i.e., the $t_{\mathbf{k}\alpha n}\rightarrow 0$) ground state as assumed here.

The approach we use is suitable only when both denominators are much larger than the band Fermi energy, an assumption that is valid in (Ga,Mn)As but not in (Ga,Mn)P or (Ga,Mn)N; in the latter case the $d^4\rightarrow d^5$ denominator even appears to be positive. The exchange interaction in Eq. (6) is invariant under spin rotation, while its wave vector dependence is specified by the factor $\Sigma_n t^*_{\mathbf{k}\alpha n}t_{\mathbf{k}'\alpha' n}$, for which we can obtain analytic expressions from tight-binding theory. In the limit $\mathbf{k},\mathbf{k}'\rightarrow 0$ for $\alpha=\alpha'=p_x,p_y,p_z$ we obtain

$$\sum_n t^*_{0\alpha n}t_{0\alpha n}=\frac{16}{27}[3(pd\sigma)^2-4\sqrt{3}(pd\sigma)(pd\pi)+4(pd\pi)^2]. \tag{8}$$

The envelope-function based phenomenological description of DMS ferromagnetism has a single phenomenological parameter J_{pd} which is the $\mathbf{k}\rightarrow 0$, $\mathbf{k}'\rightarrow 0$ limit of the general exchange interaction; the $\mathbf{k},\mathbf{k}'$ dependence is assumed to be negligible because the carrier density is sufficiently low. (The RKKY interaction anisotropy follows from the *full* $\mathbf{k},\mathbf{k}'$ dependence of this quantity as described below.) Since $1/\Delta<0$ the exchange interaction is *antiferromagnetic*, $J_{pd}<0$. $|J_{pd}|$ is minimized and the effective model has the widest range of validity when the $d^5\rightarrow d^4$ and $d^5\rightarrow d^6$ transition energies coincide. In this case we find that $J_{pd}=-48$ meV nm^3, close to but somewhat smaller than the value[33] inferred from experiment. This circumstance suggests that both energy denominators are large and therefore provides support for the validity of the weak hybridization approximations we employ in the case of GaAs. For the sake of definiteness we fix ϵ_d at the equal energy denominator value in the numerical calculations described below, although we note that our results depend on the parameters U,J_H, and ϵ_d only through the factor $1/\Delta$. Changes in these experimentally somewhat uncertain[27] parameters simply change the scale of the RKKY interactions, without altering the anisotropies on which we focus.

The expression for J_{pd}, combined with materials trends,[34] suggests that the T_c of $Ga_{1-x}Mn_xAs_{1-y}P_y$ quaternary alloys might *increase* with y since the $d^5\rightarrow d^4$ transition energy moves closer to E_F, $|1/\Delta|$ becomes larger, and the pd hybridization strengthens. Eventually larger values of $|J_{pd}|$ will lead

to a more localized disturbance of valence-band orbitals, less coupling between separated impurity spins, and the lower T_c (Refs.12 and 35–37) that appears to be seen experimentally in $Ga_{1-x}Mn_xP$, along the way invalidating the approximations we make here. There is no reason to expect that the maximum T_c occurs either at $y=0$ or at $y=1$. In our opinion varying y is certain to lead to higher T_c values provided that materials preparation issues, particularly the problem of maintaining a low density of Mn interstitials, can be solved.

We next evaluate the RKKY interaction between two Mn spins at 0 and **R**, which is sensitive to the nonlocality of the *pd* exchange interaction. We start from the Hamiltonian for two Mn impurities,

$$H = H_c + H_d(0) + H_{\rm hyb}(0) + H_d(\mathbf{R}) + H_{\rm hyb}(\mathbf{R}) \tag{9}$$

and apply the CPT. Integrating out the band electrons and expanding the action to second order in the *full* $(\mathbf{k},\mathbf{k}')$-dependent exchange coupling we obtain

$$\begin{aligned} H_{\rm RKKY} &= \frac{1}{4\beta\Delta^2}\sum_{\mu\nu} S_1^\mu S_2^\nu \frac{1}{\mathcal{N}^2}\sum_{\mathbf{k},\mathbf{k}'}\sum_{i\omega} \mathrm{Tr}\, e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \\ &\times[-i\omega+\hat{\epsilon}(\mathbf{k})-\mu]^{-1}\hat{j}^\mu(\mathbf{k},\mathbf{k}') \\ &\times[-i\omega+\hat{\epsilon}(\mathbf{k}')-\mu]^{-1}\hat{j}^\nu(\mathbf{k}',\mathbf{k}) \\ &\equiv -\sum_{\mu\nu} J_{\mu\nu}(\mathbf{R}) S_1^\mu S_2^\nu, \end{aligned} \tag{10}$$

where $\beta=1/k_BT$ is the inverse temperature, $\hat{\epsilon}(\mathbf{k})$ is the tight-binding Hamiltonian with matrix elements $\epsilon_{\alpha'\sigma';\alpha\sigma}(\mathbf{k})$, μ is the chemical potential, and $j^\mu(\mathbf{k},\mathbf{k}')_{\alpha'\sigma';\alpha\sigma} \equiv \Sigma_n t^*_{\mathbf{k}\alpha n} t_{\mathbf{k}'\alpha' n}\sigma^\mu_{\sigma'\sigma}$. The trace is over orbital and spin indices. We diagonalize $\hat{\epsilon}(\mathbf{k})=\hat{U}^\dagger_{\mathbf{k}}\hat{d}(\mathbf{k})\hat{U}_{\mathbf{k}}$, where $\hat{d}(\mathbf{k})$ is the diagonal matrix of band energies $d_{\alpha\sigma}(\mathbf{k})$, and perform the Matsubara sum. It is useful to express $J_{\mu\nu}(\mathbf{R})$ in terms of its Fourier transform. We obtain

$$\begin{aligned} J_{\mu\nu}(\mathbf{q}) &= \frac{v_{\rm uc}^2}{2\Delta^2}\int\frac{d^3k}{(2\pi)^3}\sum_{\alpha\sigma} f_{\mathbf{k}\alpha\sigma}\sum_{\alpha'\sigma'}(1-f_{\mathbf{k}-\mathbf{q},\alpha'\sigma'}) \\ &\times\frac{1}{d_{\alpha'\sigma'}(\mathbf{k}-\mathbf{q})-d_{\alpha\sigma}(\mathbf{k})}[\hat{U}_{\mathbf{k}}\hat{j}^\mu(\mathbf{k},\mathbf{k}-\mathbf{q})\hat{U}^\dagger_{\mathbf{k}-\mathbf{q}}]_{\alpha\sigma;\alpha'\sigma'} \\ &\times[\hat{U}_{\mathbf{k}-\mathbf{q}}\hat{j}^\nu(\mathbf{k}-\mathbf{q},\mathbf{k})\hat{U}^\dagger_{\mathbf{k}}]_{\alpha'\sigma';\alpha\sigma}, \end{aligned} \tag{11}$$

where $v_{\rm uc}$ is the unit-cell volume and $f_{\mathbf{k}\alpha\sigma}$ is a Fermi factor. In the following, we take the electrons to be at $T=0$.

For our parameter values, the occupied Mn *d*-orbitals lie 3.125 eV below E_F. For 5% Mn and one hole per Mn the valence band crosses these levels first at $k\approx 1.277(2/a)$ along(111), where a is the dimension of the fcc unit cell. Therefore, our calculation of $J_{\mu\nu}(\mathbf{q})$ is valid for q smaller than about twice that value and $J_{\mu\nu}(\mathbf{R})$ is quantitatively reliable for $R \gg 0.2\ a$ and should thus be reasonable for all separations except when the Mn atoms sit on neighboring cation sites. In this case we have in any event neglected superexchange interactions and Coulomb hole-confinement effects which likely play a role.[6] We return to the superexchange interaction below.

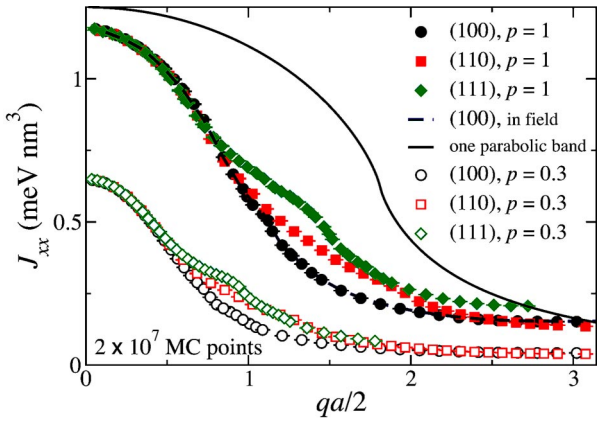

FIG. 1. (Color online) Fourier-transformed RKKY interaction $J_{xx}(\mathbf{q})$ along high-symmetry directions with numerical errors for 5% Mn substitution and $p=1$ (filled symbols) and $p=0.3$ (open symbols) holes per Mn, respectively. The dashed curve shows $J_{xx}(\mathbf{q})$ for $p=1$ in the (100) direction calculated with a band Zeeman splitting of 0.125 eV, corresponding to full polarization of Mn moments. For comparison, the solid line show the results for one (heavy-hole mass) *parabolic* band assuming a local J_{pd}.

III. RESULTS AND DISCUSSION

We have evaluated $J_{\mu\nu}(\mathbf{q})$ at $T=0$ using Monte Carlo (MC) integration with the VEGAS algorithm.[38] Figure 1 shows $J_{xx}(\mathbf{q})$ along high-symmetry directions in momentum space for two different carrier densities, $p=1$ and $p=0.3$ holes per Mn, respectively. A important aspect of our results is the rapid decline of $J_{xx}(\mathbf{q})$ at small $|\mathbf{q}|$, compared to the case of RKKY interactions calculated for a single parabolic band system. As emphasized earlier,[10] $J_{\mu\nu}(\mathbf{q})$ decays faster, the RKKY interactions has substantially longer range in real space, and mean-field theory is more accurate, because of the multiband character of the valence band. The faster fall off results from the momentum dependence of Bloch-state orbital content, i.e., from the **k** dependence of the unitary matrices $U_{\mathbf{k}}$ in Eq. (11), an effect absent in a one-band model. The momentum-space RKKY interactions are weaker at lower carrier density, and are only moderately anisotropic. Figure 1 shows that adding a realistic Zeeman splitting to the bands has little effect on $J_{\mu\nu}(\mathbf{q})$, justifying the pairwise RKKY model for the spin-spin interactions.[5,6] We note that $J_{\mu\nu}(\mathbf{q}=0)$ is isotropic; this limit determines the bulk magnetic anisotropy,[8,9] which vanishes in unstrained samples in the RKKY approximation.

$J_{\mu\nu}(\mathbf{R})$ was evaluated as a Fourier sum over $J_{\mu\nu}(\mathbf{q})$ using a grid of $(2n_k)^3/2$ points in the fcc Brillouin zone. The resulting RKKY interaction, plotted in Fig. 2, is ferromagnetic at small separations and shows Friedel oscillations at larger separations. In the range of separations we consider here[39] the amplitude does not fall off as R^{-3}, consistent with the highly anisotropic Fermi surface. It is apparent that $J_{\mu\nu}(\mathbf{R})$ has more anisotropy, in orbital space and in spin space, and less systematic behavior when looked at as a function of lattice-vector separation **R**, compared to the behavior of $J_{\mu\nu}(\mathbf{q})$ at small wave vectors. In particular, the exchange interaction is often very different in different lattice directions.

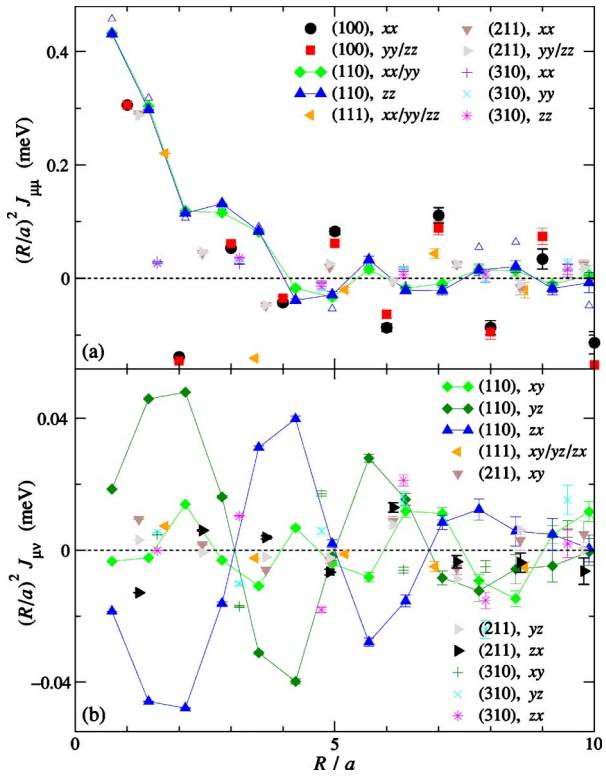

FIG. 2. (Color online) (a) Diagonal and (b) off-diagonal components of the RKKY interaction $J_{\mu\nu}(\mathbf{R})$ for 5% Mn and $p=1$ hole per Mn($E_F=-0.526$ eV) in various crystal directions, scaled by $(R/a)^2$. All results have been obtained with $n_k=36$ and 2×10^5 MC points for each $\mathbf{q}$ point except for $(qa/2)^2\leqslant0.5$, when 2×10^6 points have been used (Ref. 39). The off-diagonal components vanish along (100). The open triangles show $J_{\mu\mu}(\mathbf{R})$, $\mu=x,y,z$, along (110) for vanishing spin-orbit coupling. The off-diagonal components vanish in this case.

Robust ferromagnetism can occur in diluted moment systems only when the range of the interaction is safely longer than the typical moment separation. Mean-field theory will tend to be most reliable and, everything else being equal, ferromagnetic transition temperatures higher when the interaction range is substantially longer than the typical moment separation. As explained above, the correlation between momentum space direction and spin orientation in the valence bands increases the range of the RKKY interactions we calculate. For long range interactions, macroscopic magnetic behavior will depend primarily on the long wavelength behavior of $J_{\mu\nu}(\mathbf{q})$ which is not very anisotropic in the absence of strain.[8] There is indeed a great deal of evidence that the bulk magnetic properties of (Ga,Mn)As are well described by a long-wavelength continuum theory with magnetic anisotropy and stiffness energies as in standard micromagnetic theory. Anisotropy is effectively averaged out and weakened because each moment interactions with many other moments. Nevertheless, the short distance frustration implied by our $J_{\mu\nu}(\mathbf{R})$ results could in principle reduce the coarse-grained magnetization, as argued in Ref. 2. The anisotropy in *real* space is due to both the directionality associated with *pd* hybridization and the anisotropy of the band structure—neither effect is included in the spherical model of Ref. 2. The relatively weak anisotropy that we find at small $\mathbf{R}$ is consistent with Ref. 3; we attribute the discrepancy at larger $\mathbf{R}$ to the isotropic ansatz for the hole-impurity exchange interaction and the momentum cutoff employed in that paper. Note that we also have a large anisotropy in spin-space that is absent in any calculation that neglects spin-orbit coupling. For small separations the relative spin anisotropy is below 10% as found in Ref. 3.

Before turning to an assessment of the importance of the role of anisotropies in determining the character of the magnetic ground state, it is useful to discuss some of the uncertainties associated with the approach used here. As noted above, we have so far neglected the *superexchange* interaction between Mn moments. Our strong-coupling approach is not expected to give quantitatively correct results for the superexchange interaction since it is of short range, thus involving large momentum transfers for which the band energies become comparable to the energy of the occupied *d* levels. It does nevertheless provide a qualitatively correct description of the superexchange interaction, which appear at *fourth order* in the CPT (Ref. 31) strong-coupling expansion of the Hamiltonian (9). The fourth-order term contains products of four *d*-electron creation/annihilation operators. When the transformed Hamiltonian is projected onto the subspace with $(N,S)=(5,5/2)$ for both impurities, only terms that leave the total number of electrons in the *d*-shells invariant are nonzero. The superexchange process is possible only between Mn ions that are at neighboring fcc lattice sites and involves the virtual transfer of an electron between *d* shells by hopping via the intermediate As atom. (Formally, this stems from the fact that the c and $c^\dagger$ operators are all at equal time. They are replaced by equal-time tight-binding Green functions, which are purely local.) The terms that lead to this interaction are of the same general form as the second-order terms but contain permutations of two c, two $c^\dagger$, two d, and two $d^\dagger$ operators. Their energy denominators contain *three* powers of *d*-orbital energy differences like $E_{4,2}-E_{5,5/2}$ and $E_{6,2}-E_{5,5/2}$. The RKKY interaction is also proportional to the *pd* hybridization to the fourth power but contains *d*-orbital energy differences only to the power of -2 and an additional inverse band-electron energy difference, of the order of E_F^{-1}. The relative strength of these interactions depends on E_F/U and also on the density of band electrons. For (Ga,Mn)As we expect that the RKKY interaction is stronger.

In assessing uncertainties, it is also informative to compare our effective Mn-Mn exchange interactions with those that have emerged from numerous first-principles calculations employing either a supercell approach[15,19,21,22] or the coherent potential approximation.[16,18] All works we are aware of neglect spin-orbit coupling so that the interaction $J(\mathbf{R})$ is a scalar in spin space, whereas in Fig. 2 sizeable off-diagonal components are seen. In agreement with our results, the interaction is typically found to be ferromagnetic for small R.[16,18,19,21,22] Strong real-space anisotropy is evident in all calculations, due to the anisotropic band structure. The most detailed study of this anisotropy is presented by Kudrnovský *et al.*[18] Interestingly, in agreement with our results, the interaction in the (110) direction stays ferromagnetic up to quite large separations, whereas in the (100) direction it becomes antiferromagnetic much earlier. However,

in Ref. 18 the period of Friedel oscillations is very long, suggesting that k_F is much smaller than in our calculation. We attribute this to the Mn *d*-orbital weight at E_F found in the LDA, which in our judgement is likely unphysical. Indeed, LDA+U leads to a reduced period,[18] more consistent with valence-band densities. Generally, approaches that account for local correlations, such as LDA+U and self-interaction-corrected LDA lead to a large shift of *d*-orbital weight away from the Fermi energy,[17–20] in better agreement with experiments.[27]

Finally, we compare critical temperatures estimated from our RKKY interactions and from other approaches with experiment. Assuming the average spin polarization to be equal to $\mathbf{M}=\langle\mathbf{S}\rangle$ and to be parallel to the *z* direction at each site, the mean effective fields acting on the impurity spins are $H_\mu(\mathbf{R}_i)=M^z\Sigma_{j\neq i}J_{\mu z}(\mathbf{R}_i-\mathbf{R}_j)$, where the sum is over Mn impurity sites. Assumming that the Mn ions are distributed completely at random,[14,40] the average over all sites is $\overline{H_\mu}=(xM^z/v_{\rm uc})J_{\mu z}(\mathbf{q}=0)\propto\delta_{\mu z}$; the mean fields are parallel to the magnetization orientation on average. As noted above, $J_{\mu\nu}(\mathbf{q}=0)$ is isotropic for unstrained bulk samples. The resulting mean-field value of the spin polarization is

$$M^z = SB_S\left(\beta\frac{x}{v_{\rm uc}}J_{zz}(\mathbf{q}=0)M^zS\right), \tag{12}$$

where B_S is the Brillouin function, leading to the mean-field Curie temperature

$$k_BT_c=\frac{x}{3}\frac{J_{zz}(\mathbf{q}=0)}{v_{\rm uc}}S(S+1), \tag{13}$$

which in the present model gives $T_c\approx 44.2$ K. This critical temperature is well below the experimental value for $Ga_{.95}Mn_{.05}As$ samples which is close to 160 K. Because the perturbative approach used in RKKY theory is best justified close to T_c, since the average polarization of the local moments is small, this substantial discrepancy requires comment. It arises partly from our choice of *d*-transition energies that minimize the value of $|J_{pd}|$, which is then somewhat too small. Shifting the *d*-orbtial energy ϵ_d in any direction leads to an increase of $|J_{pd}|$ and thus of $T_c\propto J_{pd}^2$. Another effect increasing T_c is the Coulomb interaction between itinerant holes, which favors ferromagnetism. This effect can be incorporated by means of a Fermi liquid parameter.[6,9] These two effects enhance T_c by a factor of about 1.5,[6,9] not large enough to bring our theory into agreement with experiment. We believe that the most important source of error in the T_c estimate is inaccuracies in the tight-binding parametrization of the host band structure which produce unreliable values for the valence-band-edge effective mass values, and in particular underestimate the heavy-hole mass by nearly a factor of 2. We note that our value for T_c is comparable to that of Brey and Gómez-Santos[3] and to the result obtained from a single parabolic band with the heavy-hole mass. We conclude that details of the band-edge electronic structure captured most reliably by the Kohn–Luttinger approach[5–10] are critical for accurate T_c estimates. The Kohn–Luttinger theory uses a local J_{pd} interaction, however, and is not reliable for capturing the anisotropies on which we focus here.

Unlike our tight-binding model, *ab initio* theory typically overestimates T_c.[41] The discrepancy in T_c between *ab initio* and model calculations is accompanied by a discrepancy in $J_{\mu\nu}(\mathbf{R})$. The discrepancy results in part from effects present in *ab initio* calculations that are absent to varying degrees in different phenomenological models. For example, it may reflect the neglect of the nonlocal part of the Coulomb acceptor potential in existing model studies. Including it should increase the hole density at Mn sites that are close to other Mn impurities and thereby increase the Mn-Mn coupling. On the other hand, *ab initio* calculations employing the LDA typically have significant *d*-orbital weight close to E_F, as mentioned above, which leads to an overestimate of the density of states and also of the hole density at Mn sites. Partially correcting for this in the LDA+U approach leads to the decrease of the Mn-Mn exchange interaction and of T_c. On the other hand, even LDA+U does not include correlations between neighboring Mn ions. This may lead to an overestimate of the hole density close to Mn pairs and thus of the Mn-Mn coupling. Further work is clearly required to reach firm conclusions. Of course, mean-field estimates from both model and *ab initio* approaches are not completely reliable since they neglect magnetic fluctuations and also effects from the spatial distribution of Mn spins, which are known to be important for the magnetization.[14,40]

Mahadevan *et al.*[22] have recently argued on the basis of *ab initio* calculations that the interaction between Mn spins cannot be described in an RKKY picture. Their conclusion is based on the claim that the strong spatial anisotropy present in their first-principles results is inconsistent with an RKKY picture, even one that accounts for band-structure anisotropy. We have shown here that a *strongly anisotropic* Mn-Mn exchange interaction results from a realistic RKKY theory, contrary to this claim. We feel that this undermines the main basis for the authors' conclusion that the RKKY picture is not applicable to $Ga_{1-x}Mn_xAs$. They also emphasize that the nonlocality of the carrier-impurity exchange interaction must be taken into account to obtain a realistic Mn-Mn interaction.[22] This is indeed the case, but the assumption of a local carrier-impurity exchange interaction is not imposed by RKKY theory, as we show here by explicitly accounting for this nonlocality.

Keeping the theoretical uncertainties in mind, we return to the issue that has motivated the approach taken in this paper. To determine whether or not frustration due to spin and spatial anisotropies alters the character of the ordered state, we start from a fully aligned (in the *z* direction) spin configuration and consider the mean effective fields $\overline{H_\mu}=(xS/v_{\rm uc})J_{\mu z}(\mathbf{q}=0)\propto\delta_{\mu z}$, again assuming a random distribution of Mn impurities. The typical Mn tilt angle is proportional to the ratio of the *xy* plane effective-field components to $\overline{H_z}$. We find

$$\frac{\overline{H_x^2}}{(\overline{H_z})^2}=(x^{-1}-1)v_{\rm uc}\int\frac{d^3q}{(2\pi)^3}\frac{|J_{xz}(\mathbf{q})|^2}{J_{zz}^2(\mathbf{q}=0)}. \tag{14}$$

Thus the anisotropies become more important for small Mn fractions *x*. For the parameters used above we obtain

$(\overline{H_x^2})^{1/2}/\overline{H_z}=0.0057(x^{-1}-1)^{1/2}$. We conclude that the anisotropies do not cause a large moment suppression in (Ga, Mn) As even for $x\sim 0.01$. The effect is small because with relatively long-range interactions many moments contribute to the effective field, averaging the anistropies. Finally, we remark that we have neglected the indirect influence of charge scattering and local chemical shifts. These will reduce the RKKY interaction at large **R** and further reduce the importance of frustrating interactions.[13] Recently, Fiete *et al.*[4] have also found a weak suppression of the magnetization starting from a Kohn-Luttinger Hamiltonian and employing Monte Carlo simulations.

To conclude, we have used a tight-binding model of III-V DMS to calculate the momentum-dependent hole-impurity exchange interaction. We find that this interaction depends crucially on the energy of the Mn *d*-levels and suggest that $Ga_{1-x}Mn_xAs_{1-y}P_y$ might have a higher T_c than $Ga_{1-x}Mn_xAs$. Starting from the hole-impurity interaction, we have calculated the hole-mediated RKKY interaction between impurity spins. It is highly anisotropic in real and spin space, due to three factors partly ignored in previous works: spin-orbit coupling, the nonlocal hole-impurity exchange interaction, and the anisotropic band structure. However, despite the strong anisotropies, local-moment suppression is weak due to the averaging brought about by the long-range RKKY interaction.

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