Influence of non-local exchange on RKKY 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 Letter we address the strength of these anisotropies using a semi-phenomenological tight-binding model which treats the hybridization between Mn d-orbitals and As p-orbitals perturbatively and accounts realistically for the non-local exchange interaction between their spins. We show that exchange non-locality, valence-band spin-orbit coupling, and band-structure anisotropy all play a role in determining the strength of both effects. We use these results to estimate the degree of ground-state magnetization suppression due to frustrating interactions between randomly located Mn ions.

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The current interest in diluted magnetic semiconductors (DMS) is fueled by possible applications in spintronics and by basic-science issues generated by the interplay between disorder, spin-orbit coupling, and magnetic order. We concentrate on the prototypical III-V DMS $Ga_{1-x}Mn_xAs$, which, once interstitial Mn ions have been eliminated, exhibits robust homogeneous ferromagnetism [1] with critical temperatures T_c above 160K for $x \gtrsim 0.05$. It is generally agreed that the substitutional Mn ions are in Mn^{2+} valence states that have $S = 5/2$, $L = 0$ local moments, and that exchange interactions with As neighbors allow the Mn moments to interact via valence-band holes [2]. The effective exchange interaction between Mn moments is spatially anisotropic and, because of spin-orbit interactions, also anisotropic in spin space. This Letter is motivated primarily by theoretical interest [3, 4] in the role of anisotropies in determining the character of the magnetic ground state but has implications for other aspects of (III,Mn)V DMS ferromagnetism.

The theory of (III,Mn)V ferromagnetism has been developed in several directions. A simple phenomenological approach [5, 6, 7, 8, 9, 10, 11] approximates the valence-band holes by a host-semiconductor Kohn-Luttinger envelope-function Hamiltonian and couples them to randomly located Mn spins by a *local*, *isotropic* exchange interaction $J_{\rm pd}$. This leads to a semi-quantitative 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 [3] is highly anisotropic in *spin* space, *i.e.*, it depends strongly on the orientation of two spins relative to their connecting vector, but it is spatially isotropic because it starts from a *local* hole-impurity exchange interaction and uses a *spherical* approximation for the bands. Using a more realistic 6-band envelope-function Hamiltonian, Brey and Gómez-Santos [4] find that both spin and real space anisotropies are weak. Their conclusion, however, depends in part on their momentum-space cut-off [11] for the exchange interaction $J_{\rm pd}$, *i.e.*, on atomic-length-scale physics not described realistically in the envelope-function approach. First-principles calculations [12] do not have these limitations, but are hampered by their extreme sensitivity to the placement of unoccupied and occupied d-orbital energies relative to the valence and conduction bands. In this Letter we address exchange anisotropy using a realistic tight-binding model that combines virtues of these two different approaches and estimate the bulk magnetization suppression due to frustrating interactions between impurity moments. Based on our results we also suggest a possible route toward higher transition temperatures in (III,Mn)V ferromagnets.

Our theory is based on a Slater-Koster [13] tight-binding model, and on a perturbative treatment of pd hybridization, in which the band electrons are integrated out to yield a spin-only model [3, 4, 5, 14, 15, 16]. A similar model has recently been used to obtain the local density of states around Mn impurities [17]. In Slater-Koster theory, the electronic structure is specified by orbital-dependent onsite energies and hopping amplitudes that are treated as fitting parameters. Spin-orbit coupling is included [18] to obtain realistic bands and a realistic description of (III,Mn)V ferromagnetism [8].

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 [13, 18]. Here, $c^\dagger_{\mathbf{k}\alpha\sigma}$ creates an electron with wave vector $\mathbf{k}$ in orbital α with spin σ. The most important effect of Mn impurities is to introduce partially filled d-orbitals. The resulting strong electron-electron interactions are parametrized by the local Hubbard repulsion U and the Hund's-first-rule coupling J_H [19, 20]: $H_d = (\epsilon_d + J_H - U/2)\hat{N} + 1/2\,(U -$

$J_H/2)\hat{N}^2 - J_H\,\mathbf{S}\cdot\mathbf{S}$, with $\hat{N} \equiv \sum_{n\sigma} d^\dagger_{n\sigma} d_{n\sigma}$ and $\mathbf{S} \equiv \sum_{n\sigma\sigma'} d^\dagger_{n\sigma}\,(\boldsymbol{\sigma}_{\sigma\sigma'}/2)\,d_{n\sigma'}$, where $d^\dagger_{n\sigma}$ creates an electron in d-orbital n with spin σ. We assume $U \approx 3.5\,\mathrm{eV}$ [21] and $J_H \approx 0.55\,\mathrm{eV}$ [22]. $H_{\rm hyb}$ describes the hybridization between the d-orbitals and sp-bands,

$$H_{\rm hyb} = \frac{1}{\sqrt{\mathcal{N}}}\sum_{\mathbf{k}}\sum_{\alpha\sigma n} t_{\mathbf{k}\alpha n}\, c^\dagger_{\mathbf{k}\alpha\sigma} d_{n\sigma} + \mathrm{h.c.} \equiv H^-_{\rm hyb} + H^+_{\rm hyb}, \tag{2}$$

where $\mathcal{N}$ is the number of unit cells in the system. The coefficients are expressed in terms of real-space hopping matrix elements, $t_{\mathbf{k}\alpha n} = \sum_i e^{-i\mathbf{k}\cdot\mathbf{u}_i} t_{i\alpha n}$, where the sum runs over nearest-neighbor As sites of the impurity. The symmetries of $t_{\mathbf{k}\alpha n}$ are obtained from Slater-Koster theory [13], which expresses the matrix elements in terms of two-center integrals. We use $(pd\sigma) = 1.0\,\mathrm{eV}$ and $(pd\pi) = -0.46\,\mathrm{eV}$ as inferred from photoemission [21] and $(sd\sigma) = 1.5\,\mathrm{eV}$ obtained as a rough spin average of *ab-initio* calculations for zinc-blende MnAs [23].

In the large-U limit we can use canonical perturbation theory (CPT) [24] to integrate out d-shell charge fluctuations, leaving only the impurity spin degrees of freedom. We first consider a single Mn impurity. We introduce the canonically transformed Hamiltonian $\tilde{H} \equiv e^{-i\epsilon T}\,(H_c + H_d + \epsilon H_{\rm hyb})\,e^{i\epsilon T}$, where T is hermitian, and expand in ϵ. The operator T is chosen so that the linear term 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 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

$$\tilde{H} \cong H_c + \frac{H^+_{\rm hyb}H^-_{\rm hyb}}{E_{5,5/2} - E_{4,2}} + \frac{H^-_{\rm hyb}H^+_{\rm hyb}}{E_{5,5/2} - E_{6,2}}. \tag{3}$$

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 corresponding isolated-ion energy. Inserting Eq. (2) and noting that $\sum_{\sigma\sigma'} d^\dagger_{n\sigma}\,(\boldsymbol{\sigma}_{\sigma\sigma'}/2)\,d_{n\sigma'} = \mathbf{S}/5$ in the $(5,5/2)$ sector, we obtain a Hamiltonian that includes a microscopic hole-impurity exchange interaction,

$$\begin{aligned}\tilde{H} \;&=\; H_c \;+\; (\text{charge scattering}) \\ &-\frac{1}{\Delta}\frac{1}{\mathcal{N}}\sum_{\mathbf{k},\mathbf{k}'}\sum_{\alpha\alpha' n} t^*_{\mathbf{k}\alpha n} t_{\mathbf{k}'\alpha' n}\sum_{\sigma\sigma'} c^\dagger_{\mathbf{k}'\alpha'\sigma'}\frac{\boldsymbol{\sigma}_{\sigma'\sigma}}{2} c_{\mathbf{k}\alpha\sigma}\cdot\mathbf{S}\end{aligned} \tag{4}$$

with

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

The two energy denominators in $1/\Delta$ are respectively the isolated-ion $d^5 \to d^4$ and $d^5 \to d^6$ transition energies measured from the chemical potential. If either of the denominators becomes small, the interval of energy over which our approximations are justified is correspondingly reduced. Note first that the exchange interaction is quite generally invariant under spin rotation. The wavevector dependence of the exchange interaction is specified by the factor $\sum_n t^*_{\mathbf{k}\alpha n} t_{\mathbf{k}'\alpha' n}$ for which we can obtain analytic expressions from tight-binding theory. For $\mathbf{k},\mathbf{k}' \to 0$ and $\alpha = \alpha' = p_x, p_y, p_z$ we obtain

$$\sum_n t^*_{0\alpha n} t_{0\alpha n} = \frac{16}{27}\,[3(pd\sigma)^2 - 4\sqrt{3}\,(pd\sigma)(pd\pi) + 4(pd\pi)^2]. \tag{6}$$

Restoring the prefactor from Eq. (4) we find a microscopic expression for the envelope-function exchange constant $J_{\rm pd}$. By including the *full* $(\mathbf{k},\mathbf{k}')$ dependence we recover spatial anisotropies neglected in that theory.

Since both denominators in $1/\Delta$ must be negative for $(5,5/2)$ to be the isolated-ion ground state, the exchange interaction is *antiferromagnetic*, $J_{\rm pd} < 0$. $|J_{\rm pd}|$ is minimized and the effective model has the widest range of validity when the $d^5 \to d^4$ and $d^5 \to d^6$ transition energies bracket the Fermi energy E_F symmetrically. In this case $J_{\rm pd} = -48\,\mathrm{meV\,nm^3}$, close to the experimental value in (Ga,Mn)As [25]. We consider this case in what follows. The expression for $J_{\rm pd}$, combined with materials trends [26], suggests that T_c of $\mathrm{Ga}_{1-x}\mathrm{Mn}_x\mathrm{As}_{1-y}\mathrm{P}_y$ quaternary alloys might *increase* with y since their $d^5 \to d^4$ transition energy will approach E_F, increasing the value of $J_{\rm pd}$.

We employ the full $(\mathbf{k},\mathbf{k}')$-dependent hole-impurity exchange to evaluate the RKKY interaction between two Mn spins at 0 and $\mathbf{R}$ and perform the CPT as above. Integrating out the band electrons and expanding the action to second order in impurity spins we obtain

$$\begin{aligned}H_{\rm RKKY} \;&=\; \frac{1}{4\beta\Delta^2}\sum_{\mu\nu} S^\mu_1 S^\nu_2\,\frac{1}{\mathcal{N}^2}\sum_{\mathbf{k},\mathbf{k}'}\sum_{i\omega}\mathrm{Tr}\; e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \\ &\times (-i\omega + \hat{\epsilon}(\mathbf{k}) - \mu)^{-1}\,\hat{j}^\mu(\mathbf{k},\mathbf{k}')\,(-i\omega + \hat{\epsilon}(\mathbf{k}') - \mu)^{-1} \\ &\times \hat{j}^\nu(\mathbf{k}',\mathbf{k}) \;\equiv\; -\sum_{\mu\nu} J_{\mu\nu}(\mathbf{R})\,S^\mu_1 S^\nu_2,\end{aligned} \tag{7}$$

where $\hat{\epsilon}(\mathbf{k})$ is the tight-binding Hamiltonian with matrix elements $\epsilon_{\alpha'\sigma';\alpha\sigma}(\mathbf{k})$ and $j^\mu(\mathbf{k},\mathbf{k}')_{\alpha'\sigma';\alpha\sigma} \equiv \sum_n t^*_{\mathbf{k}\alpha n} t_{\mathbf{k}'\alpha' n}\,\sigma^\mu_{\sigma'\sigma}$. The trace in Eq. (7) 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 $J_{\mu\nu}(\mathbf{R}) = \int d^3q/(2\pi)^3\, e^{i\mathbf{q}\cdot\mathbf{R}}\,J_{\mu\nu}(\mathbf{q})$ in terms of its Fourier transform. Making use of the symmetries of $\hat{d}$ and $\hat{U}$ we obtain

$$\begin{aligned}J_{\mu\nu}(\mathbf{q}) = \frac{v^2_{\rm uc}}{2\Delta^2}\int\frac{d^3k}{(2\pi)^3}\sum_{\alpha\sigma} f_{\mathbf{k}\alpha\sigma}\sum_{\alpha'\sigma'}(1 - f_{\mathbf{k}-\mathbf{q},\alpha'\sigma'}) \\ \times\frac{1}{d_{\alpha'\sigma'}(\mathbf{k}-\mathbf{q}) - d_{\alpha\sigma}(\mathbf{k})}\,[\hat{U}_{\mathbf{k}}\hat{j}^\mu(\mathbf{k},\mathbf{k}-\mathbf{q})\hat{U}^\dagger_{\mathbf{k}-\mathbf{q}}]_{\alpha\sigma;\alpha'\sigma'} \\ \times[\hat{U}_{\mathbf{k}-\mathbf{q}}\hat{j}^\nu(\mathbf{k}-\mathbf{q},\mathbf{k})\hat{U}^\dagger_{\mathbf{k}}]_{\alpha'\sigma';\alpha\sigma},\end{aligned} \tag{8}$$

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 $T = 0$. We remark that Eq. (8) is unreliable when $\mathbf{q}$ is comparable to Brillouin-zone dimensions because the band eigenenergies are then as far from the Fermi energy as the d-quasiparticle levels. Correspondingly the results for $J_{\mu\nu}(\mathbf{R})$ are quantitatively reliable only for $R \gg a$, where a is the dimension of the fcc unit cell.

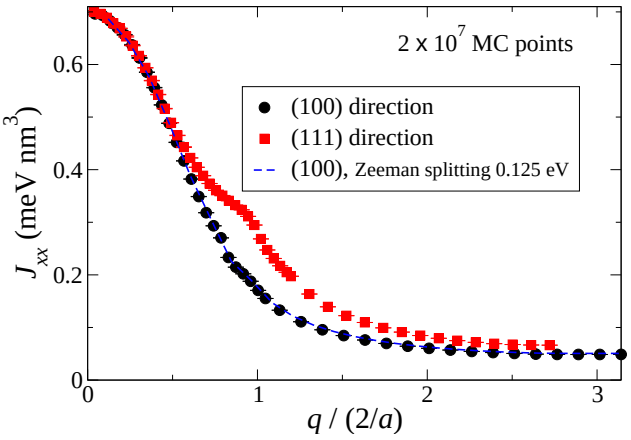

FIG. 1: (color online) Fourier-transformed RKKY interaction $J_{xx}(\mathbf{q})$ and numerical errors in the (100) and (111) directions for $E_F = -0.307$ eV relative to the valence-band top, corresponding to a hole concentration of 3.8×10^{20} cm^{-3}. The dashed curve shows $J_{xx}(\mathbf{q})$ in the (100) direction calculated with a band Zeeman splitting of 0.125 eV, corresponding to 5% Mn substitution and full polarization of Mn moments.

We have evaluated $J_{\mu\nu}(\mathbf{q})$ using Monte Carlo (MC) integration with the VEGAS algorithm [27]. Figure 1 shows $J_{xx}(\mathbf{q})$ in the (100) and (111) directions. At a nonzero Mn density, the interactions between spins are dominated by the pairwise RKKY interaction only if the mean hole-impurity exchange interaction is weak [7, 8]. This is indeed the case since Fig. 1 shows that the effect of a realistic Zeeman splitting on $J(\mathbf{q})$ is small. We note that $J_{\mu\nu}(\mathbf{q} = 0)$ is isotropic; this limit determines the bulk magnetic anisotropy [9, 10] which vanishes in the present approximation [28].

$J_{\mu\nu}(\mathbf{R})$ is evaluated as a Fourier sum over $J_{\mu\nu}(\mathbf{q})$ calculated on a cubic grid with $(2n_k)^3/2$ points in the fcc Brillouin zone, making use of all symmetries. The resulting RKKY interaction is plotted in Fig. 2. It is ferromagnetic at small separations, as expected. The near-neighbor interactions are not reliable, both because their evaluation stretches the validity of the CPT and because we neglect the *superexchange* interaction, which appears at fourth order in $H_{\rm hyb}$, and in which an electron hops virtually from a Mn d-orbital to a d-orbital on a neighboring Mn site via an intervening As p-orbital. For larger separations $J_{\mu\nu}(\mathbf{R})$ shows typical Friedel oscillations.

We find a very strong anisotropy in *real* space; $J_{\mu\nu}(\mathbf{R})$ depends on the direction of $\mathbf{R}$ for similar $R = |\mathbf{R}|$. This is

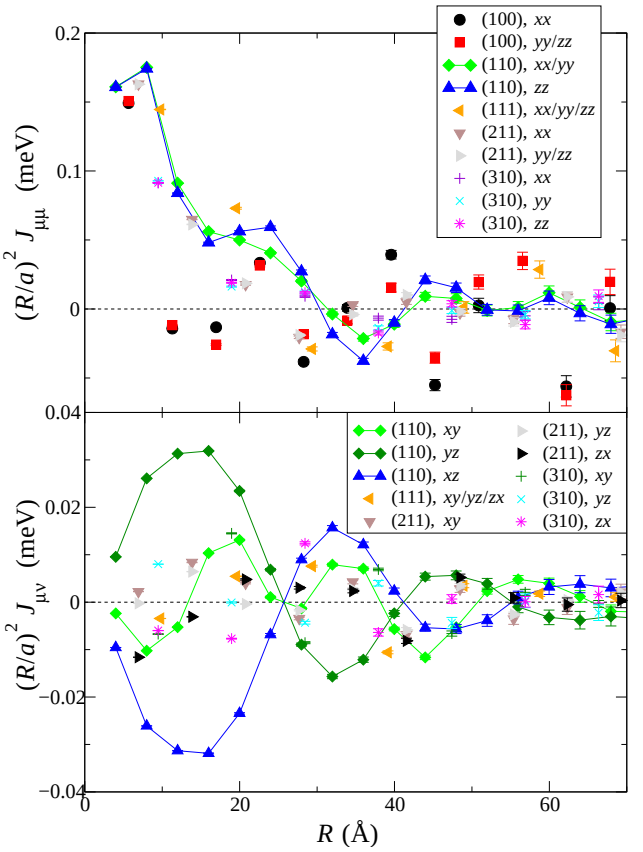

FIG. 2: (color online) (a) Diagonal and (b) off-diagonal components of the RKKY interaction $J_{\mu\nu}(\mathbf{R})$ in various crystal directions, scaled by $(R/a)^2$. All results have been obtained with $n_k = 36$ and 2×10^5 MC points for each $\mathbf{q}$ point except for $(qa/2)^2 \leq 0.5$, when 2×10^6 points have been used. The off-diagonal components vanish exactly along (100).

a consequence of both the directionality associated with *pd* hybridization and of the anisotropy of the band structure and the Fermi surface; neither effect is included in the spherical model of Ref. [3]. In Ref. [4] the real-space anisotropy is concluded to be small, based on the interaction between two spins at neighboring sites. For small R we also find relatively weak anisotropies but at larger R this conclusion does not hold. The isotropic Gaussian ansatz for the hole-impurity exchange interaction [4] contributes to this small anisotropy.

The anisotropy in *spin* space, *i.e.*, the deviation of $J_{\mu\nu}(\mathbf{R})$ from $J(\mathbf{R})\,\delta_{\mu\nu}$, is also large. For small spin-orbit coupling, the differences between diagonal components are of second order in spin-orbit coupling, whereas the off-diagonal components are linear. Only for the smallest separations is the relative anisotropy below 10% as found in Ref. [4]. At larger R the anisotropy becomes quite pronounced, as in Ref. [3].

When the anisotropies are neglected, the moments are fully aligned in the ground state. To determine whether or not the anisotropies substantially alter the

character of the ground state, we start from a fully aligned (in the z direction) spin configuration and consider the mean effective fields acting on individual spins, $H_\mu(\mathbf{R}_i) = S\sum_{j\neq i} J_{\mu z}(\mathbf{R}_i - \mathbf{R}_j)$, where the sum is over Mn impurity sites. Assumming that the Mn ions are distributed completely at random [16, 29], the average over all sites is $\overline{H}_\mu = (xS/v_{\rm uc})\, J_{\mu z}(\mathbf{q}=0) \propto \delta_{\mu z}$. On average the effective fields align with the average moment, but spatial fluctuations reduce the overall degree of spin polarization. The typical angle of the Mn tilt at a given site is proportional to the ratio of the xy plane effective-field components to $\overline{H}_z$. We find

$$\frac{\overline{H_x^2}}{(\overline{H}_z)^2} = (x^{-1}-1)\, v_{uc} \int \frac{d^3q}{(2\pi)^3} \frac{|J_{xz}(\mathbf{q})|^2}{J_{zz}^2(\mathbf{q}=0)}. \qquad (9)$$

Thus the anisotropies become more important for small Mn fractions x. For the parameters used above we get $\overline{H_x^2}/[\overline{H}_z]^2 = 3.1\times 10^{-5}\,(x^{-1}-1)$. We conclude that the anisotropies do not cause a large moment suppression in (Ga,Mn)As even for $x \sim 0.01$, despite the large anisotropies. The effect is small because many moments contribute to the effective field due to the long-range interaction, averaging over the anisotropies. We neglect the indirect influence of charge scattering, as well as Coulomb interactions and local chemical shifts. These will reduce the RKKY interaction at large separations and further reduce the importance of frustrating interactions [15].

To conclude, we have used a Slater-Koster tight-binding model of III-V DMS to calculate the full momentum dependence of the hole-impurity exchange interaction. We find that this interaction depends crucially on the position of the Mn d-levels relative to the valence band and suggest that quaternary $Ga_{1-x}Mn_xAs_{1-y}P_y$ alloys might have higher transition temperatures than $Ga_{1-x}Mn_xAs$. Starting from the hole-impurity interaction, we have calculated the hole-mediated RKKY interaction between impurity spins. This interaction is highly anisotropic in real and spin space. The anisotropy crucially depends on two factors partly ignored in previous works: the nonlocal form of the hole-impurity exchange interaction and the highly anisotropic band structure. However, despite the strong anisotropies the local-moment suppression is weak due to the averaging brought about by the long-range RKKY interaction.

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