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Exchange interaction in diluted magnetic semiconductors: Crystal-structure-induced anisotropy

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In this paper, we employ the model developed by Larson *et al.* to calculate the exchange interaction between magnetic ions in diluted magnetic semiconductors in a method that incorporates explicitly the intrinsic symmetry of the host crystals. We find that the symmetry of the wave function, and thus the interaction matrix element, could induce strong directional dependence of the exchange interaction despite the fact that superexchange is mediated via virtual excitations near the center of the Brillouin zone, where the band energy is essentially isotropic.

Recently, the magnetic properties of semiconductors containing a few percent of magnetic ions have received considerable attention.[1] For these diluted magnetic semiconductors (DMS's) that crystallize in the structure of the nonmagnetic semiconductor host (e.g., zinc-blende for $Cd_{1-x}Mn_xTe$), experimental results have shown evidence of antiferromagnetic interaction between Mn^{2+} local moments. In the high-temperature limit, the behavior of the magnetic susceptibility can be described rather well by the Curie-Weiss law with a negative paramagnetic Curie-Weiss temperature. At the early stage of theoretical research, the dominant exchange interaction in DMS's is attributed to the Bloembergen-Rowland mechanism.[2,3] The work of Larson *et al.*,[4,5] through detailed energy-band calculation, demonstrates the importance of p-d hybridization, and thus the dominance of Anderson superexchange in elaborated fourth-order perturbative formalism, which allows a unified treatment, and thus a direct comparison of Anderson superexchange and Bloembergen-Rowland mechanism. In evaluating the exchange constants, however, they use the matrix element and the band energy along Γ-X in the Brillouin zone (BZ) and then make the isotropic approximation. From our previous study,[6] it is concluded that the symmetry of the wave function, and thus the matrix element, might play an important role in the determination of the exchange interaction in DMS's. So, the purpose of this paper is to calculate the exchange interaction in a method which incorporates explicitly the symmetry of the wave function and the matrix element. We have decided to use the semiempirical linear-combination-of-atomic-orbitals (LCAO) method to simulate the band structure of the II-VI semiconductor host.[7] This method is chosen because, not only is the local symmetry of a state well represented by the atomic basis set, but, also, the crystal symmetry is reflected in LCAO band energy and wave functions. With the nonmagnetic band structure known, we calculate the exchange interaction between magnetic ions by the fourth-order perturbation theory. In evaluating the energy shift of the system, the double BZ integration is carried out by the linear analytic tetrahedron method.[8,9] With the sampling of finite energy bands near the energy-gap region as the only limitation, the variation of the symmetry of both band energy and wave functions in the realistic BZ are treated exactly.

In the LCAO formulation, the wave function of a state in the nth band is expressed as a linear combination of atomic orbitals,

$$|n\mathbf{k}\rangle=\sum_{\mu}C_{\mu}(n\mathbf{k})|\mu\mathbf{k}\rangle \ . \tag{1}$$

The LACO basis function $|\mu\mathbf{k}\rangle$ is a Bloch sum of atomic orbitals of local symmetry μ centered either at the cation (or anion) sites,

$$|\mu\mathbf{k}\rangle=N^{-1/2}\sum_{i}e^{i\mathbf{k}\cdot\mathbf{R}_i}|\mu(\mathbf{R}_i)\rangle \ . \tag{2}$$

The matrix representation of the crystal Hamiltonian H_0 with respect to LCAO basis functions (1) has been worked out by Slater and Koster[7] for various crystal structures. The energy of a band state and the mixing coefficients can therefore be obtained after diagonalizing the Hamiltonian matrix. There are eight energy integrals in the Hamiltonian matrix. We choose their values semiempirically to fit several measured optical and photoemission data. The fitted LCAO energy integrals are listed as follows: $E_{xx}(000)_{aa}=-1.146$, $E_{xx}(\frac{1}{2}\frac{1}{2}0)_{aa}=0.065$, $E_{xx}(0\frac{1}{2}\frac{1}{2})_{aa}=0.156$, $E_{xy}(\frac{1}{2}\frac{1}{2}0)_{aa}=0.070$, $E_{xy}(0\frac{1}{2}\frac{1}{2})_{aa}=-0.077$, $E_{xs}(\frac{1}{4}\frac{1}{4}\frac{1}{4})_{ac}=1.176$, $E_{ss}(000)_{cc}=0.754+1.60x$, $E_{ss}(\frac{1}{2}\frac{1}{2}0)_{cc}=0.073$ (all in eV). The Mn^{2+}-concentration dependence of the energy gap is taken into account by $E_{ss}(000)_{cc}$.

Following the model Hamiltonian proposed by Larson *et al.*,[5] we calculate the exchange interaction between magnetic ions in DMS's in the fourth-order perturbative formalism. This approach was developed previously by Falicov and co-workers in the investigation of the magnetic property of rare-earth compounds and NiO.[10,11] The model Hamiltonian to be used is written as

$$H=H_0+H' \ . \tag{3}$$

H_0 is the unperturbed crystal Hamiltonian. The perturbation H' consists of the correlation of localized Mn $3d$ electrons, the Mn $3d$–Te $5p$ hybridization (kinetic exchange), and the Mn $3d$-sp band potential exchange. If the spin-orbit interaction is neglected, the spin-$\frac{5}{2}$ Mn-Mn exchange in $Cd_{1-x}Mn_xTe$ is of the Heisenberg form,

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$$H_H=I(\mathbf{R}_{ij})\mathbf{S}_i\cdot\mathbf{S}_j\ , \tag{4}$$

with

$$I(\mathbf{R}_{ij})=-S^{-1}\times\sum_{I_1,I_2,I_3}\frac{\langle f|H'|I_3\rangle\langle I_3|H'|I_2\rangle\langle I_2|H'|I_1\rangle\langle I_1|H'|i\rangle}{(E_3-E_0)(E_2-E_0)(E_1-E_0)}\ , \tag{5}$$

where I_1, I_2, and I_3 label intermediate states; E_1, E_2, E_3, and E_0 are the energies of the intermediate states I_1, I_2, I_3, and the ground state, respectively. Depending on how the intermediate states permutate, there are three different cases to consider: hole-hole, hole-electron, and electron-electron processes. The corresponding exchange constants are denoted by $I_{hh}(\mathbf{R}_{ij})$, $I_{he}(\mathbf{R}_{ij})$, and $I_{ee}(\mathbf{R}_{ij})$. $I_{ee}(\mathbf{R}_{ij})$ would be identically equal to zero at $T=0$ K, and should be small at room temperature, and is thus neglected. By adding the contributions of all permutation of the intermediate states, $I_{hh}(\mathbf{R}_{ij})$ is then

$$\begin{aligned}I_{hh}(\mathbf{R}_{ij})=N^{-2}\sum_{(n\mathbf{k},n'\mathbf{k}')}&|M(n'\mathbf{k}')|^2|M(n\mathbf{k})|^2e^{-i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_{ij}}\\&\times\{2\Delta E^{-1}(n'\mathbf{k}')U^{-1}\Delta E^{-1}(n\mathbf{k})\\&\quad+\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})[\Delta E^{-1}(n'\mathbf{k}')\\&\quad+\Delta E^{-1}(n\mathbf{k})]^2\}+\text{c.c.},\end{aligned} \tag{6}$$

where

$$M(n\mathbf{k})=\sum_{\mu}C_{\mu}(n\mathbf{k})\sum_{\delta_j}e^{i\mathbf{k}\cdot\delta_j}V_{pd}\ ,$$

$$\Delta E^{-1}(n'\mathbf{k}')=[E_d+U-E(n'\mathbf{k}')]^{-1}\ ,$$

$$\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})=\{[E_d+U-E(n'\mathbf{k}')]+[E_d+U-E(n\mathbf{k})]\}^{-1}\ ,$$

and $\Delta E^{-1}(n\mathbf{k})=[E_d+U-E(n\mathbf{k})]^{-1}$. The double summation is carried out for the $n\mathbf{k},n'\mathbf{k}'$ pair, with n,n' restricted to the upper valence bands and $\mathbf{k},\mathbf{k}'$ over the first BZ. The definition of parameters V_{pd}, E_d, and U is the same as that of Ref. 5. $I_{he}(\mathbf{R}_{ij})$, which contains intermediate states involving the lowest conduction band and upper valence bands, can be similarly worked out, and the formula is of the same form as that of $I_{hh}(\mathbf{R}_{ij})$, but with different definition of the excitation energy: $\Delta E^{-1}(n'\mathbf{k}')=[E(n'\mathbf{k}')-E_d]^{-1}$, $\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})=[E(n'\mathbf{k}')-E(n\mathbf{k})]^{-1}$, and $\Delta E^{-1}(n\mathbf{k})=[E_d+U-E(n\mathbf{k})]^{-1}$. For computational convenience, the double summation of $\mathbf{k},\mathbf{k}'$ over the BZ is converted to the double integration via the following two equivalent versions of averaging over the BZ:

$$N^{-1}\sum_{\mathbf{k}}\leftrightarrow V_{\mathrm{BZ}}^{-1}\int_{\mathrm{BZ}}d\mathbf{k}, \tag{7}$$

where V_{BZ} is the volume of the first BZ.

In evaluating the double BZ integration, we use the linear analytic tetrahedron method.[8,9] In this method, the BZ is divided into microcells of tetrahedron geometry. When the integrand exhibits some functional form of a linear function, the contribution from one tetrahedron can be integrated analytically, and the result depends only on the values of the linear function at the vertices, not on the specific shape of the tetrahedron. The BZ integration thus reduces to the summation of the values of the analytically integrated function at the mesh points established in the BZ. In this problem, however, a double integration is required and the presence of the phase factor makes the situation more complicated. In the modified version of the linear analytic tetrahedron method, the double integration over the BZ is rewritten as the double summation of the double integration over pairs of tetrahedra with index t and t',

$$\begin{aligned}I_{hh}(\mathbf{R}_{ij})=V_{\mathrm{BZ}}^{-2}\sum_{n,n'\in v;t,t'\in\mathrm{BZ}}\int_t d\mathbf{k}\int_{t'}d\mathbf{k}'|M(n'\mathbf{k}')|^2\cos(\mathbf{k}'\cdot\mathbf{R}_{ij})\{2\Delta E^{-1}(n'\mathbf{k}')U^{-1}\Delta E^{-1}(n\mathbf{k})\\+\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})[\Delta E^{-1}(n'\mathbf{k}')+\Delta E^{-1}(n\mathbf{k})]^2\}|M(n\mathbf{k})|^2\cos(\mathbf{k}\cdot\mathbf{R}_{ij})+(\cos\rightarrow\sin)\ .\end{aligned} \tag{8}$$

The double integration over each t,t' tetrahedron pair is approximated by the product of the integration of the rapidly oscillating phase and the average values of other slowly varying terms, i.e., the matrix element and the inverse of excitation energy. The advantage of making such a separation is that the integration of the phase over the tetrahedron can be performed analytically. Since the sign of the matrix element and the inverse of excitation energy always remain the same throughout the BZ, this approximation is good even if $|\mathbf{R}_{ij}|$ is large, as long as the tetrahedra are small enough to account for the variation of the matrix element and the inverse of excitation energy in the whole BZ. Under this approximation,

$$\begin{aligned}I_{hh}(\mathbf{R}_{ij})\simeq V_{\mathrm{BZ}}^{-2}\sum_{n,n'\in v;t,t'\in\mathrm{BZ}}&|M(n't')|^2F(t')\{2\Delta E^{-1}(n't')U^{-1}\Delta E^{-1}(nt)+\Delta E^{-1}(n't',nt)[\Delta E^{-1}(n't')+\Delta E^{-1}(nt)]^2\}\\&\times|M(nt)|^2F(t)+(F\rightarrow G)\ ,\end{aligned} \tag{9}$$

where $M(nt)$ is the average of $M(n\mathbf{k})$ over the tetrahedron t, $F(t)$ and $G(t)$ are the integration of $\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$ and $\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$ over the tetrahedron t, $\Delta E^{-1}(nt)$ is the average of $\Delta E^{-1}(n\mathbf{k})$ over the tetrahedron t, and $\Delta E^{-1}(n't',nt)$ is the average of $\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})$ over the tetrahedron pair t,t'. The integration of $\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$ [or $\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$] over one tetrahedron is readily obtained via a linear transformation,[6] and the result depends only on the volume of the tetrahedron and the values of $\mathbf{k}\cdot\mathbf{R}_{ij}$ at the four vertices of the tetrahedron, as expected. The average of $\Delta E^{-1}(n\mathbf{k})$ over the tetrahedron t is, by definition,

$$\Delta E^{-1}(nt)=v_0^{-1}\int_t d\mathbf{k}\,\Delta E^{-1}(n\mathbf{k})\ , \tag{10}$$

and the integration can be carried out once the band energy is linearly interpolated inside the tetrahedron t.[9] The average of $\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})$ over the tetrahedron pair t,t' is, by definition,

$$\Delta E^{-1}(n't',nt)=v_0'^{-1}v_0^{-1}\int_{t'} d\mathbf{k}'\int_t d\mathbf{k}\,\Delta E^{-1}(n'\mathbf{k}',n\mathbf{k})\ , \tag{11}$$

and the integration can be similarly carried out. Since for the II-VI semiconductors under investigation the dispersion of the conduction band is smaller, it is sufficient to approximate the second integration by the average of the first integration at the four vertices of the tetrahedron t'. As for the average of the matrix element over one tetrahedron, we directly use the value at the center of the tetrahedron. Since $\Delta E^{-1}(n't')$, $\Delta E^{-1}(n't',nt)$, and $\Delta E^{-1}(nt)$ with t' and t taken from any of $*t'$ and $*t$ are all equal, we may first sum $|M(n't')|^2F(t')$ over $*t'$ and sum $|M(nt)|^2F(t)$ over $*t$, thus contracting the double summation of t,t' into the irreducible part of the BZ:

$$\begin{aligned}I_{hh}(\mathbf{R}_{ij})=V_{\mathrm{BZ}}^{-2}\sum_{\substack{n,n'\\ t,t'\in \text{irred. BZ}}}&\left[\sum_{t'\in *t'}|M(n't')|^2F(t')\right]\\ &\times\{\Delta E^{-1}(n't')U^{-1}\Delta E^{-1}(nt)\\ &+\Delta E^{-1}(n't',nt)[\Delta E^{-1}(n't')+\Delta E^{-1}(nt)]^2\}\\ &\times\left[\sum_{t\in *t}|M(nt)|^2F(t)\right]+(F\to G)\ ,\end{aligned} \tag{12}$$

where $*t$ indicates the set of tetrahedra derived from the tetrahedron t by applying on it the symmetry operations of the point group of the crystal. We would like to point out that it is crucial to preserve the symmetry of the matrix element while performing summation over $*t$ to contract the double integration over the BZ into over its irreducible parts. For example, if $|M(n\mathbf{k})|^2$ for $\mathbf{k}=(+k_x,+k_y,+k_z)$ is proportional to $|C_x+C_y+C_z|^2$, then $|M(n\mathbf{k})|^2$ for another symmetry related $\mathbf{k}=(+k_x,-k_y,-k_z)$ would be proportional to $|C_x-C_y-C_z|^2$. They may be quite different; the isotropic approximation, however, treats them as equal. It is seen that sphericalization of the matrix element would be improper, and may result in numerical inaccuracy. The double BZ integration program is tested in two ways via the following relation:

$$\int\int_{\mathrm{BZ}} d\mathbf{k}\,d\mathbf{k}'e^{-i(\mathbf{k}'-\mathbf{k})\cdot(\mathbf{R}_i-\mathbf{R}_j)}=V_{\mathrm{BZ}}^2\delta_{ij}\ . \tag{13}$$

First, the integration of (13) is identically equal to zero for any lattice vector $\mathbf{R}_{ij}=\mathbf{R}_i-\mathbf{R}_j$ except $\mathbf{R}_{ij}=0$. Second, if $\mathbf{R}_{ij}$ is replaced by some nonlattice vector $\mathbf{R}$, the integration is generally not equal to zero and should be independent of the total number of tetrahedra established in the BZ, since the integration formulas of $\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$ are exact. Calculated $I(\mathbf{R}_{ij})$ for $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ with a mesh of 73 728 tetrahedra established in the BZ (eight divisions along Γ-X) for $x=0.05$, 0.35, and 0.65 are listed in Table I.

Now, we compare our numerical results with experimental data. In the high-temperature limit, the magnetic susceptibility χ obeys the Curie-Weiss law

$$\chi^{-1}=C(T-\Theta)\ , \tag{14}$$

with a negative paramagnetic Curie-Weiss temperature Θ, which can be expressed in terms of the exchange coupling constant $I(\mathbf{R}_{ij})$ as

$$k_B\Theta=-\{[S(S+1)]/3N\}\sum_{i,j}I(\mathbf{R}_{ij})\ , \tag{15}$$

where N is the total number of magnetic ions and the summations of $\mathbf{R}_i$ and $\mathbf{R}_j$ run over the actual positions of magnetic ions. The experimental data of concentration-dependent $\Theta(x)$ for $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ can be fitted approximately as $\Theta(x)=\Theta_0x$.[12] This implies that a completely random distribution of Mn^{2+} ions in the host lattice is a reasonable assumption. Under this assumption, we get

$$\Theta_0=-\{[S(S+1)]/3k_B\}\sum_j I(\mathbf{R}_{ij})\ , \tag{16}$$

where the summation over $\mathbf{R}_j$ now runs over all the sites of a fcc lattice. The value of Θ_0 for $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ determined from experimental data is -470 ± 34 K,[12] which is close to the values of our calculation: -457, -461, and -464 K for $x=0.05$, 0.35, and 0.65, respectively.

From our results we see that $I_{hh}(\mathbf{R}_{ij})$, being always antiferromagnetic, mediates the dominant exchange interaction for the nearest neighbors; $I_{he}(\mathbf{R}_{ij})$, being ferromagnetic in the long range, decays more slowly than $I_{hh}(\mathbf{R}_{ij})$ and becomes dominant for $\mathbf{R}_{ij}$ beyond the nearest neighbors, which confirms the previous conclusion reached by Larson *et al.* However, there is a strong directional dependence in the obtained exchange constants. For example, the exchange constants for $\mathbf{R}_{ij}$ along [100] are much smaller than those for $\mathbf{R}_{ij}$ along other directions. As argued by Larson *et al.*,[5] the sphericalization procedure, which assumes $E(n\mathbf{k})$ and $M(n\mathbf{k})$ depend on $|\mathbf{k}|$ according to analytical expressions obtained along Γ-X in the BZ, is a reasonable approximation because the main contribution to $I(\mathbf{R}_{ij})$ arises from the central region of

TABLE I. Calculated exchange constants $I(\mathbf{R}_{ij})$ (in K) of $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ of the nearest four neighbors for $x=0.05$, 0.35, and 0.65.

$\mathbf{R}_{ij}$	$(\frac{1}{2},\frac{1}{2},0)R_0$	$(1,0,0)R_0$	$(\frac{1}{2},\frac{1}{2},1)R_0$	$(1,1,0)R_0$
		$x=0.05$		
$I_{hh}(\mathbf{R}_{ij})$	1.14×10^{1}	1.16×10^{-1}	5.72×10^{-1}	2.12×10^{-1}
$I_{he}(\mathbf{R}_{ij})$	8.77×10^{-1}	3.30×10^{-2}	-5.42×10^{-1}	-2.06×10^{-1}
$I(\mathbf{R}_{ij})$	1.23×10^{1}	1.49×10^{-1}	3.02×10^{-2}	6.02×10^{-2}
		$x=0.35$		
$I_{hh}(\mathbf{R}_{ij})$	1.16×10^{1}	1.26×10^{-1}	5.47×10^{-1}	2.01×10^{-1}
$I_{he}(\mathbf{R}_{ij})$	5.44×10^{-1}	3.68×10^{-3}	-4.52×10^{-1}	-1.72×10^{-1}
$I(\mathbf{R}_{ij})$	1.22×10^{1}	1.30×10^{-1}	9.50×10^{-2}	2.98×10^{-2}
		$x=0.65$		
$I_{hh}(\mathbf{R}_{ij})$	1.18×10^{1}	1.35×10^{-1}	5.21×10^{-1}	1.90×10^{-1}
$I_{he}(\mathbf{R}_{ij})$	3.19×10^{-1}	-1.31×10^{-2}	-3.73×10^{-1}	-1.40×10^{-1}
$I(\mathbf{R}_{ij})$	1.21×10^{1}	1.22×10^{-1}	1.48×10^{-1}	$4.97\times10^{10}{}^{-2}$

the BZ where $E(n\mathbf{k})$ and $M(n\mathbf{k})$ are isotropic. We would like to point out that, for the region near the center of the BZ, the band energy $E(n\mathbf{k})$ is essentially isotropic; the matrix element $M(n\mathbf{k})$, however, is no longer so no matter how small $|\mathbf{k}|$ is. In order to see why the strong directional dependence is the case, let us inspect more closely the integration in (12) [or (8)]. Since the LCAO basis functions $|x\mathbf{k}\rangle$, $|y\mathbf{k}\rangle$, and $|z\mathbf{k}\rangle$ transform like x, y, and z under the point group of the crystal, the matrix element $|M(n\mathbf{k})|^2$ for $\mathbf{k}=(\pm k_x,\pm k_y,\pm k_z)$ is proportional to $|\pm C_x \pm C_y \pm C_z|^2$ [refer to (2.1)]. Besides, for $\mathbf{R}_{ij}$ along [100], $\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$ for $\mathbf{k}=(\pm k_x,\pm k_y,\pm k_z)$ are all equal and $\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$ for $\mathbf{k}=(+k_x,\pm k_y,\pm k_z)$ differ by a sign from those for $\mathbf{k}=(-k_x,\pm k_y,\pm k_z)$. So, if we sum $|M(n\mathbf{k})|^2\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$ and $|M(n\mathbf{k})|^2\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$ over $*\mathbf{k}$, only $|C_\mu|^2\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$, with $\mu=x,y,z$ as well as $C_y^* C_z\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$ and $C_y C_z^*\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$ remain. If $\mathbf{R}_{ij}$ is along other directions, all 18 terms contribute. Since all these terms are of the same order of magnitude, the mutual cancellation between terms in $|M(n\mathbf{k})|^2\cos(\mathbf{k}\cdot\mathbf{R}_{ij})$ and $|M(n\mathbf{k})|^2\sin(\mathbf{k}\cdot\mathbf{R}_{ij})$ when $\mathbf{R}_{ij}$ along [100] makes the calculated $I(\mathbf{R}_{ij})$ much smaller. For example, I_2 obtained is one order of magnitude smaller than that by Larson *et al.* It is seen from the above qualitative argument that the spherical approximation of the matrix element, which neglects the interference between matrix elements of different symmetries, always overestimates $I(\mathbf{R}_{ij})$, and the overestimate is more pronounced for $\mathbf{R}_{ij}$ along [100]. So the result $I_2/I_1 \approx 0.11$, reported by Larson *et al.*, only provides an upper limit. Larson and Ehrenreich, in their subsequent paper,[13] include the spin-orbit interaction to treat the subject of anisotropic superexchange, and avoid the spherical approximation in the BZ summation by the special-points method of Baldereschi. They find that the result for I_1 is about 26% smaller, which confirms the qualitative argument given above. However, the value of I_2 is still not reported. We emphasize that the anisotropy treated here means the strong directional dependence of exchange constants of the isotropic Heisenberg interaction, and is conceptually different from the anisotropy usually referred to as departure from the $\mathbf{S}_i\cdot\mathbf{S}_j$ interaction form. So, the results presented here do not contradict the conclusion by Larson *et al.* that DM exchange is the dominant anisotropy interaction, nor the experimental data on electron-paramagnetic-resonance linewidth.

Escorne *et al.*[14] deduce from the measured x dependence of the spin-glass freezing temperature that $I_2/I_1 \approx 0.12$. In the fitting procedure, they assume an exponential decrease of $I(\mathbf{R}_{ij})$ with $|\mathbf{R}_{ij}|$ of the Bloembergen-Rowland mechanism, and equate thermal energy at the spin-glass freezing temperature with exchange energy at the mean distance between Mn^{2+} ions. Since Mn^{2+} ions are randomly distributed inside the semiconductor host and I_1 is much greater than I_2, the neglect of the formation of magnetic ion clusters would overestimate the mean distance between Mn^{2+} ions, and thus overestimate I_2. Thus, the result $I_2/I_1 \approx 0.12$ given also only provides an upper limit.

Giebultowicz *et al.*[15] study the static and dynamic spin correlation in $\mathrm{Cd_{0.35}Mn_{0.65}Te}$ by neutron scattering and by computer simulation. As they explained, in the type-III antiferromagnetic order of DMS's spins in (100) planes are aligned antiferromagnetically by I_1 whereas the between-plane coupling is maintained only by I_2. This implies that the ratio of the inverse correlation ranges should be sensitive to the ratio of I_2/I_1. By comparing computer-simulated results and measured correlation ranges, they obtain $I_2/I_1 \approx 0.12$. This also does not exclude the possibility of small I_2, since, in addition to I_2, I_3 and I_4 also contribute to interplane coupling, and, according to our calculation, they cannot be neglected (both I_3 and I_4 are of the same order of magnitude as I_2 for x being 0.65 and the number of the third and the fourth neighbors are larger than that of the second neighbors). If the effects of I_3 and I_4 are all lumped into I_2, the effective I_2 would be 0.813 K, which is close to the experimental results.

For DMS's of the zinc-blende structure, the nearest-neighbor lattice vector is along the [110] direction. If there is a lattice structure such that the nearest-neighbor lattice vector lies in the [100] direction, then the magnetic ordering predicted by a theory which takes into account the exact symmetry of the matrix element and one based on the isotropic approximation might be entirely different. We conclude that it is the symmetry of the matrix element, not the slightly anisotropic band energy, that induces the strong directional dependence of the exchange interaction between magnetic ions in DMS's. Serious calculation should preserve the symmetry of the matrix element in order that correct numerical results can be obtained.

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