Long-range superexchange: An exchange interaction through empty bands

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We derive a generalization of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction to semiconductors using perturbation theory on a nondegenerated two-impurity Anderson model. In metals the interaction is mediated by excitations of free carriers over the Fermi energy. In semiconductors, where no carriers are present, the only possible excitations are those of the localized impurity electrons (or holes) themselves. Thus a possible interaction is closely related to superexchange. We find an oscillating antiferromagnetic spin-spin coupling due to impurity electron (hole) excitations. By treating the coupling through empty bands (superexchange) along the same route as carrier-mediated interactions (RKKY), it is easy to compare these two kinds of spin-spin coupling. The interaction derived here is of special interest for diluted magnetic semiconductors.

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Usually superexchange is formulated within a cluster model consisting of three sites: two cation orbitals are partly filled, thus forming an effective spin moment, and one intermediate anion orbital is completely filled. In fourth-order perturbation theory the resulting spin-spin interaction between the cation sites reads (180° Mn-O-Mn)

$$J=-\frac{2V^4}{\Delta T_0^2}(U^{-1}+\Delta T_0^{-1}), \tag{1}$$

where ΔT_0 is the difference between the ground state and a configuration where one electron is transferred from the anion to the cation. U is the on-site Coulomb interaction at the cation, and V is the hybridization between both kinds of electrons.

However it is out of question whether the superexchange also has a long-range component. The latter is very important for diluted magnetic semiconductors, especially for doped (II,Mn)VI semiconductors. A competition between Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and superexchange is typical for these materials.[1–4] Nearest-neighbor superexchange leads to local-spin singlet that reduce the effective concentration of Mn spins. The superexchange between more distant pairs of Mn gives an antiferromagnetic coupling that competes with RKKY interaction as soon as free carriers are present, which may be generated by doping with N in (II,Mn)VI semiconductors or are present from the very beginning in (III,Mn)V semiconductors. To obtain a qualitative picture of the interplay between the different exchange interactions it is convenient to have some simple limiting expressions of the mechanisms at hand. These expressions should give an idea of the dependence of the exchange mechanisms on certain model parameters like, e.g., the intraatomic exchange coupling or the local Coulomb repulsion of Mn d electrons and the (sp)-d hybridization. For superexchange Eq. (1) shows these dependencies.

Concerning the long-range part of the interaction additional information is crucial, i.e., the dependence on the inter spin distance Δ and the dependence on the electronic band structure of the host material. Both properties cannot be deduced from Eq. (1) or from expressions derived from any other cluster model. It is the intention of this paper to derive an expression for the long-range component of superexchange, which is very comparable to the RKKY interaction. To this end we will study a toy model and adjust the parameters in such a way, that the so to say ''standard RKKY situation'' is recovered. That means two isolated spins should be located at a certain distance in a host material that is described by a nondegenerate uncorrelated band. For this limiting case we will then apply fourth-order degenerated perturbation theory.

The paper proceeds in the following way: First the most important indirect exchange mechanisms are briefly reviewed and discussed. We will concentrate on superexchange, RKKY, and Bloembergen-Rowland interactions, and a mechanism similar to the latter as well as to superexchange. Compact expressions for the last three coupling mechanisms are discussed. Then we will introduce the toy model and adjust the parameters in such a way that we reach the best comparability to the RKKY expression. In the next step we will derive an expression for superexchange that will be exact in fourth-order perturbation theory for the prepared model situation. Since the toy model establishes a well-defined limit for more complex calculations, this expression can be used as a check for certain approximations. For demonstration we will compare a work of Larson *et al.*[5] with our result. Furthermore the result should give a vivid idea of the distance and band structure dependence of superexchange. To this end we evaluate the superexchange expression numerically for some simple model lattices.

I. INDIRECT EXCHANGE MECHANISM

Indirect exchange mechanisms, i.e., effective spin-spin couplings usually between local spins at cation sites mediated by diamagnetic anions, were intensively discussed in the 1950s, e.g., by Anderson[6] and Goodenough and others.[7] The main goal of these studies was to understand magnetism in insulators such as MnO, and to justify the use of the Heisenberg model for this class of materials. These works were primarily concerned with the leading interaction of spins in adjacent lattice cells, and consequently many cluster models were adopted.

A different topic is the effective spin-spin interaction in metals. Here the interaction is mediated by free carriers. In the language of perturbation theory these carriers are virtually excited over the Fermi energy, which results in a spin-spin coupling that oscillates in sign and altitude in dependence of the interspin distance. Such an interaction is usually called RKKY coupling. It was first proposed by Ruderman and Kittel for nuclear spins,[8] and later generalized to electronic spins. It is often discussed, e.g., in heavy fermion systems. In contrast to indirect spin exchange in insulators RKKY interaction is formulated within a band picture, and its dependence on the spin-spin distance is well known.

In semiconductors both mechanisms, i.e., virtual excitations of carriers and noncarriers, may be important and even compete with each other. However for many materials the restriction to spins of neighboring lattice cells, typical for insulators, is no longer a good approximation. Let us mention Europium chalcogenides, where at least the next nearest neighbor cell is important or diluted magnetic semiconductors (DMS's). Therefore, a band formulation that is analogous to RKKY interaction is also desired for interactions caused by excitations of non-carriers, like, e.g., superexchange.

Such interaction types are widely discussed especially for semiconductors. Let us start with Bloembergen-Rowland[9] interaction, that is the band analog to the process described for clusters in Ref. 6 in Eqs. (29) and (30). A valence electron is virtually excited at site 1, and both the electron and the hole are transferred and recombine at site 2. The spin of the electron and the hole are coupled to local spins at site 1 and 2 by an intraatomic interorbital potential. This interaction is believed to be responsible for the magnetic interactions in Eu chalcogenides,[10] and is also discussed for DMS's.[11] A similar interaction [Eq. (23) of Ref. 6] was discussed by Litvinov and Dugaev for a (III,Mn)V DMS.[12] In the following we will call the this interaction ''impurity induced Bloembergen-Rowland interaction'' since the impurity electrons [Mn d electrons in a (III,Mn)V DMS] are virtually excited instead of valence electrons.

For all couplings discussed so far there is a ''standard expression'' usually derived in perturbation theory for some toy model that describes pure basic conditions for the respective interactions. For RKKY interaction this setup consists of two spins which are locally coupled to an uncorrelated partly filled electron band by an intraatomic spin-spin interaction J_{pd}. The same holds for an impurity-induced Bloembergen-Rowland interaction, but the ''spin'' is now described by partly filled localized electron orbitals and the electron band is empty instead of partly filled. The incomplete filling may be due to a strong on-site Coulomb repulsion. For the classical Bloembergen-Rowland interaction we again need two spins and a completely filled valence band as well as an empty conduction band. Again, the spin of the electrons are coupled to the local spins by an intraatomic interaction.

The resulting expressions for RKKY and Bloembergen-Rowland interaction in perturbation theory read (natural units)

$$J(\Delta)=-\frac{J_{pd}^2}{2N^2}\sum_{k,k'}\frac{\cos((k'-k)\Delta)}{\epsilon_{k'}-\epsilon_k} \tag{2}$$

for the RKKY interaction. The sum runs over all k's within the Fermi sphere and all k''s that are located outside. For parabolic bands for the Bloembergen-Rowland interaction one finds

$$J(\Delta)=-\frac{J_{pd}^2 m^2 \Delta T_0}{\pi^3 \Delta^2}K_2(2r/r_0)\quad r_0=(2m\Delta T_0)^{-1/2} \tag{3}$$

with the band gap ΔT_0, the interspin distance Δ and the reduced effective electron mass m. K_2 is the MacDonald function ($K_2 \sim 1/\Delta^2$ for $r \ll r_0$, $K_2 \sim \Delta^{-3/2}\, e^{2\Delta/r_0}$ for $r \gg r_0$). The same holds for the impurity induced Bloembergen-Rowland interaction, where ΔT_0 is now the energy difference between the impurity and the conduction band and m is now the effective electron mass in the conduction band.[12] In Sec. II we want to treat superexchange and derive a similar expression for this interaction.

II. MODEL DESCRIPTION FOR SUPEREXCHANGE

A model that can describe a ''pure'' version of long-range superexchange should be similar to the above-mentioned models, especially to the model for RKKY interaction. Thus the competition between the latter and superexchange can be studied. Our model consists of two impurity sites with an effective spin moment. This moment is due to partly filled localized orbitals, which is realized by a strong on-site Coulomb repulsion U at the impurity orbitals. Furthermore there is a free-electron band, described by the dispersion ϵ_k, that is energetically separated from the impurities by an energy ΔT_0. The chemical potential is located between the impurity orbital and the band. The latter is thus completely empty in the unperturbed ground state. The impurity orbitals and the band are kinetically coupled by a local hybridization V. The latter will constitute the perturbation in the following calculation. This is a minimal set to study superexchange. Therefore, other features, like e.g., intraorbital Coulomb exchange, are not taken into account.

The Hamiltonian for the described model reads

$$H=H_0+H_V,$$

$$H_0=\sum_{i\sigma}^{i=1,2} T_0^d n_{i\sigma}^d+\frac{U}{2}\sum_{i\sigma}^{i=1,2} n_{i\sigma}^d n_{i-\sigma}^d+\sum_k \epsilon_k^p n_{k\sigma}^p, \tag{4}$$

$$H_V=V\sum_{i\sigma}^{i=1,2}(d_{i\sigma}^{+}p_{i\sigma}+\text{H.c.}).$$

Let us note, that the construction operators can stand for holes or for electrons. If $d^\dagger$ and $p^\dagger$ create electrons, the situation is closest to the usual interpretation of RKKY interaction, i.e. that electrons are polarized by a local spin. For $S=1/2$ the model of Eqs. (4) may describe RKKY interaction as well as superexchange, if the band is partly filled instead of empty. In this case virtual excitations of band electrons

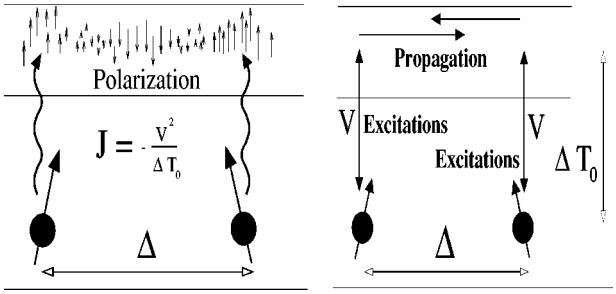

FIG. 1. Schematic picture of conventional RKKY and virtual RKKY (superexchange) interaction for a large on-site Coulomb repulsion $U\rightarrow\infty$.

over the Fermi energy contribute to the RKKY interaction, while virtual excitations of the impurity electrons lead to superexchange (see Fig. 1).

However, in most cases superexchange is constituted by virtual excitations of holes instead of electrons (e.g., in the ''classical'' case of MnO). Thus in most cases the construction operators have to be interpreted as hole creators and annihilators. T_0 and ϵ_k are now energies for holes, and U is the Coulomb repulsion between holes. The two interpretations of Eqs. (4) are connected via a particle-hole transformation

$$(p,d)_h^\dagger \rightarrow (p,d)_e \quad (p,d)_h \rightarrow (p,d)_e^\dagger, \tag{5}$$

with the well-known results

$$T_{0h}^d = -(T_{0e}^d + U_e),$$

$$\epsilon_{kh}^p = -\epsilon_{ke}^p,$$

$$U_h = U_e = U,$$

$$\mu_h = -\mu_e. \tag{6}$$

Let us discuss the following situation: The construction operators apply to holes, and thus the hole energies T_{0h}^d, ϵ_{kh}^p, and μ_h are fixed. Now let us consider the limit $U\rightarrow\infty$. In this case the one-electron energy of the impurities T_{0e}^d goes to $-\infty$ while the energy of a doubly occupied impurity orbital stays finite ($T_{0e}^d + U = -T_{0h}^d$). The same holds for the Bloch energies ϵ_{ke}^p. For superexchange we want to discuss the parameter constellation $T_{0h}^d < \mu_h < \epsilon_{kh}^p$ and $U\rightarrow\infty$ (see Fig. 1). For electrons this means

$$T_{0e}^d \ll \epsilon_{ke}^p < \mu_e < (T_{0e}^d + U). \tag{7}$$

Hence in the unperturbed ground state the impurity orbitals are filled with one electron (thus creating local spins). Furthermore, the band is completely filled. Possible excitations are from the band into the impurity orbital with an excitation energy

$$(T_{0e}^d + U) - \epsilon_{ke}^p = -T_{0h}^d + \epsilon_{kh}^p. \tag{8}$$

This describes a situation where superexchange is exclusively mediated by filled (valence) bands. Such a situation is not only found for the classical magnetic insulators (MnO), but also in (II,Mn)VI semiconductors.[5,13]

For (III,Mn)V semiconductors, too, the models of RKKY interaction and superexchange are well comparable if the construction operators apply to holes, since the important carriers are holes in these systems. However the simple model of Eqs. (4) is quite general and does not only apply to DMS's but to every situation, where virtual excitations of localized electrons are important.

Let us now derive an expression for long-range superexchange within this model. The most instructive way of considering the virtual processes leading to spin-spin interaction is perturbation theory, since one sums explicitly over all excited states. Treating the hybridization term as the perturbation we find that the free ground state is fourfold degenerated (with respect to the spin configuration). While calculating the energy corrections it is convenient to characterize the eigenstates of the free Hamiltonian H_0 by their number of impurity electrons, which is a good quantum number of the free system. Further the following property of the perturbation H_V should be considered: If H_V works on a free ground state it changes the number of conduction ($p-$) and impurity ($d-$) electrons (holes) by one (while the total number of electrons (holes) is conserved).

Due to this all odd energy corrections $E_a^{(1)} E_a^{(3)} \ldots$ vanish.[15] In second order we find an energy contribution, which does not affect the degeneracy of the ground state:

$$E_a^{(2)} = \frac{2}{N} \sum_k \frac{V^2}{T_0^d - \epsilon_k}. \tag{9}$$

The degeneracy is not broken until fourth-order perturbation theory, which gives an energy contribution $E_\alpha^{(4)}$. Besides a constant term, which does not affect the spin orientation, this is given by

$$E_a^{(4)} = \sum_{bcd} \frac{H_a^b \cdot H_b^c \cdot H_c^d \cdot H_d^a}{(E_n^{(0)} - E_m^{(0)})(E_n^{(0)} - E_l^{(0)})(E_n^{(0)} - E_o^{(0)})}, \tag{10}$$

where $H_x^y = \langle E_x^{(0)} | H_V | E_y^{(0)} \rangle$. $|E_a^{(0)}\rangle$ is one ground-state of the free system with the ground-state energy $E_n^{(0)} = 2T_0^d$. $|E_{\{b,c,d\}}\rangle$ are excited eigenstates of the free system with the energies $E_m^{(0)}$, $E_l^{(0)}$, and $E_o^{(0)}$. The sum goes over all excited states. Due to the special shape of the perturbation potential H_V the latter states must have a certain number of excited electrons (holes) to obtain a nonzero energy correction. Since we consider the limit $U\rightarrow\infty$, the excited electrons (holes) are in the conduction (valence) band. There is exactly one electron (hole) in the band in $|E_b^{(0)}\rangle$ and $|E_d^{(0)}\rangle$, and exactly two electrons (holes) are located within the band in $|E_c^{(0)}\rangle$. Thus $|E_{\{b,c,d\}}\rangle$ can be expressed in terms of construction operators working on states with two impurity electrons $|\alpha\sigma_x \beta\sigma_y\rangle$ ($\alpha,\beta = 1$ or 2; $\sigma = \uparrow$ or $\downarrow$):

$$|E_b^{(0)}\rangle = p_{k_1\sigma_1}^\dagger d_{i\sigma_1} |i\sigma_1 \alpha\sigma_x\rangle,$$

$$b = (k_1\sigma_1\alpha\sigma_x),$$

$$|E_c^{(0)}\rangle = p^\dagger_{k_2\sigma_3} p^\dagger_{k_3\sigma_4} d_{j\sigma_5} d_{l\sigma_6} |j\sigma_5 l\sigma_6\rangle, \tag{11}$$

$$c = (k_2\sigma_3 k_3\sigma_4),$$

$$|E_d^{(0)}\rangle = p^\dagger_{k_4\sigma_7} d_{m\sigma_8} |m\sigma_8 \beta\sigma_y\rangle,$$

$$d = (k_4\sigma_7 \beta\sigma_8).$$

After tedious but straightforward calculations one arrives at

$$E_a^{(4)} = \gamma \sum_{\substack{k_1 \ldots k_4 \\ o \ldots v \\ i \ldots m \\ \sigma_1 \ldots \sigma_8}} \frac{\langle E_a^{(0)}|XYZ|E_a^{(0)}\rangle e^{i\phi}}{(T_0^d - \epsilon^p_{k_1})(2T_0^d - \epsilon^p_{k_2} - \epsilon^p_{k_3})(T_0^d - \epsilon^p_{k_4})},$$

where $\gamma = 1/108N^4$, and

$$X = H_V p^+_{o\sigma_1}(1 - n^d_{i\sigma_2}) p_{p\sigma_1} H_V p^+_{q\sigma_3} p^+_{s\sigma_4},$$

$$Y = d_{j\sigma_5}(1 - n_{l\sigma_6}) d^+_{j\sigma_5}, \tag{12}$$

$$Z = p_{t\sigma_4} p_{r\sigma_3} H_V p^+_{u\sigma_7}(1 - n^d_{m\sigma_8}) p_{v\sigma_7} H_V.$$

In the sum the subscripts $i \ldots m$ denote impurity sites (1 or 2), while the subscripts $o \ldots v$ go over all lattice sites. The k summations are over the first Brillioun zone and $\sigma_1 \ldots \sigma_8$ are spin subscripts. The phase factor ϕ reads $\phi = \bar{k}_1(\bar{R}_o - \bar{R}_p) + \bar{k}_2(\bar{R}_q - \bar{R}_r) + \bar{k}_3(\bar{R}_s - \bar{R}_t) + \bar{k}_4(\bar{R}_u - \bar{R}_v)$. Performing the sum and introducing impurity-spin operators as usual,

$$S_i^z = \tfrac{1}{2}(n^d_{i\uparrow} - n^d_{i\downarrow}),$$

$$S_i^{(+/-)} = d^+_{i(\uparrow/\downarrow)} d_{i(\downarrow/\uparrow)},$$

we finally can write the energy contribution in terms of an effective Hamiltonian of Heisenberg-form that works on the free ground state $|E_\alpha^{(0)}\rangle$:

$$E_\alpha^{(4)} = \langle E_\alpha^{(0)}|H_{\text{eff}}|E_\alpha^{(0)}\rangle$$

with

$$H_{\text{eff}} = -J(\Delta)\bar{S}_1 \cdot \bar{S}_2. \tag{13}$$

For the exchange integrals $J(\Delta)$ (Δ is given in terms of the lattice constant) we find

$$J = \frac{8V^4}{N^4} \sum_{k_1 \ldots k_4} \frac{F(\Delta)}{(T_0^d - \epsilon^p_{k_1})(2T_0^d - \epsilon^p_{k_2} - \epsilon^p_{k_3})(T_0^d - \epsilon^p_{k_4})},$$

$$F(\Delta) = 2\cos[(k_2 - k_3)\Delta) + 4\cos((k_1 - k_2)\Delta] + \cos((k_1 - k_4)\Delta) + \cos[(k_1 + k_4 - k_2 - k_3)\Delta]. \tag{14}$$

This effective spin coupling is of the anticipated order $V^4/\Delta T_0^3$ for small distances Δ. Due to the fourfold sum in Eq. (14), we cannot give an analytical expression for the asymptotic behavior of $J(\Delta)$. However, since excitations over the band gap are necessary, we expect an exponential decay. In contrast to the ''classical superexchange'' where the particles fluctuate between the impurities and a single degenerated intermediate state, now the electrons may hop into different Bloch states and still cause an effective interaction.

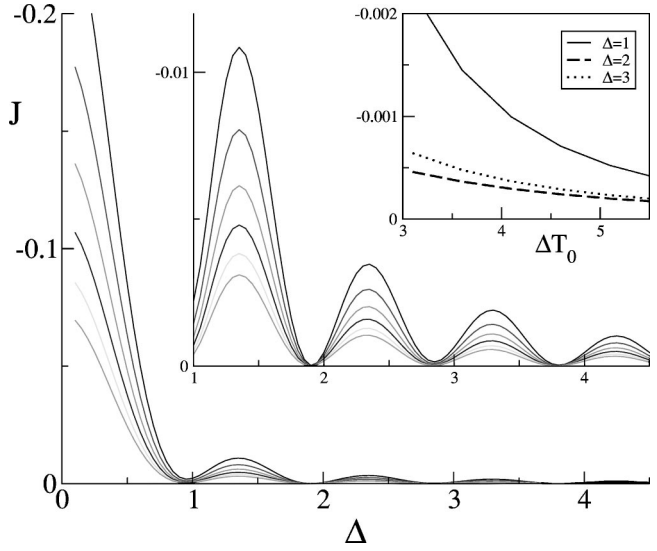

FIG. 2. Effective spin-spin coupling J as a function of the impurity distance Δ at different energy gaps. The distance is measured in units of the lattice constant. The band gaps $[\Delta T_0 - (W/2)]$ range from 0.1 to 2.6 eV (from top to bottom). The impurities are located along the [001] direction of a simple cubic tight-binding lattice. Other parameters: bandwidth $W = 6$ eV, $V = 0.16$ W. Inset: J as a function of the ΔT_0 at (001), (002), and (003).

For the zero-bandwidth limit, i.e., $\epsilon^p_k = T^p_0$ for all k, the k sum in Eq. (14) goes only over $F(\Delta)$. Since each cosine function now adds to zero, the interaction vanishes in this limit. This is the correct result, because the sites are completely decoupled in the zero-bandwidth limit. The numerical evaluation of Eq. (14) always gives an antiferromagnetic interaction that declines with the distance and shows certain oscillations (see Fig. 2). The interaction becomes even more important for systems with reduced dimensionality. For a two dimensional lattice the magnitude of the interaction increases approximately by a factor of 5. This is seen in Fig. 3, where we used the same parameters for the nearest-neighbor hoppings and the gaps between the band and the impurity-level as in Fig. 2.

Equation (14) gives an exact result in perturbation theory for a well-defined limit. Other treatments of long-range superexchange that may involve more complicated models but also some additional approximations can be compared in the limit $U_h \to \infty$ with Eq. (14).

Let us demonstrate this with an example in literature that treats (II,Mn)VI semiconductors. In Ref. 5 Larson *et al.* investigated electron- and hole-mediated superexchange and a special kind of Bloembergen-Rowland interaction (negative local J). They applied a multiband model with a realistic electronic structure, a local Coulomb repulsion U between Mn-3d electrons, and a hybridization between Mn ions and the host material. As explained in their paper, the five degen-

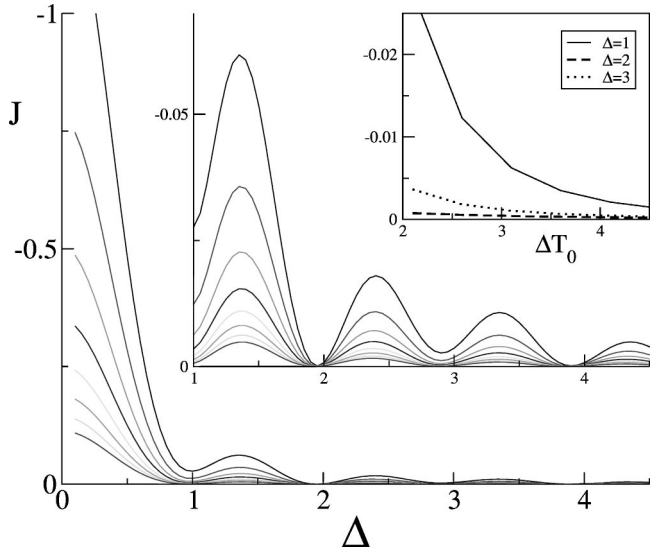

FIG. 3. As in Fig. 2, but for a two-dimensional quadratic lattice with a bandwidth $W=4$ eV, $V=0.25$ W.

erated Mn-*d* orbitals can be modeled by a single orbital plus a factor that depends on the Mn ground state only. Thus only a single orbital is considered at each Mn site. The authors found that the superexchange caused by virtual excitations of two holes dominate. After applying the limits $U\rightarrow\infty$, single nondegenerate valence band and local hybridization ($V(k)=V$) to their result, we want to compare them with Eq. (14). The result of Larson *et al.* [Eq. (4.4) of Ref. 5] is written with electronic parameters. To compare it with our result we have to perform a particle hole transformation [Eq. (6)] and apply the just mentioned limits and simplifications. Then the Mn-Mn exchange of Ref. 5 reduces to

$$J_{hh}^{dd}(\Delta)=2\sum_{kk'}\frac{V^4\cos(k-k')\Delta}{(T_0^d-\epsilon_k)^2(T_0^d-\epsilon_{k'})}. \tag{15}$$

This is quite close to the exact result in fourth-order perturbation theory [Eq. (14)]. The remaining discrepancies seem to be a fair price for the complexity of the model investigated in Ref. 5.

Finally let us discuss qualitatively the influence of free carriers on superexchange and RKKY interaction. If free carriers are doped into the band, the virtual excitations of this carriers over the Fermi energy lead to RKKY interaction. Since the energy gap is much smaller for these carriers the RKKY contribution should dominate in sum (10). Furthermore, since the band is now partly occupied there are less virtual intermediate states for superexchange. This gives a vivid explanation for the fact that superexchange is suppressed by free carriers as e.g. worked out by Qimiao Si *et al.* for CuO (Fig. 1 of Ref. 14).

In conclusion, we have derived a simple expression for long-range superexchange in a well-defined toy model. This expression is useful for qualitative discussions and constitutes a limit, which can be used to evaluate approximations in more complex models. We have given an example of one such comparison for the case of (II,Mn)VI semiconductors, where the long-range component of superexchange is very interesting. However, as in the case of RKKY interaction, the physical picture developed here is quite general and is applicable to all problems where virtual excitations of two electrons or of two holes lead to an effective spin-spin coupling between these electrons or holes.

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[15] This is most easily seen for the first-order energy contribution $\langle E_a^{(0)}|H_V|E_a^{(0)}\rangle$. There is the same state to the left and right of H_V. Since H_V changes the number of impurity electrons by 1, and free states with a different number of impurity electrons are orthogonal, this energy correction is zero.