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Ruderman–Kittel–Kasuya–Yosida interaction versus superexchange in a CuO_2 plane in the limit $U_d \to \infty$

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Abstract. The indirect exchange interaction between localized Cu spins via mobile O holes is derived from the three-band Anderson lattice model for copper oxides for infinite Hubbard repulsion U_d at Cu sites. By means of two nested canonical transformations, Ruderman–Kittel–Kasuya–Yosida (RKKY) and superexchange interactions are found in the fourth order of the Cu–O hybridization amplitude, where both O bands are consequently taken into account. For nearest neighbours the RKKY coupling J^{RKKY} and the superexchange integral $J^{sup\,ex}$, which increase with the direct O–O transfer, differ in sign and undergo a sign change upon doping. J^{RKKY} overcompensates $J^{sup\,ex}$.

1. Introduction

The competition between ferromagnetic and antiferromagnetic correlations in a doped CuO_2 layer of high-T_c superconductors can be ascribed to the interplay of a Ruderman–Kittel–Kasuya–Yosida-type (RKKY-type) interaction [1] and Anderson's superexchange [2]. In metallic copper oxides the indirect exchange between the localizable Cu (d) spins is mediated by the itinerant O (p) holes. A standard procedure for deriving an effective Cu–Cu spin coupling is to start from the Anderson lattice model, including a large or infinite Mott–Hubbard repulsion U_d at Cu sites, and to perform a Schrieffer–Wolff transformation [3] with the Cu–O hybridization amplitude t_{dp} as a small parameter. So the superexchange can be found in the fourth order of t_{dp} [4, 5]. On the other hand, the RKKY term can be obtained in the fourth order of t_{dp} by eliminating the Kondo lattice term of second order in t_{dp}; this perturbative origin was already described in [6, 7]. Both superexchange and RKKY exchange were deduced on the basis of the Anderson lattice model by means of slave-boson and path-integral approaches [8], a linked-cluster expansion [9], or two subsequent canonical transformations [10–13].

In this paper we propose two nested canonical transformations for calculating the superexchange and the RKKY exchange in the limit $U_d \to \infty$ for the CuO_2 plane under hole doping. Consequently, the band and hybridization dispersions arising from the two O bands are taken into account.

The Anderson lattice model is used to describe a single CuO_2 layer in the hole representation relative to a filled-shell ($3d^{10}$, $2p^6$) configuration. The corresponding three-band Hamiltonian [4, 5]

$$H = H_0 + H_1 \tag{1}$$

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is decomposed into the unperturbed part

$$H_0 = \epsilon_{\rm d} \sum_i n_i^{\rm d} + \sum_{\boldsymbol{k}\nu\sigma} \epsilon_{{\rm p}\boldsymbol{k}}^{\nu} p_{\boldsymbol{k}\sigma}^{\nu+} p_{\boldsymbol{k}\sigma}^{\nu} + U_{\rm d} \sum_i n_{i\uparrow}^{\rm d} n_{i\downarrow}^{\rm d} \tag{2}$$

with the O-band dispersion

$$\epsilon_{{\rm p}\boldsymbol{k}}^{\nu} = \epsilon_{\rm p} - 4t_{\rm pp}(-1)^{\nu} \sin\left(\frac{ak_x}{2}\right)\sin\left(\frac{ak_y}{2}\right) \tag{3}$$

and the perturbation

$$H_1 = \sum_{i\boldsymbol{k}\nu\sigma} (t_{i\boldsymbol{k}}^{\nu} d_{i\sigma}^{+} p_{\boldsymbol{k}\sigma}^{\nu} + t_{i\boldsymbol{k}}^{\nu *} p_{\boldsymbol{k}\sigma}^{\nu+} d_{i\sigma}) \tag{4}$$

in terms of the hybridization

$$t_{{\rm i}\boldsymbol{k}}^{\nu} = \frac{\sqrt{2}{\rm i}t_{\rm dp}}{\sqrt{N}} \left((-1)^{\nu} \sin\left(\frac{ak_x}{2}\right) + \sin\left(\frac{ak_y}{2}\right)\right) {\rm e}^{-{\rm i}ak_{\nu}/2} {\rm e}^{{\rm i}\boldsymbol{k}\cdot\boldsymbol{R}_i} = \frac{t_{\boldsymbol{k}}^{\nu}}{\sqrt{N}} {\rm e}^{{\rm i}\boldsymbol{k}\cdot\boldsymbol{R}_i}. \tag{5}$$

Within a mixed lattice–wavevector (i–$\boldsymbol{k}$) representation, the operator $d_{i\sigma}^{+}$ creates a hole with spin σ in the $3{\rm d}_{x^2-y^2}$ orbital at Cu site i and $n_i^{\rm d} = \sum_\sigma n_{i\sigma}^{\rm d} = \sum_\sigma d_{i\sigma}^{+} d_{i\sigma}$ is the corresponding number operator. The O-hole creation operator $p_{\boldsymbol{k}\sigma}^{\nu+}$ with the band index $\nu = 1, 2$ arises from the $2{\rm p}_x, 2{\rm p}_y$ orbitals in the unit cell. The parameters in (2) to (5) are the atomic energies $\epsilon_{\rm d}$ and $\epsilon_{\rm p}$ for d and p holes, the direct O–O hopping amplitude $t_{\rm pp}$, the Hubbard interaction strength $U_{\rm d}$ at a Cu site, and the Cu–O hybridization amplitude $t_{\rm dp}$. Moreover, a is the nearest-neighbour Cu–Cu distance, N the total number of Cu sites, $k_1 = k_x$, $k_2 = k_y$, and $\boldsymbol{R}_i$ denotes the position vector of the ith Cu ion.

2. Perturbative treatment of the Anderson lattice model

In view of the decomposition (1) we treat, by assuming $t_{\rm dp} \ll \epsilon_{\rm p} - \epsilon_{\rm d} \ll U_{\rm d}$, the hybridization term (4) perturbatively. This assumption of a charge-transfer insulator situation is justified in principle for the CuO_2 planes in the high-$T_{\rm c}$ superconductors. Band-structure calculations (e.g. in [14]) show that for the parent cuprate compounds the parameter inequalities $t_{\rm dp} < \epsilon_{\rm p} - \epsilon_{\rm d} < U_{\rm d}$ are valid.

Let us perform two unitary transformations nested by the definition

$$H'' = {\rm e}^{-S_2} H' {\rm e}^{S_2} = {\rm e}^{-S_2} {\rm e}^{-S_1} H {\rm e}^{S_1} {\rm e}^{S_2}. \tag{6}$$

The first generator S_1 leading to H' is determined by the condition

$$H_1 + [H_0, S_1] = 0 \tag{7}$$

in the sense of a Schrieffer–Wolff transformation [3]. Decomposing the second-order result for H' as

$$H_2' = \frac{1}{2}[H_1, S_1] = H_{2{\rm a}}' + H_{2{\rm b}}' \tag{8}$$

we quote (cf. [5]) the following expressions for $U_{\rm d} \to \infty$:

$$H_{2{\rm a}}' = -\frac{1}{N} \sum_{i\boldsymbol{k}\nu} \frac{|t_{\boldsymbol{k}}^{\nu}|^2}{\epsilon_{\boldsymbol{k}}^{\nu}} n_i^{\rm d} + \frac{1}{2} \sum_{\boldsymbol{k}\nu\nu'\sigma} t_{\boldsymbol{k}}^{\nu *} t_{\boldsymbol{k}}^{\nu'} \left(\frac{1}{\epsilon_{\boldsymbol{k}}^{\nu}} + \frac{1}{\epsilon_{\boldsymbol{k}}^{\nu'}}\right) p_{\boldsymbol{k}\sigma}^{\nu+} p_{\boldsymbol{k}\sigma}^{\nu'}$$
$$- \frac{1}{4} \sum_{i\boldsymbol{k}\boldsymbol{k}'\nu\nu'\sigma} t_{i\boldsymbol{k}}^{\nu *} t_{i\boldsymbol{k}'}^{\nu'} \left(\frac{1}{\epsilon_{\boldsymbol{k}}^{\nu}} + \frac{1}{\epsilon_{\boldsymbol{k}'}^{\nu'}}\right) n_i^{\rm d} p_{\boldsymbol{k}\sigma}^{\nu+} p_{\boldsymbol{k}'\sigma}^{\nu'} - \sum_{\substack{ij\boldsymbol{k}\nu\sigma \\ (i\neq j)}} \frac{t_{i\boldsymbol{k}}^{\nu} t_{j\boldsymbol{k}}^{\nu *}}{\epsilon_{\boldsymbol{k}}^{\nu}} d_{i\sigma}^{+} d_{j\sigma}$$

$$+\frac{1}{2}\sum_{ij\boldsymbol{k}\nu\sigma}\frac{t^{\nu}_{i\boldsymbol{k}}t^{\nu *}_{j\boldsymbol{k}}}{\epsilon^{\nu}_{\boldsymbol{k}}}d^{+}_{i\sigma}d_{j\sigma}(n^{\mathrm{d}}_{i,-\sigma}+n^{\mathrm{d}}_{j,-\sigma})$$

$$+\frac{1}{2}\sum_{i\boldsymbol{k}\boldsymbol{k}'\nu\nu'\sigma}\frac{1}{\epsilon^{\nu}_{\boldsymbol{k}}}\left(t^{\nu}_{i\boldsymbol{k}}t^{\nu'}_{i\boldsymbol{k}'}d^{+}_{i\sigma}d^{+}_{i,-\sigma}p^{\nu}_{\boldsymbol{k},-\sigma}p^{\nu'}_{\boldsymbol{k}'\sigma}+t^{\nu *}_{i\boldsymbol{k}}t^{\nu' *}_{i\boldsymbol{k}'}p^{\nu'+}_{\boldsymbol{k}'\sigma}p^{\nu+}_{\boldsymbol{k},-\sigma}d_{i,-\sigma}d_{i\sigma}\right) \tag{9a}$$

$$H'_{2\mathrm{b}}=H_2^{\mathrm{Kondo}}=\sum_{i\boldsymbol{k}\boldsymbol{k}'\nu\nu'}t^{\nu *}_{i\boldsymbol{k}}t^{\nu'}_{i\boldsymbol{k}'}\left(\frac{1}{\epsilon^{\nu}_{\boldsymbol{k}}}+\frac{1}{\epsilon^{\nu'}_{\boldsymbol{k}'}}\right)\boldsymbol{S}_i\cdot\boldsymbol{s}^{\nu\nu'}_{\boldsymbol{k}\boldsymbol{k}'} \tag{9b}$$

with

$$\epsilon^{\nu}_{\boldsymbol{k}}=\epsilon^{\nu}_{\mathrm{p}\boldsymbol{k}}-\epsilon_{\mathrm{d}}>0. \tag{10}$$

In the Kondo lattice part (9*b*), $\boldsymbol{S}_i=\frac{1}{2}\sum_{\sigma\sigma'}d^{+}_{i\sigma}\boldsymbol{\sigma}_{\sigma\sigma'}d_{i\sigma'}$ ($\boldsymbol{\sigma}$ being the vector of the Pauli matrices) denotes the localized Cu-spin operator, whereas $\boldsymbol{s}^{\nu\nu'}_{\boldsymbol{k}\boldsymbol{k}'}=\frac{1}{2}\sum_{\sigma\sigma'}p^{\nu+}_{\boldsymbol{k}\sigma}\boldsymbol{\sigma}_{\sigma\sigma'}p^{\nu'}_{\boldsymbol{k}'\sigma'}$ involves the itinerant O spin. In order to remove the Kondo term we choose the second generator S_2 from (6) to satisfy

$$H_2^{\mathrm{Kondo}}+[H_0,S_2]=0. \tag{11}$$

Now we are going over from H'' to an effective Hamiltonian

$$H_{\mathrm{eff}}=H_{0\mathrm{p}}+H_{2\mathrm{p}}+H_{4\mathrm{I}}+H_{4\mathrm{II}} \tag{12}$$

which is (i) truncated at order of t^4_{dp} and (ii) projected onto the subspace of single Cu-hole occupancy, i.e. $n^{\mathrm{d}}_i=1$, according to the limit $U_{\mathrm{d}}\to\infty$. The contributions in (12) are given, on the basis of (2) and (6) to (11), by

$$H_{0\mathrm{p}}=\sum_{\boldsymbol{k}\nu\sigma}\epsilon^{\nu}_{\mathrm{p}\boldsymbol{k}}p^{\nu+}_{\boldsymbol{k}\sigma}p^{\nu}_{\boldsymbol{k}\sigma} \tag{13}$$

$$H_{2\mathrm{p}}=\frac{1}{4}\sum_{\boldsymbol{k}\nu\nu'\sigma}t^{\nu *}_{\boldsymbol{k}}t^{\nu'}_{\boldsymbol{k}}\left(\frac{1}{\epsilon^{\nu}_{\boldsymbol{k}}}+\frac{1}{\epsilon^{\nu'}_{\boldsymbol{k}}}\right)p^{\nu+}_{\boldsymbol{k}\sigma}p^{\nu'}_{\boldsymbol{k}\sigma} \tag{14}$$

$$H_{4\mathrm{I}}=\frac{1}{8}\left[[[H_1,S_1],S_1],S_1\right]\Big|_{n^{\mathrm{d}}_i=1} \tag{15a}$$

$$H_{4\mathrm{II}}=\frac{1}{2}\left[([H_1,S_1]-H_2^{\mathrm{Kondo}}),S_2\right]\Big|_{n^{\mathrm{d}}_i=1} \tag{15b}$$

where the term of third order in t_{dp} disappears due to the constraint $n^{\mathrm{d}}_i=1$. For clarification, the projection onto the subspace $n^{\mathrm{d}}_i=1$ is carried out only at the end of the calculation, i.e. after the expansion of H'' up to the order of t^4_{dp}. This means, in particular, that the terms of third order in t_{dp}, which emerge alone from the first transformation H', are projected out after having generated the fourth-order contributions of the triple commutator. From (15) we retain only such fourth-order contributions as survive under the projection (ii).

3. Extraction of the indirect exchange interactions

The resultant H_{eff}, ascribed to localized Cu spins and mobile O holes, involves spin–spin, spin–spin–hole, spin–hole, spin–hole–hole, and hole–hole interactions. Among a lot of terms inherent in (15), we extract here only the indirect exchange interaction of the Heisenberg model form

$$H_4^{\mathrm{ind\,ex}}=\sum_{\substack{ij\\(i\neq j)}}J_{ij}\boldsymbol{S}_i\cdot\boldsymbol{S}_j \tag{16}$$

with the total exchange integral

$$J_{ij} = J_{ij}^{\text{sup ex}} + J_{ij}^{\text{RKKY}} \tag{17}$$

decomposed into the superexchange and the RKKY parts. An expression of the type (16) can be reached from the spin–spin–hole operator $\boldsymbol{S}_i \cdot \boldsymbol{S}_j n_{\boldsymbol{kq}}^{\nu\mu}$ by averaging ($\langle\cdots\rangle_0$) its p charge-density part $n_{\boldsymbol{kq}}^{\nu\mu} = \sum_\sigma p_{\boldsymbol{k}\sigma}^{\nu+} p_{\boldsymbol{q}\sigma}^{\mu}$ in a mean-field approximation with respect to $H_{0\text{p}}$ from (13), i.e. replacing $n_{\boldsymbol{kq}}^{\nu\mu}$ by $\langle n_{\boldsymbol{kq}}^{\nu\mu}\rangle_0 = \langle n_{\boldsymbol{kk}}^{\nu\nu}\rangle_0 \delta_{\boldsymbol{kq}}\delta_{\nu\mu}$.

For completeness, all the contributions arising from (15*a*) were given explicitly in [5] even at finite U_d. For $H_{4\text{II}}$ in (15*b*) we find now the same spin–spin–hole operator combinations as are listed in formula (23*d*) of [5]. The Heisenberg form of the exchange between the Cu spins is in (15) always accompanied by Dzyaloshinsky–Moriya-like interactions proportional to $2\text{i}(\boldsymbol{S}_i \times \boldsymbol{S}_j) \cdot \boldsymbol{s}_{\boldsymbol{kq}}^{\nu\mu}$. However, such terms tend to zero under the mean-field approximation mentioned above, namely in the sense of $2\text{i}(\boldsymbol{S}_i \times \boldsymbol{S}_j) \cdot \langle \boldsymbol{s}_{\boldsymbol{kq}}^{\nu\mu}\rangle_0$.

More explicitly, we derive from (15*a*) the superexchange and from (15*b*) the RKKY exchange integrals. Our result is

$$\left.\begin{matrix} J_{ij}^{\text{sup ex}} \\ J_{ij}^{\text{RKKY}} \\ J_{ij} \end{matrix}\right\} = \frac{1}{4N^2} \sum_{\boldsymbol{kq}\nu\mu} |t_{\boldsymbol{k}}^{\nu}|^2 |t_{\boldsymbol{q}}^{\mu}|^2 \cos\left[(\boldsymbol{k}-\boldsymbol{q})\cdot(\boldsymbol{R}_i - \boldsymbol{R}_j)\right]$$

$$\times \begin{cases} \left(\dfrac{3}{\epsilon_{\boldsymbol{k}}^{\nu}} + \dfrac{1}{\epsilon_{\boldsymbol{q}}^{\mu}}\right) \dfrac{(2 - \langle n_{\boldsymbol{kk}}^{\nu\nu}\rangle_0)}{\epsilon_{\boldsymbol{k}}^{\nu}\epsilon_{\boldsymbol{q}}^{\mu}} & (18a) \\ \left(\dfrac{1}{\epsilon_{\boldsymbol{k}}^{\nu}} + \dfrac{1}{\epsilon_{\boldsymbol{q}}^{\mu}}\right)^2 \dfrac{\langle n_{\boldsymbol{kk}}^{\nu\nu}\rangle_0}{\epsilon_{\boldsymbol{k}}^{\nu} - \epsilon_{\boldsymbol{q}}^{\mu}} & (18b) \\ \dfrac{4}{(\epsilon_{\boldsymbol{k}}^{\nu})^2 \epsilon_{\boldsymbol{q}}^{\mu}} \left(2 + \dfrac{\epsilon_{\boldsymbol{q}}^{\mu} \langle n_{\boldsymbol{kk}}^{\nu\nu}\rangle_0}{\epsilon_{\boldsymbol{k}}^{\nu} - \epsilon_{\boldsymbol{q}}^{\mu}}\right). & (18c) \end{cases}$$

Note that (18*a*) had already been derived in [4, 5]. It is pointed out that $J_{ij}^{\text{sup ex}}$ as well as J_{ij}^{RKKY} include the doping dependence via the occupation number $\langle n_{\boldsymbol{kk}}^{\nu\nu}\rangle_0$. The doping rate δ, i.e. the mean number of O holes per unit cell, is defined by

$$\delta = \frac{1}{N} \sum_{\boldsymbol{k}\nu} \langle n_{\boldsymbol{kk}}^{\nu\nu}\rangle_0 = \frac{2}{N} \sum_{\boldsymbol{k}\nu} \Theta(\epsilon_\text{F} - \epsilon_{\text{p}\boldsymbol{k}}^{\nu}) \tag{19}$$

where ϵ_F denotes the Fermi energy related to (13); note that $\delta_\text{max} = 4$. The temperature is assumed to be zero, hereafter.

To evaluate the indirect exchange couplings we have to insert (3), (5), (10), and (19) into (18). The procedure for exploiting (18) is outlined here as follows. (i) Consider the case where $\epsilon_\text{F} < \epsilon_\text{p}$ and put $\epsilon_\text{d} = 0$ as the origin of energy; (ii) replace the summations over the first Brillouin zones by integrations; (iii) perform the summations over the band indices by using symmetry relations; (iv) reduce the integrations to the first quadrants of the Brillouin zones. Then we get straightforwardly, e.g. from (18*b*),

$$J_{\boldsymbol{R}}^{\text{RKKY}} = -\frac{8t_{\text{dp}}^4}{\pi^4 t_{\text{pp}}} \left\{ \int_{\tilde{k}_x}^{\pi/2} \text{d}k_x \int_{\tilde{k}_y(k_x)}^{\pi/2} \text{d}k_y \int_0^{\pi/2} \text{d}q_x \int_0^{\pi/2} \text{d}q_y \;\; \cos(2k_x R_x)\cos(2k_y R_y) \right.$$

$$\left. \times \;\; \cos(2q_x R_x)\cos(2q_y R_y) \frac{(\sin k_x + \sin k_y)^2 (\sin q_x - \sin q_y)^2}{\sin k_x \sin k_y + \sin q_x \sin q_y} \right.$$

$$\times \left(\frac{1}{\epsilon_p - 4t_{pp} \sin k_x \sin k_y} + \frac{1}{\epsilon_p + 4t_{pp} \sin q_x \sin q_y} \right)^2$$
$$+ \int_{\tilde{k}_x}^{\pi/2} dk_x \int_{\tilde{k}_y(k_x)}^{\pi/2} dk_y \int_0^{\pi/2} dq_x \int_0^{\tilde{q}_y(q_x)} dq_y \ \cos(2k_x R_x) \cos(2k_y R_y)$$
$$\times \ \cos(2q_x R_x) \cos(2q_y R_y) \frac{(\sin k_x + \sin k_y)^2 (\sin q_x + \sin q_y)^2}{\sin k_x \sin k_y - \sin q_x \sin q_y}$$
$$\times \left(\frac{1}{\epsilon_p - 4t_{pp} \sin k_x \sin k_y} + \frac{1}{\epsilon_p - 4t_{pp} \sin q_x \sin q_y} \right)^2 \Bigg\} \qquad (20)$$

expressed in terms of the Fermi boundary line

$$\tilde{k}_y(k_x) = \begin{cases} \dfrac{\pi}{2} & 0 \leqslant k_x \leqslant \sin^{-1}(-\tilde{\epsilon}_F) = \tilde{k}_x \\ \sin^{-1}\left(-\dfrac{\tilde{\epsilon}_F}{\sin k_x} \right) & \tilde{k}_x \leqslant k_x \leqslant \dfrac{\pi}{2} \end{cases}$$

with $\tilde{\epsilon}_F = (\epsilon_F - \epsilon_p)/4t_{pp} < 0$. The range is measured as $\boldsymbol{R} = (\boldsymbol{R}_i - \boldsymbol{R}_j)/a$.

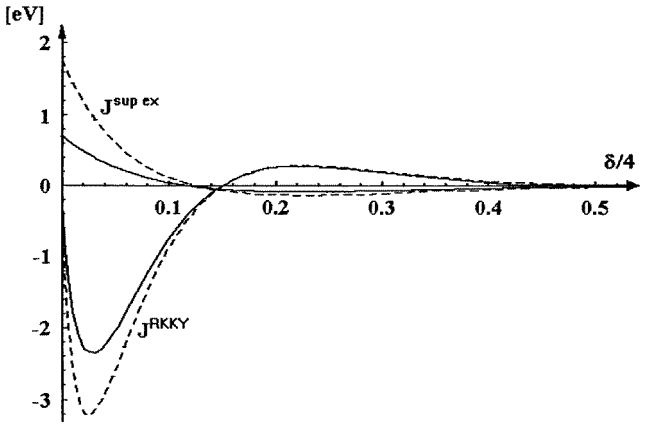

Figure 1. The dependence of the nearest-neighbour superexchange $J^{\text{sup ex}}$ and the RKKY interaction strength J^{RKKY} on the p-hole concentration $\delta/4$ for the parameters $t_{pp} = 0.4$ eV (——) and $t_{pp} = 0.55$ eV (- - -) at $\epsilon_p - \epsilon_d = 3.5$ eV, $t_{dp} = 1.3$ eV, and $U_d = \infty$.

In figure 1 the nearest-neighbour (i.e. $|\boldsymbol{R}_i - \boldsymbol{R}_j| = a$) exchange integrals $J^{\text{sup ex}}$ and J^{RKKY} resulting numerically from (18) and (20) are presented as functions of the O-hole doping δ defined in (19). The set of parameters ($\epsilon_p - \epsilon_d$, t_{dp}, t_{pp}, U_d) = (3.5, 1.3, 0.4 or 0.55, ∞) eV chosen here is similar (except the U_d-value) to that used, e.g., in [14] for studying $La_{2-x}Sr_xCuO_4$. The couplings $J^{\text{sup ex}}$ and J^{RKKY} are doping sensitive, and differ in sign and also in magnitude. The superexchange shows in figure 1 an unexpected sign variation upon doping. The source of this behaviour must be the non-trivial topology of the Fermi boundary line crossing both O bands. Numerical test calculations using only a single, symmetric band always yield $J^{\text{sup ex}} > 0$.

In figure 1 the RKKY exchange overcompensates the superexchange. The total nearest-neighbour exchange $J = J^{\text{sup ex}} + J^{\text{RKKY}}$ from (18*c*) behaves antiferromagnetically, ferromagnetically, and again antiferromagnetically with increasing δ and finally vanishes. Generally, our numerical example confirms the sign change of J^{RKKY} in [8] and that both

$J^{\sup ex}$ and J^{RKKY} increase with increasing oxygen transfer [11]. For comparison, there is no sign change of $J^{\sup ex}$ in [8]. Moreover, $J^{\sup ex}$ in [10, 13] is not renormalized by t_{pp} and is assumed to be independent of δ. It should be mentioned that we consequently used two O bands, in contrast to making calculations with a simple parabolic band [8, 10], a single O band [11, 13], or an additional apical-O ($2p_z$) orbital [12].

To conclude, we examined here only the magnetic correlations between the Cu spins in the normal state of cuprate superconductors. The present scheme implies the constraint $n_i^d = 1$, i.e. there is exactly one hole per Cu site. But the case where $n_i^d \neq 1$ is also of interest. Recent analysis of the electric field gradients in high-T_c superconductors [15] shows that the holes introduced by doping are nearly equally shared among the Cu and O ions. Considering the complete collection of the 64 operator products in (15*a*), without the prescription $n_i^d = 1$ one cannot extract Cu–Cu spin combinations weighted by n_i^d. Indeed, indirect exchange interactions are not eliminated by projection.

Note that a derivation of an effective Heisenberg-type spin interaction between localized magnetic moments via fourth-order perturbation theory in the hybridization (with conduction electrons) has been carried out in the literature also for several other (non-cuprate) systems. Work along these lines was done, e.g. to get RKKY exchanges and superexchanges from one unified approach for rare-earth compounds [16], to calculate the exchange parameters for transition-metal monoxides [17], or to determine corrections to the RKKY interaction in spin-glass systems [18].

A detailed analysis of the complete spin–hole Hamiltonian generated by the two nested unitary transformations, also at finite U_d, will be presented elsewhere [19].

References

[1] Ruderman M A and Kittel C 1954 *Phys. Rev.* **96** 99
Kasuya T 1956 *Prog. Theor. Phys. (Kyoto)* **16** 45
Yosida K 1957 *Phys. Rev.* **106** 893
[2] Anderson P W 1950 *Phys. Rev.* **79** 350
Anderson P W 1959 *Phys. Rev.* **115** 2
[3] Schrieffer J R and Wolff P A 1966 *Phys. Rev.* **149** 491
[4] Ihle D and Kasner M 1990 *Phys. Rev.* B **42** 4760
[5] Kolley E, Kolley W and Tietz R 1994 *Phys. Status Solidi* b **186** 239
[6] Proetto C and López A 1981 *Phys. Rev.* B **24** 3031
[7] Long M W 1988 *Z. Phys.* B **71** 23
[8] Si Q, Lu J P and Levin K 1989 *Physica* C **162–164** 1467
Si Q, Lu J P and Levin K 1992 *Phys. Rev.* B **45** 4930
[9] Rodriguez-Nuñez J J and Beck H 1993 *J. Phys.: Condens. Matter* **5** L613
[10] Rodriguez-Nuñez J J, Coqblin B, Beck H and Konior J 1994 *Acta Phys. Pol.* A **85** 317
[11] Tran Minh-Tien 1994 *Physica* C **223** 361
[12] Eid Kh, Matlak M and Zieliński J 1995 *Phys. Status Solidi* b **187** 589
[13] Rodriguez-Nuñez J J, Beck H, Konior J, Oleś A M and Coqblin B 1995 *Phys. Lett.* **197A** 173
[14] Hybertsen M S, Schlüter M and Christensen N E 1989 *Phys. Rev.* B **39** 9028
[15] Kupčiś I, Barišić S and Tutiš E 1996 *J. Physique* **6** 2291
[16] Gonçalves da Silva C E T and Falicov L M 1972 *J. Phys. C: Solid State Phys.* **5** 63
[17] Koiller B and Falicov L M 1975 *J. Phys. C: Solid State Phys.* **8** 695
[18] Zhang Q and Levy P M 1986 *Phys. Rev.* B **34** 1884
[19] Kolley E, Kolley W and Tietz R 1998 *Phys. Status Solidi* b at press