

# Contribution of the Non-Heisenberg Ring Exchange to the Magnetic Mechanism of High- $T_c$ Superconductivity

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The effect of non-Heisenberg four-spin ring exchange on the superconducting transition temperature in strongly correlated electron systems is studied. It is shown that the ring exchange can significantly suppress the contribution of the Heisenberg exchange to superconducting pairing.

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In high- $T_c$  superconducting cuprates, the four-spin ring exchange in the  $\text{CuO}_2$  plane is only a factor of 3–5 weaker than the Heisenberg exchange [1–14]. Therefore, we face a natural question, namely, how significant is its effect on the Cooper pairing mediated by the magnetic interactions. Let us recall the source of the ring exchange in the theory for spin 1/2. It is well known that the copper oxide-based high- $T_c$  superconductors belong to the class of strongly correlated electron systems. The simplest model for such systems is the Hubbard model [15]. It describes the splitting of the initial electron band into two Hubbard subbands. The Hamiltonian for this model contains only two parameters: the hopping integral  $t$  between the nearest sites and the on-site Coulomb repulsion energy  $U$ . Unfortunately, in spite of the apparent simplicity of the model, its properties are well understood only in the case of a one-dimensional chain [16]. In the limit of strong electron correlations ( $t \ll U$ ), the Hubbard model can be simplified [17–20] by excluding the interband transitions according to perturbation theory [21]. The corresponding effective low-energy Hamiltonian is then treated in the reduced Hilbert space without the two-particle states. Thus, in the transformed Hamiltonian, we exclude the initial local Coulomb interaction. However, instead of it, an infinite series describing the long-range exchange interactions appears. In the simplest case of the Heisenberg exchange limit  $t/U \rightarrow 0$ , only the  $t$ - $J$  model is considered [17, 18]. It contains only the first correction to the ground state energy of the unreduced Hamiltonian

$$H_{t-J} = t \sum_{i \neq j, \sigma} \tilde{a}_{i\sigma}^+ \tilde{a}_{j\sigma} + \sum_{\langle ij \rangle} J_{ij} \left( \mathbf{S}_i \mathbf{S}_j - \frac{1}{4} n_i n_j \right). \quad (1)$$

Here,  $\tilde{a}_{i\sigma}^+ = a_{i\sigma}^+ (1 - n_{i\bar{\sigma}})$  is the projection operator for the creation of an electron with spin  $\sigma$  at site  $i$ , where

$n_{i\sigma} = \tilde{a}_{i\sigma}^+ \tilde{a}_{i\sigma}$ ;  $\mathbf{S}_i \equiv (S_i^+, S_i^-, S_i^z) = (a_{i\sigma}^+ a_{i\bar{\sigma}}, a_{i\bar{\sigma}}^+ a_{i\sigma}, (n_{i\sigma} - n_{i\bar{\sigma}})/2)$  is the spin operator, where  $\bar{\sigma} = -\sigma$ ; and  $J_{ij} = 2t_{ij}^2/U$  is the effective exchange integral for the nearest neighbors. In addition to the Heisenberg exchange  $J$ , the same order of perturbation theory gives three-site terms  $H_{(3)}$  proportional to  $t_{ij}^2 t_{jk}/U$ . The latter describe the processes involving the electron hopping between three sites with the spin flip or with the conserved spin projection at the intermediate site. In contrast to the  $t$ - $J$  model given by Eq. (1), these contributions qualitatively change the quasiparticle spectrum in the normal phase [22, 23] and significantly suppress the critical temperature for the superconducting phase [24, 25]. Therefore, they should be taken into account in a consistent formulation of the theory.

In the range of intermediate correlations, where the electron mobility grows, it is necessary to take into account the higher order corrections to Hamiltonian (1), namely, those proportional to  $\sim t(t/U)^3$ . As was shown in [18, 20, 26], these corrections (accurate to constant terms) have the form

$$H_{(4)} = J_1 \sum_{\langle ij_1 \rangle} \mathbf{S}_i \mathbf{S}_{j_1} + J_2 \sum_{\langle ij_2 \rangle} \mathbf{S}_i \mathbf{S}_{j_2} + J_3 \sum_{\langle ij_3 \rangle} \mathbf{S}_i \mathbf{S}_{j_3} \\ + J_c \sum_{\langle ijkl \rangle} \{ (\mathbf{S}_i \mathbf{S}_j)(\mathbf{S}_k \mathbf{S}_l) + (\mathbf{S}_i \mathbf{S}_l)(\mathbf{S}_k \mathbf{S}_j) \\ - (\mathbf{S}_i \mathbf{S}_k)(\mathbf{S}_j \mathbf{S}_l) \}. \quad (2)$$

Subscripts  $j_1, j_2$ , and  $j_3$  enumerate the first, second, and third neighbors of site  $i$ , respectively. Sites  $i, j, k$ , and  $l$  correspond to four spins circulating clockwise within the unit  $\text{Cu}_4\text{O}_4$  square plaquette. The first three terms



$$H_{(4)} = J_c \sum_{\substack{\langle ijkl \rangle \\ \sigma\sigma'}} (\tilde{H}_{ij\sigma}^J \tilde{H}_{kl\sigma'}^J + \tilde{H}_{il\sigma}^J \tilde{H}_{kj\sigma'}^J - \tilde{H}_{ik\sigma}^J \tilde{H}_{jl\sigma'}^J).$$

Here,  $\tilde{t}_{ij}$  and  $t_{ij}$  are the intra- and interband hopping integrals, respectively, and  $\mu$  is the chemical potential.

For brevity, we introduced the notation  $H_{ij,\sigma}^J = \frac{1}{2} J_{ij} (X_i^{\sigma\bar{\sigma}} X_j^{\bar{\sigma}\sigma} - X_i^{\sigma\sigma} X_j^{\bar{\sigma}\bar{\sigma}})$  and  $\tilde{H}_{ij,\sigma}^J = H_{ij,\sigma}^J + \frac{1}{4} n_i n_j$ , where  $n_i = X_i^{\sigma\sigma} + X_i^{\bar{\sigma}\bar{\sigma}}$ .

The normal  $G(\mathbf{k}, E)$  and anomalous  $F(\mathbf{k}, E)$  Green's functions were determined using the generalized Hartree–Fock approximation in the framework of the irreducible linear operator technique [31, 32]. It is well known that the short-range antiferromagnetic order does not vanish up to the optimum doping and significantly affects the formation of the quasiparticle-related characteristics within both the low doping and optimum doping ranges [33]. At the same time, the spin dynamics at low temperatures is much slower than the electron one [34–36]. Therefore, in the self-energy term, we take into account only static spin correlation functions; i.e., we neglect the dynamics of the short-range antiferromagnetic order but consider its spatial inhomogeneity,  $\Sigma(\mathbf{k}, \omega) \rightarrow \Sigma(\mathbf{k})$ . For the  $t$ – $J^*$  model, the calculation procedure, the explicit form of the self-energy term, the approximation used, and the obtained results are described in detail in [37, 38]. The averages arising as the contributions of the non-Heisenberg ring exchange in the course of the decoupling of the equations for the Green's functions were simplified by separating the two-operator Green's function  $c_{\mathbf{q}} = \sum_i \langle \mathbf{S}_i \mathbf{S}_j \rangle \exp[-i(R_i - R_j)\mathbf{q}]$  from the product of four spin operators. We assume the isotropy of the spin liquid; hence, we have  $c_{ij} = 3 \langle S_i^z S_j^z \rangle = \frac{3}{2} \langle X_i^{\sigma\bar{\sigma}} X_j^{\bar{\sigma}\sigma} \rangle$ . In the normal phase, the corrections coming from the non-Heisenberg exchange  $H_{(4)}$  are proportional to the product  $J_c(\mathbf{k}, \mathbf{q}) c_{\mathbf{k}} c_{\mathbf{q}}$ . At the characteristic values of the ring exchange parameter,  $J_c \sim 0.3J$ , and of the nearest-neighbor spin function,  $c_{\mathbf{k}} \sim -0.3$ , these corrections are as small as several percent of the band width and do not modify the form of the dispersion law and the density of states. The main contribution of the ring exchange manifests itself in the renormalization of the coupling constant for the superconducting pairing in the equation for  $T_c$ . Deriving the latter equation, we took into account that the order parameter for the symmetrical solution corresponding to the singlet pairing has the following property:  $\Delta_{\mathbf{k}} = \Delta_{-\mathbf{k}}$ . In view of Eq. (2), the Fourier transform of the Heisenberg exchange parameter includes the neighbors up to the third coordination sphere. However, in the final equation for  $T_c$ , the contribution

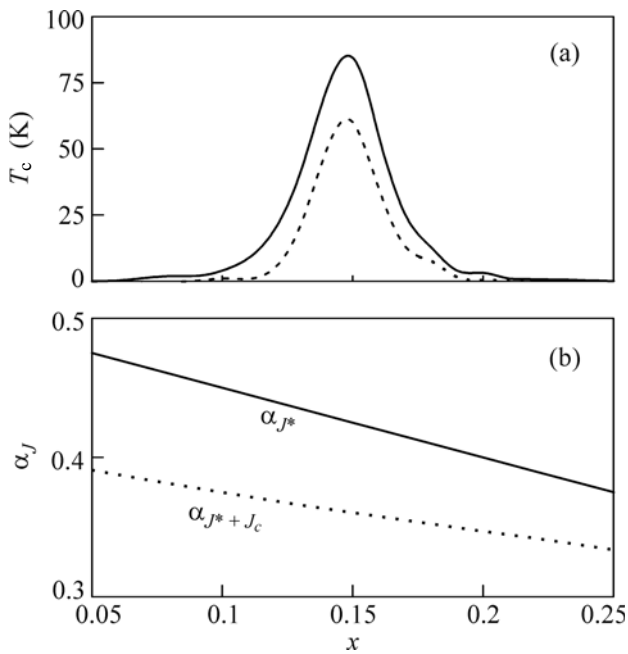
coming from the second neighbors disappears since it does not meet the  $d_{x^2-y^2}$  symmetry of the gap. We omit the contribution coming from the third neighbors generating the next harmonic of the gap because it is negligibly small. Eventually, for the BCS-type theory, the equation for the superconducting transition temperature can be represented in a form similar to that for the  $t$ – $J$  model

$$1 = \tilde{J} \frac{1}{N} \sum_{\mathbf{p}} \frac{(\cos p_x - \cos p_y)^2}{\xi_{\mathbf{p}}} \tanh \frac{\xi_{\mathbf{p}}}{2k_B T_c}. \quad (5)$$

Here,  $\xi_{\mathbf{p}}$  is the dispersion law for the quasiparticles in the superconducting phase and  $k_B$  is the Boltzmann constant. We separate the contributions from the Heisenberg exchange, three-site hopping, and ring exchange in the renormalized coupling constant:  $\tilde{J} =$

$(1 - \frac{1+x}{2} + c_{01} \frac{J_c}{J}) J$ . The antiferromagnetic nearest-neighbor spin correlation function has the negative sign. Therefore, the ring exchange, as well as the three-site hopping, suppresses the superconducting pairing induced by the Heisenberg exchange. The above expression demonstrates that the contribution of the ring exchange to the coupling constant is an order of magnitude smaller than that of the Heisenberg exchange. However, under a consistent formulation of the theory, namely, taking into account the three-site interactions, its relative role increases. At the values of parameters characteristic of the LSCO system [39] and  $J_c = (0.27-0.3)J$ , the ring exchange leads to the lowering of the maximum of the critical temperature in the  $t$ – $J$  model by only 5–7%. At the same time, the maximum critical temperature in the  $t$ – $J^*$ – $J_c$  model is 25–30% lower than that for the  $t$ – $J^*$  model (see Fig. 1a). The corresponding changes in the coupling constant in units of the Heisenberg exchange are illustrated in Fig. 1b. The ring exchange is most clearly pronounced in the range of low doping  $x$ , where it suppresses superconductivity. With growth of  $x$ , its effect decreases owing to the decay of the short-range magnetic order.

Another well-known mechanism compensating the contribution of the Heisenberg exchange to the Cooper pairing is the intersite Coulomb interaction. On the one hand, the intersite Coulomb interaction, as well as the exchange one, is proportional to the squared overlap of the orbitals of the neighboring atoms. This means that it has the same order of magnitude and can appreciably suppress superconductivity induced by the Heisenberg exchange [40, 41]. On the other hand, it is not clear from the very beginning that the intersite Coulomb interaction should also compensate the contribution of the ring exchange. Moreover, based on the structure of the Hamiltonian for the ring exchange, it is possible to suggest that the situation is just the opposite [42]. Then, the ring



Effect of the ring exchange on (a) the concentration dependence of the superconducting transition temperature and (b) the pairing parameter. The inclusion of the ring exchange  $J_c = 0.27J$  (dashed line) leads to the lowering of the maximum  $T_c$  value by 25%.

exchange can play even a more significant role than that following from the above estimates.

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