Exchange interactions and anisotropic spin waves in bilayer manganites

Andrzej M. Oleś[1,2] and Louis Felix Feiner[3,4]
[1]*Marian Smoluchowski Institute of Physics, Jagellonian University, Reymonta 4, PL-30059 Kraków, Poland*
[2]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*
[3]*Institute for Theoretical Physics, Utrecht University, NL-3584 CE Utrecht, The Netherlands*
[4]*Philips Research Laboratories, Prof. Holstlaan 4, NL-5656 AA Eindhoven, The Netherlands*
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We present a theory of the magnetic interactions in the bilayer manganites, based upon the manganite *t-J* model, and show that the ferromagnetic phase is an *anisotropic orbital liquid* disordered state. By including the correlations of the e_g electrons we explain why the *intralayer* exchange coupling increases with hole doping x. The transition from the ferromagnetic to *A*-type antiferromagnetic phase in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ at $x\simeq 0.45$ occurs when the occupancy of $3z^2-r^2$ orbitals drops and the superexchange term exceeds the double exchange in the *interlayer* coupling.

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The theoretical understanding of doped cubic manganites $La_{1-x}A_xMnO_3$, with $A=$ Sr, Ca, Pb, is among the most challenging current areas of research in condensed-matter physics. Experimental studies have revealed their fascinating physical properties and rich phase diagrams, originating from the competition between charge, spin, and orbital degrees of freedom.[1,2] The most spectacular are changes in resistivity by several orders of magnitude when applying a magnetic field, known as "colossal magnetoresistance." The basic concept is the double exchange (DE),[3] which explains why the e_g electrons, which couple strongly to the t_{2g} spins by Hund's exchange, induce a ferromagnetic (FM) metallic state in doped systems.

In all manganites strong local Coulomb interactions suppress charge fluctuations and allow only virtual transitions to excited states. These generate the superexchange (SE) interactions, which involve both the spin and orbital degrees of freedom (due to the degeneracy of the e_g orbitals, see Ref. 1), and contain both antiferromagnetic (AF) and FM terms. Although the SE is highly frustrated and quantum effects are enhanced,[4] such frustration is often removed when a combination of the SE and the Jahn-Teller (JT) effect induces a structural phase transition and stabilizes a symmetry-broken phase with orbital ordering. This occurs in undoped $LaMnO_3$,[5,6] but the cubic symmetry is restored in the highly doped metallic FM phase, when the orbital ordered state is replaced by a disordered *orbital liquid*.[7]

Recently, the spin-wave excitations in the FM bilayer manganites $La_{2-2x}A_{1+2x}Mn_2O_7$ were reported by several groups.[8–10] As the coupling between neighboring bilayers is weaker by two orders of magnitude,[9] one should be able to understand the microscopic origin of the exchange couplings by considering a single bilayer. Here the cubic symmetry is explicitly broken, and the kinetic energy favors the population of $|x\rangle\equiv|x^2-y^2\rangle$ over $|z\rangle\equiv|3z^2-y^2\rangle$ orbitals,[11,12] as for a single layer.[13] It is puzzling why the anisotropy between the intralayer and interlayer exchange coupling increases so fast with increasing hole doping x within the narrow concentration range $0.3<x<0.45$.[9,10] Thereby, the intralayer exchange $J_a(=J_b)$ *gets stronger* with x, while the interlayer J_c *decreases* rapidly and changes sign at $x\simeq 0.45$.

In this paper we formulate the theory of magnetic interactions in the FM bilayer manganites and explain their microscopic origin, showing that the orbital degrees of freedom play a fundamental role. While one might expect that the orbital ordering stabilized by the SE could be tuned by doping,[11] we will demonstrate that a *disordered* (i.e., orbital liquid) state with strong x^2-y^2 orbital polarization is realized instead.

A manganite bilayer consists of two square-lattice (a,b) planes connected by interlayer bonds along the c axis. In this geometry the (unrenormalized) kinetic energy $\propto t$ of the (as yet noninteracting) itinerant e_g electrons, including a crystal-field splitting $\propto E_z$, is given by

$$H_t=-\tfrac{1}{4}t\sum_{\langle ij\rangle_\parallel,\sigma}[3d^\dagger_{ix\sigma}d_{jx\sigma}+d^\dagger_{iz\sigma}d_{jz\sigma} \pm\sqrt{3}(d^\dagger_{ix\sigma}d_{jz\sigma}+d^\dagger_{iz\sigma}d_{jx\sigma})] -t\sum_{\langle ij\rangle_\perp,\sigma}d^\dagger_{iz\sigma}d_{jz\sigma}+\tfrac{1}{2}E_z\sum_{i\sigma}(n_{iz\sigma}-n_{ix\sigma}), \quad (1)$$

where $n_{i\alpha\sigma}=d^\dagger_{i\alpha\sigma}d_{i\alpha\sigma}$, and $\{d^\dagger_{ix\sigma},d^\dagger_{iz\sigma}\}$ are fermion creation operators.[14] The splitting E_z between $|x\rangle$ and $|z\rangle$ orbitals originates from the JT distortions of the MnO_6 octahedra and leads to the tetragonal symmetry.[15]

The large intraorbital on-site Coulomb interaction $U\simeq 5.9$ eV causes the e_g electrons to be strongly correlated, while the Hund's coupling $J_H\simeq 0.7$ eV enforces maximum total (e_g plus t^3_{2g}) spin on every site.[16] Presuming that therefore each manganese ion is either Mn^{3+} in the ^5E state or Mn^{4+} in the $^4A_2$ state we derive the bilayer *t-J* model, $\mathcal{H}_{tJ}=\tilde{H}_t+H_J$. This contains a spin-and-orbital SE term H_J, discussed in more detail below, while the kinetic energy $\tilde{H}_t$ is restricted to the subspace without multiply occupied (e_g^2, e_g^3, e_g^4) configurations and obeying Hund's rule for the total spin at each site. Formally $\tilde{H}_t$, including the crystal-field term $\propto E_z$, assumes exactly the same form as in Eq. (1) but with $d^\dagger_{i\alpha\sigma}$ replaced by the *projected* fermion operator $\tilde{d}^\dagger_{i\alpha\sigma}$, the projection excluding multiple occupancy and enforcing

Hund's rule, i.e., allowing only the ^5E state of Mn^{3+} and excluding the ^3E state. The spin restriction is taken care of by defining $\tilde{d}^\dagger_{i\alpha\sigma}=a^\dagger_{i\sigma}\tilde{c}^\dagger_{i\alpha}$, where $a^\dagger_{i\sigma}$ creates a Schwinger boson with spin σ, while the remaining (projected) operator with orbital flavor $\tilde{c}^\dagger_{i\alpha}=c^\dagger_{i\alpha}(1-n_{i\bar{\alpha}})$ adds a fermion in orbital α at an empty site i. The number of Schwinger bosons per site, $\Sigma_\sigma a^\dagger_{i\sigma}a_{i\sigma}=2S$, should then fluctuate only between four for Mn^{3+} $(S=2)$ and three for Mn^{4+} $(S=3/2)$, i.e., $2S=3+\Sigma_\alpha \tilde{c}^\dagger_{i\alpha}\tilde{c}_{i\alpha}$. Here we will use the average (mean-field) constraint, $\Sigma_\sigma a^\dagger_{i\sigma}a_{i\sigma}=2\mathcal{S}$, where $2\mathcal{S}=4-x$ at hole density x.

To describe the FM phase of the bilayer manganites we expand $\tilde{H}_t$, where the hopping terms are now of the form $\sim a^\dagger_{i\sigma}\tilde{c}^\dagger_{i\alpha}\tilde{c}_{j\beta}a_{j\sigma}$, around the FM ground state:

$$\frac{1}{2\mathcal{S}}\sum_\sigma a^\dagger_{i\sigma}a_{j\sigma}\simeq 1-\frac{1}{4\mathcal{S}}(a^\dagger_{i\downarrow}a_{i\downarrow}+a^\dagger_{j\downarrow}a_{j\downarrow}-2a^\dagger_{i\downarrow}a_{j\downarrow}). \quad (2)$$

The lowest (zeroth) order in $1/\mathcal{S}$ then gives the ground-state kinetic energy, $\tilde{H}^{(0)}_t$. At this point it is convenient to switch orbital basis, and use, instead of the real orbitals $\{|x\rangle,|z\rangle\}$, the *complex orbitals*,[17] defined by $c^\dagger_{i\pm}=(1/\sqrt{2})(c^\dagger_{iz}\mp ic^\dagger_{ix})$. In this representation,

$$\tilde{H}^{(0)}_t=-\frac{1}{2}t\sum_{\langle ij\rangle}[\tilde{c}^\dagger_{i+}\tilde{c}_{j+}+\tilde{c}^\dagger_{i-}\tilde{c}_{j-}+e^{+i\chi_\alpha}\tilde{c}^\dagger_{i+}\tilde{c}_{j-}+e^{-i\chi_\alpha}\tilde{c}^\dagger_{i-}\tilde{c}_{j+}]+\frac{1}{2}E_z\sum_i(\tilde{c}^\dagger_{i+}\tilde{c}_{j-}+\tilde{c}^\dagger_{i-}\tilde{c}_{j+}), \quad (3)$$

where $\tilde{c}^\dagger_{i\pm}=c^\dagger_{i\pm}(1-n_{i\mp})$. The phase factors χ_α follow from the orbital phases in Eq. (1) and depend on the bond direction: $\chi_{a,b}=\pm 2\pi/3$ and $\chi_c=0$.

We implemented the constraint of no double occupancy in $\tilde{H}^{(0)}_t$ by representing, following Kotliar and Ruckenstein[18] (KR), the occupied e_g orbital by a fermion $(f^\dagger_{i\pm})$ and a slave-boson $(b^\dagger_{i\pm})$ operator, $|\pm\rangle_i=\tilde{c}^\dagger_{i\pm}|0\rangle=f^\dagger_{i\pm}b^\dagger_{i\pm}|\text{vac}\rangle$, and the empty site by a slave-boson $(e^\dagger_i)$ operator describing a hole, $|0\rangle_i=e^\dagger_i|\text{vac}\rangle$, leading in Eq. (3) to the replacement $\tilde{c}^\dagger_{i\pm}=f^\dagger_{i\pm}b^\dagger_{i\pm}e_i$, with the local constraint $b^\dagger_{i+}b_{i+}+b^\dagger_{i-}b_{i-}+e^\dagger_i e_i=1$. One can now solve the electronic problem within the slave-boson functional-integral method of KR,[18] adapted here for the orbital degrees of freedom,[7] while the use of the complex-orbital representation guarantees that one would retain the cubic symmetry in the limit of a cubic system at $E_z=0$.[19] With all sites assumed equivalent, mean-field approximation now leads to

$$\tilde{H}^{(0)}_{t,\text{MF}}=-\frac{1}{2}t\sum_{ij}[q_+f^\dagger_{i+}f_{j+}+q_-f^\dagger_{i-}f_{j-}+\sqrt{q_+q_-}(e^{-i\chi_\alpha}f^\dagger_{i+}f_{j-}+e^{+i\chi_\alpha}f^\dagger_{i-}f_{j+})]+\frac{1}{2}E_z\sum_i(f^\dagger_{i+}f_{j-}+f^\dagger_{i-}f_{j+}), \quad (4)$$

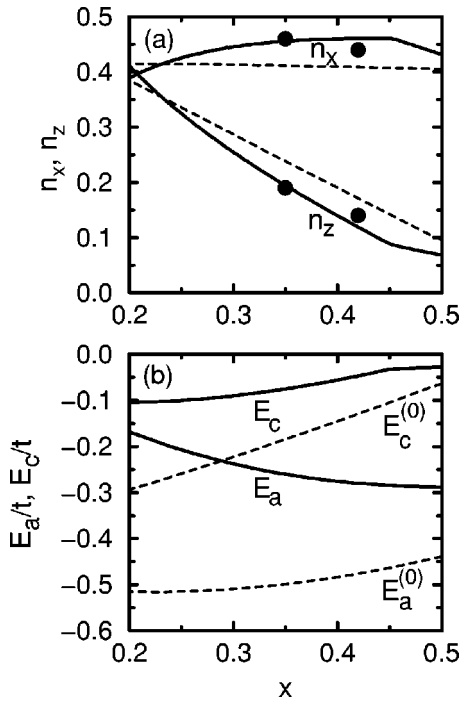

FIG. 1. Electron densities n_x and n_z (a), and kinetic energies E_a and E_c (b), as functions of hole doping x, obtained for correlated (full lines) and uncorrelated (dashed lines) e_g electrons with $E_z=-0.9t+4xt$ and $t=0.48$ eV. The experimental values of Ref. 12 are shown by filled circles in (a).

with the kinetic energy reduced by Gutzwiller factors, $\sqrt{q_\pm}=[x/(1-\langle n_{i\pm}\rangle)]^{1/2}$, given by $\langle n_{i\pm}\rangle=\langle f^\dagger_{i\pm}f_{i\pm}\rangle$.

From Eq. (4) we obtained the fermionic bands and calculated (by summing over the occupied band states) the kinetic energy at fixed E_z of the orbital-disordered state, and compared it with that of several states with orbital order [as for instance uniform real ($|x\rangle$ or $|z\rangle$) or complex ($|+\rangle$) orbitals], which can be enforced by introducing Lagrange multipliers in Eq. (4) and taking an appropriate limit. The ground state is given by *disordered* and equally populated *complex* orbitals, $n_\pm=\langle n_{i\pm}\rangle=\frac{1}{2}(1-x)$, similar to the orbital liquid in the cubic manganites.[7] However, the fermion densities in the *real* orbitals, $n_x=\langle f^\dagger_{ix}f_{ix}\rangle$ and $n_z=\langle f^\dagger_{iz}f_{iz}\rangle$, are now different. The anisotropy in the electron distribution, $\delta n=n_x-n_z$, increases with x due to the kinetic energy that favors the x^2-y^2 orbitals lying in the (a,b) planes. Actually, $\delta n>0$ is found even when $E_z=0$, similar as for a single plane,[13] but the JT effect enhances the anisotropy. Using the experimental information that the JT distortion changes linearly with x (see Ref. 15), we suggest that the orbital splitting follows a similar linear law: $E_z=-0.9t+4tx$, enhancing the anisotropy when $E_z>0$ for $x>0.22$. Indeed, the experimental electron densities, n_x and n_z, and their doping dependence,[12] are well reproduced [Fig. 1(a)], with the charge anisotropy δn being *enhanced* by the e_g electron correlations.

With all parameters, including $t=0.48$ eV, fixed now by experiment,[20] we next consider the spin dynamics, which is given by the first-order term in the $1/\mathcal{S}$ expansion of $\tilde{H}_t$. It

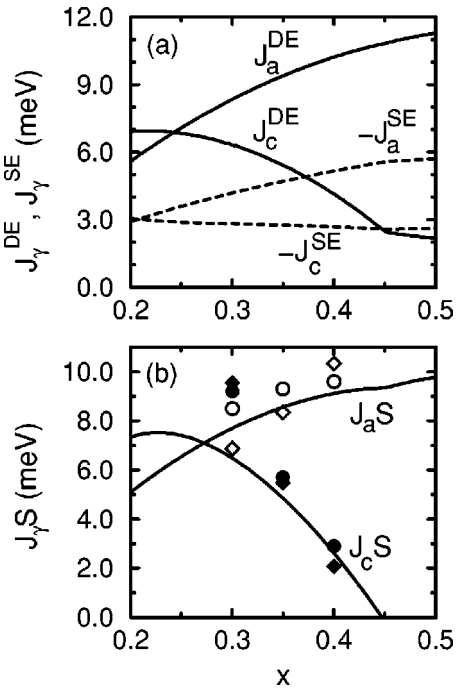

FIG. 2. (a) Magnetic DE (full lines) and SE (dashed lines) terms which contribute to the intralayer and interlayer interactions; and (b) effective exchange interactions: $J_a\mathcal{S}$ and $J_c\mathcal{S}$ (empty and filled symbols), compared with the experimental data of Refs. 9 (circles) and 10 (diamonds).

follows immediately from Eq. (2) that the DE (i.e., the kinetic-energy) contribution to the exchange constants is given by $J_a^{\rm DE}=-E_a/4\mathcal{S}^2$ and $J_c^{\rm DE}=-E_c/2\mathcal{S}^2$, where $E_\gamma=\langle\tilde{H}_t^{(0)}\rangle_\gamma/N$ is the kinetic energy along the $\gamma=a,c$ direction per atom. Since in-plane kinetic energy E_a of correlated e_g electrons is being gained, while interplanar energy E_c is being lost in the range $0.2<x<0.5$ [Fig. 1(b)], when the polarization towards the x^2-y^2 orbitals increases, $J_a^{\rm DE}$ and $J_c^{\rm DE}$ behave in the opposite way under increasing hole doping [Fig. 2(a)]. These trends agree with experiment, but *only* when the local correlations between the e_g electrons are included. In contrast, the kinetic energy for free electrons [see Eq. (1)], $E_a^{(0)}$ and $E_c^{(0)}$, would at best give $J_a^{\rm DE}$ weakly decreasing with x,[21] which contradicts the experimental data.[9,10] The electron density in the $|z\rangle$ orbitals is always finite ($n_z>0$), because the orbital (unlike the spin) quantum number is not conserved and both orbitals contribute to the electron dynamics, even when $E_z\simeq t$. Therefore, the DE term $J_c^{\rm DE}$ remains finite ($\simeq$2.2 meV at $x\simeq0.5$), and one has to consider the full t-J model including the SE term H_J in order to understand the observed crossover to the A-AF phase at $x\simeq0.45$.

The SE is derived by analyzing the individual charge excitations for $U\gg t$, $d_i^n d_j^m \rightleftharpoons d_i^{n+1} d_j^{m-1}$, starting from the high-spin ^5E ($^4A_2$) states of the Mn^{3+} (Mn^{4+}) ions. One thus finds doping-dependent SE interactions $\propto t^2/U$, having both AF and FM contributions:[22]

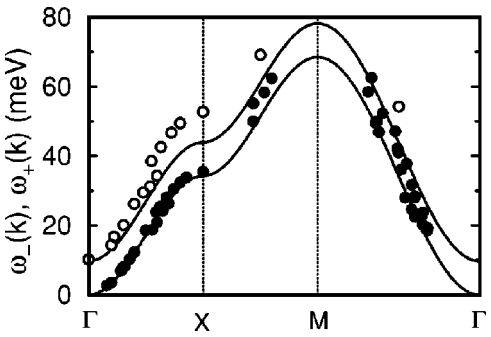

FIG. 3. Magnon dispersion $\omega_\pm(\vec{k})$ Eq. (7) as obtained in the manganite t-J model at $x=0.35$ with $J_a\mathcal{S}=8.55$ and $J_c\mathcal{S}=4.85$ meV (full lines) along the main directions in the two-dimensional Brillouin zone [$\Gamma=(0,0)$, $X=(\pi,0)$, $M=(\pi,\pi)$], and the experimental points of Ref. 9 (filled and empty circles).

$$H_J=-\sum_{\langle ij\rangle}\hat{K}_{ij}(\vec{S}_i\cdot\vec{S}_j+6)Q_{ij}^{11}+\sum_{\langle ij\rangle}\sum_{\alpha\beta}\hat{J}_{ij}^{\alpha\beta}(\vec{S}_i\cdot\vec{S}_j-c_{ij}^{\alpha\beta})Q_{ij}^{\alpha\beta}. \quad (5)$$

The operators $\hat{K}_{ij}$ and $\hat{J}_{ij}^{\alpha\beta}$ contain orbital projection operators with positive prefactors,[5,7] and have non-negative eigenvalues, resulting in $\langle\hat{K}_{ij}\rangle\geqslant0$ and $\langle\hat{J}_{ij}^{\alpha\beta}\rangle\geqslant0$. The FM interactions $\propto\hat{K}_{ij}$ occur only for Mn^{3+}-Mn^{3+} pairs.[23] In contrast, the AF interactions $\propto\hat{J}_{ij}^{\alpha\beta}$ occur for all possible configurations of e_g electrons on the bond $\langle ij\rangle$, as described by the operators $Q_{ij}^{\alpha\beta}$ with $\alpha,\beta=0,1$, e.g., $Q_{ij}^{10}=n_i(1-n_j)$. These AF terms arise both from e_g and t_{2g} electron excitations, while the constants $c_{ij}^{\alpha\beta}$ ($c_{ij}^{11}=4$, $c_{ij}^{10}=c_{ij}^{01}=3$, and $c_{ij}^{00}=\frac{9}{4}$) guarantee that they vanish in the FM state. Only a single t_{2g} term $\propto\hat{J}_{ij}^{00}$ survives from H_J in the limit of $x=1$ (with $Q_{ij}^{00}=1$), and explains the G-type AF order observed[24] in $Sr_3Mn_2O_7$.

The contributions to the energy and to the spin excitations from the SE terms are easily evaluated by applying the Schwinger boson expansion in each term of Eq. (5),

$$\vec{S}_i\cdot\vec{S}_j\simeq S_iS_j-\frac{1}{2}\sqrt{S_iS_j}(a_{i\downarrow}^\dagger a_{i\downarrow}+a_{j\downarrow}^\dagger a_{j\downarrow}-2a_{i\downarrow}^\dagger a_{j\downarrow}), \quad (6)$$

with $S_i(S_j)=2$ or 3/2. The SE terms depend on the electron densities n_x and n_z, which requires one to evaluate the averages of the orbital operators in Eq. (5); e.g., $J_c^{\rm SE}=\langle\hat{K}_{ij}\rangle_c\langle Q_{ij}^{11}\rangle_c-\Sigma_{\alpha\beta}\langle\hat{J}_{ij}^{\alpha\beta}\rangle_c\langle Q_{ij}^{\alpha\beta}\rangle_c$. While all the SE terms contribute to the magnon dispersion in order $1/\mathcal{S}$, the AF interactions dominate for realistic parameters, and thus the net SE *opposes* the DE.

The parameters of the SE (t, U, and J_H) are all fixed by the high-energy spectroscopy. The SE term in the (a,b) planes $-J_a^{\rm SE}$ is large and amounts to about half of the DE term $J_a^{\rm DE}$ [Fig. 2(a)]. It originates mainly from the largest AF (e_g and t_{2g}) contributions for Mn^{3+}-Mn^{4+} pairs. These

terms increase with x, and therefore the exchange constant $J_a\mathcal{S}=(J_a^{\rm DE}+J_a^{\rm SE})\mathcal{S}$ increases slower than the DE alone [Fig. 2(b)]. The doping dependence of $-J_c^{\rm SE}$ is quite different, as the AF e_g terms which involve now the $|z\rangle$ orbitals at Mn^{3+} ions are here much weaker. Therefore, the weak increase of the t_{2g} term with x is compensated by the decreasing e_g terms, and one finds an almost constant $J_c^{\rm SE}\simeq -2.8$ meV. So the doping dependence of the exchange constant $J_c\mathcal{S}=(J_c^{\rm DE}+J_c^{\rm SE})\mathcal{S}$ is entirely due to the DE (Fig. 2).

Finally, we demonstrate that the calculated values of $J_a\mathcal{S}$ and $J_c\mathcal{S}$ reproduce the overall doping dependence,[25] reported in Refs. 9 and 10 for $x=0.35$ and 0.40. As in experiment, the crossover to the A-AF phase ($J_c<0$) is found at $x\simeq 0.45$ (Fig. 2), when the AF SE on the interlayer bonds is stronger than the FM DE for correlated e_g electrons. The spin waves,

$$\omega_{\pm}(\vec{k})=4J_a\mathcal{S}(1-\gamma_{\vec{k}})+J_c\mathcal{S}(1\pm 1), \qquad (7)$$

where $\gamma_{\vec{k}}=\frac{1}{2}(\cos k_x+\cos k_y)$ are split into an acoustic, $\omega_-(\vec{k})$, and an optic, $\omega_+(\vec{k})$, mode for a single bilayer. The excellent agreement between the calculated and measured dispersions and splitting of these modes (Fig. 3) justifies *a posteriori* our theoretical model.[26]

Summarizing, the observed anisotropy of the spin excitations in the FM bilayer manganites is well explained by an *anisotropic orbital liquid* disordered phase, with predominantly x^2-y^2 orbitals occupied. The increase of the *intralayer* coupling $J_a\mathcal{S}$ is due to the release of the in-plane kinetic energy of the *correlated* e_g electrons under doping. The decreasing electron density in $3z^2-r^2$ orbitals explains the rapid decrease of the *interlayer* coupling $J_c\mathcal{S}$, which eventually changes its sign and triggers a transition to the A-AF phase.

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[26] The SE could also include the second-neighbor interlayer interaction $J_c'\mathcal{S}=-0.8$ meV along [1,0,1], involving the hopping of t_{2g} electrons via $O(2p_\pi)$ orbitals, and improving the agreement with the experiment for $\omega_+(\vec{k})$ (Ref. 9).