Why spin excitations in metallic ferromagnetic manganites are isotropic

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We consider the *t-J* model for doped manganites, and show that double exchange for correlated e_g electrons favors in the ferromagnetic metallic phase an orbital liquid (i.e., orbital-disordered) state. Therefore the spin-wave stiffness is *isotropic*, and *increases* with hole doping x, providing a direct measure of the kinetic energy $\propto x$ of strongly correlated e_g electrons. The superexchange interactions, which stabilize orbital order at low doping, are frustrated in the orbital liquid and only reduce the stiffness constant, leading to a quantitative explanation of the magnon dispersion.

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Strong Coulomb interactions in transition-metal oxides are responsible for numerous fascinating phenomena. They suppress charge fluctuations in the Mott-Hubbard insulators, and replace them by superexchange (SE) interactions. While these interactions are antiferromagnetic (AF) when they follow just from the Pauli principle for a single orbital, they have a richer structure when degenerate *d* orbitals are involved, as in cuprates or manganites. It is evident that in such situations the orbital and spin degrees of freedom have to be considered on equal footing,[1] and the SE interactions are highly frustrated even on a cubic lattice. This was recently recognized as the origin of interesting quantum effects both for e_g and t_{2g} electrons.[2] However, in undoped e_g systems such frustration is likely removed by the Jahn-Teller (JT) effect, which leads to a structural phase transition and stabilizes orbital ordering. The best example is undoped $LaMnO_3$ with *A*-type AF spin order which coexists with orbital order,[3,4] and can be understood only as a superposition of the cooperative JT effect and the SE.[5]

The theoretical understanding of doped manganese oxides $La_{1-x}A_xMnO_3$, with $A=$Sr,Ca,Pb, is among the most challenging current areas of research in condensed-matter physics. Experimental studies have revealed their rich phase diagrams,[3] originating from the competition between charge, spin, and orbital degrees of freedom. Charge fluctuations are thereby partly suppressed by the large Coulomb interaction $U\simeq 5.9$ eV (Ref. 6) between two e_g electrons on the same ion. In addition, the Hund's exchange $J_H\simeq 0.7$ eV couples the $s=1/2$ spins of the e_g electrons strongly to the $S_t=3/2$ core spins of the t_{2g} electrons, resulting in large $S=2$ spins of Mn^{3+} ions. When $LaMnO_3$ is doped, holes that are created in e_g orbitals can move but only when all spins are parallel, as explained by the double-exchange (DE) mechanism,[7] thus leading to a ferromagnetic (FM) state. When the orbital degeneracy is ignored, as in the original DE theory, isotropic magnon excitations are obtained.[8] However, when the orbital degrees of freedom are taken into account, one finds that the kinetic energy is lowered when orbital order occurs and the system becomes effectively lower dimensional.[9–11] Such states have broken cubic symmetry, and thus *anisotropic* magnetic excitations.[11] It is therefore puzzling why *isotropic* transport and magnetic properties are observed in the FM metallic manganites,[3,12] best exemplified by the isotropic spin waves in $La_{0.7}Pb_{0.3}MnO_3$.[13]

In this paper we investigate the microscopic origin of the isotropic magnetic properties of the FM metallic manganites,[12,13] and demonstrate that orbital fluctuations play a prominent role in this phase, stabilizing an isotropic *orbital liquid* (OL) phase with disordered orbitals. Thereby we consider (i) why the FM metallic phase is *orbital disordered* in a system which seems to have such a strong tendency towards orbital ordering, and (ii) what the physical meaning is of the *increase of the spin-wave stiffness* with hole doping x (Ref. 14). In addition, we show that the SE cannot be ignored, and that it reduces the spin-wave stiffness constant down to experimental values.

We study the manganite *t-J* model

$$\mathcal{H}_{tJ}=H_t+H_J^e+H_J^t+H_{\rm JT}, \tag{1}$$

derived for strongly correlated e_g and t_{2g} electrons at Mn^{3+} (Mn^{4+}) ions in the $t_{2g}^3e_g:{}^5E$ ($t_{2g}^3:{}^4A_2$) state determined by Hund's rule. Model (1) includes the kinetic energy in the e_g band (H_t) describing the DE at $U\rightarrow\infty$,[15] the SE which follows from virtual charge excitations due to e_g (H_J^e) and t_{2g} (H_J^t) electron hopping at large $U\gg t$, and the intersite JT interaction ($H_{\rm JT}$).

The kinetic energy H_t describes the electron hopping $\propto t\simeq 0.41$ eV (Ref. 6) between pairs of directional e_g orbitals $|\zeta\rangle$ oriented along the individual bonds $\langle ij\rangle$, i.e., $|\zeta\rangle\propto 3x^2-r^2$, $3y^2-r^2$, and $3z^2-r^2$, for $\langle ij\rangle$ along the cubic axis $\alpha=a$, *b*, or *c*, respectively (we denote by $|\xi\rangle$ the orthogonal orbital $\propto y^2-z^2$, z^2-x^2, and x^2-y^2, respectively, perpendicular to the bond). In order to derive the spin excitations we separate the spin dynamics from the charge and orbital dynamics, representing the electron creation operator $d_{i\zeta\sigma}^\dagger\propto a_{i\sigma}^\dagger c_{i\zeta}^\dagger$ by a fermion with orbital flavor $c_{i\zeta}^\dagger$ and a Schwinger boson $a_{i\sigma}^\dagger$. The number of bosons per site, $\Sigma_\sigma a_{i\sigma}^\dagger a_{i\sigma}$, depends on the electron configuration, and thus we introduce a slave boson for a hole, with the boson number operator $e_i^\dagger e_i=0$ ($e_i^\dagger e_i=1$) for Mn^{3+} (Mn^{4+}) ions, respectively. The physical space is defined by the local constraint $\Sigma_\sigma a_{i\sigma}^\dagger a_{i\sigma}+e_i^\dagger e_i=4$. Here we use the average

constraint $\Sigma_\sigma a_{i\sigma}^\dagger a_{i\sigma}=2\mathcal{S}$ with $2\mathcal{S}=4-x$ and $\langle e_i^\dagger e_i\rangle=x$. The kinetic energy, from the electron hopping in the subspace without double occupancies of e_g orbitals, is then

$$H_t=-\frac{t}{2\mathcal{S}}\sum_\alpha \sum_{\langle ij\rangle\|\alpha,\sigma} a_{i\sigma}^\dagger \tilde{c}_{i\zeta}^\dagger \tilde{c}_{j\zeta} a_{j\sigma}, \tag{2}$$

where $\tilde{c}_{i\zeta}^\dagger=c_{i\zeta}^\dagger(1-n_{i\bar{\zeta}})$. The band energy and the spin dynamics in a FM metallic state are then obtained by expanding the Schwinger bosons in Eq. (2):

$$\frac{1}{2\mathcal{S}}\sum_\sigma a_{i\sigma}^\dagger a_{j\sigma}\simeq 1-\frac{1}{4\mathcal{S}}(a_{i\downarrow}^\dagger a_{i\downarrow}+a_{j\downarrow}^\dagger a_{j\downarrow}-2a_{i\downarrow}^\dagger a_{j\downarrow}). \tag{3}$$

In analogy to frustrated magnetic systems, the kinetic energy [Eq. (2)] is orbitally frustrated, i.e., the contributions coming from different bonds cannot be minimized simultaneously, as the directional orbitals $\{|\zeta\rangle\}$ point in different directions. In order to demonstrate that the ground state is an OL, we rewrite H_t using the *complex orbitals* $|+\rangle$ and $|-\rangle$, which are linear combinations of the conventional real orbitals $\{|x\rangle=|x^2-y^2\rangle,|z\rangle=|3z^2-r^2\rangle\}$, with creation operators $c_{i\pm}^\dagger=(1/\sqrt{2})(c_{iz}^\dagger \mp ic_{ix}^\dagger)$. In lowest (zeroth) order in $1/\mathcal{S}$, one obtains

$$H_t^{(0)}=-\tfrac{1}{2}t\sum_{\langle ij\rangle}[\tilde{c}_{i+}^\dagger\tilde{c}_{j+}+\tilde{c}_{i-}^\dagger\tilde{c}_{j-}+e^{-i\chi_\alpha}\tilde{c}_{i+}^\dagger\tilde{c}_{j-} +e^{+i\chi_\alpha}\tilde{c}_{i-}^\dagger\tilde{c}_{j+}], \tag{4}$$

with the phase χ_α depending on the bond direction: $\chi_{a,b}=\pm 2\pi/3, \chi_c=0$. As Eq. (4) is manifestly invariant under cubic rotations, it provides a good starting point to analyze the metallic FM phase of doped manganites.

In order to treat $H_t^{(0)}$, we use the slave-boson functional integral method of Kotliar and Ruckenstein[16] in the limit $U\to\infty$, and adapt it for the orbital degrees of freedom.[17] Here the occupied e_g orbitals $|\pm\rangle=\tilde{c}_{i\pm}^\dagger|0\rangle$ are represented by fermion ($f_{i\pm}^\dagger$) and boson ($b_{i\pm}^\dagger$) operators, $\tilde{c}_{i\pm}^\dagger\propto f_{i\pm}^\dagger b_{i\pm}^\dagger e_i$, with the local constraint $b_{i+}^\dagger b_{i+}+b_{i-}^\dagger b_{i-}+e_i^\dagger e_i=1$. With all sites assumed equivalent, mean-field approximation leads to

$$H_t^{(0)}=-\sum_{i,\lambda=\pm}\mu_{i\lambda}n_{i\lambda}-\tfrac{1}{2}t\sum_{ij}[q_+f_{i+}^\dagger f_{j+}+q_-f_{i-}^\dagger f_{j-} +\sqrt{q_+q_-}(e^{-i\chi_\alpha}f_{i+}^\dagger f_{j-}+e^{+i\chi_\alpha}f_{i-}^\dagger f_{j+})], \tag{5}$$

with $n_{i\pm}=f_{i\pm}^\dagger f_{i\pm}$ and band narrowing Gutzwiller factors $\sqrt{q_\pm}=[x/(1-\langle n_{i\pm}\rangle)]^{1/2}$. A similar expression is obtained for two-sublattice states, as for instance for alternating orbital $|+\rangle/|-\rangle$ (AO$\pm$) order. The Lagrange multipliers $\mu_{i\lambda}$ originate from the local constraints in the boson space, and have been found in a self-consistent solution.

We calculated the kinetic energy $E_{\rm kin}=\langle H_t^{(0)}\rangle$ for several metallic phases [Fig. 1(a)], reproducing, *inter alia*, the reported transition from $|x\rangle$ to $|z\rangle$ orbital polarization.[11] However, this is not a transition between ground states, because at any finite doping $x=1-n$ disordered orbitals give an even lower $E_{\rm kin}$. Indeed, considering the total energy [see Fig. 1(b)] one finds that when the orbital ordering induced by SE (see below), which is so pronounced in the undoped system,[5,9–11] becomes unstable at larger doping where the kinetic term H_t dominates, it is replaced by the OL state. This OL phase is characterized by *equal* fermion densities for *any* pair of orthogonal orbitals, so $\langle n_{i\pm}\rangle=\langle n_{i\zeta(\bar{\zeta})}\rangle=\frac{1}{2}(1-x)$, and is qualitatively different from the one considered before.[18,19] Its kinetic energy is obtained by summing over the occupied states in the effective bands, $\varepsilon_{\mathbf{k}\pm}=-tq(x)[A_{\mathbf{k}}\pm B_{\mathbf{k}}]$, which are *narrowed by the Gutzwiller factor* $q(x)=q_{i\pm}(x)=2x/(1+x)$ [so that $E_{\rm kin}\propto q(x)$], with the dispersion given by $A_{\mathbf{k}}=c_x+c_y+c_z=3\gamma_{\mathbf{k}}$ and $B_{\mathbf{k}}^2=c_x^2+c_y^2+c_z^2-(c_xc_y+c_yc_z+c_zc_x)$, with $c_x=\cos k_x$, etc. These bands are isotropic, and interpolate between the limit of an almost empty uncorrelated band ($x\simeq 1$) and localized e_g electrons at half-filling ($x=0$).

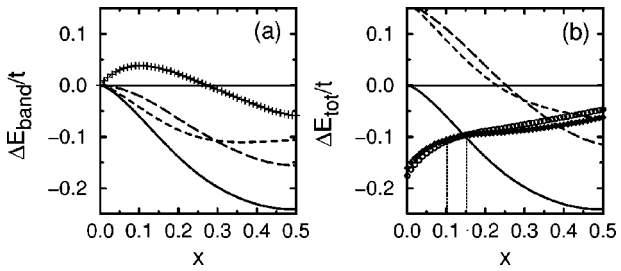

FIG. 1. Energy gains with respect to the polarized $|+\rangle$ phase as functions of doping x: (a) kinetic energy $\Delta E_{\rm kin}/t$, and (b) total energy $\Delta E_{\rm tot}/t$, for polarized $|x\rangle$ (dashed lines), $|z\rangle$ (long-dashed lines), staggered $|+\rangle/|-\rangle$ (pluses), and OL (full lines) phases. At low doping the orbital ordered states *A*-AF (circles) and FI (diamonds) states are more stable (b).

The energy of the spin excitations is determined by the first order term in the $1/\mathcal{S}$ expansion [Eq. (3)] of H_t. After averaging over the fermions, one finds the effective DE interaction, $J_{\rm DE}=\langle H_t^{(0)}\rangle/2z\mathcal{S}^2$, where $z=6$ for a cubic lattice. The magnons are *isotropic* in the OL state, with $\omega_{\mathbf{k}}^{\rm DE}=J_{\rm DE}z\mathcal{S}(1-\gamma_{\mathbf{k}})$. The spin-wave stiffness $D_{\rm DE}=J_{\rm DE}\mathcal{S}$ (note that we do not include a factor a^2 in the definition of D, where a is the lattice constant) is determined by the *kinetic energy of the correlated e_g electrons*, which in the OL state is therefore proportional to the band narrowing factor $q(x)$—it *vanishes* in the undoped $x\to 0$ limit, and *increases with hole doping x* (Fig. 2), in qualitative agreement with the observations in FM metallic manganites.[14] This expresses the gradual release of the kinetic energy in a correlated system when the carrier (hole) concentration increases. We emphasize that this experimental finding is reproduced *only* when the local correlations between e_g electrons are included, while the opposite and incorrect result, $J_{\rm DE}$ decreasing with x, is obtained in electronic structure calculations[20] that ignore these correlations.

The SE is somewhat intricate, as it involves both AF and FM contributions, most of them orbital dependent, and is therefore highly frustrated. Whereas the largest Mn^{3+}-Mn^{3+} interaction is FM and thus would add to the DE,[19] it achieves its full strength only for a particular orbital orientation. In the OL phase, when the e_g orbitals are disordered (and the SE is isotropic), the AF interactions dominate and the net SE *opposes* the DE.

Explicitly, the SE is obtained by starting from the high-

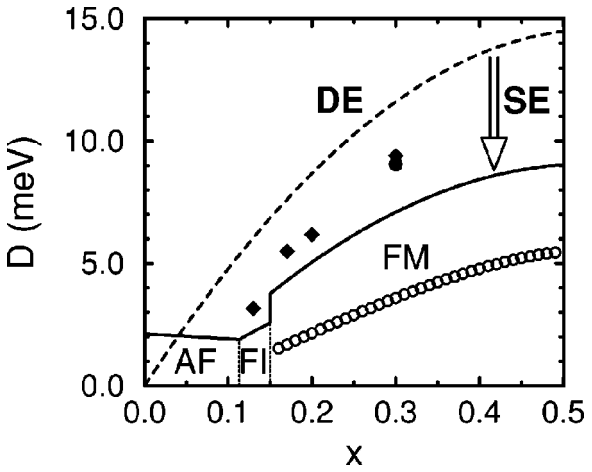

FIG. 2. Spin-wave stiffness D as a function of hole doping x from DE (dashed line), and including also the SE terms (full line) in Eq. (9), for A-AF (AF), FI, and FM metallic phases. Experimental points correspond to $La_{1-x}Sr_xMnO_3$ (Ref. 14) (diamonds) and $La_{0.7}Pb_{0.3}MnO_3$ (Ref. 13) (filled circle). The staggered AO± state is shown for $x>0.15$ (open circles).

spin ^5E and $^4A_2$ states of the Mn^{3+} and Mn^{4+} ions, with the spin operators $\vec{S}_i$ being $S=2$ or $S=3/2$, respectively, and considering the charge excitations $d_i^n d_j^m \rightleftharpoons d_i^{n+1} d_j^{m-1}$. Such processes have to be analyzed carefully, and the intermediate states projected on the eigenstates of the respective ions: Mn^{2+}, Mn^{3+}, and Mn^{4+}.[21] Consider first the SE induced by the e_g electrons,

$$H_J^e=\sum_{\langle ij\rangle}\{[-\hat{J}_1^e(\vec{S}_i\cdot\vec{S}_j+6)+\hat{J}_2^e(\vec{S}_i\cdot\vec{S}_j-4)]Q_{ij}^{11} + \hat{J}_3^e(\vec{S}_i\cdot\vec{S}_j-3)Q_{ij}^{01}\}, \quad (6)$$

with $Q_{ij}^{11}=n_in_j$, $Q_{ij}^{01}=n_i(1-n_j)+(1-n_i)n_j$, and $Q_{ij}^{00}=(1-n_i)(1-n_j)$ indicating the local e_g electron occupation. The Mn^{3+}-Mn^{3+} SE interactions ($\hat{J}_1^e$ and $\hat{J}_2^e$) are $\propto t^2/U$, whereas the Mn^{4+}-Mn^{3+} SE is strongest with $\hat{J}_3^e \propto t^2/\overline{J}_H$, because the excited Mn^{3+} low-spin ^3E state involved has only excitation energy $\propto\overline{J}_H$.[21] All interactions depend on the orbital configuration at the Mn^{3+} site(s): for Mn^{3+}-Mn^{3+} pairs this was analyzed in Ref. 5, e.g., $\hat{J}_1^e=\frac{1}{10}\{t^2/\varepsilon(^6A_1)\}(n_{i\zeta}n_{j\xi}+n_{i\xi}n_{j\zeta})$, while for Mn^{4+}-Mn^{3+} pairs one finds $\hat{J}_3^e=\frac{1}{8}\{t^2/\varepsilon(^3E)\}[(1-n_i)n_{j\zeta}+n_{i\zeta}(1-n_j)]$. Next, consider the SE induced by t_{2g} hopping,

$$H_J^t=\sum_{\langle ij\rangle}[\hat{J}_1^t(\vec{S}_i\cdot\vec{S}_j-4)Q_{ij}^{11}+\hat{J}_2^t(\vec{S}_i\cdot\vec{S}_j-3)Q_{ij}^{01} + \hat{J}_3^t(\vec{S}_i\cdot\vec{S}_j-\tfrac{9}{4})Q_{ij}^{00}], \quad (7)$$

which is weaker than H_J^e by almost an order of magnitude because of the smaller hopping amplitude $\simeq t/3$. Again the SE interactions $\hat{J}_1^t$ and $\hat{J}_2^t$ for Mn^{3+}-Mn^{3+} and Mn^{3+}-Mn^{4+} pairs, respectively, are orbital dependent (but only weakly so). Therefore, in any state with ordered real orbitals the SE given by $H_J=H_J^e+H_J^t$ would be anisotropic. The intersite JT interaction H_{JT} favors alternation of occupied e_g orbitals,[5] but being spin independent does not contribute to the magnon energy.

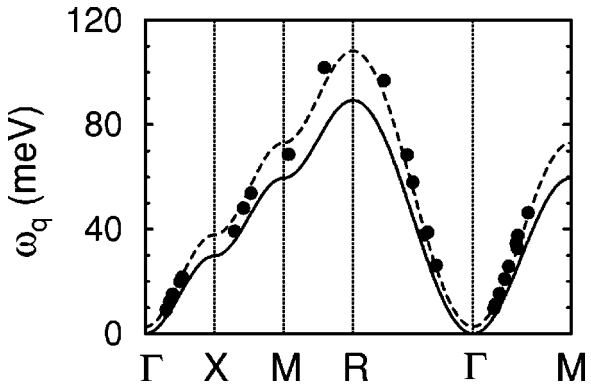

FIG. 3. Magnon dispersion [Eq. (9)] as obtained at $x=0.30$ (full line), and the experimental points for $La_{0.7}Pb_{0.3}MnO_3$ (circles and dashed line) after Ref. 13.

The contributions to the energy and the spin excitations due to the SE terms are easily evaluated using again the Schwinger bosons $\{a_{i\uparrow}^\dagger, a_{i\downarrow}^\dagger\}$, leading in the FM state to

$$\vec{S}_i\cdot\vec{S}_j\simeq S_iS_j-\tfrac{1}{2}\sqrt{S_iS_j}(a_{i\downarrow}^\dagger a_{i\downarrow}+a_{j\downarrow}^\dagger a_{j\downarrow}-2a_{i\downarrow}^\dagger a_{j\downarrow}), \quad (8)$$

with $S_i(S_j)=2$ or 3/2. One finds that all SE terms contribute to the magnon dispersion, which is *isotropic* in the OL state as $\langle n_{i\zeta}\rangle=\langle n_{i\xi}\rangle$ for any pair $\{|\zeta\rangle,|\xi\rangle\}$, and

$$\omega_{\mathbf{k}}=(J_{DE}-J_{SE})zS(1-\gamma_{\mathbf{k}}), \quad (9)$$

with $J_{SE}>0$, reducing the dispersion from that given by the DE. The strongest AF interactions $\propto\hat{J}_3^e$ come from the e_g excitations for Mn^{3+}-Mn^{4+} pairs. As a result, the stiffness constant $D_{eff}=(J_{DE}-J_{SE})S$ is significantly reduced, and this reduction increases with x (Fig. 2). The calculated values reproduce the increase of D with increasing x observed in the FM manganites for $x>0.15$ (Ref. 14).

The spin-wave stiffness $D_{eff}=7.45$ meV obtained at $x=0.3$ is thus *isotropic*,[22] and agrees well with the universal values reported by Fernandez-Baca *et al.*[12] at $x\simeq0.3$ doping. Thus it is also close to $D_{expt}=8.79$ meV measured in $La_{0.7}Pb_{0.3}MnO_3$ (Ref. 13) (see Fig. 3). This agreement is gratifying indeed, as no fit was used but all parameters were derived from spectroscopic data,[6,21] and further improvement is only possible by including the quantum fluctuations beyond the mean-field approximation in the OL phase. Note that although the states with polarized complex orbitals, i.e., the ferro orbital state with $|+\rangle$ orbitals at all sites (FO+) or the staggered AO± state,[10] would give isotropic magnons as well, they do not offer a valid explanation,[23] as they are both unstable against the OL state.

The manganites at $x\leqslant0.15$ are insulating, and are orbital ordered, with either A-AF or FM spin order. This behavior is generally understood to be due to polaronic effects,[24] which dominate at low carrier concentration. Considering a polaronic model with localized e_g electrons,[25] we find that the SE interactions in combination with the JT interaction

stabilize orbital order in either the *A*-AF phase at doping $x \lesssim 0.11$, or in the FM insulating (FI) phase at $0.11 \lesssim x < 0.15$ [Fig. 1(b)]. The anisotropic magnons in this regime are explained by the SE alone (Fig. 2), while the DE is blocked by the polaronic localization of the e_g electrons.[26]

Summarizing, the isotropy of the spin excitations in the FM metallic phase of doped manganites is naturally explained by the *orbital liquid* state of disordered e_g orbitals, triggered by doping. In this state the magnon excitation energy is proportional to the kinetic energy of the e_g electrons, and thus measures directly the *band narrowing due to the strong correlations*. This explains the increase of the spin-wave stiffness with increasing hole doping, observed in $La_{1-x}Sr_xMnO_3$ compounds.

Note added in proof. Recently, we became aware of the recent data for the magnetic coupling in insulating and metallic ferromagnetic $La_{1-x}Ca_xMnO_3$.[27] In particular it was found that (i) also for Ca doping the spin-wave stiffness D is proportional to x, but only in the metallic regime $x > 0.2$ dominated by the DE, while (ii) D is smaller (typically by a factor larger than two) in the insulating regime and exhibits a discontinuity at the transition of the metallic phase.[27] Our theoretical description (see Fig. 2 in particular) reproduces both these experimental features.

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