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SPIN EXCITATIONS IN DOPED MANGANITES

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We generalize the double exchange to describe holes doped in degenerate e_g orbitals in $La_{1-x}A_xMnO_3$ manganites, where A are divalent alkaline earth ions. Assuming an orbital liquid of disordered e_g orbitals we find *isotropic* ferromagnetic exchange interactions which increase with hole doping. The magnon dispersion agrees very well with the experimental data in ferromagnetic metallic manganites.

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The ferromagnetic (FM) metallic phase in doped $La_{1-x}A_xMnO_3$ compounds, where A are divalent alkaline earth ions, and $0.1 < x < 0.5$, is believed to be stabilized by the double-exchange (DE) mechanism [1], using purely magnetic interactions. However, there are indications that this concept is oversimplified, as it does not allow to reproduce the experimentally observed temperature dependence of the resistivity, and the value of the FM transition temperature itself [2]. Further evidence comes from the optical conductivity in the FM metallic phase, with a very weak Drude peak and strong intensity of the incoherent part distributed uniformly in a broad range of $0 < \omega < 1$ eV, both features indicating the crucial role of another degree of freedom — the orbital variable for e_g electrons [3, 4].

Double exchange in *degenerate* e_g orbitals is expected in the first instance to produce anisotropic phases, and indeed the antiferromagnetic (AF) layered structures of the A-type, and the chain-like C-type structures are stabilized at low electron concentration $n \ll 1$ (i.e., $x = 1 - n \approx 0$) [5]. The case of the FM metallic systems ($n \approx 0.7$) is different, as here electrons feel a strong Coulomb repulsion U, and the system becomes an AF charge-transfer insulator at $n = 1$. Here also the magnetic interactions are strongly anisotropic, and their experimental values and the A–AF order observed in $LaMnO_3$ can be reproduced only by considering superexchange (SE) between Mn^{3+} ions with degenerate e_g orbitals [6].

Here we investigate a novel effective t–J model for doped $La_{1-x}A_xMnO_3$ compounds in the FM regime [7],

$$\mathcal{H} = H_t + H_J^e + H_J^t, \tag{1}$$

where H_t describes the correlated hopping in the e_g-band, and H_J^e (H_J^t) stands for the SE term due to virtual hopping of e_g (t_{2g}) electrons. The Hilbert space in the large-U limit contains only d^4 (Mn^{3+}) configurations: $|i\theta, M\rangle_4 = f_{ix}^\dagger b_{ix}^\dagger |M\rangle$ and $|i\epsilon, M\rangle_4 = f_{iz}^\dagger b_{iz}^\dagger |M\rangle$ with M being the component of spin $S = 2$, and d^3 (Mn^{4+}) configurations, $|i, m\rangle_3 = b_{i0}^\dagger |m\rangle$, with m being the component of the core t_{2g}-spin $S = 3/2$. We use a local basis for the occupied e_g orbitals: $|x\rangle \equiv |x^2 - y^2\rangle$ and $|z\rangle \equiv |3z^2 - r^2\rangle$. The fermion operators $\{f_{ix}^\dagger, f_{iz}^\dagger\}$ describe the orbitals, while we represent the (local) spin state $|M\rangle$ ($|m\rangle$) by means of the Schwinger bosons at site i, $a_{i\uparrow}^\dagger$ and $a_{i\downarrow}^\dagger$. The orbital bosons for the Mn^{3+} sites, $b_{ix}^\dagger$ and $b_{iz}^\dagger$, and for the empty Mn^{4+} sites, $b_{i0}^\dagger$, are introduced to implement the local constraints,

$$a_{i\uparrow}^\dagger a_{i\uparrow} + a_{i\downarrow}^\dagger a_{i\downarrow} + b_{i0}^\dagger b_{i0} = 4, \qquad b_{ix}^\dagger b_{ix} + b_{iz}^\dagger b_{iz} + b_{i0}^\dagger b_{i0} = 1. \tag{2}$$

They restrict the physical space to contain no double occupancy in the e_g orbitals. In addition, the number of fermions is equal to the number of bosons for each orbital, $f_{i\lambda}^\dagger f_{i\lambda} = b_{i\lambda}^\dagger b_{i\lambda}$, $\lambda = x, z$, which gives two additional terms with the Lagrange multipliers in the Hamiltonian H_t.

The occupied e_g orbital at site i, $|i\rangle = \cos\theta |iz\rangle + \sin\theta |ix\rangle$, may be represented by an orbital angle θ. The hopping term H_t reduces in the mean-field approximation for the slave bosons [8] to the fermion problem coupled to the Schwinger boson operators (for $S = 2$),

$$H_t = -\frac{1}{2S} \sum_{\langle ij \rangle} \sum_{\lambda\rho\sigma} q_{i\lambda} q_{j\rho} t_{i\lambda, j\rho} a_{i\sigma}^\dagger a_{j\sigma} f_{i\lambda}^\dagger f_{j\rho}, \tag{3}$$

where the hopping elements $t_{i\lambda,j\rho}$ for the orbitals $\lambda, \rho = x, z$ are renormalized by $q_{ix} = \sqrt{2\delta/[1 + \delta + (1-\delta)\cos 2\theta]}$ and $q_{iz} = \sqrt{2\delta/[1 + \delta - (1-\delta)\cos 2\theta]}$. Replacing the Schwinger bosons by the Holstein–Primakoff bosons, one finds up to lowest order $\sum_\sigma a_{i\sigma}^\dagger a_{j\sigma} = 2S - \frac{1}{2}(a_{i\downarrow}^\dagger a_{i\downarrow} + a_{j\downarrow}^\dagger a_{j\downarrow} - 2a_{i\downarrow}^\dagger a_{j\downarrow})$. The zeroth-order term $\propto 2S$ gives a band Hamiltonian $H_t^{(0)}$ which describes two fermionic bands narrowed by correlation effects. The remaining first-order terms $H_t^{(1)}$ may be written as products of bilinear terms in fermion ($f_{i\lambda}^\dagger$) and magnon ($a_{i\sigma}^\dagger$) operators. Averaging over the correlated band structure yields the magnon dispersion due to the DE mechanism $\propto (t/2S) R_{ab}(2 - \cos q_x - \cos q_y)$ and $\propto (t/2S) R_c(1 - \cos q_z)$ in the main directions of the cubic Brillouin zone, with $t = 0.40$ eV being the largest local hopping element between two $3z^2 - r^2$ orbitals along the c-axis. The lattice sums R_{ab} and R_c depend on the average occupancy of x and z orbitals. For an orbital liquid, with $\cos 2\theta = n_x - n_z = 0$, they are approximately equal, reproducing the qualitative result of the Kondo lattice model with a non-degenerate band [9].

The hopping of e_g electrons between a pair of Mn^{3+} ions creates a double occupancy in the e_g orbitals at one site and is included in a perturbative way in the manganite t–J model (1). By considering the $d_i^4 d_j^4 \rightleftharpoons d_i^3(t_{2g}^3) d_j^5(t_{2g}^3 e_g^2)$ excitations

one finds the magnetic part of the e_g SE between spins $S = 2$ [6],

$$H_J^e = \frac{1}{16}\sum_{\langle ij\rangle} n_i n_j \left\{ -\frac{8}{5}\frac{t^2}{\varepsilon(^6A_1)}\mathcal{P}_{\langle ij\rangle}^{\zeta\xi}\boldsymbol{S}_i\cdot\boldsymbol{S}_j \right.$$

$$\left. + \left[\left(\frac{t^2}{\varepsilon(^4E)} + \frac{3}{5}\frac{t^2}{\varepsilon(^4A_1)}\right)\mathcal{P}_{\langle ij\rangle}^{\zeta\xi} + \left(\frac{t^2}{\varepsilon(^4E)} + \frac{t^2}{\varepsilon(^4A_2)}\right)\mathcal{P}_{\langle ij\rangle}^{\zeta\zeta}\right]\boldsymbol{S}_i\cdot\boldsymbol{S}_j \right\}, \quad (4)$$

where $\varepsilon(^6A_1) = U - 5J_H$, $\varepsilon(^4A_1) = U$, $\varepsilon(^4E) = U + \frac{2}{3}J_H$, $\varepsilon(^4A_2) = U + \frac{10}{3}J_H$ are the excitation energies of high spin ($^6A_1$) and low spin ($^4A_1$, ^4E, and $^4A_2$) configurations at Mn^{2+} ions, expressed in terms of $U \approx 7.3$ eV and $J_H = 0.69$ eV [6], and $n_i = f_{ix}^\dagger f_{ix} + f_{iz}^\dagger f_{iz}$. This part of the SE depends on the occupied orbitals of both sites via the projection operators, $\mathcal{P}_{\langle ij\rangle}^{\zeta\xi} = P_{i\zeta}P_{j\xi} + P_{i\xi}P_{j\zeta}$ and $\mathcal{P}_{\langle ij\rangle}^{\zeta\zeta} = 2P_{i\zeta}P_{j\zeta}$, where the on-site projections give an orbital parallel/perpendicular ($P_{j\xi(\zeta)} = \frac{1}{2} \mp \tau_j^\alpha$) to the bond direction, with $\alpha = a, b, c$ for three cubic axes. The orbital operators τ_i^α are defined in a 2×2 pseudospin space and are given by the combinations of two Pauli matrices σ_i^z and σ_i^x: $\tau_i^{a(b)} = \frac{1}{4}(-\sigma_i^z \pm \sqrt{3}\sigma_i^x)$, $\tau_i^c = \frac{1}{2}\sigma_i^z$.

The t_{2g}-hopping leads in leading order to an isotropic SE [7],

$$H_J^t = \sum_{\langle ij\rangle}\left\{ \frac{1}{4}J_t n_i n_j\,(\boldsymbol{S}_i\cdot\boldsymbol{S}_j - 4) + \frac{4}{9}\widehat{J}_t(1-n_i)(1-n_j)\left(\boldsymbol{S}_i\cdot\boldsymbol{S}_j - \frac{9}{4}\right)\right.$$

$$\left. + \frac{1}{3}\bar{J}_t\,[n_i(1-n_j) + (1-n_i)n_j]\,(\boldsymbol{S}_i\cdot\boldsymbol{S}_j - 3)\right\}, \quad (5)$$

with the AF SE constants $J_t = 2.1$ meV, $\widehat{J}_t = 4.6$ meV, and $\bar{J}_t = 5.5$ meV for the pairs of Mn^{3+}–Mn^{3+}, Mn^{4+}–Mn^{4+}, and Mn^{3+}–Mn^{4+} ions, respectively, and $\boldsymbol{S}_i$ is an $S = 2$ ($S = 3/2$) spin operator for a Mn^{3+} (Mn^{4+}) ion with $n_i = 1$ ($n_i = 0$) e_g electrons.

Both SE terms (4) and (5) may be expanded using the Schwinger bosons around the FM state and lead to an isotropic reduction of the effective FM DE interactions. For the numerical evaluation we adopted the realistic parameters of $LaMnO_3$ as given in Ref. [6]. One finds that H_J^t gives a larger contribution, while the FM and AF terms in H_J^e almost compensate each other in the orbital liquid state with $\langle\mathcal{P}_{\langle ij\rangle}^{\zeta\xi}\rangle = \langle\mathcal{P}_{\langle ij\rangle}^{\zeta\zeta}\rangle = \frac{1}{2}$, and give a weak net AF interaction. The theory predicts an observed increase in the magnon width W with increasing doping [10] due to the DE which dominates in the metallic regime of $x > 0.08$ (Fig. 1). At small doping $x \leq 0.08$ we show instead W for a polaronic A–AF phase [6]. The DE vanishes in the $x \to 0$ limit, in contrast to the non-physical result of band structure calculations that ignore electron correlations, where the largest FM interactions occur at $x = 0$ [11], precisely at the point of the A–AF insulator $LaMnO_3$. The exchange interactions found at $x = 0.3$: $J_{ab}S = 8.20$ and $J_cS = 8.26$ meV are almost isotropic and reproduce well (Fig. 2) the experimental points for $La_{0.7}Pb_{0.3}MnO_3$ and the value of $JS = 8.79$ meV given in Ref. [12].

In conclusion, the magnon dispersion derived from DE for degenerate e_g orbitals supplemented by smaller SE terms agrees well with the experimental findings in FM metallic manganites [10, 12].

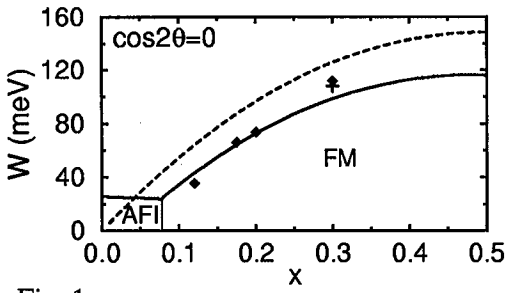

Fig. 1

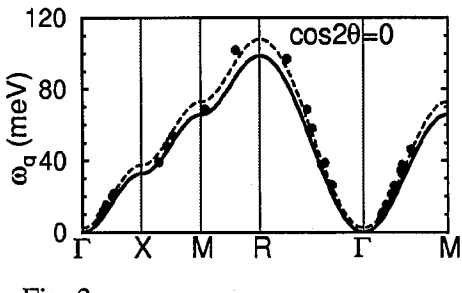

Fig. 2

Fig. 1. Width W of the magnon dispersion in FM manganites $La_{1-x}A_xMnO_3$ as a function of doping x, as obtained including only the DE mechanism (dashed line), and both DE and SE contributions from Eq. (1) (full line). In the AF insulating (AFI) phase at $x < 0.08$ only the anisotropic SE interactions contribute. Experimental points correspond to: $La_{1-x}Sr_xMnO_3$ [10] (diamonds) and $La_{0.7}Pb_{0.3}MnO_3$ [12] (cross).
Fig. 2. Magnon dispersion $\omega_{\boldsymbol{q}}$ as obtained at $x = 0.3$ doping level using DE and SE contributions (heavy line); parameters as in the text and in Ref. [6]. Experimental data for $La_{0.7}Pb_{0.3}MnO_3$ (circles and dashed line) are reproduced from Ref. [12].

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