Spin excitations in ferromagnetic manganites with orbital order

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New Journal of Physics 6 (2004) 190
Received 21 September 2004
Published 7 December 2004
Online at http://www.njp.org/
doi:10.1088/1367-2630/6/1/190

Abstract. The ferromagnetic metallic phase of doped cubic manganites with orbital order is investigated in a two-band double exchange model extended by Jahn–Teller (JT) terms. Evaluating the spin wave self-energy in the lowest order $1/S$ expansion, one finds that spin excitations are anisotropic and depend on the type of occupied $e_g$ orbitals. In the case of a small splitting between $e_g$ orbital energies, magnon dispersion exhibits a pronounced asymmetry between low and high hole doping, while a strong JT effect leads to the alternating orbital order which can reduce the spin stiffness by one order of magnitude. For a bilayer system, the kinetic energy induces an $x^2 - y^2$ orbital character and is responsible for a drastic change in the interplane exchange coupling in the doping range $0.3 < x < 0.5$. 
1. Introduction

The theoretical description of doped manganese perovskites $R_{1-x}A_x\text{MnO}_3$ (e.g. $R=$La, Nd, Pr; $A=$Sr, Ca, Pb) is among the most important current issues in condensed matter physics [1]. Although in a canonical double-exchange (DE) system, like $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ at $x \sim 0.3$, the DE model [2] can explain the magnetic and transport properties even quantitatively by a proper fitting of its parameters, such a model is not sufficient for other dopings or for compounds with smaller bandwidths [3]. Therefore, in the realistic model for doped manganites one has to consider not only spin but also orbital and lattice degrees of freedom. Strong competition between all these degrees of freedom [4] leads to rich phase diagrams [1], as revealed experimentally in different compounds.

The present study is motivated by the experimentally observed signatures of the orbital order (OO) in ferromagnetic (FM) manganites at low doping [5], which lead to interesting anomalies in the spin wave dispersion [6]. We will investigate the microscopic origin of the anisotropic magnetic properties of the FM metallic phase in cubic and bilayer (BL) manganites. Thereby we examine to what extent the spin excitation spectrum in these doped compounds, when dictated by the DE mechanism, depends on the type of possible order of occupied $e_g$ orbitals. While the DE mechanism, due to a strong coupling between spins of $t_{2g}$ and $e_g$ electrons by Hund’s exchange, leads to the FM order, it is frustrated by the superexchange (SE) interactions between $t_{2g}$ core spins which are antiferromagnetic (AF). Such a frustration can be removed to some extent by a structural phase transition due to the Jahn–Teller (JT) effect which also stabilizes a given type of the OO. Here, we calculate the magnon dispersion using the diagrammatic approach introduced for a one-band model first in the limiting case of infinite Hund’s exchange interaction ($J_H = \infty$) by Kubo and Ohata [7], and next extended to finite $J_H$ by Furukawa [8].

The paper is organized as follows. In section 2.1 we present the model Hamiltonian, and describe in section 2.2 the diagrammatic expansion for the magnon self-energy. Section 3 contains numerical results for spin wave dispersion and spin stiffness calculated at zero temperature. These quantities are evaluated as functions of doping and the $e_g$ orbital splitting for different types of the OO. Some comparison with data of neutron inelastic scattering experiments [9, 10] is made. The results are summarized in section 4.

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2. Model Hamiltonian

2.1. Effective spin–orbital model

We consider the FM Kondo lattice model for manganites \cite{11, 12} with the Mn e\textsubscript{g} electrons coupled to the core t\textsubscript{2g} spins via the Hund coupling $J_H$. The model is extended by cooperative JT interactions included on the mean field level as the splitting of two e\textsubscript{g} states in a given orbital basis which leads to the alternating OO:

$$H = H_t + H_{\text{Kondo}} + H_{\text{OO}}.$$  \hspace{1cm} (1)

The first term describes the motion of the e\textsubscript{g} electrons,

$$H_t = - \sum_{\langle ij \rangle \xi \zeta \sigma} \left( t_{ij}^{\xi \zeta} d_{i \xi \sigma}^{\dagger} d_{j \zeta \sigma} + \text{H.c.} \right),$$  \hspace{1cm} (2)

where the hopping matrix elements depend on the basis chosen. For the commonly used \{|$\mathbf{x}$\rangle,|$\mathbf{z}$\rangle\} basis, with

\begin{align*}
|\mathbf{x}\rangle &\sim |x^2 - y^2\rangle, \\
|\mathbf{z}\rangle &\sim |3z^2 - r^2\rangle,
\end{align*}  \hspace{1cm} (3)

orbitals, they are given by

$$\begin{pmatrix} t_{ij}^{\mathbf{x}} \\ t_{ij}^{\mathbf{z}} \end{pmatrix}_{|\mathbf{a}(b)\rangle} = \frac{t}{4} \begin{pmatrix} 3 & \mp \sqrt{3} \\ \mp \sqrt{3} & 1 \end{pmatrix}.$$  \hspace{1cm} (4)

in the (a, b) plane and by

$$\begin{pmatrix} t_{ij}^{\mathbf{c}} \\ t_{ij}^{\mathbf{e}} \end{pmatrix} = t \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix},$$  \hspace{1cm} (5)

for the nearest neighbours along the c direction. Here, + (−) in equation (4) refers to the a(b) direction. The orbital quantum number is not conserved (due to the presence of nondiagonal elements in equation (4)), and both orbitals contribute to the electron dynamics even in the limit of large orbital splitting.

In the ground state with robust OO, it is convenient to introduce local orbital bases on both sublattices. Therefore, we perform two different transformations depending on a sublattice \cite{13},

$$\begin{pmatrix} |i\mu\rangle \\ |i\nu\rangle \end{pmatrix} = \begin{pmatrix} \cos \left( \frac{\pi}{4} \pm \phi \right) & \sin \left( \frac{\pi}{4} \pm \phi \right) \\ -\sin \left( \frac{\pi}{4} \pm \phi \right) & \cos \left( \frac{\pi}{4} \pm \phi \right) \end{pmatrix} \begin{pmatrix} |i\mathbf{z}\rangle \\ |i\mathbf{x}\rangle \end{pmatrix},$$  \hspace{1cm} (6)

where + (−) refers to $i \in A$ ($i \in B$) sublattice, respectively. We use here a simplified notation, with |$i\mathbf{x}$\rangle and |$i\mathbf{z}$\rangle standing for the basis orbital (3) at site $i$. The new orbitals |$i\nu\rangle$ and |$i\mu\rangle$ are the occupied and empty orbital on the $A(B)$ sublattice, respectively. It is now straightforward to rewrite the hopping term (2) in terms of these new orbital states.
The second term in Hamiltonian (1) stands for the Hund interaction between the \( s = 1/2 \) spin of an \( e_g \) electron and the \( S = 3/2 \) core spin \( S_i \) due to \( t_{2g} \) electrons at site \( i \),

\[
H_{\text{Kondo}} = -J_H \frac{1}{S} \sum_{\mathbf{k},\sigma} S_i \cdot \vec{d}_{\mathbf{k},\sigma} \vec{\sigma}_{\sigma\sigma} \vec{d}_{\mathbf{k},\sigma'},
\]

where \( \lambda = \mu, \nu \), resulting in the parallel alignment of \( e_g \) electron spins and \( t_{2g} \) ones at each site in the regime of \( J_H \gg t \) which corresponds to the realistic situation in manganites. Finally, the orbital–orbital interaction is induced by the JT effect:

\[
H_{\text{OO}} = 3\kappa \sum_{\mathbf{k} \neq \mathbf{Q}} T_i^+ T_j^-, \tag{8}
\]

with pseudospin operators

\[
T_i^\pm = \frac{1}{2}(n_{i\mu} - n_{i\nu}), \tag{9}
\]

where \( n_{i\mu} = \sum_\sigma d_{i\mu\sigma}^\dagger d_{i\mu\sigma} \) refers to the occupied orbital, and \(|\nu\rangle\) stands for the (unoccupied) orbital orthogonal to \(|\mu\rangle\) at site \( i \) (see equation (6)). \( H_{\text{OO}} \) is treated on a mean-field level. The form of the interaction \( H_{\text{OO}} \) is adjusted to an alternating \( \text{OO} \) with on-site orbital states split by the energy \( \Delta E_{\text{OO}} = 12\kappa \), where \( \kappa \) is determined by the strength of the JT coupling on a given \( \text{Mn–O–Mn bond} \) [14]. We do not consider here a similar term which originates from the SE and would further increase this splitting [15]. Such a splitting suppresses charge fluctuations in the system in a similar manner as the Coulomb repulsion does [16]. Note that the first two terms of the total Hamiltonian, \( H_t + H_{\text{Kondo}} \), represent the DE mechanism [2], and yields at finite hole doping by \( x = 1 - n \) holes in \( e_g \) orbitals a FM ground state which is favoured by the gain in the kinetic energy of the remaining \( e_g \) electrons.

2.2. Spin excitations

We use the linear order spin wave theory to describe spin excitations in the FM state. Hence, we use an approximate transformation:

\[
S_i^+ \simeq \sqrt{2S} a_i, \quad S_i^- \simeq \sqrt{2S} a_i^\dagger, \quad S_i^z = S - a_i^\dagger a_i, \tag{10}
\]

from spin to boson operators \( \{a_i, a_i^\dagger\} \). Now the Hamiltonian given by equation (1), after transforming it to the momentum space, has the following form:

\[
H_{\text{LSW}} = \sum_{\mathbf{k} \sigma} \left[ T_{ab} c_{\sigma}^\dagger(\mathbf{k}) + T_c c_{\sigma}(\mathbf{k}) - \sigma J_H \right] c_{k1\sigma}^\dagger c_{k1\sigma} + \sum_{\mathbf{k} \sigma} \left[ T_{\sigma} c_{\sigma}^\dagger(\mathbf{k}) + T_c c_{\sigma}(\mathbf{k}) - \sigma J_H + 12\kappa \right] c_{k2\sigma}^\dagger c_{k2\sigma}
\]

\[
+ \sum_{\mathbf{k} \sigma} \left[ T_{\sigma} c_{\sigma}^\dagger(\mathbf{k}) + T_c c_{\sigma}(\mathbf{k}) \right] \left( c_{k1\sigma}^\dagger c_{k2\sigma} + \text{H.c.} \right) + \sum_{\mathbf{k} \sigma} T_{\sigma} c_{\sigma}(\mathbf{k}) \left( c_{k1\sigma}^\dagger c_{k2\sigma} + \text{H.c.} \right) + J_H \frac{1}{2SN} \sum_{\mathbf{k} \xi} \left( a_{q1}^\dagger a_{q2} c_{\mathbf{k}+\mathbf{Q}+\xi}^\dagger c_{\mathbf{k}+\mathbf{Q}-\xi} + a_{q1}^\dagger a_{q2} c_{\mathbf{k}+\mathbf{Q}-\xi}^\dagger c_{\mathbf{k}+\mathbf{Q}+\xi} \right) \pm J_H \frac{1}{SN} \sum_{\mathbf{k} q \xi} a_{q1}^\dagger a_{q2} c_{\mathbf{k}-\mathbf{Q}-\xi} c_{\mathbf{k}-\mathbf{Q}+\xi}, \tag{11}
\]

where, \( c_{\pm}(\mathbf{k}) = \frac{1}{2}(\cos k_x \pm \cos k_y) \), \( c_{\xi}(\mathbf{k}) = \cos k_z \) and \( +(-) \) in the last term refers to \( \sigma = \uparrow(\downarrow) \), respectively. Here \( \mathbf{Q} = (\pi, \pi, 0) \) is the nesting vector. The orbital index \( \xi = 1, 2 \) (referring to a
given orbital basis set by the rotation angle $\phi$ is unchanged in the scattering processes as both $e_g$ orbitals are orthogonal at each site. Moreover,

$$T_{ab} = -t \left[ 1 - 2 \sin(2\phi) \right], \quad T'_{ab} = -t \left[ 1 + 2 \sin(2\phi) \right], \quad T''_{ab} = -2t \cos(2\phi),$$

are in-plane hopping elements, and

$$T_c = -t \left[ 1 + \sin(2\phi) \right], \quad T'_c = -t \left[ 1 - \sin(2\phi) \right], \quad T''_c = t \cos(2\phi)$$

are hopping elements in the $c$ direction, while $T_Q = \sqrt{3} t/4$ stands for the Umklapp scattering factor. The Hamiltonian (11) describes fermions scattered on spin waves and the scattering processes can be classified as in the one-band model by two kinds of diagrams contributing to the magnon self-energy (corresponding to processes with one ($\sim a_{\xi}^\dagger c_k^\dagger c_{-\xi \sigma}^\dagger$) or two ($\sim a_{\xi}^\dagger a_{\eta}^\dagger c_k^\dagger c_{-\xi \sigma} c_{-\eta \sigma} c_{\eta \sigma}$) magnons involved in a scattering event [7, 8]).

Next, we diagonalize the hopping part of the Hamiltonian by a linear transformation by introducing operators

$$a_{k\nu\sigma}^\dagger = \sum_{\xi=1}^2 V_{\xi,\nu}(k)c_{k\xi\sigma}^\dagger + \sum_{\xi=1}^2 V_{\xi+2,\nu}(k)c_{k+Q\xi\sigma}^\dagger.$$  

The spin wave Green’s function is given by

$$D(q, \omega) = \{\omega - \Pi(q, \omega) + i\epsilon\}^{-1},$$

and magnon excitations are evaluated from the poles of this Green’s function,

$$\omega_q = \Pi(q, \omega_q),$$

where the magnon self-energy for completely polarized electrons in the FM phase ($f_{k \xi \sigma} = 0$) in the lowest order of $1/S$ expansion (as schematically illustrated in figure 1) is given by

$$\Pi(q, \omega) = \frac{1}{2SN} \sum_k \sum_{\xi=1}^4 f_{k\xi} \left\{ \sum_{\mu=1}^4 V_{\mu\xi}(k) + 2J_H^2 \sum_{\eta=1}^4 \left[ \sum_{\mu=1}^4 \frac{V_{\mu\xi}(k)V_{\mu\eta}(k+q)}{\omega + \epsilon_{k\xi} - \epsilon_{k\eta} - 2J_H} \right]^2 \right\}. \quad (17)$$
Here, \( f_{\sigma \xi}(k) \) is the Fermi distribution function for the \( e_g \) electron with spin \( \sigma = \uparrow, \downarrow \). The coefficients \( V_{\mu \xi}(k) \) follow from the diagonalization of \( H_t \) (see equation (1)), which involves momenta \( k (\xi = 1, 2) \) and \( k + Q (\xi = 3, 4) \), leading to the band energies \( \varepsilon_{k \xi} \). Numerical calculations were performed on three-dimensional (3D) \( 50 \times 50 \times 50 \) and BL \( 50 \times 50 \times 2 \) meshes in momentum space. As presented in the next section, although the interaction vertices are proportional to \( J_H \) in the physically interesting range, the magnon self-energy (17) does not show singularities in the limit \( J_H/t \to \infty \).

3. Numerical results and discussion

3.1. Cubic manganites

In the present study, we focus on the spin wave dispersion \( \omega_q \) and on the magnon stiffness \( D \) in the FM phase of doped manganites with the OO, controlled by the \( e_g \) level splitting \( \sim \kappa \). Thereby we assume the limit of strong Hund’s coupling \( J_H \gg t \). The effective Mn–Mn hopping \( t \) strongly depends on the lattice parameters (e.g. on the Mn–O bond length, and on the Mn–O–Mn bond angle) [17]. In previous studies its value has been only roughly estimated as lying in the range between \( t = 0.2 \) and 0.7 eV [11, 15, 18, 19]. Finally, the SE interaction between \( t_{2g} \) spins \( J_{AF} \) was estimated to be only a few meV [15], so we will consider only \( J_{AF} \ll t \) (used in section 3.2), while the JT coupling \( \kappa \) will be treated as a free parameter.

Firstly, we concentrate on the dependence of the magnon dispersion on the Hund coupling. In the strong coupling limit \( (J_H \gg t) \), when \( e_g \) and \( t_{2g} \) electron spins are parallel to each other at each Mn site, the spin excitation is localized and the effective spin–spin interaction has a short ranged character [8]. In such a case, the DE model can be mapped on the Heisenberg model with nearest neighbour interactions between spins \( S \),

\[
H = J_a \sum_{\langle ij \rangle_a} S_i \cdot S_j + J_b \sum_{\langle ij \rangle_b} S_i \cdot S_j + J_c \sum_{\langle ij \rangle_c} S_i \cdot S_j,
\]

(18)

where \( a, b, c \) refer to different directions in the cubic lattice. One can deduce the values of the effective exchange constants \( J_\gamma (\gamma = a, b, c) \) by comparing the results of the numerical calculations with the model dispersion,

\[
\omega_q^H = 2J_a S(1 - \cos q_x) + 2J_b S(1 - \cos q_y) + 2J_c S(1 - \cos q_z).
\]

(19)

For the \( \{(|x| \pm |z|)/\sqrt{2}\} \) OO with large orbital splitting \( (12\kappa \gg t) \) (see figure 2(a)) the in-plane (out-of-plane) effective hopping element is \( t/4 \) \( (t/2) \), respectively, which leads to \( J_a \approx J_b \approx J_c / 2 \) in agreement with our numerical calculation (see figure 2(a)), whereas in the regime of small splitting \( 12\kappa \sim t \) both \( e_g \) bands contribute to the electron motion and one finds an almost isotropic dispersion \( J_a \approx J_b \approx J_c \) (see figure 2(b)). With decreasing \( J_H \) the carriers are more itinerant and the range of effective interactions (mediated by the coherent motion of electrons) increases and the nearest-neighbour Heisenberg model approximation is no longer valid. Another complication (not considered here) is the effect of randomness in (R,A) ion substitution which induces some anomalies in the excitation spectrum near the Fermi wave number [20].

The spin wave bandwidth \( E_{SW} \) evaluated for different strengths of the JT term \( \propto \kappa \) is shown in figure 3 as a function of hole doping. Strong asymmetry in \( E_{SW} \) between doping by \( x \) holes...
Figure 2. Spin wave dispersion $\omega_q$ obtained in the 3D model for the alternating $\{(|x\rangle \pm |z\rangle)/\sqrt{2}\}$ orbital order ($\phi = 0$) at doping $x = 0.3$ for different values of the Hund coupling $J_H/t = 50, 20, 10, 5$ (curves from top to bottom), assuming (a) $\kappa = t$, and (b) $\kappa = 0.1t$. Note a different scale for large and small value of $\kappa$.

Figure 3. The magnon bandwidth $E_{SW}$ in the 3D model for the alternating $\{(|x\rangle \pm |z\rangle)/\sqrt{2}\}$ orbital order ($\phi = 0$) as a function of doping $x$, as obtained for different values of the JT coupling $\kappa/t = 1.0, 0.5, 0.3, 0.1$ (curves from bottom to top) evaluated for $J_H = 10t$. Arrows indicate the maxima in $E_{SW}$.

(e.g. a La compound) and doping by $1 - x$ holes (e.g. a Ca compound) is found for small values of $\kappa$ when both $e_g$ orbitals contribute to carrier propagation in a similar way. In contrast, in the limit $\kappa \to \infty$ one recovers qualitatively the results of the one-band model [8]. However, in this case the magnon bandwidth $E_{SW}$ is anisotropic between different directions, as dictated by the geometry of the lower (partially occupied) $e_g$ orbital.

The model predicts a rapid decrease of the magnon bandwidth $E_{SW}$ with increasing $\kappa$ due to the decrease of the kinetic energy when the OO develops. Moreover, the maximum of $E_{SW}$ moves from $x = 0$ to $1/2$, when the JT orbital splitting increases from 0 to $\infty$. This reflects a transition from the two-band to an effective one-band model, with a half-filled case in the latter model being only a quarter-filled case in the former one. We remark that the width $E_{SW}$ found here for small
Figure 4. Spin wave dispersion $\omega_q$ obtained in the 3D model for the alternating $\{(|x| \pm |z|)/\sqrt{2}\}$ orbital order (solid line), uniform $|x|$ phase (dashed line), and uniform $|z|$ phase (dotted line), respectively. Parameters: $\kappa = t$, $J_H = 10t$, $x = 0.3$.

level splitting (at $\kappa \ll t$) would be further reduced, if the correlation effects (due to finite $U$) were included [21]. For large $\kappa \sim t$, the effect of Coulomb repulsion is included on the mean-field level by the orbital splitting. As a result, for the doping $x = 0.3$ we find $0.3 < E_{SW}/t < 0.7$ (see figure 3) and comparing these values with the experimental data for Pr$_{0.63}$Sr$_{0.37}$MnO$_3$ ($E_{SW} \simeq 80$ meV) or La$_{0.7}$Pb$_{0.3}$MnO$_3$ ($E_{SW} \simeq 100$ meV) [22], one can obtain the estimated values for the hopping element: $115$ meV $< t < 270$ meV or $140$ meV $< t < 330$ meV, respectively. These estimations do not include SE contributions which oppose the DE [21], so higher values of $t$ than those estimated above are expected. We note that the effective Mn–Mn hopping can vary as different ion substitutions in doped manganites affect the length and the angle of Mn–O–Mn bonds.

The dependence of the magnon dispersion $\omega_q$ on the OO is presented in figure 4, in the limit of large orbital splitting ($\kappa = t$). When the uniform $|x|$ orbital phase is stabilized and the effective dimension of the system is reduced to two, one finds large dispersion in the $(a, b)$ planar directions, and almost dispersionless magnon excitation in the $c$ direction. Such an $(x^2 - y^2)$-type of OO, typical for two-dimensional (2D) manganites [12], was reported in the metallic phase of a 3D compound Nd$_{0.45}$Sr$_{0.55}$MnO$_3$ [23]. The present analysis shows that this 2D dispersion indicates strong JT effect. In contrast, for the uniform $|z|$ orbital phase at higher doping ($x \sim 0.7$) [3], the electrons in $e_g$ orbitals propagate mostly along the $c$ direction which gives a strong magnon dispersion for the out-of-plane momenta.

For an intermediate OO ($|\phi| < \pi/4$) one finds anisotropic dispersion (solid line in figure 4). This case might represent the anisotropic FM insulating phase of La$_{1-x}$Ca$_x$MnO$_3$ with $x \leq 0.2$, while isotropic spin waves are observed for $x \geq 0.22$ [6]. The experimental stiffness increases discontinuously at the insulator–metal transition [6], which can be explained by increasing kinetic energy when $\kappa$ decreases (figure 3). Within the present model (1), one comes close to isotropic spin waves with $\kappa = t$ (not shown) for $\phi \simeq -0.04\pi$. Taking the magnon bandwidth, one finds $\omega_{(\pi,\pi,\pi)}/\omega_{(\pi,0,0)} \simeq 3.0$ and $\omega_{(\pi,\pi,0)}/\omega_{(\pi,0,0)} \simeq 2.03$ in $(a, b)$ planes, indeed close to an isotropic dispersion obtained from equation (19), but an orbital liquid [19, 21] or the OO with complex orbital states occupied [24] are yet other possibilities. Isotropic spin waves are distinctly seen in La$_{0.7}$Pb$_{0.3}$MnO$_3$ [9].

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Figure 5. Spin stiffness $D_a$ (see equation (20)) for the doping $x = 0.3$ as a function of inverse Hund coupling $(1/J_H)$ as calculated for the alternating orbital order $\{(|x\rangle \pm |z\rangle)/\sqrt{2}\}$ (a) and the uniform $|x\rangle$ phase (b). Curves (from top to bottom) are for $\kappa/t = 0.1$, 0.3, 0.5, 1.0 and $\infty$.

In the one-band model with nearest-neighbour electron hopping, one finds the isotropic magnon dispersion, and for small momenta $\omega_q = Dq^2$, where $D$ is the spin stiffness constant. In the present two-band model the situation is more complex, leading to the directional dependence of the spin stiffness,

$$\omega_q \simeq D_a q_x^2 + D_b q_y^2 + D_c q_z^2.$$  \hspace{1cm} (20)

In figure 5 we show the spin stiffness as a function of the Hund coupling for different values of the $c_g$ orbital splittings ($\sim \kappa$) evaluated along the (100) direction in the lattice for two different types of OO, realized in doped manganites with low concentration of holes ($x < 0.5$). Assuming the orbital rotation as set by equation (6) one finds $D_a = D_b$.

Similar to the magnon bandwidth discussed above, increasing orbital splitting $\propto \kappa$ causes a decrease in the magnon stiffness (figure 5). This decrease is most pronounced in the planar directions for the alternating OO, where for large $\kappa$ the carrier propagation is only through the $\{(|x\rangle \pm |z\rangle)/\sqrt{2}\}$ channel with a small effective hopping element $t/4$. A similar trend was found in the total magnon dispersion (compare figure 5(a) with figure 4 for the doping $x = 0.3$). In contrast, for the OO with $|x\rangle$ orbitals favoured over the $|z\rangle$ ones, the change in the spin stiffness with increasing $\kappa$ is not so pronounced and undergoes a reduction by about 50% when the orbital splitting is increased. The change of $D$ with $\kappa$ expresses here a gradual reduction of the kinetic energy when the orbital splitting increases. For a realistic $\kappa \sim 0.3t$ one finds $D \sim 0.05t$ for the $\{(|x\rangle \pm |z\rangle)/\sqrt{2}\}$ OO, being only 10 meV for small $t = 200$ meV, indeed close to the values measured in La$_{0.7}$Pb$_{0.3}$MnO$_3$ and in La$_{0.7}$Sr$_{0.3}$MnO$_3$ ($D_{exp} \sim 9$ meV) [9, 10].

3.2. Antiferromagnetic superexchange between core spins

The measured spin wave dispersion results from a superposition of DE and SE terms. Here we investigate the effect of the isotropic AF SE interactions between $t_{2g}$ core spins by adding to the

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spin wave self-energy in equation (16) the term resulting from the Heisenberg Hamiltonian,

\[ H_{SE} = J_{AF} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \]  

with an AF interaction \( J_{AF} > 0 \) which opposes the DE mechanism, leading to the intrinsic frustration of magnetic interactions in doped manganites [21, 25]. The spin wave dispersion estimated in the LSW order in the FM state is now obtained from the self-consistent equation for the self-energy of the form:

\[ \omega_q = \Pi(q, \omega_q) - 4SJ_{AF}[1 - c_+(q)] - 2SJ_{AF}[1 - c_+(q)]. \]  

Using the lowest 1/\( S \) order expansion and assuming \( \Pi(q, \omega_q) \approx \Pi(q, 0) \), equation (22) gives directly the magnon dispersion.

The SE contribution to the effective spin–spin interaction softens the FM magnon spectrum in the entire Brillouin zone (see figure 6). For the doping \( x = 0.3 \) considered by us and large \( e_g \) orbital splitting \( 12\kappa = 1.2t \), the FM ground state gets unstable already for \( J_{AF} \approx 0.04t \) with \( \omega_{(\pi,\pi,\pi)} \rightarrow 0 \). Assuming the SE element \( J_{AF} = 9.2 \text{ meV} \), as estimated in [15], and the Mn–Mn effective hopping element \( t = 400 \text{ meV} \) obtained from the basic electronic structure parameters for the Mn ion [15, 26], one finds the spin wave bandwidth of figure 5 reduced by about 50% due to the SE contribution. The dispersion relation of the spin waves measured in \( \text{La}_{0.7}\text{Pb}_{0.3}\text{Mn}_2\text{O}_7 \) is then quite well reproduced by the dispersion shown in figure 5, assuming a small value of \( J_{AF} \lesssim 0.02t \); yet the dispersion would change when the correlations between \( e_g \) electrons were included [21].

3.3. Bilayer manganites

The spin wave excitations were also measured in FM BL manganites \( \text{La}_{2-x}\text{A}_{x+2}\text{Mn}_2\text{O}_7 \) with \( A = \text{Sr, Ca, Pb} \) [27]–[29], showing that the anisotropy between the intralayer and interlayer
exchange coupling increases very fast with increasing hole doping in the narrow concentration range $0.3 < x < 0.45$ [28, 29]. We limit ourselves to a single BL as the coupling between two neighbouring bilayers is by two orders of magnitude weaker [28], and can be neglected. Thus, in numerical simulations a single BL could be considered to show the microscopic origin of the observed strong asymmetry in the exchange couplings in different directions [30].

Here, we investigate a BL system evaluating the spin wave dispersion on the $50 \times 50 \times 2$ lattice, with the cubic symmetry broken and the $|x\rangle$ orbitals favoured over $|z\rangle$ ones by the crystal-field splitting $\sim 12\kappa$. Consider first the magnon dispersion for the intermediate $|x\rangle - |z\rangle$ orbital splitting ($12\kappa = 3t$) in the doping range $0.1 < x < 0.4$, as shown in figure 7. For $x = 0.4$ we find the in-plane spin stiffness $D \simeq 0.06t$ (for moderate $t = 200$ meV being $D \simeq 12$ meV) close to the value reported for $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ where $D \simeq 9.8$ meV (assuming the lattice constant $a = 1$) [31]. When the doping increases from $x = 0.1$ to $x = 0.4$ the interplane magnetic exchange decreases by a factor of five (due to the decreasing electron density within $|z\rangle$ orbitals [32]), while the intraplane exchange interaction simultaneously increases by about 10%. Both the above trends in the doping dependence of the exchange elements are consistent with the results of neutron experiments [28, 31]. Thus, including the SE processes, the FM interlayer coupling can change sign for larger doping $x \sim 0.45$ [30], leading to the observed crossover from the FM to the A-AF phase [28].

For stronger JT effect ($\kappa \gtrsim 0.3t$), the inter-plane coupling is negligible in the whole range of doping, while for a weak JT coupling ($\kappa \lesssim 0.2t$) the interplane exchange steadily decreases with increasing $x$. Note that by including the electron correlations due to the Coulomb repulsion (excluding multiple occupancy on each Mn site) the in-plane exchange would further decrease for small $x$.

4. Summary and conclusions

In this paper, we have studied the spin excitations in a two-band spin–orbital model with an effective orbital splitting due to the JT coupling which may persist in the FM phase. Our results (most relevant for wide band compounds [3]) predict a drastic change in the spin spectrum when the OO and the type of occupied $e_g$ orbitals change. For finite orbital splitting, the magnon
dispersion, as well as its $q \rightarrow 0$ limit (spin stiffness), reflects the form of the OO in the system, being correlated with the kinetic energy of the $e_g$ electrons in a given lattice direction. While the OO leads to generic anisotropy in the spin excitations, for nearly degenerate $e_g$ orbitals, almost isotropic magnetic excitations were found, similar to those found for the orbital liquid state of disordered $e_g$ orbitals [21].

For the BL system with the $x^2 - y^2$ character of the OO and the orbital level splitting comparable with the electron bandwidth, we found the interlayer coupling changing even by an order of magnitude on increasing hole doping (in the range of $0.2 < x < 0.5$). Similar changes in the exchange elements were measured in La$_{2-x}$Sr$_{1+x}$Mn$_2$O$_7$ [28]. Recent experiments on Pr-doped BL system show that applying a magnetic field might change a paramagnetic insulator into a FM metallic state [31]—here once again the kinetic energy changes, and the DE mechanism plays a prominent role.

Summarizing, the spin wave spectrum could help to identify the character of occupied orbitals in the ferromagnetic manganites with orbital order induced by strong JT coupling. In contrast to the one-band model considered before by Furukawa [8], here a strong anisotropy in the excitation spectrum is generic and reflects the OO, stabilized by the JT terms at larger values of orbital splitting, $\Delta E_{OO} \gtrsim t$ (see figure 4). An extension of the present theory, treating both spin and orbital degrees of freedom dynamically [33], would be required to explain the reported softening of the high frequency spin waves at zone boundary [22].

Acknowledgments

We would like to thank P Horsch, M Hennion and F Moussa for stimulating discussions and for valuable comments. This work was supported by the Polish State Committee of Scientific Research (KBN) under Project No 1 P03B 068 26.

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