Interplanar coupling and the weak ferromagnetic transition in $La_{2-x}Nd_xCuO_4$

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(Received 20 August 1993)

A detailed theoretical analysis of the weak ferromagnetic (WF) transition in the low-temperature orthorhombic and *Pccn* phases of the $La_{2-x}Nd_xCuO_4$ system is presented. This transition arises from a subtle interplay between intraplanar anisotropic Dzyaloshinski-Moriya (DM) interactions and the interplanar coupling between adjacent layers. We model the DM interactions using a modified form of a spin Hamiltonian recently derived by Shekhtman *et al.* [Phys. Rev. Lett. **69**, 836 (1992)], and use symmetry arguments to show that there are only two possible interplanar coupling mechanisms: (1) a distortion-induced isotropic interaction and (2) a pseudo-dipolar interaction. For (1) to account for the antiferromagnetic arrangement of WF moments in low field the interplanar coupling must be *ferromagnetic*, and an explicit calculation for the effective interaction including antiferromagnetic superexchange, ferromagnetic Hund's-rule interaction, and direct ferromagnetic exchange, shows that this is plausible. In the *Pccn* phase, we show that the critical magnetic field required to induce the WF transition depends strongly on whether (1) or (2) is the relevant interplanar coupling mechanism. These results are discussed in the context of recent experiments on the magnetic properties of $La_{2-x}Nd_xCuO_4$ system.

I. INTRODUCTION

After the discovery of superconductivity in Ba and Sr doped La_2CuO_4 it was quickly established through a combination of experimental and theoretical work that the spin degrees of freedom in the undoped material could be approximately described by a two-dimensional, isotropic, spin-1/2 quantum antiferromagnet.[1] There are, however, important corrections to this model, and in this paper we will be concerned with two of these: the intraplanar anisotropic interactions, arising from spin-orbit coupling [Dzyaloshinski-Moriya (DM) interactions[2,3]], and the interplanar magnetic interaction, arising primarily from superexchange processes between the layers. While there has been considerable recent work on the physics of the DM interactions,[4] the interplanar interactions are still poorly understood.

In the low-temperature orthorhombic (LTO) phase of La_2CuO_4, the CuO_6 octahedra making up each CuO layer tilt in a staggered fashion about the $\langle 1\bar{1}0\rangle$ axis. This structural distortion gives rise to DM interactions which cause a small weak ferromagnetic (WF) moment, perpendicular to the CuO plane, to appear in each layer. The size of this WF moment ($\sim 0.003\mu_B$) can be measured by applying a magnetic field B perpendicular to the CuO planes.[5] In the LTO phase, when this field is less than a critical field, $B < B_c \sim 6$ T, the WF moments in successive layers are aligned antiferromagnetically, and when $B > B_c$ this alignment becomes ferromagnetic. At B_c there is a discontinuous jump in the magnetization of the sample which is a measure of the WF moment. In this paper we will refer to this transition as a WF transition. Because the antiferromagnetic alignment of the WF moments in low field is due to the interplanar coupling, measurements of the WF transition provides a potentially useful probe of the physical nature of these interactions.

In this paper we present a detailed theory of the WF transition in La_2CuO_4 and a related series of antiferromagnetic insulating compounds $La_{2-x}Nd_xCuO_4$. When $x \lesssim 0.4$ the latter materials undergo a second structural transition from the LTO phase into a new phase with space group *Pccn*.[6] The interplanar DM interactions induced by the structural distortions in these phases are modeled using a modified version of the spin Hamiltonian recently derived by Shekhtman *et al.*[4] for the LTO phase and the generalization for the *Pccn* phase in Ref. 7. The modification is necessary in order for the classical ground state of the system to agree with recent experiments. Symmetry considerations are used to make a number of nontrivial observations about the interplanar interactions. Because of the crystal structure of La_2CuO_4, any isotropic interplanar coupling between Cu spins is highly frustrated. We find, however, that there is an anisotropic interplanar spin interaction, allowed by symmetry, which couples the in-plane spin components and is *not frustrated* even in the absence of an orthorhombic distortion. When this "pseudodipolar" interplanar coupling is combined with the intraplanar DM interaction, the WF moments are effectively coupled. Rough estimates of the magnitude of this pseudodipolar term indicate that it could be large enough to account for the observed WF transition in the LTO phase of La_2CuO_4. In the LTO phase the orthorhombic distortion partly removes the frustration of the isotropic interaction, and gives rise to an effective coupling mechanism between the WF moments.[8,9] We find that, in order to stabilize the observed zero-field antiferromagnetic arrangement of the WF moments in the LTO phase, this isotropic interaction must be *ferromagnetic*, and by estimating this interaction in perturbation theory we show that this is

plausible. These two coupling mechanisms are the only relevant ones allowed by symmetry, and they are far more effective than the direct isotropic coupling between the small WF moments. Finally, we show that observations of the WF transition in the *Pccn* phase can, in principle, determine which of these mechanisms is dominant.

This paper is organized as follows. In Sec. II, the spin Hamiltonian we use to model the Cu spins in $La_{2-x}Nd_xCuO_4$ is introduced, and a phenomenological discussion of the interplanar coupling mechanisms (distortion-induced isotropic coupling and pseudodipolar coupling) is presented. In Sec. III, the general mean-field equations governing the WF transition are worked out, and it is shown that in the LTO phase either interplanar coupling mechanism could account for the observed critical magnetic field. Section IV then presents calculations of the order of magnitude of these interplanar interactions and shows that either could be strong enough to be the dominant interplanar interaction in the LTO phase. Section V contains a comparison of our results with recent experiments on the magnetic structure of $La_{2-x}Nd_xCuO_4$, comparing the ground state spin configuration and the critical WF transition field with our results. Finally, Sec. VI summarizes the the results of the paper.

II. MODEL HAMILTONIAN

Before proceeding, we state two key assumptions that will be used throughout the paper. First, because the *Pccn* phase is stabilized by randomly placed Nd ions on La sites, this phase naturally contains more structural disorder then the LTO phase of pure La_2CuO_4. While it is possible that this disorder is important for determining the magnetic properties of the *Pccn* phase, in what follows we work within a "virtual crystal" approximation; that is, we assume that there is no disorder and take the average structure to be the overall structure. Second, we assume that the Nd^{3+} moments decouple completely from the Cu spins.

The Hamiltonian that we have used is of the form

$$H = \sum_{\mu} H^{\mu}_{\rm plane} + \sum_{\{\mu\nu\}} H^{\mu\nu}_{\rm interplane} + H_{\rm Zeeman}, \qquad (1)$$

where the indices μ and ν label the CuO planes. For $H_{\rm plane}$ we assume a form based on previous theoretical work.[4,7] Very recently there has appeared controversy about the detailed form of $H_{\rm plane}$ due to experimental observations on the *Pccn* phase.[10,11] We shall show, however, that our main results for $H_{\rm interplane}$ are essentially independent of the yet uncertain features of $H_{\rm plane}$.

A. Intraplanar interactions

For the intraplanar spin-spin coupling we adopt the following Hamiltonian:

$$H^{\mu}_{\rm plane} = \sum_{\{ij\}\in\mu} [J\vec{S}_i\cdot\vec{S}_j + K\vec{S}_i\cdot(\vec{\lambda}_{ij}\vec{\lambda}_{ij} - \vec{\lambda}_{ij}\cdot\vec{\lambda}_{ij}\,\mathbf{I})\cdot\vec{S}_j + D\vec{\lambda}_{ij}\cdot(\vec{S}_i\times\vec{S}_j)], \qquad (2)$$

where the sum is over nearest-neighbor bonds $\{ij\}$ within the CuO plane μ and where

$$\vec{\lambda}_{i,i+\hat{x}} = \sqrt{2}\,(-1)^{x_i+y_i}\Big(0,\sin\chi,0\Big),$$
$$\vec{\lambda}_{i,i+\hat{y}} = -\sqrt{2}\,(-1)^{x_i+y_i}\Big(\cos\chi,0,0\Big), \qquad (3)$$

or

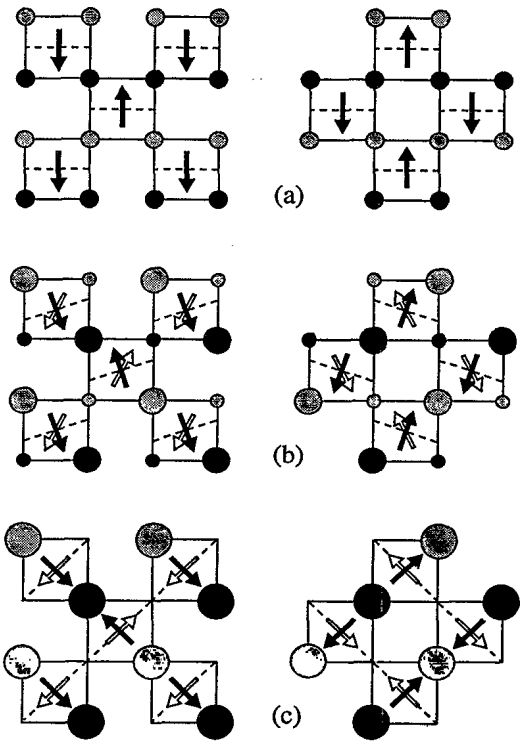

FIG. 1. Tilting patterns of the CuO_6 octahedra and zero-field spin configurations for (a) $\chi \approx \pi/4$ (LTO), (b) $\chi = \pi/8$ (*Pccn*), and (c) $\chi = 0$ (LTT). Two pictures beside one another illustrate the two alternating CuO_2 layers. Tilting axes are shown by dashed lines. O atoms displaced up (down) from the planes are illustrated by black (grey) circles; a large (small) circle corresponds to a large (small) displacement. In-plane magnetic moments are represented by black or white arrows corresponding to the values $C_{JDK} = 1$ or $C_{JDK} = -1$, respectively. In the LTO phase (a), the two configurations are similar. When χ is decreased (b), the in-plane moments remain perpendicular to the tilting axes for $C_{JDK} = -1$, whereas for $C_{JDK} = 1$ they rotate in the opposite direction as a function of χ. In the LTT phase (c), spins are parallel to the tilting axis for $C_{JDK} = 1$ and perpendicular to it for $C_{JDK} = -1$.

$$\vec{\lambda}_{i,i+\hat{x}} = \sqrt{2}\,(-1)^{x_i+y_i}\Big(0,\cos\chi,0\Big),$$
$$\vec{\lambda}_{i,i+\hat{y}} = -\sqrt{2}\,(-1)^{x_i+y_i}\Big(\sin\chi,0,0\Big), \tag{4}$$

when μ is even or odd, respectively. Here the angle χ parametrizes the tilting distortion. The CuO_6 octahedra tilt through an angle δ about first the $(-\cos\chi,\sin\chi,0)$ and then the $(\sin\chi,-\cos\chi,0)$ axis in successive CuO layers (see Fig. 1). In the LTO phase $\chi=\pi/4$, in the *Pccn* phase $0<\chi<\pi/4$, and the case $\chi=0$ corresponds to the low-temperature tetragonal (LTT) phase (space group $P4_2/ncm$) which occurs, for example, in the doped material $La_{1.88}Ba_{0.12}CuO_4$.[13] For the LTO phase, tilting angles $\delta\approx 3°$-$5°$ have been reported; here we take $\delta=5°$ according to the measurements by Jorgensen *et al.*[14] and also use their lattice parameters. The tilting angle δ appears to be essentially independent of χ.[13,11]

Hamiltonian (2) was derived for the LTO phase by Shekhtman *et al.*[4] who calculated the spin-orbit correction to the superexchange within fourth-order perturbation theory in the Cu-O hopping parameter t_{dp}. According to their results, the relative magnitudes of the symmetric and antisymmetric terms of the anisotropic intraplanar coupling should be related by

$$K/J = 1-\sqrt{1-(D/J)^2}. \tag{5}$$

This relation arises from the assumption that the only important source of intraplanar interactions is superexchange via the O p_σ orbitals and the only source of anisotropy is the spin-orbit corrections to this superexchange. The possible importance of other superexchange paths as well as direct exchange is ignored. The relation (5) guarantees that a unitary transformation can be used to transform the spin interaction on a single bond to an isotropic exchange interaction. Any physical anisotropy due to the DM terms (2) is then due to the frustration of these bonds.[4,7]

The classical ground state of (2) as a function of χ was discussed in Ref. 7. If the direction of the antiferromagnetic moments in the xy plane with respect to the x axis is denoted by θ, and the tilting angle with respect to the xy plane is denoted by ϕ, then, to lowest order in ϕ, we have

$$\theta\approx\chi,\qquad \phi\approx\frac{1}{2\sqrt{2}}\frac{D}{J}\sin 2\chi. \tag{6}$$

Figures 1 (a)–(c) (white arrows) show the zero-field configurations for $\chi=\pi/4$ (LTO), $\chi\approx\pi/8$ (*Pccn*), and $\chi=0$ (LTT). In the LTO phase, spins are pointing almost perpendicularly to the tilting axis ($\theta+\chi=\pi/2$), while in the LTT phase they are are parallel to it ($\theta+\chi=0$). The WF moment disappears in the LTT phase.

Experimental studies of the spin configuration and the WF transition in the LTO phase[5] are in good agreement with this picture. They yield for the magnitude of the WF moment $\sin\phi\approx 0.01$,[5] which gives $D/J\approx 0.01$. Recent measurements on the *Pccn* phase,[10,11] however, contradict these predictions. Both Shamoto *et al.*[10] and Keimer *et al.*[11] attempted to determine the ground state spin configuration in the *Pccn* phase of $La_{2-x}Nd_xCuO_4$, $x=0.35$, using neutron scattering. The measured angle of the tilting axis, χ, in both samples was close to zero, corresponding to an almost tetragonal crystal structure. Both groups were able to fit their neutron intensity data by assuming a phase of two coexisting magnetic domains, one in which the WF moments are aligned ferromagnetically, and another in which they are aligned antiferromagnetically. Keimer *et al.* only discuss the possibility that the spins in these domains were perpendicular to the tilt axis, while Shamoto *et al.* were unable to determine whether the spins are nearly parallel or perpendicular to the tilt axis. However, Shamoto *et al.* have also measured the magnetization of their sample. Upon extrapolating to zero temperature they find a spontaneous magnetization corresponding to $1.8\times10^{-3}\ \mu_B$/Cu—a number consistent with a spin canting angle of $\phi\simeq 0.005$, the value observed for the LTO phase of La_2CuO_4, and a domain structure in which only half the moments contribute to the spontaneous magnetization. This result is in conflict with the prediction in Ref. 7 that the WF moment should go to zero as χ goes to zero suggesting that the Hamiltonian (2) should be modified.

The observed spin structures, both in the LTO and in the *Pccn* phases, can be accounted for by a Hamiltonian of the form (2) if only the condition (5) is relaxed. Without this constraint, to lowest order in ϕ, Eqs. (7) are to be replaced by

$$\tan\theta\approx\frac{-C_{JDK}\cos(2\chi)\pm\sqrt{C_{JDK}^2\cos^2(2\chi)+4\sin^2\chi\cos^2\chi}}{2\sin\chi\cos\chi},\qquad \tan\phi\approx\frac{1}{2\sqrt{2}}\frac{D}{J}\sin(\theta+\chi), \tag{7}$$

where $C_{JDK}=4KJ/D^2-1$. The alignment of the in-plane components of the spins for any χ is governed by the parameter C_{JDK}. For any value of C_{JDK}, Eqs. (7) have two solutions, which we refer to by subscripts + and − according to the choice of the sign ± in Eqs. (7). It is easy to show analytically that for any C_{JDK}, the (+)-branch gives the minimum energy in the LTO and LTT phases; we checked numerically that this is the case for intermediate χ also. Figures 2 and 3 give the solutions for some values of C_{JDK}, with $D/J=0.01$ fixed. The case $C_{JDK}=1$ corresponds to the predicted result[7] illustrated in Fig. 1 (white arrows). For $C_{JDK}=-1$, also shown in Fig. 1 (black arrows), spins remain perpendicular to the tilt axis at any χ ($\theta+\chi=\pi/2$), and the canting angle has a nonzero value which is independent of χ. When $|C_{JDK}|$ approaches zero, the spins tend to

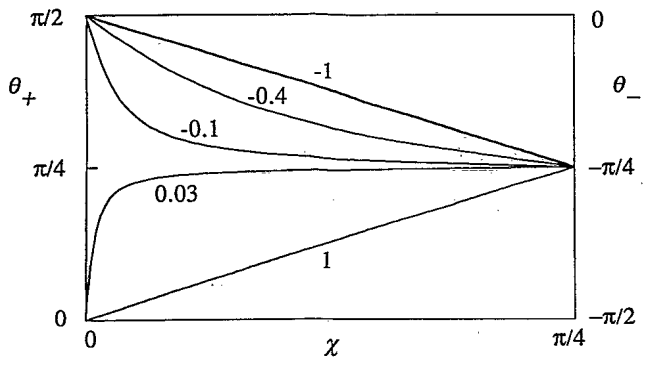

FIG. 2. Angle θ of the in-plane moment alignment as a function of the tilting axis angle χ, shown for five values of C_{JDK}. The θ_+ axis corresponds to the ground state while the θ_- axis shows a saddle-point solution. For $C_{JDK} = -1$, the moments are perpendicular to the tilting axis ($\chi + \theta = \pi/2$) at any χ. For $C_{JDK} = 1$, in-plane moments are perpendicular to the tilting axis in the LTO phase but turn to be parallel to it in the LTT phase ($\chi + \theta = 0$). With decreasing $|C_{JDK}|$, the moments have a growing tendency to direct themselves close to the LTO alignment.

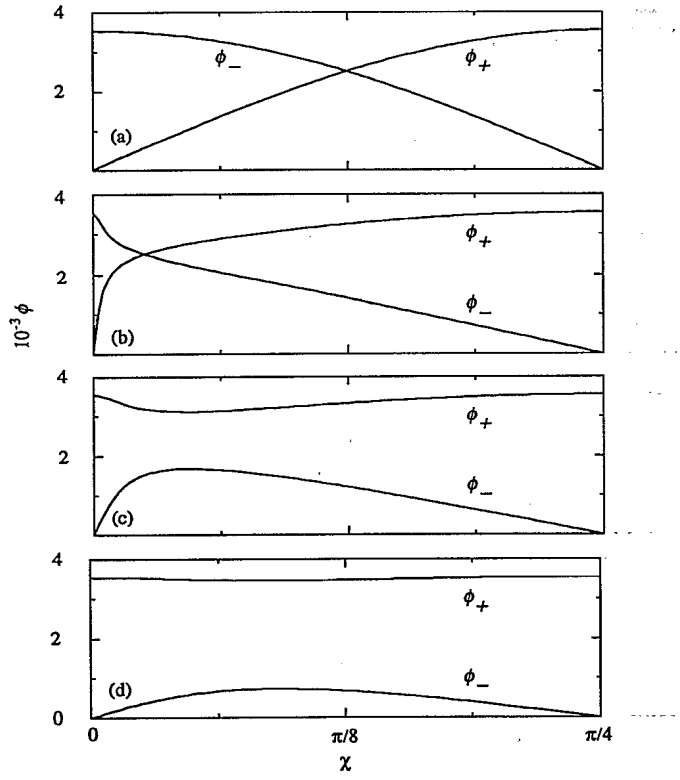

FIG. 3. Canting angle ϕ as a function of the tilting axis angle χ for (a) $C_{JDK} = 1$, (b) $C_{JDK} = 0.03$, (c) $C_{JDK} = -0.1$, and (d) $C_{JDK} = -0.4$. The ground state result is denoted by ϕ_+ while ϕ_- stands for a saddle-point solution. For $C_{JDK} > 0$ [(a) and (b)], ϕ increases from zero at $\chi = 0$ (LTT) to a maximum value at $\chi = \pi/4$ (LTO). For $C_{JDK} < 0$ [(c) and (d)], ϕ is nonzero for all χ, and independent of χ when $C_{JDK} = -1$. The value of C_{JDK} was varied by changing K while $D/J = 0.01$ was fixed.

retain the LTO alignment along the direction $\theta = \pi/4$, and a crossover takes place at $C_{JDK} = 0$.

The experimental observations for the *Pccn* phase obviously indicate that C_{JDK} should be negative, in contradiction with the constraint (5). The value of $J \approx 150$ meV has been well established by various experiments,[12] and the measured value of the WF moment in the LTO phase fixes the ratio $D/J \approx 0.01$; only K is unknown. According to the result shown in Fig. 3, the magnitude of the WF moment depends only very weakly on χ when C_{JDK} is negative. Therefore, observations of the WF moment in the *Pccn* phase do not help to fix K. The constraint $C_{JDK} < 0$ with the above values for J and D gives $K < 0.004$ meV; i.e., the strength of the symmetric anisotropic interaction appears to be less than half of the value predicted by (5).

For the particular case $\chi = 0$, the pure LTT phase, the crossover between two configurations was discussed already by Koshibae *et al.*[15] They argued that spin-orbit corrections to the superexchange via the O p_z orbitals result in an anisotropic interaction which stabilizes an LTT configuration with a nonzero canting angle and an in-plane moment perpendicular to the tilt axis. Shekhtman *et al.*[16] have shown, however, that this contribution is negligible. Very recently, two groups[17,18] have calculated the isotropic interaction term beyond the fourth-order perturbation theory. It is not presently known if spin-orbit corrections to these improved models could lead to relevant changes in the anisotropic interaction terms.

Because of the low symmetry of the crystal, the assumed form of the Hamiltonian (2) cannot be deduced from symmetry arguments. It is, however, a reasonable starting point for our analysis, as (i) all microscopic interactions mechanisms suggested so far fall into this framework and (ii) it includes Hamiltonians consistent with the experimental observations discussed above. In what follows, we will not fix the value of C_{JDK}, and we will consider both cases $C_{JDK} > 0$ and $C_{JDK} < 0$.

Figure 1 clearly shows that the mutual alignment of spins in adjacent layers is largely determined by the angle between the respective tilting axes; symmetry alone dictates, for example, that the antiferromagnetic in-plane moments in the LTO phase are parallel to those in an adjacent layer, while the in-plane moments in the LTT phase are perpendicular to their interplanar neighbors. The unknown details of the intraplanar interactions are, therefore, not very important for our analysis. The most crucial ingredient in the intraplanar coupling to the mechanism of the WF transition arises from the antisymmetric DM interaction, which provides the necessary link between the Zeeman energy and the interplanar interaction.

B. Interplanar interactions

We consider a general bilinear spin-spin interaction between two nearest neighbors in adjacent CuO layers,

$$H^{\mu\nu}_{\text{interplane}} = \sum_{\substack{\{ij\} \\ i\in\mu, j\in\nu}} \vec{S}_i \cdot \mathbf{A}_{ij} \cdot \vec{S}_j, \qquad (8)$$

where $\mathbf{A}_{ij}$ is 3×3 matrix. Constraints on the form of $\mathbf{A}_{ij}$ are imposed by the symmetry of the lattice. First, since the lattice is invariant under an inversion which exchanges the positions of the spins i and j, the matrix $\mathbf{A}_{ij}$ is symmetric. Second, $\mathbf{A}_{ij}$ transforms like a second rank tensor with respect to $\vec{r}_{ij}$, the vector joining spins i and j.

The symmetry properties of $\mathbf{A}_{ij}$ have an important consequence. Let us first consider the interaction in an undistorted lattice ($\delta = 0$). If the staggered magnetizations in the four sublattices are parametrized as

$$\begin{aligned}
\vec{S}_{A1} &= S\,(\cos\theta_1 \cos\phi_1, \sin\theta_1 \cos\phi_1, \sin\phi_1)\,, \\
\vec{S}_{A2} &= S\,(\sin\theta_2 \cos\phi_2, \cos\theta_2 \cos\phi_2, \sin\phi_2)\,, \\
\vec{S}_{B1} &= S\,(-\cos\theta_1 \cos\phi_1, -\sin\theta_1 \cos\phi_1, \sin\phi_1)\,, \\
\vec{S}_{B2} &= S\,(-\sin\theta_2 \cos\phi_2, -\cos\theta_2 \cos\phi_2, \sin\phi_2)\,,
\end{aligned} \tag{9}$$

then the coupling energy arising from (8) takes the form

$$\begin{aligned}
E^{\delta=0}_{\text{interplane}}/N = 4S^2[&J_p \sin\phi_1 \sin\phi_2 \\
&+ d\cos(\theta_1 - \theta_2)\cos\phi_1 \cos\phi_2],
\end{aligned} \tag{10}$$

where $d = A^{xy}_{ij}$ for $x_{ij}y_{ij} > 0$ and $d = -A^{xy}_{ij}$ for $x_{ij}y_{ij} < 0$, and $J_p = A^{zz}_{ij}$. Any other elements of $\mathbf{A}_{ij}$ give zero coupling. In the following we shall therefore assume

$$\mathbf{A}_{ij} = \begin{pmatrix} J_p & d\frac{x_{ij}y_{ij}}{|x_{ij}y_{ij}|} & 0 \\ d\frac{x_{ij}y_{ij}}{|x_{ij}y_{ij}|} & J_p & 0 \\ 0 & 0 & J_p \end{pmatrix}. \tag{11}$$

It is well known that the antiferromagnetic spin components in the CuO_2 planes, $\cos\phi$, are frustrated with respect to an isotropic interaction; the J_p term only couples the WF moments $\sin\phi$. Here we find that the anisotropic "pseudodipolar" interaction d contributes to a direct interaction between the large in-plane components. The strength of this interaction is independent of χ because the x components of the spins are coupled to the y components of the spins in the adjacent layers and vice versa (see Fig. 1).

In the LTO phase, frustration associated with the isotropic interplanar interaction is to some extent removed by the orthorhombic distortion.[8] In the distorted lattice, superexchange paths from a Cu atom to its nearest neighbors in the adjacent layers are not exactly equivalent (see Fig. 4), so that interactions between the large in-plane moments no longer cancel with each other. The distortion can be described as an almost rigid tilting of the CuO_6 octahedra by 5°. The octahedra are also deformed so that the distances between Cu atoms perpendicular to the direction of the tilting axis are slightly larger than those measured along the tilting axis: The ratio of the two is 1.008.[14,19] Because of the latter effect, the central Cu spin in the top layer of Fig. 4 is closer to its interplanar neighbors with opposite moments ($B1$ and $B2$) than those parallel to it ($A1$ and $A2$).

At first sight, this simple picture suggests that the distortion enhances the interaction between antiparallel interplanar neighbors, so that an antiferromagnetic in-

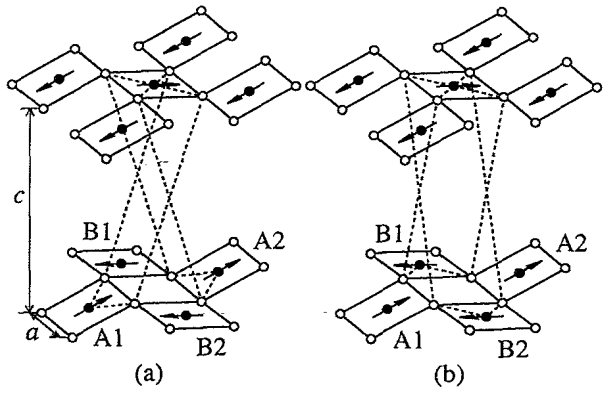

FIG. 4. Example of an interplanar superexchange pattern illustrated for the LTO phase. Open and solid spheres denote O and Cu atoms, respectively. O atoms at the top and bottom of the CuO_6 octahedra are not shown as they are not relevant to the interplanar superexchange mechanism. The octahedra tilt by an angle of 5°. The small canting angle of the spins, $\phi \sim 0.01$, is exaggerated for clarity. Superexchange paths, represented by dashed lines, connect a Cu spin to its interplanar neighbors with (a) parallel ($A1$,$A2$) or (b) antiparallel ($B1$,$B2$) in-plane magnetic moments. Tilting distortion weakens the coupling to $A1$ and enhances it to $A2$, while interactions with $B1$ and $B2$ are unaltered. The net effect enhances superexchange between ferromagnetically aligned in-plane moments.

terplanar interaction is needed in order to the observed ground state configuration to be stable. This was assumed by Berger and Aharony,[9] who modeled the interaction with two unequal exchange constants, $|J_{AA}| < |J_{AB}|$, for interplanar neighbors belonging to the same or to different sublattices. A closer look at the superexchange process shows that this picture is incorrect, and the effect of the distortion actually enhances the interaction between *parallel* interplanar neighbors requiring, consequently, a *ferromagnetic* coupling. Here we give a simple justification of this result; the microscopic calculation is presented in Sec. IV A. The interplanar interaction is predominantly mediated by hopping processes via the O atoms in the CuO_2 layers. Instead of comparing straight distances between Cu atoms, one must consider how the superexchange paths are altered by the distortion. Because of symmetry, hopping processes from a Cu atom to any of its neighboring O's are equivalent. The effect which removes frustration arises, therefore, solely from the interplanar hopping processes between these O atoms. The orbitals of these O's do not tilt, because they are shared by two octahedra which tilt in opposite directions. The effect of the distortion is then only to displace rigid p orbitals relative to each other. Hopping integrals t_{pp} between any two p orbitals depend on distance r as $1/r^2$, which gives a factor $1/r^4$ in the strength of the superexchange. Let us consider the effective geometrical factor

$$\left[\frac{1}{r^4}\right]_{\text{eff}} = \frac{1}{4}\left(-\frac{1}{r^4_{A1}} - \frac{1}{r^4_{A2}} + \frac{1}{r^4_{B1}} + \frac{1}{r^4_{B2}}\right), \tag{12}$$

where $r_{A1}r_{A2}$ and $r_{B1}r_{B2}$ are the distances between two O atoms connecting Cu atoms to its parallel or antiparallel interplanar neighbors. The most simple hopping pattern, in which holes are transferred between O atoms located directly on top of each other, obviously gives $[1/r^4]_{\rm eff} = 0$. Figure 4 shows an example of a hopping pattern which contributes to a nonzero $[1/r^4]_{\rm eff}$. Owing to the tilting distortion, r_{A1} decreases and r_{A2} increases, while r_{B1} and r_{B2} are unaltered: Instead of $|J_{AA}| < |J_{AB}|$ we have $|J_{AA2}| < |J_{AB1}| = |J_{AB2}| < |J_{AA1}|$. The net effect enhances interaction between parallel interplanar neighbors: $|\frac{\delta}{\delta r}(1/r^4)|$ is largest at small r so that the decrease of r_{A1} gives the dominant effect, and $|J_{AA1}| + |J_{AA2}| > |J_{AB1}| + |J_{AB2}|$. For rigid tilting of the octahedra by an angle δ with respect to a tilting axis in an angle χ,

$$\left[\frac{1}{r^4}\right]_{\rm eff} \sim -\sin(2\chi)\sin^2\delta. \tag{13}$$

The displacements of the O atoms are proportional to $\sin\delta$, but the first-order terms in $\sin\delta$ are canceled out in Eq. (13). We must, therefore, also take into account the mentioned small deformation of the octahedra. This correction weakens the coupling due to tilting as it increases r_{A1} and r_{A2} relative to r_{B1} and r_{B2}. In the calculation in Sec. IV A this correction is taken into account.

Although the effective interaction is not a direct consequence of the orthorhombic distortion—which exists in the LTO phase but *not* in the LTT phase—but rather a consequence of the tilting octahedra—which takes place in *both* the LTO and in the LTT phase—it still disappears in the LTT phase. This is due to the fact that the tilting pattern in the LTT phase modifies the superexchange paths to sublattice A precisely in the same way as those to sublattice B (see Fig. 1). The factor $\sin(2\chi)$ in Eq. (13) arises from this effect.

The resulting effective interaction energy has the form

$$E^{\delta^2}_{\rm interplane}/N = -4S^2\gamma J_p \sin^2\delta\sin(2\chi) \times \sin(\theta_1+\theta_2)\cos\phi_1\cos\phi_2. \tag{14}$$

The geometrical factor γ and the coupling strength J_p must be calculated separately for all possible superexchange paths. The minus sign implies that in order to stabilize the ground state spin configuration observed in experiments, J_p should be negative; i.e., the superexchange process should be *ferromagnetic*. This question will be considered in detail in Sec. IV A.

Both the pseudodipolar and the distortion-induced interplanar coupling mechanisms are essentially independent of whether we assume that $C_{JDK} > 0$ or $C_{JDK} < 0$. For $C_{JDK} > 0$, $\theta_1 = \theta_2 \approx \chi$, and for $C_{JDK} < 0$, $\theta_1 = \theta_2 \approx \pi/2 - \chi$ (except at very small values of $|C_{JDK}|$ which yield $\theta_1 = \theta_2 \approx \pi/4$). In both cases, $\cos(\theta_1 - \theta_2) = 1$ and $\sin(\theta_1 + \theta_2) \approx \sin(2\chi)$. Equations (10) and (14) then show that the pseudodipolar interaction is independent of χ whereas the distortion-induced interaction disappears in the LTT phase.

III. WEAK FERROMAGNETIC TRANSITION

We now consider the total energy of the spin system. We first introduce a function $E_{\rm plane}(\phi,\theta)$ which gives the energy due to intraplanar interactions only:

$$E_{\rm plane}(\phi,\theta) = -2J\cos(2\phi) - (\sqrt{2}D)\sin(\chi+\theta)\sin(2\phi) + 2K[(\sin^2\chi\cos^2\theta + \cos^2\chi\sin^2\theta)\cos^2\phi - \sin^2\phi]. \tag{15}$$

The intraplanar Hamiltonian, Eq. (2), is not invariant under a reflection with respect to the xy plane. This asymmetry is caused by the tilting CuO_6 octahedra, breaking the symmetry corresponding to an inversion which interchanges the positions of two neighboring Cu atoms. Consequently, the factor $\sin(2\phi) \sim \sin\phi\cos\phi$ in the D term of Eq. (15) requires that a flip of the z components of the spins, $\sin\phi \to -\sin\phi$, be accompanied by a flip of the xy components also, $\cos\phi \to -\cos\phi$.

Using Eqs. (10), (14), and (15), the total energy can be written

$$E/N = S^2\{4J_p[\sin\phi_1\sin\phi_2 - C\sin^2\delta\sin(2\chi)\sin(\theta_1+\theta_2)\cos\phi_1\cos\phi_2] + 4d\cos(\theta_1-\theta_2)\cos\phi_1\cos\phi_2 + \tfrac{1}{2}[E_{\rm plane}(\phi_1,\theta_1) + E_{\rm plane}(\phi_2,\theta_2)] - (B/2S)(\sin\phi_1+\sin\phi_2)\}. \tag{16}$$

The first J_p term (with $J_p > 0$) favors an antiferromagnetic alignment of the WF moments, $\sin\phi_2 = -\sin\phi_1$, while the external magnetic field B tends to align them ferromagnetically, $\sin\phi_2 = \sin\phi_1$. At first sight of Eq. (16), this seems to be the only term to play any role in the WF transition since the two succeeding interplanar terms only couple the in-plane $\cos\phi$ components. Indeed, in the first theoretical studies of the WF transition only this coupling was considered.[20] Because of the mentioned asymmetry of the D term in Eq. (15), however, also $\cos\phi_1\cos\phi_2$ is forced to change sign as the transition takes place. The pseudodipolar interaction and the distortion-induced coupling are thus, indirectly, connected to the Zeeman energy. These interplanar interaction mechanisms provide the coupling between the in-plane moments in adjacent CuO_2 layers, the DM interaction couples the in-plane moments to WF moments, and the latter are finally coupled to the external field. A comparison of the orders of magnitude of the relevant geometrical factors in Eq. (16), $\sin^2\phi \sim 10^{-5}$ (direct),

$\sin^2\delta \sim 10^{-2}$ (distortion-induced), and $\cos^2 \sim 1$ (pseudodipolar), shows that the two indirect interaction mechanisms are far more effective than the direct isotropic coupling. The remaining question is whether it is the distortion coupling or the pseudodipolar interaction that plays the dominant role in the WF process.

Equations for the stable configurations, as functions of the field, were found by minimizing E in Eq. (16):

$$\pm K\cos(2\chi)\sin(2\theta_l)\cos\phi_l - \sqrt{2}D\cos(\theta_l+\chi)\sin\phi_l + 4d\sin(\theta_1-\theta_2)\cos\phi_m - 4J_p\gamma\sin^2\delta\sin(2\chi)\cos(\theta_1+\theta_2)\cos\phi_m = 0,$$

$$4(2J-K-K\sin^2\chi)\cos\theta_l\sin\phi_l\cos\phi_l \pm 2\sqrt{2}D[\sin\chi\sin^2\phi_l - \sin(\theta_l+\chi)\cos\theta_l\cos^2\phi_l] \tag{17}$$

$$+8J_p\cos\theta_l\cos\phi_l\sin\phi_m + 8d\cos\theta_m\cos\phi_m\sin\phi_l - 8J_p\gamma\sin^2\delta\sin(2\chi)\sin\theta_m\sin\phi_l\cos\phi_m - (B/S)\cos\theta_l\cos\phi_l = 0.$$

The $\pm$ sign corresponds to $+$ for $(l,m) = (1,2)$ and $-$ with $(l,m) = (2,1)$, respectively. Equations (17) were solved numerically as a function of the field. We then evaluated the required strengths of the two interactions to account for the observed WF transition in the LTO phase. For the coupling parameters, we chose $J = 150$ meV and $D = 1.5$ meV which give $D/J = 0.01$, consistent with measurements of $\sin\phi$. In order to compare the effects of the isotropic distortion-induced coupling and the pseudodipolar interaction we considered two models, model I with $J_p < 0$ and $d = 0$ and model II with $J_p = 0$ and $d > 0$. The magnitudes of the nonzero J_p and d were fitted to reproduce a value of $B_c = 5.7$ T consistent with the experimental results for the critical field of the WF transition in the LTO phase.[5] For model I we obtained $J_p\gamma\sin^2\delta = -6.4\times10^{-4}$ meV which corresponds to $J_p \sim -0.1$ meV. With $d = 6.4\times10^{-4}$ meV for model II, the two models yield coinciding results for the LTO phase. The parameter K does not enter into these considerations; they thus hold for any value of C_{JDK} as long as J and D are fixed.

IV. INTERPLANAR INTERACTIONS: MICROSCOPIC THEORY

A. Distortion-induced isotropic interaction

We shall now consider in detail the effective interplanar coupling due to the tilting distortion. The O atoms at the top and at the bottom of the octahedra can be omitted, as it was found that they do not essentially contribute to the interplanar superexchange: All the three $2p$ orbitals of these atoms are orthogonal to the $3d_{x^2-y^2}$ orbital of the Cu atom, inhibiting direct hopping to these O's, and the hopping processes to these O's via the four O atoms in the CuO_2 layer almost cancel each other by destructive quantum interference. Tilting of the octahedra gives rise to a small net transfer by the latter process but the resulting coupling is too small to be relevant. Instead, the largest contributions to the interplanar superexchange arise from processes such as the one shown in Fig. 4, in which a hole first hops from the Cu $3d_{x^2-y^2}$ orbital to the $2p_\sigma$ orbital of a neighboring O atom in the CuO_2 layer, and continues directly to one of the three $2p$ orbitals of an O atom in an adjacent plane. A hole from a neighboring Cu atom in this layer hops to the p_σ orbital of the same O atom, giving rise to superexchange, or interacts by direct exchange.

We systematically considered all possible superexchange paths which connect two neighboring Cu atoms in adjacent layers via the O atoms that belong to the corresponding octahedra. We ruled out (i) paths which give zero contribution due to destructive quantum interference. Also we omitted (ii) paths for which the total coupling to the two sublattices remains equal even in the presence of the orthorhombic distortion or (iii) which give a contribution of higher order than δ^2; furthermore, (iv) some hopping patterns exactly cancel with each other. Applying these criteria, only few of the vast number of initial possibilities turned out to be relevant.

The paths which give rise to an unfrustrated interplanar exchange of order δ^2 fall into four groups according to the hopping pattern

$$\begin{aligned}
&(1)\ \vec{R}_1 = \frac{a}{2}\hat{\mathbf{y}},\\
&\qquad \vec{R}_2 = \frac{a}{2}(\hat{\mathbf{x}}+\hat{\mathbf{y}}) + c\hat{\mathbf{z}},\\
&\qquad \vec{R}_3 = -\frac{a}{2}\hat{\mathbf{y}},\\
&(2)\ \vec{R}_1 = -\frac{a}{2}\hat{\mathbf{x}},\\
&\qquad \vec{R}_2 = a(\hat{\mathbf{x}}+\hat{\mathbf{y}}) + c\hat{\mathbf{z}},\\
&\qquad \vec{R}_3 = -\frac{a}{2}\hat{\mathbf{y}},\\
&(3)\ \vec{R}_1 = -\frac{a}{2}\hat{\mathbf{x}},\\
&\qquad \vec{R}_2 = \frac{3a}{2}\hat{\mathbf{x}} + \frac{a}{2}\hat{\mathbf{y}} + c\hat{\mathbf{z}},\\
&\qquad \vec{R}_3 = -\frac{a}{2}\hat{\mathbf{x}},\\
&(4)\ \vec{R}_1 = \frac{a}{2}\hat{\mathbf{y}},\\
&\qquad \vec{R}_2 = a\hat{\mathbf{x}} + c\hat{\mathbf{z}},\\
&\qquad \vec{R}_3 = -\frac{a}{2}\hat{\mathbf{x}}.
\end{aligned} \tag{18}$$

The vector $\vec{R} = \vec{R}_1 + \vec{R}_2 + \vec{R}_3$ connects two Cu atoms in adjacent layers. The particular hopping pattern shown in Fig. 4 corresponds to path (1). In each hopping scheme, the hop along $\vec{R}_1$ takes place from the $3d_{x^2-y^2}$ orbital of a Cu atom to the $2p_\sigma$ orbital of an O atom in the same CuO layer; hopping integrals into the two other $2p$ orbitals vanish. The hole continues along $\vec{R}_2$ to any of the three p orbitals of an O atom in the adjacent CuO layer. A hole from a neighboring Cu atom in this layer hops along $-\vec{R}_3$ to the p_σ orbital of the same O atom. The paths to and from the adjacent plane can differ; possible combinations of the paths (1)−(4) then give rise to nine hopping patterns. Path (4) only gives a nonzero contribution in combination with another path (1)−(3).

Geometrical considerations show that for these superexchange patterns the tilting distortion enhances superexchange between parallel interplanar neighbors relative to that between antiparallel ones. The conclusion is that in order to stabilize the observed zero-field state in the LTO phase, the interaction should be *ferromagnetic.*

We now consider this possibility. In insulators, there are three distinct mechanisms which lead to effective spin-spin interactions.[21] Superexchange processes involving virtual intermediate states with two holes in one atomic orbital give rise to an antiferromagnetic coupling due to the Pauli exclusion principle. It is this mechanism which usually dominates in insulators and makes them antiferromagnets. If the dominant superexchange path is such that the holes interact at mutually orthogonal orbitals of the same atom, however, ferromagnetic coupling results (see Fig. 5). In this case the Coulomb force between the holes causes a mixing of the orbitals into singlet and triplet states and, according to the Hund's rule, triplet states with parallel spins are favored. The ferromagnetic interaction is in general weaker than the antiferromagnetic one by a factor of $\Delta E_{\rm st}/U$, $\Delta E_{\rm st}$ is the singlet-triplet splitting and U is the Coulomb repulsion. The third and typically the weakest mechanism is the ferromagnetic direct exchange arising from singlet-triplet splitting of orbitals located on separate atoms.

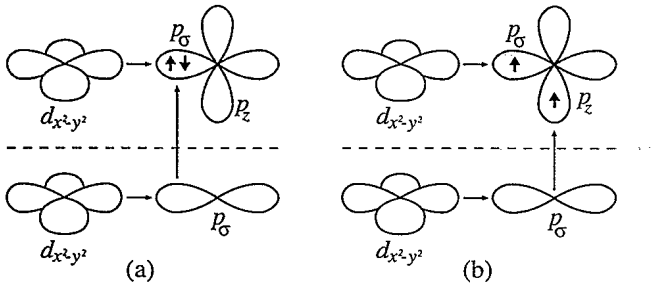

FIG. 5. Schematic illustration of superexchange processes giving rise to antiferromagnetic and ferromagnetic interplanar interactions. A hole hops from the $d_{x^2-y^2}$ orbital of a Cu atom to the p_σ orbital of a neighboring O. (a) An interplanar hop between two O's which takes place into a p_σ orbital gives rise to an antiferromagnetic coupling. (b) It is about 4 times more probable that an interplanar hop ends up into a p_z orbital which extends far below and above the O site. This process gives rise to ferromagnetic coupling. The third interaction mechanism arises from direct ferromagnetic exchange between one hole remaining at the $d_{x^2-y^2}$ orbital and another entering a neighboring p orbital by an interplanar hop.

Details of the microscopic calculation are presented in the Appendix. The resulting effective interactions can be written as products of two factors: $J_{\rm eff} = K_{\rm Cu-O} \times K_{\rm hopping}$. $K_{\rm Cu-O}$ stands for the strength of the coupling between a hole at the Cu atom and another at a neighboring O, while $K_{\rm hopping}$ is proportional to the effective interplanar hopping amplitude t_{pp}^2 evaluated for the nonfrustrated part of the process. We find that $K_{\rm Cu-O}$ for the antiferromagnetic superexchange mechanism is, indeed, 6 times as large as that for the ferromagnetic Hund's-rule superexchange. The direct ferromagnetic exchange is twice as strong as the Hund's-rule ferromagnetic interaction. However, because of the particular mutual orientations of orbitals in $La_{2-x}Nd_xCuO_4$, the interplanar hopping process strongly favors the Hund's-rule interaction: $K_{\rm hopping}$ for the Hund's-rule coupling is 4 times as large as that for the antiferromagnetic superexchange and that for the direct interaction. As illustrated in Fig. 5, p_z orbitals extend far below and above the O sites, so that interplanar hopping processes $p_\sigma \to p_z$ which bring the hole to an orbital orthogonal to a p_σ orbital are dominant. The resulting antiferromagnetic (AFM) and ferromagnetic (FM) interactions are then approximately equal: $[J^{\rm AFM}]_{\rm eff} = 8 \times 10^{-4}$ meV, $[J^{\rm FM}_{\rm Hund}]_{\rm eff} = 5 \times 10^{-4}$ meV, and $[J^{\rm FM}_{\rm direct}]_{\rm eff} = 3 \times 10^{-4}$ meV, giving $J^{\rm AFM} = J^{\rm FM} = 8 \times 10^{-4}$ meV. The above value for $[J^{\rm FM}_{\rm direct}]_{\rm eff}$ only includes the effect involving the O p_σ orbitals. Direct exchange between a p_π orbital and a Cu $d_{x^2-y^2}$ orbital gives rise to an additional ferromagnetic correction for which we found a rough estimate $[J^{\rm FM\pi}_{\rm direct}]_{\rm eff} \sim 1 \times 10^{-4}$ meV.

We conclude that it is indeed plausible that the isotropic interplanar interaction is ferromagnetic and thus it may play the relevant role in the WF transition. We remark also that the obtained values are of the same order of magnitude as the required coupling strength, 6×10^{-4} meV, to obtain the measured critical field of the WF transition.

The presented estimates include the effect of frustration. It is also of interest to find the plain magnitude of J_p, the interplanar interaction between two Cu atoms; in particular, its magnitude is needed to estimate the strength of the pseudodipolar coupling. To evaluate J_p, the frustration factor $\sin^2\delta$ must be divided out. Another correction arises from the fact that all superexchange paths which give no δ^2 contribution were omitted in the above calculation. There are approximately 4 times more superexchange patterns actually contributing to J_p, which leads to a rough estimate $|J_p| \sim (4/\sin^2\delta) \times 10^{-4}$ meV−0.1 meV. We conclude that the ferromagnetic interaction is indeed roughly equal in strength to the antiferromagnetic interaction, and within the accuracy of our calculation the net interaction can have either sign.

B. Pseudodipolar interaction

As discussed in Sec. II, it is also possible that the relevant interplanar coupling is due to the pseudodipolar

interaction, assuming that this is the only source of interplanar coupling which would require that $d \sim 6\times10^{-4}$ meV. Two possible contributions to d are the direct magnetic dipolar interaction and the "pseudodipolar" interaction arising from spin-orbit corrections to the isotropic interplanar exchange discussed above and in the Appendix.

The direct magnetic dipolar interaction between nearest-neighbor Cu spins on adjacent layers yields a value of $d \simeq 1.4 \times 10^{-4}$meV, a result which is close to the required value. Unfortunately, when the long-range $(1/r^3)$ contribution of the dipolar interaction is included in the calculation, the effective d is reduced by a factor of 6. We conclude, therefore, that the direct magnetic dipolar interaction is too small to account for the observed WF transition.

In principle, it is possible to calculate the pseudodipolar contribution to d by including spin-orbit coupling in the calculation of the isotropic coupling. Rather than performing this calculation, it is possible to make a rough estimate of the magnitude of this term by noting that, on general grounds, symmetric anisotropic corrections, both to superexchange and direct exchange, are expected to be on the order of $\sim (\Delta g/g)^2 J_p$ where g is the g factor of the Cu spins, and Δg is the shift in the g factor due to spin-orbit coupling.[2,22] In the cuprates $\Delta g/g \sim 0.1$,[5] which, taking our rough estimate of $J_p \sim 0.1$ meV, yields $|d| \sim 10 \times 10^{-4}$ meV, which is indeed sufficiently large in comparison with the required value $|d| \sim 6 \times 10^{-4}$ meV. We conclude that the pseudodipolar contribution to d can, in principle, provide an interplanar coupling which is strong enough to account for the observed WF transition.

V. COMPARISON WITH EXPERIMENT

Since we cannot *a priori* determine whether the relevant interplanar coupling mechanism is due to the distortion-induced isotropic interaction, the pseudodipolar interaction, or both, it is of interest to find an experimental test which is sensitive to the physical nature of these couplings. Such a test is provided in principle by experiments on the magnetic structure and weak ferromagnetic transition in the *Pccn* phase of $La_{2-x}Nd_xCuO_4$,[10,11] discussed already in Sec. II A.

The two models I ($J_p < 0, d = 0$) and II ($J_p = 0, d > 0$) result in qualitatively different behavior of the critical field B_c as a function of the structural parameter χ. Let us consider the system when $B = B_c$. Approximating $|\phi_1^{\rm LF}| = |\phi_2^{\rm LF}| = |\phi^{\rm HF}|$ and $\theta_1^{\rm LF} = \theta_2^{\rm LF} = \theta^{\rm HF}$ gives $E_{\rm plane}^{\rm LF} = E_{\rm plane}^{\rm HF}$, where LF and HF stand for the low- and high-field phases, respectively. Equation (16) then gives

$$B_c \approx 8S\left(-\frac{J_p}{\sin\phi}\sin^2\delta\sin(2\chi)\sin(\theta_1+\theta_2)+\frac{d}{\sin\phi}\right). \tag{19}$$

For $C_{JDK} = 1$, the zero field estimates (7) yield $\sin\phi \propto \sin(2\chi)$ and $\sin(\theta_1+\theta_2) \propto \sin(2\chi)$, from which we obtain

$$\begin{aligned} &\text{(I)}\ \ J_p < 0\ ,\ d = 0 : B_c \propto \sin(2\chi), \\ &\text{(II)}\ \ J_p = 0\ ,\ d > 0 : B_c \propto \frac{1}{\sin(2\chi)}. \end{aligned} \tag{20}$$

For $C_{JDK} = -1$, Eqs. (7) give $\sin\phi \propto 1$ and $\sin(\theta_1 + \theta_2) \propto \sin(2\chi)$, yielding

$$\begin{aligned} &\text{(I)}\ \ J_p < 0\ ,\ d = 0 : B_c \propto \sin^2(2\chi), \\ &\text{(II)}\ \ J_p = 0\ ,\ d > 0 : B_c \propto 1. \end{aligned} \tag{21}$$

Figures 6 and 7 illustrate the magnetizations calculated for $C_{JDK} = 1$ and $C_{JDK} = -1$; results for models I and II are plotted in (a) and (b), respectively. The three curves correspond to structural phases with $\chi = 0$ (LTT), $\chi = \pi/8$ (*Pccn*), and $\chi = \pi/4$ (LTO).

For an arbitrary C_{JDK}, $-1 \le C_{JDK} \le 1$, we find the following result: Systems with χ lower than $\pi/4$ should have critical fields lower than that measured for the LTO phase; model II predicts an opposite behavior of increasing or at least constant B_c when χ is lowered. This provides an experimental scheme to decisively test whether the origin of the interplanar coupling mechanism is primarily anisotropic or not.

The coexistence of magnetic domains in which the WF moments are antiferromagnetically aligned with those in which they are ferromagnetically aligned indicates that in the *Pccn* phase of $La_{2-x}Nd_xCuO_4$ the effective interplanar coupling is much smaller than in the LTO phase; consequently, $B_c \approx 0$. If this observation is interpreted

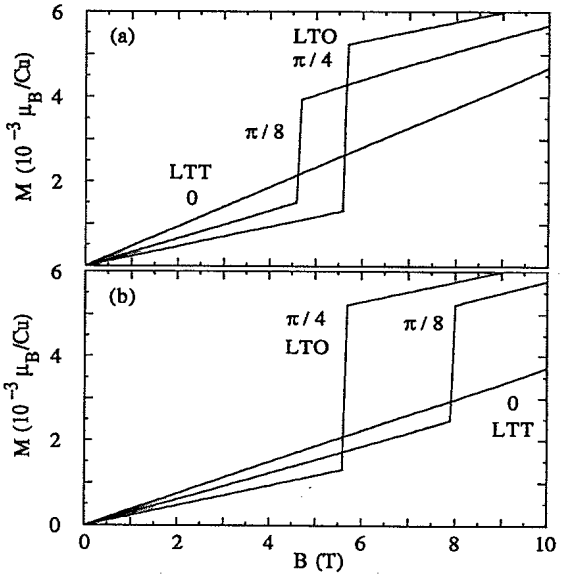

FIG. 6. Induced magnetization M as a function of external field B with $C_{JDK} = 1$, calculated for three values of χ. (a) Model I with $J_p\gamma\sin^2\delta = -6.4 \times 10^{-4}$ meV ($J_p \sim -0.1$ meV) and $d = 0$: $B_c \propto \sin(2\chi)$ decreases with decreasing χ. (b) Model II with $J_p = 0$ and $d = 6.4 \times 10^{-4}$ meV: $B_c \sim 1/\sin(2\chi)$ increases with decreasing χ.

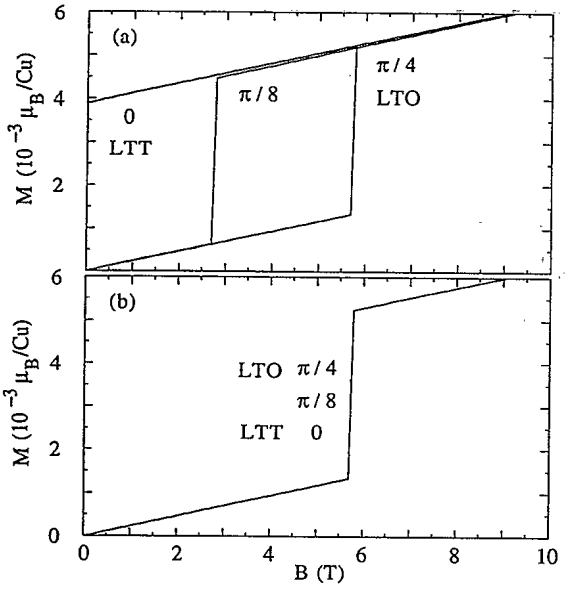

FIG. 7. Induced magnetization M as a function of external field B with $C_{JDK} = -1$, calculated for three values of χ. (a) Model I with $J_p\gamma\sin^2\delta = -6.4\times10^{-4}$ meV ($J_p \sim -0.1$ meV) and $d = 0$: $B_c \sim \sin^2(2\chi)$ decreases with decreasing χ. (b) Model II with $J_p = 0$ and $d = 6.4\times10^{-4}$ meV: Magnetization and B_c are independent of χ.

in terms of the calculations presented above, it suggests strongly that the dominant interplanar coupling is due to the distortion-induced isotropic interaction and not the pseudodipolar interaction.

VI. SUMMARY

To conclude, we have presented a detailed theory of the WF transition in the LTO and *Pccn* phases of the $La_{2-x}Nd_xCuO_4$ system. The interplanar spin interactions were modeled using a modified form of a recently derived Hamiltonian[4,7] which includes the DM interactions due to spin-orbit corrections to superexchange. Symmetry considerations were then used to show that there are two possible interplanar coupling mechanisms: the pseudodipolar coupling and the distortion-induced isotropic coupling. In order for the distortion-induced isotropic coupling to be consistent with the observed AFM arrangement of the WF moments in low field, this interaction must be ferromagnetic, and a microscopic calculation of the interplanar superexchange, including antiferromagnetic superexchange, ferromagnetic Hund's-rule interaction, and direct ferromagnetic exchange, showed that this is indeed plausible. The dependence on the structural parameter χ of the critical field required to induce the WF transition was then shown to be a sensitive test of the relative importance of these two coupling mechanisms. Comparison with recent experiments on the $La_{2-x}Nd_xCuO_4$ system show that it is likely that the distortion-induced interaction is the dominant source of interplanar coupling.

ACKNOWLEDGMENTS

We wish to thank R. Fehrenbacher, L. Shekhtman, R. Hlubina, and T. M. Rice for useful conversations. In particular we would like to thank T. M. Rice for bringing the possibility of ferromagnetic superexchange to our attention. This work was supported by grants from the Magnus Ehrnrooth Foundation (H.E.V.) and the Swiss National Fund (N.E.B. and H.E.V.).

APPENDIX: ISOTROPIC INTERPLANAR SUPEREXCHANGE IN $La_{2-x}Nd_xCuO_4$

Let us consider a single superexchange path which connects two neighboring Cu atoms in adjacent layers via two O atoms. The Cu-Os-O-Cu bond is described by the Hamiltonian

$$H = H_0 + H', \tag{A1}$$

with

$$\begin{aligned} H_0 = {} & \Delta_{dp}\sum_{i\alpha} p^\dagger_{i\alpha}p_{i\alpha} \\ & + U_{dd}(n_{d1\uparrow}n_{d1\downarrow} + n_{d2\uparrow}n_{d2\downarrow}) + U_{pp}\sum_i n_{i\uparrow}n_{i\downarrow} \\ & + \sum_{\{i,j\}}\left[J_{\rm Hund}\vec{s}_i\cdot\vec{s}_j + U_{\rm Hund}\sum_{\alpha,\beta} n_{i\alpha}n_{j\beta}\right] \\ & + J_{\rm direct}\left[\vec{S}_{d1}\cdot\sum_{i(d1)}\vec{s}_{i(d1)} + \vec{S}_{d2}\cdot\sum_{i(d2)}\vec{s}_{i(d2)}\right], \end{aligned} \tag{A2}$$

$$\begin{aligned} H' = {} & \sum_{i\alpha}\{t_{dp}(d^\dagger_{1\alpha}p_{i\alpha} + d^\dagger_{2\alpha}p_{i\alpha}) + \text{H.c.}\} \\ & + \sum_{ij}\sum_{\alpha}\{t_{ij}p^\dagger_{i\alpha}p_{j\alpha} + \text{H.c.}\}. \end{aligned}$$

The operators $d^\dagger_{1\alpha}$, $d^\dagger_{2\alpha}$, and $p^\dagger_{i\alpha}$ create holes with spin α on the $3d_{x^2-y^2}$ orbitals of the two copper atoms (1 and 2) and the oxygen $2p$ orbitals, respectively; $\vec{S}_{d\{1,2\}} = d^\dagger_{\{1,2\}}\vec{\sigma}d_{\{1,2\}}$ and $\vec{s}_i = p^\dagger_i\vec{\sigma}p_i$. Summations over index i are taken over all p orbitals of O atoms in the superexchange path, and $\{i,j\}$ denote mutually orthogonal p orbitals on one O atom. Summations over indices $i(d1)$, $i(d2)$ are taken over the four p_σ orbitals surrounding Cu atoms 1 or 2, respectively. Parameter values $\Delta_{dp} = 3.6$ eV, $U_{dd} = 10.5$ eV, $U_{pp} = 4$ eV, and $t_{dp} = 1.3$ eV calculated by Hybertsen *et al.*[23] were used. While we take $\Delta_{dp} = \Delta_\sigma = 3.6$ eV for holes on O p_σ orbitals, we use a slightly larger value $\Delta_{dp} = \Delta_\pi = 4.6$ eV for the two p_π orbitals, consistent with the result by Stechel and Jennison.[24] The Coulomb force acting on holes at orthogonal p orbitals results in a singlet-triplet splitting with $J_{\rm Hund} = -5.50$ eV and $U_{\rm Hund} = 3.17$ eV.[25] The direct ferromagnetic interaction between a Cu $3d_{x^2-y^2}$ orbital and a neighboring O p_σ orbital corresponds to $J_{\rm direct} = -0.44$ eV.[24] Hopping integrals t_{ij} between O

orbitals are to be calculated separately for each superexchange path.

The unperturbed Hamiltonian (A2) is first diagonalized; this involves only the Hund's term and the direct ferromagnetic term for which singlet-triplet splitting mixes states with different eigenvalues $n_{i\alpha}$. The resulting interaction is then evaluated in this basis by treating H' as a perturbation. For the antiferromagnetic superexchange interaction and the ferromagnetic Hund's-rule interaction, perturbation expansion up to sixth order is required; the direct ferromagnetic interaction gives a contribution in fourth order. The analysis leads to the following formulas for the three coupling mechanisms:

$$
\begin{aligned}
[J^{\mathrm{AFM}}]_{\mathrm{eff}} &= \frac{16 t_{dp}^4 [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{AFM}}}{\Delta_\sigma^4 (2\Delta_\sigma + U_{pp})}, \\
[J_{\mathrm{Hund}}^{\mathrm{FM}}]_{\mathrm{eff}} &= \frac{2}{\Delta_\sigma^2}\left(\frac{1}{\Delta_\sigma^2} + \frac{2}{\Delta_\sigma \Delta_\pi} + \frac{1}{\Delta_\pi^2}\right) \\
&\quad \times t_{dp}^4 [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{FM(Hund)}} \\
&\quad \times \left(\frac{1}{\Delta_\sigma + \Delta_\pi + U_{\mathrm{Hund}} + (1/4) J_{\mathrm{Hund}}}\right. \\
&\quad \left. - \frac{1}{\Delta_\sigma + \Delta_\pi + U_{\mathrm{Hund}} - (3/4) J_{\mathrm{Hund}}}\right), \\
[J_{\mathrm{direct}}^{\mathrm{FM}}]_{\mathrm{eff}} &= \frac{2 t_{dp}^2 [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{FM(direct)}}}{\Delta_\sigma^2} \\
&\quad \times \left(\frac{1}{\Delta_\sigma + (1/4) J_{\mathrm{direct}}} - \frac{1}{\Delta_\sigma - (3/4) J_{\mathrm{direct}}}\right).
\end{aligned}
\tag{A3}
$$

Inserting the parameter values we get

$$
\begin{aligned}
[J^{\mathrm{AFM}}]_{\mathrm{eff}} &= 0.0243\ (\mathrm{eV})^{-1}\ [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{AFM}}, \\
[J_{\mathrm{Hund}}^{\mathrm{FM}}]_{\mathrm{eff}} &= 0.0038\ (\mathrm{eV})^{-1}\ [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{FM(Hund)}}, \\
[J_{\mathrm{direct}}^{\mathrm{FM}}]_{\mathrm{eff}} &= 0.0084\ (\mathrm{eV})^{-1}\ [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{FM(direct)}}.
\end{aligned}
\tag{A4}
$$

The result shows that the antiferromagnetic coupling mechanism is twice as strong as the two ferromagnetic mechanisms together. Next the effective hopping factors were evaluated, including the contributions from all the superexchange patterns summarized in Eqs. (19). For each hopping pattern, an effective value which corresponds to the unfrustrated contribution was computed:

$$
\begin{aligned}
[t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}} &= \frac{1}{4}([t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{B1} + [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{B2} \\
&\quad - [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{A1} - [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{A2}),
\end{aligned}
\tag{A5}
$$

where for the interatomic matrix elements the formulas by Slater and Koster[26] were used with the parameter values of Ref. 27. As a result, we obtained

$$
\begin{aligned}
[t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{AFM}} &= [t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{FM(direct)}} = 0.034\ (\mathrm{eV})^2, \\
[t_{pp}^{1\to 2} t_{pp}^{2\to 1}]_{\mathrm{eff}}^{\mathrm{FM(Hund)}} &= 0.138\ (\mathrm{eV})^2.
\end{aligned}
$$

The hopping factor for the ferromagnetic Hund's-rule interaction is 4 times as large as that for the two other mechanisms. This result arises from the fact that interplanar hole transfer to the p_π orbitals is much more effective than to the p_σ orbitals. As illustrated schematically in Fig. 5, the p_z orbitals extend far below and above the O sites, so that interplanar hopping processes $p_\sigma \to p_z$ which bring the hole to an orbital orthogonal to a p_σ orbital are dominant. For the particular hopping pattern in Fig. 4, for example, hopping integral from the p_σ orbital to the p_z orbital gives a ferromagnetic superexchange contribution $t_{\sigma\to z}^2 = 0.026\ (\mathrm{eV})^2$ while the antiferromagnetic contribution from hops to p_σ orbital is only $t_{\sigma\to\sigma}^2 = 0.006\ (\mathrm{eV})^2$. The resulting antiferromagnetic and ferromagnetic interactions are then approximately equal: $[J^{\mathrm{AFM}}]_{\mathrm{eff}} = 8\times 10^{-4}$ meV, $[J_{\mathrm{Hund}}^{\mathrm{FM}}]_{\mathrm{eff}} = 5\times 10^{-4}$ meV, and $[J_{\mathrm{direct}}^{\mathrm{FM}}]_{\mathrm{eff}} = 3\times 10^{-4}$ meV, which yields $J^{\mathrm{AFM}} = J^{\mathrm{FM}} = 8\times 10^{-4}$ meV.

We have so far neglected the interaction mechanism arising from direct interaction between a hole on a p_π O orbital and another on a Cu $d_{x^2-y^2}$ orbital. While there exists, to our knowledge, no reliable estimate for the exchange parameter $J_{\mathrm{direct}}^{\sigma}$ in the literature, we estimated the effective interplanar interaction using $J_{\mathrm{direct}}^{\sigma} = -0.033$ eV which is consistent with a cluster calculation by Guo *et al.*[28] Because of the large hopping integrals $t_{\overline{pp}}$ involving the p_π orbitals, this weak exchange would give rise to a relevant ferromagnetic correction $[J_{\mathrm{direct}}^{\mathrm{FM}\pi}]_{\mathrm{eff}} = 1\times 10^{-4}$ meV.

Quite recently, Eskes and Jefferson[18] have shown that O-O hopping processes within CuO_2 layers give rise to significant contributions to the intraplanar superexchange J. In our calculation for the interplanar interaction these processes were omitted. Were they taken into account, they would lead to some amount of redistribution of holes among the O p orbitals, but this would not affect the conclusion $J^{\mathrm{FM}} \approx J^{\mathrm{AFM}}$.

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