

# Spin order accompanying loop-current order in cuprate superconductors

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The theory of the long-range order of orbital current loops in the pseudogap phase is generalized to include the effects of spin-orbit scattering. It is shown by symmetry arguments as well as by microscopic calculation that a specific in-plane spin order must necessarily accompany the loop-current order. The microscopic theory also allows an estimate of the magnitude of the ordered spin moment. Exchange coupling between the generated spins further modifies the in-plane direction of the spin moments. The structure and form factor for the spin and orbital moments combined with the induced spin order are consistent with the direction of moments deduced from polarization analysis in a recent neutron-scattering experiment.

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## I. INTRODUCTION

In all underdoped cuprates thermodynamic, transport, and spectroscopic measurements reveal the formation of a new state of matter which is commonly referred to as the “pseudogap” state. Any credible theory of cuprates must specify the nature of this state. One of the theories proposed predicts that this phase breaks time reversal through ordered current loops in O-Cu-O plaquettes without breaking translational symmetry.<sup>1–3</sup> The fluctuations about this state are such that there is no specific-heat singularity at the transition.<sup>4</sup> This removes a major difficulty in regarding that the pseudogap state represents a broken symmetry, since no specific-heat anomaly has been observed experimentally. The unit cell for YBCO is shown in Fig. 1(a). The predicted loop-current order in the copper oxide planes has the pattern shown in Fig. 1(b) for one of the four possible domains. Evidence for such a state was obtained from angle resolved photoemission spectroscopy using circularly polarized light<sup>5</sup> in BISCCO, and more directly by recent polarized neutron-scattering diffraction in YBCO.<sup>6</sup>

While the spatial symmetry of moments of Fig. 1(b) is borne out by the neutron experiments, the direction of the magnetic moments is not consistent with the predictions. The orbital moments should be normal to the O-Cu-O plaquettes. The plaquettes are not coplanar with the two-dimensional Cu planes due to the buckling of the planes [see Fig. 1(a)]; for YBCO in which the neutron-scattering experiments were done the nearest-neighbor O-Cu-O plaquettes make an angle of about  $7^\circ$  with respect to the Cu planes. Therefore a tilting of only about  $7^\circ$  of the moments with respect to the normal to the Cu planes is expected. However, this angle has been deduced to be  $45^\circ \pm 20^\circ$ .<sup>6</sup>

The purpose of this paper is to resolve this matter. The basic physical point we draw on<sup>7</sup> is that spin-orbit interaction can lead to spin ferromagnetism in states with orbital currents.<sup>8</sup> We present general symmetry arguments supporting this and calculate microscopically the nature of spin order in YBCO for states of the symmetry consistent with the observations.<sup>6</sup> The magnitude of the spin moment is estimated to be only 10%–20% of the orbital moment. But the neutron-scattering intensity depends on the spatial distribution or the form factor of the moments besides their magni-

tudes. For the [011] Bragg peak studied in experiments, we find that the spin form factor is significantly larger than the orbital current form factor because the latter are spread out more inside a unit cell than the former. The existing experimental results may thus be reconciled with the theory but a definitive confirmation awaits the measurement of the form factors in experiments.

The direction of the ordered in-plane spin moments is affected also by the exchange interaction between the moments. We present a rough order of magnitude for this effect which suggests that the spin order may be quite complicated. Reliable theoretical estimates on the actual spin order are very difficult to make at this point. However, there are some general features of the results which are expected to be robust. The details of the magnetic order suggested as possible here can only be resolved in experiments which have greater accuracy than the one performed to date. A companion to this paper contains the details of the experimental results which were published as a short report earlier as well as an analysis of the data applying the ideas in this paper.<sup>9,10</sup>

## II. SYMMETRY CONSIDERATIONS

We can deduce through arguments using the symmetry of the crystal structure, and of the orbital order parameter, Fig.

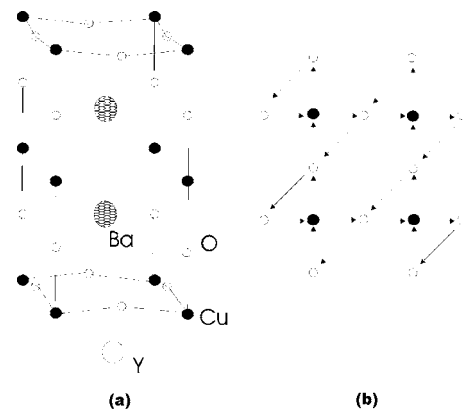


FIG. 1. (a) Crystal structure of YBCO (additional oxygens which form chains along the  $b$  axis are not shown for simplicity) and (b) the current pattern in the observed time-reversal violating states.

1, that spin-orbit coupling must induce an in-plane order of the spins. First, we present an argument by constructing a free-energy invariant which is a product of the spin-orbit tensor, the orbital order, and the possible spin order. We will show that the same conclusions can also be obtained from general arguments patterned after those due to Dzyaloshinskii.<sup>11</sup> These arguments allow one to deduce the symmetry of the spin order parameter but the physical basis of these general arguments requires a microscopic theory which also allows us to obtain the magnitude of the effective moments.

The crystal structure of YBCO is shown in Fig. 1(a). Notice that the oxygen atoms are displaced from the plane formed by the copper atoms; the copper oxide plane is not flat but dimpled. Cu's do not sit at a point of *inversion*. This leads to a linear coupling between the spin and orbital degrees of freedom.

Consider first the symmetries of the crystal structure. Due to the buckling of the oxygens, we observed the following in the crystal structure in Fig. 1(a).

(i) It breaks inversion symmetry through copper and oxygen.

(ii) It preserves reflection symmetry about  $\hat{x}$ ,  $\hat{y}$ ,  $\hat{x}+\hat{y}$  and  $\hat{x}-\hat{y}$ . Note that the existence of oxygen chains [not shown in Fig. 1(a)] and difference in lattice constants along the  $a$  and  $b$  axes of the crystal renders the crystal orthorhombic rather than tetragonal. This implies that the reflection  $\mathbf{x}\pm\mathbf{y}$  is also broken. While this changes the net spin-orbit coupling along the two directions, the net spin moment deduced remains, its direction albeit is modified. For the purposes of the symmetry argument here, this complication can be ignored.

The spin-orbit coupling tensor  $\Lambda$ , which couples linearly the spin and the momentum of the electrons, respects these symmetries. Let us now look at the symmetries of the loop-current order. The order parameter  $\mathbf{M}_O$ , corresponding to the domain in Fig. 1(b) (i) breaks inversion symmetry through copper and oxygen, (ii) preserves the reflection symmetry about  $(\hat{x}-\hat{y})$ , and (iii) breaks reflection symmetries about  $(\hat{x}, \hat{y}, \hat{x}+\hat{y})$ .

Let  $M_S$  specify the distribution and direction of possible spin order which obeys the translational symmetry of the crystal and other symmetries so that it *must* accompany the orbital order. An invariant term in the free energy of the form

$$f_{so} = \Lambda^{\alpha\beta} M_O^\alpha M_S^\beta + \frac{M_S^2}{\chi} \quad (1)$$

must then exist. Here  $\chi$  is the spin susceptibility for the order specified by  $M_S$ . Given Eq. (1),

$$M_S^\alpha = -\chi \Lambda^{\alpha\beta} M_O^\beta \quad (2)$$

will be realized.

Consider the symmetries that need to be satisfied by  $\mathbf{M}_S$ . The product of  $\Lambda$  and  $M_O$  preserves inversion through copper and oxygen, but breaks time reversal and the one mentioned reflection. Hence  $\mathbf{M}_S$  must be (i) odd under reflection about  $(\hat{x}, \hat{y}, \hat{x}+\hat{y})$ , (ii) even under reflection about  $(\hat{x}-\hat{y})$ , and (iii) even under inversion about copper and oxygen.

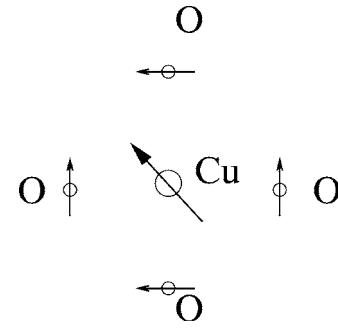


FIG. 2. Calculated spin order in YBCO for the domain shown in Fig. 1(b) in the absence of exchange interaction.

A spin order  $\mathbf{M}_S$  consistent with these requirements is shown for one of the two Cu-O bilayers in Fig. 2. Under inversion through copper, the spins remain the same thus satisfying inversion (since spin is an axial vector). All reflections other than  $\mathbf{x}=\mathbf{y}$  are broken. Since the oxygen layers are below the Cu layers in one of the layers of the bilayer and above it in the other, the spin-orbit coupling has an opposite sign in the two layers of the bilayer. It follows that the direction of the moments specified by  $\mathbf{M}_S$  is opposite in the two layers of the bilayer so that the net moment per unit cell is zero.

In the argument above  $\langle \Lambda M_O \rangle$  acts as a net magnetic field on the spins and the order is stabilized by the quadratic term in the free energy. One can also give an argument, which we find a bit more abstract, following Dzyaloshinskii-Moriya<sup>11,12</sup> (DM) for  $\mathbf{M}_S$ . In DM, one asks whether an antisymmetric interaction between magnetic moments  $\mathbf{M}_A$  and  $\mathbf{M}_B$  of the form

$$\mathbf{D}_{AB} \cdot (\mathbf{M}_A \times \mathbf{M}_B) \quad (3)$$

has the symmetries of the lattice. The direction of  $\mathbf{D}_{AB}$  is specified by the crystal symmetry in relation to positions  $A$  and  $B$  of  $\mathbf{M}_A$  and  $\mathbf{M}_B$ . The general conditions on  $\mathbf{D}_{AB}$  have been given by Moriya.<sup>12</sup> We take  $\mathbf{M}_A$  and  $\mathbf{M}_B$  to be the moments at the position of the centroid of the two triangles within the unit cell with the currents shown in Fig. 1(b). Due to the buckling of the planes, there is no center of inversion in the vector connecting these two moments. Then  $\mathbf{D}_{AB} \neq 0$ . A mirror plane perpendicular to  $AB$  bisects  $AB$ . Then  $\mathbf{D}_{AB}$  must be parallel to the mirror plane. There exists also two-fold rotation axis perpendicular to  $AB$  which passes through the midpoint of  $AB$ . Then  $\mathbf{D}_{AB}$  must be perpendicular to the twofold axis. Thus  $\mathbf{D}_{AB}$  is along the  $c$  axis and in the mirror plane specified by its normal  $\hat{x}-\hat{y}$  passing through Cu. Given such a  $\mathbf{D}_{AB}$ , a tilt of  $\mathbf{M}_A$  and  $\mathbf{M}_B$  so that they have a finite in-plane component is mandated by the term (3). This is consistent with the direction of the spin order deduced from the previous argument and shown in Fig. 2.

An important point to emphasize here is that the symmetry considerations do not specify the relative orientation of the spin on the copper and oxygen atoms. The moment in the unit cell has a component in the  $-\mathbf{x}+\mathbf{y}$  direction but the relative directions of the spin moment on oxygen and on copper are not specified. The microscopic calculation in the

next section based purely on spin-orbit scattering provides the result that these moments are parallel. However, once a spin moment is generated by the “effective field” provided by spin-orbit coupling and orbital order, one must consider also the exchange interaction between them. The actual spin arrangement depends on the relative magnitude of the exchange interactions to the effective fields for spin order due to orbital order and spin-orbit coupling. The exchange interactions will be considered later.

### III. MICROSCOPIC MODEL

The theory of loop-current order is derived from a microscopic model on the basis of the copper  $d$  and the two oxygen  $p_{x,y}$  orbitals in each unit cell.<sup>2</sup> The unit cell is shown in Fig. 2. The Hamiltonian for the system is

$$H = \sum_{i\alpha} [\bar{t}_{pd} d_{i\alpha}^\dagger (p_{i+x\alpha} - p_{i-x\alpha} + p_{i+y\alpha} - p_{i-y\alpha}) + \bar{t}_{pp} (p_{i+x\alpha}^\dagger - p_{i-x\alpha}^\dagger)(p_{i+y\alpha} - p_{i-y\alpha})] + V \sum_{i\alpha\beta} d_{i\alpha}^\dagger d_{i\alpha} (p_{i+x\beta}^\dagger p_{i+x\beta} + p_{i+y\beta}^\dagger p_{i+y\beta}) + \text{c.c.}, \quad (4)$$

where  $\alpha$  and  $\beta$  are spin indices, the sum is over the position of the copper atoms,  $\bar{t}_{pd}$  and  $\bar{t}_{pp}$  are renormalized hopping which include the effects of the large on site repulsions, and  $V$  is the Coloumb repulsion between the charges on the copper and oxygen. The symmetry breaking is captured by considering the mean-field decoupling of the quartic term. An operator identity for the interaction terms is

$$d_{i\alpha}^\dagger d_{i\alpha} p_{i+x\beta}^\dagger p_{i+x\beta} = \frac{1}{2} (-|j_{i,i+x\alpha\beta}|^2 + d_{i\alpha}^\dagger d_{i\alpha} + p_{i+x\beta}^\dagger p_{i+x\beta}). \quad (5)$$

Here  $j_{i,i+x\alpha\beta}$  is the current tensor between the sites  $i$  and  $i+x$ .

$$j_{i,i+x\alpha\beta} = i(d_{i\alpha}^\dagger p_{i+x\beta} - p_{i+x\beta}^\dagger d_{i\alpha}). \quad (6)$$

The mean-field ansatz is

$$H = \begin{pmatrix} 0 & 2i(\bar{t}_{pd}s_x(\mathbf{k}) + Rc_x(\mathbf{k}))\mathbf{I} + i\lambda_0 c_x \sigma_y & 2i(\bar{t}_{pd}s_y(\mathbf{k}) + Rc_y(\mathbf{k}))\mathbf{I} - i\lambda_0 c_y \sigma_x \\ 2i(\bar{t}_{pd}s_x(\mathbf{k}) + Rc_x(\mathbf{k}))\mathbf{I} + i\lambda_0 c_x \sigma_y & 0 & 4\bar{t}_{pp}s_x(\mathbf{k})s_y(\mathbf{k})\mathbf{I} \\ 2i(\bar{t}_{pd}s_y(\mathbf{k}) + Rc_y(\mathbf{k}))\mathbf{I} - i\lambda_0 c_y \sigma_x & 4\bar{t}_{pp}s_x(\mathbf{k})s_y(\mathbf{k})\mathbf{I} & 0 \end{pmatrix}, \quad (10)$$

where  $s_{x,y}(\mathbf{k}) = \sin(k_x a/2, k_y a/2)$ ,  $c_{x,y}(\mathbf{k}) = \cos(k_x a/2, k_y a/2)$ ,  $\mathbf{I}$  is the identity matrix, and  $\sigma$ 's are the Pauli matrices. Consider first no spin-orbit coupling, i.e.,  $\lambda_0 = 0$ . This mean-field Hamiltonian leads to the time-reversal breaking of the loop-current phase with order parameter  $R$ , but for  $\lambda = 0$  it preserves spin rotational invariance. The minima of the band are shifted from the  $\Gamma$  point corresponding to the fact that the ground state breaks inversion and the reflection symmetry

$$\sqrt{V} j_{i,i\pm x\alpha\beta} = \pm \sqrt{V} j_{i,i\pm y\alpha\beta} = \psi \delta_{\alpha\beta} = R \exp(i\varphi). \quad (7)$$

So an order parameter in terms of ordinary current (and not spin current) is sought. Symmetry requires  $\varphi = \pi/2$ .

Due to the buckling of the planes there is a finite overlap between the oxygen ( $p_x, p_y$ ) and the copper ( $d_{xz}, d_{yz}$ ) orbitals, respectively. The spin-orbit interaction on copper couples the  $d_{x^2-y^2}$  orbital with the  $d_{xz}$  and  $d_{yz}$  orbitals but such matrix elements are off diagonal in spin. Thus processes are allowed wherein the electron can hop from the ground-state orbital of the copper to that of oxygen and also flip its spin in the process. The effective Hamiltonian generated by such processes is of the form<sup>13,14</sup>

$$H_{so} = \sum_{i,\delta} i d_{i\alpha}^\dagger \vec{\lambda}_{i,i+\delta} \cdot \vec{\sigma}_{\alpha\beta} p_{i+\delta\beta}^\dagger + \text{c.c.}, \quad (8)$$

where  $\delta = (\pm x, \pm y)$ ,  $\lambda_{i,i\pm x} = \lambda_0 \hat{y}$ , and  $\lambda_{i,i\pm y} = -\lambda_0 \hat{x}$ . The coupling constant is given by  $\lambda_0 = \langle d_{x^2-y^2} | \mathbf{L} | d_{xz} \rangle t_{xz} / \epsilon_0$ , where  $\mathbf{L}$  is the angular momentum operator,  $t_{xz}$  is the hopping matrix element between the  $d_{xz}$  and  $p_x$  orbitals, and  $\epsilon_0$  is the energy difference between  $d_{x^2-y^2}$  and  $d_{xz}$  orbitals. Note that the spin-orbit Hamiltonian is generated to second order in perturbation theory which eliminates the  $d_{xz}$  and  $d_{yz}$  orbitals. The total Hamiltonian for the system is

$$H = H_{mf} + H_{so} + H_{ex}. \quad (9)$$

We have included the spin exchange term in the Hamiltonian,  $H_{ex}$  which we will discuss later. The mean-field Hamiltonian<sup>2</sup> is obtained from from Eq. (4) with the mean-field ansatz [Eq. (5)] made to decouple the quartic interaction.

We first determine the spin state (in the absence of the exchange coupling discussed below) and see that it indeed reproduces the results from general symmetry grounds obtained above. Fourier-transforming the Hamiltonian, in the basis  $\{d_{\mathbf{k}\uparrow}, d_{\mathbf{k}\downarrow}, p_{\mathbf{x}\mathbf{k}\uparrow}, p_{\mathbf{x}\mathbf{k}\downarrow}, p_{\mathbf{y}\mathbf{k}\uparrow}, p_{\mathbf{y}\mathbf{k}\downarrow}\}$   $H = H_{mf} + H_{so}$ , is given by

$-\mathbf{x} + \mathbf{y}$ . Note that the orthorhombicity implies that the hopping matrix element, spin-orbit coupling, and the magnitude of the order parameter are different along the  $a$  and  $b$  axes which changes the the position of the  $\Gamma$  point to  $-\alpha\mathbf{x} + \beta\mathbf{y}$  where  $\alpha$  and  $\beta$  are determined by the degree of orthorhombicity, but the lack of inversion implies that such a vector always exists. The qualitative discussion of the spin moments is not modified by the orthirhombicity and will be

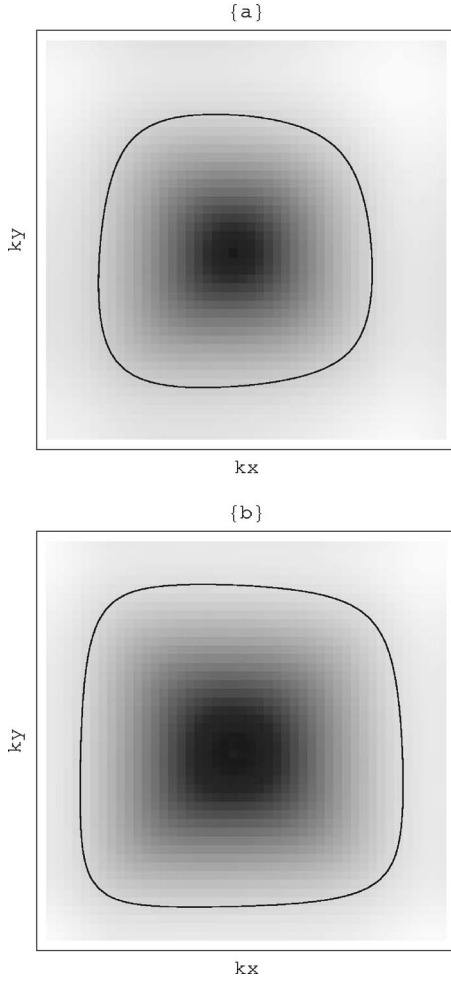


FIG. 3. Energy for the two topmost bands, plotted in gray scale, and their respective Fermi energy contour. Splitting shown is due to the presence of spin-orbit coupling. Notice that band (b) is shallower than band (a) and that the minima of the bands are shifted from the  $\Gamma$  point reflecting the broken inversion symmetry.

ignored. The particular direction of wave vector picked out by the ground state depends on the choice of domain.

#### IV. SPIN ORDER

We now estimate numerically the direction and magnitude of the spin moment in the absence of exchange coupling. To do so we discretize the Brillouin zone and for each wave vector,  $\mathbf{k}$ , we find the six eigenstates and corresponding eigenvalue of Eq. (10). Given the eigenstates we can compute the contribution to the spin moment at the copper and oxygen sites by taking the expectation value of their respective spin operators. We then sum the contributions from all states below the chemical potential. For  $t_{pd}=1$ ,  $t_{pp}=0.4$ ,  $R=0.1$ , and  $\lambda=0.1$ , we find, for the domain shown in Fig. 1(b), that the moment is distributed as shown in Fig. 2. To understand the origin of the spin moment we plot in Fig. 3 the energy of the two topmost bands which are near half filling. For the occupied states of the bands the corresponding spins on the copper atoms are shown in Fig. 4. All other bands are fully

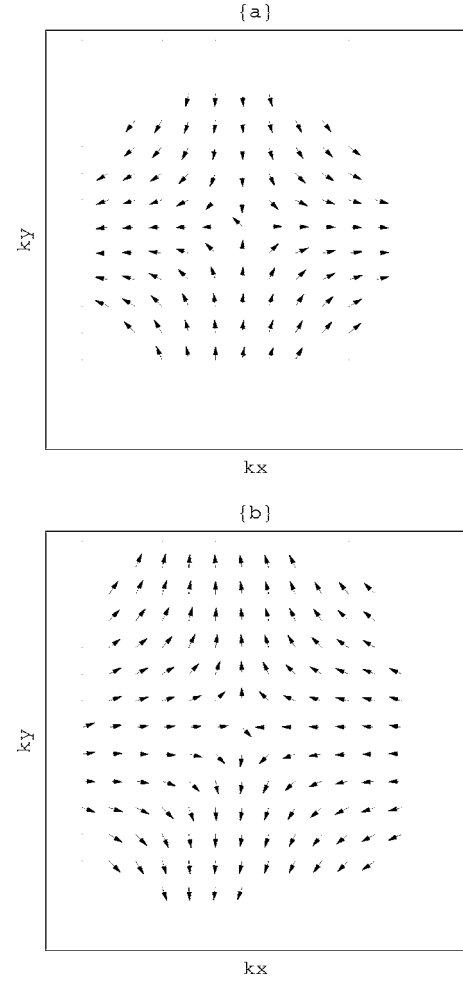


FIG. 4. Orientation of spins on the copper atom for the two topmost bands for the occupied states.

filled and do not contribute to total spin. By summing over occupied states we get the net spin and from Fig. 4 we see that there is a net spin along the  $-\mathbf{x} + \mathbf{y}$  direction.

Notice, as required by our earlier symmetry argument, the spin order breaks the  $\mathbf{x} = -\mathbf{y}$  reflection symmetry but not the  $\mathbf{x} = \mathbf{y}$  reflection symmetry. The magnetic moment due to the orbital current is estimated as  $\sim I(a^2/8)$  where  $I$  is the current and  $a$  is in-plane lattice constant. The current density is related to the order parameter as  $(R/\bar{t}_{pd})ev_F a_0^{-3}$  where  $v_F$  is the Fermi velocity and  $a_0$  is the typical size of the atomic orbital. Thus the net moment is  $\sim (R/\bar{t}_{pd})(\hbar m v_F / \epsilon_F a_0) \mu_B$ , where  $m$  is the mass of the electron and  $\epsilon_F$  is the Fermi energy. For the band structure of YBCO,  $(\hbar m v_F / \epsilon_F a_0) \sim \mathcal{O}(1)$ , we estimate the orbital moment to be  $0.1 \mu_B$  for the values of parameters chosen. The magnitude of the spin moment for the same parameters is estimated from the calculations represented in Fig. 4 to be  $0.01 \mu_B$  on the oxygen and  $0.02 \mu_B$  on the copper.

As discussed in Sec. II, including exchange changes the relative orientation of the copper and the net oxygen spins and might also affect the magnitude.

##### A. Effect of exchange interaction on the order of spins

The microscopic calculation above provides the result that the net spins on the two oxygens and on Cu, generated due to



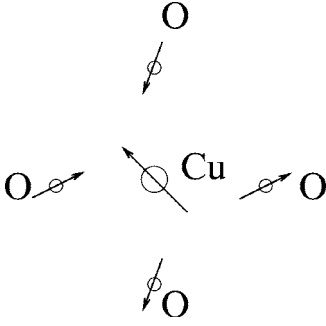


FIG. 5. Schematic spin order in YBCO for the domain shown in Fig. 1(b) in the presence of exchange interaction. The spins need not be collinear and the angle between the net oxygen spins (summed over all oxygens in a unit cell) and the copper atom is dictated by the relative strength of the exchange and ordering fields.

spin-orbit coupling, are parallel. However, once a spin moment is generated by the effective field provided by spin-orbit coupling and orbital order, one must consider also the exchange interaction between them to determine the direction of the moments. Exchange introduces a coupling whose leading terms are of the form

$$H_{ex} = J_{\text{Cu-O}} \sum_i \mathbf{S}_i^{\text{Cu}} \cdot (\mathbf{S}_{i+x}^{\text{O}} + \mathbf{S}_{i-x}^{\text{O}} + \mathbf{S}_{i+y}^{\text{O}} + \mathbf{S}_{i-y}^{\text{O}}) + J_{\text{Cu-Cu}} \sum_{\langle i,j \rangle} \mathbf{S}_i^{\text{Cu}} \cdot \mathbf{S}_j^{\text{Cu}}, \quad (11)$$

where  $J_{\text{Cu-O}}$  and  $J_{\text{Cu-Cu}}$  are the exchange couplings between nearest-neighbor Cu and O and nearest-neighbor Cu's, respectively. We expect the exchange interaction to be antiferromagnetic. Hence the induced order via the effective field discussed above competes with the the coupling  $J$ . In particular, one would expect a long-wavelength modulation of the net moment depending on the relative magnitude of the spin-orbit interaction and the exchange coupling. The in-plane moment is found to be of order  $10^{-2} \mu_B$  from which we can estimate the net effective field  $\langle \Delta M_O \rangle \sim \langle M_S \rangle E_F = 10^{-2} \text{ eV} / \mu_B$ . The exchange coupling  $J_{\text{Cu-Cu}}$  is given by  $4t_{pd}^4 / \Delta^3$ , where  $\Delta$  is the charge-transfer gap, and is of order 0.15 eV. The exchange coupling  $J_{\text{Cu-O}}$  is given by  $2t_{pd}^2 / \Delta$  which is equal to 0.67 eV for  $t_{pd}=1$ . This  $J_{\text{Cu-Cu}} / J_{\text{Cu-O}} \sim 0.22$ . The effective exchange field defined as  $J_{\text{Cu-O}} \langle M_S \rangle$  is then also of order  $10^{-2} \text{ eV} / \mu_B$ . In Fig. 5 we schematically draw the effect of exchange interaction on the in-plane spins. Symmetries dictate the existence of in-plane moments while the orientation of their ordering depends on the relative strengths of the exchange and ordering fields. It should also be clear that the competition between these effects will in general change the translational symmetry of the spin pattern. Depending on the relative exchange parameters and the spin-orbit coupling, the spin pattern can be very complicated and in general incommensurate. However, since these small moments are daughters of the orbital order, the latter is expected to be modified only weakly. At this point, it is not worthwhile to speculate on the details of the spin order of the small spin moment of  $O(10^{-2}) \mu_B$  since the exchange energies can be estimated only very imprecisely in the metallic or

pseudogap state. We must rely on the details of the magnetic structure to be obtained from the neutron experiments but for such small moments, this is no easy task.

## V. EXPERIMENTS

From the symmetry and microscopic analysis above, we have shown that there are two sources of modulated magnetic fields within the sample: current loops and spin order. Using polarized neutron scattering, Fauque *et al.*,<sup>6</sup> performed a detailed study of five different samples of YBCO (four underdoped and one overdoped) to look for magnetic ordering. The most detailed measurements are on an untwinned sample where the uncertainty is smaller.

The principle conclusions reached were the following.

(i) A magnetic contribution to scattering intensity arose at the [011] Bragg peak in all underdoped samples below a temperature which increases as the sample is progressively underdoped.

(ii) No signal was seen at the [002] Bragg peak.

(iii) No Bragg peaks appear ruling out breaking of translation invariance.

(iv) The data could be fitted to the loop-current model provided one assumed that the moments were located at the centroid of the triangles.

(v) Assuming that all the signal was due to a single source of magnetic ordering, i.e., it has a unique form factor and structure factors, the moment had to be tilted away from the  $c$  axis. The angle was largest for the detwinned sample with the moment being at  $\sim 45^\circ \pm 20^\circ$ . Our finding that the in-plane moment is due to spins while the out-of-plane component is due to orbital order necessitates a reevaluation of these numbers.

To fit the neutron-scattering data one has to assume a model for the magnetic moments in the system. From the observations above we conclude that within the experimental uncertainties, ( $\sim 0.01 \mu_B$ ), the moment is commensurate with the lattice and that the net moment in a copper oxide plane is zero. The latter follows from the absence of observable magnetic signal at [002]. For loop-current order, the spin-flip signal should appear at Bragg Peaks  $[0, K, L]$ ,  $[H, 0, L]$ ,  $[H, H, L]$ , and  $[H, -H, L]$ . The dimpling of the plane implies that these moments are at an angle of  $\sim 7^\circ$  with the  $c$  axis. To understand the origin of the larger deduced angle as stated in point (v) above we have to take into account the fact that the in-plane moments arise due to ordering of spins while the out-of-plane component is due to current loops. The corresponding moments have very different form and structure factors. For any given ordering of moments  $\mathbf{M}(\mathbf{r})$  the spin-flip scattering intensity at Bragg peak  $\mathbf{Q}=[H, K, L]$  for polarization of the incident neutron parallel to  $\mathbf{Q}$  is<sup>15</sup>

$$I(\mathbf{q}) \propto |f_Q|^2 |S(\mathbf{Q})|^2 |M_\perp|^2$$

$$\mathbf{M}_\perp = \mathbf{Q} \times (\mathbf{M} \times \mathbf{Q}) / Q^2, \quad (12)$$

where  $f_Q$  is the form factor, given by the Fourier transform of the spread of individual moment,  $S(\mathbf{Q})$  is the structure factor, given by the Fourier transform of the distribution of

these localized moments in the crystal, and  $M$  is the magnitude of the localized moment. For the current loops, the magnitude of the moments is of order  $0.1\mu_B$ , while for the spins it is  $0.02\mu_B$ . The structure factor for the current loops is proportional to  $\cos(\pi zL) \sin[2\pi x_0(H \pm K)]$ , where  $z \sim 0.29$  is the ratio of the interlayer CuO spacing to the lattice constant  $c$  and  $x_0$  is the position of the centroid of the triangular plaquette given by  $(x_0, x_0)$ . The cosine factor reflects the fact that the orbital moments are identical in the two layers of the bilayer while the sine factor arises due to the antiferromagnetic orientation between the two triangles in the unit cell. For the spins, the in-plane structure factor cannot be completely determined without knowing the precise relative angles of the spins in a unit cell. But the fact that the spins are oppositely oriented in the two layers implies that the structure factor can be as large as  $\sin(\pi zL)$ . Thus at  $[011]$  the spin structure factor can be as large as 1.8 times the moment structure factor.

For current loops, the effective orbital moment generated is spread over the area of the triangular plaquette which is  $a^2/8$ , where  $a$  is the lattice constant. For spins on copper and oxygen atoms, the moment is distributed over the  $d_{x^2-y^2}$  and  $p_{x,y}$  orbitals, respectively. Since the atomic orbitals are more localized their Fourier transforms are weaker functions of  $\mathbf{Q}$  as compared to the orbital moments. To estimate the form factors we model the time-reversal violating state with current wires along the  $x$ ,  $y$  and  $-x-y$  directions with thickness  $\delta$ . The Fourier transform of this pattern of currents is expressed in terms of a combination of form and structure fac-

tors. Then the form factor is  $2 \exp(-\pi^2 \delta^2/a^2)/\pi$  where  $a$  is the lattice constant. We have assumed a Gaussian profile for the current in the wires. Since the width of the current wire is related to the overlap of the copper and oxygen orbitals, we take it to be of order 1 Å. Thus the form factor for the current loop is  $\sim 0.3$ . The form factor for the spins is  $\sim 0.9$  implying that the net geometric factor for spins is approximately six times larger than those for the current loops. Given the estimate for the magnitude of the spin and orbital moments, the rough estimate for the geometric factors implies that indeed the resulting neutron-scattering intensities due to the two orderings will be of the same order of magnitude.

In conclusion, we find that given the orbital order, an in-plane spin order is mandated. With reasonable assumptions about the relative form factors for loop currents and spin moments and the calculated magnitude of the ordered spin moment, the observed polarized diffraction can be understood. A detailed test awaits experimental refinements. Given the spin structure shown in Fig. 5, DM interactions can induce a tilting of the spins leading to a small ferromagnetic moment. This is also under further investigation.

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