Evidence for skyrmions in the high-temperature superconductors

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I. INTRODUCTION

Since nuclear magnetic resonance studies have revealed that damped spin correlations exist across the entire range of doping for the copper oxides, and with sufficient intensity to mediate superconductivity, new questions have been raised for the coexistence of superconductivity, magnetism and charge flow in the layers that leads to the breaking of the time reversal symmetry below the pseudogap line. The core of the skyrmions form pockets of local magnetic field piercing the superconducting layers in opposite direction to the rest of the unit cell.

We claim that the charge density wave recently found by resonant soft x-ray scattering in layered copper oxides is the tetragonal symmetry defined by the distance between neighbor Cu^{++} ions in the CuO$_2$ layer that determines the critical temperature. We find evidence that this tetragonal symmetry is a skyrmionic state, which is responsible for an unusual magnetic order and charge flow in the layers. We claim that damped spin correlations exist across the entire range of doping for the copper oxides, and with sufficient intensity to mediate superconductivity, new questions have been raised for the coexistence of superconductivity, magnetism and charge flow in the layers that leads to the breaking of the time reversal symmetry below the pseudogap line. The core of the skyrmions form pockets of local magnetic field piercing the superconducting layers in opposite direction to the rest of the unit cell.

I. INTRODUCTION

Since nuclear magnetic resonance studies have revealed that damped spin correlations exist across the entire range of doping for the copper oxides, and with sufficient intensity to mediate superconductivity, new questions have been raised for the coexistence of superconductivity, magnetism and charge flow in the layers that leads to the breaking of the time reversal symmetry below the pseudogap line. We find evidence that this tetragonal symmetry is a skyrmionic state, which is responsible for an unusual magnetic order and charge flow in the layers. This transformation of the Fermi surface has been suggested to be a consequence of a new periodicity that sets in the superconductivity phase diagram. In the absence of doping the copper oxides are antiferromagnetic Mott insulators based on Cu^{++} (d^9) spins. Upon hole or electron doping by chemical substitution or oxygenation at out-of-layer sites this antiferromagnetic state is rapidly destroyed and its associated Neel transition temperature brought to zero. Above a certain doping level superconductivity emerges, and the critical temperature ($T_c$) increases with doping up to a maximum value, associated to the optimal doping. Beyond this maximum value further doping results in a reduction of $T_c$. Thus according to the doping level with respect to the maximum $T_c$, the superconducting state is called underdoped, optimally doped, and overdoped, respectively. The pseudogap state emerges at a temperature $T^*$, that is claimed to be a phase transition line. In the underdoped regime this temperature is above $T_c$ and decreases with increasing doping level. At some doping $T^* = T_c$, and beyond, one expects that the pseudogap line enters the superconducting dome to finally reach a quantum critical point at $T = 0$. The microscopic nature of the pseudogap remains controversial, but some of its characteristics are being unveiled by recent experiments. Time-reversal symmetry is spontaneously broken below the pseudogap line, because left-circularly polarized photons give a different photocurrent from right-circularly polarized photons. The Fermi surface breaks apart in the pseudogap phase, above the superconducting critical temperature $T_c$, leading to the emergence of electron pockets for hole-doped cuprates. This transformation of the Fermi surface has been suggested to be a consequence of a new periodicity that sets in the system. During the early days of superconductivity Bloch and Landau suggested that superconductivity was governed by a ground state with spontaneous circulating currents. This idea was soon to be disproved by the fair argument that circulating currents have an energetic cost that increase the kinetic energy. In terms of an order parameter approach mean the presence of a gradient term, $\nabla \psi$. There is no question that a constant order parameter is the preferred ground state solution in this case. Interestingly many experiments done in the high-temperature superconductors indicate the presence of a corrugated superconducting state, called by some PDW (pairing density state). Thus for an inhomogeneous state it is possible to entertain the idea of spontaneously circulating currents in the ground state. Nevertheless the question remains how to prevent their decay into the homogenous state, which has lower energy. Here we claim that topological stability is the key for the stability of heterogeneous superconducting state. Therefore we claim that skyrmions are behind the stability of corrugated superconducting states and set topological protectorates.

In this paper we conjecture that near to the optimal doping (maximum $T_c$), and below the pseudogap line, there is a tetragonal skyrmion state in the copper oxides. Skyrmions have been found in several non-superconducting compounds, such as the antiferromagnet La$_2$Cu$_{1-x}$Li$_x$O$_{12}$, the helimagnet MnSi and the doped semiconductor Fe$_{1-x}$Co$_x$Si. In this paper we describe qualitatively our conjecture of a skyrmion state based on an order parameter approach.

II. TETRAGONAL SYMMETRY

Recently G. Ghiringhelli et al. have found signals of a charge-density-wave instability that competes with superconductivity using resonant soft x-ray scattering. This two-dimensional charge-density-wave in the under-
doped compound YBa$_2$Cu$_3$O$_{6+x}$ has an incommensurate periodicity of nearly 3.2$a$, (atomic unit cell parameters $a = 0.39$ nm and $c = 1.17$ nm, as shown in Table 1 of Ref.$^{38}$supplementary material and in Ref.$^{39}$). This periodicity sets a tetragonal lattice because it is found to exist in orthogonal directions, namely, along and perpendicular to the so-called CuO chains, which act as charge reservoirs for the CuO$_2$ layer in this material. Interestingly they find that this structure holds both above and below $T_c$ and has correlations that reach the size of $\lambda = (16\pm 2)a$ for the underdoped compound YBa$_2$Cu$_3$O$_{6.6}$. The compound with $T_c = 57$ can be regarded as the optimally doped in case of depleted CuO chains. We point out that this resonant elastic x-scattering (REXS) correlation length of $\lambda = 6.24 \pm 0.78$ nm, found by G. Ghiringhelli et al., is equal, within error range, to the length that defines the tetragonal structure at optimal doping (maximum $T_c$), claimed by R.P. Roessler et al.$^{46}$. According to R.P. Roessler et al. this tetragonal structure “acts as a resonator for a coherent phase transition from a particle gas to a superconducting state” at $T_c$. The average distance between neighbor Cu$^{3+}$ ions in the CuO$_2$ layer of the YBa$_2$Cu$_3$O$_{6.5}$ compound is $L = 3.49$ nm. Basically H.P. Roessler et al. claim that this length really sets a de Broglie wavelength $\lambda = 2L$ that resonates in the unit cell, namely, defines $T_c$ through the free particle in a box expression, $\pi k T_c = p^2/2m$, where $p = \hbar/\lambda$ and $m$ is the Cooper pair mass. The relation $T_c \propto 1/L^2$ holds for the copper oxide superconductors.$^{47}$ Thus we claim here that the charge density correlation length of G. Ghiringhelli et al. is nothing but the de Broglie wavelength of Roessler et al., namely, $\lambda = \lambda$. Next we argue that this tetragonal symmetry is indicative of a skyrmionic state.

III. BREAKING OF TIME REVERSAL SYMMETRY

Below the pseudogap line one must seek a tetragonal symmetric state that breaks the time-reversal symmetry. Next we show that this breaking is a straightforward consequence of the following assumptions: (i) superconductivity arises in the layers and evanesces away from them; (ii) there are spontaneous superficial currents ($J_s$) confined to the CuO$_2$ layers (or a superficial magnetization, $M_s = -c \hat{\epsilon}_3 \times J_s$, where $c$ is the speed of light), and (iii) in between the layers there may exist a metallic state able to sustain three-dimensional superconductivity. The breaking of the time reversal symmetry is a straightforward consequence of $J_s$ in the layers, which produce a magnetic field, $\mathbf{h}$, in space, whose parallel component, $h_{\parallel} = h_1 \hat{x}_1 + h_2 \hat{x}_2$ is discontinuous across the CuO$_2$ layer, while the perpendicular one, $h_3$, is continuous. According to Maxwell’s equations for a layer at $x_3 = 0$, $\hat{x}_3 \times [\mathbf{h} (0^+) - \mathbf{h} (0^-)] = 4\pi J_s/c$, and $\hat{x}_3 \cdot [\mathbf{h} (0^+) - \mathbf{h} (0^-)] = 0$. A time reversal operation ultimately corresponds to the exchange of $t$ into $-t$, but in a static treatment, this operation is simply played by $\mathbf{h}$ into $-\mathbf{h}$. Hence time reversal is no longer a symmetry nor is the spatial reflection $\hat{x}_3 \rightarrow -\hat{x}_3$, under a fixed $J_s$, but their product is still a symmetry. The presence of spontaneous circulating currents in the superconducting ground state has been considered long ago by Felix Bloch and Lev Landau during the early days of superconductivity.$^{12}$ Recently C. M. Varma$^{19}$ has proposed that microscopic orbital currents within the CuO$_2$ atomic cell describe properties of the pseudogap. In the present approach we consider a state with tetragonal symmetry, namely, with spontaneous circulating currents in an area larger, than that claimed by Varma, which is confined to the atomic cell $(a)$. The present area is defined by the optimal doping length $(L)$. We claim here that $\mathbf{h}$, created by $J_s$ is such that there are skyrmions, defined by their topological charge, given by,

$$Q = \frac{1}{4\pi} \int_{x_3=0} \left( \frac{\partial \mathbf{h}}{\partial x_1} \times \frac{\partial \mathbf{h}}{\partial x_2} \right) \cdot \mathbf{h} \, d^2x,$$

for $\mathbf{h} = \hbar/|\hbar|$. Skyrmions cannot decay into other configurations because of this topological stability no matter how close they are in energy to any other configuration. Clearly the time reversal symmetry, $(\hbar \rightarrow -\hbar)$, is broken by the skyrmions. The skyrmions are chiral magnetic excitations with a well defined core, where the rotation sense is in the opposite direction of the rest, such that there is no net current flowing out of the unit cell area. This core has the size of nearly $3a$, thus close to the proposed periodicity of G. Ghiringhelli et al.$^{54}$. In the tetragonal symmetric state the cells carry skyrmions with topological charge $Q$. The preferred chirality of the skyrmions will rotate circularly polarized light passing through the layers and lead to the dichroism observed below the pseudogap line.$^2$

IV. STRUCTURE OF THE SKYRMION LATTICE

For the compound YBa$_2$Cu$_3$O$_{6.5}$ the ratio between the unit cell length and the atomic unit cell height is $L/c = 2.98$ nm.$^{36}$ We interpret this ratio as a consequence of the skyrmion lattice. To reach this conclusion consider, for simplicity, the copper oxide superconductor as made of a stack of identical CuO$_2$ layers separated by a fixed distance $c$. Thus in each layer there is an identical tetragonal lattice whose cells have $Q$ skyrmions each. Hence there is an elaborate spatially magnetic field arrangement in between the layers whose magnetic field energy density, $\hbar^2/8\pi$, stored in space, must be considered. We find that this magnetic energy integrated over the unit cell is optimally reduced for some values of the unit cell length $L$, that will be discussed in detail in a future publication. Thus the present approach shows that the distance $c$ between layers plays a key role in defining $L$, and consequently $T_c$, as stressed by some authors.$^{20}$ According to our theory this minimum is reached at $L/c \sim 3$.
TABLE I. Single layer cuprate superconductors containing the atomic unit cell height (c nm), the maximum critical temperature ($T_c$ K), the average dopant distance ($L$ nm) obtained from $L \equiv (26.3 \text{ nm} K^{1/2})/\sqrt{T_c}$ and the ratio $L/c$.

| Compound | $T_c$ (K) | $L$ (nm) | $c$ (nm) | $L/c$ |
|----------|-----------|----------|----------|--------|
| Bi$_2$(Sr$_{1.6}$La$_{0.4}$)CuO$_{4+\delta}$ | 34 | 4.51 | 1.220 | 3.70 |
| Tl$_2$Ba$_2$CuO$_6$ | 80 | 2.94 | 1.162 | 2.53 |
| HgBa$_2$CuO$_{4+\delta}$ | 95 | 2.70 | 0.950 | 2.84 |

and 4 for $d$ and $s$ wave symmetries, respectively. Table I displays this ratio $L/c$ for some single layered compounds, according to Roeser et al. $^{16,17}$, to show that its value is similar for different compounds. We suggest that this ratio truly reflects some inner property of the tetragonal state, since it falls within our theoretical calculations, which are a consequence of the stored magnetic energy of the skyrmion lattice.

Let us analyze in more details the magnetic field associated to the skyrmion assuming, for simplicity, that there should exist only two kinds of magnetic field stream lines in the layered structure. Since $\nabla \cdot \mathbf{h} = 0$ there are those that pierce a particular layer twice and those that pierce all the layers only once, never to return again, like in a solenoid. Consider that the skyrmion has a core, this core a center, and all the magnetic stream lines of the first kind should pass through it. As shown in Fig. this center acts as a sinkhole, as seen from the top of a layer, and a source, as seen from the bottom of the very same layer. All the stream lines that pass through the core of the skyrmion must return and cross the layer again piercing the remaining area at some point in the opposite direction. Hence the neighborhood of the skyrmion center, which is the core, has $h_3 < 0$, while the remaining unit cell area has $h_3 > 0$. $\mathbf{J}_s$ around the center is very strong in comparison to the rest of the unit cell, where it is weak and flows in the opposite direction, thus the areas associated to flow and counterflow have very distinct sizes. Thus the core of the skyrmion forms a pocket of local magnetic field piercing the superconducting layers in opposite direction to the rest. We determine that these pockets correspond to 9.4 % and 5.3 % of the total area, in case of $d$ and $s$ wave respectively, for the optimal value of the unit cell size.

It must be emphasized that the local magnetic field created by the skyrmion is very weak. Indeed it is fundamental to estimate the value of $\mathbf{h}$ since NMR/NQR$^{21,22}$ and $\mu$SR$^{23,24}$ experiments put several bounds on its maximum value. An estimate of the maximum modules of $h_3$ can be obtained by calculating its value at the center of the skyrmion. It follows by considering the core of the skyrmion as a disk of radius $R$, such that $\mathbf{J}_s$ grows linearly proportional away from the center of the skyrmion. The magnetic moment generated by such a disk satisfies $\mu = h_3 \pi R^3/8$. This relation shows that an extended ob-

![FIG. 1. (color online) Pictorial view of the local magnetic field of a skyrmion very near to a superconducting layer. Both views, slightly above (red) and below (blue) the layer, are shown. The discontinuity of the tangential field component results in the superficial current which is also displayed here (below).](image)

ject (assume, as an example, that $R \sim L/4$, such that

for $L \sim 3.0$ nm, $R \sim 0.75$ nm) can have a large magnetic moment, $\mu \sim 0.01 \mu_B$, attached to a very weak field, $h_3 \sim 0.02$ mT. This magnetic field and magnetic moment is in the threshold limit of local fields from dynamical currents measured by NMR/NQR$^{21,22}$ and by $\mu$SR$^{23,24}$ measurements. Because of the elaborate circulating currents, the skyrmion lattice manifests itself as a periodic charge arrangement.

V. CONCLUSIONS

We claim that there is a lattice of skyrmions in the CuO$_2$ layers that explains the tetragonal symmetry and the breaking of time-reversal symmetry near to the optimal doping where the critical temperature reaches a maximum.

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1. J. L. Miller *Phys. Today*, vol. 64, no. 9, pp. 13–14, 2011.
2. E. Berg, E. Fradkin, S. A. Kivelson, and J. M. Tranquada *New J. Phys.*, vol. 11, p. 115004, 2009.
3. M. Vojta *Adv. Phys.*, vol. 58, no. 6, pp. 699–820, 2009.
4. R. Daou, J. Chang, D. Leboeuf, O. Cyr-choiniere, F. Laliberte, N. Doiron-Leyraud, B. J. Ramshaw, R. Liang, d. A. Bonn, W. N. Hardy, and L. Taillefer *Nature*, vol. 463, no. 7280, pp. 519–522, 2010.
5. G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N. B. Brookes, G. M. De Luca, A. Frano, D. G. Hawthorn, F. He, T. Loew, M. M. Salh, D. C. Peets, M. Salluzzo, E. Schierle, R. Sutarto, G. A. Sawatzky, E. Weschke, B. Keimer, and L. Braicovich *Science*, vol. 337, no. 6096, pp. 821–825, 2012.
6. R.-H. He et al. *Science*, vol. 331, no. 6024, pp. 1579–1583, 2011.
7. S. Kaminski, A. Rosenkranz, H. M. Fretwell, J. C. Campuzano, Z. Li, H. Raify, W. G. Cullen, H. You, C. G. Olson, C. M. Varma, and H. Hochst *Nature*, vol. 416, pp. 610–613, 2002.
8. J. Xia, E. Schemm, G. Deutscher, S. A. Kivelson, D. A. Bonn, W. N. Hardy, R. Liang, W. Siemons, G. Koster, M. M. Fejer, and A. Kapitulnik *Phys. Rev. Lett.*, vol. 100, p. 127002, 2008.
9. M. R. Norman *Physics*, vol. 3, p. 86, Oct 2010.
10. L. Taillefer *Journal of Physics: Condensed Matter*, vol. 21, no. 16, p. 164212, 2009.
11. I. Raicévić et al. *Phys. Rev. Lett.*, vol. 106, p. 227206, 2011.
12. C. Pfleiderer et al. *Nature*, vol. 427, pp. 227–231, 2004.
13. W. Münzer et al. *Phys. Rev. B*, vol. 81, p. 041203, 2010.
14. M. M. Doria, A. R. d. C. Romaguera, and F. M. Peeters *Europhys. Lett.*, vol. 92, no. 1, 2010.
15. A. A. Vargas-Paredes, M. M. Doria, and J. A. H. Neto *Journal of Mathematical Physics*, vol. 54, no. 1, p. 013101, 2013.
16. H. Roeser, F. Hetfleisch, F. Huber, M. von Schoenermark, M. Stepper, A. Moritz, and A. Nikoghosyan *Acta Astronautica*, vol. 62, no. 12, pp. 733 – 736, 2008.
17. *Journal of Superconductivity and Novel Magnetism*, vol. 24, pp. 1443–1451, 2011.
18. J. Schmalian *preprint*, 2010.
19. C. M. Varma *Phys. Rev. B*, vol. 73, p. 155113, 2006.
20. D. Innocenti, N. Poccia, A. Ricci, A. Valletta, S. Caprara, A. Perali, and A. Bianconi *phys. Rev. B*, vol. 82, p. 184528, 2010.
21. S. Strässle, J. Roos, M. Mali, H. Keller, and T. Ohno *Phys. Rev. Lett.*, vol. 101, p. 237001, 2008.
22. S. Strässle, B. Graneli, M. Mali, J. Roos, and H. Keller *Phys. Rev. Lett.*, vol. 106, p. 097003, 2011.
23. G. J. MacDougall, A. A. Aczel, J. P. Carlo, T. Ito, J. Rodriguez, P. L. Russo, Y. J. Uemura, S. Wakimoto, and G. M. Luke *Phys. Rev. Lett.*, vol. 101, p. 017001, 2008.
24. J. E. Sonier, V. Pacradouni, S. A. Sabok-Sayr, W. N. Hardy, D. A. Bonn, R. Liang, and H. A. Mook *Phys. Rev. Lett.*, vol. 103, p. 167002, 2009.