Diagonal charge modulation in the insulating $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

O. P. Sushkov$^1$

$^1$School of Physics, University of New South Wales, Sydney 2052, Australia
(Dated: February 2, 2008)

We show that, due to the Dzyaloshinskii-Moriya and the XY anisotropies, the disordered diagonal spin spiral in the insulating $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ generates a diagonal charge density wave (CDW) with the wave vector twice of that of the spin spiral. The amplitude of the CDW depends on values of the anisotropies, the doping level, and on the density of states at the chemical potential. Based on available experimental data we estimate that for 4% doping the amplitude of CDW is about 1/10 of the doping level. We believe that this mechanism explains the CDW observed recently in Zn-codoped detwinned $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$.

The three-dimensional antiferromagnetic Néel order in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) disappears at doping $x \approx 0.02$ and gives way to the so-called spin-glass phase which extends up to $x \approx 0.055$. In both the Néel and the spin-glass phase the system essentially behaves as an Anderson insulator and exhibits only hopping conductivity $\text{(a)} \text{[1, 2].}$ Superconductivity then sets in for doping $x \gtrsim 0.055$. The incommensurate magnetic order has been observed at low temperature in elastic and inelastic neutron scattering. According to experiments in the Néel phase, the incommensurability is almost doping-independent and directed along the orthorhombic $b$ axis [3]. In the spin-glass phase, the shift is also directed along the $b$ axis, but scales linearly with doping [4, 5, 6]. Finally, in the underdoped superconducting state, the shift still scales linearly with doping, but it is directed along one of the crystal axes of the tetragonal lattice [7].

In the present work we discuss only the insulating spin-glass phase, $0.02 \leq x \leq 0.055$. The theory for the insulating phase has been suggested in Refs. [8, 9, 10]. The theory has the following essential components:

1) Due to strong antiferromagnetic correlations, the minima of dispersion of a mobile hole are at points $(\pm \pi/2, \pm \pi/2)$ of the Brillouin zone, so the system can, to some extent, be considered as a two valley semiconductor. The hole does not have a usual spin, but it possesses a pseudospin that describes how the hole wave function is distributed between two magnetic sublattices.

2) At low temperature, each hole is trapped in a hydrogen-like bound state near the corresponding Sr ion, the binding energy is about $10 - 15 \text{meV}$ and the radius of the bound state is about 10 Å.

3) Due to the orthorhombic distortion of LSCO, the matrix elements $t'_a$ and $t'_b$ describing the diagonal hopping of the hole are slightly different, and this makes the $b$ valley, $(-\pi/2, \pi/2)$, deeper than the $a$ valley, $(\pi/2, \pi/2)$. So all the hydrogen-like bound states are built with holes from the $b$-valley. In what follows, we refer to these bound states as impurities.

4) Each impurity creates a spiral distortion of the spin background in the orthorhombic $b$-direction. The distortion is observed in neutron scattering. So the state at $0.02 < x < 0.055$ is not a spin glass, it is a disordered spin spiral.

5) At the point of overlapping of bound states ("percolation" point) the direction of the spiral must rotate from the diagonal to parallel because the Pauli principle. Simultaneously the superconducting pairing is getting possible. Hence we conclude that $x = 0.055$ is the percolation point.

Intrinsically this picture does not contain any charge ordering and this is qualitatively different from the stripe scenario [11]. Charge modulation in the spin spiral picture is certainly possible, but this can only be a secondary effect that is due to the spin-orbit interaction. Stimulated by the recent discovery [12] of the charge modulation in $\text{La}_{1.95}\text{Sr}_{0.05}\text{Cu}_0.95\text{Zn}_{0.05}\text{O}_4$ we suggest in the present work a specific mechanism for CDW that is due to the Dzyaloshinskii-Moriya (DM) and the XY anisotropies.

The spiral pitch. Calculation of the spiral pitch can be performed within the mean-field approximation. Mobile holes are trapped by Sr ions in hydrogen-like bound states ("impurities") . The ground state of the “hydrogen atom” is four-fold degenerate: (two-fold pseudospin) $\times$ (two-fold valley). The orthorhombic distortion lifts the valley degeneracy, so all the impurities reside in the $b$-valley [8]. Impurity pseudospin interacts with the spiral distortion of the spin fabric [8]. The interaction energy is $\sqrt{2}gQ_b$ where $Q$ is the wave vector of the spiral, $Q_b$ is the component along the orthorhombic $b$-direction, and the coupling constant is approximately equal to the antiferromagnetic exchange, $\approx J \approx 140 \text{meV}$. We set the tetragonal lattice spacing equal to unity, so the wave vector $Q$ is dimensionless. The pseudospin degeneracy is lifted as soon as the spiral is established, all pseudospins are aligned and the corresponding energy gain per unit area is $-x\sqrt{2}gQ$. Here $x$ is concentration of impurities that is practically equal to doping. The elastic energy of the spin fabric deformation is $\rho_sQ^2/2$. Here $\rho_s \approx 0.18J$ is the spin stiffness. Thus the total energy is $\rho_sQ^2/2 - x\sqrt{2}gQ_b$. Minimization with respect to $Q$ gives

$$Q = Q_b = \frac{\sqrt{2}g}{\rho_s}x. \quad (1)$$
To fit the experimental data \[4, 5, 6\] we need \( g = 0.7J \) that agrees with the \( t - J \) model estimate, \( g \approx J \). The presented mean-field picture does not address the stability of the state, broadening of the line due to disorder, topological defects, etc. These issues have been studied in Refs. [8, 10]. The stability depends on the localization length (size of the impurity) that does not appear in the mean-field picture. However, as soon as we know that the disordered spiral state is stable, then the above description is correct. We would like to stress that the spiral picture does not necessary assume a static spiral. The spiral can be dynamic. In particular in a pure 2D system the spiral is dynamic at any nonzero temperature. In LSCO, due to anisotropies and a weak 3D coupling, the spiral becomes dynamic at a small finite temperature, \( T \sim 20K \). However, the absence of the static spiral at \( T \geq 20K \) does not mean that the spiral is not there, it just becomes dynamic. The important components in the above picture are 1) the hole binding, 2) the a-valley depopulation, and 3) the height of the spin-wave dome \( E_{\text{cross}} \) observed in neutron scattering. Both the binding energy and the valley anisotropy energy are about 10-15meV [8, 13]. The value of \( E_{\text{cross}} \) depends on doping, and for \( x = 0.03 - 0.05 \) it is also about 15mev [11]. Therefore the spiral description is valid up to characteristic temperature \( T_h \sim 150K \).

**Density of states.** In the case of uniform doping the diagonal spiral (unlike the parallel spiral) always has a tendency towards charge modulation [15, 16]. In the case of the disordered state, the problem of charge instability was resolved in Refs. [8, 9, 10], assuming that at zero temperature all the Sr-hole bound states are filled and hence there is no room for compression. So, implicitly the picture of energy levels shown in Fig.1A was assumed: all the bound states are below chemical potential. However, Mott variable range hopping (VRH) formula [1]

\[
\sigma \sim \exp\{- (T_0/T)^{1/3}\}.
\]

This implies that the chemical potential is within the range of impurity energies, as it is shown in Fig.1B. Hence some bound states are unoccupied and this gives room for CDW built on the bound states. Let us denote the concentration of unoccupied bound states by \( \delta x \). It is well established that the hole doping level is pretty close to concentration of Sr ions, therefore, \( \delta x \ll x \). On the other hand \( \delta x \) is not that small because it is sufficient for VRH. It is reasonable to assume that

\[
\frac{\delta x}{x} \sim 0.1.
\]

This is the maximum possible relative amplitude for charge density modulation, there are no more quantum states within the impurity band to develop a larger amplitude.

The characteristic VRH temperature \( T_0 \) in Eq. (2) depends on doping and sample quality and generally decreases when doping increases (and thus conduction becomes easier). At 4% doping the data of Ref. [1] are well fit with \( T_0 \approx 500 \text{ K} \). Analyzing the curves from Ref. [2] we have found \( T_0(x = 0.02) \sim 8000 \text{ K} \), \( T_0(x = 0.03) \sim 2000 \text{ K} \), \( T_0(x = 0.04) \sim 300 \text{ K} \). The temperature \( T_0 \) is related to the 2D density of states \( G \),

\[
G = \frac{13.8}{T_0^{0.5}},
\]

see Ref. [18]. Here \( l \) is the localization length (the “Bohr radius” of the bound state). We set the Boltzmann constant equal to unity. The localization length is about 2.5 lattice spacings [1]. Note that the density of states determined in this way is valid up to \( T \sim E_b \sim 100 - 150K \) (ionization of bound states) in spite of the fact that the VRH resistivity formula is valid only up to \( T \sim 20 - 30K \). The density of states is related to the width of the impurity band \( \delta E \) shown in Fig.1

\[
G = x/\delta E.
\]

From here we find values of \( \delta E \) at different doping levels: \( \delta E(x = 0.02) \sim 6 \text{ meV} \), \( \delta E(x = 0.03) \sim 2 \text{ meV} \), \( \delta E(x = 0.04) \sim 1 \text{ meV} \).

**Spin anisotropies and generation of the second harmonics of the spiral.** It is very convenient to use the \( \sigma \)-model notation. The energy density in this notation reads [10]

\[
\frac{\rho_s}{2} \left( \nabla \vec{n}(r) \right)^2 - \sqrt{2} \bar{g} \sum_i \xi_i \cdot [\vec{n} \times (e_b \cdot \nabla)] \bar{n} \delta(r-r_i) + \frac{\rho_s}{2e^2} \left[ D^2 n_{\alpha}^2 (r) + \Gamma_e n_{\alpha}^2 (r) \right] .
\]

Here \( \vec{n}, n^2 = 1 \), is the staggered field that describes spins, \( \xi_i \) is direction of pseudospin of i-th impurity, \( \xi^2 = 1 \), \( e_b \) is the unit vector along the orthorhombic b-axis. As we have already mentioned, we neglect here the impurity
size. We mention once again that what we call “the impurity” is the occupied bound state. The last two terms in [5] describe anisotropies induced by the spin-orbit interaction [19, 20]. The anisotropies “want” to direct \( \vec{n} \) along the b-axis. The DM vector is \( D \approx 2.5 \text{ meV} \) and the XY anisotropy \( \sqrt{\Gamma_c} \approx 5 \text{ meV} \). The spin wave velocity is \( c \approx \sqrt{2J} \).

As we described above, the two first terms in [5] generate the spiral

\[
\vec{n} = (0, \sin(Q \cdot r + \varphi), \cos(Q \cdot r + \varphi)) .
\]  

(6)

Here we assume that the spins are in the bc-plane. However, we will argue below that the actual plane of the spiral is not important, moreover, spins can slowly fluctuate without any static spiral. The disorder and topological defects give random phase \( \varphi \) that broadens the line [10], however the broadening is a separate issue and we disregard it here. The XY-term in [5] is

\[
\sim \frac{c^2}{G^2} \left[ \cos(Q \cdot r + \varphi) \right]^2 - \frac{1}{c^2} \left[ \cos(2Q \cdot r + 2\varphi) \right] \rightarrow -\varphi \sin(2Q \cdot r). \]

Here we have assumed that \( \varphi \ll 1 \). Thus, the interaction does not vanish after integration over space only if

\[
\varphi = A \sin(2Q \cdot r) .
\]  

(7)

This is the mechanism for generation of the second harmonics in the spin pattern. Substitution of [5] in [6] gives the following energy density

\[
\frac{\rho_s}{2} (Q + \varphi')^2 - \sqrt{\frac{\sigma_0^2}{g}} (Q + \varphi') - \frac{\rho_s \Gamma_c}{2c^2} \varphi \sin(2Q \cdot r) ,
\]  

(8)

where \( \varphi' = (e_b \cdot \nabla) \varphi \) and \( \sigma(r) = \sum_i \delta(r - r_i) \) is the density of impurities per unit area of the plane. In the case of distribution of states shown in Fig. 1A the density of impurities is constant because there is no room for a density modulation, \( \sigma = \bar{x} \). In this case minimization of [5] with respect to the amplitude of the second harmonics of the spin spiral gives

\[
A = \frac{\Gamma_c}{8c^2 Q_s^2} .
\]  

(9)

The value of \( A \) is small: \( A \approx 10^{-3} - 10^{-2} \), so it is hardly possible to observe it directly in neutron scattering.

**Charge density wave.** We know that the correct picture of states is shown in Fig. 1B and in this case the CDW is possible. According to Eq. 15 the energy of a single impurity is shifted due to the second harmonics of \( \varphi \) as

\[
\delta \varepsilon = -\sqrt{2g} \varphi' = -2\sqrt{2g} Q A \cos(2Q \cdot r) .
\]  

(10)

Hence, the variation of density of impurities is

\[
\delta \sigma = -G \delta \varepsilon = B \cos(2Q \cdot r) ,
\]

\[
B = 2G\sqrt{2g} QA = \frac{13.8}{8T^2 x J T_0} \approx 0.3 \frac{\Gamma_c}{T_x J T_0} .
\]  

(11)

Here \( G \) is the density of states given by Eq. (1). Substituting numerical values of parameters in [11] we find that at \( x = 0.04 \) the amplitude of charge modulation is

\[
B \sim 10^{-2} .
\]  

(12)

This value is in units of elementary charge per unit cell of the square lattice. Thus, the modulation is about 20% of the doping level. This dramatic enhancement of CDW is due to the very high density of states (4). In doing the estimate we assumed the static spin spiral in the bc-plane, see Eq. (6). However, since the effect comes from the phase \( \varphi \), the static order is not essential. In the case of fully dynamic spiral we have to replace in [11] \( \Gamma_c \rightarrow \frac{1}{2} (\Gamma_c + D^2) \). Since \( \Gamma_c \sim D^2 \), this does not influence the estimate [12]. According to [12] the CDW is so strong that it is quite possible that the real limitation on the amplitude \( B \) comes from the available Hilbert space limit given by Eq. (3). Anyway, both estimates [12] and [9] give CDW amplitude of about 10% of the doping level.

Interestingly, a naive selfconsistent treatment of the mean-field Eq. (9) gives an instability with respect to unlimited increasing of the CDW amplitude. However we know from the analysis [5, 10] that the stability issue cannot be resolved within the mean-field approximation. This is where we rely on perturbation theory. In any case, the amplitude is bound from the top by the condition [33] and since the perturbation theory result [12] is of the same value as the upper bound we believe that this is a reliable estimate of the effect. The effect depends on temperature due to depopulation of the b-valley as well as due to ionization of bound states. The expected dependence is roughly \( B \propto \tanh(T_J/T_0) \). The characteristic temperature is \( T_J \sim 150K \).

**Coulomb interaction, phonons, and large correlation lengths.** The mechanism considered above explains the CDW amplitude, but it does not explain large correlations lengths observed in [12]. One needs an additional weak interaction to coordinate the phase of the CDW. There are two candidates for the “coordination interaction”, 1)Coulomb interaction, 2)Interaction with phonons (lattice deformation). Let us first consider the Coulomb interaction. The dielectric constant \( \kappa \) is strongly anisotropic [21]. For the direction along the c-axis it is equal to the ionic value \( \kappa_c \sim 30 - 70 \). The in-plane value is much larger because polarizabilities of impurities contribute to the screening of the electric field [21]. This contribution is proportional to doping, and extrapolating from data [21] we get that at \( x = 0.04 \) the value is \( \kappa_{ab} \sim 2000 \). Therefore, for estimates we will use the effective isotropic dielectric constant that is average between \( \kappa_c \) and \( \kappa_{ab} \). \( \kappa \sim 1000 \). Here we have in mind the zero temperature value.

If the Coulomb interaction is important then it must establish the CDW antiphase between the CuO2 layers. On the other hand we know that the in-plane modulation is very slow, \( 2\pi/(2Q) \ll d_c \), where \( d_c = 13.15 \AA \).
is the separation between the planes. A straightforward electrostatic calculation shows that the Coulomb energy per unit area of the plane in this geometry is $E_C \approx \frac{1}{2\kappa}3.2d_e(e\delta\sigma)^2$, where $e\delta\sigma$ is the charge density per unit area, $e$ is the elementary charge and $\kappa$ is the dielectric constant (we use CGS units). The density wave is $\delta\sigma = B\cos(2Q \cdot r)$. Therefore, with account of the electrostatic energy the impurity energy shift is changed from (11) to

$$\delta\epsilon = -2\sqrt{2}gQA\cos(2Q \cdot r) + 3.2e^2/\kappa d_e B\cos(2Q \cdot r).$$

The density variation is $\delta\sigma = -G\delta\epsilon$. Hence, we find from (13) that the density wave amplitude is

$$B = \frac{2G\sqrt{2}gQA}{1 + 3.2(e^2/\kappa)d_e G}.$$  

The difference from Eq. (11) is in the Coulomb factor $F_C = [1 + 3.2(e^2/\kappa)d_e G]^{-1}$. The value of this factor at $x = 0.04$ is $F_C \sim 0.5$. Thus, the Coulomb interaction does not qualitatively influence the estimate (12). It can change the estimate by at most a factor $\sim 2$. This conclusion is supported by the following experimental observation: The CDW amplitude is not very sensitive to the change of the screening regime and hence the Coulomb interaction is not important. The most powerful confirmation of this point comes directly from experiment [12]. In the observed CDW the layers are in-phase and this implies that the Coulomb interaction is negligible.

Thus, as it has been pointed out in Ref [12], we are left with phonons to coordinate the CDW phase. Most likely this is also related to the DM interaction. The DM vector $D$ is proportional to the oxygen octahedra tilting angle, so we can write $D \rightarrow D + \delta D$, where $\delta D$ is coupled to the soft phonon responsible for variation of the tilting angle. The $D^2n^2_\sigma$-term in (13) generates coupling to the spiral $D^2n^2_\sigma \rightarrow (D + \delta D)^2[\sin(Q \cdot r + \varphi)]^2 \rightarrow D\delta D\cos(2Q \cdot r + 2\varphi)$. This generates the lattice deformation at the second harmonics and this is the “coordination” interaction. Clearly, this mechanism gives the same phase of the CDW for nearest CuO$_2$ layers.

In conclusion, the disordered diagonal spin spiral in the insulating phase of LSCO generates a charge density wave with the wavelength half that of the spin spiral. The charge modulation is due to relativistic Dzyaloshinskii-Moriya and the $XY$ anisotropies. At $x = 0.04$ we estimate the amplitude of the modulation at $\sim 10\%$ of the doping level. The effect survives up to characteristic temperature $T_k \sim 150K$. We believe that this theory explains the CDW observed in Ref [12].

I am grateful to P. Abbamonte and K. Yamada for communicating their results prior to publication. I am also grateful to V. Kotov and A. I. Milstein for helpful discussions.

[1] B. Keimer, A. Aharony, A. Auerbach, R. J. Birgeneau, A. Cassanho, Y. Endoh, R. W. Erwin, M. A. Kastner, and G. Shirane, Phys. Rev. B 45, 7430 (1992); M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. 70, 897 (1998).

[2] Y. Ando, K. Segawa, S. Komiyi, and A. N. Lavrov, Phys. Rev. Lett. 88, 137005 (2002).

[3] M. Matsuda, M. Fujita, K. Yamada, R. J. Birgeneau, Y. Endoh, and G. Shirane, Phys. Rev. B 65, 134515 (2002).

[4] S. Wakimoto, G. Shirane, Y. Endoh, K. Hirota, S. Ueki, K. Yamada, R. J. Birgeneau, M. A. Kastner, Y. S. Lee, P. M. Gehring, and S. H. Lee, Phys. Rev. B 60, R769 (1999).

[5] M. Matsuda, M. Fujita, K. Yamada, R. J. Birgeneau, M. A. Kastner, H. Hiraka, Y. Endoh, S. Wakimoto, and G. Shirane, Phys. Rev. B 62, 9148 (2000).

[6] M. Fujita, K. Yamada, H. Hiraka, P. M. Gehring, S. H. Lee, S. Wakimoto, and G. Shirane, Phys. Rev. B 65, 064505 (2002).

[7] K. Yamada, C. H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R. J. Birgeneau, M. Greven, M. A. Kastner, and Y. J. Kim, Phys. Rev. B 57, 6165 (1998).

[8] O. P. Sushkov and V. N. Kotov, Phys. Rev. Lett. 94, 097005 (2005).

[9] A. Lüscher, G. Misguich, A. I. Milstein, and O. P. Sushkov, Phys. Rev. B 73, 085122 (2006).

[10] A. Lüscher, A. I. Milstein, and O. P. Sushkov, Phys. Rev. Lett. 98, 037001 (2007).

[11] S. A. Kivelson, E. Fradkin, V. J. Emery, Nature 390, 550 (1998).

[12] A. Rusydi, S. Smadici, J. C. Lee, S. Wang, P. Abbamonte, M. Enoki, M. Fujita, M. Rubhausen, K. Yamada, to be published.

[13] O. P. Sushkov, Wenhui Xie, O. Jepsen, O. K. Andersen, G. A. Sawatzky, arXiv:0710.5325 Phys. Rev. B, to be published.

[14] K. Yamada, private communication.

[15] A. V. Chubukov and K. A. Musaelian, Rev. B 51, 12605 (1995).

[16] O. P. Sushkov and V. N. Kotov, Phys. Rev. B 70, 024503 (2004).

[17] E. Lai and R. J. Gooding, Phys. Rev. B 57, 1498 (1998).

[18] A. L. Efros and B. I. Shklovskii, in Electron-Electron Interactions in Disordered Systems, edited by A. L. Efros and M. Pollak, p. 409 (North-Holland, Amsterdam, 1985).

[19] J. Chovan and N. Papanicolaou, Eur. Phys. J. B 17, 581 (2000).

[20] M. B. Silva Neto, L. Benfatto, V. Juricic, C. Morais Smith, Phys. Rev. B 73, 045132 (2006).

[21] C. Y. Chen, R. J. Birgeneau, M. A. Kastner, N. W. Preyer, and Tineke Thio, Phys. Rev. B 43, 392 (1991); C. Y. Chen, E. C. Branlund, ChinSung Bae,
K. Yang, M. A. Kastner, A. Cassanho, and R. J. Birgeneau, Phys. Rev. B 51, 3671 (1995).