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Is There Any Hidden Symmetry in the Stripe Structure of Perovskite High-Temperature Superconductors?

Vladimir A. Gavrichkov,*[†] Yury Shan'ko,[‡] Natalia G. Zamkova,[†] and Antonio Bianconi*[§,∥,⊥]

[†]Kirensky Institute of Physics, Federal Research Center KSC Siberian Branch of the Russian Academy of Sciences, 660036 Krasnoyarsk, Russia

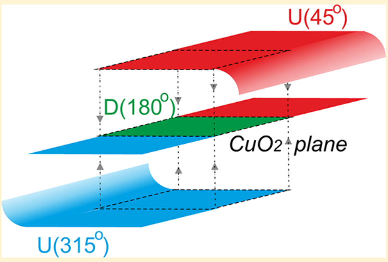
[‡]Institute of Computational Modeling of the Siberian Branch of the Russian Academy of Sciences, 660036 Krasnoyarsk, Russia

[§]Rome International Center for Materials Science Superstripes (RICMASS), Via dei Sabelli 119A, 00185 Rome, Italy

[∥]Institute of Crystallography, Consiglio Nazionale delle Ricerche, CNR, I-00015Monterotondo, Italy

[⊥]National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 115409 Moscow, Russia

ABSTRACT: Local and fast structural probes using synchrotron radiation have shown nanoscale striped puddles and nanoscale phase separation in doped perovskites. It is known that the striped phases in doped perovskites are due to competing interactions involving charge, spin, and lattice degrees of freedom. In this work, we show that two different stripes can be represented as a superposition of a pair of stripes, $U(\theta_n)$ or $D(\theta_n)$, characterized by perovskite tilts where one of the pair is rotated in relation to the other partner by an angle $\Delta\theta_n = \pi/2$. The spatial distribution of the U and D stripes is reduced to all possible maps in the well-known mathematical four-color theorem. Both the periodic striped puddles and random structures can be represented by using planar graphs with a chromatic number $\chi \leq 4$. To observe the colors in mapping experiments, it is necessary to recover variously oriented tilting effects from the replica. It is established that there is an interplay between the annihilation/creation of new stripes and ordering/disordering tilts in relation to the θ_n angle in the CuO$_2$ plane, where the characteristic shape of the stripes coincides with the tilting-ordered regions.



A characteristic feature of perovskite structures ABO$_3$, known as Glazer's systems, is a variety of tilting effects.[1,2] The latter can be caused by external factors, for example, by pressure,[3] by temperature,[4] by a discrepancy between the lattice parameters of a substrate and the material,[5] or by the misfit strain between different layers in complex materials made of stacks of atomic layers.[6−9] The unique functionality of several quantum complex materials with perovskite structure, like cuprates,[10−15] manganites,[16,17] and bismuthates,[18] can be tuned by atomic substitutions, tolerance factor, misfit strain and pressure which control local structural tilts, bond disproportionation, and nanoscale phase separation. A similar complex lattice fluctuations have been identified by extended X-ray absorption fine structure (EXAFS) in BaPb$_{1-x}$Bi$_x$O$_3$ perovskite superconductors showing bond disproportionation and correlated tilts.[19,20]

While the competition between disorder and superconductivity has been a topic of intense interest for decades, there are several exemplary systems can be found in the literature that demonstrate how correlated disorder,[21−25] manifested through doping[26] vacancies and strain[27] in both two and three dimensions,[28,29] plays an important role in the pairing mechanism[20,30,31] and can favor the enhancement of critical temperature[32−34] toward room temperature.[35,36]

It is known that cuprates show a universal dome of T_c versus doping, little is known about the mechanism beyond the variation of the maximum critical temperature of the dome, T_{c-max}, which has been found to be controlled by pressure or strain. Using advanced scanning micro X-ray diffraction, the space distribution of inhomogeneity in the nanoscale has been well understood in cuprates. It has been found that a correlated disorder induced by doping and strain leads to charge puddles creating an inhomogeneous lattice, and superconductivity is the result of percolation in a hyperbolic space through filamentary pathways defined by the puddles[37,38] which has been confirmed in iron based superconductors,[29] in bismuth sulfide[39] and it is expected also in oxyselenide[40] perovskite superconductors.

Since high-T_c superconductors are superlattices of metallic CuO$_2$ layers, an appropriate physical variable describing elastic effects is not a tolerance factor for three-dimensional ABO$_3$ systems, but the superlattice misfit strain between different layers.[41,42] Using resonant Cu K-edge X-ray diffraction,[43] Cu K-edge X-ray absorption near edge structure (XANES), and Cu K-edge EXAFS,[44,45] it was shown that the local structure of the CuO$_2$ plane of doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+y}$ (called BSCCO or Bi2212)[43−46] and of doped La$_{2-x}$Sr$_x$CuO$_{4+y}$ (called LSCO or La124)[47−50] perovskite families show a periodic anharmonic incommensurate lattice modulation described as a particular nanoscale phase separation due to the formation of alternate stripes of undistorted U stripes and distorted D stripes with

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different tilts (see Figure 1) and disproportionated Cu−O bonds in cuprates.[47] For the superlattice of CuO_2 atomic layers separated by rocksalt LaO spacer layers in the hole-doped layered perovskite family La_2CuO_4 (LCO), the relevant physical quantity is the misfit strain between the active $[CuO_2]$ and the spacer rocksalt $[La_2O_{2+y}]$ layers. For the BSCCO superlattice of perovskite bilayers $[CaCu_2O_4]$ separated by spacer layers $[Sr_2O_2]$ and $[Bi_2O_2]$, the relevant physical quantity is the misfit strain between the active CuO_2 and the spacer $[Bi_2O_2]$ layers.

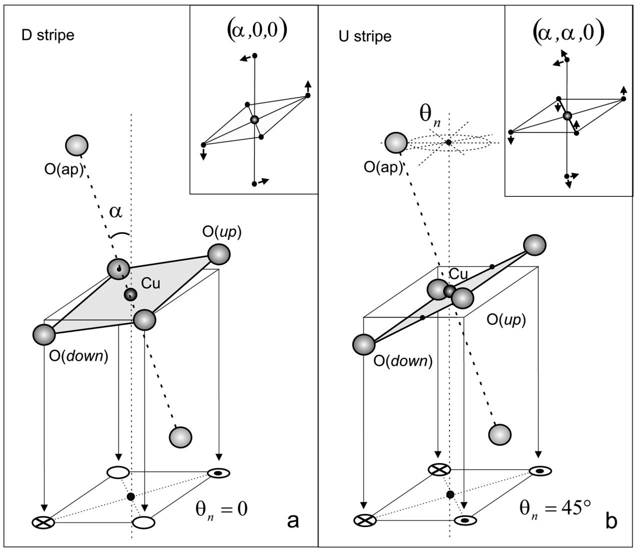

Figure 1. View of the distorted octahedra of the *D* (a) and *U* (b) stripes with different tilting effects in LSCO.[47] The planar projections in the lower part of the figure corresponds to the positions of oxygen atoms in the CuO_4 planar complex. Insets show the tilting effects in accordance with Glazer's notations.[2]

Recent experimental studies using scanning tunneling conductivity (STM) over a wide range of temperatures and doping[51] evidence that there is a clear connection between the stripe structure and electronic structure of Cu oxides supported by the fact that the pseudogap state and the stripe structure detected by EXAFS[45,48,50] and scanning micro X-ray diffraction[37] have a similar temperature region of existence.

This research was inspired by the study presented in the work,[52] where hidden symmetries were investigated in the stripe structure of high-temperature superconductors. Here, we focus on the tilting effects as a required attribute of observed stripes.[47] Based on the large isotope effect in high-T_c superconductors with the stripe nanostructure,[53−55] we tried to study the phonon nature of the stripe structure in high-T_c cuprates, and to identify the stripes according to the type of their tilting effects. For example, the *U* and *D* stripes in Figure 1 show the $(a^-a^-a^0)$ and $(a^-a^0a^0)$ tilts in Glazer's notations.[2] Unfortunately, in these notations, it is impossible to classify the stripes rotated about the axis *z* by different angles θ_n. Therefore, we introduce new notations in which an orientation of the tilting angle α can vary within a single Glazer's notation. To do this, we replace *a* by the angle $\pm\alpha$, and omit the top indices + and − as they do not change under the action of rotation about the axis *z*. Then, all the possible *U* and *D* stripes $D(0) = (0,\alpha,0)$, $U(45°) = (\alpha,\alpha,0)$, $D(90°) = (\alpha,0,0)$, $U(135°) = (\alpha,-\alpha,0)$, $D(180°) = (0,-\alpha,0)$, $U(225°) = (-\alpha,-\alpha,0)$, $D(270°) = (-\alpha,0,0)$, and $U(315°) = (-\alpha,\alpha,0)$ form a group of a complex root of unity $\sqrt[g]{e^{(2\pi i)}}$, where $g = 8$. This is the symmetry group of an isolated CuO_6 octahedron with respect to its rotation by an angle $\theta_n = n(\pi/4)$. In the LCO and LSCO layered materials, this group reduces to the two $U(\theta_n)$ and $D(\theta_n)$ groups because of different orientation of the CuO_4 planar complex relative to the rocksalt spacer layers.

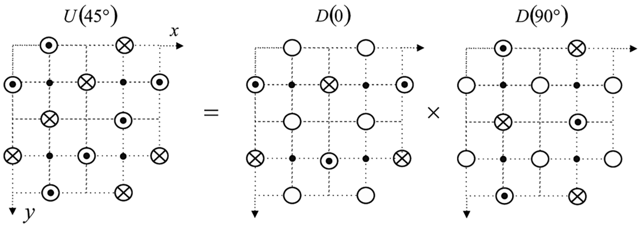

Figure 2. *U* type structure in the undoped orthorhombic LCO presented as a superposition of two *D* tetragonal structures rotated one relative to the other by an angle $\pi/2$.

In accordance with the study,[56] the undoped LCO has the orthorhombic A_{bma} (D_{2h}^{18}) structure at a temperature below 500 K, where the tilts of the perovskite octahedra are ordered into a $(a^-a^-a^0)$ sequence. The latter can formally be presented here as a superposition $(a^-a^-a^0) = (a^-a^0a^0) + (a^0a^-a^0)$, and we further consider the mono *U* stripe structure of the undoped LCO as the confinement of a pair of tetragonal structures rotated relative to each other by an angle $\Delta\theta_n = \pi/2$ (Figure 2). As a consequence, the number of stripes, in the doped LSCO, is reduced to one species (*U* stripes) with extensive overlapping areas (*D* stripes) between them. To study the nature of the spatial stripe distribution, we take into account that the $U(\theta_n)$ and $D(\theta_n)$ structures have a common origin and differ only by orientation (i.e., θ_n) of the tilting angle α. Let us collect the $U(\theta_n)$ and $D(\theta_n)$ stripes into a new group $G(\alpha)$ built from the stripes rotated relative to each other by an angle $\theta_n = n(\pi/4)$

$$\begin{aligned} U(45°) &= D(0) \times D(90°) = (0,\ \alpha,\ 0) + (\alpha,\ 0,0) \\ D(180°) &= D(0)^2 = (0,\ \alpha,\ 0) + (0,\ \alpha,\ 0) \\ O &= D(0) \times D(180°) = (0,\ \alpha,\ 0) + (0,\ -\alpha,\ 0) \end{aligned} \tag{1}$$

and so on. Following the superposition for the $U(\theta_n)$ and $D(\theta_n)$ stripes, we derive the rules of group multiplication for the stripes: $U(45°)$, $U(135°)$, $U(225°)$, $U(315°)$ and $D(0)$, $D(90°)$, $D(180°)$, $D(270°)$ (see Table 1, where the group unit *O* refers to the untilted prototype in which there are no tilts at all). The group $G(\alpha)$ of the stripe pairs presented in Table 1 appears if we assume the replacement $(2\alpha) \leftrightarrow -\alpha$ in the squared group element $D(0)^2 = (0,\alpha,0) + (0,\alpha,0) = (0,2\alpha,0) = D(180°)$, i.e., there can be only one specific angle $\alpha \sim$ const in the material. In this case, we obtain an abelian group $G(\alpha)$ of the ninth order, which is a direct product of any two cyclic subgroups

$$\begin{aligned} G &= D_1 \times D_2 = U_1 \times U_2 = D_1 \times U_1 = D_2 \times U_2 \\ &= D_1 \times U_2 = D_2 \times U_1 \end{aligned} \tag{2}$$

of the four possible:

Table 1. Multiplication Table for the Ninth Order Abelian Group $G(\varphi)$, Where the Group Unit Is an Untilted Prototype O

group elements	$D(0)$	$D(90°)$	$D(180°)$	$D(270°)$	$U(45°)$	$U(135°)$	$U(225°)$	$U(315°)$	O
$D(0)$	$D(180°)$	$U(45°)$	O	$U(315°)$	$U(135°)$	$D(90°)$	$D(270°)$	$U(225°)$	$D(0)$
$D(90°)$	$U(45°)$	$D(270°)$	$U(135°)$	O	$U(315°)$	$U(225°)$	$D(180°)$	$D(0)$	$D(90°)$
$D(180°)$	O	$U(135°)$	$D(0)$	$U(225°)$	$D(90°)$	$U(45°)$	$U(315°)$	$D(270°)$	$D(180°)$
$D(270°)$	$U(315°)$	O	$U(225°)$	$D(90°)$	$D(0)$	$D(180°)$	$U(135°)$	$U(45°)$	$D(270°)$
$U(45°)$	$U(135°)$	$U(315°)$	$D(90°)$	$D(0)$	$U(225°)$	$D(270°)$	O	$D(180°)$	$U(45°)$
$U(135°)$	$D(90°)$	$U(225°)$	$U(45°)$	$D(180°)$	$D(270°)$	$U(315°)$	$D(0)$	O	$U(135°)$
$U(225°)$	$D(270°)$	$D(180°)$	$U(315°)$	$U(135°)$	O	$D(0)$	$U(45°)$	$D(90°)$	$U(225°)$
$U(315°)$	$U(225°)$	$D(0)$	$D(270°)$	$U(45°)$	$D(180°)$	O	$D(90°)$	$U(135°)$	$U(315°)$
O	$D(0)$	$D(90°)$	$D(180°)$	$D(270°)$	$U(45°)$	$U(135°)$	$U(225°)$	$U(315°)$	O

$$D_1 = D(0),\ D(180°),\ O;\ U_1 = U(45°),\ U(225°),\ O;$$
$$D_2 = D(90°),\ D(270°),\ O;\ U_2 = U(135°),\ U(315°),\ O \tag{3}$$

In contrast to the observed orthorhombic symmetry with the (a^-,a^-,a^0) tilting effects in the undoped LCO cuprates,[56] described only by the subgroup U_1 (or U_2) while in the doped cuprates LSCO, the effects $(a^-a^-a^0)$ and $(a^-a^0a^0)$, $(a^0a^-a^0)$ from the direct product $U_1 \times U_2$ might be observed.[47]

The irreducible representations of the group $G(\alpha)$ are homomorphisms from this group into a group of three elements: 1, $e^{2\pi i/3}$, and $e^{4\pi i/3}$, that is, a third degree root of unity. The homomorphism is completely defined by how it acts on the generators of a group. In our case, these are two elements which do not belong to the same subgroup, for example, $D(0)$ and $U(45°)$. If $D(0)$ is mapped to $e^{2\pi i/3}$ and $U(45°)$ is mapped to $e^{4\pi i/3}$, then $U(135°) = D(0)U(45°)$ is mapped to 1 and similarly for all other elements of the group. The three variants of the homomorphism for each of the pair of generators of the group result in the nine different representations, and the latter are numbered by pairs of indices $\{i,j\}$, where $i(j) = 1,2,3$.

Since the active CuO_2 plane does not contain any untilted prototypes, there is no neighborhood of U stripes rotated by 180° relative to each other. All of them are separated by the $U(\theta_n)$ stripes with $\Delta\theta_n = 90°$ (or $\Delta\theta_n = 270°$), which have an overlapping region ($D(\theta_n)$ stripes). For example, $U(45°)/D(270°)/U(135°)/D(0)/U(225°)/D(90°)/U(315°)/D(180°)/U(45°)$, or $U(45°)/D(270°)/U(135°)/D(270°)/U(45°)/D(270°)/U(135°)$ with a period of the stripe structure less than the order of the group $G(\alpha)$.

The spatial distribution of the stripes in Figures 3 and 4 directly follows from the well-known four color theorem on the map.[57] In four colors, $U(45°)$, $U(225°)$, red (R); $U(135°)$, $U(315°)$, blue (B) and also $D(0)$, $D(180°)$, green (G); $D(90°)$, $D(270°)$, yellow (Y), we can always color an arbitrary map on the plane.[57] The four colors R, G, B, Y are just four subgroups: D_1, D_2 and U_1, U_2, from which the untilted prototypes O have been removed.

For example, the unidirectional charge stripe order and a chessboard electronic crystal[58,59] can correspond to one of the color maps: $R/Y/B/Y/R$, $R/Y/B/G/R/Y/B$ in Figures 3 and 4, respectively. Any stripe structures, including a random structure, are more convenient for representation using planar graphs with a chromatic number $\chi \leq 4$.[57] In accordance with the group $G(\alpha)$, all the maps are symmetric with respect to the replacement of U by D and vice versa. Recently, the four color theorem has been used to interpret the domain topology of the magnetic material Fe_xTaS_2.[60]

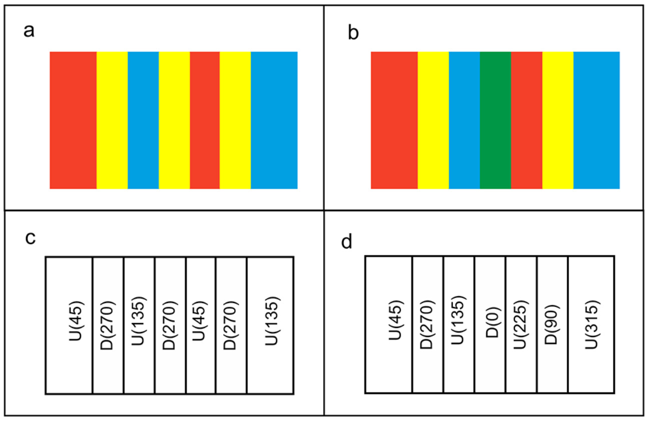

Figure 3. Color maps with the chromatic number χ = 2 for the unidirectional charge-stripe order.

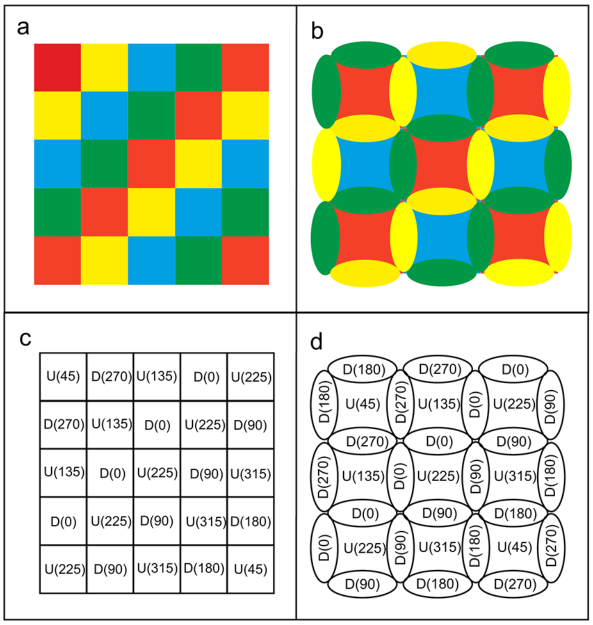

Figure 4. Color maps with the chromatic number χ = 3 for the chessboard electronic crystal. The maps (a,c) and (b,d) are topologically equivalent.

The periodic stripe maps in Figures 3b,d and 4a,c with U and D stripes equivalent in shape are invariant with respect to the simultaneous rotation of all CuO_6 octahedra by angle θ_n = 45° around the z axis. The maps in Figure 4b,d with different U and D stripes are invariant only with respect to rotation by the angle θ_n = 90°. Any random and monochrome stripe maps with $\chi = 4$ and $\chi = 1$ does not have a rotational symmetry for θ_n angle.

In conclusion, in this paper, we have demonstrated that the overlapping between the stripes may be an alternative for Bloch and Neel type of domain walls in oxides[61] at the subatomic scale. The overlapping looks like the D stripes, whose properties follow from the group multiplication in Table 1. In this group, the overlapping areas of the D and U stripes are equal, and the untilted prototype O with $\alpha = 0$ is the group unit. In the undoped LCO cuprate, the structure of one large U stripe is observed, and the material is at the state with $(a^{-}a^{-}a^{0})$ tilting effects. Under doping, the disappearance of the long-range stripe order is accompanied by the generation of the D stripes in the region of the immediate boundary between the $U(\theta_n)$ stripes oriented at different angles ($\Delta\theta_n = \pi/2$) relative to each other. The spatial distribution of the $D(\theta_n)$ and $U(\theta_n)$ stripes is reduced to possible maps in the four color problem for the CuO_4 plane, where the color represents the $D(\theta_n)$ or $U(\theta_n)$ stripes combined into four subgroups of the group $G(\alpha)$. Any observed spatial distributions, namely, the unidirectional charge-stripe order, chessboard electronic crystal or totally random structures can be represented by planar graphs with different chromatic numbers, where $2 \leq \chi \leq 3$ and $\chi = 4$, respectively. At the long-range stripe ordering, $\chi = 1$ in the undoped material LCO. By their origin, the boundaries between the U and D stripes should atomically be sharp. Moreover, in layered materials the reduced symmetry of the CuO_4 square in the D stripes agrees well with the symmetry of the local b_{1g} vibrations that are active in the $(^2a_1 + {}^2b_1) \otimes (b_{1g} + a_{1g})$ JT effect.[62,63] However, unlike the usual local b_{1g} mode, there no contribution to the multimode JT effect from the tilting mode in the CuO_6 octahedron in the isotropic environment. In the U stripes, the local symmetry of all four plane oxygens is identical even in layered materials. We expect the inhomogeneous distribution of electron density over the U and D stripes since the JT energy depends on the electron concentration only in the D stripes. To observe the colors using STM spectroscopy, it is necessary to recover variously oriented tilting effects from the STM charge replica of the CuO_2 plane. Note, compatible types of cooperative JT and tilting effects within a single structure were investigated in the work,[64] and an interplay between the tilting and JT distortions was studied previously in $LaMnO_3$, YVO_3, and $YTiO_3$.[65]

What is the physical meaning of the group $G(\alpha)$? It is not a symmetry group of the lattice Hamiltonian since the group elements $U(\theta_n)$ and $D(\theta_n)$ do not reproduce any initial stripe distribution. The group $G(\alpha)$ is just the symmetry of a pair of interacting stripes. By analogy with the Woodward–Hoffmann's rules for orbital symmetry conservation in chemical reactions,[66] we can associate the initial and final reagents in a reaction with the stripes $U(\theta_n)$ and $D(\theta_n)$ respectively. In this reversible "reaction" the orbital symmetry changes, but the tilting angle α remains a constant. Due to the group, we have established the relationship between the annihilation/creation of new stripes and ordering/disordering process of the tilts in the CuO_2 plane, where the characteristic shape of the stripes coincides with the regions ordered by the θ_n angle.

The whole variety of tilting domains in the three-dimensional ABO_3 perovskites[67] cannot be derived in the simplified approach. Indeed, we used the four-color theorem for the planar stripe maps to reflect a nature of the superconducting layered perovskites, and the group $G(\alpha)$ most likely exists due to a certain ratio between the tilting effects and JT effect.

■ AUTHOR INFORMATION

Corresponding Authors

*E-mail: gav@iph.krasn.ru.
*E-mail: antonio.bianconi@ricmass.eu.

ORCID

Vladimir A. Gavrichkov: 0000-0002-5340-7803
Antonio Bianconi: 0000-0001-9795-3913

Notes

The authors declare no competing financial interest.

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