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Out of Equilibrium Dynamics

Soliton-mediated compression density waves and charge density in 2d layers of underdoped cuprate-like lattices

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ABSTRACT

We study here the landscape of the excitations (compression density) of the nonlinear oxygen vibrations in a two-dimensional CuO₂ layer. After studying regular lattices we investigate lattices with bonds which are weakly distorted due to an influence of weak doping (below the HTSC regime). We estimate the density of compressions (strain density) in dependence of a misfit of the Cu–O-bonds. We show that with increasing misfit the nonlinear oscillations of the O-atoms are organized in stripes. Assuming that the density of doping charges follows at least qualitatively the compression density, we discuss the stripe structures of the strain density as a possible origin for the experimentally observed stripe structures.

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1. Introduction

The excitations in nonlinear lattices form a rather complex nonuniform and fast changing in time landscape of dynamic compressions. One of the basic results of the Davydov theory about solitons in molecular systems [1] is that the probability density of electrons $\rho_e(x, t)$ introduced by doping is closely related to the compression density $\rho(x, t)$ in nonlinear molecular systems. In the special case of one-dimensional (1d) supersonic solitons Davydov could show that both these (normalized) densities are for the ground state one-electron wave function of a supersonic “electrosoliton” identical in adiabatic approximation. For subsonic “electrosolitons”, Davydov found more complicated relations. In other more general situations, e.g. when having many excitations, in the case of higher dimension or with other model assumptions, the identity may not hold, but the similarity between compression density and the charge density remains a valid approximation [2,3]

$$\rho_e(\mathbf{r}, t) \simeq \rho(\mathbf{r}, t)$$

At least the structure of the landscape is the same, the maxima are at nearly the same places, the correlation is always quite high. This was demonstrated in quite a number of computer simulations for 1d systems [2,3]. This is the basic assumption upon which we built here. Recently, we have studied soliton-like excitations in two-dimensional (2d) atomic layers [4–6] in triangular lattices as well as the corresponding charge densities by using different methods like solving, e.g. Pauli master equations [2,7,8]. Here we will study more complicated two-component 2d lattices. We consider two interacting quadratic superposed sublattices or otherwise said embedded into each other of the type we found in cuprate layers. One is defined by rather motionless Cu-atoms which interact with eventually oscillating oxygen atoms.

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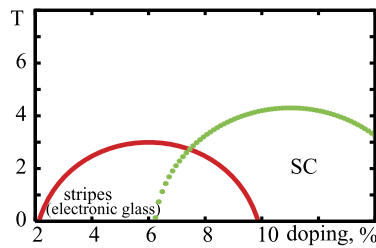


Fig. 1. Schematic temperature (Kelvin) versus doping phase diagram of cuprates as doping increases.

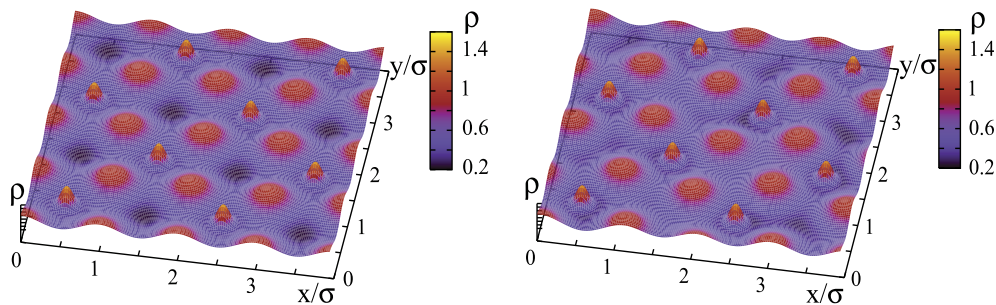


Fig. 2. The core densities of the Cu-atoms (narrower peaks) and of the oxygen atoms (broader peaks) in an equilibrium state in a (nearly) regular CuO₂ (cuprate, in short) lattice with $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.707$ (left panel) and $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.8$ (right panel) at the rather low temperature $T = 0.0002D_{\text{O-O}}$. In the first case O-atoms form practically an ideal quadratic lattice with axes oriented along x - and y -axes in addition to an ideal Cu-lattice (with axes oriented along diagonals of O-lattice). In the second case O-atoms are shifted relatively the middle point on a bond between two nearest Cu-atoms (see also Fig. 4, left panel).

Most high T_c superconductors (HTSC) on cuprate basis have a layered structure and several studies support the idea that nonlinear excitations in these layers play an essential role for the mechanisms of superconduction. The idea that nonlinear excitations may play a role for superconductivity is supported by several experimental observations which suggest that at least in some such systems anharmonic effects may play a significant role. Long ago Müller [9,10] underlined the possible role of “strongly anharmonic phonons due to spin excitations”. He proposed a model of anharmonic vibrations of apical oxygen interacting with carriers in CuO₂ planes. Special models of anharmonic oscillations were also proposed by Bussmann-Holder and colleagues [11–13].

Recent topographic and other studies give us a lot of detailed information about the microscopic charge distributions. Typically one sees stripes and other outstanding patterns. Following Tranquada, Kohsaka and others [14–22] we assume that below the HTSC regime for very low densities and outside the superconducting dome exists a regime of stripes (electronic glass). Here the long range order seems to be broken and one observes a kind of “tessellated patterns”. Our working hypothesis is that those structures may be related to the localized nonlinear excitations in the lattices. As already noted in systems of charges and atoms with nonlinear interactions, the charges (electrons or holes) densities $\rho_e(r)$ and lattice compression $\rho(r)$ are strongly correlated. We refrain from arguing about the reasons for this correlation. Let us just assume that the compression density may be at least a caricature of the charge distribution in cuprates. Our study is first restricted to underdoped cuprates as in the case of very low doping charge so that charge interactions and electron degeneration effects can be neglected as there are specific experimental studies for this region yet to be explained [17]. Thus we consider the underdoped regime below charge concentrations which may lead to pair formation (Fig. 1). The feedback of the charges to the lattice dynamics is taken into account by merely introducing elements of the fluctuating bond model [23,24], where doping changes the binding length and leads to bistable oxygen configurations. For a qualitative (mean field) account of this effect we will assume that the length parameter of the Cu–O-bond increases with the doping density. This assumption seems to be justified in the region of very low doping.

To simplify we start with an investigation of nonlinear oscillations of CuO₂ lattices without doping and study then the influence of charges in the limit of very low density. This excludes indeed all effects of pairing and quantum degeneration.

2. Model for atom–atom interactions in the lattice and the method of visualization

For simplicity we place all Cu-atoms and O-atoms in the same plane, as two superposed planes, as a kind of projections of the real atoms to the pseudo-atoms which oscillate in our plane. The players in our nonlinear dynamics are the O-atoms. In a triangular lattice the oscillating atoms each have six nearest neighbors. Here four of the neighbors are O-atoms and two of them are Cu-atoms which are fixed to a quadratic lattice on the plane (Fig. 2, left panel) [23].

The influence of light doping is small and is reflected only in a small change of the bond length of Cu–O. We write the lattice Hamiltonian as the sum of internal and onsite terms:

$$H_a = \frac{m}{2} \sum_i v_i^2 + \sum_{ik} V_{ik}(r_i, r_{k0}) + \frac{1}{2} \sum_{i,j} V(r_i, r_j) \quad (1)$$

The subscripts locate atoms sitting near to lattice sites and the summations over i and j run from 1 to N_O while the summations over k run from 1 to N_{Cu} . The positions of the Cu-atoms are all fixed at the edges of the quadratic lattice unit. For the potential we use the Morse “ansatz”

$$\begin{aligned} V_{ik}(r_i, r_{k0}) &= D_{Cu-O} \{ \exp[-2b_{Cu-O}(r_{ik0} - \sigma_{Cu-O})] - 2 \exp[-b_{Cu-O}(r_{ik0} - \sigma_{Cu-O})] \} \\ V(r_i, r_j) &= D_{O-O} \{ \exp[-2b_{O-O}(r_{ij} - \sigma_{O-O})] - 2 \exp[-b_{O-O}(r_{ij} - \sigma_{O-O})] \} \end{aligned} \quad (2)$$

Here $r_{ik0} = |r_i - r_{k0}|$, $r_{ij} = |r_i - r_j|$, with indices Cu–O and O–O referring to Cu–O and O–O interactions, respectively.

Putting the Cu-atoms and O-atoms together and giving full freedom to the O-atoms, we obtain a complex multistable potential landscape. In order to observe the rather complex dynamics in such regular or irregular lattices we need a suitable method of visualization. Here we model the lattice units or atoms as points on a plane which are surrounded by little spheres formed by the core electrons. We will assume that the latter may be represented by a Gaussian distribution centered on each lattice site and hence save a constant normalization factor

$$\rho(Z, t) = \sum_{i; |Z - Z_i(t)| < 1.5} \exp\left[-\frac{|Z - Z_i(t)|^2}{2\lambda^2}\right] \quad (3)$$

where λ is the reciprocal radius of the core of oxygen atoms. Note that for the purpose of visualization the representation of the atom positions by Gaussians is more useful than the representations by lattice points.

Here the $Z(t) = (x + iy)/\sigma_{O-O}$, the Z_i are the complex oxygen (dimensionless) coordinates which move according to the Hamiltonian (1). For the density of the Cu-atoms we set, save for a constant normalization factor,

$$\rho_k(Z) = \sum_{|Z - Z_{k0}| < 1.5} \exp\left[-\frac{|Z - Z_{k0}|^2}{2\lambda_c^2}\right] \quad (4)$$

Here the $Z_{k0}(t)$ are the complex copper coordinates, which are fixed on a quadratic lattice. The overlap of these functions gives us the compression density which is the one we focus on this work.

Fig. 2 shows an example of visualization of two equilibrium cases with a small Cu-core $\lambda_c/\lambda = 0.266$ and very low temperature $T = 0.0001$, $\eta = \sigma_{Cu-O}/\sigma_{O-O} = 1/\sqrt{2} = 0.707$ (left panel) and $\eta = 0.8$ (right panel). (Temperature is measured in $2D_{O-O}$ units. Low-temperature simulations are used to check stability of equilibrium states.) This provides a picture in which the Cu-atoms are seen only as small peaks in a landscape of moving O-density peaks. With these assumptions we may easily follow the motion of oxygen seeing the fixed copper positions only in enlarged views. For simplicity we take $\lambda_c = \lambda\sigma_{Cu-O}/\sigma_{O-O}$ assuming that the λ_c and σ_{Cu-O} parameters are proportional to each other.

In order to represent the bonds between Cu and O and their evolution we shall play with the still free λ parameter looking for values which allow an easy representation of bond structure.

The parameters $\sigma_{O-O, Cu-O}$ specify the bond lengths, $b_{O-O, Cu-O}$ the stiffness of the pair repulsion and $D_{O-O, Cu-O}$ the depth of the attracting potential. We will take σ_{O-O} as the length unit. We introduce now as our significant and tunable parameter the ratio $\eta = \sigma_{Cu-O}/\sigma_{O-O}$. The other parameters are held fixed in given values.

Two cases are of interest:

- (i) In the limit of zero doping, the O-atoms are sitting at equilibrium on a quadratic lattice where they are located half-way between the Cu-atoms (Fig. 2, left panel). Including temperature, hence heating the system, the O-atoms perform nonlinear oscillations. Assuming that there is just one charge (an electron or a hole) we may assume that the charge density follows the compression density. As a first approximation there is no feedback of the charge density on the lattice dynamics. In an ideal regular lattice we would need according to Pauling a ratio of the equilibrium distance of O–O to Cu–O as 1 to $1/\sqrt{2}$. In other words the effective binding radii should be in the ratio of $\sqrt{2}$. In fact the actual ratio of the lattice radii is around $0.96/1.40 \simeq 0.69$ near to $1/\sqrt{2}$. In other words the Pauling radii are near to the ratios which are ideal for regularity though the O–O radii are a bit larger but this has only a small influence.
- (ii) At non-vanishing but yet undercritical doping we have according to the fluctuating bond model [23,24] a small increase of the Cu–O-bond length leading to a linear relation $\eta(n) = \sigma_{Cu-O}/\sigma_{O-O} > 1/\sqrt{2} = 0.707$. This brings specific effects (formation of stripes) which we are going to study now.

3. Simulation of the oxygen motions

We start with some computer simulations of the regular or nearly regular case (Fig. 3). Newns and colleagues [23,24], calculated with quantum-chemical methods the potential landscape in which the oxygen atoms are moving, and estimated, in particular, the influence of doping. It was shown that a finite amount of doping changes the binding length and leads to the possibility of bistable configurations. The potential landscape was locally approximated by quartic potentials. We used

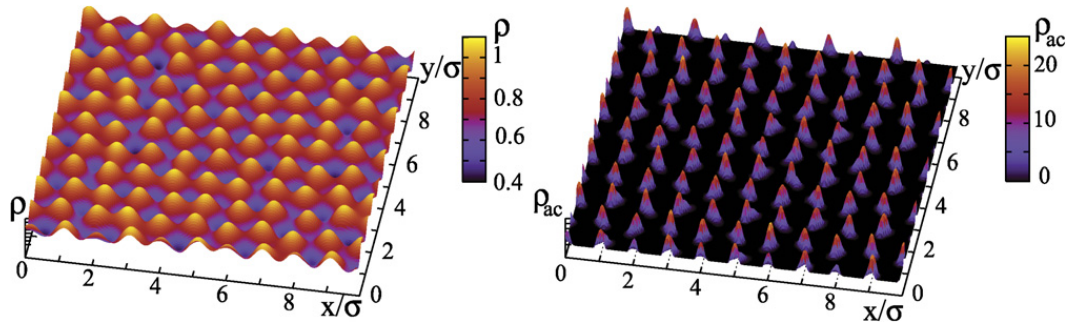


Fig. 3. Snapshot of the compression density of oxygen in a rather regular cuprate lattice with $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.707$ at low albeit finite temperature when nonlinear oscillations are excited (left panel) and corresponding averaging (in time) distribution of excess-density $\rho_{ac} = \rho - 1$ (right panel). One may observe that O-atoms oscillate with small amplitude near equilibrium positions in-between two neighbor Cu-atoms.

a different approximation of such potential landscape, by using Morse potentials for the Cu–O-bonds. We have shown that bistable landscapes of the oxygen potential may be created by Morse potentials with increased length for the Cu–O-bonds. For a qualitative (mean field) account of the bistability we will assume that the length parameter of the Cu–O-bond increases monotonically with doping density,

$$\eta(n) = 1/\sqrt{2} + \alpha n + \dots$$

where n is the (low) doping density. We assume that at least for small doping the deviation of the oxygen positions at rest from the line connecting two Cu-atoms goes linearly with the doping density.

For a regular lattice corresponding to zero doping, as already noted, the ideal ratio is $1/\sqrt{2} \simeq 0.707$. Now we assume that the ratio of the radii is a bit larger $\eta = \sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} > 0.707$, and increases slowly with density. Let us further assume that $(b\sigma)_{\text{O-O}} \sim 5$ characterizes the stiffness of the O–O potential and $(b\sigma)_{\text{Cu-O}}$ the stiffness of the Cu–O potential which is assumed to have a similar value. For special sets of parameter values we observe bistability and the potential reminds then the quartic type potential along the axis perpendicular to the Cu–Cu-bond given by Newns and Tsuei [23]. We underline that this is a different potential model but leading to nearly the same potential landscape, leading in particular to quartic potentials for the oxygen atoms are moving, but of the consequences of a locally bistable behavior on the nonlinear oxygen dynamics. This is the central point of our study and we want to underline, that the potential landscape created by our Morse model is flexible enough (using adapted parameters) to model even fine details of bistable and globally multistable landscape but the consequence of such structures for the nonlinear dynamics.

We investigate now the energy landscape of a system admitting some small misfit of the parameters and show that this may lead indeed to fluctuating bonds. We remind that we take the equilibrium distance between two O-atoms $\sigma = \sigma_{\text{O-O}}$ as the unit of the distance. Furthermore in our model the equilibrium distance of Cu–O is $\sqrt{2}\sigma_{\text{O-O}}/2$ and the Cu–Cu equilibrium distance is $\sigma_{\text{O-O}}\sqrt{2}$. In the fluctuating bond model we have deviations from these regular bond lengths. Let us analyze the consequences of small changes of the bond length. We expect that an increased bond length Cu–O will expel out the oxygen from the axes and will create a positive or negative elongation. Recall that we consider one O-atom in an equilibrium cage surrounded by four O-atoms and two Cu-atoms. The potential in the center of the cage may be represented by 6 contributions from the 6 neighbors and may show indeed bistability. Note, however, that the O-atom may be influenced by other nearby atoms while it moves far enough from its initial equilibrium position.

The “fluctuating bond” condition is that the extremum connected with the regular position is higher than the global minimum what means in practice that the originally regular position is unstable. The condition that the side minima are deeper as the central extremum may be fulfilled for a large set of parameter values. The search for an appropriate set of parameter values may be done by adjusting the Morse system to fit the fourth-order polynomial calculated by Newns and colleagues [24].

Let us look at various possibilities. As Fig. 4 shows irregular bonds are created and in the case of multistability the system may fluctuate between these bonds. Further we see that with increasing misfit quite interesting stripe structures appear which remind a kind of “tessellation” which is due to the nonlinear oscillations as shown in Figs. 5 and 6.

4. Discussion and conclusions

Our interpretation of the potential obtained by Newns and Tsuei [23,24] is that the observed bistability is due to an increase of the bond length with increasing doping. In order to take this effect into account we have introduced a misfit of the bond length using our Morse potential.

The main result found here is that the dynamic structure hence the evolution of the cuprates is strongly affected by any misfit of the bond length. Therefore if one assumes that this is the result of doping, then it gives some explanation for the observed stripe structures. We assumed here that the changes due to doping have the physical meaning that in the lowest approximation the ratio of the length of the Cu–O-bond to the O–O-bond is changed. In order to check this idea we played

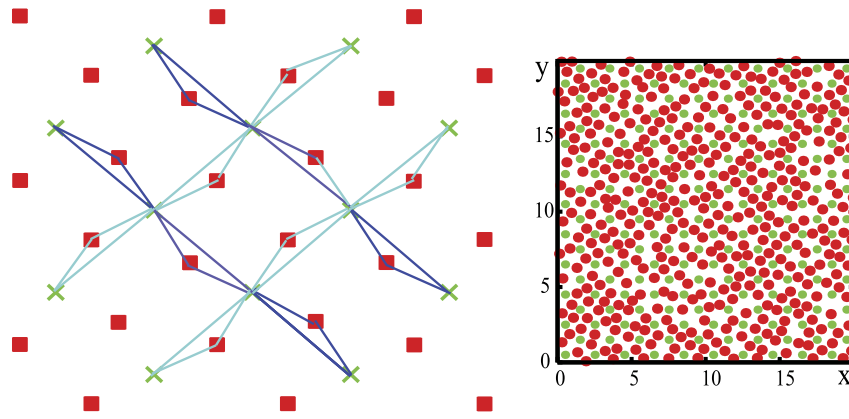


Fig. 4. Cu–O-lattice with irregular, i.e., fluctuating bonds. Left panel: Schematic representations of the oxygen atoms (squares) in the plane sitting in equilibrium on a quadratic lattice and interacting with some Cu-atoms (crosses) with a small misfit (fluctuating bonds) of the Cu–O interactions. The Cu-atoms are fixed at the edges of a quadratic lattice. Right panel: Snapshot from a large fragment of a lattice with fluctuating bonds; most of the O-atoms (red) are displaced off the mid-point of the connecting line between the Cu–Cu and form stripes oriented along lines parallel to axes of Cu-lattice.

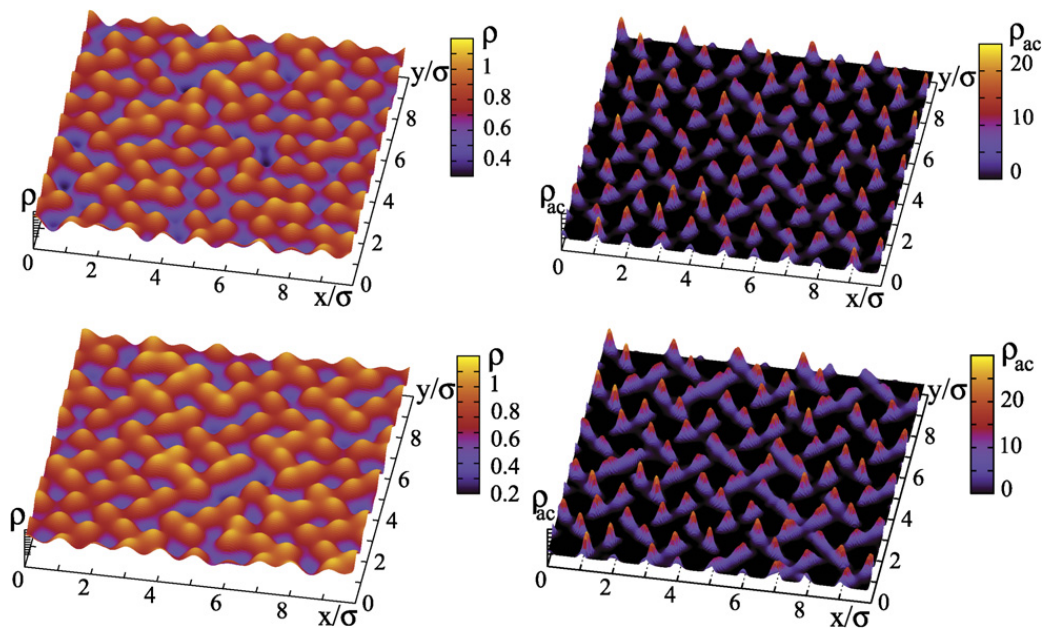


Fig. 5. The same as in Fig. 3 but with $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.74$ (upper panel: oscillations amplitudes of O-atoms increase in comparison with a case in Fig. 3) and $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.8$ (lower panel: O-atoms oscillate in each of two possible potential minima jumping between them from time to time).

with the bond length of the original system where according to Pauling this ratio is given by $1/\sqrt{2}$. Within this concept, the change of the bond length is the main cause for the nonlinear oscillations in the lattice.

Our significant parameter is the ratio of the radii $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}}$. For a regular lattice the ideal ratio is $1/\sqrt{2} \simeq 0.707$. The quantity $b_{\text{O-O}}$ is the stiffness of the O–O potential and $b_{\text{Cu-O}}$ is the stiffness of the Cu–O potential. For special sets of parameter values we observe bistability as in the case of the quartic potential along the axis perpendicular to the Cu–Cu-bond proposed by Newns and Tsuei [23].

The Cu-lattice gives the frame for our game, the O-atoms are quite free and are looking for the “best place” according to given Morse-type interactions. Note that an O-atom has six nearest neighbors in the plane. A special property of the model is the multistability, the existence of a high number of ground states instead of one ground state in the case of a regular lattice. What we see from our computer simulations, is the basic role played by the crystallographic axes formed by the O-atoms. Along these axes nonlinear oscillations of the oxygen atoms are performed. It remains to look for the formation of soliton-like structures. Note that we see always finite excited pieces of the O axes. Since there are two axes, perpendicular to each other, the direction of the oscillations may change, so that we see structures which are like tessellated patterns. The Cu–O-bonds, where presumably most of the added excess electrons are accumulated, are located right and left from the O–O–O, ... axes. So we may imagine the existence of some “electronic streets” right and left from the excited O axes.

In summary we may conclude that in our rather simplified model we observe quite complex motions of the O-atoms oscillating in the field of the other O-atoms and in the field of the Cu-atoms. The structure includes at finite (but low) doping multistable potential landscape which is responsible for the existence of multistable positions of the oxygen atoms according

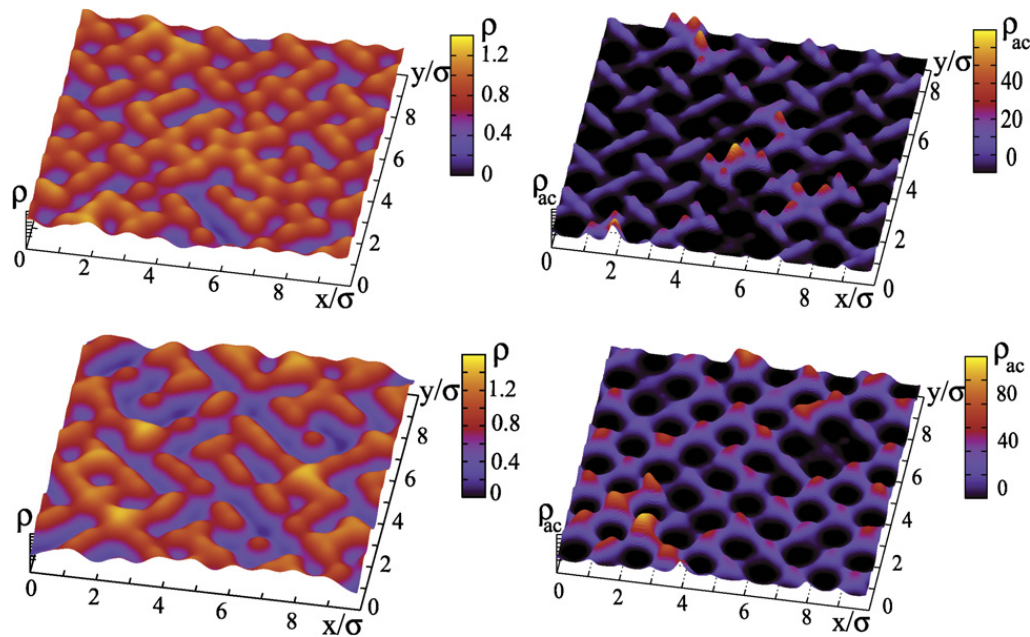


Fig. 6. The same as in Fig. 3 but with $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.85$ (upper panel: O-atoms move between two minima mainly) and $\sigma_{\text{Cu-O}}/\sigma_{\text{O-O}} = 0.95$ (lower panel: trajectories of O-atoms form a dynamical lattice with axes parallel axes of Cu-lattice). The last gives a base to suppose that motion of excitations (possibly, localized ones) along axes of this dynamical lattice may stimulate transport of electrons along axes of Cu-lattice.

to [23,24]. Our computer simulations show that this leads to a transient tessellation of the 2d structure offering transient oxygen “streets” which may have a strong influence on the electron transport. The parameter space is so wide that we could explore so far only a small subset of all possibilities of nonlinear excitations in particular those leading to stripe structures. We have seen here several types of such structures for different choices of the misfit parameter $\eta(n)$. We note that for the charge transport the nonlinear structures oriented on the O–O axes maybe of interest. Since the hopping transport is in zeroth approximation from copper to copper position, it remains to clarify how the electronic streets are related to the “streets of excitations”. Our view is that the nonlinear excitations reflected in the compression density significantly influence electron or hole dynamics, eventually leading to the observed dynamic patterns of the electron density [25].

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