Modelling quasiparticles and inhomogeneous pseudogap emergence in cuprates in presence of charge ordering potential

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Abstract. We are modelling the ground normal state of cuprate high-temperature superconductors, taking into account that they have strongly polarizable ionic lattice, with the aim to study the pseudogap (PG) nature. At strong Fröhlich electron-phonon interaction autolocalized carriers form charge ordering and coexist with delocalized ones. We consider quasiparticles (QPs) that emerge in cuprates in presence of additional charge ordering potential. We show that this potential transforms Bloch QPs into distributed wave packets with different momentums in areas with different potential. Modelling dispersion of the hole-doped cuprates and constructing the momentum space trajectories of the new QPs we found that topology of hole-like dispersion forbids QPs with average momentums near antinodes. Considering photoemission of carriers from permitted QP states, we recently have demonstrated that calculated antinodal spectrum of angle-resolved photoemission spectroscopy (ARPES) has all the features characteristic of the PG behaviour in cuprates. Using the approach defined above, here we calculate influence of fluctuations in dopant ion density on the PG width. As a result, we demonstrate that the suggested model of the PG emergence in cuprates reproduces all the details of the PG display not only in ARPES but also in scanning tunneling microscopy experiments, where the prominent feature is inhomogeneity of the PG width over the crystal.

1. Introduction
Hole-doped cuprate superconductors keep two main enigmas for researchers: pairing mechanism and pseudogap (PG) nature [1], here we study the second one. PG is a gap in the carriers’ spectrum near Fermi surface (FS) in the vicinity of points ($\pm \pi, 0$), (0, $\pm \pi$) (so called antinodes) in the First Brillouin zone (FBZ). PG is observed at temperatures up to $T^*$ which is higher than the temperature of superconducting transition, except overdoped systems. The superconducting transition in cuprates is a transition from the very unusual PG state into the superconducting state [1], therefore unveiling the PG nature is necessary to understand the ground normal state of cuprates and may shed light on pairing mechanism in them. Two existing models of the PG emergence in cuprates are conventional charge density wave model and recent pair density wave scheme [2]. However, they both have discrepancies with experiments. The first cannot describe the so-called Fermi momentum misalignment, or particle-hole asymmetry recently found in angle-resolved photoemission spectroscopy (ARPES) experiments [2,3], the second leads to superconductivity at very high temperatures due to pairing of the antinodal carriers [2], which is not observed in cuprates.
We reveal a third possible mechanism of the PG emergence in cuprates. It is neither scattering of Bloch carriers on each other (as in conventional charge density wave model) nor their pairing (as in pair density wave scheme). Charge ordering (CO) with short coherence length is well known phenomenon in cuprates [4]. In systems with strong long-range (Fröhlich) electron-phonon coupling such CO is formed by large bipolarons whose size is much larger than the unit cell, depends on doping level and determines the CO period [5]. They create potential that influences the delocalized carriers which coexist with autolocalized ones provided the latter are of the large radius (it is the case for Fröhlich coupling occurring in strongly ionic lattices). Here we model this influence (Section 2) and demonstrate how it results in PG appearance in systems with hole-like dispersion (Section 3). Then we study the emergence of PG spatial inhomogeneity by incorporating inhomogeneous distribution of dopant ions [6] into the model and compare the results of calculation with experiments on cuprates (Sections 4 and 5). As will be shown, in the suggested approach the PG width coincides with the amplitude of the potential additional to the lattice potential. Therefore, the PG width becomes spatial dependent in presence of inhomogeneously distributed dopant ions. As a result, we demonstrate that the suggested model of the PG emergence in cuprates reproduces all the details of the PG display not only in ARPES spectra [3,7] (as is described in detail in [8]) but also in scanning tunneling microscopy (STM) experiments [6,9], where the prominent feature is PG inhomogeneity over the crystal.

2. The model and methods
Taking advantages of two-dimensional character of the electron dispersion in cuprates (where conductivity occurs in CuO planes and is negligible in the direction perpendicular to the CuO planes [10]) we develop a method to obtain stationary states of delocalized electrons in additional potential created by bipolaronic CO and by both CO and inhomogeneity in dopant ions distribution. As experiments show, in the CO phase of cuprates charge density in the conducting CuO planes is proportional to the sum $cos(Kx)+cos(Ky)$ [11] albeit with a short coherence length. The CO period along axes x and y is bipolaron diameter: $C=2\pi/K=2R$ [5]. Together with the known total charge of a bipolaron $2e/\varepsilon_0$ [12], where $\varepsilon_0$ is static dielectric constant, this allows calculating the amplitude of the charge density in the conducting planes (equating the integral from the charge density over the bipolaron volume to $2e/\varepsilon_0$) and the CO potential.

It is natural that resulting CO potential $U$ is also quasi-periodic function with the same period as CO and short coherence length. Below we use $R=7.8\text{Å}$ that corresponds to $K$ measured in BSCO2201 at the doping level $p=0.11$ [4]. At such $R$ and $\varepsilon_0=50$ the amplitude of the calculated (as is described in detail in Section 4) electron energy in the CO potential is $U_0=0.06$ eV. It coincides with the PG width at such doping observed experimentally [6], the reason of the coincidence is discussed in Section 3.

Slightly overestimated value of the static dielectric constant is caused by simplified model used (as is discussed in Section 4). Amplitude $U_0$ of the additional potential energy becomes spatial dependent if inhomogeneous distribution of dopant ions is taken into account. Examples of additional potential created by CO and by CO together with inhomogeneous distribution of dopant ions (calculated as is described in Section 4) are presented in Figure 1, panels a and b, respectively.
Figure 1. (a) and (b) Examples of the additional potential energy of electrons due to CO potential and due to joint potential of CO and inhomogeneously distributed dopant ions, respectively.

To solve the Schrödinger equation with additional CO potential $U$

$$
\left[-\frac{\hbar^2 \nabla^2}{2m} + V + U\right] \Psi = E\Psi
$$

we develop a method [8] reminiscent of finite elements one. We divide the conducting CuO$_2$ plane into small squares where the additional potential energy $U_j$ of an electron can be considered constant. In each square, we look for the solution of the Schrödinger equation (1), corresponding to one and the same quasiparticle (QP) energy $E$ in all the squares, as an expansion in terms of Bloch waves $\psi_k$:

$$
\Psi_j = \sum_k C_{jk} \psi_k \equiv \psi_k, \quad (2)
$$

$$
\left[-\frac{\hbar^2 \nabla^2}{2m} + V\right] \psi_k = \varepsilon(k) \psi_k, \quad (3)
$$

where $\varepsilon(k)$ is Bloch electron dispersion. Let us consider influence of the CO potential on a Bloch electron state with some wave vector $k_0=(k_{x0}, k_{y0})$. In a square with zero CO potential equation (1) is reduced to (3) so that its solution is a linear combination of $\psi_{k0}$ and Bloch waves reflected from the square boundaries and thus having the same absolute values of the wave vector projections $(\pm k_{x0}, \pm k_{y0})$. In adjacent squares with non-zero CO potential the total QP energy is the same whereas the potential energy is changed and, accordingly, the effective kinetic energy too. Therefore, in adjacent squares as well as in any subsequent squares the stationary state of the carrier is described by Bloch wave function with the absolute values of the wave vector projections that satisfy the following system of equations (as one projection of the momentum is parallel to the square boundary it is conserved at the boundary):

$$
\begin{cases}
\varepsilon(k_j) = E - U_j \\
k_{y,j-1} = k_{y,j} \\
k_{x,j-1} = k_{x,j}
\end{cases}
$$

for the boundary parallel to $y$ and $x$ axes, respectively. These equations (together with the standard boundary conditions for the wave function and its derivative) completely determine the carrier stationary states in additional CO potential. One can see that distribution of the QP state over wave vectors resembles a wave packet. However, in this distributed wave packet the components with different wave vectors are present in different areas of the coordinate space with different CO potential. The QP state can be characterized by the momentum in areas with zero additional potential, denoted as the average momentum for the sake of brevity.

As it will be seen below the carrier dispersion determines whether the real solution of Eqs.(4) exist in different areas. We model carrier dispersion near FS in the hole-doped cuprates with a function that provides arc-shaped curves of the constant energy, including FS, (some of them are represented in Figure 2 as dotted curves) and sufficiently flat dispersion near the FS:

$$
\varepsilon(k) = E_0 - c * \left(\sqrt{(k_{jx} - b)^2 + (k_{jy} - b)^2} - \tilde{k}_0\right)^d, \varepsilon \leq E_0
$$

$$
\varepsilon(k) = E_0 + c' * \left(\tilde{k}_0 - \sqrt{(k_{jx} - b)^2 + (k_{jy} - b)^2}\right)^{d'}, \varepsilon > E_0.
$$

The centers of the four arcs, according to experimental data [13] on cuprates, may be located outside the FBZ (as is demonstrated in Figure 2), therefore the absolute value of the center coordinates $b=\pi/a=0.813\AA^{-1}$, where $a$ is the lattice constant in the conducting plane. $\tilde{k}_0$ is the radius of the arc corresponding to the QP energy $E_0$. Below we use the following values of the dispersion parameters: $E_0=0.5$ eV, $c=1.5$, $d=0.99$, $c'=0.5$, $d'=0.26$, $b=0.86\AA^{-1}$, $\tilde{k}_0=0.59\AA^{-1}$.

Below it will be convenient to characterize QP state by its energy $E$ and angle $\alpha$ between the QP momentum and $x$ axis (instead of the momentum projections $k_{x0}$, $k_{y0}$) in areas with zero additional potential, as it is shown in Figure 2(a,b). Angle $\alpha$ is related with the angle $\gamma$ (also shown in Figure 2) which is a generalization of the FS angle notion used in ARPES.
3. Momentum space trajectories of the QPs and construction of the antinodal ARPES spectrum

Now when the way to obtain the stationary states of carriers in additional CO potential are clear it is possible to discuss emergence of near-antinodal PG characteristic of hole-doped cuprates [3,7]. To do this let us construct momentum-space trajectories of the new QPs with average momentums near antinodes solving Eqs.(4) with the dispersion (5). Two examples of them are shown in Figure 2 (a,b). They demonstrate geometrically that system of equations (4) with hole-like dispersion ε(\(k\)) have no real roots for average momentums (momentums in areas with zero additional potential) near antinodes. Momentum space trajectory presented in Figure 2(a) corresponds to QP with the energy \(E\) and momentum directed at the angle \(\alpha_1\) with respect to x-axis in areas with zero additional potential. It reaches both surfaces corresponding to the minimum \((E-U_0)\) and the maximum \((E+U_0)\) kinetic energy, that corresponds to unimpeded QP propagation through areas with the maximum and minimum additional potential, respectively.

The trajectory presented in Figure 2(b) corresponds to the same energy \(E\) and a smaller angle \(\alpha_2<\alpha_1\) between the momentum and x-axis in zero-potential areas and, accordingly, smaller angle \(\gamma_2<\gamma_1\). As Figure 2(b) shows this trajectory is interrupted at the First Brillouin zone (FBZ) boundary not reaching the surface of the maximum kinetic energy (corresponding to the minimum additional potential energy \(-U_0\)). This occurs at any mesh size demonstrating that such QP cannot propagate. The reason is impossibility for kinetic energy of the state \((E, \alpha_2)\) to reach values near the maximum \((E+U_0)\) value along the QP momentum trajectory in the FBZ at hole-like dispersion. Thus, the carrier states with the energy \(E\) and angle \(\alpha_2<\alpha_1\) (or \(\gamma_2<\gamma_1\)) correspond to real QPs whereas the stationary states with \(\alpha<\alpha_1\) (or \(\gamma<\gamma_1\)) cannot exist in systems with the hole-like dispersion (5) [8].

How the revealed absence of near-antinodal QPs will display itself in experiments? Ordinarily, PG is studied experimentally with STM and ARPES. In the present approach the PG width measured with STM method coincides with the minimal value of the carrier energy in the CO potential. Indeed, the bias equal or higher than the amplitude of the CO potential compensates the negative half-wave of the CO potential responsible for exclusion of near-antinodal QPs. Earlier we have calculated antinodal ARPES spectrum as a set of narrow stripes which are intersections of the momentum space trajectories of permitted QPs having different energies (like that in Figure 2(a)) with the FBZ face (more precisely...
with a line parallel to FBZ face but shifted by a small value about 0.02 Å⁻¹ to the origin) [8]. As ARPES measures the probability of the electron to be in a state with certain energy and momentum, the intensity of the stripe corresponding to given QP energy is inversely proportional to the area of the momentum-space trajectory of the QP with this energy.

The obtained in such a way spectrum demonstrates shift of the spectral weight down in the binding energies and giant broadening [8]. Just the same features are characteristic of the PG display in ARPES spectra of cuprates [3,7]. At the momentum equal to \( k_F \) at \( T>T^* \) where PG is absent the spectrum is shifted approximately by the amplitude \( U_0 \) of the CO potential; thus, the PG width is approximately equal to \( U_0 \) [8]. The developed approach allowed to calculate doping dependence of the PG width (caused by doping dependence of the CO period) and doping evolution of the PG onset temperature \( T^* \) (as a temperature corresponding to thermal decay of 95% of bipolarons responsible for the CO potential) [8]. Calculated dependences are in good agreement with that observed in cuprates. Besides, in agreement with the experiments [1] in the suggested approach the calculated \( T^* \) is higher than the temperature of the CO onset \( T_{CO} \), although CO in it is a reason of the PG. Indeed, for experimental observation of the CO much larger number of bipolarons than 5% of their maximum number is necessary [5], such bipolaron densities take place at essentially lower temperature.

### 4. Modelling emergence of spatial inhomogeneity of the PG width

Intriguingly, that PG width measured by STM varies not only with doping but also in space: it demonstrates deviations from the average value by about 0.01-0.02eV in the areas of several unit cell size [6,9]. Experimental data revealed also that the deviation areas correlate with inhomogeneities in dopant ions distribution in charge reservoir layer [6,9]. The developed approach relating the PG width with the additional potential for charge carriers makes possible direct taking into account a contribution into it stemming from inhomogeneity of dopant ions distribution. Thus, the approach may allow clarifying emergence of spatial inhomogeneity of the PG width observed in STM studies.

We model inhomogeneity in dopant ions distribution in the continual approximation. Fluctuation of the density \( \rho \) of dopant ions’ charge in a reservoir layer we model as its increase by \( \Delta \rho \) (in comparison with the average charge density \( \rho_0 \), \( \Delta \rho=\rho-\rho_0 \)) in the square area with the center in \((0,0)\) point (were the minimum of the electron energy in CO potential is) and side \( L \) and \( \rho \) decrease, according to the electroneutrality condition, in neighboring area of different shapes shown in Figures 3 and 5. Due to electroneutrality the average charge density \( \rho_0 \) in the charge reservoir layers is equal in the absolute value to the average charge density in the conducting \( \mathrm{CuO}_2 \) planes. For the simplicity we consider the influences of these constant parts are compensating each other at homogeneous distribution of dopant ions. This simplification results in slight overestimation of static dielectric constant providing coincidence of the calculated additional potential amplitude with the experimentally observed PG width. It should be noted that in the hole-doped cuprates in the conducting plane there are electron bipolarons and hole bipolarons as well as compensating charge of electron bipolarons, whereas the compensating charge of hole bipolarons is in the charge reservoir layers. As study with the generalized variational method [5] has shown at doping level corresponding to CO presence the densities of electron and hole bipolarons coincide.

Calculating additional potential generated by to both CO and fluctuations of the dopant ions density we take into account two conducting planes with the energetically profitable antiphase CO in them, that provides electroneutrality of the considered area. The system is represented by Figure 3(a). Interestingly, addition of other pairs of conducting planes and corresponding pairs of the charge reservoir layers do not change the CO potential obtained. The energy of an electron in the CO potential is:

\[
U(\mathbf{r}) = -e \int \frac{\rho^{\text{CO}}(\mathbf{r}')|d^3\mathbf{r}'|}{|\mathbf{r}-\mathbf{r}'|},
\]

where \( \rho^{\text{CO}}(\mathbf{r}) \) is the charge density in the conducting planes and integration is carried out over a so large area in the \( xy \) plane (parallel to conducting planes) that its size does not influence the result and includes two conducting planes and two charge reservoir layers in the direction perpendicular to the \( xy \)
plane as is shown by Figure 3(a). The electron energy due to potential of excess dopant ions charge is calculated according to expression similar to (6) but with constant charge density inside the square areas (red and black) shown in Figure 3(a).

Contribution to $U(x)$ function from the fluctuations of dopant ions density shown in Figure 3(a) is represented by Figure 3(b). The sum of the fluctuation contribution to $U(x)$ for different size $L$ of the fluctuation area having the shape shown in Figure 3(a) and different values of the relative fluctuation of the dopant charge density $\Delta \rho/\rho_0$ and the electron energy in bipolaronic CO is depicted by Figure 3(c-f). The CO period used in Figures 3-6 is 15.6 Å that is the CO period measured in BSCO2201 at the doping level $p=0.11$ [4]. The interlayer distances used in Figures 4-6 are equal to 6.15 Å.

As Figure 3 shows, noticeable changes in the PG width, which is equal to the carrier potential energy in the minima, appear when the size of the fluctuation area becomes of the order of CO period or larger. The PG width in systems corresponding to panels (e) and (f) is about 0.05 eV in (e) and 0.04 eV in (f) in the areas with increased density of dopant ions' charge and about 0.075-0.08 eV in the areas with decreased density, in areas without fluctuations the PG width is 0.06 eV. These values are in good consent with those experimentally observed [6,9] with STM method in different areas of one and the same sample. Figure 4 represents the electron potential energy in the field of CO and square fluctuations of dopant charge density the opposite sign of the fluctuations in comparison with those.

Figure 3. (a) Charge distribution in the model: circles represent schematically bipolarons (hole bipolarons are blue and electron ones are grey), grey planes depict charge reservoir layers, red and black squares are the areas with increased and decreased charge density, respectively; (b) the electron potential energy in the field of fluctuations in dopant ions distribution, shown in (a); red line 1 and black line 2 are due to red (central) and black squares, line 3 is their sum; the relative deviation of the dopant charge density from the average value $\Delta \rho/\rho_0=(\rho-\rho_0)/\rho_0=1$, the size of the fluctuation area $L=16\AA\approx 2R$. (c)-(f) The electron potential energy in the filed of CO and square fluctuations of density of dopant ions’ charge, (c) for $\Delta \rho/\rho_0=0.5$, $L=8\AA\approx R$, (d) for $\Delta \rho/\rho_0=1$, $L=8\AA$, (e) for $\Delta \rho/\rho_0=0.5$, $L=16\AA\approx 2R$, (f) for $\Delta \rho/\rho_0=1$, $L=16\AA$. 

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**Figure 4.**
shown in Figure 3. As Figure 4 demonstrates the permutation of the fluctuation charges results simply in the permutation of areas with increased and decreased PG width.

Figure 4. Calculated electron energy in the potential created by CO and inhomogeneity in dopant ions density as function of x coordinate in the system shown in Figure 3(a) but with permutation of signs of the fluctuation charge (the decrease of the charge density is in the red area and the increase is in the black one). Panel (a) \( L=8\text{Å} \approx R, \Delta \rho/\rho_0 = 1 \), (b) \( L=16\text{Å} \approx 2R, \Delta \rho/\rho_0 = 0.5 \), (c) \( L=16\text{Å}, \Delta \rho/\rho_0 = 1 \). The case \( L=8\text{Å} \approx R, \Delta \rho/\rho_0 = 0.5 \) is not shown as it is completely coinciding with Figure 3 (c).

Figure 5 depicts the electron energy in the potential of dopant density fluctuations and CO in the case of frame-like shape of the area with decreased dopant density (shown with black in the inset, Figure 5(a)). In such case for the PG width change becomes noticeable the larger area with decreased dopant charge density is necessary. When the area of the frame-like region with decreased charge density is twice the area of the square with increased charge density the PG width change in the area where it is decreased becomes the same as in the case of square areas. And even in the case of increased frame-like area the PG width in the area with decreased dopant charge density does not increase noticeably, in distinct from the square-shaped areas. Smearing the region with decreased dopant density over the frame-like region vanishes the PG increase in this area as the area becomes narrow in comparison with the CO period.

Figure 5. (a) The electron energy in the potential of dopant charge density fluctuations with frame-like shape of the area with decreased dopant density shown with black in the insets of Figure 5(a,b). Panel (a) \( L=8\text{Å} \approx R, \Delta \rho/\rho_0 = 2 \) in the red area and \( \Delta \rho/\rho_0 = 1 \) in the black one (the black area is twice the red one in all the panels), (b) \( L=16\text{Å} \approx 2R, \Delta \rho/\rho_0 = 1 \) in the red area and \( \Delta \rho/\rho_0 = 1 \) in the black one, (c) \( L=16\text{Å}, \Delta \rho/\rho_0 = 2 \) in the red area and \( \Delta \rho/\rho_0 = 1 \) in the black one.
5. Discussion and conclusion

The PG width is clearly seen in STM spectra since the tunneling differential conductance $dI/dV$ as a function of the bias $V$ directly represents the local density of states [6]. Therefore, let us compare results of our modelling the PG spatial inhomogeneity with the STM spectra of hole-doped cuprates [6,9]. As was mentioned above, in the present approach the PG width is the minimal value of the additional potential. In presence of inhomogeneities in the dopant distribution the minimal values become different in different areas as is demonstrated by Figures 3-5. Namely, in the area with increased density of dopants the minimum value is decreased in the absolute value and in the areas of decreased dopants density it is increased in the absolute value. To make a noticeable impact on the PG width the size of the fluctuation area should be close or larger than the CO period. The obtained results are in consent with the PG width variation experimentally observed with STM method [6,9]. The experimentally observed distribution of the PG width over the crystal represents grain structure consisting of grains with increased, average and decreased PG width with the size of the order or larger than the CO period [6,9]. The calculated degree of the PG width change due to variation of dopants density demonstrated by Figures 3-5 is also in agreement with one observed experimentally [6,9].

In summary, strong Fröhlich electron-phonon coupling results in bipolaronic CO which coexists with delocalized carriers. Potential created by CO reconstructs the QPs into distributed wave packets having different momentums in areas with different CO potential. Hole-like dispersion forbids new QPs with the momentums near antinodes. This displays itself as PG in ARPES and STM spectra [8], and all the characteristic features of this PG coincides with those observed experimentally in hole-doped cuprates [3,6,7,9]. In particular, the inhomogeneities in the dopant ions distribution over the charge reservoir layers were suggested to be related with the PG width spatial variation [6]. The present approach allows directly take the inhomogeneity into account in calculating the additional potential for charge carriers. The calculated variation of the PG width is in consent with STM experiments [6,9] both in absolute value of the variation and in the size of the fluctuation areas. Thus, in the frames of the suggested approach that takes into account effects of strong Fröhlich electron-phonon coupling we not only reveal a possible source of the PG in hole-doped cuprates but also demonstrate the mechanism of influence of inhomogeneity in dopant ion density on the PG width.

Acknowledgements

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