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Relaxation of strongly coupled electron and phonon fields after photoemission and high-energy part of ARPES spectra of cuprates

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Abstract

An approach to considering systems with a high concentration of correlated carriers and strong long-range electron–phonon interaction and to calculating the high-energy part of the angle-resolved photoemission spectroscopy (ARPES) spectra of such systems is suggested. Joint relaxation of strongly coupled fields—a field of correlated electrons and phonon field—after photoemission is studied to clarify the nature of characteristic features observed in the high-energy part of the ARPES spectra of cuprate superconductors. Such relaxation occurs in systems with strong predominantly long-range electron–phonon interaction at sufficiently high carrier concentration due to the coexistence of autolocalized and delocalized carriers. A simple method to calculate analytically a high-energy part of the ARPES spectrum arising is proposed. It takes advantage of using the coherent states basis for the phonon field in the polaron and bipolaron states. The approach suggested yields all the high-energy spectral features like broad Gaussian band and regions of ‘vertical dispersion’ being in good quantitative agreement with the experiments on cuprates at any doping with both types of carriers. Demonstrated coexistence of autolocalized and delocalized carriers in superconducting cuprates changes the idea about their ground state above the superconducting transition temperature that is important for understanding transport and magnetic properties. High density of large-radius autolocalized carriers revealed may be a key to the explanation of charge ordering in doped cuprates.

Keywords: polaron, cuprate high-temperature superconductors, angle-resolved photoemission spectroscopy (ARPES), bipolaron, electron–phonon interaction, correlated electrons

(Some figures may appear in colour only in the online journal)

1. Introduction

Thirty years of theoretical modeling of cuprates demonstrating high-temperature superconductivity by separating one ‘main’ interaction failed to explain their properties. Obviously this points out that simultaneous interplay of charge, spin and lattice degrees of freedom is crucial. Recently such complex approach was successfully applied to quasi-1D cuprates: experimental results on resonant inelastic x-ray scattering turned out to be in complete accordance with the theoretical

description based on simultaneous consideration of electron relaxation and electron–phonon interaction (EPI) [1].

Here we consider photoemission from a system modeling cuprate high-temperature superconductors taking into account joint relaxation of two fields—a field of correlated electrons and phonon field—coupled by strong long-range EPI. The choice of the model is mainly due to the results of the ARPES experiments on superconducting cuprates, which have made a significant breakthrough in recent years. Broad bands of Gaussian shape [2–5] and ‘vertical dispersion’ patterns [6–10]

were observed in ARPES spectra of cuprates universally on both sides of cuprates' phase diagram (in hole- and electron-doped systems) as well as in undoped parent compounds [11]. Progress in qualitative theoretical description of broad bands in ARPES spectra of cuprates was achieved due to consideration of joint influence of electron correlations (in the frames of t - J model) and strong Holstein (short-range) EPI resulting in small polaron (SP) formation [12, 13]. However, 'vertical dispersion' patterns do not appear in this approach. Consideration in the frames of Hubbard model or other models without strong EPI [7, 9, 10] yielded the low-energy part of the ARPES band in good agreement with the experiments at essential doping but did not yield the broad band of Gaussian shape observed experimentally well below the Fermi energy at low doping [2–5, 11].

Simultaneous presence of broad Gaussian bands and 'vertical dispersion' emerges in a model with strong long-range (Frohlich) EPI [14] favoring large polarons (LP), whose radius is larger than the lattice constant [15]. The main distinction of systems with strong long-range EPI at high carrier concentration is coexistence of autolocalized and delocalized carriers [14] due to large size of the autolocalized state (AS). Such coexistence influences strongly the properties of a system. In particular, the electron subsystem can participate in the system relaxation after photoemission. The relaxation pathways are different at photoemission from AS and from delocalized state (DS), with different energy cost. Since according to Pauli exclusion rule ASs and DSs occupy different regions in the momentum space [14] their coexistence displays itself in ARPES spectrum as presence of 'vertical dispersion' universally observed in superconducting cuprates [6–10]. Reconstruction of the phonon vacuum during relaxation when initial or final carrier state or both these states are autolocalized results in broad ARPES bands of Gaussian shape [16]. Therefore below we consider photoemission at arbitrary carrier concentration in a system with strong long-range EPI generalizing the relaxational approach [13, 16–20] to higher doping case.

However, to obtain the ARPES spectrum features being in quantitative agreement with the experiments on cuprates simultaneous taking into account carriers correlations and strong Frohlich EPI is necessary. Therefore here we develop a relaxational approach based on joint results of Hubbard or t - J model with the model of strong long-range EPI. As is shown below the dispersion of correlated carriers can be taken into account in the effective mass approximation at considering autolocalization of carriers with momentums near extrema of the Hubbard bands. The approach under consideration allows simple *analytical* calculation of high-energy part (HEP) of the ARPES spectrum according to Fermi Golden rule. The suggested method takes advantages of using the coherent states basis for description of the phonon field state in AS [14, 16, 19, 21] in the following way. The phonon vacuum reconstruction during the system relaxation after photoemission from AS (or at formation of AS by a photohole) is accompanied with radiation of different number of phonons in different acts with Poissonian distribution of the probability; the average number of phonons is determined by a change of the phonon vacuum

energy [16, 19]. The energy conservation equation rigidly ties the number of radiated phonons with the binding energy of photoelectron.

Only two system characteristics are necessary for the calculation: dispersion of the correlated carriers and effective dielectric constant $1/\varepsilon^* = 1/\varepsilon_\infty - 1/\varepsilon_0$ characterizing the strength of the long-range EPI [22]. The former can be taken either from theoretical models (for example, t - J model [23] or Hubbard model [9]) or extracted from ARPES spectrum. Indeed, below we show that in highly doped cuprates in certain area of the momentum space the ARPES band dispersion follows the 'bare' (without EPI) carrier dispersion; for undoped or slightly doped with holes cuprates this was demonstrated earlier [12]. The latter, in principle, can be calculated using static and high-frequency dielectric constants. However, some renormalization of the EPI strength due to its interplay with carrier correlations caused by their strong Coulomb interaction can occur [24]. Besides, the value of ε_∞ is determined from optical spectra ambiguously: supposing the mid-infrared band has a contribution from EPI, ε_∞ should be taken from $\text{Re}\varepsilon(\omega)$ spectrum above the mid-infrared band frequency. The value obtained (about 3 [25]) differs from one taken above the phonon frequencies but below the mid-infrared band frequency. Therefore here we use effective dielectric constant deduced from ARPES spectrum of undoped or low-doped cuprates.

Predicted energetic and momentum position and line-width of the features in the HEP of ARPES spectrum of cuprates are in good *quantitative* agreement with the experiments at any doping with both types of carriers [2–11]. In particular, the momentum position of a high-energy anomaly (HEA) and the double scale of energy of HEA in electron doped cuprates [9, 10] with respect to that in hole doped ones [6–8] receive natural explanation. Thus, combination of two models—Hubbard or t - J model with the model of strong Frohlich EPI—generates new approach that results in significantly improved agreement of calculated HEP of ARPES spectrum of cuprates with experiments. The approach suggested modifies our notion about the ground state of cuprates (in the normal state) by demonstrating coexistence of autolocalized and delocalized carriers. In future works this finding may help fitting better temperature and doping dependence of other characteristics of cuprates (e.g. transport, optical and magnetic). The results obtained allow us also making some notes on such long discussed problems as appearance/disappearance of HEA in some cuts in different BZs at photon energy change [26–28] (sometimes considered as confirmation of matrix-elements nature of HEA) and charge ordering in doped cuprates [29–34].

The article is organized as following. First we discuss the methods that allow considering high carrier concentrations in a system with simultaneous presence of strong electron correlations and strong long-range EPI and develop analytical method to calculate the HEP of ARPES spectrum from such systems. Then it is applied to systems doped with electrons and holes to any doping level including undoped system. The calculated spectra are compared with the experimental ARPES spectra of cuprates from undoped up to overdoped

with electrons or holes, good quantitative agreement is demonstrated. Then we briefly discuss the implications of the results obtained.

2. Methods

The complexity of the system under consideration demands applying a non-trivial combination of several models and methods and some their development to reach the goal of the study. We start from the results of the Hubbard model [9] or t - J model [23] for the correlated carriers dispersion in the lower and upper Hubbard bands and consider autolocalization of carriers with the momentums near the Hubbard bands extremes. Variational method is used to calculate the binding energy of the AS and the energy of the deformed phonon vacuum in dilute and high-density systems; in the average energy functional the electronic correlations are taken into account in the effective mass approximation [22]. The vector of the system state is written in adiabatic approximation with using coherent states representation for the phonon field [21]. Applying this representation makes possible analytical calculation of the HEP of the ARPES spectrum according to Fermi Golden rule [16].

The ground state of the system at varying carrier density (in the normal state) is determined by the distribution function for systems with strong Frohlich EPI constructed with Gibbs method [14] and modified here to take into account compressibility of the large bipolaron liquid. Its main feature is possible coexistence of autolocalized and delocalized carriers occupying different regions in the momentum space. As a result photoemission from these different regions of the momentum space has different energetic cost. It is calculated according to the energy conservation equations including the change in the electron and phonon fields energy due to post-photoemission relaxation.

2.1. A model of system with strong electron correlations and strong Frohlich EPI and methods of its analysis

As strongly correlated systems are under intensive discussion for many years let us begin with brief introduction concerning systems with LP. The usage of terms LP and SP in literature is ambiguous: sometimes LP is understood as weak-coupling polaron (delocalized carrier state) and SP as strong-coupling (localized) one. We use more traditional terminology in which the LP is AS of a charge carrier, whose size is larger than the unit cell [22, 35] whereas the SP is localized inside the unit cell. LP can be studied in continual approximation, it can move coherently with sufficiently high mobility as distinct from SP [15, 36]. Although the LP's ground state properties were calculated in late forties [22, 35], experimental observation of LP was complicated due to necessity of doped strongly polarizable ionic crystals of high quality to prevent carrier trapping by defects. The first success in experimental test of the polaron theory was achieved by Feynman and Thornber. They found a system needed—oxide covering of cold cathode devices—and managed to calculate giant losses of carriers

energy in them (with a path integral method) [37] consistent with experiments. Similar result was later obtained in LP model considering Cherenkov radiation of the polarization waves [21].

Doped cuprates are systems where polarons can occur, and broad bands observed in their optical absorption and ARPES spectra [2–5, 11, 15, 38, 39] were discussed in this context. However, the initial LP theory considered polarization field as classical one with the energy equal to its average value (doubled binding energy [22]). As a result the width of the predicted band in optical absorption spectrum caused by LP photodissociation was determined only by electron wave function in LP [15]. Taking into account quantum fluctuations of the phonon field in LP with using quantum coherent states basis [21] allowed analytical calculation of broad Gaussian bands in optical conductivity [19] and ARPES spectra [16], resulting from generation of different number of phonons in different acts of the LP photodecay with Poissonian probability. The optical conductivity band caused by carrier photoexcitation into excited state in the polaronic polarization potential well was calculated with other methods [20, 40]. Bands caused by LP photodecay and photoexcitation along with Drude contribution form optical conductivity spectrum of cuprates (below the charge-transfer band) close to ones observed experimentally [38, 39], their partial spectral weight at different doping are to be calculated. Finally, the region where LP and large bipolaron exist is limited in temperature [14, 41]. Together with their enhanced effective mass this can cause an upturn in the underdoped cuprates' resistivity (in normal state) at lowering temperature [42] like one observed experimentally [23].

Now let us discuss a model of a system with strong carrier correlations and strong Frohlich EPI and its applicability to cuprates demonstrating high-temperature superconductivity. LP is formed by a carrier at strong long-range EPI, if the 'bare'-carrier bandwidth W (i.e. without EPI) exceeds the carrier average kinetic energy in the polaron [15, 36] which is equal to the polaron binding energy E_p [22]. In the opposite case as well as at the dominance of short-range EPI the SP is formed [15, 36]. Let us check which of these conditions is satisfied in the cuprates doped with holes where carriers are considered to appear in the so called lower Hubbard band (LHB) and in cuprates doped with electrons introduced into upper Hubbard band (UHB).

For the LHB case both W and E_p values can be extracted from the dispersion of ARPES band at zero or low doping with holes. Indeed, it follows the 'bare' carrier dispersion [12, 13] shifted deeper due to hole polaron formation, and the binding energy $\varepsilon_{\max}$ in its maximum is about $2E_p$ for SP [36] and $3E_p$ [22], or, more precisely, about $3.2E_p$ for LP [16]. The polaron binding energy E_p in the UHB is obtained similarly from the position of the ARPES band maximum at low doping with electrons [4, 5]. The UHB bandwidth W can be deduced from the ARPES spectrum at high doping with electrons in which the dispersion of the ARPES band crossing the Fermi level also follows 'bare' carrier dispersion as is shown below. Since in cuprates $W \approx 0.4 \div 0.5$ eV for the LHB [11] and is larger for the UHB [9], whereas $|\varepsilon_{\max}| \approx 0.42$ eV [4, 5, 11]

in both LHB and UHB, E_p does not exceed W , and LP model is appropriate for both bands. This conclusion is confirmed by comparison of the predictions obtained below with the experiments.

Carrier tunneling between nodes inside the LP polarization potential well is much quicker (adiabatical) than the ions motion, as the ratio of the carrier average kinetic energy in LP $E_{\text{kin}} = E_p \approx \varepsilon_{\text{max}}/3.2$ [16] to the average phonon energy [13] shows. Therefore at considering the effects of strong EPI the electron correlations can be taken into account in the effective mass approximation. The dispersion of correlated carriers can be taken either from theoretical calculations [9, 23] or from experimental ARPES spectra of cuprates [2–11] (excluding the region separated by ‘vertical dispersion’), the more so that they demonstrate good correspondence. The fact that dispersion of the polaronic ARPES band (observed in experimental spectra at low or zero doping) follows the ‘bare’ carrier dispersion (shifted downward by $|\varepsilon_{\text{max}}|$) was revealed in [12]. Below we show that the dispersion of the ARPES band crossing the Fermi level at high doping (both with electrons or holes) in systems with strong Frohlich EPI also follows the ‘bare’ (without EPI) carrier dispersion, or quasi-particle dispersion, in terms of [9] (please, see¹ for refining the use of terms LHB and UHB dispersion below).

Therefore here we use the UHB dispersion in nodal direction ($k_x = k_y$) which is taken from the lower-energy band in experimental ARPES spectrum of electron-doped cuprate (near optimal doping) [9] in nodal direction (in eV):

$$E_{\text{UHB}}^{\text{nodal}}(\mathbf{k}) = -0.5(\cos(k_x a) + \cos(k_y a)) + 1, \quad (1)$$

where a is the in-plane lattice constant. Analogously, the LHB dispersion in nodal direction is taken from the dispersion of the lower-energy band in experimental ARPES spectrum of highly overdoped Bi2201 in nodal direction [6]. The experimental ARPES spectrum of undoped cuprates (shifted upward by $|\varepsilon_{\text{max}}|$) follows approximately the same dispersion in nodal direction [11]. In the region $k_x = k_y \leq 0.5\pi/a$ considered below it can be approximated as following (in eV):

$$E_{\text{LHB}}^{\text{nodal}}(\mathbf{k}) = -0.125(\cos(2k_x a) + \cos(2k_y a)) - 0.25. \quad (2)$$

It is also consistent with theoretical t - J model dispersion [23]. Zero energy in (1) and in (2) is in the bottom of UHB and top of LHB, respectively, that corresponds to very low doping with electrons (equation (1)) or holes (equation (2)). Dispersions of UHB and LHB modified by strong Frohlich EPI as it is discussed below are shown by figures 1(a) and (b).

¹ Theoretical study of the Hubbard model spectrum at high doping [9] reported presence of dispersing quasi-particle band and incoherent LHB or more precisely oxygen valence band [9]. Here we deal only with the former, quasi-particle band. The filled states from the incoherent band obviously have large effective mass even without EPI, so that at taking into account strong EPI they (if charged) form SPs as described in the beginning of section 2. As a consequence they do not participate in the phase space division between large ASs and DSs (due to Pauli exclusion rule) studied in the article as a reason of HEA. Therefore we do not consider them below and for short denote the dispersion of the quasi-particle bands [9] as UHB and LHB dispersion. Nevertheless, the filled states from the incoherent band [9] can be the source of features observed in ARPES experiments at high binding energy which are not restricted in the momentum space.

Strong Frohlich EPI results in autolocalization of carriers with the momentums near extremes of the correlated carrier bands determined by equations (1) and (2). The binding energies of the large-radius ASs of charge carriers are calculated with variational method, first used at classical description of the polarization field [22], then developed applying quantum coherent states representation for the phonon field in the polaron [21] and bipolaron [14]. Besides carrier dispersion, the only values we need to calculate the polaron and bipolaron binding energy are effective dielectric constant ε^* and high-frequency dielectric constant ε_∞ . As discussed in the Introduction ε^* should be deduced from experimental ARPES spectrum using the relation between the binding energy ε_{max} in the ARPES band maximum and the polaron binding energy E_p : $E_p \cong \varepsilon_{\text{max}}/3.2$ [16, 22] and the well-known Pekar result [22, 35] (the second equation is in eV):

$$E_p = 0.0544m^*e^4/\hbar^2\varepsilon^{*2} = 1.47(\varepsilon^*)^{-2}m^*/m_e. \quad (3)$$

Equation (3) is obtained for the isotropic case but contains the carrier effective mass which is anisotropic in the considered cuprates. Nevertheless, we can use equation (3) with ‘effective’ isotropic m^* to determine ‘effective’ ε^* without introducing essential mistake into further calculations as the value of ε^* enters in them only in expressions for energies (bipolaron binding energy and the phonon field energy in the bipolaron, equation (4) below) which contain ε^* and m^* similarly to equation (3). Therefore we use below the carrier effective mass $m^* \approx m_e$ near the bands extremes according to equations (1) and (2). Then equation (3) yields $(\varepsilon^*)^{-1} \approx 0.3$ for both electron- and hole-doped cuprates and we use this value in further calculations.

For the ARPES spectrum calculation we will also need the value of the energy stored in the deformed phonon vacuum in the polaron $E_{\text{pvd}}^{\text{pol}} = 2E_p$ [22] and in the bipolaron $E_{\text{pvd}}^{\text{bip}}$ and the binding energy of the bipolaron. The binding energy per carrier E_b and the energy of the deformed phonon vacuum $E_{\text{pvd}}^{\text{bip}}$ in the one-center bipolaron with taking into account carriers correlation [14] are determined by the same medium parameters m^* , ε^* and ε_∞ . They can be calculated approximately with the mistake lower than 5% according to the following expressions obtained with the variational method (in eV):

$$E_b = -1.44(c^2 + \varepsilon^*cd + 1/80)m^*/m_e, \\ E_{\text{pvd}}^{\text{bip}} = 8 * 1.44(c/\varepsilon^* + d - 1/140)m^*/m_e, \quad (4)$$

where $c = 2/\varepsilon^* - 1/\varepsilon_\infty$, $d = 2/(21\varepsilon_\infty^2)$. We use below the value of $\varepsilon_\infty = 3$ [25] observed at frequencies above the mid-infrared band in optical spectra as was discussed in the Introduction.

2.2. Methods of studying the system with strong Frohlich EPI at high carrier concentration

Here we study the HEP of the ARPES spectrum which does not change at the superconducting transition. Therefore below we limit ourselves to consideration of the normal state of the system. The ground state of the system at high carrier density

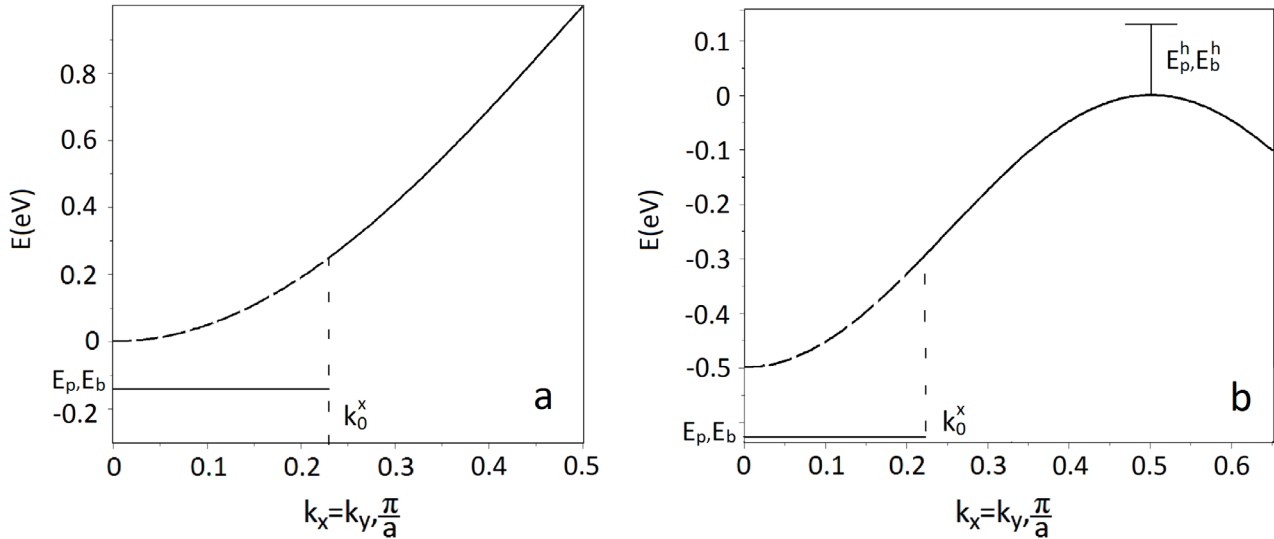


Figure 1. (a) and (b) Upper and lower Hubbard band dispersion in nodal direction (expressed by equations (1) and (2)) modified by strong Frohlich EPI, respectively; k_0^x is $\mathbf{k}_0$ projection on x or y axis; E_p, E_b are binding energies of electron polaron and bipolaron, respectively, E_p^h, E_b^h stand for binding energy of hole polaron and bipolaron.

is described with the carrier distribution function in systems with strong Frohlich EPI obtained with Gibbs method [14]. The carrier momentum in LP and in large bipolaron has large uncertainty $\hbar k_0$ tied with the (bi)polaron volume V_0 (the LP and bipolaron have close volumes [14]) by the uncertainty relation:

$$\frac{4}{3}\pi(\hbar k_0)^3 V_0 = (2\pi\hbar)^3. \quad (5)$$

k_0 is ordinarily about $\pi/(4a) \div \pi/(3a)$, therefore the momentum is not suitable to characterize the carrier AS. Spontaneous breaking of the translational symmetry in a system with strong EPI results in the ground state degeneration as polarons can be located in different regions of the crystal [43]. Thus, ASs can be characterized by their center position, and different carrier states take place when the distance between the AS centers is larger than their diameter or when the momentums of the ASs with the same location differ to the value higher than k_0 . The latter is, however, impossible due to limitation of the AS velocity by the group velocity of phonons [21]. Therefore there exists a maximum ASs density [14, 44, 45] $n_0 = 2/V_0$, where V_0 is the bipolaron volume at high carrier concentration.

At carrier concentration $n > n_0$ delocalized carriers are present in the system even at zero temperature. But Pauli exclusion rule limits the momentums of the delocalized carriers: they cannot be lower than $\hbar k_0$ if the carrier density is higher than $n'_0 = 2/V'_0$ where V'_0 is the bipolaron volume in the dilute system. The DS unavailable at $n > n'_0$ are shown with dashed line on figures 1(a) and (b). Two different values of the critical carrier density— n_0 and n'_0 —appear due to the fact that at high carrier density the bipolaron volume depends essentially on the carrier density. For example, at $(\varepsilon^*)^{-1} = 0.3$, $m^* = m_e$ and $\varepsilon_\infty = 3$ [25] the single bipolaron radius $R'_{bip} \approx 13.5 \text{ \AA}$ [14] (we estimate it as the radius of the region, that contains 0.9 part of the polarization charge) whereas at high carrier density $n \geq n_0$ the calculation yields

$R_{bip} \approx 6.5 \div 7 \text{ \AA}$ [46]. For clarity, let us estimate doping levels p'_0 and p_0 corresponding to the carrier densities n'_0 and n_0 in a system with $(\varepsilon^*)^{-1} = 0.3$, $\varepsilon_\infty = 3$, $m^* = m_e$. Two slightly different estimates are obtained in the models with square grid of bipolarons and at their most close packing: $p'_0 = a^2 * 2/(2R'_{bip})^2 \approx 0.04$, $p_0 = a^2 * 2/(2R_{bip})^2 \approx 0.176$ and $p'_0 = a^2 * 2/\pi(R'_{bip})^2 \approx 0.05$, $p_0 = a^2 * 2/\pi(R_{bip})^2 \approx 0.225$, respectively.

The value of the bipolaron radius at high carrier concentration is obtained by minimizing the total system energy. First the energy of ‘compressed’ bipolaron as function of its radius is calculated by means of conditional (at fixed bipolaron radius) minimization of the average value of bipolaron Hamiltonian [14] with respect to parameters of the electron wave function in the bipolaron. The dependence of the bipolaron energy on its radius is used to calculate the total energy of the system (comprising the maximal number of bipolarons at given their radius and the rest carriers in DS with momentums higher than k_0) as function of the bipolaron radius at given carrier concentration. The minimum of this function yields the equilibrium bipolaron radius as function of the carrier concentration. At $(\varepsilon^*)^{-1} = 0.3$, $m^* = m_e$ and $\varepsilon_\infty = 3$ and high carrier density $R_{bip} \approx 6.5 \div 7 \text{ \AA}$ [46]. According to equation (5) the corresponding k_0 value is about $0.35\text{--}0.37 \text{ \AA}^{-1}$. When the momentum is along the nodal direction the corresponding value of $\mathbf{k}_0$ projection on k_x or k_y axis is $0.2 \div 0.27 \text{ \AA}^{-1}$, where different possible values of the momentum projection onto the normal to the crystal surface are taken into account.

The binding energy per carrier in large bipolaron calculated with taking into account carrier correlation is close to that in LP, therefore at low doping the ground state of the system with strong Frohlich EPI can be either polarons or bipolarons, or they coexist [14]. At $n'_0 < n < n_0$ it is a system of bipolarons at zero temperature or bipolarons and delocalized carriers at non-zero temperature and carrier density essentially higher than n'_0 , at $n > n_0$ it is a system of bipolarons and delocalized

carriers. Below we will denote $n > n_0$ the case when delocalized carriers are present in the system, however, if the temperature is non-zero, it occurs at slightly lower carrier density.

Screening of EPI by delocalized carriers in the systems under consideration is prohibited by Pauli exclusion rule. Indeed, at $n > n_0$ delocalized carriers are present in the system (or at $n > n'_0$ at non-zero temperature). However, to form a screening charge the squared modulus of a DS wave function should be non-zero in a localized area whose size is of the order of the polaron size. Then the carrier momentum should be from the interval $k < k_0$, that is prohibited by Pauli exclusion rule at $n > n'_0$ ($n'_0 < n_0$).

Ground state of a single hole at strong EPI is a hole polaron. However, at strong EPI completely filled band contains about $n_0/2$ electron bipolarons. Then where in the coordinate space do the hole ASs locate? Obviously the hole is not localized inside the electron bipolaron as it is not profitable energetically. The more so that between electron bipolarons there are areas of negative polarization charge attracting the holes whereas inside the electron bipolarons the positive polarization charge repulsing the holes is concentrated. The coexistence of the hole ASs with the electron ones allows understanding the peculiar dependence of the charge ordering period in hole-doped cuprates on the carrier density. The charge ordering period does not depend on the holes concentration in wide interval of the latter [30, 32] since at hole density lower than n_0 it is dictated by the electron AS size. It is worth noting that due to large radius of the carrier ASs in cuprates the electron bipolarons and the hole ASs contain phonon vacuum deformation in mainly different harmonics with the wave vectors around Γ and $(\pi/2, \pi/2)$ points of the first Brillouin zone, respectively.

2.3. The method of calculating the HEP of ARPES spectrum of the systems under study

The simplest analytical calculation of the band in ARPES spectrum caused by photodissociation of the LP [16] is based on Fermi golden rule

$$W_{if} = \frac{2\pi}{\hbar} \left| \langle f | \hat{H}_{int} | i \rangle \right|^2 \delta(E_i - E_f),$$

$$\langle f | \hat{H}_{int} | i \rangle \propto \int d\mathbf{r} \exp(-i\mathbf{k}\mathbf{r}) \hat{H}_{int} \psi(\mathbf{r}) \prod_{\mathbf{q}} \langle \nu_{\mathbf{q}} | d_{\mathbf{q}} \rangle, \quad (6)$$

with the vector of the system initial state written in the adiabatic approximation as a product of the electron wave function $\psi(\mathbf{r})$ in the polaron and a vector of the phonon field state $\prod_{\mathbf{q}} |d_{\mathbf{q}}\rangle$ in the coherent states representation. Parameters $d_{\mathbf{q}}$ of the phonon vacuum deformation in the $\mathbf{q}$ th harmonics due to EPI are simply expressed through the corresponding Fourier-transform of the squared electronic wave function [21], $\nu_{\mathbf{q}}$ is a number of phonons in the $\mathbf{q}$ th harmonics radiated at decay of the coherent state after the photoelectron escape [16, 19].

The energy conservation law (δ -function argument in equation (6)) relates the photoelectron energy and the number ν of radiated phonons: $E_p + \hbar\Omega = E_{kin} + \Phi + \nu\hbar\omega$, where E_p , $\hbar\Omega$, E_{kin} and Φ are the polaron binding energy, photon

energy, photoelectron energy and work function, respectively. Ordinarily in ARPES studies three latter values are replaced by binding energy $\varepsilon = E_{kin} - (\hbar\Omega - \Phi)$ so that the energy conservation law has the form $E_p = \varepsilon + \nu\hbar\omega$. It (and, consequently, the argument of δ -function in equation (6)) contains only two variables: ε and ν . Then in neglecting the phonon dispersion a probability $A(\mathbf{k}, \varepsilon)$ to catch a photoelectron with the momentum $\mathbf{k}$ and binding energy ε results from summing the probabilities (6) over all the cases with equal total number ν of radiated phonons [16, 19]:

$$A(\mathbf{k}, \varepsilon) \propto |\psi_{\mathbf{k}}|^2 P_{\nu(\varepsilon)}, \quad (7)$$

$$P_{\nu(\varepsilon)} = \sum_{\{\nu_{\mathbf{q}}\}=\nu} \prod_{\mathbf{q}} |\langle \nu_{\mathbf{q}} | d_{\mathbf{q}} \rangle|^2 = \frac{\bar{\nu}^{\nu(\varepsilon)-1}}{(\nu(\varepsilon)-1)!} e^{-\bar{\nu}}, \quad (8)$$

$$\nu(\varepsilon) = (E_p - \varepsilon)/\hbar\omega, \bar{\nu} = \sum_{\mathbf{q}} |d_{\mathbf{q}}|^2 = \Delta E_{pvd}/\hbar\omega, \quad (9)$$

where $\psi_{\mathbf{k}}$ is Fourier-transform of the electron wave function in the initial state, $\bar{\nu}$ is the average number of radiated phonons and ΔE_{pvd} is the change in the phonon vacuum energy due to deformation caused by EPI. For the photoemission from the polaron state $\Delta E_{pvd} = E_{pvd}^{pol}$, the average polarization field energy in the polaron state due to the phonon vacuum deformation. According to known Pekar results [22] $E_{pvd}^{pol} = 2E_p$, and E_p is determined by equation (3). Due to phonon dispersion the energy distribution curves $A(\mathbf{k} = \text{const}, \varepsilon)$ are envelopes of points obtained according to equations (7)–(9). Such a calculation yields broad bands of Gaussian shape [16] located in the momentum region $k < k_0$ where $\psi_{\mathbf{k}}$ is noticeable, which are in good agreement with the experimental results [4, 5].

The method (6)–(9) is applicable to calculate the photoemission from the large bipolaron state [14], with the first equation (9) replaced by

$$E_b = E_p + \varepsilon + \nu\hbar\omega \quad (10)$$

and

$$\Delta E_{pvd} = E_{pvd}^{bip} - E_{pvd}^{pol}, \quad (11)$$

where ΔE_{pvd} is the difference between the energies of the phonon vacuum deformation in the bipolaron and in the polaron. However, if delocalized carriers appear in the system (at zero temperature this occurs at carrier concentration higher than the double maximum density of bipolarons, $n > n_0$, this case will be supposed below) they have energy higher than that of degenerated [43] bipolaron states, and electron subsystem starts to participate in the post-photoemission relaxation. It turns out that the method (6)–(9) can be applied to this case too. Let us discuss first the pathways of the resulting relaxation and then the corresponding energy conservation equation and change of the phonon filed energy used to replace the first of equations (9) and for substitution into the second of them, respectively, at $n > n_0$.

At high carrier density $n > n_0$, if photoelectron originates from bipolaron state so that its in-plane momentum $k < k_0$ (supposing the band minimum is in $k = 0$ point), the

intermediate state with one carrier remained in bipolaronic polarization potential well (it does not change during electron phototransition according to Frank–Condon principle) in presence of delocalized carriers relaxes due to EPI in other way than in their absence. Namely, relaxation results in restoration of the bipolaron and emptying the state from the Fermi surface (or hole appearance at the Fermi surface). If photoelectron comes from DS, i.e. its in-plane momentum $k > k_0$, the system relaxation is absolutely different and simple, without changing the phonon vacuum. A ‘boundary’ $k = k_0$ between the two types of the system relaxation with different relaxation energy displays itself in ARPES spectrum as ‘vertical dispersion’. It should be noted that in systems where SPs are formed there is not a coexistence of localized and delocalized carriers and correspondingly there is not ‘vertical dispersion’ pattern in the ARPES spectrum.

Even more interesting relaxation occurs in undoped or hole-doped systems. The hole left after the photoelectron escape relaxes to the Fermi surface near the LHB maximum and forms AS due to strong EPI. Such final state occurs for doping lower than the ASs maximum concentration n_0 . At higher doping the final state is delocalized hole at the Fermi surface (similarly to the electron doping case with $n > n_0$). If the photoelectron originates from the electron AS the electron bipolaron is restored in the final state. Each relaxation way is discussed in details below at calculating its display in the ARPES spectrum.

To calculate the HEP of the ARPES spectrum of systems with high carrier concentration the described above method is generalized on the base of developing the relaxational approach [13, 16–20]. The energy conservation equation describing the relaxation result includes (apart from the change of the phonon field energy caused by formation or decay of the phonon vacuum deformation) the energies of two electronic quasiparticles which change their states in the final state in comparison with the initial one: the photoelectron and the electron with the maximum energy in the initial state (at the Fermi surface). The energy of the latter (varied with doping) is transferred to the phonon field in the final state. The matrix element of phototransition determines the position of the spectral weight in the momentum space whereas the energy conservation equation determines its position with respect to the binding energy axis.

At $n > n_0$, $k < k_0$ the initial state is electronic bipolaron. Therefore $|\psi_{\mathbf{k}}|^2$ in (7) results in the same localization of the bipolaron band in the momentum space as at $n < n_0$. However, the other relaxation way changes its position in the binding energies. Similarly to $n < n_0$ case after photoelectron escape the bipolaron polarization cloud begins to decay down to the polaron one with radiation of phonons, their average number is $\bar{\nu}_1 = \Delta E'_{\text{pvd}}/\hbar\omega$, provided the process is completed. The stroke designates that at $n > n_0$ the difference between the polarization field energy in the bipolaron and in the polaron is slightly larger than in the single bipolaron case due to ASs contraction in the system ground state at high carrier density.

At $n > n_0$ the relaxation due to strong EPI restores the bipolaron after photoelectron with $k < k_0$ escape. First a transition of a delocalized carrier towards the minimum energy

state (or relaxation of the photohole) occurs. This process and decay of the bipolaron polarization ‘coat’ into polaron one are both multiphonon processes and occur during the characteristic phonon times. Therefore there will be a distribution of probabilities for the degree of the latter process completeness. As a result some decrease of $\bar{\nu}_1$ will take place. However, this decrease is compensated to some degree by increase of $\Delta E'_{\text{pvd}}$ due to bipolarons contraction at high carrier density. Therefore for rough estimate of energetic position of the spectral weight at $k < k_0$ we suppose $\bar{\nu}_1 \approx \Delta E'_{\text{pvd}}/\hbar\omega$, where $\Delta E'_{\text{pvd}}$ is defined by equation (11). The possible mistake is not essential in comparison with extremely large width of the ARPES band at $k < k_0$.

Relaxation empties the Fermi-surface state, its excess energy and momentum are transferred to the phonon field with creating the appropriate average number $\bar{\nu}' = \hbar^2 k_F^2/(2m^* \hbar\omega)$ of phonons. The bipolaron restoration from the polaron is also accompanied with the radiation of phonons, their average number $\bar{\nu}_2 \approx \bar{\nu}_1$ since the initial and final phonon vacuums for the cases of decay and restoration are simply interchanged. According to Gaussian distribution property the average number of radiated phonons

$$\bar{\nu} = \bar{\nu}_1 + \bar{\nu}_2 + \bar{\nu}' \cong 2\bar{\nu}_1 + \bar{\nu}' \cong 2(E_{\text{pvd}}^{\text{bip}} - E_{\text{pvd}}^{\text{pol}})/\hbar\omega + \hbar^2 k_F^2/(2m^* \hbar\omega). \quad (12)$$

The energy conservation equation at $n > n_0$, $k < k_0$ has the form

$$E_b + 0 = E_b + \varepsilon + \nu\hbar\omega. \quad (13)$$

Using it to express $\nu(\varepsilon)$ for substitution into P_ν (8) simultaneously with $\bar{\nu}$ value (12) we calculate the spectrum at $k < k_0$ according to equation (7).

At $n > n_0$ and the photoelectron momentum $k > k_0$ the relaxation does not affect the phonon vacuum. In this case two variants of the calculation yield the same result presented by figure 2(c): traditional one where the final state is a hole with the in-plane momentum k and one including relaxation of the hole to its minimum energy state on the Fermi surface. The energy conservation equation for the second calculation is $0 + E(k) = E(k) + \varepsilon + \nu\hbar\omega$, where $E(k) = E_{\text{UHB}}(k)$ is determined by equation (1) with zero of energy at the Fermi surface (changing with the doping), $\nu = (0 - E(k))/\hbar\omega$ is the number of radiated phonons. This yields $\varepsilon(k) = E(k)$, i.e. spectral weight in the region $k > k_0$ follows ‘bare’ (i.e. without EPI) electron dispersion crossing the Fermi level.

Thus, the correlated electrons dispersion can be extracted from the lower-energy part of the experimental ARPES spectrum of highly doped samples as we have done to obtain equation (1). Broadening of the lower-energy part of the ARPES band is not related with changing the phonon vacuum. As we do not calculate here such-type broadening, to visualize the lower-energy part of the ARPES spectrum in figure 2(c) (the region $k > k_0$) we use the dispersion (1) as bare band dispersion and real and imaginary parts of the self-energy extracted from the experimental spectrum according to [47]. It should be noted that the broadening of the $k > k_0$ part of ARPES spectrum (as it is not associated with change in the phonon

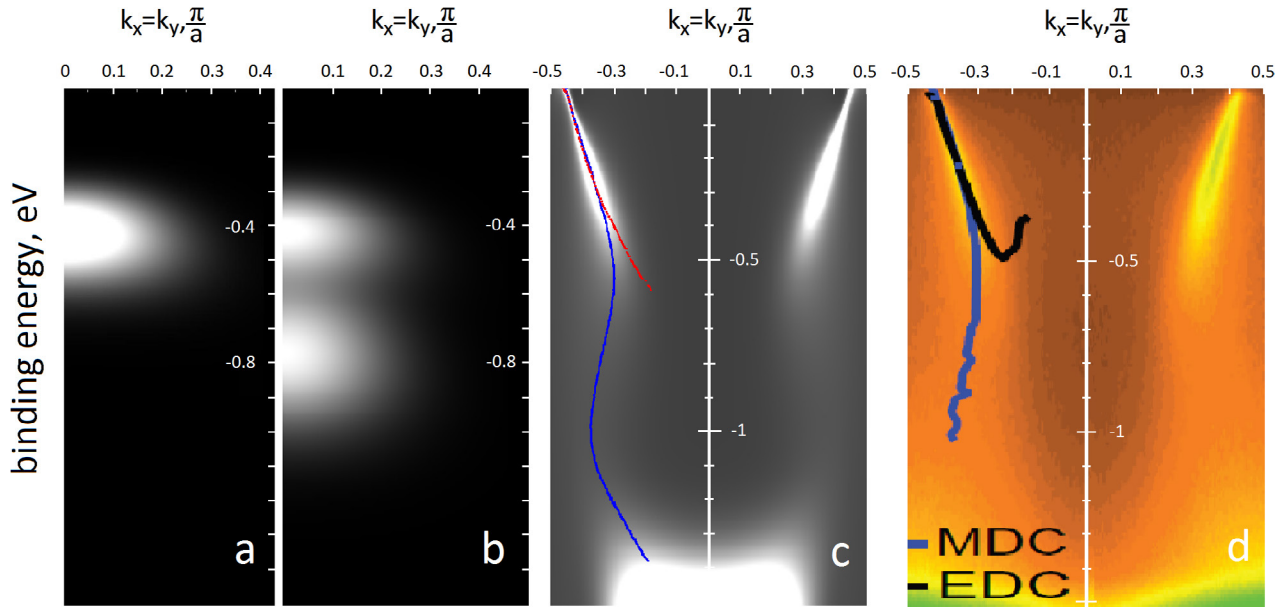


Figure 2. (a)–(c) Calculated HEP of the ARPES spectrum of electron-doped cuprate at low doping, at doping corresponding to coexistence of polarons and bipolarons and at electron concentration $n > n_0$, respectively, red (shorter) and blue (longer) lines on panel c are dispersions inferred from the calculated EDCs and MDCs, respectively; (d) experimental spectrum along the nodal cut of $\text{Nd}_{1.83}\text{Ce}_{0.17}\text{CuO}_4$ [9].

vacuum) as well as ‘kinks’ in it are successfully calculated in the weak-coupling EPI model [48]. Thus, the supposition often made in the literature in relation with the observed (in ARPES) dichotomy of the momentum space that EPI strength in cuprates depends on the carrier momentum is in good agreement with the picture of strong Frohlich EPI.

Now let us consider the boundary $k = k_0$. Obviously, existence of a sharp boundary in the momentum space between the AS and DSs is a consequence of the too rough model. In reality the wave function of a carrier in the AS vanishes exponentially both in the coordinate and momentum spaces, i.e. the autolocalized carrier can have the momentum k slightly higher than k_0 , but with small probability. Accordingly, the HEP of the ARPES band slightly extends into the momentum region $k > k_0$, but with low intensity. Then delocalized carriers with momentums about k_0 cannot penetrate into the very inner region of the ASs where the momentum of autolocalized carriers reaches the values about k_0 . Thus, the volume which can be occupied by delocalized carriers decreases with their momentum from the whole volume of the system for carriers with $k \gg k_0$ to zero for carriers with k slightly lower than k_0 . This change in the number of available DSs with k can be approximated with corresponding Gaussian function of the momentum, as we have done calculating figure 2(c).

3. Results

3.1. Electron-doped systems at increasing doping

The ARPES spectrum of system of single polarons (electron doped system at low carrier concentration) calculated according to equations (7)–(9) is demonstrated by figure 2(a) in the form of intensity map. It is in good agreement with the experimental ARPES spectrum of electron-doped cuprates at low doping [4, 5] (of course, as we use the experimental

band maximum to determine ε^* the comparison at low doping makes sense for the width and shape of the calculated band in the energy and momentum space).

Equations (6)–(9) were first generalized to describe photoemission from large bipolaron when its binding energy and wave function are calculated with taking into account carriers correlation inside polarization potential well [14]. However, the exact expression obtained was cumbersome. To simplify it one can calculate the electronic matrix element neglecting carriers correlation in the bipolaron (electronic wave function in the bipolaron is approximated as a product of two polaron ones), but still taking it into account calculating bipolaron binding energy. Then $|\psi_{\mathbf{k}}|^2$ in (7) coincides with the polaron case and the difference of bipolaronic $A(\mathbf{k}, \varepsilon)$ from the polaronic one is in the energy conservation equation (10) which determines $\nu(\varepsilon)$ to be substituted in equation (8) and in average number of radiated phonons (11).

The average number of phonons (11) radiated at the bipolaron polarization cloud decay down to polaron one is larger than that in the case of photoemission from the polaron state, therefore the bipolaronic band in ARPES spectrum calculated according to equations (7), (8), (10) and (11) lies essentially deeper than the polaronic band. Figure 2(b) shows them both that corresponds to carrier concentration characteristic for coexistence of polarons and bipolarons (occurring in a narrow interval of the carrier concentration [14]). Similar smearing of the spectral weight over a wide area of binding energy values has been managed to observe in a cuprate severely underdoped with holes ($T_c = 5$ K) [49], it is represented in figure 3(d).

At $n > n_0$ as it was discussed above the system relaxation after photoemission involves electron subsystem, at $k < k_0$ the initial state is bipolaron whereas at $k > k_0$ the initial state is delocalized carrier. Different relaxation after photoemission from these different initial states results in two distinct parts in the calculated ARPES spectrum. The calculated according

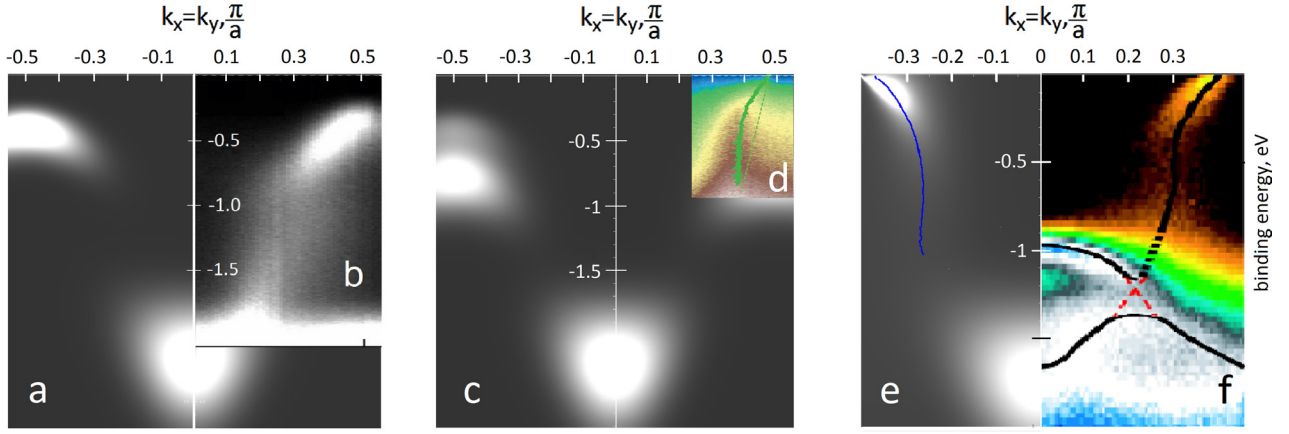


Figure 3. (a) and (b) Calculated and experimental (along nodal cut of $\text{Ca}_2\text{CuO}_2\text{Cl}_2$) [11] ARPES spectra of undoped parent compound, respectively, in the same scale; (c) calculated ARPES spectrum at hole doping corresponding to two possible final states: hole polaron and bipolaron; (d) experimental spectrum along nodal cut of highly underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ($T_c = 5$ K) [49] in the same scale as panel (c); (e) calculated ARPES spectrum at hole concentration $n > n_0$, blue line is the dispersion inferred from the calculated MDCs; (f) experimental ARPES spectrum of highly overdoped $\text{Bi}2201$ in nodal direction [6] in the same scale as panel (e).

to described above method with using equations (1), (7), (8), (12) and (13) HEP of the ARPES spectrum in nodal direction for the case $n > n_0$ is demonstrated by figure 2(c). It is in good *quantitative* agreement with the experimental spectrum of slightly overdoped with electrons $\text{Nd}_{1.83}\text{Ce}_{0.17}\text{CuO}_4$ [9] shown in figure 2(d) in the same scale. The left half of both figures 2(c) and (d) shows also dispersions inferred from the calculated energy distribution curves (EDC, intensity as function of the binding energy at fixed momentum) and momentum distribution curves (MDC, intensity as function of the momentum at fixed binding energy) and dispersions obtained from experimental EDCs and MDCs, respectively.

The dispersions of the calculated EDC and MDC are in good agreement with those of experimental ones. It may seem that such a comparison is meaningless, since the dispersion and the self-energy for the calculation of the lower-energy part of the spectrum are extracted from the experimental spectrum. However, the dispersion of the calculated MDCs in the energy region where EDC-dispersion vanishes is the result of joint influence of the lower- and higher-energy parts of the spectrum, the latter is calculated according to equations (7), (8), (12) and (13). Remarkably, the back-bending MDCs dispersion observed in experiments [7, 9] arises naturally in the suggested approach, confirming the supposition made in literature that MDC-inferred dispersion does not obligatory represent the quasiparticle dispersion, unlike the EDC-inferred dispersion which follows equation (1).

3.2. Undoped and hole-doped systems

To calculate photoemission from undoped and hole-doped cuprates we use the correlated carriers dispersion (2) modified by strong Frohlich EPI (figure 1(b)). Similarly to the electron-doped systems with $n > n_0$ there are two types of the initial state: autolocalized (bipolaron) for photoelectrons with the momentums $k < k_0$ and delocalized for $k > k_0$. The relaxation process is also similar to high ($n > n_0$) electron doping case with the only difference in the final state. The system states with delocalized hole or electron polaron which are created at

the photoemission from the states $k > k_0$ and $k < k_0$, respectively, are not its stationary state. Strong EPI results in hole polaron formation (with preceding hole transition into a state near the electron band maximum). Deformation of the phonon vacuum during the hole polaron formation is accompanied by multiple phonon radiation. As a result the whole ARPES spectrum both in $k < k_0$ and $k > k_0$ regions is shifted deeper in the binding energy by about $3E_p^h$ (where E_p^h is the hole polaron binding energy) and in the region $k > k_0$ the ARPES band is broad analogously to polaronic band in electron-doped system.

At $k < k_0$ the energy conservation equation and the average number of radiated phonons are

$$E_b + 0 = E_b + \varepsilon + E_p^h + \nu\hbar\omega, \bar{\nu} \cong (2\Delta E_{\text{pvd}} + 2E_p^h - E_{\text{LHB}}(k))/\hbar\omega, \quad (14)$$

where $E_{\text{LHB}}(k)$ is determined by equation (2) with zero energy at the Fermi level (changing with doping). Thus, in undoped cuprates the ARPES band in the region $k < k_0$ lies deeper than in the electron doped ones at high doping by the energy released at hole polaron formation and preceding hole relaxation towards its minimum energy state. At $k > k_0$

$$E_{\text{LHB}}(k) + 0 = \varepsilon + E_{\text{LHB}}(k) + E_p^h + \nu\hbar\omega, \bar{\nu} \cong (2E_p^h - E_{\text{LHB}}(k))/\hbar\omega. \quad (15)$$

The ARPES band dispersion at $k > k_0$ obtained as average of the first equation (15) has the form

$$\bar{\varepsilon}(k) = -E_p^h - \bar{\nu}\hbar\omega = -3E_p^h + E_{\text{LHB}}(k), \quad (16)$$

i.e. it follows the ‘bare’ carrier dispersion shifted deeper by $3E_p^h$. This is similar to the SP model [12, 13] result but in SP model this dispersion is predicted for the whole k axis whereas in the LP model the ‘vertical dispersion’ emerges at $k \approx k_0$ due to different system relaxation ways in different k regions. Figures 3(a) and (b) demonstrate calculated and experimental [11] ARPES spectrum from undoped cuprate in one and the same scale. They are in good quantitative agreement except low calculated spectral weight in the intermediate energy region which may be caused by the fact that the calculation does not take into account the band broadening due electron correlations.

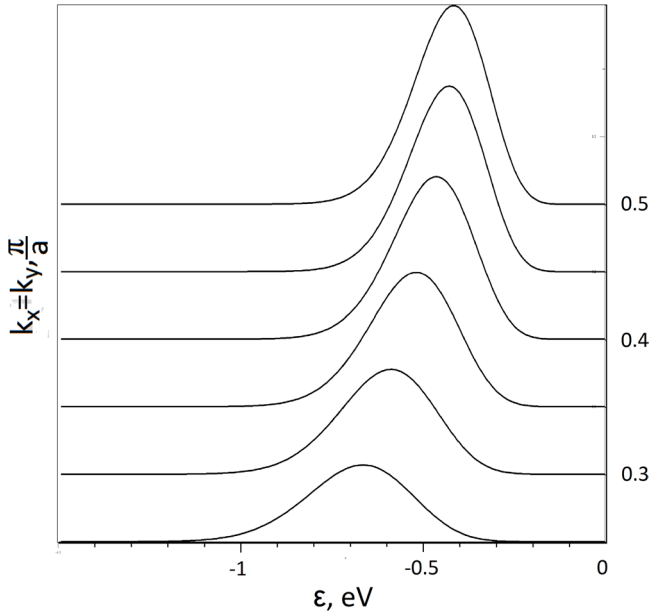


Figure 4. EDCs calculated for undoped system with the nodal dispersion (2) and $(\varepsilon^*)^{-1} = 0.3$.

Figure 4 represents the calculated EDCs obtained from the spectral function shown by figure 3(a) at constant values of $k \geq k_0$. The shape of the calculated band is Gaussian, its width (determined by $\bar{\nu}$ value (15)) increases at moving away from the band extremum in accordance with the experiment [3]. In experimental ARPES spectra this effect was highlighted in [3] where the ratio of the binding energy in the EDC maximum to the EDC halfwidth (width at the half height) was obtained approximately constant and equal to 1.8. In the calculated EDCs presented in figure 4 this ratio depends on the phonon energy and for the average phonon energy 0.04 eV it is 1.8 at $k_x = k_y = 0.5\pi/a$ (in the ARPES band maximum) and 1.9 at $k_x = k_y = 0.25\pi/a$ (where the ‘vertical dispersion’ emerges) in good agreement with the experiment [3].

When at increasing doping the hole polaron states become filled the final photohole state is no longer a hole polaron but a hole bipolaron. The energy conservation equations read

$$E_b + E_p^h = E_b + \varepsilon + E_b^h + \nu\hbar\omega, \\ \bar{\nu} \cong (2\Delta E_{pvd} + \Delta E_{pvd}^h - E_{LHB}(k))/\hbar\omega, k < k_0, \quad (17)$$

$$E_{LHB}(k) + E_p^h = \varepsilon + E_{LHB}(k) + E_b^h + \nu\hbar\omega, \\ \bar{\nu} \cong (\Delta E_{pvd}^h - E_{LHB}(k))/\hbar\omega, k > k_0, \quad (18)$$

where $E_{LHB}(k)$ is determined by equation (2) with zero of energy at the Fermi level. Similarly to the electron doping system the spectral weight in the case of bipolaron final state is shifted deeper in comparison with the polaron one since the energy $\Delta E_{pvd}^h + E_b^h - E_p^h$ (where h denotes hole ASs) is larger than $3E_p^h$. Its location in the momentum space does not change.

If due to close binding energy per carrier in the polaron and bipolaron state the photohole can appear in some concentration interval with comparative probability in both these states

the spectral weight will be smeared over more wide area of energies. Interestingly, that this stage of the spectral weight transferring deeper upon doping the authors of [49] managed to observe on the very underdoped cuprate with $T_c = 5$ K. Calculated and experimental ARPES spectra demonstrating smearing the spectral weight over very wide area of energies are shown on figures 3(c) and (d), respectively, in one and the same scale. Smaller intensity of the polaronic band in comparison with the bipolaronic one in experimental spectrum (figure 3(d)) likely means that the doping level of the pattern corresponds to almost filled hole polaron states, the same situation is chosen for calculated spectrum shown on figure 3(c).

At hole concentration $n > n_0$ the final state of the photohole is DS at the Fermi surface. Since it does not induce the phonon vacuum deformation the whole HEP of the ARPES spectrum is lifted (in comparison with the $n < n_0$ case) along the energy axis by the value $3.2E_p^h + |E(\pi/2a) - E(k_F)|$. Energy conservation equations have the form

$$E_b + 0 = E_b + \varepsilon + \nu\hbar\omega, \bar{\nu} \cong (2\Delta E_{pvd} - E_{LHB}(k))/\hbar\omega, k < k_0, \quad (19)$$

$$E_{LHB}(k) + 0 = \varepsilon + E_{LHB}(k) + \nu\hbar\omega, \nu = -E_{LHB}(k)/\hbar\omega, k > k_0. \quad (20)$$

As equation (20) show, the ARPES band dispersion at $k > k_0$ follows ‘bare’ (without EPI) carrier dispersion similarly to the electron doping case at $n > n_0$. Analogously to the electron doping case, as we do not calculate here the band broadening not related with changing the phonon vacuum, to visualize the lower-energy part of the spectrum at high doping on figure 3(e) we use the dispersion (2) and real and imaginary parts of the self-energy extracted from the experimental spectrum. Generally, the situation at high doping with holes is very close to that at high doping with electrons, the difference is in the dispersion: equation (2) is used instead of equation (1).

The calculated HEP of the ARPES spectrum in nodal direction at hole doping $n > n_0$ is presented in figure 3(e). Figure 3(f) shows the experimental spectrum of highly overdoped Bi2201 in nodal direction [6] in the same scale. The comparison is somewhat complicated due to crossing of several bands in the experimental spectrum [6], but similar dispersion was also reported in [7], the similar dispersion and band crossing were observed in [8]. Taking into account that the LHB dispersion can be slightly different in $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ studied in [3, 11] and in Bi2201 and Pb-Bi2212 studied in [6–8] whereas we use one and the same equation (2) to model all these systems, the calculated HEPs of the ARPES spectra are in good agreement with experimental results. The MDC-inferred dispersion shown in figure 3(e) with blue line demonstrates back-bending in agreement with experiments [7] whereas the quasi-particle dispersion (equation (2)) displays itself in the EDC dispersion (not shown).

4. Discussion

Comparing the results obtained with the previous models’ results, one can note two main points in which the Hubbard model with taking into account strong Frohlich EPI

demonstrates advance in comparison with the Hubbard-only model (without EPI) [9]. First, the matrix elements vanishing near Γ point are no longer needed to obtain HEA. Indeed, vanishing spectral weight near Γ point in the Hubbard-only approach [9] was obtained after multiplying the calculated spectral function by the matrix elements obtained in a special way [47]. Namely, the matrix elements are fitted from the experimental spectrum in supposition that it is formed by continuous spectral function of the Lorentzian form (representing the LDA band broadened due to many-body interactions) multiplied by the matrix elements [47]. Such method obligatory yields the matrix elements vanishing near Γ point if the experimental spectral weight vanishes there. However, several strong arguments against using vanishing matrix elements near Γ -point to obtain HEA are given in [11].

Second, the lowest-lying band observed in experimental ARPES spectra of cuprates at low doping with electrons [4, 5] and zero or low doping with holes [2, 3, 11] is the broad band of Gaussian shape [3]. The models without strong EPI do not yield the broad bands of Gaussian shape. The calculations with taking into account strong EPI according to equations (7)–(9) in the electron-doped case and according to equations (7) and (8) in the undoped or low-doped with holes cases result in broad Gaussian bands (shown on figures 2(a) and (b) for electron doping and on figures 3(a), (c) and 4 for zero and low doping with holes) whose position and width are in good agreement with the experiments [2–5, 11].

Moreover, there is quantitative agreement between the calculated and experimental relation of the Gaussian band widths and the position of its maximum. The measured experimentally ratio of the binding energy in the EDC maximum to the EDC halfwidth (width at the half height) was approximately independent on k and equal to 1.8 [3]. In the calculated EDCs presented in figure 4 this ratio depends on the phonon energy and for the average phonon energy 0.04 eV it is 1.8 at $k_x = k_y = 0.5\pi/a$ (in the ARPES band maximum) and 1.9 at $k_x = k_y = 0.25\pi/a$ (where the ‘vertical dispersion’ emerges) in good agreement with the experiment [3].

The experimentally observed double scale of HEA energy in electron-doped cuprates with respect to that in hole-doped cuprates arises naturally in the frames of model under consideration. Indeed, in this approach the momentum k_0 of the break in the delocalized carrier dispersion at essential doping due to Pauli exclusion rule is invariant in electron and hole-doped cuprates due to close size of the bipolarons in them. But the corresponding energy $E(k_0)$ (the energy of HEA) is determined by the LHB and UHB dispersion which are different as it is illustrated for nodal direction by equations (1) and (2) and figures 1(a) and (b).

The back-bending MDC-inferred dispersion observed in experiments [7, 9] also naturally appears in the suggested approach (figures 2(c) and 3(e) for systems with high doping with electrons and holes, respectively) as a result of superposition of spectral weight from two contributions present mainly in different but partially interpenetrating areas of the momentum space (the lower-energy part of ARPES spectrum caused by delocalized carriers and present mainly in the region of momentums $k > k_0$, and the HEP of the ARPES spectrum

caused by autolocalized carriers and located basically in the region $k < k_0$). Thus, the results of the present consideration confirm the supposition made in literature that MDC-inferred dispersion does not obligatory represent the quasiparticle dispersion, unlike the EDC-inferred dispersion which follows equation (1) in the electron-doped case (figure 2(c)) and equation (2) in the hole-doped case (not shown).

Comparing the results of the present approach with those of Hubbard model with taking into account strong short-range (Holstein) EPI [12, 13] one can note that both approaches result in the same dispersion of the hole-polaronic band in ARPES spectrum which follows the ‘bare’ band dispersion. The broad Gaussian bands arise at low doping in both approaches (with strong long-range and short-range EPI), too. However, the ‘vertical dispersion’ patterns appear in the ARPES spectrum only in the model with strong long-range EPI, as the large radius ASs formed in such systems can coexist with delocalized carriers at high carrier density.

The present approach allows also understanding several experimental findings being unclear in some earlier models. First, manganites are also characterized by broad bands in ARPES spectrum but do not demonstrate ‘vertical dispersion’ patterns. This fact was sometimes considered as proof that strong EPI cannot be the reason of this feature [50]. As we have shown strong long-range EPI causes ‘vertical dispersion’ at sufficiently high doping due to coexistence of autolocalized and delocalized carriers, whereas strong short-range EPI resulting in SP formation does not. However both types of EPI result in broad Gaussian bands in ARPES spectrum [12, 13, 16] provided the EPI is strong. Thus, the mentioned difference in ARPES spectra of cuprates and manganites is likely caused by different dominating type of EPI, long-range in the former case and short-range in the latter, albeit EPI is strong in both cases.

Second, some notes concerning the observation conditions of HEA can be made. They are important because changes in HEA manifestation in some cuts in different BZs at change of the photon energy observed experimentally [26–28] posed a question whether it is not an effect of the matrix elements only. We show that the break of the delocalized carrier dispersion in essentially doped systems due to strong EPI exists in restricted region of the k space: at $|\mathbf{k}| < k_0$. Thus, two of three cuts studied in [26] are out of the region where HEA occurs and the third cut $k_x = 3\pi/(8a)$ is just on the boundary $|\mathbf{k}| = k_0$. Likely, this is the reason of unordinary behavior of the spectrum [26] taken from this cut at the photon energy change. Earlier the similar behavior was observed in the second and third BZs [27, 28]. However, if the HEA nature is related with the strong long-range EPI breaking the translational symmetry due to ASs formation then the appearance of HEA in the second and third BZs in the same form as in the first one is questionable.

Finally, demonstrated broken by strong EPI translational symmetry in cuprates allows also discussion of charge ordering observed in doped cuprates [29–34] in terms of the large-radius ASs formation. Indeed, the estimated radius of the bipolaron at their maximum density in cuprates is $R_{\text{bip}} \approx 6.5 \div 7 \text{ \AA}$ [46]. This value is in good agreement both with the k_0 value marking the HEA position in the momentum space [6–11] and with

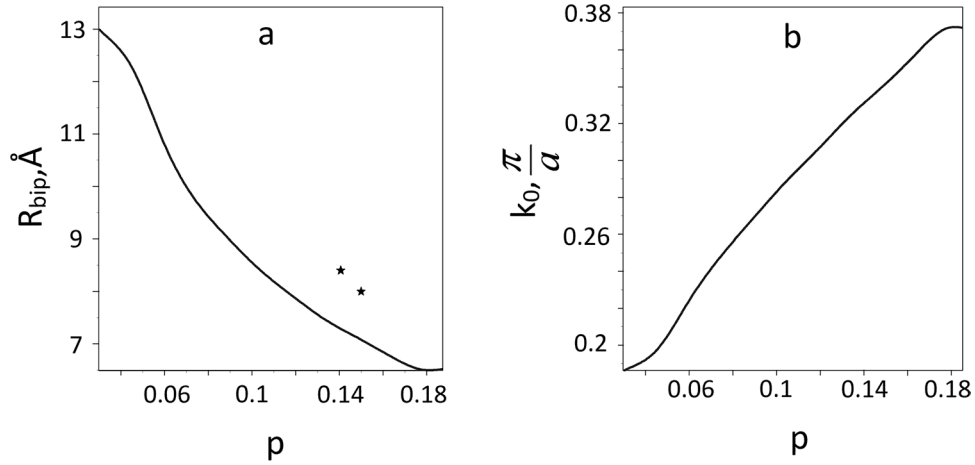


Figure 5. (a) and (b) Doping dependence of the bipolaron radius (determining charge ordering period) and k_0 value, respectively, in electron-doped systems. The system parameters used are $(\epsilon^*)^{-1} = 0.3$, $\epsilon_\infty = 3$, $m^* = m_e$. The doping level p is calculated in a model with square grid of bipolarons. Stars on panel (a) demonstrate a half of the charge ordering period experimentally measured on electron-doped cuprate at two doping levels [34].

measured experimentally period of charge ordering ($3.3 \div 4a$) [29–34] in cuprates. Moreover, the experimental finding that the charge ordering period in hole-doped cuprates is independent on the carrier density in wide interval of it [30–33] is quite understandable in the present approach where the hole ASs are located in-between the electron ASs, whose density is maximal. Thus, the electron ASs location dictates the hole ASs one, and charge ordering period does not depend on the holes density while it is lower than n_0 . For the electron-doped systems the present approach predicts change of the charge ordering period with doping: the ASs size becomes smaller with increasing doping (an example of such dependence is presented in figure 5(a) [46]), rendering the wave vector of the charge ordering larger. Similar behavior was observed in the electron doped cuprate experimentally [34], as it is illustrated with stars in figure 5(a). Of course, charge ordering in a system of ASs is a subject for separate consideration as we used here the simplest quasi-isotropic model whereas to obtain the square grid of ordered charges or stripes [29–34] one should take into account the system anisotropy.

Doping dependence of the bipolaron radius in electron-doped systems results in doping dependence of k_0 value (the boundary momentum separating autolocalized and delocalized carriers in the momentum space and representing the ‘vertical dispersion’ momentum in the ARPES spectra) in them, illustrated by figure 5(b). It should be noted that $\mathbf{k}_0$ projection k_0^x onto x or y axis used as the momentum axis scale in ARPES spectra (figures 2 and 3) and shown on figure 1 is distributed in the limits $k_0/\sqrt{3} \div k_0/\sqrt{2}$ due to uncertainty of z -projection of $\mathbf{k}_0$. In undoped and hole-doped systems the value of k_0 determining the position of ‘vertical dispersion’ in the ARPES spectrum does not depend on the doping level as electron density in these systems is higher than n_0 .

5. Conclusion

In summary, we suggest a new approach to studying the normal state of cuprate superconductors which allows simultaneous

taking into account high concentration of correlated charge carriers and strong long-range EPI. We show that in such systems photoemission is accompanied by joint relaxation of strongly coupled fields—a field of correlated electrons and phonon field—due to coexistence of autolocalized and delocalized carriers. Sharing of the momentum space between two these types of carrier states according to Pauli exclusion rule together with different relaxation pathways for autolocalized and delocalized initial (final) states result in fragmentation of the band in ARPES spectrum into two parts in different regions of the wave vector space. Predicted theoretically value of the ‘vertical dispersion’ momentum position (the k_0 value) is in good agreement with the experiments on cuprates with both types of doping.

Besides realizing the possible reason of fragmentation of the HEP in ARPES spectrum of cuprates we suggest a method to calculate analytically the HEP of ARPES spectrum in systems with strong carrier correlations and strong long-range EPI at arbitrary carrier concentration. It takes advantages of applying coherent states basis for the phonon field state description and the fact that only two electrons change their state together with the phonon field as the result of photoemission and subsequent system relaxation. The calculated position, width and shape of the bands in the HEP of ARPES spectrum of cuprates are consistent with the experiments at any level of doping with both types of carriers.

The agreement of the calculated HEP of the ARPES spectrum with the experiments on cuprates confirms presence of ASs of the large radius and coexistence of autolocalized and delocalized carriers in them. This result changes the idea about the ground state of the superconducting cuprates in the normal state. It may be useful for understanding temperature and doping dependence of transport and magnetic properties of cuprates [23]. Suggested approach may be also effective in theoretical modeling the evolution of the optical conductivity spectra of cuprates with doping and temperature [38, 39]. Finally, it allows to describe charge ordering in systems with strong long-range EPI at high carrier density, the predicted value of the charge ordering period and its concentrational

behavior (independence on the carrier density in wide interval in hole-doped systems and change with doping in electron-doped system) are in agreement with the experimental data.

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