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A simple model for the checkerboard pattern of modulated hole densities in underdoped cuprates

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Abstract. A simple model is proposed as a possible explanation for the checkerboard pattern of modulations in the hole density observed in recent tunneling experiments on underdoped cuprates. Two assumptions are made; first, an enhanced hole density near the acceptor dopants and secondly short range correlations in the positions of these dopants caused by their electrostatic and anisotropic elastic interactions. Together these can lead to a structure factor in qualitative agreement with experiment.

PACS. 74.25.Jb Electronic structure – 74.72.-h Cuprate superconductors (high-T_c and insulating parent compounds)

Recent studies which measured the tunneling conductance into underdoped cuprates found an unexpected spatial modulation at higher bias voltages with a 4 × 4 periodicity. The most comprehensive studies [1] have been made on the $Ca_{2-x}Na_xCuO_2Cl_2$ compounds where a good surface can be prepared deep into the underdoped region. These scanning tunneling microscopy (STM) studies have led to interpretations of the data as signals of superlattices of single holes or hole pairs [2–6]. Since these materials are Mott insulators doped randomly with acceptors it is not clear if superlattice ordering can persist in the presence of the random electric fields of the acceptor ions. In this letter we will explore an alternative explanation that starts from the assumption of enhanced hole densities localized in the vicinity of acceptors and examines the consequences of short range correlations in the positions of the acceptors, e.g. the Na-ions in $Ca_{2-x}Na_xCuO_2Cl_2$.

Our first assumption of enhanced hole densities close to the acceptors is consistent with the standard behavior of doped semiconductors. These undergo a Mott transition when the dopant concentration is reduced and hole wavefunctions become localized around acceptors. Here we make a simple assumption of an enhanced hole density and explore the consequences. Then we simulate the positions of the Na ions under the combined influence of inter-ion Coulomb repulsion and elastic forces, using a Monte Carlo algorithm. Together these two effects determine the form of the modulations in the hole density and so the STM structure factor. As we shall see the calculated structure factor shows qualitative agreement with the STM experiments.

The outermost layer of the crystal consists of interpenetrating square lattices of Cl and $(Ca_{1-x/2}Na_{x/2})$ ions. The STM tip will couple most effectively to the outermost wavefunctions of the surface layer which should be the $3p_z$ orbitals of the Cl ions. The final state of the injected electron (+ve bias) is in the hybridized antibonding band of $3d_{x^2-y^2}$ orbitals centered on Cu sites and the $2p_{x,y}$ O-orbitals. Since the Cl ions lie directly above the Cu-sites there is no matrix element to tunnel into the state with $d_{x^2-y^2}$-symmetry centered of the Cu-site directly below. There will be however a finite matrix element to couple into the 4 nearest neighbor Cu sites generated through the overlap of the $3p_z$–Cl and $2p_{x,y}$–O states on neighboring Cl–O pairs. This situation is similar to the case of tunneling through an outermost BiO layer in BSSCO compounds which has important consequences for the form factor of substitutional impurities in the CuO_2-plane as was pointed out by Martin, Balatsky and Zaanen [7]. Returning to the case of $Ca_{2-x}Na_xCuO_2Cl_2$ the injection of an electron (or hole) through the $3p_z$ orbital of a Cl ion neighboring a Na-ion leads to finite amplitudes for injection into the 4 Cu sites which are nearest neighbors (nn) to the Cu underneath the Cl ion. Of these 4 sites 2 are nn sites of the Na ion and 2 are n.n.n. sites (see Fig. 1).

The next question that arises concerns the tunneling probability is whether the tunneling amplitude into these 4 sites should be added coherently or incoherently. Consider the case of positive bias or electron injection from the tip. Clearly the Coulomb attraction between a hole and the Na-acceptor leads to increased occupation of states centered on the 4 n.n. Cu sites and to a lesser extent on the 8 n.n.n. sites. Explicit form of hole wavefunction around a dopant was numerically calculated for the case of Li^+ dopant in ladder system [8]. Similar

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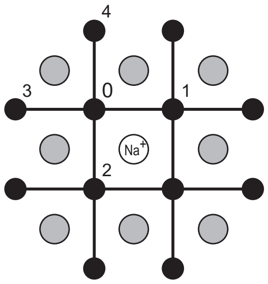

Fig. 1. Projected view of crystal structure of $Ca_{2-x}Na_xCuO_2Cl_2$ around a doped Na^+ ion. Black and gray balls represent Cu and Ca ions. An electron (or hole) injected to Cl $2p_z$-orbital above site 0 is transferred to one of the Cu sites 1–4.

calculation is possible for this case but this is beyond the scope of this paper. This in turn enhances the hole density on these sites. When we calculate the electron spectral function $A(\mathbf{x},\omega)$ that determines the conductance

$$A(\mathbf{x},\omega)=\sum_{\alpha,\sigma}\left|\left\langle\Psi_\alpha^{0\mathrm{h}}\right|\sum_{\mathbf{x}'}h(\mathbf{x},\mathbf{x}')c^\dagger_{\mathbf{x}',\sigma}\left|\Psi_0^{1\mathrm{h}}\right\rangle\right|^2 \times\delta\left(\omega-E_\alpha^{0\mathrm{h}}+E_0^{1\mathrm{h}}\right). \quad (1)$$

We are interested in an energy range stretching up to several hundred meV. Therefore the relevant matrix element will be an incoherent superpositions of tunneling processes into the 4 Cu sites which are n.n. to the Cl site. Here $|\Psi_0^{1\mathrm{h}}\rangle$ is the one-hole groundstate with energy $E_0^{1\mathrm{h}}$ and $|\Psi_\alpha^{0\mathrm{h}}\rangle$ are eigenstates of a stoichiometric system, and $h(\mathbf{x},\mathbf{x}')$ is the amplitude of the hole wave function centered on the Cu-site $\mathbf{x}'$, at the Cl site at position, $\mathbf{x}$. For calculating the spectral function, we average the energy over the allowed range for the injected electron so that we may treat the sum over the one-hole eigenstates as a sum over a complete set of states. Under these incoherent conditions the $\langle A(\mathbf{x},\omega)\rangle_{\omega-\mathrm{av}}\sim\sum_{\mathbf{x}'}h^2(\mathbf{x},\mathbf{x}')n(\mathbf{x}')$ where $n(\mathbf{x}')$ is the hole density at $\mathbf{x}'$. In the neighborhood of a Na^+-ion the Coulomb attraction leads to an enhanced hole density on the inner ring of 4 nearest neighbor sites and to a lesser extent on the outer ring of 8 next nearest neighbor sites (see Fig. 1). This in turn will lead to an enhanced tunneling signal on the inner ring (f_1) and the outer ring (f_2). The Fourier transformed cluster form factor is then

$$f(\mathbf{k})=4f_1\cos\frac{k_x}{2}\cos\frac{k_y}{2} +4f_2\left[\cos\frac{3k_x}{2}\cos\frac{k_y}{2}+\cos\frac{k_x}{2}\cos\frac{3k_y}{2}\right]. \quad (2)$$

We do not attempt detailed calculations of f_1, and f_2 but treat them as phenomenological parameters. Note in this letter we will concentrate on electron injection only and so our calculations are for $A(\bar{\omega})$, where $\bar{\omega}$ is an energy of order 100 (meV) or above, where the experimental data do not depend on the energy so much. Strictly speaking this should be determined from experiment by normalizing the spectra in the − ve bias (hole injection) side rather than the +ve bias side chosen by Hanaguri et al. [1]. An explicit proposal along these lines has been made by Randeria and coworkers [9].

The STM signal intensity is given by

$$I(\mathbf{x})=\int d^2\mathbf{R}\,\rho(\mathbf{R})f(\mathbf{x}-\mathbf{R}) \quad (3)$$

where $f(\mathbf{x}-\mathbf{R})$ is the intensity of a cluster at Cl site $\mathbf{x}$ around a Na-acceptor at $\mathbf{R}$ and it is the inverse Fourier transform of $f(\mathbf{k})$ in equation (2). $\rho(\mathbf{R})$ is the distribution function for the Na-ions. At the relevant densities $x\approx0.1$ the concentration of the Na ions in the outermost $Ca_{2-x}Na_xCuO_2Cl_2$ layer is not in the dilute regime so that it is possible that the Na–Na interactions lead to short range order in the positions of the Na acceptors. To estimate the effects of these interactions on the distribution we simulated the annealing of an initial random 2-dimensional distributions through inter-ion interactions using a Monte Carlo algorithm. The interaction has a Coulomb repulsion component and an elastic component

$$V(\mathbf{R})=\frac{1}{R}-A\frac{\cos4\theta}{R^2}. \quad (4)$$

The former is radially symmetric but the second term is in general anisotropic. We estimate it following Eshelby [10] for a 2-dimensional square lattice and we ignore effects associated with the fact that the Na-acceptors lie both above and below the topmost CuO-plane. Here θ is the angle of vector $\mathbf{R}$ measured from the principal axis of the lattice. The intensity $A=2\pi c^2\eta(\lambda+\mu)/(\lambda+2\mu)$ is given by Lamé constants, $\lambda=\lambda_{xx,yy}$ and $\mu=\lambda_{xy,xy}$, and the tetragonal anisotropy, $\eta=\lambda_{xx,xx}-(\lambda+2\mu)$, with c being a constant proportional to local volume change due to impurity doping. This is normalized such that the coefficient of the Coulomb term is set to be unity.

We have investigated stable configuration of doped Na^+-ions with the above interaction potential. For this simulated annealing, we have used a Monte Carlo algorithm. Starting from a random initial configuration, we choose a Na-ion in turn and find a vacant site randomly. We then calculate how much the interaction energy changes, ΔE, if the chosen ion is to move. If $\Delta E<0$, we always move that ion to achieve a stabler configuration, whereas even if $\Delta E>0$, we accept the new configuration with the Boltzmann probability $e^{-\Delta E/T}$ at temperature T. This update process is applied to all the ions in turn, and the procedure is repeated many times to obtain a stable ion configuration. Typically, we have set the temperature $T=0.01$–0.05, and repeated the Monte Carlo procedure 10^3–10^4 steps for each initial configuration. We have checked that obtained results do not depend on these parameters sensitively. In Figure 2 we show typical distributions resulting from Monte Carlo annealing of samples with 10^4 sites and 10^3 ions. Periodic boundary conditions are imposed to both directions and the interaction $V(\mathbf{R})$ is tailored for the boundary conditions by using the mirror charge technique.

Once the annealed configuration is obtained, we calculate the structure factor of Na ions by taking the Fourier transform. Result for 10% doping and $A=1.0$ is plotted in Figure 3a, and the Fourier transform of random initial configurations is also shown in Figure 3b in comparison.

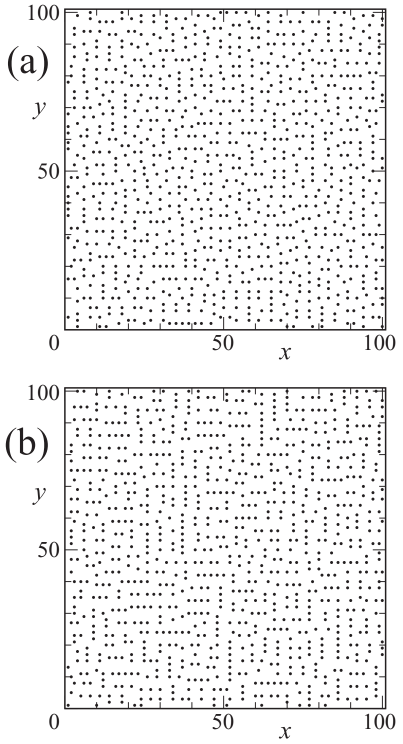

Fig. 2. Typical configuration of Na ion distribution simulated by Monte Carlo calculations. The system size is 100×100 sites and the Na ion density is 0.10. Strength of elastic component of Na–Na interaction is (a) $A = 0.5$, and (b) $A = 1.0$.

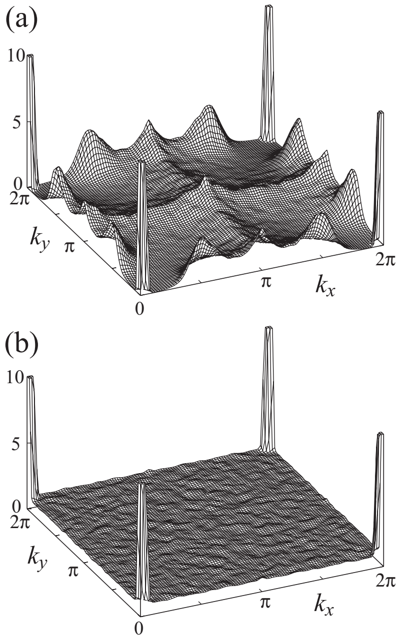

Fig. 3. (a) Fourier transform of Na-ion distribution, $\langle|\rho(\mathbf{k})|^2\rangle$, annealed for the potential $V(\mathbf{R})$ with $A = 1.0$. Ensemble average is taken over 10^2 configurations generated by Monte Carlo calculations and the results are smoothed by convoluting with Gaussian function with width 0.03π. The system size is 100×100 sites and the Na ion density is 0.10. (b) Fourier transform of random initial configurations at the same density before annealing.

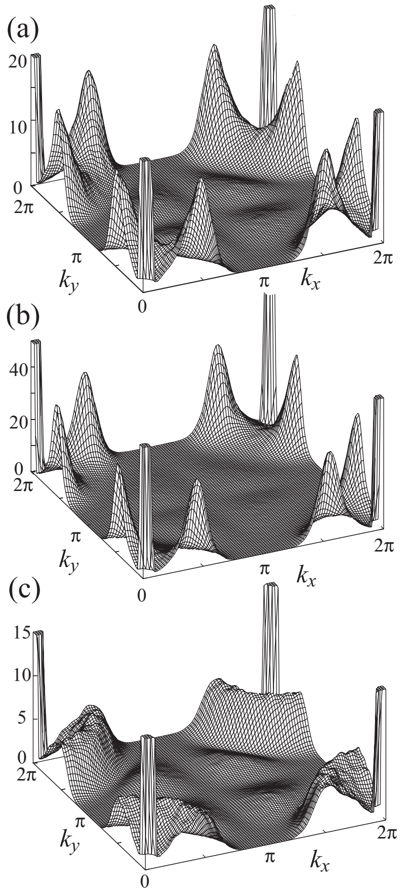

Fig. 4. Structure factor of holes localizing around doped Na ions, $\langle|I(\mathbf{k})|^2\rangle$. Ensemble average is taken over 10^2 configurations generated by Monte Carlo calculations and the results are smoothed by convoluting with Gaussian function with width 0.03π. The system size is 100×100 sites and the Na ion density is 0.10. Coupling constant is (a) $A = 0.5$, (b) $A = 1.0$, and (c) $A = 0.0$.

We have also checked that ensemble average over a larger set of samples (e.g., 10^3) gives essentially the same results. The Na-ion structure factor is averaged over initial configurations, and then smoothed by Gaussian broadening with width 0.03π.

The final structure factor is calculated by multiplying it with the form factor of hole density localizing around Na ions

$$|I(\mathbf{k})|^2 = |\rho(\mathbf{k})|^2\,|f(\mathbf{k})|^2\,. \tag{5}$$

In Figures 4a and 4b, we show representative sets of data for two values of the relative strength of the elastic and Coulomb interactions. The data for the case of pure Coulomb interaction are also shown in Figure 4c in comparison. We see that the main characteristic of the checkerboard pattern observed in the experiments [1], namely peaks at wavevectors $(\pi/2, 0)$ and $(0, \pi/2)$ are nicely reproduced when the anisotropic elastic energy is considered $A > 0$, especially for the case with stronger elastic interactions. The stronger tendency for the Na-ions to line up along the crystal axes in these simulations is reflected in the structure factor by the suppression of weight along the diagonal leading to the peaks being concentrated near the vertical and horizontal axes.

Our calculations show that at least the broad features of the data can be reproduced by our two assumptions, enhanced hole density around the acceptor and short range correlations in the positions of the Na-acceptors. Such short range order necessarily induces a corresponding modulation in the hole distribution. In addition the hole density will be distributed over a cluster of sites around each acceptor. Both effects influence the final structure factor. The STM data contain a lot of more detailed spectroscopic information, e.g. small peaks in the conductance. These features are beyond the scope of the calculations presented here. The STM experiments give us really detailed information on an unprecedented scale but for now at least, we can only analyze the main features. A key point in our model is the enhanced hole density in the vicinity of an acceptor which hopefully can be explicitly tested in experiment.

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