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1995 J. Phys.: Condens. Matter 7 3741

(http://iopscience.iop.org/0953-8984/7/19/008)

J. Phys.: Condens. Matter 7 (1995) 3741–3748. Printed in the UK

Charge fluctuation properties of a 1D p–d model

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Received 16 November 1994, in final form 1 March 1995

Abstract. We study a one-dimensional p–d model with a finite bandwidth under the unrestricted Hartree–Fock approximation. It can be seen that charge fluctuations play an important role in the system, and the finite bandwidth will not qualitatively change the characteristics of the charge fluctuations. The appearance of the phase separation depends on the relative strength of the nearest-neighbour intersite Coulomb repulsion V_{pd}. The finite bandwidth turns the divergent behaviour of χ_{CT} into a sharp peak in the curve of χ_{CT} against ϵ.

1. Introduction

After the discovery of copper-oxide superconducting materials, in recent years much effort has been expended in the study of the normal state properties and the mechanism of superconductivity. One of the most notable features of high-T_c superconducting materials is their unusual normal state behaviour, an understanding of which is important in the search for the mechanism of superconductivity. It is now generally agreed that strong electron–electron correlations play an important role in these materials. A copper-oxide superconductor will be an insulator in a half-filled situation due to an energy gap. One of the interesting questions here is: what is the nature of the gap for the parent insulator? Multi-band models in which both Cu and O orbitals are contained have been stressed by many authors [1–8]. As suggested by Varma and co-workers [2], the effective copper and oxygen levels are close in energy and the nearest-neighbour copper and oxygen Coulomb repulsion is comparable with the hybridization energy. This is regarded as one of the key ingredients for high-T_c superconductivity. It also confirms that the on-site Coulomb repulsion in Cu is the largest energy scale in the problem; the materials are charge-transfer insulators in a half-filled situation. Many authors have paid attention to the charge fluctuations of the system. It has been shown by Littlewood and co-workers [9, 10] that, in the weak-coupling limit, the charge-transfer susceptibility will diverge with the increase of the nearest-neighbour copper and oxygen Coulomb repulsion. Using the large-N expansion method [11, 12], the phase separation of the model is found to occur in the region of the phase diagram surrounding the metal–insulator transition. The problem has also been studied by numerical diagonalization and Monte Carlo simulations [13, 14]. Much useful information has resulted from finite-size studies. Scalettar and co-workers [13] have researched the qualitative behaviour of the spin and charge correlations of the three-band Hubbard model; their results show that attractive interactions exist in both the d-wave and extended s*-wave channels near the antiferromagnetic boundary. Sano and Ono [14] pointed out that in the proximity of the phase boundary towards the phase separation, the superconducting correlation is dominant compared with the charge density wave (CDW) and spin density wave (SDW) correlations.

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The system size is a limit to small-cluster studies, due to the complexity of the structure. Recently, Sudbo and co-workers [15] have obtained an exact solution of the one-dimensional p–d model with zero bandwidth by the transfer matrix technique. They showed that the phase separation will exist in this system and that there will be a sharp peak in the charge transfer susceptibility which responds to the charge transfer instability. Their results also confirmed that the nearest-neighbour Coulomb repulsion cannot be ignored in this kind of strongly correlated electronic system. No exact solution is obtained when the hybridization is introduced.

In order to address this issue, in this work we study a one-dimensional p–d model with a finite bandwidth. The consideration is that, on the one hand, the hopping integral is normally taken as the energy scale in the problem; it plays a role in the theoretical framework and cannot be ignored. On the other hand, previous mean-field studies have usually been performed with the uniform Hartree–Fock approximation, but the local character of the model is ignored under this approximation. To overcome this drawback, we apply the unrestricted Hartree–Fock approximation to the model, and the properties of the charge fluctuation of the system are obtained. The paper is organized as follows. In section 2 the formalism and the description of calculations is given. The results and discussion are presented in section 3.

2. The model

The model Hamiltonian we consider has the following form:

$$H = -t\sum_{\langle il\rangle,s}(d_{is}^{\dagger}p_{ls} + p_{ls}^{\dagger}d_{is}) - \mu\sum_{is} n_{is} + (\epsilon - \mu)\sum_{ls} n_{ls} + U_{\mathrm{d}}\sum_{i} n_{i\uparrow}n_{i\downarrow} + U_{\mathrm{p}}\sum_{l} n_{l\uparrow}n_{l\downarrow} + V_{\mathrm{pd}}\sum_{\langle il\rangle} n_i n_l \tag{1}$$

where $d_i^{\dagger}$ and $p_l^{\dagger}$ are the hole creation operators on Cu site i and O site l respectively, and $n_{is} = d_{is}^{\dagger}d_{is}$, $n_{ls} = p_{ls}^{\dagger}p_{ls}$. The site energy of O is ϵ (which is in fact the level difference between O and Cu, because the site energy of Cu is set to zero and $\epsilon > 0$ is assumed in this work), t is the Cu–O hybridization, U_{d} and U_{p} are the on-site Coulomb repulsion on Cu and O sites respectively, and V_{pd} is the nearest-neighbour Cu–O intersite Coulomb interaction.

In previous studies, most of the mean-field analysis has been performed under the uniform Hartree–Fock approximation. However, this approximation is not reasonable when the local properties of the system become important and some kind of non-linear excitation starts to play an important role in the system. In this case a non-uniform Hartree–Fock approximation is a good starting point for the local characteristics of the model. In fact, recent works have addressed this aspect [16–18]. Following these works, in this paper we consider the model Hamiltonian under the unrestricted Hartree–Fock approximation. Then, the many-body terms in (1) can be decoupled:

$$\langle n_{i\uparrow}n_{i\downarrow}\rangle = \langle n_{i\uparrow}\rangle n_{i\downarrow} + \langle n_{i\downarrow}\rangle n_{i\uparrow} - \langle n_{i\uparrow}\rangle\langle n_{i\downarrow}\rangle \tag{2a}$$

$$\langle n_{i\uparrow}n_{l\downarrow}\rangle = \langle n_{i\uparrow}\rangle n_{l\downarrow} + \langle n_{l\downarrow}\rangle n_{i\uparrow} - \langle n_{i\uparrow}\rangle\langle n_{l\downarrow}\rangle \tag{2b}$$

$$\langle n_{i\uparrow}n_{l\uparrow}\rangle = n_{i\uparrow}\langle n_{l\uparrow}\rangle + n_{l\uparrow}\langle n_{i\uparrow}\rangle - \langle n_{i\uparrow}\rangle\langle n_{l\uparrow}\rangle + d_{i\uparrow}^{\dagger}p_{l\uparrow}\langle d_{i\uparrow}p_{l\uparrow}^{\dagger}\rangle + p_{l\uparrow}^{\dagger}d_{i\uparrow}\langle p_{l\uparrow}d_{i\uparrow}^{\dagger}\rangle - \langle d_{i\uparrow}^{\dagger}p_{l\uparrow}\rangle\langle d_{i\uparrow}p_{l\uparrow}^{\dagger}\rangle. \tag{2c}$$

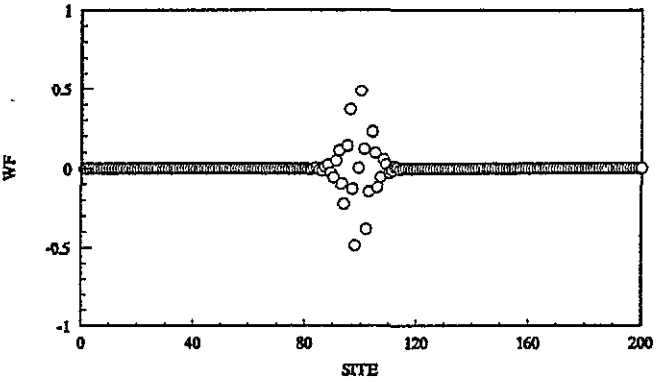

Figure 1. One-hole wavefunction of the spinless fermion model when one more hole is added to the half-filled situation with $\epsilon = 0.1$, $V_{pd} = 2.0$.

The Hamiltonian is written in the following form:

$$H = H_\uparrow + H_\downarrow + H_b \tag{3}$$

where

$$H_\uparrow = \sum_i \Big(-\mu + U_d\langle n_{i\downarrow}\rangle + \sum_l V_{pd}(\langle n_{l\downarrow}\rangle + \langle n_{l\uparrow}\rangle)\Big) d_{i\uparrow}^\dagger d_{i\uparrow} + \Big((\epsilon - \mu) + U_p\langle n_{l\downarrow}\rangle + \sum_i V_{pd}(\langle n_{i\downarrow}\rangle + \langle n_{i\uparrow}\rangle)\Big) p_{l\uparrow}^\dagger p_{l\uparrow} + (-t + V_{pd}\langle d_{i\uparrow} p_{l\uparrow}^\dagger\rangle) d_{i\uparrow}^\dagger p_{l\uparrow} + (-t + V_{pd}\langle p_{l\uparrow} d_{i\uparrow}^\dagger\rangle) p_{l\uparrow}^\dagger d_{i\uparrow} \tag{4a}$$

$$H_\downarrow = \sum_i \Big(-\mu + U_d\langle n_{i\uparrow}\rangle + \sum_l V_{pd}(\langle n_{l\downarrow}\rangle + \langle n_{l\uparrow}\rangle)\Big) d_{i\downarrow}^\dagger d_{i\downarrow} + \{(\epsilon - \mu) + U_p\langle n_{l\uparrow}\rangle + \sum_i V_{pd}(\langle n_{i\downarrow}\rangle + \langle n_{i\uparrow}\rangle)\} p_{l\downarrow}^\dagger p_{l\downarrow} + (-t + V_{pd}\langle d_{i\downarrow} p_{l\downarrow}^\dagger\rangle) d_{i\downarrow}^\dagger p_{l\downarrow} + (-t + V_{pd}\langle p_{l\downarrow} d_{i\downarrow}^\dagger\rangle) p_{l\downarrow}^\dagger d_{i\downarrow}. \tag{4b}$$

Here the spin-flip process is considered to be small, and then the up and down spin Hamiltonian decouples; they will influence each other in a self-consistent way. The term H_b is an energy shift:

$$H_b = \sum_{il} [-U_d\langle n_{i\uparrow}\rangle\langle n_{i\downarrow}\rangle - U_p\langle n_{l\uparrow}\rangle\langle n_{l\downarrow}\rangle - V_{pd}(\langle n_{i\downarrow}\rangle + \langle n_{i\uparrow}\rangle)(\langle n_{l\downarrow}\rangle + \langle n_{l\uparrow}\rangle) + V_{pd}\langle d_{i\uparrow}^\dagger p_{l\uparrow}\rangle\langle p_{l\uparrow}^\dagger d_{i\uparrow}\rangle + V_{pd}\langle d_{i\downarrow}^\dagger p_{l\downarrow}\rangle\langle p_{l\downarrow}^\dagger d_{i\downarrow}\rangle]. \tag{4c}$$

A lot of experimental and theoretical investigation shows that the doping dependence of the electronic state is not rigid-band-like, but rather an intensity transfer from the high-energy region to the low-energy region is induced, and the charge excitation gap is smaller than the uniform Hartree–Fock gap [16]. A non-uniform solution of the Hamiltonian (1) would therefore be interesting, and we will show in section 3 that a non-uniform solution is more stable than a uniform solution due to a lowering of energy.

To illustrate the local character of the unrestricted Hartree–Fock approximation, we give here a simple example: the spinless fermion model considered in [16]. Figure 1 is an illustration of the one-hole wavefunction of the one-dimensional spinless fermion model when one more hole is added to the half-filled case. Clearly, it is a polaron-like solution, and the charge distribution of the whole system is highly non-uniform. When spin is introduced, as in the present model, the ground-state configuration is more complicated, and the hole filling is also non-uniform. The details of the ground-state configuration will be discussed elsewhere.

As mentioned above, the properties of the system rely on its gap character. When the system is in the charge transfer regime, the band structure for either a uniform or a non-uniform solution will be changed with hole doping. In the case of a non-uniform solution, there still exists a region where the average total filling $\langle n \rangle$ remains constant as the chemical potential is moved. The width of the region with $\langle n \rangle$ remains constant in terms of the chemical potential μ and is regarded as the charge transfer gap in the system [7]. In figure 2, we show the gap character as a function of the filling in two regions: the charge transfer regime (figure 2(a)) and the Mott–Hubbard regime (figure 2(b)). It is clearly seen that in the charge transfer regime, when additional holes are added to the $n = 1$ system, they will go mainly to O sites. When the holes decrease from the $n = 1$ system, they are removed largely from the Cu sites. On the other hand, in the Mott–Hubbard regime, the occupation of holes on O sites is very small and remains almost unchanged when the total filling is varied. It is certainly the case that the properties of the system will be strongly influenced by the difference. In this work we pay attention mainly to the charge transfer regime.

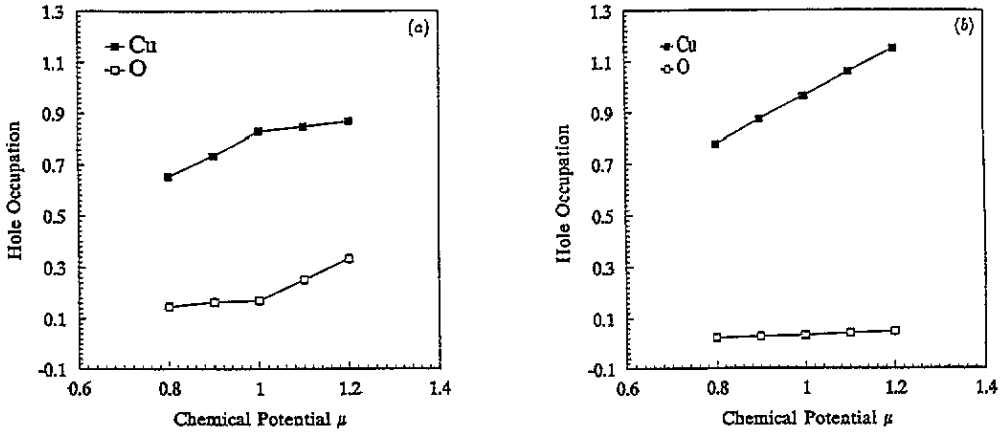

Figure 2. (a) Hole occupation of the Cu and O sites against total filling n_{tot} in the charge transfer regime: $U_d = 12$, $\epsilon = 2.0$. (b) Same as (a), but with $U_d = 6$, $\epsilon = 8.0$ (the Mott–Hubbard regime).

To capture the charge fluctuation character of the model, we define the phase-separation susceptibility χ_{PS} and the charge transfer susceptibility χ_{CT} as usual [15]:

$$\chi_{PS} = \frac{\partial}{\partial \mu}(n_{Cu} + n_O) \tag{5}$$

$$\chi_{CT} = \frac{\partial}{\partial \epsilon}(n_{Cu} - n_O)|\mu = \text{constant}. \tag{6}$$

These two quantities determine the possibility of phase separation when the system is doped beyond half filling and the response of the system to the transfer of charges between Cu and O sites respectively. The divergence of χ_{CT} will signal the onset of charge transfer instability, and the discontinuity in the total filling n_{tot} against the chemical potential μ will respond to a phase separation. The criterion is based on the following idea: states with densities inside the gap will be unstable if a discontinuity in n_{tot} is observed, i.e. the system will evolve into two regions having different densities, one poor in holes with $n_{tot} = 1$, and the other rich in holes with $n_{tot} = n_h$; this is the so-called phase-separated state.

Having equations (4*a*) and (4*b*) in hand, the Hamiltonian can be diagonalized for a finite system in a self-consistent way. In fact, the calculation of χ_{PS}, or equivalently the curve of total filling against chemical potential, is not difficult. The convergence is easy to realize. As a comparison, much CPU time is needed to ensure the convergence of χ_{CT}. One thing that should be noted here is that when two or more configurations are obtained the real one will have the lowest energy.

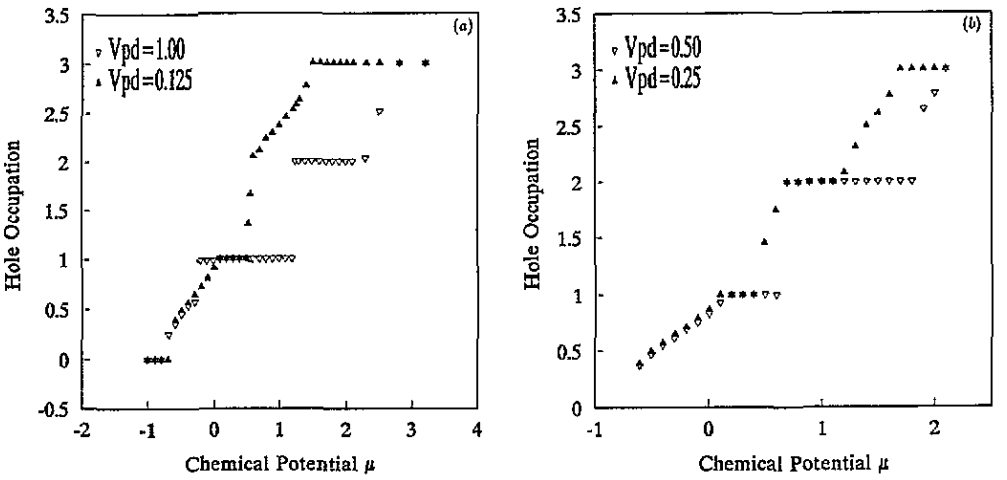

Figure 3. (*a*) The total filling $n_{tot} = n_{Cu} + n_O$ against chemical potential μ on a 1D Cu–O chain with $U_d = 100$, $U_p = 0$, $\epsilon = 1.0$. (i) $V_{pd} = 1.00$ for filled triangles. (ii) $V_{pd} = 0.125$ for open triangles. (*b*) Same as (*a*), but with $U_d = 12$, $U_p = 0$, $\epsilon = 1.0$. (i) $V_{pd} = 0.50$ for filled triangles. (ii) $V_{pd} = 0.25$ for open triangles.

3. Results and discussion

The Hamiltonian is solved for a one-dimensional Cu–O chain with a finite number of sites. The number of sites is taken to be 200: this size is adequate and large enough for our problem. Periodic boundary conditions are used during the calculation. In the following discussion we limit our consideration to two sets of parameters: (i) $U_d = 100$, $U_p = 0$ and (ii) $U_d = 12$, $U_p = 0$, although other Coulomb parameters can be adopted within our numerical studies. The value of the on-site Coulomb repulsion on a Cu site considered in the first case is large enough to be regarded as infinite. In fact, the larger value of U_d

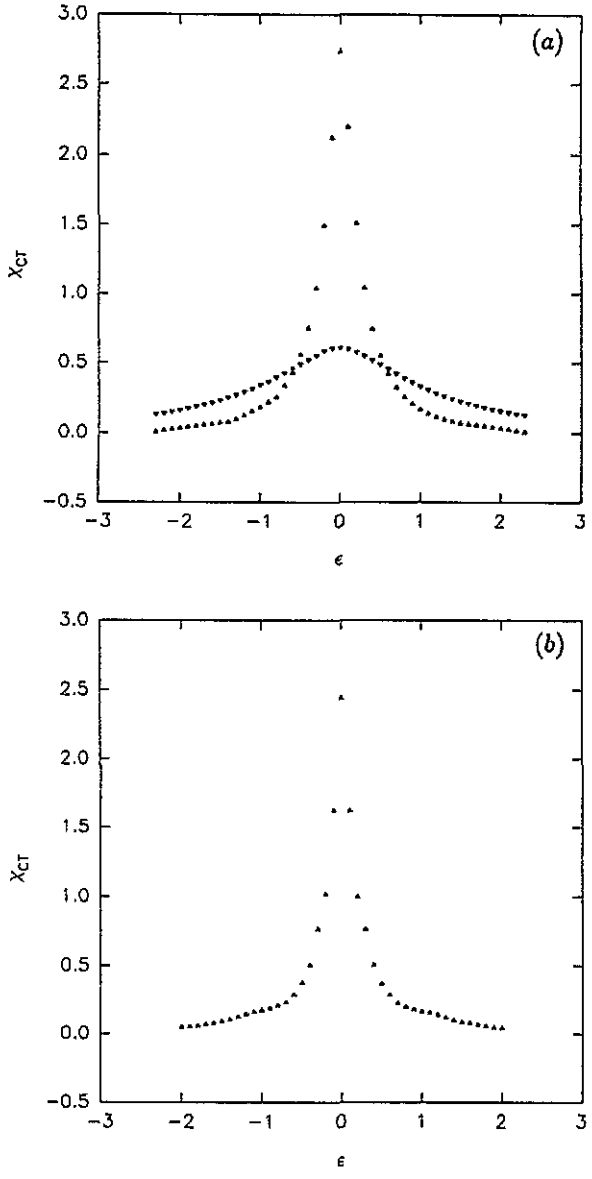

Figure 4. Charge-transfer susceptibility χ_{CT} against ϵ with $U_d = 100$, $U_p = 0$, $n = 1.0$. (*a*) $V_{pd} = 0.25$, $t = 0.25$ for filled triangles; $V_{pd} = 1.00$, $t = 1.00$ for open triangles. (*b*) Same as (*a*), but with $V_{pd} = 1.00$, $t = 0.25$.

will result in only a quantitative change of the characteristic properties of the model; no qualitative variation will occur. It will then be easy to compare our results with other works.

First, we make a numerical comparison of the total energy of non-uniform and uniform solutions, as an example. The special case we consider is the situation in which one more hole is added to a half-filled system. The energy difference is about $37.7t$ for the given parameters $U_d = 12$, $U_p = 0$, $V_{pd} = 1.5$ and $\epsilon = 2.0$. The non-uniform configuration should then be more stable when the system is doped beyond half filling.

The main properties of the charge fluctuation of the system are shown in figures 3 and 4. In figure 3, the curve of total filling against chemical potential is plotted for various parameters. Here the hopping integral t is taken as the energy scale, and $\epsilon = 1.0$ is assumed for all the cases. The result for $V_{pd} = 1.00$ is denoted by open triangles in figure 3(a). An obvious sudden change appears for hole filling n_{tot} when the chemical potential approaches a critical value. As discussed in the definition of χ_{PS}, the phase separation manifests itself as a discontinuity in n_{tot} as a function of μ; this figure is a typical illustration showing the phase separation character of the system. Compared with the zero-bandwidth results [15], the density of the two phases between which the phase separation happens is unchanged, but the distribution of charge is altered in a way that decreases the total energy. The result shows that if the system is initially prepared with a density between the two plateaux for given parameters, it will finally evolve into the phase-separated state: one region with $n_{tot} = 1$, where the charge resides on Cu, and O sites are empty, and another phase with $n_{tot} = 2$, where the charge resides on O, and Cu sites are empty. The plot with filled triangles in figure 3(a) corresponds to the case with $V_{pd} = 0.125$. It is clear that the variation of the total filling n_{tot} depends strongly on the value of V_{pd}. When V_{pd} is small enough, the discontinuity in n_{tot} is strongly suppressed and the upper plateau in n_{tot} is almost completely washed out; no phase separation will take place. As an illustration, in figure 3(b) a similar result is given with $U_d = 12$, $V_{pd} = 0.50$ and 0.25 respectively. It is a situation in which the relative values of the parameters are closer to the practical case. It can be seen that the larger value of U_d prefers phase separation, and only a quantitative change takes place compared with the $U_d = 100$ result. In both cases, the phase separation will occur over a wide range of doping $1 < n_{tot} < 2$ for large enough V_{pd}.

The charge-transfer susceptibility χ_{CT} against ϵ is shown in figure 4. All the calculations are performed under the half-filled situation, $n = 1.0$. The result for $V_{pd} = 0.25$ and $t = 0.25$ is given in figure 4(a) (filled triangles). Compared with the results of Subdo and co-workers [15], the divergent behaviour of the susceptibility χ_{CT} against ϵ is replaced by a sharp peak in the curve. The peak responds to charge transfer instability. It can also be seen that the value of the peak is sensitive to the magnitude of the bandwidth. With increasing bandwidth, the peak broadens. When $t = 1.00$ (figure 4(a), open triangles), the peak almost disappears, and a smooth curve is obtained instead. Therefore a relatively narrow band is necessary to ensure the appearance of the charge transfer instability. On the other hand, the influence of V_{pd} on χ_{CT} is relatively small (figure 4(b)).

In summary, in the present work we have studied a one-dimensional p–d model with a finite bandwidth. The Hamiltonian was solved for a finite, but large enough, system under the unrestricted Hartree–Fock approximation. The advantage of a non-uniform Hartree–Fock approximation is that the local character of the system remains. The system undergoes a phase separation and charge transfer instability when a finite bandwidth is introduced. The appearance of phase separation depends on the relative strength of the nearest-neighbour repulsion V_{pd}. A finite bandwidth makes the divergent behaviour of χ_{CT} into a sharp peak in the curve of χ_{CT} against ϵ. The occurrence of this sharp peak is sensitive to the magnitude of the bandwidth: the larger the bandwidth, the broader the peak.

Acknowledgments

The work described in this paper is supported in part by the National Centre for Research and Development on Superconductivity of China and by grant LWTZ-1298 of the Chinese Academy of Sciences.

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