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ON THE POSSIBILITY OF PHASE SEPARATION IN THE EXTENDED HUBBARD MODEL

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The one-band extended Hubbard model on the square lattice in the strong interaction limit is investigated using an exact diagonalization technique. The main emphasis was on the possibility of a phase separation. It is shown that the instability of the system against separation into a no-hole antiferromagnetic phase and an hole-rich phase that takes place at low hole concentrations is effectively destroyed through the inclusion of a moderate interatomic Coulomb interaction. In the two-phase region, i.e. at small interaction energies, a negative hole-binding energy indicates that the hole-rich phase is build up by hole-pairs. We comment on the relevance of our results for the description of the copper-oxide superconductors.

1. INTRODUCTION

THE PHYSICS of strongly correlated fermion systems is at present extensively studied, mainly triggered by the attempt to understand a mechanism underlying high temperature superconductivity in the cuprates [1]. It is now generally believed that the relevant electronic properties are determined by the copper oxide planes, where holes or electrons are introduced by doping the parent compounds as La_2CuO_4 or Nd_2CuO_4, which are antiferromagnetic insulators. Anderson [2] suggested that a two-dimensional Hubbard model with strong on-site Coulomb interactions and a nearly half-filled band may be an appropriate starting point for the theoretical description of this system. It was also shown [3] that a more realistic two-band Hamiltonian [4] in the relevant parameter regime can be reduced to an effective single-band Hamiltonian, and so the simplest one-band prototype model of the t–J form [2] has been the subject of intensive study. The central problem in this model is the strongly correlated motion of doped holes in a quantum antiferromagnet. The spin configuration that minimizes the total energy of the system can in principle be found from a complicated many-body problem caused by the interplay of antiferromagnetic correlations in the spin background and the local ferromagnetic order near the holes, i.e. from the tendency of the formation of ferromagnetic polarons or domains in an antiferromagnet [5, 6]. Therefore the investigation of the ground state properties of the t–J and related models is required. In these models, however, the possibility of an instability towards phase separation cannot be ruled out *a priori*. Along this line Emery *et al.* [7] recently addressed the question whether for the t–J model dilute holes have a uniform density or separate into a hole-rich phase and a phase without holes. The authors found indeed phase separation for $J > 0$ which implies that the doped system cannot be regarded as a weakly disordered antiferromagnet. Even if the hole-rich phase is built up by electron pairs as suggested in [7] this result would make the application of the simple t–J model to ceramic superconductors questionable.

In view of these important consequences the aim of this communication is to discuss three effects:

(i) The importance of next-nearest neighbour virtual hopping processes has recently [8, 9] been pointed out; we therefore start from the large-U expansion of the Hubbard model including three-site terms and show that qualitatively the same results were found as in the t–J model, i.e. the large-U Hubbard model is in the sense of [7] unstable against phase separation.

(ii) In the two-phase regime the structure of the hole-rich phase is investigated by calculating various binding energies as well as the energy of the hole-rich phase as function of the hole density.

(iii) However, for small additional nearest and next-nearest neighbour Coulomb interaction phase separation is rapidly destroyed making the system homogeneous.

The model studied here is the strong coupling approximation (intra-atomic Coulomb interaction $U \gg$ transfer amplitude t) of the two-dimensional

extended Hubbard Hamiltonian given by

$$H = H_1 + H_2 + H_3 + H_4, \tag{1}$$

$$H_1 = -t \sum_{\langle ij \rangle,\sigma} \tilde{c}^+_{i\sigma} \tilde{c}_{j\sigma} + (i \leftrightarrow j), \tag{2}$$

$$H_2 = \frac{4t^2}{U} \sum_{\langle ij \rangle} \left(\mathbf{S}_i \mathbf{S}_j - \frac{n_i n_j}{4} \right), \tag{3}$$

$$H_3 = -\frac{t^2}{U} \sum_{\langle ijk \rangle,\sigma} [(\tilde{c}^+_{i\sigma} \tilde{c}^+_{j-\sigma} \tilde{c}_{j-\sigma} \tilde{c}_{k\sigma} + \tilde{c}^+_{i-\sigma} \tilde{c}_{j-\sigma} \tilde{c}^+_{j\sigma} \tilde{c}_{k\sigma}) + (i \leftrightarrow k)], \tag{4}$$

$$H_4 = \sum_{ij} V_{ij} n_i n_j, \tag{5}$$

where $\mathbf{S}_i = \frac{1}{2} \Sigma_{\alpha\beta} \tilde{c}^+_{i\alpha} \boldsymbol{\sigma}_{\alpha\beta} \tilde{c}_{i\beta}$, $\tilde{c}_{i\sigma} = c_{i\sigma}(1 - n_{i-\sigma})$, $n_i = n_{i\uparrow} + n_{i\downarrow}$, $n_{i\sigma} = \tilde{c}^+_{i\sigma} \tilde{c}_{i\sigma}$, and the factor of $(1 - n_{i-\sigma})$ enforces the constraint of no double occupancy. The terms H_1 to H_3 have been derived on many occasions (cf. [10]). $H_1 + H_2$ correspond to the t–J model with $J = 4t^2/U$. Including H_3, i.e. virtual three site processes where $\langle ij \rangle$ and $\langle jk \rangle$ are nearest neighbours, we obtain the so-called t–t'–J model [10, 11] with $t' = t^2/U$. H_4 involves the interatomic part of the Coulomb repulsion between nearest and next nearest neighbours denoted by V_{nn} and V_{nnn}, respectively. For the undoped system (H_1 and H_3 vanish, while H_4 and the second term of H_2 result in a constant energy shift) it is generally believed that the ground state on the square lattice shows antiferromagnetic long range order. Upon doping the situation is much more complicated except in the Nagaoka limit [5].

Following [7] (similar ideas have been given in the earlier work of Visscher [13]) ground state properties of a system with different phases can be obtained by minimizing the total energy with respect to the portions of the various homogeneous phases. In the low doping regime relevant for the high-T_c cuprates Emery's proposal of a two-phase model with one antiferromagnetic phase containing no holes (according to the Brinkman–Rice argument [14]) and a second hole-rich phase seems to be appropriate. The energy of such a system with N_s sites is then given by

$$E = (N_s - N)\, e_{\mathrm{afm}} + N\, e_{\mathrm{h}}(x), \tag{6}$$

where N is the number of sites in hole-rich phase. The energy per site of the hole-rich phase depends only on the hole concentration $x = N_h/N$, N_h being the number of holes, and one has $e_h(0) = e_{\mathrm{afm}}$. The phase separation in terms of electrons [13] could be pictured in such a way that starting from a dilute electron gas and increasing the electron density (at fixed t, U, V), an antiferromagnetic solid with fixed density (one electron per site) condenses out if the instability condition $\mathrm{d}E/\mathrm{d}N > 0$ becomes fulfilled. Thus $\mathrm{d}E/\mathrm{d}N = 0$ fixes the critical density x_m of the hole-rich phase which is equivalent to Emery's condition. This phase separation takes place for hole concentrations $x < x_m$, provided that the function

$$e(x) = \frac{e_h(x) - e_h(0)}{x} \tag{7}$$

exhibits a minimum at x_m. In the limit $x_m = 0$ which is realized at $U/t \to \infty$ there is no phase separation (i.e. $N \to N_s$) whereas in the opposite case $x_m = 1$ the separation becomes complete, i.e. the hole phase contains no electrons.

An exact evaluation of equation (7) can be made only for finite-size clusters interpreting the energy of a cluster with N sites and N_h holes as the energy of the hole-rich phase $e_h(x)$ with the hole concentration N_h/N. Using periodic boundary condition we carry out the numerical Lanczos diagonalization [15] in the subspace of minimal S_z of the full Hamiltonian (1) for a $\sqrt{10} \times \sqrt{10}$ and a 4×4 lattice at $U/t = 20$ (all energies in units of t) where the approximation leading to (1) seems reasonable The translational invariance of the considered finite systems determines the allowed **k**-vectors of the wave function and we can use the symmetries contained in the little group of each selected **k** to reduce the dimension of the Hilbert space drastically. Note that in the representation of the Hamiltonian as a matrix more than half of the nonvanishing matrix elements arise from the H_3 term.

Our results are depicted in Fig. 1. The main con-

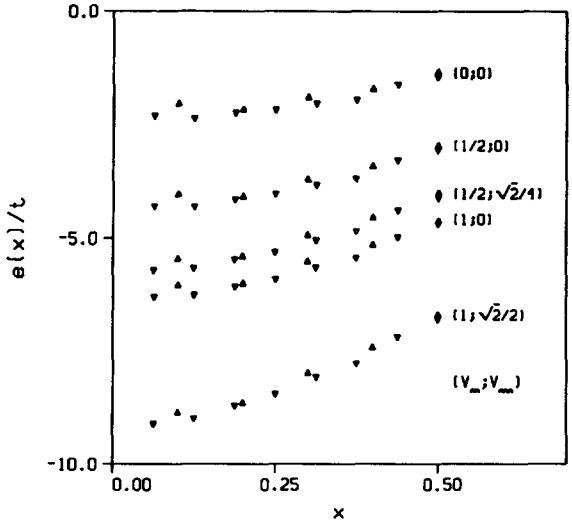

Fig. 1. The function $e(x)$ (see text) is shown at several hole fractions in the hole-rich phase for $U = 20$ and different interatomic Coulomb interactions, where the upper and lower triangles correspond to the lattice with 10 and 16 sites, respectively.

clusion of this figure is that the minimum of $e(x)$ which is maintained also for the t–t'–J model vanishes rapidly for increasing V_{nn} and V_{nnn}. That means the inclusion of moderate interatomic Coulomb repulsion restores the homogeneity of the system. Besides the strong influence of the longer range Coulomb interaction the curves at various lattice sizes indicate that in the parameter region where $e(x)$ shows a minimum, this minimum is weakened and shifted to lower hole concentrations if the size of the system is raised. This is due to the fact that in all present finite-size calculations the minimum of $e(x)$ (in the strong coupling regime $U \gg t$) is always connected to the two-hole ground state. Therefore it is interesting to speculate on whether in this limit Emery's [7] identification of N_h holes on a finite lattice with a hole concentration $x = N_h/N_{lattice}$ of the hole-rich phase makes sense, i.e. really supports the qualitative arguments for the occurrence of phase separation in the large-U Hubbard model given by Visscher [13] and by Ioffe and Larkin [16].

The structure of the hole-rich phase can be discussed in more detail if we calculate the energy of various hole bound states. Our results for the binding energies

$$E_B^2 = E(2) + E(0) - 2E(1),$$
$$E_B^3 = E(3) + E(0) - E(2) - E(1),$$
$$E_B^4 = E(4) + E(0) - 2E(2)$$

of two, three and four holes, respectively, are given in Table 1, where $E(N_h)$ denotes the total energy in the hole phase with N_h holes. As reported by several exact diagonalization studies [17], the negative value of E_B^2 in the relevant U region indicates the tendency of two holes to bind together. Including the longer ranged Coulomb forces this binding is weakened and finally lifted for larger values of V_{ij}. Because E_B^3, $E_B^4 > 0$ the result of Emery *et al.* [7] that in the hole-rich phase the first instability is due to hole pairs seems reasonable in the case where phase separation is favourable. However, increasing the size of the system the absolute value of these energies decreases and one can speculate whether they remain finite for an infinite system (cf. [8, 11]).

Let us now consider the behaviour of the energy per site in the hole-rich phase $\Delta E = e_h(x) - e_{afm}$ as a function of x (Fig. 2). For the pure t–t'–J model ($H_4 = 0$) we can compare our results with other diagonalization studies [17] up to $N_h = 4$ and get good agreement. Obviously our data are only weakly size dependent. The interatomic Coulomb interaction suppresses the well known minimum [18] of the large-U Hubbard model at an almost quarter-filled band. The influence of the H_4 term on the ground state symmetry is by no means trivial, e.g., the **k**-degeneracy of the

Table 1. Binding energies E_B^2, E_B^3 and E_B^4 of two, three and four holes, respectively, in the ground state of the Hamiltonian (1) for the 4 × 4 lattice with periodic boundary conditions at different interaction energies U, V_{nn} and V_{nnn}. To illustrate the size dependence of the binding energies the corresponding values for the $\sqrt{10} \times \sqrt{10}$ lattice are given in rectangular brackets

U	V_{nn}	V_{nnn}	E_B^2	E_B^3	E_B^4
10	0	0	−0.274	0.126	0.493
			[−0.502]	[0.094]	[1.240]
	0.5	0	−0.142	0.302	0.820
			[−0.283]	[0.515]	[1.978]
	0.5	$0.5/\sqrt{2}$	−0.043	0.493	1.189
			[−0.107]	[0.876]	[2.726]
	1	0	−0.028	0.462	1.133
			[−0.108]	[0.905]	[2.687]
	1	$1/\sqrt{2}$	0.164	0.837	1.860
			[0.247]	[1.651]	[4.275]
20	0	0	−0.132	0.304	0.757
			[−0.250]	[0.678]	[1.845]
	0.5	0	−0.025	0.463	1.097
			[−0.087]	[1.095]	[2.691]
	0.5	$0.5/\sqrt{2}$	0.079	0.644	1.452
			[0.089]	[1.455]	[3.428]
	1	0	0.068	0.604	1.413
			[0.061]	[1.879]	[3.459]
	1	$1/\sqrt{2}$	0.257	0.969	2.133
			[0.418]	[2.224]	[5.018]

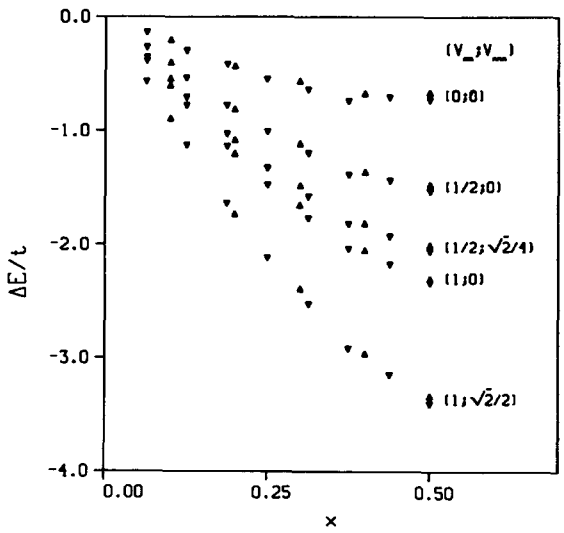

Fig. 2. Ground state energy per site of the hole phase as a function of the hole concentration for the $\sqrt{10} \times \sqrt{10}$ and the 4 × 4 lattice with periodic boundary condition. The symbols are the same as in Fig. 1.

ground state (cf. [17]) is lifted by V_{nnn}. The details of this **k**-dependence of the ground state will be reported in a subsequent publication. On general stability arguments the energy of an homogeneous system should be a convex function of the density: otherwise a phase separated state can be obtained by a Maxwell construction [19]. That means the critical density x_m given by the minimum of (7) can be alternatively determined from the turning point of $\Delta E(x)$, i.e. $e_h''(x_m) = 0$ or $\Delta E(x)$ is a concave function at $x < x_m$.

In any case it would be desirable to evaluate $e(x)$ at smaller x values to decide whether the minimum of $e(x)$ at low hole concentrations is really lifted, but this means diagonalization of much larger clusters. However, we believe that in general the inclusion of moderate nearest or next-nearest Coulomb interaction restores the phase homogeneity of the system. That may be important for the application of one-band Hubbard models to the high-T_c superconductors.

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REFERENCES

1. J.G. Bednorz & K.A. Müller, *Z. Phys.* **B64,** 189 (1986).
2. P.W. Anderson, *Science* **235,** 1196 (1987).
3. F.C. Zhang & T.M. Rice, *Phys. Rev.* **B37,**3759 (1988).
4. V.J. Emery, *Phys. Rev. Lett.* **58,** 2794 (1987).
5. Y. Nagaoka, *Phys. Rev.* **147,** 392 (1966).
6. D. Foerster, *Z. Phys.* **B74,** 295 (1989).
7. V.J. Emery, S.A. Kivelson & H.Q. Lin, *Phys. Rev. Lett.* **64,** 475 (1990).
8. J.A. Riera & A.P. Yang, *Phys. Rev.* **B39,** 9697 (1989).
9. P.A. Lee, *Phys. Rev. Lett.* **63,** 680 (1989).
10. E. Kaxiras & E. Manousakis, *Phys. Rev.* **B37,** 656 (1988).
11. S.A. Trugman, *Phys. Rev.* **B37,** 1597 (1988).
12. It should be stressed that the terminology t–t'–J model is not unique; starting from two- or three-band models the mapping procedure may give an effective one-band model, where t' includes in addition to the second order hopping process of H_3 an intra-sublattice contribution due to the direct transfer between O-sites (cf. M.S. Hybersten *et al.*, preprint).
13. P.B. Visscher, *Phys. Rev.* **B10,** 943 (1974).
14. W.F. Brinkman & T.M. Rice, *Phys. Rev.* **B2,** 1324 (1970).
15. A. Mistriotis, H. Büttner & W. Pesch, *J. Phys. Cond. Matter* **1,** 891 (1989).
16. L.B. Ioffe & A.I. Larkin, *Phys. Rev.* **B37,** 5730 (1988).
17. J. Bonca, P. Prelovsek & I. Sega, *Phys. Rev.* **B39,** 7074 (1989); J.A. Riera & A.P. Young, *Phys. Rev.* **B40,** 5285 (1989); M. Ogata & H. Shiba, *J. Phys. Soc. Japan* **58,** 2836 (1989); Y. Hasegawa & D. Poilblanc, *Phys. Rev.* **B40,** 9035 (1989).
18. H. Yokoyama & H. Shiba, *J. Phys. Soc. Japan* **56,** 3570 (1987).
19. M. Marder, N. Papanicolaou & G.C. Psaltakis, *Phys. Rev.* **B41,** 6920 (1990).