Phase Separation in the Extended Hubbard Model at Weak Coupling

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The Hubbard model including nearest-neighbor interaction is studied at $T = 0$ on a d-dimensional hypercubic lattice ($d \geq 3$) close to half filling. It is shown that the ground state at weak coupling is *phase separated,* consisting of homogeneous lower-density and antiferromagnetic or charge density wave higher-density regions. Incommensurate phases are unstable. The exact critical doping level and the exact order parameter are renormalized relative to the Hartree results. The renormalization factor at weak coupling turns out to be *identical* to the result found previously for the same model at half filling.

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In recent years the Hubbard model [1–3] has advanced to one of the most important standard models for interacting electrons on a lattice. The success of the Hubbard model is based on its ability to explain a number of important phenomena in condensed matter physics. Among these are the (Mott-Hubbard) metal-insulator transition [1,4], antiferromagnetism [5,6], and several normal-state properties of high-T_c materials [7,8]. Unfortunately, little is known *exactly* about the ground state or the thermodynamics of the Hubbard model. Nevertheless, the model at half filling is qualitatively well understood, mainly as a consequence of the exact solution in one dimension [9,10] and several rather general rigorous results [11].

The situation away from half filling is more severe. Here there exists basically only one rigorous result, due to Nagaoka [12], stating that the ground state for a single hole in a half-filled band is ferromagnetic in the extreme strong-coupling limit ($U \rightarrow \infty$). Furthermore, various approximate (Hartree-Fock [13,14] and slave-boson [15]) methods have been applied to study possible commensurate and incommensurate phases. Much work has also been done on the t-J model, which is a simplified strong-coupling version of the Hubbard model. The t-J model displays, apart from commensurate and incommensurate phases [16], various indications of phase separation at small doping [17–20]. The latter result is particularly important in view of the experimental evidence for phase separation in high-T_c superconductors [21]. On the basis of these results it has been conjectured by Emery and Kivelson [18] that phase separation occurs rather generally in antiferromagnets at small doping. Indeed we will see below that the ground state of the Hubbard model at weak coupling is phase separated. This conclusion remains true if one considers the *extended* Hubbard model, which contains also nearest-neighbor interaction.

The purpose of the present Letter is to present new exact results on the phase diagram of the doped extended Hubbard model in the *weak-coupling* limit. The main result to be found below is that the ground state of extended Hubbard model at weak coupling is *phase separated,* consisting of regions with long-range antiferromagnetic (AFM) or charge density wave (CDW) order at density $n = 1$ and disordered regions at density $n = 1 - \delta_c < 1$. Moreover, we show that the *exact* results for the order parameter and for the critical doping concentration δ_c are strongly reduced by fluctuations relative to the corresponding Hartree results. The renormalization factor can be calculated exactly and turns out to be identical to that found previously [22] for the model at half filling. These results will be derived for the extended Hubbard model on a hypercubical lattice in high dimensions ($d \gg 1$). The application kept in mind is that of the model in $d = 3$; finite dimensionality will be accounted for in a $1/d$ expansion.

The extended Hubbard model describes hopping of electrons (H_t), interacting with each other through on-site (H_U), and nearest-neighbor (H_V) Coulomb repulsion. Accordingly, its Hamiltonian has the form [23]

$$H = H_t + H_U + H_V + H_\mu , \tag{1}$$

where

$$H_t = -\frac{1}{\sqrt{2d}} \sum_{(\mathbf{ij}),\sigma} c^\dagger_{\mathbf{i}\sigma} c_{\mathbf{j}\sigma}, \qquad H_\mu = -\mu \sum_{\mathbf{i}} n_{\mathbf{i}} ,$$

$$H_U = U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}, \qquad H_V = \frac{V}{d} \sum_{\langle \mathbf{ij} \rangle} n_{\mathbf{i}} n_{\mathbf{j}} .$$

Here $c^\dagger_{\mathbf{i}\sigma}$ ($c_{\mathbf{i}\sigma}$) creates (destroys) an electron with spin σ at site $\mathbf{i}$, $n_{\mathbf{i}\sigma} \equiv c^\dagger_{\mathbf{i}\sigma} c_{\mathbf{i}\sigma}$, $n_{\mathbf{i}} \equiv n_{\mathbf{i}\downarrow} + n_{\mathbf{i}\uparrow}$, and d is the space dimension. The prefactors in H_t and H_V are chosen such that a finite energy contribution is obtained in the limit $d \rightarrow \infty$ [24]. In taking the weak coupling limit $U, V \downarrow 0$ we keep the ratio $v = V/U$ fixed. Moreover, we assume $U > 0$ and $V > 0$ (corresponding to Coulomb *repulsion*).

In order to demonstrate that phase separation occurs we proceed as follows. We first determine the phase diagram in Hartree approximation, for both commensurate (AFM or CDW) and incommensurate phases. Already in Hartree approximation phase separation is favored at weak coupling. We then show that the only effect of the fluctuations at weak coupling is to renormalize the

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Hartree results, implying that the *exact* ground state of the extended Hubbard model, too, is phase separated.

The Hartree approximation for commensurate (AFM or CDW) phases is defined by the usual decoupling scheme [22] for the interaction terms in (1). The averages $\langle n_{i\sigma}\rangle$ occurring in the Hartree Hamiltonian are determined by $\langle S_i^z\rangle = \frac{1}{2}\lambda\Delta$ (in the AFM phase) or by $\langle n_i\rangle = n + \lambda\Delta$ (in the CDW phase). Here Δ denotes the order parameter and $\lambda = +1$ on one sublattice [labeled by (+)] and $\lambda = -1$ on the other [labeled by (−)]. The order parameter Δ can be calculated from the consistency requirement $\langle n_{i\uparrow}\rangle_H = \frac{1}{2}(n \pm \Delta)$ if $\mathbf{i} \in (\pm)$. One finds a nontrivial solution $\Delta > 0$ only for $\delta < \delta_1$, where δ_1 is the critical density of holes.

The Hartree approximation yields the following results. At weak coupling both the order parameter and the critical doping concentration δ_1 are *exponentially small*. The scale for both is set by the Hartree gap parameter H_0 at half filling, which is at weak coupling, including the $1/d$ corrections, given by [22]

$$H_0 \sim 2\sqrt{2}\exp\left(-\frac{1}{2\alpha U \nu_d(0)} - \frac{1}{2}\gamma + \frac{1}{4d}\right).$$

Here $\alpha = \frac{1}{2}$ ($\alpha = 2v - \frac{1}{2}$) in the AFM (CDW) phase, $\nu_d(\varepsilon)$ is the density of states for $U = 0$, and $\gamma \simeq 0.577$ is Euler's constant. In terms of H_0, the order parameter $\Delta(\delta)$ and the critical density δ_1 are given by

$$\Delta(\delta) = \frac{H_0}{\alpha U}\sqrt{1 - \delta/\delta_1}, \qquad \delta_1 = \nu_d(0)H_0 .$$

The same value δ_1 is obtained if one calculates the divergence of the RPA susceptibility in the homogeneous phase at wave vector $\mathbf{q} = \mathbf{Q} \equiv (\pi, \pi, \pi, \ldots)$. Note that the order parameter has a mean-field critical exponent of $1/2$ near $\delta = \delta_1$. One can also calculate the energy gain per site E_S due to symmetry breaking. One finds

$$E_S(\delta) = -\tfrac{1}{2}(H_0)^2\nu_d(0)(1 - \delta/\delta_1)^2 .$$

This result shows that symmetry breaking leads to a small energy gain, of order H_0^2. The *lowest* ground state energy is obtained for the phase with the *largest* value of α. Thus, like at half filling, the AFM (CDW) phase has lower energy if $v < \frac{1}{2}$ ($v > \frac{1}{2}$). Even more interesting than $E_S(\delta)$ is the difference between the ground state energy $E(\delta)$ at concentration δ and the ground state energy at half filling. The result is

$$E(\delta) - E(0) = -(2V + \tfrac{1}{2}U)\delta + 2\nu_d(0)(H_0)^2\Phi(\delta/\delta_1) ,$$

where

$$\Phi(x) = \begin{cases} \frac{1}{2}x(1 - \frac{1}{4}x) & (x < 1), \\ \frac{1}{4}(1 + \frac{1}{2}x^2) & (x > 1). \end{cases} \tag{2}$$

A sketch of $E(\delta)$, or rather $\Phi(\delta/\delta_1)$, is given in Fig. 1. Equation (2) clearly shows that the ground state energy is *not convex* as a function of particle number, at least not for

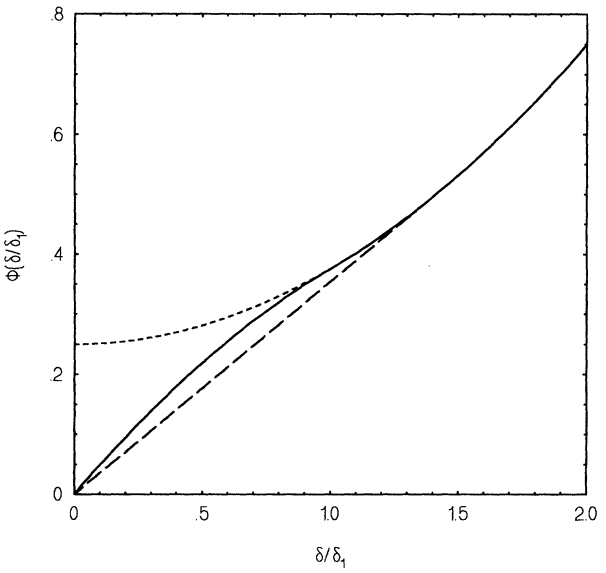

FIG. 1. The energy of the antiferromagnetic state as a function of doping (solid curve), compared to the energy of the homogeneous phase (dashed curve), and that of the phase separated state (long-dashed curve).

$\delta < \delta_1$. Since this is thermodynamically impossible (it implies a negative compressibility), one has to conclude that pure AFM or CDW symmetry breaking is not stable.

A priori there are two possible solutions to this problem. The first possibility is that the broken-symmetry phase is indeed commensurate, but that the transition is *first* rather than *second* order. From (2) one can construct a thermodynamically stable (phase separated) state by means of Maxwell's construction (long-dashed line in Fig. 1): The stable state consists of a mixture of AFM or CDW regions (at density $n = 1$) and disordered regions (at density $n = 1 - \delta_3$). The critical density of holes δ_3, below which such a phase separated state is stable, follows from (2) as $\delta_3 = \sqrt{2}\delta_1$. The second possible solution to the nonconvexity problem is that the broken-symmetry phase is *incommensurate*, rather than commensurate. This possibility will be considered next.

The simplest way to investigate possible incommensurate symmetry breaking is to calculate the spin-dependent density-density susceptibility in RPA approximation in the homogeneous phase. The calculations are very similar to those carried out for spinless fermions by Uhrig and Vlaming [25]. One finds that the homogeneous phase becomes unstable at wave vector $\mathbf{q}$ if either $1 = U\chi_0(\mathbf{q})$ or $1 = (-4V\rho_{\mathbf{q}} - U)\chi_0(\mathbf{q})$, where $\chi_0(\mathbf{q})$ is the susceptibility of the noninteracting system and $\rho_{\mathbf{q}} \equiv d^{-1}\sum_{n=1}^{d}\cos(q_n)$. The analysis of the RPA criterion is simple only in high dimensions ($d = \infty$), since in this case $\chi_0(\mathbf{q})$ depends upon $\mathbf{q}$ only through $\rho_{\mathbf{q}}$. One finds that the optimal value ρ_{opt} of $\rho_{\mathbf{q}}$, for which the RPA susceptibility di-

verges first, is related to the effective chemical potential $\mu_0 \equiv \mu - (2V + \frac{1}{2}U)n$ as $\mu_0/\sqrt{1+\rho_{\rm opt}} \simeq 0.924$. The same relation was found in Ref. [25] for spinless fermions. From this relation between μ_0 and $\rho_{\rm opt}$ one can calculate the critical concentration of holes δ_2 for the optimal incommensurate phase. The result is $\delta_2/\delta_1 \simeq 1.278$. However, since phase separation occurs already at $\delta_3 = \sqrt{2}\delta_1$, one concludes that at weak coupling $\delta_1 < \delta_2 < \delta_3$, so that incommensurate phases are suppressed and phase separation dominates.

Although these arguments clearly demonstrate that phase separation must occur, still the *nature* of the phase separated state could be more complicated than the mixture of AFM or CDW and homogeneous domains discussed above. One could imagine other scenarios, such as the coexistence of incommensurate and homogeneous domains (near δ_3) or a coexistence of incommensurate and AFM or CDW domains (at low doping). Such scenarios depend crucially on the form of the energy as a function of doping in the incommensurate broken-symmetry phase. To rule out such more complicated scenarios we investigate $E(\delta)$ for $\delta < \delta_2$. We consider an incommensurate spin density wave [26] spiraling in the x-z plane at wave vector $\mathbf{q} \neq \mathbf{Q}$. The order parameter is then defined through $\langle S_{\mathbf{i}}^z\rangle + i\langle S_{\mathbf{i}}^x\rangle = \frac{1}{2}\Delta e^{i\mathbf{q}\cdot\mathbf{i}}$, where $\mathbf{S_i}$ denotes the Heisenberg spin. The corresponding Hartree Hamiltonian can be diagonalized by standard methods. One finds equations for the order parameter, the ground state energy, and the Green functions. The analysis of these equations for $\delta < \delta_2$ is rather awkward unless $d = \infty$. In this case one can show that the optimal solution has the properties $\Delta \to \Delta_0$, $\mu_0 \to -H_0$ and $\rho_{\rm opt} \to -1$ if $\delta/\delta_2 \to 0$. Moreover, the ground state energy of the optimal incommensurate phase is nonconvex as a function of particle number: $\partial^2 E/\partial n^2 \sim -\sqrt{\pi/2}\ln(1/\delta) \to -\infty$ at small doping. Hence, this phase, too, cannot be stable. As a consequence, the more complicated scenarios, discussed above, can be ruled out: The stable (phase separated) ground state is indeed a mixture of purely AFM and homogeneous domains.

Next we address the effects of fluctuations on the Hartree results, which will be labeled by a superscript "H" from now on. From Ref. [22] we know that, to obtain *exact* results in the broken-symmetry phase at weak coupling ($U, V \downarrow 0$), one has to go to second order in perturbation theory. There are various ways to do this [27–29]. Here we use perturbation theory at constant order parameter [27]. The diagrams to be calculated in this approach away from half filling are the same as those considered in the half-filled case [22]; the interpretation of the lines in the diagrams (Green functions) is of course different. Below we present the main results of this calculation. Details will be published elsewhere.

The main result of the diagrammatic study is that, in the AFM or CDW phase, the exact critical concentration of holes δ_1, the exact order parameter Δ, and the exact energy gain $E_S(\delta)$ can be expressed in terms of their Hartree equivalents and a scaling factor $q(v) = \exp(-C_0 - C_1 d^{-1})$. Here C_0 and C_1 take the form

$$C_0 = \frac{1}{2\sqrt{2}\alpha^2}\ln(\sqrt{2}+1),$$

$$C_1 = \frac{(\alpha-1)v}{2\alpha^2} - \frac{1}{64\alpha^2} + C_0\left(\frac{3}{32} + v + 4v^2\right),$$

which is *identical* to the result found at half filling [22]. Numerical values in $d = 3$ are $q \simeq 0.282$ for the pure Hubbard model ($v = 0$) and $q \simeq 0.179$ for $v = \frac{1}{2}$. The exact expressions for δ_1, $\Delta(\delta)$, and $E_S(\delta)$ are given by

$$\delta_1 \sim q\delta_1^H,$$

$$\Delta(\delta) \sim q\Delta^H(\delta/q) \qquad (U, V \downarrow 0),$$

$$E_S(\delta) \sim q^2 E_S^H(\delta/q).$$

Clearly the system can still gain energy by breaking the symmetry ($E_S < 0$), so that fluctuations do not suppress the tendency toward long range order. Furthermore, the exact energy difference $E(\delta) - E(0)$ is given by

$$E(\delta) - E(0) = -(2V + \tfrac{1}{2}U)\delta + 2\nu_d(0)q^2 H_0^2 \Phi(\delta/\delta_1),$$

with $\Phi(x)$ given by (2). Hence, $\partial^2 E/\partial n^2 < 0$ for all $\delta < \delta_1$ also when fluctuations are taken into account. As a consequence the pure AFM or CDW phases are again unstable. This demonstrates that, if only commensurate phases are considered, phase separation will actually occur in the extended Hubbard model and is *not* an artifact of the Hartree approximation. The critical concentration of holes, below which phase separation occurs, is $\delta_3 = \sqrt{2}\delta_1 = \sqrt{2}q\delta_1^H$.

Incommensurate phases might interfere with phase separation only if the incommensurate q factor is significantly larger that the one found above for commensurate phases. To investigate whether this happens I calculated $q(v)$ in the incommensurate broken-symmetry phase. In $d = \infty$ one can show that the commensurate and incommensurate q factors are, in fact, equal. The conclusion is therefore that, at weak coupling, incommensurate phases do *not* interfere with phase separation, so that the ground state of the extended Hubbard model is *phase separated*.

Concerning the relevance of our results for finite U, V, from a simple continuity argument it is clear that phase separation, found here in the weak-coupling limit, will be dominant for a finite range of interactions. The size of the phase separation region can be estimated within the Hartree approximation and turns out to be strongly doping dependent. Near half filling both the commensurate and incommensurate phases are unstable for all $U, V > 0$. On the other hand, near δ_c incommensurate phases are stable from $U_0 \simeq 0.56/(4v+1)$ onwards. Hence, phase separation is to be expected at weak coupling ($U \leq U_0$) and probably also at larger U for small doping

($\delta \ll \delta_c$). Recently, the magnetic phase diagram of the ∞-dimensional Hubbard model has been studied by Freericks and Jarrell in a Monte Carlo simulation [30]. In the simulation stable incommensurate phases are found (at positive temperatures) for $\delta \geq 0.025$, $\sqrt{2} \leq U \leq 7\sqrt{2}$. The present work shows that phase separation is to be added to the phase diagram in the region where both U and δ are small. A detailed comparison to the simulation requires the extension of our results to $T > 0$, which is, in principle, straightforward. Clearly, the renormalization effects in the simulation (at finite values of U) will be more complex than the simple scaling behavior in terms of a q factor, found here for $U \to 0$. In general, the renormalization effects will depend on the interaction strength U, the doping concentration, the temperature, and the ordering channel. It seems likely that such nonuniversal effects can be well described by self-consistent perturbation theory provided U is not too large ($U \lesssim 2$).

To summarize the results, we studied the phase diagram of the extended Hubbard model away from half filling at weak coupling on a hypercubic lattice in dimensions $d \geq 3$. Finite dimensionality was accounted for in a $1/d$ expansion. Our main result is that the ground state of the extended Hubbard model at weak coupling is phase separated, consisting of AFM or CDW higher-density and disordered lower-density domains. Incommensurate phases are suppressed. This result adds support to the conjecture [18] that phase separation occurs generally in slightly doped antiferromagnets. We also found that, even in the limit $U, V \to 0$, the Hartree results for the critical doping level δ_c and for the order parameter $\Delta(\delta)$ are renormalized by a factor $q < 1$. The implication of this result is that thermodynamic and transport properties cannot be calculated reliably in Hartree approximation for any $U, V > 0$, at least not in the broken-symmetry phase. We estimated that phase separation persists up to $U_0 \simeq 0.56/(4v + 1)$ near δ_c; the phase separation region for $\delta \ll \delta_c$ may well be larger. These exact results, which are based on a $1/d$ expansion at weak coupling, will hopefully lead to more insight into the doped Hubbard model in low dimensions ($d = 3$, perhaps also $d = 2$) and its relevance for real materials.

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