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The negative-U Hubbard model with long-range Coulomb interaction: metal–insulator transition far from half-filling

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Abstract

It is shown that a metal–insulator transition can occur far from half-filling in the negative-U Hubbard model in the presence of long-range repulsive interactions. Specifically, we consider the bcc lattice at an electron concentration of $2/3$ and show that a CDW insulating state exists which is energetically favoured over the relevant metallic states. The repulsive interaction plays the same role as it does in stabilizing a Wigner crystal. Despite the absence of Fermi surface nesting, the CDW insulator appears at rather small values of the interaction, preceded by a CDW semimetal at even smaller values. This places severe restrictions on the region of the parameter space where superconductivity may exist. We believe that the model will show similar behaviour for other electron densities and other lattices.

1. Introduction

The negative-U Hubbard model has frequently been used to study superconductivity [1–3]. The model is characterized by a nonretarded on-site attraction U which promotes local pairing, and gives rise to charge-density-wave (CDW) instabilities. In the U-only model, these instabilities arise from lattice effects, and are strongest near half-filling, but usually are not strong enough to destabilize a superconducting state. However, long-range Coulomb repulsion $V(\mathbf{r})$ is expected to favour a CDW over superconductivity and thus has to be considered in describing real systems. Since a CDW is a dielectric screening instability (see below), one cannot invoke metallic screening beforehand, and use short-range repulsion to start with. In practice, theoretical calculations are based on the so-called *extended* Hubbard model in which $V(\mathbf{r})$ is taken to be short-ranged, and usually restricted to nearest neighbours. This model has been studied widely; and has recently been applied, e.g., to barium bismuthate [1, 4]. The combination of a short-ranged V and Fermi surface nesting in a bipartite lattice is sufficient to stabilize a CDW insulator at half-filling. Even in nonbipartite lattices an insulator can exist if V is large enough [2]. However, the insulator does not exist away from half-filling. It evolves into a CDW metal or a coexisting phase, which soon gives way to the ordinary metal (and hence, a superconductor). Experimentally, however, barium bismuthate is not only an

insulator at half-filling, it continues to be one far from half-filling, in disagreement with the prediction of the short-range model [1].

In this paper we study the instability of a normal metal toward a CDW, in the presence of a true long-range repulsion $V(\mathbf{r}) \propto 1/r$. In this case, the existence of insulator far from half-filling, while possible in principle, is yet to be established theoretically. Here we provide an example of such a state based on a microscopic calculation. We are primarily interested in the $U < W$ regime, where W is the bandwidth. A simplifying feature of the CDW instability is that it occurs at the Hartree level. Hence a self-consistent Hartree approximation is sufficient to establish the existence of the CDW state. However, for long-range $V(\mathbf{r})$, even the Hartree approximation runs into serious mathematical difficulties. Since the CDW is a crystal on top of a crystal, finding the correct self-consistent structure for an arbitrary value of the electron concentration n_c is exceedingly difficult; and for an incommensurate n_c, the problem is mathematically intractable.

We therefore consider a specific (bcc) lattice at a specific density, $n_c = 2/3$, which allows us to construct reasonably simple commensurate structures. Since the lattice is bipartite, the Fermi surface is perfectly nested at half-filling ($n_c = 1$), leading to a CDW insulator. At $n_c = 2/3$ there is no Fermi surface nesting, and there is no stable insulating state if V is short-ranged. However, for long-range V, we find an indirect gap CDW insulator (different from the one found at half-filling), which is always stable relative to the CDW metallic state that evolves out of half-filling. This conclusion is shown to be consistent with the results of an analysis in the strong-coupling ($U = \infty$) limit. In this case, electrons are paired into bosons which do not move; the problem involves only potential energy so that the Hartree approximation is exact. The elementary excitations are found to be gapped, implying that the CDW state is locally stable. The case of large (but finite) U is very difficult to handle, and is only discussed qualitatively.

Our main results can be summarized in terms of an effective interaction u (to be defined later). The ordinary metal is stable only for $u < 0.142W$, and is separated by a CDW semimetal from the insulator, which appears for $u > 0.174W$. Thus the insulator exists over a rather large region of the parameter space. These results, though derived here for a specific density, are likely to be more general since the reason for the stability of the CDW insulator is the combination of two factors: long-range repulsion which favours 'Wigner' crystals at lower densities [5], and lattice effects which promote CDWs toward half-filling. However, unlike the Wigner crystal, this is a crystal formed by pairs, rather than electrons.

Previous work on the short-range problem, in which both the CDW and superconducting order parameters are included, has shown that the CDW insulator remains stable except when V is very small [2]. This conclusion is likely to be valid in the long-range case as well. However, a similar calculation is not attempted here since, for long-range $V(\mathbf{r})$, the problem becomes mathematically impractical. We discuss these issues in the concluding section 4. In section 2, we discuss the occurrence of a CDW instability in the ordinary metal, and the role of metallic screening. We then consider the two-sublattice insulator which is the ground state at half-filling, and which, away from half-filling, becomes metallic. In section 3, we identify a different, three-sublattice, CDW state using a strong-coupling treatment. The CDW states are then studied by a self-consistent Hartree approximation. The energies are compared with those metallic phases, with or without a CDW. Our results are summarized in section 4.

2. The two-sublattice CDW

We consider the Hamiltonian

$$H = -t \sum_{ij\sigma} c^{\dagger}_{i\sigma} c_{j\sigma} - U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \tfrac{1}{2} \sum_{i \neq j} V(\mathbf{r}_i - \mathbf{r}_j) n_i n_j, \tag{1}$$

where $c^{\dagger}_{i\sigma}$ creates an electron with spin σ and $n_i = \sum_\sigma n_{i\sigma}$. The first term describes nearest-neighbour hopping, the last term, long-range repulsion, and $U > 0$. For uniform states, the Hartree part of the interaction energy is cancelled by the neutralizing positive background. However, in the presence of a CDW, the Hartree term dominates, and can be used to distinguish different states. This is done by using the self-consistent Hartree approximation. We do not expect higher-order contributions (e.g., exchange and correlation) to change our result qualitatively.

Let us first consider the onset of a CDW instability from the metallic side. In a metal the effective electron–electron interaction is screened. It is then tempting to start with a screened, i.e., short-ranged, interaction and then look for the CDW instability. Such a procedure is not valid, however. To see this, consider the screened interaction in the random-phase approximation (RPA): $v_{\rm scr}(\mathbf{q}) = v(\mathbf{q})/D(\mathbf{q})$. Here $v(\mathbf{q}) = -U/2 + V_{\mathbf{q}}$ is the charge part of the interaction potential, where

$$V_{\mathbf{q}} = \sum_{\mathbf{r}\neq 0} \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\mathbf{r}} V(\mathbf{r}), \tag{2}$$

is the repulsive part. $D(\mathbf{q})$ is the static dielectric constant in the RPA:

$$D(\mathbf{q}) = 1 - v(\mathbf{q})\chi_0(\mathbf{q}), \tag{3}$$

where $\chi_0(\mathbf{q})$ is the charge susceptibility for the noninteracting system.

The metallic state becomes unstable to a CDW with a wavevector $\mathbf{Q}$ when its charge susceptibility $\chi(\mathbf{Q})$ diverges. Now, the introduction of CDW gives rise to screening which must be included in χ. Again using RPA, we have $\chi(\mathbf{q}) = \chi_0(\mathbf{q})/D(\mathbf{q})$. Thus, the CDW instability is a dielectric instability associated with screening. It occurs when $D(\mathbf{Q})$ vanishes for some $\mathbf{Q}$. This is precisely the condition obtained from our self-consistent Hartree approximation, which is therefore consistent with screening. Note that $v(\mathbf{q})$ appearing in $D(\mathbf{q})$ is the *unscreened* interaction. It is therefore not permissible to do the screening first and then look for a CDW instability. Although this discussion is based on a single wavevector $\mathbf{Q}$, the arguments are based on the theory of linear response and hence are valid for general density modulations.

The noninteracting susceptibility $\chi_0(\mathbf{q})$ is negative. In general, there are $\mathbf{Q}$s such that $V_{\mathbf{Q}}$ (and hence $v(\mathbf{Q})$) is also negative, so that a CDW instability can occur. At half-filling, the Fermi surface is perfectly nested for $\mathbf{Q} = (0, 0, \frac{2\pi}{a})$, leading to a logarithmically divergent $\chi_0(\mathbf{Q})$. The metal is then unstable to a two-sublattice CDW, with a density modulation

$$\rho(\mathbf{r}) = \rho_0 \cos \mathbf{Q}\cdot\mathbf{r}, \tag{4}$$

for any value of $u \equiv -v(\mathbf{Q}) = U/2 - V_{\mathbf{Q}}$. For this $\mathbf{Q}$, $V_{\mathbf{Q}}$ is negative so that $u > 0$. The behaviour is essentially the same as in the bipartite simple cubic lattice [1]. In the absence of V, the CDW and s-wave superconductivity are degenerate. Inclusion of $V(r)$ stabilizes the CDW.

In the self-consistent Hartree approximation, the mean-field Hamiltonian has the form $H_{\rm MF} = H_0 - \sum_i \phi_i n_i$, where H_0 is the hopping part and

$$\phi_i = -\frac{U}{2}\rho(\mathbf{r}_i) + \sum_{j\neq i} V_{ij}\rho(\mathbf{r}_j) \tag{5}$$

is the Hartree potential and $\rho(\mathbf{r}_i) = \langle n_i\rangle - n_{\rm c}$ is the average electron density, to be determined self-consistently. Substitution of $\rho(\mathbf{r})$ from equations (4) in (5) leads to a Hartree potential of the same form: $\phi_i = u\rho_0 \cos(\mathbf{Q}\cdot\mathbf{r}_i)$ where, as before, $u = U/2 - V_{\mathbf{Q}}$. In the momentum space, the MF Hamiltonian becomes

$$H_{\rm MF} = \sum_{\mathbf{k}\sigma} \epsilon(\mathbf{k}) c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} - \frac{u\rho_0}{2}\sum_{\mathbf{k}\sigma}[c^{\dagger}_{\mathbf{k}+\mathbf{Q},\sigma} c_{\mathbf{k}\sigma} + \text{h.c.}], \tag{6}$$

where

$$\epsilon(\mathbf{k}) = -8t \cos\frac{k_x a}{2} \cos\frac{k_y a}{2} \cos\frac{k_z a}{2}$$

is the single-particle energy and a is the side of the cubic cell. In the two-sublattice case, $\mathbf{k}$ is paired with $\mathbf{k} + \mathbf{Q}$, and one has a 2×2 matrix problem in each $\mathbf{k}$ subspace. This leads to two bands with energy $E_\pm = \pm R$, with $R(\mathbf{k}) = (\epsilon^2(\mathbf{k}) + u^2\rho_0^2)^{1/2}$. At half-filling, the lower band is completely filled at $T = 0$, and the state is an insulator with a gap $2u\rho_0$.

The two-sublattice state continues to be a self-consistent solution for $n_c < 1$. But now the lower band is partially filled, and the system is a CDW metal. The ground state energy is given by: $E_G = E_{MF} + \frac{1}{2}u\rho_0^2$. The parameter ρ_0 is determined self-consistently from

$$\frac{1}{u} = \frac{1}{N}\sum_{\mathbf{k}} \frac{f(E(\mathbf{k}) - \mu) - f(E(\mathbf{k}) - \mu)}{R(\mathbf{k})}, \tag{7}$$

where f is the Fermi function, and μ is the chemical potential. We have solved the MF equations numerically for $n_c = 2/3$, with the result that the energy is larger than that of a different CDW state described below.

3. The CDW states at $n_c = 2/3$

3.1. Strong-coupling limit

To find a suitable CDW state it is useful to set $U = \infty$ (or, $t = 0$), which is the strong-coupling limit. The electrons form local pairs (hard-core bosons); however there is no kinetic energy, as neither bosons nor electrons can hop. Hence the Hartree approximation is exact in this limit. The problem is reduced to distributing pairs of density $n_c/2$ such that the interaction energy is a minimum. The contribution from the U term is the same for all configurations. Hence we need only to calculate the energy E_V corresponding to the repulsive part, $V(\mathbf{r})$.

At half-filling, this is exactly the problem of the ionic crystal [6] of alternating charges, which leads to the same two-sublattice structure: $\rho(\mathbf{r}) = \rho_0 \cos \mathbf{Q} \cdot \mathbf{r}$. The corresponding cohesive energy is

$$E_V = \frac{V_\mathbf{Q}\rho_0^2}{2N}\sum_i \cos^2(\mathbf{Q} \cdot \mathbf{r}_i). \tag{8}$$

Since $V_\mathbf{Q}$ is negative, so is E_V, whereas for the uniform state $E_V = 0$. Hence, by itself, $V(\mathbf{r})$ leads to the same CDW ground state.

Away from half-filling, the lowest-energy structure will be complex, particularly for incommensurate n_c. However, for $n_c = 2/3$, we can construct simple periodic arrays. Since the density of pairs is $1/3$, the natural period of a charge modulation is 3. Indeed, it is easy to see that, for $d = 1$, the minimum-energy configuration has every third site occupied by a pair: 02002002002.... For the bcc lattice, we consider a similar period-3 modulation in which every third plane perpendicular to the z-axis (i.e., located at $z = 3ma/2$, $m =$ integer) is occupied by pairs; the other two planes are unoccupied. This is a simple generalization of the two-sublattice state, in which every other plane is occupied.

Interestingly, this structure is also described by a single cosine: $\rho(\mathbf{r}) = \rho_0 \cos \mathbf{Q} \cdot \mathbf{r}$. But, now $\mathbf{Q} = (0, 0, \frac{4\pi}{3c})$, and $\rho_0 = 4/3$. Here we do not do a global search, but show that the chosen structure is energetically favoured over the two-sublattice CDW and the ordinary metal. Nonetheless, it is interesting to note that this is the simplest structure of period 3. It is locally stable (see below), and has no nearest-neighbour sites occupied, and thus is likely to have the lowest energy.

Using equation (8), we obtain $E_V = V_{\mathbf{Q}}\rho_0^2/4$, with $\rho_0 = 4/3$. Here, V_Q is the cosine transform of $V(\mathbf{r})$. Let us consider a $1/r$ potential:

$$V(\mathbf{r}) = V_{\rm nn}\frac{r_0}{r},$$

where $V_{\rm nn} = V(r_0)$, is the value of the potential at the nearest-neighbour distance $r_0 = a\sqrt{3}/2$, and sets the energy scale for the repulsive interaction. Then, $V_{\mathbf{Q}} = -\alpha V_{\rm nn}$, where α is a generalized Madelung constant:

$$\alpha(\mathbf{Q}) = -\sum_{\mathbf{r}\neq 0}\frac{\cos(\mathbf{Q}\cdot\mathbf{r})}{r/r_0}.$$

Evaluating the sum accurately, we obtain $\alpha \approx 1.440$, so that $E_V = -0.64V_{\rm nn}$.

It is interesting to compare with the two-sublattice case. Now, at $n_{\rm c} = 2/3$, there is no single configuration of this type. For comparison, we can simply take the results of the Hartree approximation with the choice of ρ_0 which gives the lowest energy. Since $n(\mathbf{r})$ is nonnegative, the largest value of ρ_0 is $2/3$. Also, in this case, $\alpha \approx 1.76$ [6], so that $E_V = -0.39V_{\rm nn}$. Therefore, the period-3 CDW has a lower energy.

On the other hand, if $V(\mathbf{r})$ is restricted to nearest neighbours, the two states have the same energy, and are degenerate with infinite number of other configurations, obtained, e.g., by moving an occupied layer up or down by one step. Then the system can lower its energy by constructing a CDW metal from a linear combination of such states.

It is useful to consider the elementary excitations for the three-sublattice state. The important one is obtained by moving a pair from i to an empty site j. The corresponding energy is given by

$$E_{\rm ex} = 4\alpha V_{\rm nn} - 4V_{ij},$$

where, $V_{ij} = V(\mathbf{r}_i - \mathbf{r}_j)$. Clearly this is lowest when i, j are nearest neighbours, with $E_{\rm ex} = 4(\alpha - 1)V_{\rm nn}$, which is positive since $\alpha > 1$. We have calculated other excitations, which are also found to be gapped—with a scale set by $V_{\rm nn}$. Single-electron excitations which are obtained by breaking a pair are also gapped; in this case the gap is larger, as one also has to add U to the energy. Hence, in the strong-coupling limit, the three-sublattice state is locally stable.

This analysis also gives us some insight into the behaviour of the model for large but finite $U \gg t$, when electrons remain paired as hard-core bosons. This limit is best understood in pseudo-spin formalism [7], described in terms an occupied ('up') and an empty ('down') site. The Hamiltonian is then reduced to a generalized antiferromagnetic Heisenberg Hamiltonian:

$$H = J\sum_{(i,j)}\mathbf{S}_i\cdot\mathbf{S}_j + 4\sum_{i,j}V_{ij}S_{iz}S_{jz}, \tag{9}$$

where $J = 4t^2/U$, and each bond i, j is to be counted once. Fixing the number of electrons (hence, bosons) is equivalent to fixing the z-component of magnetization: $M_z = \sum_i S_{iz}$, which is zero at half-filling and finite away from half-filling. The Ising (second) term describes the long-range repulsion between the bosons. The transverse (xy) part of the first term describes boson hopping. Bose condensation gives rise to superconductivity which appears as AF ordering in the transverse direction. The z-linear part of the J term represents a short-range repulsive interaction which, together with the long-range V term, favours a z-linear SDW corresponding to the CDW in the original model. In the absence of V, superconductivity wins away from half-filling (M_z finite). In this case, there is no energy scale at $T = 0$, and the wavefunctions are all independent of J. Therefore, the ground state remains superconducting as $U \to \infty$, i.e., as $J \to 0$.

The situation is different in the presence of V. Now, there is a new energy scale, $J/V_{\rm nn}$. At $U = \infty$ ($J = 0$), there is no hopping, and hence no metallic or superconducting behaviour. This is the strong-coupling limit, which has a CDW ground state. Furthermore, as shown above, elementary excitations that involve hopping of bosons are gapped. It follows that the CDW state is stable against the superconducting as well as the bosonic metal states—as long as J is small compared with the excitation energies $\sim V_{\rm nn}$. Now, by definition, $J \ll t \ll U$, and thus J is small in the entire large-U region. Hence, for reasonable values of $V_{\rm nn}$, the region of stability of the CDW state can be quite large. In fact it is larger than suggested by this simple analysis since the CDW gains additional energy from the J term as well. A more accurate analysis of the large-U limit is beyond the scope of the paper.

3.2. Hartree approximation

The local pairing picture does not apply for $U < W$, which is our main region of interest. In this case, we use the self-consistent Hartree approximation, with the mean-field Hamiltonian given by equation (7). For the three-sublattice CDW, each $\mathbf{k}$ is coupled to $\mathbf{k}+\mathbf{Q}$ and $\mathbf{k}-\mathbf{Q}$, which form a closed subspace. In each $\mathbf{k}$ subspace we have to diagonalize a 3×3 matrix of the form

$$\mathbf{M} = \begin{pmatrix} \epsilon(\mathbf{k}) & -\Delta & -\Delta \\ -\Delta & \epsilon(\mathbf{k}+\mathbf{Q}) & -\Delta \\ -\Delta & -\Delta & \epsilon(\mathbf{k}-\mathbf{Q}) \end{pmatrix}, \tag{10}$$

where $\Delta = u\rho_0/2$. This leads to three bands of energy $E_i(\mathbf{k})$, $i = 1, 2, 3$. The order parameter ρ_0 is determined self-consistently from

$$\rho_0 = \frac{4}{3N}\sum_{i\mathbf{k}}[\psi_{i1}(\mathbf{k})\psi_{i2}(\mathbf{k}) + \psi_{i2}(\mathbf{k})\psi_{i3}(\mathbf{k}) + \psi_{i3}(\mathbf{k})\psi_{i1}(\mathbf{k})]f(E_i(\mathbf{k}) - \mu), \tag{11}$$

where ψ_{ij} are the eigenvectors, with $i, j = 1, 2, 3$ and j corresponding to $\mathbf{k}$, $\mathbf{k}+\mathbf{Q}$ and $\mathbf{k}-\mathbf{Q}$, respectively. The ground state energy is then given by

$$E_{\rm MF} = \frac{2}{3N}\sum_{i\mathbf{k}} E_i(\mathbf{k}) f(E_i(\mathbf{k}) - \mu) + \frac{1}{4}u\rho_0^2.$$

We have solved the MF equations numerically with very good accuracy (using up to 10^6 k points). The results can be expressed in terms of the parameter $u = U/2 + \alpha V_{\rm nn}$. At low u, the state is an ordinary metal, the CDW appears only above $u_1 = 2.27t$. As shown in figure 1, the corresponding value for the two-sublattice case is $u_1 \approx 3.1t$, which is much higher. Note, however, that the parameter u is not the same in the two cases for the same values of the interaction parameters, U and $V_{\rm nn}$, since α is different. To correct for this we can eliminate $V_{\rm nn}$ and use the calculated value of α to obtain

$$u(3) = 0.82u(2) + 0.09U,$$

where $u(n)$ denotes u for the n-sublattice state. Let $E(n)$ be the corresponding energy. Then, $E(2)$ at $u(2)$ should be compared with $E(3)$ at $u(3)$. This can be done by shifting the two-sublattice curve to the left from $u(2)$ to $u(3)$ in figure 1. Clearly, for a given $u(2)$, the smallest $u(3)$ occurs for $U = 0$, and the corresponding $E(3)$ is the highest. The shifted $E(2)$ curve is shown as the intermediate line in figure 1. Note that $E(3)$ is still lower, which shows that the three-sublattice state always wins over the two-sublattice state.

In the three-sublattice case, Δ becomes nonzero above u_1. As shown in figure 2, the transition is continuous. The state however remains metallic (CDW) up to $u_2 = 2.78t$. This can be seen from figures 3(a) and (b), where we have plotted the density of states (DOS). There

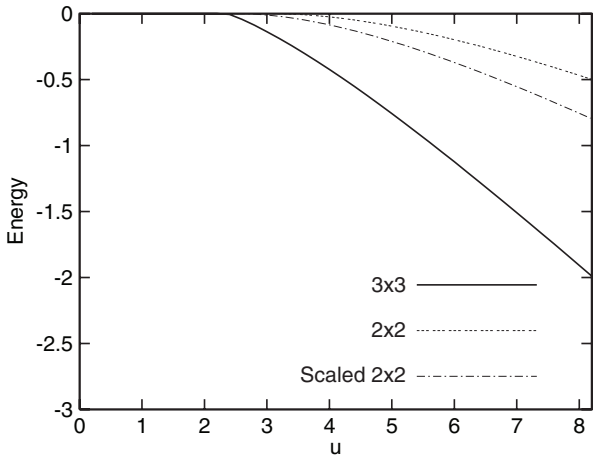

Figure 1. Ground state energy per site for the two-sublattice CDW state ($E(2)$—dashed line) and the three-sublattice CDW state ($E(3)$ solid line) as a function of $u = U/2 + \alpha V_{\rm nn}$. The energies are measured relative to the energy of the ordinary metallic (no CDW) state. In each case the CDW state appears above a critical u where its energy becomes negative. Note that u is not the same for the two cases (for given U and $V_{\rm nn}$). The intermediate line (dash–dotted) shows $E(2)$, shifted to the left appropriately (see the text), so that two energies can be compared directly. Clearly, $E(3)$ is still lower.

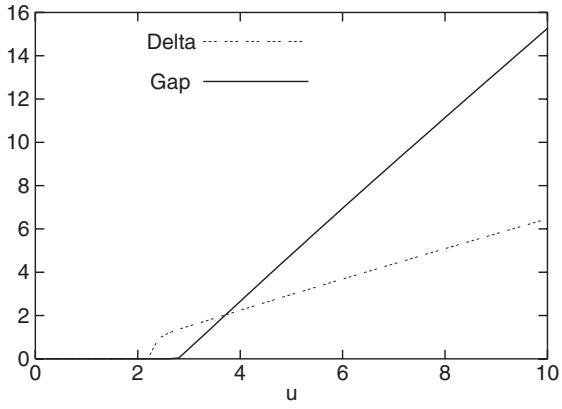

Figure 2. The direct gap parameter Δ and the indirect gap parameter $E_{\rm gap}$ as functions of u ($t = 1$). The CDW semimetal ($\Delta \neq 0$, $E_{\rm gap} = 0$) exists between $u_1 = 2.27t$ and $u_2 = 2.78t$. For $u < u_1$, the state is an ordinary metal and for $u > u_2$ it is a CDW insulator with Δ and $E_{\rm gap}$ both nonzero.

is a large depletion of the DOS near the Fermi energy, but no true gap (figure 3(a)). For $u > u_2$, a gap develops (figure 3(b)) and the system becomes an insulator. The gap and the parameter Δ are shown in figure 2. At large u, both are linear in u.

The CDW metallic state that exists between u_1 and u_2 is different from the one in the two-sublattice case. In the latter state, there is always a gap between the two bands, and the lower band becomes partially filled away from half-filling. In the period-3 case, there is also a direct gap (at the same **k** points). But the bands overlap along some **k** directions. The maximum energy of the lowest band is larger than the minimum energy (which is at a different **k** point) of the intermediate band, as shown in figure 4. Above u_2, the two bands finally separate,

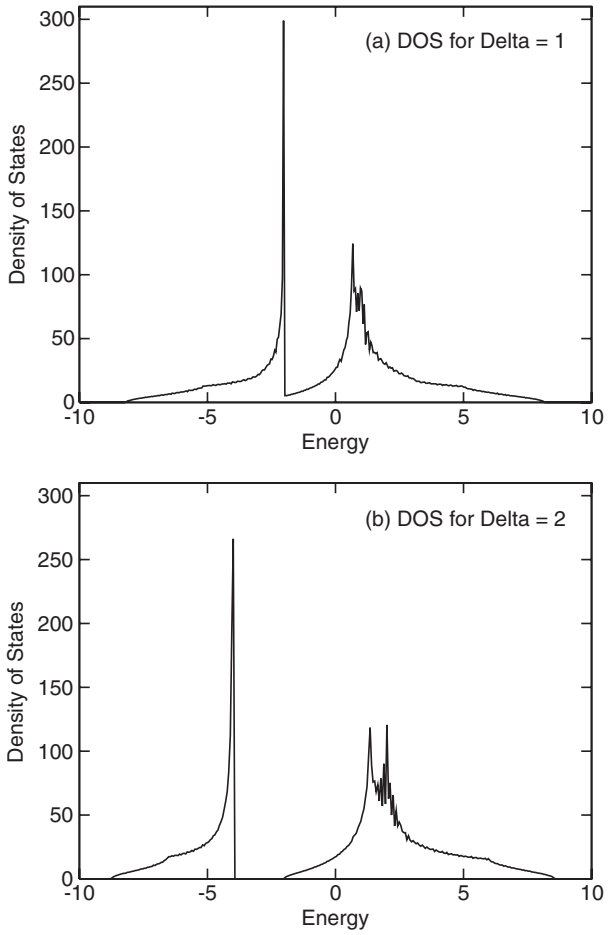

Figure 3. Density of states for the three-sublattice CDW semimetal (a) and the CDW insulator (b). In the semimetallic case, Δ is small, and the bands overlap, so that there is no true gap. In the insulator, Δ is larger, and a gap opens. The lower band is completely filled, and the upper two bands are empty.

leading to an indirect gap insulator. A similar situation occurs in fcc lattice at half-filling [2]. Such a state is in fact a semimetal, characterized by an interesting fermi surface topology and an effective carrier density which decreases with increasing u. Also, at a given u, the carrier density and the shape of the Fermi surface is expected to vary rapidly with temperature as Δ decreases increasing T. This would lead to unusual transport properties.

Our main result is the existence of a CDW insulator far from half-filling. Also, the CDW appears at a rather small value of u since $u_1/W = 0.142$, and $u_2/W = 0.174$, where $W = 16t$, is the noninteracting bandwidth. Hence, the insulating state exists over a fairly large region of the parameter space.

The insulating state above u_2 is different from the CDW insulator found at half-filling. The latter is characterized by a single gap ($=2\Delta$) in the one-particle spectrum, whereas the CDW insulator of period 3 is characterized by two one-particle gaps, a direct gap $\sim\Delta$ which will show up in optical conductivity, and an indirect gap $E_{\rm gap} = E_{1,\rm min} - E_{2,\rm max}$, which sets the scale for the thermodynamic properties.

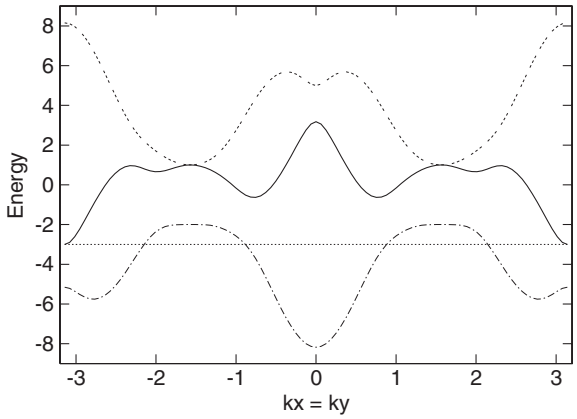

Figure 4. Energy bands $E_i(\mathbf{k})$ along the direction $k_x = k_y$, with $k_z = 0$ in the semimetallic region. The lowest band is separated from other two by a direct gap (at the same $\mathbf{k}$), but overlaps with the intermediate band, as its maximum is clearly above the minimum of the latter (at a different $\mathbf{k}$). As Δ increases, these two bands separate, leading to an indirect gap insulator.

4. Discussions

To summarize, the combination of long-range Coulomb repulsion and lattice effects can stabilize a CDW insulator far from half-filling in the negative-U Hubbard model. Although our calculation is for a particular density, we expect similar results for other lattices and densities, particularly since there are no Fermi surface nesting effects. However, because the CDW is crystal on top of crystal, commensurability will always be important. Because of this, solving the Hartree problem for a general density will be difficult.

In the present case, the fact that a CDW insulator exists at such a small u has serious consequences for superconductivity, which has not been considered in our mean-field analysis. A calculation including both CDW and superconducting order parameters is not feasible since long-range repulsion makes the numerical problem intractable. However, the central issue—namely, the stability of the insulting state relative to the superconductor—can be easily understood on the basis of earlier work on the short-range model.

Previously, we have carried out a detailed mean-field analysis including both superconducting and CDW order parameters for the fcc lattice at half-filling [2], but with a nearest-neighbour repulsion. In that case, the phase diagram in the absence of superconductivity is similar to the one found here at $n_c = 2/3$: an indirect gap CDW insulator is separated from an ordinary metal by a CDW semimetal. Thus the problem is essentially the same. Besides the ordinary metal, the CDW semimetal has a superconducting ground state over some part of the parameter space where superconductivity and CDW coexists. However, the insulating phase is found to be stable against superconductivity except very close to the metal–insulator phase boundary or for very small V (compared with W and U), as one would expect. The insulator is also stable in the large-U limit ($U \gg W$) for relatively small V/W. We expect a similar behaviour in the present case also. In fact, long-range repulsion is likely to be even more helpful in stabilizing the insulator.

The reason for the relative stability of the CDW insulator is that in the presence of V, the excitation spectrum in the pairing channel is gapped, except when V is small. Note that the CDW is favoured by both the U and V terms. However, only the U term favours superconductivity. In addition, V term opposes superconductivity since, in the

superconducting state, it contributes an energy

$$\tfrac{1}{2}\sum_{i\neq j} V(\mathbf{r}_i - \mathbf{r}_j)|\langle c_{j\downarrow}c_{i\uparrow}\rangle|^2,$$

which is always positive. Hence, for fixed U, the CDW always wins if V is large enough. This argument is general. For example, Taraphder *et al* calculated the energy spectrum in the pairing channel for the cubic crystal with a short-range V. They found a collective excitation ('cooperon') which has charge 2 and spin zero [1], and which appears in the gap of the insulator. In the pseudo-spin language, the cooperons are the analogues of spin waves. They are gapped because their is no pseudo-spin symmetry in the presence of V. While the calculations in [2, 1] are based on the short-range model, it is obvious that long-range repulsion would make superconductivity even less favoured.

Similarly, in the large-U (i.e., local pairing) limit, the superconducting (and, metallic) energy scale is set by $J = 4t^2/U$, whereas the CDW energy scale is set by 4V. As discussed earlier, the relevant excitations in the CDW insulating phase are again gapped. Hence, as long as V is large enough compared with J, superconductivity cannot win.

Experimentally the appearance of the insulator is relevant for bismuthates, although in that case the lattice is cubic. Another potential candidate is the alkali fulleride A_4C_{60} (A = alkali atom), which is a nonmagnetic insulator [8]. It has a body-centre-tetragonal structure which is bipartite, and the electron density is nominally $2/3$. In principle, the insulator can be a CDW. It is however not clear that the one-band negative-U Hubbard model can be applied to this system.

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