Phase Separation Close to the Density-Driven Mott Transition in the Hubbard-Holstein Model

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The density-driven Mott transition is studied by means of dynamical mean-field theory in the Hubbard-Holstein model, where the Hubbard term leading to the Mott transition is supplemented by an electron-phonon (e-ph) term. We show that an intermediate e-ph coupling leads to a first-order transition at $T = 0$, which is accompanied by a phase separation between a metal and an insulator. The compressibility in the metallic phase is substantially enhanced. At quite larger values of the coupling, a polaronic phase emerges coexisting with a nonpolaronic metal.

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For a long time it has been known that strong electron-electron (e-e) interactions can drive a metallic system insulating [1]. The correlation caused metal-insulator transition (MIT), usually known as the Mott transition, occurs in several compounds such as V_2O_3 and $CaTiO_3$, and is often theoretically investigated in the framework of the Hubbard Hamiltonian. However, although the latter certainly captures the fundamental properties of correlated electronic systems, it lacks the lattice degrees of freedom, which can play a crucial role in many respects. Therefore, not only is the interplay between the electronic correlation and the electron-phonon (e-ph) interaction a relevant issue in general [2,3], but interesting specific effects can arise from it, such as in doped fullerenes, where a substantial e-ph coupling associated with Jahn-Teller distortion can be synergetic with the large electron-electron interaction to produce the observed rather large superconducting temperatures [4]. Many experimental results also point toward the important role of phonons in the strongly correlated cuprates. In this light, it has also been proposed that the e-ph coupling could induce charge instabilities in the cuprates [2,5] thereby relating the interplay between strong e-e and e-ph interactions to the issue of electronic phase separation (PS), stripe formation, and to the isotopic dependence of the pseudogap crossover temperatures in these materials [6]. This connection between e-ph coupling and charge instabilities suggest that lattice degrees of freedom may greatly influence the thermodynamical stability of a strongly correlated electron system. The present work precisely focuses on the role of the e-ph coupling in modifying or emphasizing the physical properties of electrons close to their Mott insulating status. Specifically, we study the role of phonons in changing the order of the density-driven MIT and in inducing PS close to the transition.

From the technical point of view we exploit the great progress made in this field with the dynamical mean-field theory (DMFT), which emerged as the first theoretical tool able to give a complete characterization of the Mott transition [7]. This technique, which becomes exact in the infinite coordination limit [8], is in fact able to treat with the same accuracy both the metallic and the insulating phase, and it has been successfully applied to the Mott transition in the Hubbard model. Moreover, DMFT allows us to treat on the same footing the e-e and e-ph interactions, which is crucial to our study. The main drawback of this approach is the momentum independence of the self-energy and the neglect of nonlocal interactions. This is not expected to introduce significant biases when the interactions are local, such as in the model we study.

One of the most striking features of the DMFT treatment of the Mott transition is that both the interaction-driven transition at half-filling [9,10] and the density-driven transition [11] are characterized by the coexistence of a solution in some region of the parameters. In the latter case, which we address in this Letter, there is a region of chemical potentials, delimited by two curves $\mu_{c1}(U)$ and $\mu_{c2}(U)$, in which a metallic solution with $n \neq 1$ and an insulating one with $n = 1$ coexist [11]. Naturally, the stable solution is the one that minimizes the grand-canonical free energy. At $T = 0$ the metal is stable in the whole coexistence region and is continuously connected to the insulator [11]. Moreover, the metal joins onto the insulator with a finite slope in the n-μ curve; i.e., it has a finite compressibility $\kappa = \partial n / \partial \mu$ [11,12]. At finite temperature the energetic balance gets more involved, and a first-order transition occurs along an intermediate line between μ_{c1} and μ_{c2}, leading to PS between the two phases [13].

In this work we study how this scenario is modified by the inclusion of a short-range interaction with phonons. We consider the e-ph interaction in its simplest model realization, the Holstein molecular crystal model, in which tight-binding electrons interact with local modes of constant frequency [14]. The Hamiltonian is

$$H = -t\sum_{\langle i,j\rangle} c_i^\dagger c_j + \text{H.c.} + U\sum_i n_{i\uparrow} n_{i\downarrow} - g\sum_i n_i (a_i + a_i^\dagger) + \omega_0 \sum_i a_i^\dagger a_i, \qquad (1)$$

where c_i ($c_i^\dagger$) and a_i ($a_i^\dagger$) are, respectively, destruction (creation) operators for fermions and for local vibrations of frequency ω_0 on site i, t is the hopping amplitude, and g is an e-ph coupling.

Previous studies have identified two different dimensionless e-ph couplings: $\lambda = 2g^2/\omega_0 t$, which measures the energetic convenience to form a bound state and $\alpha = g/\omega_0$, which controls the number of excited phonons. The relevance of each coupling depends on the adiabatic ratio $\gamma = \omega_0/t$: If γ is small λ is the control parameter, while for large γ the physics is mainly controlled by α [15]. Here we work with $\gamma = 0.2$, and therefore we use λ to measure the strength of the e-ph coupling.

In DMFT, the lattice model is mapped onto an impurity problem subject to a self-consistency condition, which contains all the information about the lattice. In the case of the Hubbard-Holstein model we have an Anderson impurity with a local phonon on the impurity site. If we work in the Bethe lattice of half-bandwidth t, the self-consistency enforcing the DMFT solution is given by

$$\frac{t^2}{4} G(i\omega_n) = \sum_k \frac{V_k^2}{i\omega_n - \epsilon_k}, \tag{2}$$

where ϵ_k and V_k are the energies and the hybridization parameters of the Anderson impurity model. We use exact diagonalization to solve the impurity model [16]. The technique consists in restricting the sum in Eq. (2) to a finite and small number of levels $N_s - 1$. The discretized model is then solved using the Lanczos method, which allows us to compute the Green's function at $T = 0$. The method converges rapidly as a function of N_s, and just a few levels are enough to achieve convergence. Here we mainly present the results for $N_s = 10$, having checked in specific cases that no measurable change occurs in the physical quantities by increasing N_s [17].

As anticipated above, we discuss how the density-driven Mott transition is affected by the e-ph interaction. The main trend, which can be anticipated on intuitive grounds, is that the e-ph interaction favors the insulating phase with respect to the metal. We note that in Ref. [13] the same effect is obtained by raising the temperature, because in that case the insulating state has a larger entropy. Here, when the e-ph interaction is turned on in the metallic state, two competing effects take place. On one side, the electron gains potential energy if the lattice is distorted, while on the other side, the effective mass is enhanced, leading to a loss in kinetic energy. As a result of this competition, the electrons cannot completely exploit the e-ph coupling to lower the energy. On the other hand, the electrons in the Mott insulator are already localized, so that they can gain potential energy by coupling with phonons without losing kinetic energy. This simple argument already tells us that the energetic balance between the two phases will be changed in favor of the insulator. For the same reasons we can also expect the region of μ where the insulator exists to be larger, i.e., that μ_{c1} decreases with increasing λ [18].

We notice that simplified approaches, such as the one based on Lang-Firsov and squeezing transformations [19], also suggest that the insulating behavior may be favored by the electron-phonon interaction. Within this approach, the Hubbard-Holstein model is transformed into a Hubbard model with reduced hopping, in which the insulating behavior is clearly favored with respect to the case where the electron-phonon interaction is absent.

These naive arguments are confirmed and based on solid ground by the DMFT calculations. We have studied in detail both the solutions as a function of λ for values of U/t larger than the critical value $U/t \simeq 3$ for the Mott transition at half-filling [7]. In Fig. 1 we plot the density as a function of the chemical potential for $U/t = 10$ and various values of λ. For $\lambda = 0$ (not shown) we recover the known results for the Hubbard model, with two solutions in a really small μ interval, and the metal is stable in the whole coexistence region, as it can be checked by comparing the grand-canonical potentials $\Omega = E - \mu N$ of the two phases. In the insets of the various panels we plot the difference $\Omega_{\text{met}} - \Omega_{\text{ins}}$. For small values of λ we observe a slight increase of the coexistence region, but the metal stays at lower energy than the insulator (see upper panel in Fig. 1). For $\lambda > 1$ the two energy curves cross and the grand-canonical potential of the insulator becomes lower than the one of the metal at some value of the chemical potential (in the middle panel we show the result for $\lambda = 2$), leading to a jump of the occupation number by continuously varying the chemical potential.

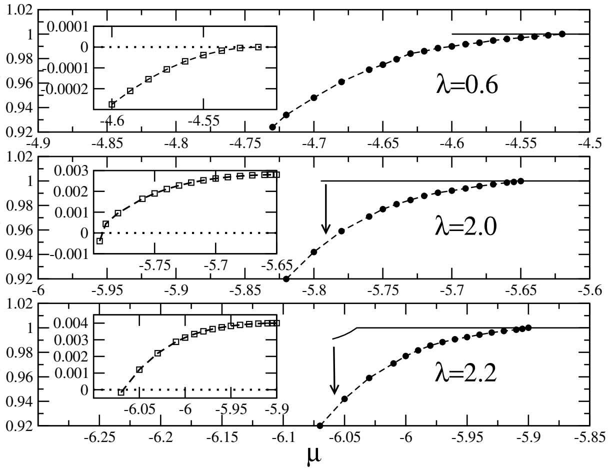

FIG. 1. The n-μ curves for $U/t = 10$ for $\lambda = 0.6, 2, 2.2$. Solid circles joined by a dashed line denote the metallic solutions, and the solid line marks the insulating one. The insets show the difference between the grand canonical potentials of the two phases $\Delta\Omega = \Omega_{\text{met}} - \Omega_{\text{ins}}$. In the two lower panels this quantity is positive, signaling the stability of the insulator. An arrow marks the transition point on the n-μ curves.

The transition becomes therefore of first order, and the chemical potential has a plateau with respect to the density. As a result, the system is not stable in the interval of density between which the jump occurs, and it undergoes PS between the insulating solution with $n = 1$ and a metallic solution with some density $n_{\mathrm{ps}}(\lambda)$. n_{ps} is found to be a decreasing function of λ. In Fig. 2 we show the PS region for $U/t = 10$.

If we further increase the coupling, the effect of the e-ph interaction becomes even more dramatic. In the case of $U/t = 10$, and $\lambda \simeq 2.2$, the insulating solution continuously evolves into a phase with a density slightly smaller than 1. Also for this coupling the insulator is energetically favored in the coexistence region (see the inset of the lowest panel in Fig. 1). An inspection to other physical quantities allows us to better characterize this strong-coupling phase as a polaronic state. In fact, if we compute the phonon displacement distribution function $P(X) = \langle\psi_0|X\rangle\langle X|\psi_0\rangle$, the evolution from the $n = 1$ insulator to this state is accompanied by a change from a unimodal distribution of the phonon displacement to a bimodal distribution characteristic of the polaronic state. On the other hand, the metallic solution does not display polaronic features for these values of λ and so the PS in this region takes place between a polaronic insulator and a nonpolaronic metal.

Our DMFT results show that the Hubbard-Holstein model displays PS close to half-filling, due to the first-order phase transition between the insulator and the metal. Even if PS occurs close to half-filling, just like in the large-N treatment of the same model, the present result is physically quite different. In Ref. [20] a divergent compressibility was indeed found within the metallic phase. Here this instability is prevented by the stabilization of the insulator. Nevertheless, we can follow the metallic solution in the regime where it is metastable and check whether the e-ph interaction may lead to a divergence or at least to a sizeable enhancement of the charge compressibility.

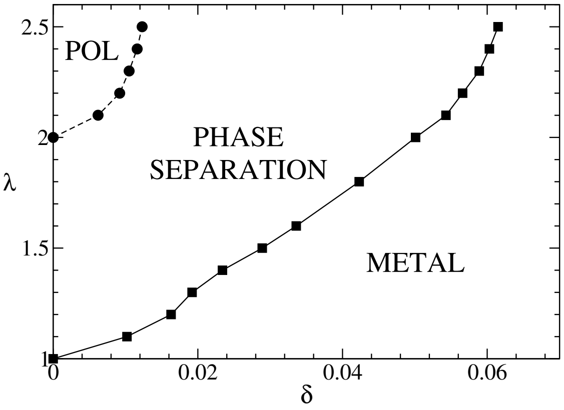

FIG. 2. The phase diagram for $U/t = 10$ as a function of doping δ ($n = 1 - \delta$) and λ.

In the infinite-U limit, the large-N approach has shown that the charge compressibility is well represented by a simple random-phase-approximation-like formula,

$$\kappa(\lambda) = \frac{2N^*}{1 + (F_0^S)_e + (F_0^S)_{\mathrm{ph}}}, \qquad (3)$$

where $\kappa(\lambda)$ is the compressibility for a given value of the e-ph coupling λ, N^* is the quasiparticle density of states per spin at the Fermi level, $(F_0^S)_{e(\mathrm{ph})}$ is the electronic (phononic) contribution to the symmetric Landau amplitude, and $(F_0^S)_{\mathrm{ph}} = -4N^* g^2/\omega_0$, which differs from our λ for the presence of the quasiparticle density of states (DOS) instead of the real DOS. From (3) it is easy to derive $\kappa(\lambda)/\kappa(0) = 1/[1 - \lambda\kappa(0)t]$, in which all the correlation effects are contained in the purely electronic compressibility calculated at $\lambda = 0$. This implies that, if the electronic compressibility is large, a small or moderate e-ph coupling is sufficient to make the system unstable in the charge channel.

Coming back to the results shown in Fig. 1, the n-μ curves of the metallic solutions have a small finite slope when they approach $n = 1$. However, if we consider the compressibility at fixed density as a function of λ, it turns out that the e-ph interaction is substantially increasing the compressibility. In Fig. 3 we present the inverse of the normalized compressibility as a function of λ at $U/t = 5$ for different values of the electron density, ranging from $n = 0.70$ to $n = 0.95$. An enhancement of $\kappa(\lambda)/\kappa(0)$ that varies from almost 6 in the $n = 0.70$ case to around 3 in the $n = 0.95$ case corresponds to the decreasing linear behavior of all the curves seen in the figure. We also calculated the values of the renormalization factor Z which is inversely proportional to the quasiparticle density of states. Since we found that Z varies much less than

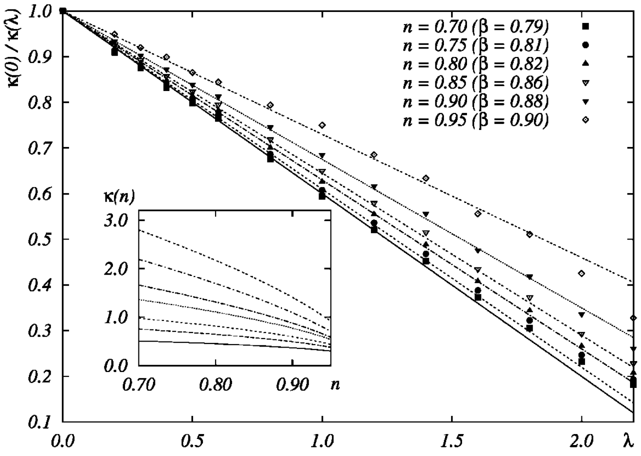

FIG. 3. Normalized inverse compressibility $[\kappa(\lambda)/\kappa(0)]^{-1}$ as a function of λ at $U/t = 5$ and for different densities, compared with the fit described in the text. Inset: Compressibility vs filling for various values of $\lambda = 0.0, 0.8, 1.2, 1.6, 1.8, 2.0, 2.2$ (from the bottom to the top).

κ as a function of λ, such an enhancement of the compressibility cannot be due to mass renormalization alone. This also indicates that, within the Landau Fermi liquid approach, the total F_0^S is expected to be renormalized by the e-ph interaction like in the large-N calculations. We extract information on this renormalization of the Landau amplitudes by simply assuming that a relation similar to Eq. (3) holds also for the DMFT solution. We therefore fitted the DMFT data with the relation

$$\frac{\kappa(\lambda)}{\kappa(0)} = \frac{1}{1 - \beta\lambda\kappa(0)t}, \tag{4}$$

where β is the only fitting parameter. The results of this fit, together with the values of $\kappa(\lambda)$ in the inset, are shown in Fig. 3. For almost the whole range of densities the fit is extremely accurate up to $\lambda = 2.2$, with just a small correction to the ideal large-N result $\beta = 1$. The closer we get to half-filling the more β approaches 1, even if at $n = 0.95$ the strong-coupling data display some fluctuations. The values of β are reported in the figure.

In light of the ability of the simple Eq. (4) to describe the way the e-ph interaction modifies the scenario determined by electron correlation, the small values of λ needed to obtain a divergent compressibility in the large-N approach [2,20] can be related to the smaller value of the electronic F_0^S found in the large-N with respect to the one found in the present DMFT approach.

Our findings could be related to some experimental observations. First of all, the presence of a (phonon-induced) PS close to a Mott insulating phase is suggestive of a chemical potential plateaux inferred from photoemission experiments in underdoped cuprates [21]. The formation of doping-induced metallic states with a chemical potential slowly varying with doping inside a Mott gap was also recently deduced from tunneling experiments [22]. In the same experiment, the small mixing of the metallic (superconducting) phase with the antiferromagnetic insulating one indicates that a spatial separation of the two phases can have small interfaces, suggesting that the presence of long-range Coulomb forces can easily induce small-scale (presumably stripe-like) domain formation. We also remarkably found that for rather large values of the e-ph coupling ($\lambda > 2$ in the $U = 10t$ case) a PS can occur between an insulator away from half-filling with a finite density of polarons and a good metal with nearly free carriers. Also this situation of coexisting (but spatially separated) polarons and free carriers could find a realization in underdoped cuprates [23].

In conclusion, we have studied the effect of the e-ph interaction on the density-driven Mott transition for large values of U. The e-ph interaction favors the insulating solution with respect to the metal. This effect, besides a quantitative modification of the coexistence region between the two solutions, determines a first-order transition between an insulator and a metal at some value of the chemical potential μ_c and to a PS between the Mott insulator with $n = 1$ and a metal with a finite density $n \neq 1$. This PS occurs for $\lambda \simeq 1$. For substantially larger values of the e-ph coupling a polaron crossover occurs very close to half-filling and PS takes place between this polaronic state and a nonpolaronic metal.

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