Charge-fluctuations in lightly hole-doped cuprates: effect of vertex corrections

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Identification of the electronic state that appears upon doping a Mott insulator is important to understand the physics of cuprate high-temperature superconductors. Recent scanning tunneling microscopy of cuprates provides evidence that a charge-ordered state emerges before the superconducting state upon doping the parent compound. We study this phenomenon by computing the charge response function of the Hubbard model including frequency-dependent local vertex corrections that satisfy the compressibility sum-rule. We find that upon approaching the Mott phase from the overdoped side, the charge fluctuations at wave vectors connecting hot spots are suppressed much faster than at the other wave-vectors. It leads to a momentum dependence of the dressed charge susceptibility that is very different from either the bare susceptibility or from the susceptibility obtained from the random phase approximation. We also find that the paramagnetic lightly hole-doped Mott phase at finite-temperature is unstable to charge ordering only at zero wave-vector, confirming the results previously obtained from the compressibility. Charge order is driven by the frequency-dependent scattering processes that induce an attractive particle-hole interaction at large interaction strength and small doping.

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I. INTRODUCTION

Immediately following the discovery of cuprate high-temperature superconductors, and the suggestion by Anderson of the importance of the proximate Mott insulating phase, the study of hole-doped Mott insulators became a central theme of research. Early Hartree-Fock studies of the hole-doped Mott insulators provide evidence of charge order, often accompanied by spin order, so-called stripes, which survives even in presence of frustrating long-range interactions accompanied by spin order, so-called stripes, doped Mott insulators provide evidence of charge order, of the theme of research. Early Hartree-Fock studies of the hole-doped Mott insulators provide evidence of charge order, of the stripe phases in La1−xNd0.4SrxCuO4, the experimental neutron scattering discovery of charge-fluctuations in lightly hole-doped cuprates: effect of vertex corrections

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teraction values comparable with the electronic bandwidth of the system and becomes more prominent at larger interactions. A real charge instability occurs only for interactions larger than what is required to drive a Mott transition at half-filling. This indicates that the charge instability is an instability of the doped Mott insulator, and not of the Slater antiferromagnet.

We present the model and method in section II. The single-site DMFT results for the compressibility are recovered in section III. Section IV describes the unusual structure of the frequency-dependent vertex in the charge channel, in particular that it can become attractive. This sets the stage for the calculation of the physically observable susceptibilities in section V. After concluding remarks, details of the calculation for the susceptibilities in various approximations are given in appendix A. Appendix B shows the results for the compressibility at lower interaction strength and higher temperature. It is shown in detail in appendix C that the compressibility obtained from single-site DMFT is identical to that deduced from the zero-wave vector density-density response function at zero Matsubara frequency, in other words that the compressibility sum-rule is satisfied.

II. MODEL AND METHOD

We consider the Hubbard model on the square lattice with $U$ the onsite Coulomb interaction and appropriate hopping parameters for cuprates, i.e., $t' = -0.3t$, $t'' = 0.2t$ the second and third nearest-neighbor hopping amplitudes, $t$ being the nearest-neighbor hopping $t$. The Hamiltonian is solved using DMFT and the exact diagonalization (ED) method. The DMFT(ED) algorithm is also used to compute the local part of the irreducible vertex function. The lattice response function can then be computed as described in appendix A. Namely, the DMFT self-energy is included in the propagators and the vertices are obtained from four point functions on the self-consistent impurity. The resulting correlation functions are the building blocks of the ladder dynamical vertex approximation. We focus on the charge channel because the charge and magnetic channels are independent in this approach. As in experiment, we see the charge order as an instability of the Mott insulator that appears independently of magnetic or superconducting long-range order. Appendix A contains more details on the method.

III. DMFT COMPRESSIBILITY

We first present the DMFT results, which are in qualitative agreement with previous DMFT results obtained at $t' = t'' = 0$. In the DMFT framework, the possibility of a first order phase transition can be assessed by direct calculation of the local charge compressibility, defined as $\kappa = (1/n^2)\partial n/\partial\mu$, as a function of hole-doping $p$. A vanishing compressibility characterizes the Mott insulator, while a divergence indicates a second-order transition and a discontinuity a first-order transition. Let $U_c$ denote the DMFT critical interaction beyond which a Mott insulating phase appears at half-filling.

The main panel of Fig. 1 illustrates the compressibility as a function of hole density for $U = 12t > U_c$ at various temperatures. At $T = 0.1t$ (in blue), $\partial n/\partial\mu$ continuously decreases upon approaching half-filling indicating the suppression of charge fluctuations. However, at a lower temperature, $T = 0.04t$ (in red), after an initial decrease, the compressibility increases and exhibits a peak around $p \approx 0.01$ before dropping to zero at half-filling. This increase suggests the proximity to the critical end-point of a first-order transition at a locus $(T_c, p_c)$. That first-order transition becomes visible at a lower temperature, $T = 0.02t$ (in green) where the compressibility is discontinuous. The inset in Fig. 1 shows the hole density as a function of chemical potential at $T = 0.02t$. This illustrates clearly that there is a value of the chemical potential for which there are two possible values of the hole doping, as expected in the coexistence region of a first-order transition. The critical doping, $p_c$, does not depend on $U > U_c$ sensitively but $T_c$ decreases upon increasing $U$. The phase transition is interpreted as a uniform phase separation ($q = 0$) since it occurs in the compressibility. The question arises whether this actually occurs at $q = 0$ or if a more sophisticated calculation could reveal a finite wave-vector instability at a higher temperature. The answer to this question demands a calculation of the density-density response function at finite wave vector.

![Fig. 1. Compressibility, $\partial n/\partial\mu$, as a function of hole density, $p$, for $U = 12t$, $t' = -0.3t$, $t'' = 0.2t$ at various temperatures, either computed directly within the DMFT framework, labeled 1P in the figure and discussed in section III, or computed from the lattice density-density correlation function, indicated as 2P and discussed in section IV. Inset: The hole density as a function of chemical potential for $T = 0.02t$ has a jump for a chemical potential $\mu_c \approx 4.8$ with a discontinuous first-order derivative at that chemical potential, implying the discontinuous compressibility seen in the main panel.](image-url)
IV. DYNAMICAL VERTEX

No charge instability is predicted by approximations with a static irreducible vertex, such as the random phase approximation (see appendix A 2). Hence we go beyond these approximations to obtain a reliable calculation for $U > U_c$. In a diagrammatic approach, the full vertex function can be decomposed into the irreducible vertex function and the generalized bubble susceptibility (see appendix A). One can diagonalize the irreducible vertex in spin space to exploit the conservation of spin in two-body scattering processes and rewrite it in the charge and magnetic channels, defined as $\Gamma^{c(m),\text{irr}} = \Gamma^{\uparrow\uparrow,\text{irr}} + (-1)\Gamma^{\uparrow\downarrow,\text{irr}}$. In a normal system, there is a range and a characteristic relaxation time, beyond which $\Gamma^{c(m),\text{irr}}$ becomes negligible. Hence, the spatially local part of the irreducible vertex function, $\Gamma^{\text{irr}}_{\text{loc}}(\nu_n)$, is the dominant part. DMFT gives an accurate estimate of its dependence on three frequencies, $\nu_n$ the center of mass bosonic frequency and $\omega_m, \omega_{m'}$ the fermionic frequencies.

In an interacting system, the electron spectral function demonstrates a coherent (quasi-particle) peak and high energy incoherent (Hubbard) satellites. Upon increasing $U$, the spectral weight is transferred from the coherent peak to incoherent satellites. Thus, the irreducible vertex function contains two sets of scattering processes: (i) the scattering of the coherent part with itself (ii) the scattering of the incoherent part with itself and with the coherent part. The latter contribution is smooth and possibly featureless at low $U$, but increases and requires frequency dependence upon increasing interaction strength, leading to the nontrivial frequency dependence of $\Gamma^{c,\text{irr}}$ for $U \geq W$.

Figure 2 displays the real and imaginary parts of the irreducible local vertex in the density channel at zero bosonic frequency, $\nu_n$, for doping level $p = 0.11$ and $U = 6t < W$ where $W$ is the bandwidth. The $\nu_n = 0$ component measures the amplitude of scattering processes that occur at all time scales. For $U = 6t$, the system is in the perturbative regime and the behavior of $\Gamma^{c,\text{irr}}_{\text{loc}}(\nu_n = 0)|_{\omega_m,\omega_{m'}}$ can be understood from low-order diagrams. The real part of the vertex function for $|\omega_m| = \omega_0$, and large $|\omega_{m'}|$ or vice versa approaches its static limit, which is $U$ in the Hubbard model. Here, $\omega_0$ denotes the lowest fermionic Matsubara frequency.

Furthermore, the real part of the $\Gamma^{c,\text{irr}}_{\text{loc}}(\nu_n = 0)|_{\omega_m,\omega_{m'}}$ is repulsive and is large on the primary diagonal, i.e., when $\omega_m = \omega_{m'}$. This enhancement is caused by p-h scattering processes involving the emission and reabsorption of pairs of fluctuations when the induced p-h excitations live on the Fermi surface.

On the other hand, the scattering rate along the secondary diagonal $\omega_m = -\omega_{m'}$ is due to particle-particle (p-p) scattering processes with energies $\omega + \nu$ and $\omega' + \nu$ and opposite spin in the Hubbard model. These scattering processes have a large amplitude for total energies at the Fermi level, i.e., $\omega' = -\omega - \nu$ with a maximum at $\nu = 0$. The amplitude of the p-p scattering is smaller than the p-h scattering in the repulsive Hubbard model and large $U$. The right-hand panel of Fig. 2 shows the imaginary part of the irreducible vertex function. The absolute value of the imaginary part is proportional to the p-h asymmetry. It increases along the primary diagonal and changes sign between positive and negative frequencies. The secondary diagonal peak is missing in the imaginary part. For interaction strengths smaller than the bandwidth, $U < W$, these characteristics of the $\Gamma^{c,\text{irr}}_{\text{loc}}(\nu_n = 0)|_{\omega_m,\omega_{m'}}$ are common for all doping levels.

In the non-perturbative regime at larger interaction strengths, $U \geq W$, the irreducible vertex in the charge channel gradually changes, as illustrated in Fig. 3. Although the previously mentioned characteristics of $\Gamma^{c,\text{irr}}_{\text{loc}}(\nu_n = 0)|_{\omega_m,\omega_{m'}}$ are generally maintained in the high energy region, the low-frequency behavior strongly depends on the interaction strength and doping level. In the low doping region, the real part of $\Gamma^{c,\text{irr}}_{\text{loc}}(\nu_n = 0)|_{\omega_m,\omega_{m'}}$ around the primary diagonal begins to show at intermediate fermionic frequencies a sign change between low and high frequencies: this means that, surprisingly, the emission and reabsorption of p-h pairs causes an effective interaction which is attractive for intermediate frequencies (see left-hand panel of the Fig. 3 for $U = 8t$). At larger $U = 12t$, this behavior also includes the very low frequency region, as can be seen from the right-hand panel of Fig. 3. Indeed, a behavior different from the perturbation theory prediction occurs on an energy scale of order $U$ at low temperatures. Upon increasing $T$, the non-perturbative low-
frequency region shrinks. The change in $\Gamma^{c,irr}_{loc}$ for $U > W$ could be a manifestation of the gradual replacement of the perturbative branch of the self-energy with the non-perturbative one at the physical self-energy$^{87}$. 

V. CHARGE SUSCEPTIBILITY

From the irreducible vertex in the charge channel, we can compute the charge susceptibility. Then, instead of differentiating $n(\mu)$, we compute the compressibility from the density-density correlation function, $\chi^c_{ph}$, using the compressibility sum rule (with $K \equiv (k, \omega_m)$)

$$\partial n/\partial \mu = 2 \lim_{\nu \to 0} (1/\beta)^2 \sum_{KK'} |\chi^c_{ph}(q, \nu_n)|_{K,K'}.$$  

(1)

The derivative of density with respect to chemical potential and the right-hand side of the above equation evaluated with the DMFT local self-energy and irreducible vertex function should give same results as long as the perturbation expansion for the dressed susceptibility is valid. However, several studies have shown that calculating physical quantities such as double occupancy, kinetic or potential energy, directly from the impurity model or from the two-particle response functions yield different results$^{88,89}$. The density of the self-consistent impurity depends on $\mu$ explicitly and implicitly through the hybridization function, $\Delta(\mu)$, hence, the impurity compressibility is given by $(\partial n/\partial \mu)_\Delta + (\partial n/\partial \Delta)_\mu (\partial \Delta/\partial \mu)$.

Going back to our original question regarding whether or not the system undergoes a uniform phase separation, the momentum dependence of the dressed susceptibility in the charge channel at a hole doping slightly larger than the critical doping is shown in the right-hand panel of Fig. 4 for $U = 12t$ at several temperatures. As can be seen, upon reducing $T$ the charge susceptibility at $q = 0$ grows faster than at the other wave-vectors, indicating a charge instability at lower $T$ at this momentum. This confirms previous results from compressibility studies$^{87}$.

Finally, it is worth mentioning that the largest eigenvalue of the (dimensionless) matrix

$$(1/\beta^2) \sum_{\omega_m^{''}} |\chi^c_{ph}(q, 0)|_{\omega_m^{''}, \omega_m}$$

with $\chi^c_{ph}(Q)_{\omega_m, \omega_m^{''}} \equiv (1/\beta)^2 \sum_{k,k'} |\chi^c_{ph}(Q)|_{K,K'}$ is called the Stoner factor. When it is a real number, it measures the distance from a continuous phase transition. With a static vertex function, as in the RPA approximation, the Stoner factors in the magnetic and charge channels are purely real. However, the eigenvalue problem that needs to be solved to search for an instability is a non-Hermitian eigenvalue problem in general. Nevertheless, the susceptibilities are always real. While our numerical results show that the Stoner factor in the magnetic channel is always real and increases with increasing $U$, eventually approaching unity, in the charge channel it is real only for $U < W$. For $U > W$, the eigenvalues appear mostly in complex conjugate pairs, with large real and imaginary parts, in particular at low doping. Therefore, in this regime, the Stoner factor is not well defined and cannot serve as a probe of an instability.
VI. CONCLUDING REMARKS

In summary, we have calculated the dressed charge susceptibility of strongly correlated metals to investigate possible charge instabilities. We verified that even for strong interactions, the compressibility sum-rule is satisfied. Our most important result is that the momentum-dependent dressed charge susceptibility of a doped Mott insulator has a completely different peak structure than what an RPA analysis would predict. This is a consequence of the complicated frequency dependence of the irreducible vertex function. Indeed, the charge susceptibilities with wave vectors connecting hot spot decreases faster than at the other wave vectors upon approaching the Mott phase. Furthermore, the phase transition of a lightly hole-doped Mott insulator occurs at the 0 = 0 wave-vector in agreement with the DMFT compressibility studies. Although the single-band Hubbard model does not show any charge instability with a finite momentum, including more degrees of freedom, such as Oxygen p orbitals, may change the physics. In Ref. 56, finite momentum instabilities were found. It may also be that the q = 0 instability corresponds to a more realistic model to an intra unit cell charge pattern.

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Appendix A: Response functions

The response function of an interacting system can be decomposed into the bubble response and vertex corrections. The bubble response describes independent, but interaction-renormalized, propagation of a particle-hole (p-h) excitation created by the field, while the vertex corrections introduce changes in the response functions due to scattering processes in which propagating particle and hole exchange multiple real or virtual p-h excitations. Denoting the amplitude of all these scattering events by the full vertex function, defined as\( \Gamma_{\text{full}} = \Gamma_{\text{charge}} + (-)^{L-1} \Gamma_{\text{magnetic}} \) for charge and magnetic channels, the dressed susceptibility is given by

\[
[\chi_{\text{ph}}^{\epsilon/m}(Q)]_{K,K'} = [\chi_{\text{ph}}^{0}(Q)]_{K,K'} - \frac{1}{N^2 \beta^2} \sum_{K_1,K_2} \left[ \chi_{\text{ph}}^{0}(Q) \right]_{K_1,K_1} \Gamma^{\epsilon/m,f}(Q) \left[ \chi_{\text{ph}}^{0}(Q) \right]_{K_2,K_2},
\]

where the bubble susceptibility is

\[
[\chi_{\text{ph}}^{0}(Q)]_{K,K'} = -(N\beta)G(K + Q)G(K)\delta_{K,K'}. \tag{A2}
\]

Here, \( G(K) \) is the dressed particle propagator, \( K \equiv (k, i\omega_n) \) denotes momentum/energy three-vectors (the lattice is two-dimensional), \( N \) is number of \( k \)-points and \( \beta = 1/(k_BT) \).

In Eq. (A1) the bosonic variable \( Q \equiv (q, i\nu_n) \) is always inactive in the multipole expansion, or conserved during the scatterings within each channel. Eq. (A1) is the common part of the response to an external field and solely depends on the electronic structure of the system. An observable response function, on the other hand, is obtained by closing the external legs of Eq. (A1) using appropriate oscillator matrix elements, \( O(Q) \) and \( O(-Q) \), which depends on the field wave-vector and frequency. The oscillator matrix elements for the charge channel are given by (\( SU(2) \) symmetric case)

\[
O_{R_1,R_2}(q) = \frac{1}{\sqrt{V}} \int d\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} \phi_{R_1}(\mathbf{r})\phi_{R_2}(\mathbf{r}), \tag{A3}
\]

where \( \phi_{R_1}(\mathbf{r}) \) the atomic orbital residing at the lattice point \( R_1 \).

In a correlation-driven phase transition, it is the full vertex function, \( \Gamma^{\epsilon/m,f}(Q) \), that causes a singular or discontinuous response. It consists of all connected diagrams. Some of these diagrams are two-particle fully irreducible. Other diagrams are reducible, i.e., cutting two Green function lines separates the diagram into two pieces. Indeed, each diagram is either fully irreducible or reducible in exactly one channel (particle-hole \( ph \), particle-hole transversal \( ph \), and particle-particle \( pp \)), so

\[
[\Gamma^{\epsilon/m,f}(Q)]_{K,K'} = [\Lambda^{\epsilon/m}(Q)]_{K,K'} + [\Phi_{ph}^{\epsilon/m}(Q)]_{K,K'} + [\Phi_{pp}^{\epsilon/m}(Q)]_{K,K'}. \tag{A4}
\]

Here, \( \Lambda^{\epsilon/m} \) and \( \Phi_{i}^{\epsilon/m} \) denote, respectively, the fully irreducible vertex in all channels and reducible vertex in \( i \) channel. Moreover, one can define the irreducible diagrams in a certain channel \( i \) as \( \Gamma^{\epsilon/m,f}_{i,i} = \Gamma^{\epsilon/m,f}_{i,i,\text{irr}} + \Phi_{i}^{\epsilon/m} \). For example, for the \( ph \) channel

\[
[\Gamma^{\epsilon/m,f}_{ph,i}](Q)]_{K,K'} = [\Lambda^{\epsilon/m}(Q)]_{K,K'} + [\Phi_{ph}^{\epsilon/m}(Q)]_{K,K'} + [\Phi_{pp}^{\epsilon/m}(Q)]_{K,K'}. \tag{A5}
\]

Having the irreducible vertex in a given channel \( i \), the reducible one can be obtained from the Bethe-Salpeter equa-
lations (BSE) as\(^{93}\)

\[
\left[\Gamma^{c/m,f}(Q)\right]_{K,K'} = \left[\Gamma^{c/m,irr}_{ph}(Q)\right]_{K,K'} - \frac{1}{N^2 b^2} \sum_{K_1,K_2} \left[\chi^0_{ph}(Q)\right]_{K_1,K_2} \left[\Gamma^{c,m,irr}_{loc}(Q)\right]_{K_1,K_2'.}
\]

(A6)

The irreducible vertex function can be evaluated using various quantum many-body approaches.

In general, \(\left[\Gamma^{c/m,irr}(Q)\right]_{K,K'}\) depends on the transferred momentum/frequency in a scattering process, \(Q\), and on the incoming momentum/frequency variables. The out-going variables are determined by conservation laws. \(\left[\Gamma^{c/m,irr}(Q)\right]_{K,K'}\) describes the irreducible interaction of the two elementary excitations. The spatially local part of the irreducible vertex function in channel \(l\), \(\left[\Gamma^{c/m,irr}_{loc,l}(\nu_n)\right]_{\omega_m,\omega_m'}\), can be calculated in framework of the DMFT approximation\(^{83,84}\). A common approximation is substituting \(\Gamma^{c/m,irr}_{loc}(\nu_n)\) by \(\Gamma^{c/m,irr}_{loc}(\nu_n)\) and neglecting the non-local part\(^{60}\). This allows to perform the summations over momentum of the internal legs in Eq. (A6), leading to a full vertex function that satisfies a similar equation but with the bubble susceptibility replaced by

\[
[\chi^0_{ph}(Q)]_{\omega_m,\omega_m'} = \left(\frac{1}{N}\right)^2 \sum_{k,k'} \left[\chi^0_{ph}(Q)\right]_{K,K'}.
\]

(A7)

Hence, the resulting full vertex depends on three frequencies but only one momentum (transferred momentum). This approximation captures the dynamics of the screening effects, which plays a significant role in correlated electron systems.\(^{96}\)

Focusing on the particle-hole channel, note that due to the fully irreducible local vertex, the momentum dependence of the reducible vertices in the transverse particle-hole, \(\Phi^{\perp}_{\mu\nu'}\), and particle-particle channels, \(\Phi^{\parallel}_{\mu
u}\), are neglected (see Eq. (A5)). On the other hand, the dependence of the reducible \(ph\) vertex on the transferred momentum is taken into account.

1. Numerical considerations

We employed the exact diagonalization (ED) technique to solve the DMFT equations and to calculate the irreducible impurity vertices. The latter calculation is very expensive and grows very fast with the number of bath levels in the ED method. Furthermore, calculating the dressed susceptibility requires \(\left[\Gamma^{c/m,irr}_{loc}(\nu_n)\right]_{\omega_m,\omega_m'}\), calculated on a large number of fermionic Matsubara frequencies. For instance, for compressibility calculations, we took 512 positive frequencies. Hence, we only consider three bath levels, \(n_b\), to calculate the irreducible impurity vertices. We checked that the results do not change when increasing the number of bath level by performing calculations with five bath levels, \(n_b = 5\), for some interaction strengths and doping levels, albeit on a much smaller frequency range. For instance, Fig. 5 demonstrates the dressed impurity susceptibility in the charge channel, \(\left[\chi^{loc}_{\nu_n}(\nu_n = 0)\right]_{\omega_m,\omega_m'}\), at zero bosonic frequency and \(\omega_m = \omega_0 = \pi/\beta\) as a function of \(\omega_m\), calculated with \(n_b = 3\) and \(n_b = 5\). As one can see the difference between the two calculations is negligible. The DMFT calculation of compressibility, presented in Fig. 1, on the other hand, are done using five bath levels.

2. Random phase approximation

Calculations with a dynamical irreducible vertex function, as in the ladder dynamical vertex approximation, differ from those done in RPA with a static vertex. In RPA, the irreducible vertex is approximated with a static, though screened, interaction which is smaller than the bare one and remains repulsive. It is parametrized by a screened Hubbard interaction \(U_s\). With a static vertex, the summation over internal frequencies can be done and hence in RPA the internal bubble susceptibilities are replaced by

\[
\chi^{0,ph}_{RPA}(Q) = \left(\frac{1}{\beta}\right)^2 \sum_{m,m'} \left[\chi^0_{ph}(Q)\right]_{\omega_m,\omega_m'}.
\]

(A8)

This makes the instability eigenvalue problem a real-symmetric one.\(^{86}\) Furthermore, while \(\left[\chi^{0,ph}_{ph}(Q)\right]_{\omega_m,\omega_m'}\) has both real and imaginary parts, the RPA susceptibility, \(\chi^{0,ph}_{RPA}(Q)\), is purely real.

To gain insight into the problem, consider the prediction of RPA when dressed propagators are used. The left panels of Fig. 6 display the locus of maxima of the spectral weight at zero frequency, \(A(k,\omega = 0)\), for two hole-doping values: one in at large doping \(p = 0.18\) (red) and one at small doping \(p = 0.04\) (blue) for \(U = 12t\) and \(T = 0.1t\). Note that the depicted \(A(k,\omega = 0)\) should not be interpreted as the Fermi surface (FS) since that concept is strictly defined only for a Fermi liquid at zero temperature. At large doping, the spectral weights intersect the antiferromagnetic (AF) Brillouin zone (BZ) at the so-called hot spots, i.e., regions of FS where the
FIG. 6. Spectral weight at zero frequency (left panel) for hole doping levels $p = 0.18$ (red), and $p = 0.04$ (blue) for $U = 12t$, $t' = -0.3t$, $t'' = 0.2t$ at $T = 0.1t$ on the square lattice. The dash-line shows the antiferromagnetic Brillouin zone. The wave vectors $Q_{1-4}$ connect hot spots when $p = 0.18$. The corresponding RPA bubble susceptibility is on the right-hand panel.

probability of Umklapp and $(\pi, \pi)$ scattering events is appreciable. The bubble susceptibility calculated in RPA shows peaks at the wave-vectors connecting hot spots. At smaller $U$, such as $U = 8t$, $A(k, \omega = 0)$ crosses the AF-BZ for both doping values. At $U = 12t$, however, the hot spots disappear at low doping due to the combination of interactions, which make the spectrum less coherent, and finite temperature. This influences the RPA bubble susceptibilities, as shown on the right-hand panel of Fig. 6.

In RPA, the charge susceptibility is small and the leading instability occurs in the magnetic channel with wave-vectors $(1, 1 - \delta)\pi/a$ and $(1 - \delta, 1)\pi/a$, where $\delta$ is small and vanishes at half-filling.

**Appendix B: Compressibility at lower interaction strength and higher temperature**

The transition to the charge ordered state, discussed in the main text, is absent at lower interaction strengths. Fig. 7 shows the compressibility for $U = 8t$ and $T = 0.1t$ (top panel) and $U = 12t$ and $T = 0.4t$ (bottom panel) computed from the density and with respect to chemical potential and from the uniform density-density correlation function, labeled by $\chi_{0,ph}$. Both methods again agree very well at all doping values considered here and they do not show a tendency towards phase separation.

**Appendix C: Thermodynamic derivative of the density in the DMFT approximation**

In this section we show that the two approaches we have employed to calculate the compressibility – namely the derivative of the density with respect to the chemical potential and the zero-frequency zero-momentum lattice charge susceptibility – are equivalent within a local self-energy, local vertex approximation, as long as the Luttinger-Ward functional remains single-valued. For an exact calculation, the two results would obviously be equal, as required by a thermodynamic sum-rule.

1. **Preliminary considerations**

The density is given by

$$n = \frac{1}{\beta} \sum_{\omega_n, \sigma} e^{-\omega_n 0^-} g_\sigma(i\omega_n)$$

$$= \frac{1}{\beta N} \sum_{k\omega_m, \sigma} e^{-i\omega_m 0^-} \frac{1}{i\omega_m + \mu - \epsilon_k - \Sigma_\sigma(i\omega_m)}, \quad (C1)$$

where the first line is for the impurity and the second line is for the lattice. Since the DMFT self-consistency equation is,

$$g_\sigma(i\omega_n) = \frac{1}{N} \sum_k G_{k\sigma}(i\omega_n), \quad (C2)$$

the density on the impurity is equal to the density on the lattice for all values of the chemical potential and $(\partial n/\partial \mu)_T$ will be identical in the two cases.

However we can also obtain $(\partial n/\partial \mu)_T$ from the susceptibility on the lattice, using the irreducible particle-hole vertex. That quantity is calculated from correlation functions on the impurity by inverting the Bethe-Salpeter equation.
Remembering that the chemical potential is space-independent and imaginary-time independent, the susceptibility that is needed on the lattice to compute \( \frac{\partial F}{\partial \mu} \) is a special case of the susceptibility considered in the previous section, namely

\[
\frac{\partial G_{k\sigma}(i\omega_n)}{\partial \mu} = -G_{k\sigma}(i\omega_n)G_{k\sigma}(i\omega_n) + G_{k\sigma}(i\omega_n)G_{k\sigma}(i\omega_n) \\
\times [\Sigma_{k\sigma}^{\text{irr}}(\nu_n = 0)]_{\omega,\omega'} \frac{1}{N} \sum_{k',\sigma'} \frac{\partial G_{k'\sigma'}(i\omega_{n'})}{\partial \mu}.
\] (C3)

where \([\Sigma_{k\sigma}^{\text{irr}}(\nu_n = 0)]_{\omega,\omega'}\) is the irreducible particle-hole vertex, which we is momentum independent since it comes from a calculation on the impurity. Summing over wave vectors on both sides of the equation and using the notation

\[
G_{\sigma}^{\text{loc}}(i\omega_n) = \frac{1}{N} \sum_k G_{k\sigma}(i\omega_n).
\] (C4)

this gives the following closed equation that can be solved by considering \(\omega_m, \omega'_m\) as matrix indices and inverting:

\[
\frac{\partial G_{\sigma}^{\text{loc}}(i\omega_n)}{\partial \mu} = -\frac{1}{N} \sum_k G_{k\sigma}(i\omega_n)G_{k\sigma}(i\omega_n) + \frac{1}{N} \sum_k G_{k\sigma}(i\omega_n)G_{k\sigma}(i\omega_n) \\
\times [\Sigma_{k\sigma}^{\text{irr}}(\nu_n = 0)]_{\omega,\omega'} \frac{\partial G_{\sigma}^{\text{loc}}(i\omega_{n'})}{\partial \mu}.
\] (C5)

If instead of the above approach, we compute the derivative of the impurity Green function \(g_{\sigma}(i\omega_n)\) (taking into account the fact that the hybridization function depends on \(\mu\) as well) and use the self-consistency equation Eq. (C2), we find the same equation as above with the following two replacements

\[
[\Sigma_{\sigma\sigma'}^{\text{irr}}(\nu_n = 0)]_{\omega,\omega'} \rightarrow \delta_{\sigma\sigma'} \frac{\partial G_{\sigma}^{\text{loc}}(i\omega_{n'})}{\partial \mu},
\] (C6)

and

\[
G_{\sigma}^{\text{loc}}(i\omega_n) \rightarrow g_{\sigma}(i\omega_n).
\] (C7)

\(G_{\sigma}^{\text{loc}}(i\omega_n)\) and \(g_{\sigma}(i\omega_n)\) are in fact equal because of the self-consistency equation Eq. (C2). By contrast, for Eq. (C6) to be an equality, we have to take the functional derivative with respect to \(g_{\sigma}(i\omega_{n'})\) on the physical branch when \(\Sigma_{\sigma}(i\omega_n)\) is not single-valued. That \(\Sigma_{\sigma}(i\omega_n)\) can be multi-valued has been documented\(^9\). In addition there are two other possible difficulties. First, the vertex can diverge at points where two branches of the solution cross\(^9\), a problem that is avoided for the case we consider. Second, the impurity Green function depends also on the self-consistent value of the hybridization function. That self-consistency condition can lead to phase transitions, such as the Mott transition at half-filling. In that case, precursors of the phase transition can appear in the vertex function\(^9\). A more careful look at the separate effect of the hybridization function is given in the following section.

2. Detailed derivation

The first derivative of the free energy is related to the Green’s function as follows

\[
n = -\frac{\partial F}{\partial \mu} = \frac{1}{N \beta} \text{Tr} G = \frac{1}{N \beta} \sum_{\sigma} G_{\sigma}(r, r +),\n\] (C8)

where \(F\) denotes free energy density and \(\mu\) the chemical potential. The second derivative of the free energy with respect to the chemical potential gives the electron compressibility.

To obtain an integral equation for the charge susceptibility, we apply a perturbation \(\phi(1') = -\mu \delta_{\eta r r'}\delta_{\tau r''} (\text{where we have used the compact notation } 1 \equiv (r, \tau)\) and calculate the response in the DMFT approximation. The dimensionless thermodynamic derivative of interest is (with \((1', 1'') = (r_1, \tau_1'))

\[
\frac{\partial G_{\sigma}(1')}{\partial \phi(2')} = -G_{\sigma}(13) \frac{\partial G_{\sigma}^{-1}(33'; \phi)}{\partial \phi(2')},
\] (C9)

where we used the identity \(G_{\sigma}(13; \phi)G_{\sigma}^{-1}(31'; \phi) = \delta_{11'}\) and a summation over repeated indices is assumed.

The propagator has the following form

\[
G_{\sigma}(1'; \phi) = -[\partial_{\tau} + H_0 - \phi + \Sigma_{\sigma}(G(\phi))]_{11'}^{-1},
\] (C10)

where \(\Sigma\) is the self-energy and the inverse should be understood as a matrix inversion in space and time coordinates. The field couples to the electron density and therefore it appears only on diagonal elements of Eq. (C10). The inverse propagator depends on the field explicitly and implicitly through the dependence of the self-energy on the propagator. From Eq. (C9) and Eq. (C10), one can see that the explicit field dependence contribution at the derivative is given by

\[
-G_{\sigma}(12)G_{\sigma}(2'1),
\] (C11)

where we used the identity \(\partial G_{\sigma}^{-1}(33'; \phi)/\partial \phi(2') = \delta_{33} \delta_{r r'}\).

In the DMFT approximation, the self-energy is fully local, i.e., \(\Sigma_{\sigma}^{\text{DMFT}}(1') = \Sigma_{\sigma}(1')\delta_{\eta r r'}\). In other words, the DMFT self-energy is only a functional of the local propagator \(\Sigma_{\sigma}^{\text{DMFT}} = \Sigma_{\sigma}^{\text{DMFT}}(G_{\text{loc}}(\phi))\), where \(G_{\text{loc}}\) denotes the local propagator \(G_{\text{loc}}(r, r +) = (1/N) \sum_{\sigma} G_{\sigma}(r, r +)\). Employing the fact that the DMFT self-energy is a functional of \(G_{\text{loc}}\), the derivative of the self-energy with respect to the field can be written as

\[
-G_{\sigma}(12)G_{\sigma}(2'1),
\] (C11)
\[
G_\sigma(13) \frac{\delta_{\tau \tau'} \partial \Sigma^{DMFT}_\sigma (33'; \phi)}{\partial \phi(22')} G_\sigma(3'1') = G_\sigma(13) \frac{\partial \Sigma^{DMFT}_\sigma (r_3 \tau_3, r_3 \tau_3'; \phi)}{\partial \phi(22')} G_\sigma(3'1')
\]

\[
= G_\sigma(13) \frac{\partial \Sigma^{DMFT}_\sigma (r_3 \tau_3, r_3 \tau_3'; \phi)}{\partial \phi(22')} G_\sigma(3'1') \frac{\partial G_{\text{loc}, \sigma'} (\tau_4, \tau_4')}{\partial \phi(22')},
\]  

(C12)

where

\[
\frac{\partial \Sigma^{DMFT}_\sigma (r_3 \tau_3, r_3 \tau_3'; \phi)}{\partial G_{\text{loc}, \sigma'} (\tau_4, \tau_4')} \equiv \Gamma^{\text{irr}}_{\sigma, \sigma'; \sigma'} (\tau_3 \tau_3'; \tau_4 \tau_4').
\]  

(C13)

Diagrammatically, \(\Gamma^{\text{irr}}_{\sigma, \sigma'; \sigma'}\) is obtained by removing one internal line from the self-energy in all possible ways. The complete equation for the susceptibility then takes the form

\[
\frac{\partial G_\sigma (11'; \phi)}{\partial \phi(22')} = -G_\sigma (12) G_\sigma (2'1') + G_\sigma (13) \frac{\partial G_{\text{loc}, \sigma'} (\tau_3 \tau_3', \tau_4 \tau_4')}{\partial \phi(22')},
\]  

(C14)

for every Matsubara frequency. We leave the two imaginary times \(1\) and \(1'\) free in Eq. (C15), but since the derivative we are interested in is for \(\phi\) equal to the chemical potential, which is independent of imaginary time, the derivation goes through if the self-consistency Eq. (C15) depends only on imaginary-time difference. Note that the inverse should be understood as a matrix inversion in imaginary time coordinates (indices are not bold anymore). Furthermore, we have \(g_\sigma (12) g^{-1}_\sigma (21') = \delta_{11'}\).

The impurity Green’s function depends on \(\phi\) explicitly and implicitly through the hybridization function. Then the variation of the impurity density with respect to the field is given by (with \(1' = 1\))

\[
\frac{\partial g_\sigma (11')}{\partial \phi(22')} \Delta + \frac{\partial g_\sigma (11')}{\partial \Delta (33')} \phi \frac{\partial \Delta (33')}{\partial \phi(22')}
\]

(C17)

Using the definition of the impurity Green’s function, one can easily show that

\[
\frac{\partial g_\sigma (11')}{\partial \phi(22')} = \chi_{\text{loc}, \sigma}(11'; 22') - \chi_{\text{loc}, \sigma}(11'; 33') \phi \frac{\partial \Delta (33')}{\partial \phi(22')} 
\]

(C18)

where \(\chi_{\text{loc}, \sigma}(11'; 22') = g_\sigma (12) g_\sigma (2'1') \delta_{\sigma \sigma'}\). Note that the relevant part of the above expression is the part with \(2 = 2'\) since \(\phi\) is diagonal.
Note that all imaginary time $3, 3'$ must be considered.

One can iterate Eq. (C18) and Eq. (C19) to find the corresponding dressed susceptibilities. The susceptibility obtained from Eq. (C18) describes the response of a non-self consistent impurity model to a change in the chemical potential. It is a physical response and therefore positive definite at a stable state. On the other hand, the susceptibility obtained from the Eq. (C19) does not describe any physical response and therefore it is not necessarily positive definite.

The self-energy and the hybridization function are functionals of the Green’s function. For strong interactions, these functionals change from a non-perturbative functional at low frequency to a perturbative functional at high frequency, i.e., $\Sigma \equiv \Sigma_{\text{per}}[g]$ or $\Sigma \equiv \Sigma_{\text{non-per}}[g]$. This causes an ambiguity in defining the vertex functions in Eq. (C18) and Eq. (C19) when one frequency is on the perturbative branch of the self-energy (or hybridization function) and the other frequency is on the non-perturbative branch. Nevertheless, our numerical verification of the compressibility sum-rule confirms that Eq. (C18) and Eq. (C19) remain valid, at least in the range of parameters considered here.

Further progress requires evaluating the derivative of the hybridization function with respect to the field. This can be found from the self-consistency condition, i.e.,

$$[F_\sigma(\phi, \Delta(\phi))]|_{11'} \equiv g_\sigma(11'; \phi) + \frac{1}{N} \sum_k [H_0(k) - g^{-1}_\sigma(\phi) - \Delta_\sigma(\phi)]|_{11'}^0 = 0, \quad (C20)$$

where we define $F_\sigma(\phi, \Delta(\phi))$ for convenience. The variation of the above equation with respect to the field is

$$\frac{\partial F_\sigma(11')}{\partial \phi(22')}|_\Delta + \frac{\partial F_\sigma(11')}{\partial \Delta(33')}|_\phi \frac{\partial \Delta(33')}{\partial \phi(22')} = 0, \quad (C21)$$

which gives

$$\frac{\partial \Delta(33')}{\partial \phi(22')} = - \left[ \frac{\partial F_\sigma(33')}{\partial \phi(22')} \right]^{-1} \sum_k G_{k\sigma}(13)G^{-1}_{\sigma}(34) \frac{\partial g_\sigma(44')}{\partial \phi(22')}|_\Delta \frac{\partial \Delta(33')}{\partial \phi(22')}|_\Delta, \quad (C22)$$

where we assume $\partial F/\partial \Delta$ is invertible. One the other word, we are assuming that $\partial g/\partial \Delta$ is finite (see the discussion after Eq. (C19)). At the vicinity of a Mott phase $F(\Delta)$ may go through an extremum with zero derivative, breaking down the above assumption.

The derivative of $F_\sigma(\phi, \Delta(\phi))$ with respect to the field at constant hybridization function can be written as follow, using Eq. (C18),

$$\frac{\partial F_\sigma(11')}{\partial \phi(22')}|_\Delta = \left[ \frac{\partial \Delta(33')}{\partial \phi(22')} \right]^{-1} \sum_k G_{k\sigma}(13)G^{-1}_{\sigma}(34) \frac{\partial g_\sigma(44')}{\partial \phi(22')}|_\Delta \frac{\partial \Delta(33')}{\partial \phi(22')}|_\Delta, \quad (C23)$$

where

$$\left[ X_{\sigma\sigma'}(q = 0) \right]|_{11'} = (-1/N) \sum_k G_{k\sigma}(13)G_{k\sigma'}(31')\delta_{\sigma\sigma'}. \quad (C24)$$

The derivative of the $F_\sigma(\phi, \Delta(\phi))$ with respect to the hybridization function at constant field is

$$\frac{\partial F_\sigma(11')}{\partial \Delta(33')}|_\phi = \left[ \frac{\partial \Delta(33')}{\partial \phi(22')} \right]^{-1} \sum_k G_{k\sigma}(13)G^{-1}_{\sigma}(34) \frac{\partial g_\sigma(44')}{\partial \phi(22')}|_\Delta \frac{\partial \Delta(33')}{\partial \phi(22')}|_\Delta, \quad (C23)$$

where

$$\left[ X_{\sigma\sigma'}(q = 0) \right]|_{11'} = (-1/N) \sum_k G_{k\sigma}(13)G_{k\sigma'}(31')\delta_{\sigma\sigma'}. \quad (C24)$$

The derivative of the $F_\sigma(\phi, \Delta(\phi))$ with respect to the hybridization function at constant field is

$$\frac{\partial F_\sigma(11')}{\partial \Delta(33')}|_\phi = \left[ \frac{\partial \Delta(33')}{\partial \phi(22')} \right]^{-1} \sum_k G_{k\sigma}(13)G^{-1}_{\sigma}(34) \frac{\partial g_\sigma(44')}{\partial \phi(22')}|_\Delta \frac{\partial \Delta(33')}{\partial \phi(22')}|_\Delta, \quad (C23)$$

where

$$\left[ X_{\sigma\sigma'}(q = 0) \right]|_{11'} = (-1/N) \sum_k G_{k\sigma}(13)G_{k\sigma'}(31')\delta_{\sigma\sigma'}. \quad (C24)$$

The derivative of the $F_\sigma(\phi, \Delta(\phi))$ with respect to the hybridization function at constant field is

$$\frac{\partial F_\sigma(11')}{\partial \Delta(33')}|_\phi = \left[ \frac{\partial \Delta(33')}{\partial \phi(22')} \right]^{-1} \sum_k G_{k\sigma}(13)G^{-1}_{\sigma}(34) \frac{\partial g_\sigma(44')}{\partial \phi(22')}|_\Delta \frac{\partial \Delta(33')}{\partial \phi(22')}|_\Delta, \quad (C23)$$

where

$$\left[ X_{\sigma\sigma'}(q = 0) \right]|_{11'} = (-1/N) \sum_k G_{k\sigma}(13)G_{k\sigma'}(31')\delta_{\sigma\sigma'}. \quad (C24)$$
where we have used Eq. (C19).

To show that the Eq. (C17) for $\partial n/\partial \mu$ on the impurity is identical to the $q = 0$ susceptibility on the lattice, note that the second term in Eq. (C17) can be rewritten using Eq. (C22)

$$\frac{\partial g_\sigma(11')}{\partial \Delta_\sigma(33')}|_{\phi} \left( \frac{\partial F}{\partial \Delta} \right)^{-1}_{\sigma'\sigma''\sigma'''} (33';44') = (1 + |\chi^{0,ph}(q = 0)|\Gamma^{ph,irr})^{-1}_{\sigma\sigma'\sigma''}(11';44'),$$

where $1_{11'\sigma';44''\sigma''} = \delta_{\sigma\sigma'}\delta_{1,4}\delta_{1',4'}$. Therefore, the Eq. (C17) as

$$-\frac{\partial g_\sigma(11')}{\partial \Delta_\sigma(33')}|_{\phi} \left[ \frac{\partial F}{\partial \Delta} \right]^{-1}_{\sigma'\sigma''\sigma'''} (33';44') \frac{\partial F^{irr}(44')}{\partial \phi(22')} |_{\Delta}.$$  

(C26)

Using the Eq. (C25), the multiplication of the first two terms at the Eq. (C27) is

$$\frac{\partial g_\sigma(11')}{\partial \phi(22')} |_{\Delta} - (1 + |\chi^{0,ph}(q = 0)|\Gamma^{ph,irr})^{-1}_{\sigma\sigma'\sigma''}(11';44') \left[ \chi^{0,ph}(q = 0)|\Gamma^{ph,irr} |_{\sigma\sigma'\sigma''}(11';44') |\chi^{0,ph}(q = 0)|\Gamma^{ph,irr} |_{\sigma\sigma'\sigma''}(11';44') \right]$$

(C28)

Therefore, the above expression for $\partial n/\partial \mu$ on the self-consistent impurity equals to the lattice BSE only when the self-energy is a functional of Green’s function described by the perturbative branch.
Note that our RPA analysis is different from the standard one because we use the dressed propagator in calculating $\chi_{0,ph}^{RPA}(Q)$.