Theory of Antibound States in Partially Filled Narrow Band Systems

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We present a theory of the dynamical two-particle response function in the Hubbard model based on the time-dependent Gutzwiller approximation. The results are in excellent agreement with exact diagonalization on small clusters and are reliable even for high densities, where the usual ladder approximation breaks down. We apply the theory to the computation of antibound states relevant for Auger spectroscopy and cold atom physics.

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Much of our understanding of strongly correlated electronic systems comes from dynamical responses like the one-particle spectral function which, under certain approximations, is probed by photoemission and inverse photoemission experiments. Less explored is the two-particle spectral function where one studies how the system responds to the addition or removal of two particles. In the case of two holes in an otherwise filled band, the response was computed exactly by Cini [1,2] and Sawatzky [3] (CS) in connection with Auger spectroscopy. They showed that for strong enough on-site repulsion the spectral function gets dominated by antibound states in which the two holes propagate together paying a large Coulomb cost.

Despite the interest of the problem, the dynamical two-particle response and the formation of antibound states in *partially filled* correlated systems are not well understood. Cini and collaborators [4,5] have compared approximations for the spectral function developed by several groups with exact diagonalization on finite clusters. They observed that any attempt to improve the single-fermion propagators with self-energy corrections or making them self-consistent leads to worse results due to the lack of vertex corrections which, if included, would tend to "undress" Green's functions. Thus for small filling, the best approximation corresponds to a trivial generalization of the original theory, namely, summing a ladder series with bare Green's functions. For moderate filling and for large interactions, this bare ladder approximation (BLA) breaks down and no reliable theory is available.

Several effects are expected to be relevant in the case of a partially filled band. First, strong correlations produce band narrowing, which should help to split off antibound states from the two-particle continuum. Second, the spectral weight of the antibound state should depend on doping, since the probability to find an empty site where to create an antibound pair depends on the filling. Third, the other holes present in the system are expected to affect the effective interaction among the added holes, which may lead to a renormalization of the position of the antibound state with respect to the continuum. Last, the chemical potential has a jump as a function of doping across the Mott insulating phase of narrow-band systems, which should show up in the position of the two-particle continuum with respect to the antibound state.

In this Letter we present a theory of antibound states for the Hubbard model, which incorporates these effects. It is based on the computation of pairing fluctuations within the time-dependent Gutzwiller approximation (TDGA) [6]. Our approach reproduces the effects discussed above, while keeping the simplicity of CS theory. Interestingly, we find that the effect of a finite density is to enhance the effective interaction which becomes singular as the Mott phase is approached [cf. Fig. 1(a)]. The comparison of

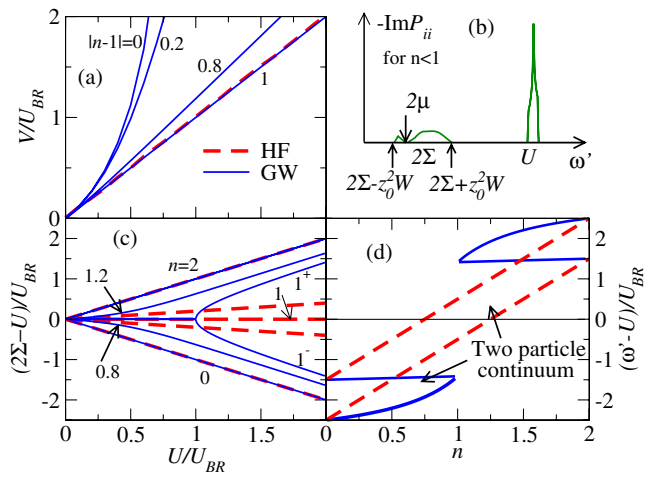

FIG. 1 (color online). Panels (a), (c), and (d) are obtained within a model with a flat density of states and a bare bandwidth W ($U_{\mathrm{BR}} \equiv 2W$) in HF (dashed line) and GA (full lines). (a) Effective particle-particle interaction V for different dopings $n-1$. Negative and positive dopings coincide. (b) Sketch of the principal energy scales of $\mathrm{Im}P_{ii}$. (c) Energy distance between the center of the two-particle scattering states (2Σ) and the doublon energy U for different fillings. The 1^+, 1^- fillings are infinitesimal deviations from half-filling and coincide in HF and in GA for $U < U_{\mathrm{BR}}$. The same plot represents the effective interaction rescaled by the doping $V(n-1)$ [see Eq. (6)]. (d) Boundaries of the two-particle continuum $\omega' = 2\Sigma \pm z_0^2 W$ for $U = 2U_{\mathrm{BR}}$ as a function of band filling.

TDGA with exact diagonalizations shows that the former is reliable even at high densities where the BLA breaks down (cf. Fig. 3).

Our starting point is the Hubbard Hamiltonian:

$$\hat{H} = \sum_{ij\sigma}(t_{ij} - \mu\delta_{ij})c^{\dagger}_{i\sigma}c_{j\sigma} + U\sum_{i} n_{i\uparrow}n_{i\downarrow}, \qquad (1)$$

where $c^{\dagger}_{i\sigma}$ creates a fermion with spin σ at site i, $n_{i\sigma} = c^{\dagger}_{i\sigma}c_{i\sigma}$, U is the on-site repulsion, t_{ij} denotes the hopping amplitude, and μ is the chemical potential.

We are interested in the following two-particle response:

$$P_{ij}(\omega) = \frac{1}{\imath}\int_{-\infty}^{\infty} dt e^{\imath\omega t}\langle \mathcal{T} c_{i\uparrow}(t)c_{i\downarrow}(t)c^{\dagger}_{j\downarrow}c^{\dagger}_{j\uparrow}\rangle, \qquad (2)$$

the imaginary part of which yields the two-particle addition (removal) spectra for $\omega > 0$ ($\omega < 0$). For the Auger application one should consider other effects which have been extensively discussed [4,7] and will not be treated here.

Our approach is based on the Gutzwiller wave function [8,9]: $|\Phi\rangle = P_g|\phi\rangle$, where P_g partially projects out doubly occupied sites from $|\phi\rangle$, which we assume to be a Bogoliubov vacuum. We define the single-particle density matrix $\rho_{i\sigma j\sigma'} \equiv \langle\phi|c^{\dagger}_{j\sigma'}c_{i\sigma}|\phi\rangle$ and pair matrix $\kappa_{i\sigma,j\sigma'} \equiv \langle\phi|c_{j\sigma'}c_{i\sigma}|\phi\rangle$, which satisfy the constraints [10]:

$$\rho^2 - \rho = \kappa\kappa^{*}, \qquad [\rho, \kappa] = 0. \qquad (3)$$

The first step is to construct the charge rotationally invariant energy functional $E \equiv \langle\Phi|H|\Phi\rangle$ in the Gutzwiller approximation (GA). This is more easily done by rotating at each site the fermion annihilation and creation operators to a basis where the anomalous expectation values vanish [11]. Then, one derives the GA with one of the known techniques [12,13] and rotates back to the original operators. For a nonmagnetic state one finds

$$E[\rho, \kappa, D] = \sum_{ij\sigma} t_{ij}z_i z_j \rho_{i\sigma,j\sigma} + U\sum_{i} D_i, \qquad (4)$$

with the hopping renormalization factors

$$z_i = \frac{\sqrt{\frac{1}{2} - D_i + J_{iz}}\left(\sqrt{D_i - J_{iz} - J_i} + \sqrt{D_i - J_{iz} + J_i}\right)}{\sqrt{\frac{1}{4} - J_i^2}}.$$

Here we defined $J_{ix} = (\kappa_{i\uparrow,i\downarrow} + \kappa^{*}_{i\uparrow,i\downarrow})/2$, $J_{iy} = \imath(\kappa_{i\uparrow,i\downarrow} - \kappa^{*}_{i\uparrow,i\downarrow})/2$, $J_{iz} = (\rho_{i\uparrow,i\uparrow} + \rho_{i\downarrow,i\downarrow} - 1)/2$, $J_i \equiv |\mathbf{J}_i|$, and the double occupancy $D_i = \langle\Phi|n_{i\uparrow}n_{i\downarrow}|\Phi\rangle$. The ground state is found by minimizing Eq. (4) with the constraints (3), leading to the static ρ^0, κ^0, $\mathbf{J}^0$, and D^0. We will consider a paramagnetic normal metal; thus $\kappa^0 = J^0_x = J^0_y = 0$.

To compute the response function we add a weak time-dependent pairing field $F(t) = \sum_i (f_i e^{-\imath\omega t}c_{i\downarrow}c_{i\uparrow} + \text{H.c.})$ to Eq. (1). This produces small time-dependent deviations $\delta\rho(t) = \rho(t) - \rho^0$. In addition, since F does not conserve the particle number, it induces pairing correlations κ, which we compute in linear response.

Previously [6,9,14], the energy was expanded to second order in terms of particle-hole fluctuations, leading to effective matrix elements for charge and spin excitations. For a normal paramagnet neither those channels nor δD fluctuations mix with the particle-particle channel, thus simplifying the formalism. The remaining part follows textbook computations in nuclear physics [10].

Expanding the energy up to second order in $\delta\rho$ and κ one finds

$$\delta E = \sum_{\mathbf{k}\sigma}(\varepsilon_{\mathbf{k}} - \mu)\delta\rho_{\mathbf{k}\sigma,\mathbf{k}\sigma} + V\sum_{i}(J^2_{ix} + J^2_{iy}). \qquad (5)$$

Here $\varepsilon_{\mathbf{k}} \equiv z_0^2 e_{\mathbf{k}} + \Sigma_G$ denotes the GA dispersion relation ($e_{\mathbf{k}}$ is the bare one), Σ_G coincides with the Lagrange parameter of the slave boson method [12] and is given by $\Sigma_G = z_0 z_0' \bar{e}$, with $\bar{e} \equiv \sum_{i\sigma} t_{ij}\rho^0_{i\sigma j\sigma}$, z_0 is the hopping renormalization factor at the saddle point, and z_0' is its density derivative. Our notation emphasizes the fact that Σ_G can be interpreted as a local GA self-energy. Finally, the effective on-site particle-particle interaction is

$$V = \frac{U - 2\Sigma_G}{1 - n}, \qquad (6)$$

where n denotes the particle concentration. At half-filling ($n = 1$), both the numerator and the denominator tend to zero and one finds $V = U(1 - U/2U_{\mathrm{BR}}) \times (1 + U/U_{\mathrm{BR}})/(1 - U/U_{\mathrm{BR}})$, which coincides with the particle-hole case [6,9,14]. Here $U_{\mathrm{BR}} \equiv 8\bar{e}$ is the critical interaction for the Brinkman-Rice metal insulator transition [9,15].

The response function can be readily derived from the equations of motion of the pair matrix in a normal system after using the constraint (3) to express the first term in Eq. (5) as a quadratic contribution in κ [10]. The pair-correlation function is given by the usual ladder expression but with the effective interaction of Eq. (6):

$$P(\mathbf{q}, \omega) = \frac{P^0(\mathbf{q}, \omega)}{1 - VP^0(\mathbf{q}, \omega)}, \qquad (7)$$

$$P^0(\mathbf{q}, \omega) = \frac{1}{N_s}\sum_{\mathbf{k}} \frac{1 - f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}})}{\omega - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}} + 2\mu + \imath\eta_{\mathbf{k},\mathbf{k}+\mathbf{q}}}, \qquad (8)$$

where N_s is the number of sites, $f(\varepsilon_{\mathbf{k}})$ is the Fermi distribution function, and $\eta_{\mathbf{k},\mathbf{k}'} \equiv 0^{+}\mathrm{sign}(\varepsilon_{\mathbf{k}} + \varepsilon_{\mathbf{k}'} - 2\mu)$.

Equations (6)–(8) constitute our main result. Our approach leads to the same formal ladder structure as in the CS theory [1,3,16] but with the HF self-energy ($\Sigma_{\mathrm{HF}} \equiv Un/2$, $z_0 = 1$) replaced by the GA one and the Hubbard repulsion U replaced by an effective interaction V. Notice that the "new" Eq. (6) is valid in the BLA provided one replaces $\Sigma_G \to \Sigma_{\mathrm{HF}}$, leading to $V = U$.

Figure 1(a) shows V [cf. Eq. (6)] as a function of U for a band of bare width W and a flat density of states. For

fillings close to 0 or 2 or small U, the effective interaction is close to the bare U, as expected. By contrast, as the Mott phase is approached, V diverges. This singular behavior is essential for the correct description of dense systems close to the Mott insulator.

Following Refs. [17,18], one can compute *exactly* $P(\mathbf{Q}, \omega)$ at $\mathbf{Q} \equiv (\pi, \ldots, \pi)$ for the particular case of nearest-neighbor hopping. For the following analysis we shift the origin of energies to eliminate the chemical potential in Eq. (8) and define $\omega' \equiv \omega + 2\mu$. The *exact* spectral function is exhausted by a single pole at $\omega' = U$. The antibound state consists of a doublon, i.e., an on-site pair. This provides a quick and instructive check of the theory. Indeed, by using that $e_{\mathbf{k}} + e_{\mathbf{k}+\mathbf{Q}} = 0$, one can verify that both BLA and the present theory reproduce the exact result. For general momenta and large U, Eq. (7) has a single pole for $\omega' \sim U$ (i.e., the antibound state) and a continuum at low (high) energy for $n < 1$ ($n > 1$). The local response is obtained as $P_{ii}(\omega) = 1/N_s \sum_{\mathbf{q}} P(\mathbf{q}, \omega)$. Figure 1(b) shows a sketch of $\mathrm{Im}P_{ii}(\omega')$. The dip in the continuum separates the addition part for $\omega' > 2\mu$ from the removal part for $\omega' < 2\mu$ [19].

The problem within the BLA is not so much the energy of the antibound state but rather the position of the two-particle continuum, which is given by the HF eigenvalues. This affects the antibound state because, as the continuum approaches the energy U, the antibound pair becomes less localized in the relative coordinate and eventually disappears. The distance between the continuum ($\sim 2\Sigma$) and the antibound pole ($\sim U$) as a function of U is shown in Fig. 1(c). The picture can be easily understood by noticing that by rescaling the y axis by $1/2$ one obtains one-particle energies. For an almost filled or an almost empty band, as well as for small U, the position of the HF and the GA bands coincide. At half-filling, the HF self-energy satisfies $2\Sigma_{\mathrm{HF}} - U = 0$. This coincides with the GA for $U < U_{\mathrm{BR}}$; however, for $U > U_{\mathrm{BR}}$ the GA self-energy bifurcates in two solutions, corresponding to infinitesimal positive and negative deviations from half-filling, due to the opening of the Mott-Hubbard gap. Thus for U larger than U_{BR} and moderate filling, the HF band is close to U, whereas the GA band is well separated from it which should produce quite different spectra. This dramatic difference is also illustrated in Fig. 1(d) where the boundaries of the continuum with respect to U are shown as a function of filling for $U = 2U_{\mathrm{BR}}$. The HF self-energy leads to a linear evolution. By contrast, in GA the band remains nearly at the same energy and narrows when $n \to 1$ due to correlation. At $n = 1$ the band jumps due to the Mott-Hubbard gap and the situation reverses. Clearly the GA continuum is always far from the antibound state whereas, in HF, it is generally much closer and overlaps the $\omega' = U$ line in a large range of filling near $n = 1$. Therefore, the formation of tight antibound states will be much more favored in the GA case than in HF.

Figure 2 compares the local two-particle spectral function for an infinite two-dimensional system and $n < 1$, within TDGA and BLA. The line shapes are dominated by the antibound state at $\omega' \sim U$. The intensity of the continuum at low energies has been multiplied by 10^3 to make the line shape visible. As anticipated, in the GA, the two-particle continuum is far from the antibound state, whereas, in the BLA, it quickly approaches it starting to overlap around $n = 0.7$.

The antibound state can propagate and forms a band which gives the width of the high-energy feature. The lower edge of this band corresponds to $\mathbf{q} = (\pi, \pi)$ for $n < 1$ and is at $\omega' = U$. For large U, the bandwidth is of order t^2/U for $n = 0$ [3] but becomes of order t [specifically $2z_0^2|\bar{e}/(1-n)|$] for finite n, since the kinetic energy can move a doublon at first order if there is a singly occupied site next to it.

The pair-correlation function satisfies the sum rule

$$\mathcal{S} = -\frac{1}{\pi}\int_{2\mu}^{\infty} d\omega' \mathrm{Im}P_{ii}(\omega') = 1 - n + \langle n_{i\uparrow} n_{i\downarrow}\rangle, \quad (9)$$

which can be used to evaluate ladder corrections to the GA or HF double occupancy shown in the upper inset of Fig. 2. The BLA introduces a strong correction to the HF double occupancy which for small filling is in accord with GA and then deviates. In contrast the TDGA double occupancy and the GA double occupancy almost coincide indicating a much higher degree of self-consistency.

In the TDGA, as the system approaches half-filling for $U > U_{\mathrm{BR}}$ the spectral weight decreases [cf. Eq. (9) and

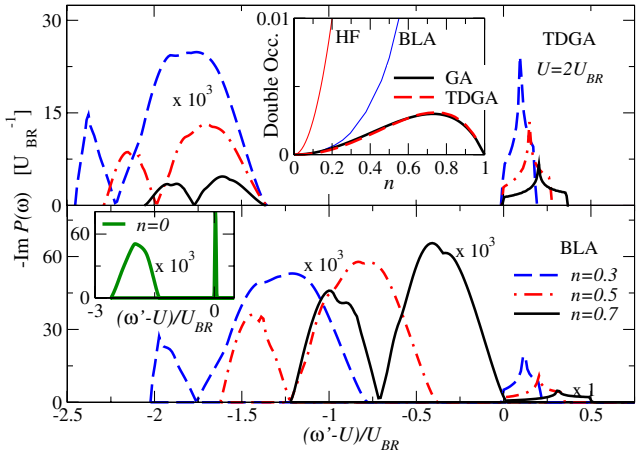

FIG. 2 (color online). Local spectral function for different fillings in TDGA and BLA. Results are for the Hubbard model on a square lattice with nearest-neighbor hopping ($U_{\mathrm{BR}} \equiv 128t/\pi^2$). The intensity of scattering states (low-energy feature) has been multiplied by 10^3. The dip in the scattering states corresponds to $\omega' = 2\mu$. A weak bound state contribution in the removal part has been omitted for clarity and will be discussed elsewhere [20]. Upper panel inset: Double occupancy vs filling in HF, GA and in BLA and TDGA after Eq. (9). Lower panel inset: The $n = 0$ case which coincides in BLA and TDGA.

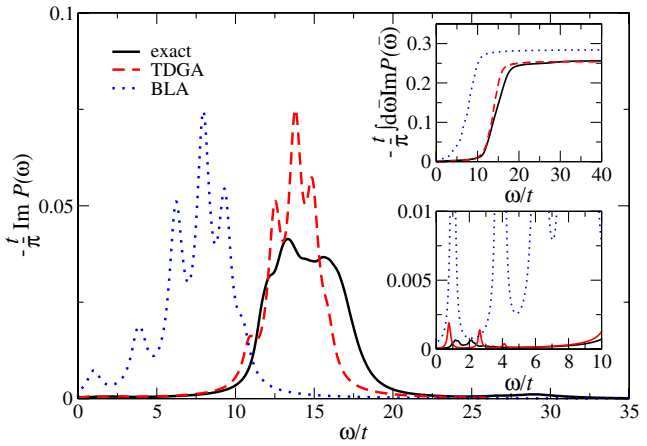

FIG. 3 (color online). Imaginary part of the pair-correlation function for the Hubbard model with $U/t = 15$ and 12 particles on a 4×4 square lattice obtained by exact diagonalization, TDGA, and BLA. The origin of energy is at 2μ. The lower inset enlarges the region of low-energy band excitations and the upper inset shows the frequency evolution of the integrated spectra. The broadening of the delta peaks is $0.5t$ in the main panel and the upper inset and $0.1t$ in the lower inset.

Fig. 2] reflecting the vanishing of the double occupancy in the GA description of the Mott phase. (Clearly in an exact computation, a small finite double occupancy and spectral weight will remain in the Mott phase.) In the BLA one gets a much larger intensity, specially in the scattering states, which contribute to a larger double occupancy.

In order to validate our results, we have computed the exact two-particle addition spectra for 12 particles on a 4×4 lattice with only nearest-neighbor hopping t and $U/t = 15$, by using exact diagonalization. Systems with 10 and 14 particles yield similar spectra. Figure 3 shows a comparison between the present theory and BLA. Here, we go back to our original variables and fix the origin of energy at 2μ. Despite the large value of the Hubbard repulsion, TDGA yields excellent agreement with exact diagonalization concerning the location, width, and intensity of the high-energy antibound states. On the other hand, BLA predicts that these excitations have a much lower energy when referenced to 2μ and no clear separation with the band states is visible (lower inset). The weight and position of the latter is also well reproduced by TDGA in contrast to the BLA. The upper inset demonstrates that the double occupancy given by Eq. (9) is accurate within TDGA, whereas BLA overestimates it as expected. The excellent performance of TDGA is not restricted to this particular value of U but persists to even larger (and of course lower) on-site repulsions.

To conclude, we have presented a computation of pair fluctuations for the Hubbard model exhibiting antibound states for large Coulomb repulsion. Our approximation gives reliable results even for large densities, where we are not aware of any accurate theory. The simplicity of the method suggests its application to the computation of Auger spectra on top of realistic Gutzwiller calculations [21]. Our theory can also be applied to ultracold fermion atoms in optical lattices, which can be described by the Hubbard model as well [22]. The possibility to observe antibound states has already been demonstrated in the Bose case [23]. Finally, the present TDGA scheme can be extended toward the computation of intersite pairing fluctuations where we also obtain a striking improvement over the standard ladder approximation [20].

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