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 give reliable results 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. A special bonus of the theory is its computational simplicity.

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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 in which one studies how the system responds to the addition or removal of two particles. In the case of two holes in an otherwise filled system, the response was computed exactly by Cini1,2 and Sawatzky3 (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 collaborators4,5 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” the 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 correlation produces 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 screen 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 work, 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 antiscreen the Hubbard $U$ interaction, i.e., the effective interaction is larger than the bare one and becomes singular as the Mott phase is approached [c.f. Fig. 1(a)]. The comparison of our results with exact diagonalizations shows that TDGA is reliable even at high densities where the BLA breaks down (c.f. Fig. 1).

Our starting point is the Hubbard Hamiltonian:

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

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

We are interested in the following two-particle response

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

for $\omega > 0$ ($\omega < 0$) the imaginary part of Eq. (2) gives the two-particle addition (removal) spectra. For the Auger application one should consider other effects which have been extensively discussed in the literature4 and will not be treated here (e.g., finite life time of the core hole7 and the interaction of the core hole with the valence electrons8).

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Here we defined \( J_{ix} = (\kappa_{i1,i1} + \kappa_{i1,i1}^*)/2 \), \( J_{iy} = i(\kappa_{i1,i1} - \kappa_{i1,i1}^*)/2 \), \( J_{iz} = (\rho_{i1,i1} + \rho_{i1,i1} - 1)/2 \), \( J \equiv \|J_i\| \), and the double occupancy \( D_i = \langle \langle \rho_{i1,i1} \rangle \| \Phi \rangle \). The ground state is found by minimizing Eq. (1) with the constraints (3), leading to the static \( \rho^0 \), \( \kappa^0 \), \( J^0 \) and \( D^0 \). We will consider a paramagnetic normal metal thus \( \kappa^0 = J^0 = 0 \).

To compute the response function we add a weak time-dependent pairing field \( F(t) = \sum_{i}(f_i e^{-i\omega t_i} \epsilon_{i1} + h.c.) \) 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 \( \kappa \), which we compute in linear response.

Previously [6, 10, 15], 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 \( \delta D \) fluctuations mix with the particle-particle channel which simplifies the formalism. The remaining part follows text book computations in nuclear physics [11].

Expanding the energy up to second order in \( \delta \rho \) and \( \kappa \) one finds:

\[
\delta E = \sum_{\sigma \sigma'} (\epsilon_k - \mu) \delta \rho_{k\sigma, \kappa \sigma'} + V \sum_i (J_{iz}^2 + J_{iz}^0). \tag{5}
\]

Here \( \epsilon_k \equiv \sqrt{2 \epsilon_k c_k} + \Sigma_G \) denotes the GA dispersion relation (\( \epsilon_k \) is the bare one), \( \Sigma_G \) coincides with the Lagrange parameter of the slave boson method [13] and is given by \( \Sigma_G = z_0 \bar{e} \rho_{k\sigma, \kappa \sigma'} \) with \( \bar{e} \equiv \sum_{i} t_{ij} \rho_{i\sigma, j\sigma'} \), \( z_0 \) is the hopping renormalization factor at the saddle point and \( z_0^* \) is its density derivative. Our notation emphasizes the fact that \( \Sigma_G \) can be interpreted as a local GA self-energy. Finally, the effective on-site particle-particle interaction is

\[
V = \frac{U - 2 \Sigma_G}{1 - n}, \tag{6}
\]

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 - \( U/2U_{BR} \)) (1 + \( U/U_{BR} \))/(1 - \( U/2U_{BR} \)), which coincides with the particle-hole case [6, 10, 15]. Here \( U_{BR} = 8 \sqrt{\epsilon} \) is the critical interaction for the Brinkman-Rice metal insulator transition [10, 16].

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 \( n \) [11]. The momentum dependent pair-correlation function is given by the usual ladder expression but with the effective interaction of Eq. (6):

\[
P(q, \omega) = \frac{P^0(q, \omega)}{1 - V P^0(q, \omega)}, \tag{7}
\]

where \( P^0(q, \omega) \) is the non-interacting two-quasiparticle correlation function

\[
P^0(q, \omega) = \frac{1}{N_s} \sum_k \frac{1 - f(\epsilon_k) - f(\epsilon_{k+q})}{\omega - \epsilon_k - \epsilon_{k+q} + 2\mu + \eta_{k,k+q}}. \tag{8}
\]

Our approach is based on the Gutzwiller wave function: \[ |\Phi\rangle = P_g |\phi\rangle \] 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 \( \rho_{\sigma\sigma'} \equiv \langle \langle \phi | c_{i\sigma}^+ c_{i\sigma'} | \phi \rangle \) and pair matrix \( \kappa_{\sigma\sigma'} \equiv \langle \langle \phi | c_{i\sigma}^+ c_{i\sigma'} | \phi \rangle \), which satisfy the following constraints [11]

\[
\rho^2 - \rho = \kappa \kappa^*, \quad [\rho, \kappa] = 0. \tag{3}
\]

The first step is to construct the charge rotationally invariant energy functional \( E \equiv \langle \langle \Phi | H | \Phi \rangle \) 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 [12]. Then, one derives the GA with one of the known techniques [13, 14] and rotates back to the original operators. Restricting to a paramagnetic state one finds

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

with the hopping renormalization factors

\[
z_i = \frac{\sqrt{\frac{1}{2} - D_i + J_{iz}} (\sqrt{D_i - J_{iz} - J_i} + \sqrt{D_i - J_{iz} + J_i})}{\sqrt{\frac{1}{4} - J_i^2}}.
\]
as the continuum at low (high) energy can move a doublon at first order if there is a 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 HF band is close to $U$, whereas the GA band is well separated from it. In this case we can anticipate quite different two-particle spectra in the two approximations. This dramatic difference is also illustrated in Fig. 1(d) where the boundaries of the continuum with respect to the doublon energy are shown as a function of filling for $U = 2U_{BR}$. The HF self-energy leads to a linear evolution. By contrast, in GA the band remains nearly at the same energy and narrows when $n \rightarrow 1$ 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.

Fig. 2 compares the local two-particle spectral function for an infinite two-dimensional system and $n < 1$, within TDGA and BLA. The inset shows the $n = 0$ case where TDGA and BLA coincide. Differences occur at finite concentrations (main panel) where the line shapes are dominated by the antibound state at $\omega' \sim U$ (as in CS), which is significantly stronger in the TDGA. The intensity of the continuum at low energies has been multiplied by $10^3$ to make the line shape visible. As anticipated the two-particle continuum is far from the antibound state in GA, whereas it quickly approaches it in the BLA.

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 $q = (\pi, \pi)$ for $n < 1$ and is at $\omega' = U$. For large $U$, the bandwidth is of order $t^2/U$ for $n = 0$ but becomes of order $t$ (specifically $2\tilde{\epsilon}^2/(1 - n)$) for finite $n$, since the kinetic energy can move a doublon at first order if there is a
single occupied site next to it.

The pair correlation function satisfies the sum rules:

\[ -\frac{1}{\pi} \int_{2\mu}^{\infty} d\omega' \text{Im} \, P_{i\bar{i}}(\omega') = 1 - n + \langle n_{i\uparrow} n_{i\downarrow} \rangle \quad (9) \]

\[ -\frac{1}{\pi} \int_{-\infty}^{2\mu} d\omega' \text{Im} \, P_{i\bar{i}}(\omega') = \langle n_{i\uparrow} n_{i\downarrow} \rangle. \quad (10) \]

This can be used to evaluate ladder corrections to the GA or HF double occupancy. The fact that the area of the removal part is much larger in the BLA than in TDGA reflects a larger double occupancy in the former. This is not surprising since at zero order BLA neglects correlations at all. Furthermore, in TDGA, as the system approaches the Mott phase, the hopping renormalizes to zero and the system becomes more “atomic” like. This explains the vanishing of two-particle scattering states as \( n \to 1 \). (Clearly in an exact computation, a small finite double occupancy and scattering intensity will remain in the Mott phase). Contrary, in the BLA the system becomes more “band” like as the filling is increased due to the closing of the gap between the scattering states and the doublon energy. Indeed for \( n = 0.85 \) the antibound state exists only for some values of the momentum.

In order to validate our results, we have computed the exact two-particle addition spectra for 10 particles on a \( 4 \times 4 \) lattice with only nearest-neighbor hopping \( t \) and \( U/t = 15 \), by using exact diagonalization. Fig. 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 \( 2\mu \). 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 \( 2\mu \) and no clear separation with the band states is visible (see upper-left inset). For the system under consideration, there are three band-like two-particle energies which are very well reproduced by TDGA in contrast with BLA. The upper-right inset demonstrates that the double occupancy after 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[20]. Our theory can also be applied to ultracold fermion atoms in optical lattices, which can be described by the Hubbard model as well[21]. The possibility to observe antibound states has already been demonstrated in the Bose case[22].

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