Angle-Resolved Photoemission Spectroscopy in the Hubbard Model Based on Cluster Perturbation Theory (Postprint)

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Abstract

Angle-resolved photoemission spectra (ARPES) are calculated in the Hubbard model by using cluster perturbation method. It is found that in a cluster of 12 sites, the local density of states displays the phase transition from normal conductor to Mott insulator with the increase of the electron-electron coupling. We show that a pseudogap develops from the metallic phase to the insulating phase. Evidence of spin-charge separation is also verified in the calculated single particle spectral functions.

Full Text

Preamble

Angle-resolved photoemission spectroscopy in the Hubbard model based on cluster perturbation theory

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Abstract: Angle-resolved photoemission spectra (ARPES) are calculated for the Hubbard model using the cluster perturbation method. In a 12-site cluster, the local density of states exhibits a phase transition from normal conductor to Mott insulator as the electron-electron coupling increases. We demonstrate that a pseudogap develops continuously from the metallic phase to the insulating

phase. Evidence of spin-charge separation is also observed in the calculated single-particle spectral functions.

Keywords: Angle-resolved photoemission spectra, Cluster perturbation theory, Exact diagonalization
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Introduction

Strong correlations arising from electron-electron interactions represent a central issue in condensed matter physics, underpinning fascinating phenomena such as high-temperature superconductivity, colossal magnetoresistance, and the metal-insulator transition known as the Mott transition. Both theoretical and experimental efforts have been extensively devoted to studying these phenomena. Research on the Mott transition has proven particularly valuable for understanding fundamental physical phenomena, including the mechanism of high-temperature superconductivity.

Angle-resolved photoemission spectroscopy (ARPES) serves as a powerful experimental tool for investigating electronic structures in solids, even those with strong electron-electron interactions. With its high energy and momentum resolution, ARPES enables bulk-sensitive observations and provides reliable information about the electronic structure of materials. It has been successfully employed to study various systems, including Mott insulators.

However, theoretical calculations sometimes fail to provide reasonable interpretations of ARPES experiments for strongly correlated electron systems, where conventional approximations of many-body quantum mechanics become inadequate. To describe the properties of low-dimensional strongly correlated cuprate materials in ARPES experiments, the Hubbard model with nearest-neighbor hopping and on-site Coulomb repulsion is frequently employed. In this paper, we calculate ARPES spectra using the cluster perturbation theory (CPT) method and present our model, theoretical approach, numerical results, and discussion.

Model and Method

A. Hubbard Model

The Hubbard model provides a fundamental description of correlated electron systems, capturing the essential physics of electrons with spin σ hopping between lattice sites. Its Hamiltonian is given by

$$H = -t \sum_{\langle i,j\rangle;\sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + c^{\dagger}_{j\sigma} c_{i\sigma}) + U \sum_{i} n_{i\uparrow} n_{i\downarrow},$$

where $\langle i, j\rangle$ denotes nearest-neighbor sites, t is the hopping energy, U is the on-site Coulomb repulsion, $c^{\dagger}_{i\sigma}$ and $c_{i\sigma}$ are creation and annihilation operators for

electrons with spin σ on site i, and $n_{i\sigma} = c^{\dagger}_{i\sigma}c_{i\sigma}$ is the electron number operator. The average electron density per site is $n = N_e/N$, where N is the number of sites and N_e is the number of electrons, with $n/2$ representing the band filling. When $U = 0$, the model reduces to the tight-binding model, while for $U \gg t$, strong repulsion suppresses double occupancy at each site.

B. Cluster Perturbation Theory

Cluster perturbation theory is an approximation scheme for lattice models with local interactions. The fundamental idea is to partition the infinite lattice into identical, disconnected clusters, each containing N_c lattice sites. The clusters are solved exactly, while inter-cluster hopping terms are treated perturbatively to first order in strong-coupling perturbation theory. We employ CPT to calculate one-particle properties of the Hubbard model, using exact diagonalization for the unit cluster to compute the Green' s function, which is then extended to the full infinite lattice.

The complete system Hamiltonian can be expressed as the sum of the one-body part H_0 and the interaction part V:

$$H = H_0 + V,$$

where

$$H_0 = \sum_R H^0_R, \quad V = \sum_{R,R',i,j} c^{\dagger}_{Ri} V^{ij}_{RR'} c_{R'j}.$$

Here H^0_R is the Hubbard Hamiltonian for the cluster located at superlattice vector R, and V represents nearest-neighbor hopping between adjacent clusters.

The quantity of interest is the one-particle Green' s function, defined as

$$G(k,z) = G^e(k,z) + G^h(k,z),$$

with

$$G^e(k,z) = \langle\Omega|c(k)\frac{1}{z - H + E_0}c^{\dagger}(k)|\Omega\rangle,$$

$$G^h(k,z) = \langle\Omega|c^{\dagger}(k)\frac{1}{z + H - E_0}c(k)|\Omega\rangle,$$

where $z = \omega + i\eta$, $c^{\dagger}(k)$ and $c(k)$ are creation and annihilation operators in momentum space, E_0 is the ground-state energy, and $|\Omega\rangle$ is the ground state. From these definitions, we obtain the single-particle spectral function

$$A(k,\omega) = -2 \lim_{\eta \to 0^+} \operatorname{Im} G(k, \omega + i\eta),$$

which gives the probability for an electron with momentum $\mathbf{k}$ to have energy ω. The negative-frequency part originates from G^h and corresponds to the signal measured in ARPES experiments.

The lowest-order approximation for the full single-particle Green' s function in mixed representation is

$$G_{ij}(Q,z) = \left(\frac{1}{1 - V(Q)G(z)} \right)_{ij} G_{ij}(z),$$

where Q is the cluster superlattice wave vector and $V(Q)$ is the Fourier transform of the $N_c \times N_c$ hopping matrix. Finally, the Green's function can be transformed from the mixed representation (real space within a cluster and reciprocal space between clusters) to obtain

$$G(k,z) = \sum_{i,j} e^{-ik(i-j)} G_{ij}(N_c k, z).$$

C. Basis States Construction

For a lattice of N sites with $N_\uparrow$ spin-up electrons and $N_\downarrow$ spin-down electrons, basis states are constructed by applying creation operators to the vacuum state in a specific order. For the Hubbard model, we first sort electrons by spin index and then by site index, with spin-up operators placed to the left of spin-down operators, and site indices increasing from left to right:

$$c^\dagger_{1\uparrow} \cdots c^\dagger_{i\uparrow} \cdots c^\dagger_{k-1\uparrow} c^\dagger_{k\uparrow} c^\dagger_{1\downarrow} \cdots c^\dagger_{i\downarrow} \cdots c^\dagger_{k-1\downarrow} c^\dagger_{k\downarrow} |0\rangle = |N_{k\uparrow}, N_{k-1\uparrow} \cdots N_{i\uparrow} \cdots N_{2\uparrow}, N_{1\uparrow}; N_{k\downarrow}, N_{k-1\downarrow} \cdots N_{i\downarrow} \cdots N_{2\downarrow}, N_{1\downarrow}\rangle.$$

A site occupied by one electron is coded as 1, otherwise as 0. The basis state $|N_k, N_{k-1} \cdots N_i \cdots N_2, N_1\rangle$ can thus be represented by a binary number $(N_k N_{k-1} \cdots N_i \cdots N_2 N_1)$. Once all basis states are identified, we can apply the Hamiltonian to obtain the matrix elements.

D. Exact Diagonalization

With the matrix representations of the basis states and Hamiltonian H, we obtain the ground state using the Lanczos algorithm, which efficiently computes a few extreme eigenvalues of a sparse matrix without full diagonalization. The algorithm constructs a projection of the full Hamiltonian matrix onto a Krylov subspace. Starting from a random normalized initial state $|\psi_0\rangle$, we build the Krylov subspace by iteratively applying H:

$$\mathcal{K} = \text{span}\{|\psi_0\rangle, H|\psi_0\rangle, H^2|\psi_0\rangle, \dots, H^n|\psi_0\rangle\}.$$

We then orthogonalize these states to obtain a basis for the Krylov space through the recursion relation

$$|\psi_{n+1}\rangle = H|\psi_n\rangle - a_n|\psi_n\rangle - b_n^2|\psi_{n-1}\rangle,$$

where

$$a_n = \langle\psi_n|H|\psi_n\rangle, \quad b_n^2 = \frac{\langle\psi_n|\psi_n\rangle}{\langle\psi_{n-1}|\psi_{n-1}\rangle}, \quad b_0 = 0, \quad |\psi_{-1}\rangle = 0.$$

At each step, only three state vectors $|\psi_{n+1}\rangle$, $|\psi_n\rangle$, and $|\psi_{n-1}\rangle$ are stored in memory. The projected Hamiltonian matrix is tridiagonal and formed by coefficients a_n and b_n:

$$T = \begin{pmatrix} a_0 & b_1 & 0 & \cdots \\ b_1 & a_1 & b_2 & \cdots \\ 0 & b_2 & a_2 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}.$$

This tridiagonal matrix can be diagonalized using standard routines. Iterations continue until the residual $||E_0|\Omega\rangle - H|\Omega\rangle||$ falls below a preset tolerance. The number of converged low-lying eigenvalues depends on the Krylov subspace dimension. As recursion order increases, the lowest eigenvalue and eigenvector of T converge to the ground-state energy E_0 and ground state $|\Omega\rangle$ of the original Hamiltonian H. Since $|\Omega\rangle$ is expressed in the reduced basis $\{|\psi_n\rangle\}$ and these vectors are not stored, we repeat the Lanczos recursion with the same initial vector $|\psi_0\rangle$ and reconstruct the ground state progressively using coefficients $\langle\Omega|\psi_n\rangle$.

Convergence is rapid at the spectrum edge where extreme eigenvalues are well-separated, but deteriorates quickly in the bulk spectrum. Thus, the Lanczos algorithm is suitable primarily for obtaining the ground state and a few low-lying excited states. Typically, a small number of iterations (ranging from a few tens to 200) suffices.

Results and Discussion

We apply the cluster perturbation method to investigate the spectral properties of a one-dimensional half-filled Hubbard model with 12 sites, the largest size permitting systematic study. As demonstrated below, our calculations successfully capture the essential features of strongly correlated electronic systems. In our numerical work, we set the electron hopping energy t as the unit of energy. The

system bandwidth is $4t$, which corresponds to a few eV in real solids, making $t \approx 1$ eV a reasonable estimate. We assume the parameter $\eta = 0.1$, giving the δ-peaks finite width.

Figure 1 [Figure 1: see original paper] shows the local density of states $N(\omega)$ at various U values, obtained by summing the electron spectral function $A(k, \omega)$ over all momenta. Three distinct evolution stages are observed with increasing U. Figures 1(a) and 1(b) correspond to a gapless phase, where electronic states feature a continuum around the Fermi energy ($\omega = 0$). Figures 1(c) and 1(d) show a pseudogap phase, where $N(\omega)$ displays a small dip at the Fermi energy, indicating pseudogap formation. Further increasing U causes the pseudogap' s depth and width to grow until a well-defined Mott insulating gap emerges, representing the gapped phase in Figures 1(e) and 1(f). This fully gapped state appears when $U > 3t$, while the positions of the lower and upper Hubbard bands remain essentially unchanged. These results clearly describe a metal-insulator transition from a normal metallic state at weak coupling to a Mott-Hubbard insulator at strong coupling.

Figure 2 [Figure 2: see original paper] presents the single-particle spectral functions at various wave vectors k (from $k = 0$ to $k = \pi$) for $U = 3$ and half-filling. These momentum-dependent spectra provide comprehensive insight into electronic band dispersion and structure. Since U is large, a Mott gap is nearly developed at the Fermi energy. All spectra separate into upper and lower parts on the positive and negative energy sides, respectively. The separation between these parts varies with k, reaching a minimum at $k = \pi/2$.

As noted in Section I, the spectral function directly corresponds to experimental photoemission and inverse photoemission spectra. The former probes occupied states below the Fermi energy and can be compared with the lower part of our calculated spectra, while the latter reveals unoccupied states above the Fermi energy. Therefore, our theoretical predictions of pseudogap states and gap-opening phenomena in strongly correlated systems are, in principle, experimentally verifiable. As shown in Figure 2 of Ref. [19], a sharp dip in the spectral density of states is observed at 30 K, which becomes more pronounced at 15 K. This dip at the Fermi energy indicates pseudogap formation at low temperatures. In Figure 1, such a pseudogap appears when U becomes comparable to t.

Beyond the Mott gap, another intriguing feature is spin-charge separation. The interacting one-dimensional electron system forms a Luttinger liquid, where elementary excitations consist of independent spin and charge waves with energy quanta called spinons and holons. These excitations propagate at different velocities with distinct dispersion relations. Such behavior manifests in the single-particle spectral function, which describes the excited states of the Luttinger liquid upon electron addition or removal. In Figure 2, the upper and lower spectral regions comprise two substructures with different energy dispersions, consistent with Luttinger liquid theory predictions of spin-charge separation. These features have been observed experimentally [20]. The high-energy edge of the spectral function corresponds to spin excitations, while the low-energy

edge relates to charge excitations.

Summary

In this work, we employ cluster perturbation theory to calculate the Green' s function and single-particle spectral function of strongly correlated electronic systems. Applying this method to the half-filled Hubbard model, we find that the calculated local density of states exhibits a smooth evolution corresponding to a phase transition from normal conductor to Mott insulator. Additionally, spin-charge separation is clearly verified in our calculations. These results should prove helpful for interpreting experimental ARPES data.

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Note: Figure translations are in progress. See original paper for figures.