Spectral evolution of angle-resolved photoemission due to Holstein-type electron-phonon scattering within the adiabatic approximation

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The angle-resolved photoemission spectra (ARPES) of the spin-1/2 Holstein model at non-half-filling are calculated by using the adiabatic approximation and the classical Monte Carlo method. It is found that in the metallic state, the spectra take broad Gaussians at the band bottom, while evolving to sharp Lorentzians near the Fermi level, commonly in one, two, and three dimensions. We justify that this evolution in ARPES is connected with an electronic state transition from quasi-localized ones to the extended ones due to the scattering of electrons with the static lattice distortion.

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I. INTRODUCTION

The role of electron-phonon (e-p) coupling in the solid state physics has gained considerable interest in recent years. Besides the fact that strong e-p interaction is crucial for the explanation of renormalization and line shapes of phonons observed in the neutron scattering experiments,1 the photo-emission spectroscopy (PES), and angle-resolved photoemission spectroscopy (ARPES) data have revealed that the electronic energy band structure is significantly modulated by the e-p interaction.2−4

The PES is widely regarded as a powerful tool to investigate the structure of occupied electronic valence bands in solids. As one of the most standard and contemporary experimental techniques, PES has been applied to study various normal metals and superconductors.5,6 The e-p coupling in the metallic systems has been studied in detail by PES for the cases of low-dimensional electronic states at surfaces7,8 and quantum-well states.9 The PES investigations on the e-p effects in the three-dimensional solids are recently reported on Pb,10 which has quite unconventional physical properties. For the case of ARPES, it can measure the binding energy as a function of each given momentum with an energy resolution of a few meV. By this means, the nature of electronic properties now can be clarified more precisely than before. For example, we can compare the information of e-p interaction thus obtained to the normal state properties like electrical resistivity and electronic heat capacity, and can clarify its contribution to the intrinsic quasiparticle linewidth.

In connection with the most up-to-date high resolution ARPES experiments on various one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) materials, at least two common features of ARPES for the e-p coupled systems have been noticed.11,12 (i) The spectra are sharp Lorentzians near the Fermi level (EF), which means the electrons are in the coherent states like plane waves extending over the whole crystal, while become broad Gaussians at the band bottom, indicating the electrons are in the incoherent state; (ii) the coherent and incoherent regimes are formally separated by a dispersion “kink”, where the spectral weight is exchanged between them so as to produce a two-headed or dip-hump structure at the crossover.

The two-headed structure is now recognized as a key evidence for the e-p interaction in the normal metals and superconductors, because it reflects the emission or absorption of an energy quanta of lattice vibration by the electron, and appears at a binding energy of 40−80 meV, which just corresponds to the characteristic phonon energy in the crystal. In this sense, the two-headed structure has a complete quantum origin. While for the coherent-incoherent evolution in ARPES, generally speaking, it represents the momentum-dependent variation of electronic states due to the many-body effects. As one of such many-body effects, in a previous paper,13 we have studied the effect of e-p scattering process via a quantum Monte Carlo (QMC) simulation on the Holstein model.14 It was shown there the e-p scattering plays a decisive role in the coherent-incoherent spectral evolution. Meanwhile a dependence of the line shape on the phonon mass indicated that the spectral evolution is related to the quantum nature of the phonon. However, because of a high computing cost of QMC, our previous calculation was only limited to the 1D and 2D cases. Moreover, in that study, since we adopted the QMC method, both quantum and classical effects of e-p scattering were fully taken into account. Hence, it could not be justified by that work whether the spectral evolution is a purely quantum phenomenon like the two-headed structure or not.

In this paper, in order to have a clear understanding on the mechanism of coherent-incoherent spectral evolution, we re-examine the spectral behavior of the Holstein model in the adiabatic limit, where the phonon mass is assumed to be infinite and only the scattering of electrons with the rigid lattice distortion is considered. We study the ARPES of 1D, 2D, and 3D cases at non-half-filling, as well as their relationship with the change of electronic states by using a classical Monte Carlo (CMC) method,15 which is applicable from weak to strong e-p coupling regimes with no approximation. Our calculation shows that the spectral evolution (refers to the calculated one hereafter if not specified) exists in all these adiabatic cases, thus demonstrating that the coherent-incoherent spectral evolution observed in experiments is essentially an admixture of quantum and classical effects.

Our paper is organized as follows: In Sec. II, we present our model and method. The numerical results are discussed
in Sec. III. In Sec. IV we give a summary of the this work.

II. MODEL AND METHOD

The e-p coupled systems with many electrons are often modeled by the Holstein model. In the adiabatic limit, the Holstein Hamiltonian reads

\[ H = -t \sum_{\langle i,j \rangle} (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) - \mu \sum_{i,\sigma} n_{i\sigma} + \frac{\omega_0}{2} \sum_{i} q_i^2 + g \sum_{i,\sigma} q_i (n_{i\sigma} - \frac{N}{2}). \]

(1)

Here \( c_{i\sigma}^\dagger \) (\( c_{i\sigma} \)) is an operator which creates (annihilates) an electron of spin \( \sigma \) on the site \( i \), and \( n_{i\sigma} := c_{i\sigma}^\dagger c_{i\sigma} \) is the electron number operator. The sum denoted by \( \langle i,j \rangle \) in the first term is over the sets of nearest neighbors with a one-electron hopping energy \( t \). \( q_i \) is the dimensionless position operator for the local Einstein phonon on the \( i \)th site with a frequency \( \omega_0 \). \( g \) is the e-p coupling constant, and \( \mu \) is the chemical potential of electrons.

From the theoretical point of view, the spectral function can be obtained from the single-particle Green’s function,

\[ G_{i,j,\sigma}(\tau) = -\langle T_F e^{i\tau c_{j\sigma}^\dagger} \rangle, \]

(2)

where \( \tau \) means imaginary time, \( c_{i\sigma}(\tau) \) is the Heisenberg representation of \( c_{i\sigma} \), and \( T_F \) is the time ordering operator. In terms of the path integral theory, the average of an operator \( \langle \cdots \rangle \) in Eq. (2) can be written as

\[ \langle \cdots \rangle = \frac{1}{Z} \int \mathcal{D}q e^{-\beta H(q)} \langle \cdots \rangle_q, \]

(3)

where the inverse temperature \( \beta := 1/k_B T \) is related to the effective one \( \beta’ \) by the following semiclassical relation,

\[ \frac{1}{2} \beta’ \omega_0 \coth \left( \frac{1}{2} \beta \omega_0 \right) = 1, \]

(4)

\( e^{-\beta H(q)} := \text{Tr} e^{-\beta H(q)} \) is the free energy for a given path \( q \) which is a classical quantity here, \( H(q) \) is the Hamiltonian on this path, and

\[ Z = \int \mathcal{D}q e^{-\beta H(q)} \]

is the partition function. On the r.h.s. of Eq. (3), the operator average \( \langle \cdots \rangle_q \) is calculated only on the given path \( q \),

\[ \langle \cdots \rangle_q = \frac{\text{Tr} e^{-\beta H(q)} \cdots}{\text{Tr} e^{-\beta H(q)}} \]

(6)

where

\[ H_{e}(q) = -t \sum_{\langle i,j \rangle} (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) - \mu \sum_{i,\sigma} n_{i\sigma} + g \sum_{i,\sigma} q_i n_{i\sigma}, \]

(7)

is the electronic Hamiltonian corresponding to this given lattice configuration. Equation (7) can be diagonalized by a canonical transformation \( \{ c_{i\sigma}^\dagger, c_{i\sigma} \} \rightarrow \{ \psi_{r\sigma}, \psi_{r\sigma}^\dagger \} \) as following,

\[ c_{i\sigma} = \sum_{r} O_{i\sigma}(q) \psi_{r\sigma}, \]

(8)

\[ c_{i\sigma}^\dagger = \sum_{r} O_{i\sigma}^*(q) \psi_{r\sigma}^\dagger, \]

(9)

\[ \sum_{r} O_{i\sigma}^*(q) O_{i\sigma}(q) = \delta_{r}, \]

(10)

Then we get the diagonalized electronic Hamiltonian as

\[ \tilde{H}(q) = \sum_{r,\sigma} \epsilon_{r}(q) \psi_{r,\sigma}^\dagger \psi_{r,\sigma}. \]

(11)

After performing the Fourier transformation

\[ G_{i,j,\sigma}(i\omega) = \frac{1}{Z} \int d\tau e^{i\omega \tau} G_{i,j,\sigma}(\tau), \]

(12)

\[ \omega_n = (2n + 1) \pi/\beta, \]

(13)

and the analytic continuation \( i\omega \rightarrow \omega + i0^+ \), we get the retarded single-particle Green’s function as,

\[ G_{i,j,\sigma}^R(\omega) = \frac{1}{Z} \int \mathcal{D}q e^{-\beta H(q)} \sum_{r} O_{i,\sigma}(q) O_{j,\sigma}^*(q) \frac{1}{\omega - \epsilon_{r}(q) + i0^+}. \]

(14)

Transforming it to the momentum space by

\[ G_{\sigma}(k,\omega) = \frac{1}{N} \sum_{i,j} e^{ik(r_j - r_i)} G_{i,j,\sigma}^R(\omega), \]

(15)

where \( N \) is the total number of sites, we obtain the spectral function as,

\[ A_{\sigma}(k,\omega) = -\frac{1}{\pi} \text{Im} G_{\sigma}(k,\omega) \]

\[ = \frac{1}{Z} \int \mathcal{D}q e^{-\beta H(q)} \sum_{i} \sum_{r} e^{ik(r_j - r_i)} O_{i,\sigma}(q) O_{j,\sigma}^*(q) \times \delta(\omega - \epsilon_{r}(q)). \]

(16)

In the numerical calculation, the \( \delta \) function is replaced by the Lorentzians

\[ \delta(\omega - \epsilon_{r}(q)) \rightarrow \frac{\eta}{\pi [\omega - \epsilon_{r}(q)]^2 + \eta^2}, \]

(17)

where \( \eta \) is taken as 0.01 in this work. Finally, by imposing the Fermi function \( f(\omega) = 1/(e^{\beta \omega} + 1) \), we have the normalized ARPES intensity as

\[ l(k,\omega) = \sum_{\sigma} A_{\sigma}(k,\omega) f(\omega). \]

(18)

Starting from Eq. (16), we numerically calculate the ARPES of the finite dimensional systems by the classical Monte Carlo method. The probability distribution of phononic configuration is sampled with a modified Monte Carlo.
Carlo algorithm. One Monte Carlo step consists of a proposed move and an accept/reject procedure. The possible movement is generated by a Gaussian-distributed random generator with zero mean. We found that 20,000 steps of the modified Monte Carlo algorithm are adequate to equilibrate the system for 64-site chain lattice. With 10,000 samples, the statistical errors are less than 0.7% in the spectral function $A(\mathbf{k}, \omega)$.  

III. RESULTS AND DISCUSSIONS

It is already known the Holstein model is subject to the Peierls instability at low temperature and strong $e\-p$ coupling, which leads to a charge-density-wave (CDW) gap at $E_F$. When the CDW gap opens, the change in spectra cannot be seen clearly, especially nearby $E_F$. To avoid such an ambiguity in ARPES and keep the system in the normal or quasi-metallic states, in this work, we restrict our attention to the weak and intermediate $e\-p$ couplings at non-half-filling with a little high temperature. In the numerical calculation, we choose $t=1$ to set the energy scale, and assume the phonon energy to be $\omega_0=0.1$. The periodic boundary condition are imposed on all the 1D, 2D, 3D systems.

First we consider the 1D 64-site chain lattice at 37.5%-filling. The plot of ARPES intensity $I(\mathbf{k}, \omega)$ versus $\omega$ is shown in Fig. 1 for various $\mathbf{k}$, where the coupling constant and inverse temperature are $g=0.4$ and $\beta'=5$, respectively. We can see the spectrum takes a broad form at the bottom $\mathbf{k}=0$. As $\mathbf{k}$ increases, the spectral shape gradually becomes sharp, and at $E_F$, corresponding to $\mathbf{k}=12\pi/32$, the Lorentzian form dominates. Figure 2 shows the spectra of 1D 64-site system with fixed filling and temperature, but different coupling constants. Both Figs. 2(a) and 2(b) show the same tendency of spectral evolution as that represented in Fig. 1. In Fig. 3, the temperature dependence of spectra is plotted for the 1D 64-site system with coupling strength fixed at $g=0.3$. Comparing the spectra at band bottom and $E_F$ for different temperatures, we find that increasing temperature has similar effect on the spectral line shape as decreasing the coupling strength. From Figs. 1–3, we note that all of these results are qualitatively consistent with the experimental observations.

Now let us proceed to the calculations of the 2D and 3D systems. In Fig. 4, the spectra of 2D $8 \times 8$ square lattice at 19.53%-filling with different temperatures and coupling constants are shown. In the 2D case, the same characteristics of spectral evolution can also be clearly seen. Moreover, increasing the coupling constant and temperature cause a notable broadening in the spectral shape. These features are identical with the description on the 1D case. For the 3D case, we study a $4 \times 4 \times 4$ cubic lattice. In Fig. 5, we show two different filling cases: Figure 5(a) is for the $4 \times 4 \times 4$ cubic lattice with 10 electrons, and Fig. 5(b) with 18 electrons. In Fig. 5, we notice that the spectral evolution still exists, although the spectra show a little weak change in the shapes.
Let us now qualitatively clarify the origin of these marked spectral evolution in connection with our numerical results. In the photoemission, the incident electromagnetic radiation on a solid excites the electrons from materials so that the outgoing photoelectrons are measured as a function of their energies and emission angles. Via both energy and momentum conservation, the measured quantities are directly related to the energy and momentum of the initial electronic state inside the materials. In an e-p coupled system, when a photoelectron is emitted from the interacting many-electron system, a hole is created at the same time. The lifetime of this hole state is determined by its location in the energy band and the scattering with nearby phonons. Therefore, the coherent-incoherent spectral evolution of ARPES is a consequence of the interaction between the phonon and hole left in the solid as a final state. In the coherent part, the spectra show sharp quasi-particle forms, indicating the hole of this regime is in an extended plane wave state. While, in the incoherent continuum, the spectra are highly broadened, which means the hole is heavily dressed by the phonon cloud.

In the adiabatic limit, one can see in Figs. 1–5, there is no well-defined coherent-incoherent crossover of two-headed structure. But the spectral evolution still exists and becomes more smooth. To understand the origin of this characteristic property, we introduce the concepts of extended and localized states in our illustrations below. For the adiabatic Holstein model, the retardation effect of phonons is dominant and the phonons cannot respond to the charge transfer of electrons. In this sense, the scattering of electrons due to the phonons is analogous to the scattering with the static impurities in the crystal, and correspondingly, the electronic spatial extent is expected to undertake an extended-localized transition in the band. Here we should note that, the static e-p scattering is essentially different from the ordinary electron-impurity scattering, as the ions are placed only around their equilibrium positions, while the impurity configurations are completely random. In spite of this difference, we can introduce the participation ratio \( \alpha = (\langle |u_i|^2 \rangle^2 / \langle |u_i|^4 \rangle) \) of an electronic eigenstate with amplitude \( u_i \) to measure the spatial range of this state, as suggested in Ref. 20. Obviously, \( \alpha \) depends on the spatial extent of electronic state. For an extended state \( \alpha \sim 1 \), while for a localized one \( \alpha \) should decrease as the site number \( N \) increases.

Keeping these points in mind, let us return to our case. Shown as in Fig. 6(a), the site-dependence of \( \alpha \) gives the site dependence of \( \alpha \) for two different electronic states: the solid curve corresponds to an electronic state near the band bottom, and the dashed curve is for a state close to \( E_F \). Figure 6(b) displays \( \alpha \) vs electronic binding energy, for a 64-site chain.
and it changes little with the increase of site number. It quantum and classical effects of the coherent-incoherent spectral evolution observed in the experiments of ARPES. Since our above analysis is greatly suppressed, so that the energy designated in ARPES calculation under various conditions demonstrates that the adiabatic Holstein model can bring out the characteristic spectral evolution from a broad Gaussian at the band bottom to a sharp Lorentzian near \( E_F \), commonly in 1D, 2D, and 3D cases; (ii) we quantitatively investigate the mechanism of this spectral evolution at adiabatic limit by introducing the participation ratio. The spectral evolution is illustrated to be connected with an electronic state transition from quasi-localized incoherent ones to extended coherent ones, as a result of the scattering of electrons with the static lattice distortion; (iii) our calculation suggests that the experimentally observed coherent-incoherent spectral evolution essentially has both quantum and classical origins. While the complete quantum nature of two-headed structure is manifested by its absence in the classically calculated ARPES of present work.

IV. SUMMARY

In conclusion, we have calculated the ARPES of the spin-
1/2 Holstein model under the adiabatic approximation for various conditions in the 1D, 2D, and 3D cases at non-half-filling, by using the CMC method. The prime results obtained are the following: (i) The theoretical treatment with adiabatic approximation partially explains the spectral evolution observed in the experiments of ARPES. The detailed analysis under various conditions demonstrates that the adiabatic Holstein model can bring out the characteristic spectral evolution from a broad Gaussian at the band bottom to a sharp Lorentzian near \( E_F \), commonly in 1D, 2D, and 3D cases; (ii) we quantitatively investigate the mechanism of this spectral evolution at adiabatic limit by introducing the participation ratio. The spectral evolution is illustrated to be connected with an electronic state transition from quasi-localized incoherent ones to extended coherent ones, as a result of the scattering of electrons with the static lattice distortion; (iii) our calculation suggests that the experimentally observed coherent-incoherent spectral evolution essentially has both quantum and classical origins. While the complete quantum nature of two-headed structure is manifested by its absence in the classically calculated ARPES of present work.

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