Invited Paper

Theoretical description of pump/probe experiments in nesting induced charge density wave insulators

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ABSTRACT

We present a theoretical description of time-resolved photoemission in charge-density-wave insulators that derive their ordering from electron nesting effects. In these pump/probe experiments, a large amplitude (but short duration) pump pulse excites the system into nonequilibrium and then a higher frequency low amplitude probe pulse photoexcites electrons, which are measured at the detector. We describe effects of electron correlations on the photoelectron spectroscopy and provide details for the theoretical techniques used to solve these problems. We also show how the gap fills in as the system is excited, even though the order parameter does not go to zero. The theory is developed for the Falicov-Kimball model, which can be solved exactly with nonequilibrium dynamical mean-field theory.

Keywords: Charge-density-wave ordered phase, Nonequilibrium dynamical mean-field theory, Pump-probe spectroscopy, Strongly correlated electrons, Falicov-Kimball model

1. INTRODUCTION

Recently, a number of different charge density wave (CDW) materials have been investigated with pump-probe time-resolved and angle resolved photoemission (PES). This includes TaS_2,[1–4] $TbTe_3$,[5,6] and $TiSe_2$.[4,7] In addition, it has been found that pumping these systems, especially TaS_2, can lead to new hidden phases, that are not part of the equilibrium phase diagram.[8–10] One of the interesting behaviors is that the PES can show the transient destruction of the CDW gap, even while the CDW order parameter (determined by the difference in the electronic charge on the different sublattices) remains nonzero. An initial theoretical study on the simplest noninteracting CDW shows the transient destruction of the gap near the time when the pump is on (and the order parameter remains nonzero), but does not show long-time oscillations of the PES signal after the pump is gone, as have been observed experimentally. It is believed that these oscillations arise from the electron-phonon coupling with the "ordering" phonon that goes soft at the transition. In this work, we examine the time-resolved PES in a more complicated electronic model for the CDW described by the Falicov-Kimball model.[11] This model is interesting because even in equilibrium, the density of states (DOS) shows a partial, then complete fill in of the gap from subgap states[12–14] instead of a continuous reduction of the magnitude of the gap (as occurs in the BCS theory[15]). Hence, this all electronic model has additional correlation effects not seen in previous modeling. Because we do not couple to a phonon, the solutions are not expected to show long-time oscillations. This work goes beyond the simplest calculations of the transient PES in a noninteracting CDW.[16]

We choose the Falicov-Kimball model because it has a nontrivial CDW phase (which exhibits a triple-point quantum phase transition) in equilibrium[17,18] and it can be solved in nonequilibrium exactly[19,20] by employing the Kadanoff-Baym-Keldysh formalism[21,22] within dynamical mean-field theory (DMFT).

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2. FORMALISM

We highlight the theory for the calculation of the PES response in the CDW ordered phase of the Falicov-Kimball model within the nonequilibrium DMFT approach. We start with a short overview of the CDW ordered state; next we introduce the time-dependent Hamiltonian of the system and the Kadanoff-Baym-Keldysh contour; and we end with the sum rules for the Green's function and formal resuls for the time-resolved PES response function.

2.1 Time-dependent Hamiltonian in the CDW ordered state

We assume the electrons move on an infinite dimensional hypercubic lattice with nearest-neighbor hopping only. The electronic system is ordered with a modulation of the charge density on a bipartite lattice. This ordering arises from a nesting instability of the Fermi surface. In this case, the Brillouin zone splits in half, the modulation vector is equal to $\mathbf{Q} = (\pi, \pi, \dots)$, and the momenta $\mathbf{k}$ and $\mathbf{k} + \mathbf{Q}$ are coupled together to produce two effective bands. In real space, this corresponds to introducing the two subattices A and B which are defined via the modulation vector $\mathbf{Q}$ as follows:

$$e^{i\mathbf{Q}\cdot\mathbf{R}_i} = \begin{cases} 1, & \mathbf{R}_i \in A, \\ -1, & \mathbf{R}_i \in B. \end{cases} \tag{1}$$

Here $\mathbf{R}_i$ denotes the position vector for the ith lattice site. To emphasize the sublattices, we enhance the site index i notation with an additional sublattice index $\alpha = A,\ B$, so that the electron annihilation and creation operators are denoted by

$$c_i \to c_{i,\alpha}, \quad \text{and} \quad c_i^\dagger \to c_{i,\alpha}^\dagger, \quad \alpha = A, B. \tag{2}$$

Similarly, one can define the CDW state in reciprocal space for the momentum-dependent operators. In our theory we use both a real space, and a momentum space representation (details[23,24] are presented elsewhere), but here we restrict ourselves to the real space description for brevity.

The Falicov-Kimball model is in a bipartite CDW state when $T = 0$ and the filling of the light and the filling of the heavy electrons are each exactly 0.5 (on average). Taking into account the above notation, we write down the time-dependent Hamiltonian of the system as the sum of two terms

$$\mathcal{H}(t) = \sum_{i\alpha} \mathcal{H}_i^\alpha - \sum_{ij\alpha\beta} t_{ij}^{\alpha\beta}(t) c_{i\alpha}^\dagger c_{j\beta}. \tag{3}$$

The symbols $c_{i\alpha}^\dagger$ and $c_{i\alpha}$ are the fermionic creation and annihilation operators for itinerant electrons at site $i\alpha$, with $\alpha = A$ or B; similar operators using an f notation are employed for the heavy electrons. The local term describes the Coulomb interaction (and the chemical potential)

$$\mathcal{H}_i^\alpha = U n_{ic}^\alpha n_{if}^\alpha - \mu^\alpha n_{ic}^\alpha, \tag{4}$$

where $n_{ic}^\alpha = c_{i\alpha}^\dagger c_{i\alpha}$ and $n_{if}^\alpha = f_{i\alpha}^\dagger f_{i\alpha}$ are the number operators of the itinerant and localized electrons, respectively. The hopping term is time-dependent due to the introduction of the Peierls' substitution for the time-dependent (but spatially uniform) electric field, which creates a complex valued and time-dependent hopping term as described below. We choose the physical constants to satisfy $\hbar = c = e = a = 1$.

The order parameter for the heavy electrons $\Delta n_f = (n_f^A - n_f^B)/2$ is defined by the difference in the occupations of the f-electrons on the A and B sublattices. The order parameter for the mobile electrons is similarly given by $\Delta n_c = (n_c^A - n_c^B)/2$.

To drive the system out of the thermal equilibrium, we hit it with an intense ultrafast pulse. We model that pulse as an electric field with a Gaussian envelope of the form

$$\mathbf{E}(t) = \mathbf{E}_0 \cos[\omega_p(t - t_0)] \exp\left[-(t - t_0)^2/\sigma_p^2\right], \tag{5}$$

where E_0 is the magnitude of the field at time $t = t_0$ (the maximum of the pump pulse). We assume that the field is spatially uniform with its direction along the diagonal (1,1,...,1). We also ignore all magnetic field and

relativistic effects. This allows us to describe the electric field via a spatially uniform vector potential in the Hamiltonian gauge:

$$\mathbf{E}(t) = -\frac{d}{dt}\mathbf{A}(t). \tag{6}$$

The interaction with the external field in Eq. (5) is then described with a Peierls' substitution to the kinetic-energy term of the Hamiltonian:

$$t_{ij}^{\alpha\beta}(t) = t_{ij}^{\alpha\beta}\exp\left(-i\int_{\mathbf{R}_{i,\alpha}}^{\mathbf{R}_{j,\beta}}\mathbf{A}(t)\cdot d\mathbf{r}\right), \tag{7}$$

where $t_{ij}^{\alpha\beta}$ is the hopping matrix in the absence of the external electric field.

Since the hopping is between lattice sites which belong to different sublattices, the time-dependent kinetic-energy term of the Hamiltonian is off-diagonal in real space. Performing a Fourier transformation to momentum space, we write it in the form

$$\hat{\mathcal{H}}_{kin}(t) = \sum_{k}\begin{bmatrix}c_{\mathbf{k}A}^{\dagger} & c_{\mathbf{k}B}^{\dagger}\end{bmatrix}\hat{\epsilon}(\mathbf{k}-\mathbf{A}(t))\begin{bmatrix}c_{\mathbf{k}A}\\ c_{\mathbf{k}B}\end{bmatrix}, \tag{8}$$

where $\hat{\epsilon}(\mathbf{k}-\mathbf{A}(t))$ is the off-diagonal matrix of the generalized band energies[25]

$$\hat{\epsilon}(\mathbf{k}-\mathbf{A}(t)) = \begin{Vmatrix} 0 & \epsilon(\mathbf{k})\cos(A(t))+\bar{\epsilon}(\mathbf{k})\sin(A(t)) \\ \epsilon(\mathbf{k})\cos(A(t))+\bar{\epsilon}(\mathbf{k})\sin(A(t)) & 0\end{Vmatrix} \tag{9}$$

with $\epsilon(\mathbf{k}) = -\lim_{d\to\infty} t^*\sum_{r=1}^{d}\cos k_r/\sqrt{d}$ and $\bar{\epsilon}(\mathbf{k}) = -\lim_{d\to\infty} t^*\sum_{r=1}^{d}\sin k_r/\sqrt{d}$ (of course, we apply the same scaling to the hopping term as in equilibrium DMFT).

2.2 Photoemission response

We follow the theory for time-resolved, pump-probe, PES developed previously.[26] In the presence of an external electric field, the Hamiltonian depends explicitly on time and we must employ the Kadanoff-Baym-Keldysh formalism to describe the nonequilibrium behavior of the system. We begin with the contour-ordered Green's function defined on the Kadanoff-Baym-Keldysh contour in Fig. 1:

$$G_{\mathbf{k}}^{c}(t,t') = -i\langle\mathcal{T}_c c_{\mathbf{k}}(t)c_{\mathbf{k}}^{\dagger}(t')\rangle, \tag{10}$$

where $\langle\mathcal{O}(t)\rangle = \mathrm{Tr}\exp[-\beta\mathcal{H}(t\to-\infty)]\mathcal{O}(t)/\mathcal{Z}$ and the partition function is $\mathcal{Z} = \mathrm{Tr}\exp[-\beta\mathcal{H}(t\to-\infty)]$. Here, $\beta = 1/T$ is the inverse of the initial equilibrium temperature before the system is driven by the electric pulse, and we assume the Hamiltonian becomes time independent at early times. The Green's function in Eq. (10) is a "block scalar" because we discretize the time interval and it corresponds to the normal-state. When we generalize to the ordered phase, we have a 2×2 block matrix

$$\hat{G}_{\epsilon,\bar{\epsilon}}^{c}(t,t') = \begin{Vmatrix} G_{\epsilon,\bar{\epsilon}}^{c,AA}(t,t') & G_{\epsilon,\bar{\epsilon}}^{c,AB}(t,t') \\ G_{\epsilon,\bar{\epsilon}}^{c,BA}(t,t') & G_{\epsilon,\bar{\epsilon}}^{c,BB}(t,t')\end{Vmatrix}. \tag{11}$$

We apply the DMFT approach to solve for the contour-ordered Green's function. A complete derivation for the DMFT procedure in nonequilibrium was developed elsewhere for both the normal[19] and the CDW ordered[23] states of the Falicov-Kimball model. Here we only summarize that in the case of a two-sublattice system, we deal with matrix equations whose components are also matrices in time. Hence, the Green's function is a continous matrix operator. To work with it numerically, we first discretize the system on a time grid with a fixed spacing Δt and then extrapolate the results to the zero spacing limit $\Delta t\to 0$. To benchmark our results, we check the spectral moment sum rules which continue to hold in the ordered phase and in nonequilibrium. The moments are defined to be

$$\mu_n^{R,a}(t_{ave}) = -\frac{1}{\pi}\mathrm{Im}\left[i^n\frac{\partial^n}{\partial t_{rel}^n}G^{R,\alpha}(t_{ave},t_{rel})\big|_{t_{rel}\to 0^+}\right] = \int d\omega A^{\alpha}(t_{ave},\omega)\omega^n \tag{12}$$

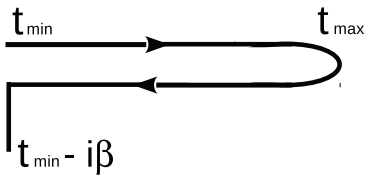

Figure 1. Kadanoff-Baym-Keldysh time contour, which runs from a minimum time to a maximum time along the real time axis, then backwards to the minimum time, and then along the imaginary axis for a length given by the inverse of the initial equilibrium temperature.

with $\alpha = (A, B)$ and with the retarded Green's function expressed in terms of the Wigner coordinates $t_{ave} = (t + t')/2$ and $t_{rel} = t - t'$. The symbol $A^{\alpha}(t_{ave}, \omega) = -\mathrm{Im}G^{R,\alpha}(t_{ave}, \omega)/\pi$ is the local density of states on the α sublattice (which has been Fourier transformed with respect to the relative time). We have calculated the zeroth, first and second moments, which satisfy[27]

$$\mu_0^{R,\alpha}(T) = 1, \tag{13}$$

$$\mu_1^{R,\alpha}(T) = -\mu + Un_f^{\alpha}, \tag{14}$$

$$\mu_2^{R,\alpha}(T) = \frac{1}{2} + \mu^2 - 2U\mu n_f^{\alpha} + U^2 n_f^{\alpha}, \tag{15}$$

where $n_f^A = 1/2 + \Delta n_f$ and $n_f^B = 1/2 - \Delta n_f$; we only show results for the first two moments below.

The time-resolved PES response function[26] $P_{\alpha}(\omega, t_0')$ is calculated from a double-time Fourier transform of the lesser Green's function weighted by the probe pulse envelope function $s(t)$ as follows:

$$P_{\alpha}(\omega, t_0') = -i \int_{t_{\min}}^{t_{\max}} dt \int_{t_{\min}}^{t_{\max}} dt' s(t) s(t') e^{-i\omega(t-t')} G_{\alpha}^{<}(t, t'), \tag{16}$$

where $\alpha = A, B$. The PES is different for each sublattice, but, generically, an experiment will measure the sum of these two responses, providing the average response. We assume the envelope function $s(t)$ is a Gaussian

$$s(t) = \frac{1}{\sigma_b \sqrt{\pi}} e^{-(t-t_0')^2/\sigma_b^2}, \tag{17}$$

where t_0' is the time when the probe pulse has its maximum and it defines the time delay relative to the pump pulse maximum at t_0 (defined above) and σ_b defines the effective width of the probe pulse. The broader the width of the probe pulse, the better the energy resolution and the worse the time resolution, and *vice versa* for narrower probe pulses.

In the CDW ordered state, we can choose two different order parameters. One is the order parameter of the heavy electrons Δn_f, which we introduced above and it is the difference of the heavy electron filling on the A and B sublattices. It starts at 0 at T_c and increases all the way to 1/2 at $T = 0$. This order parameter remains unchanged as the field is applied, because the heavy electrons do not couple to the external electric field, since they are localized. The other order parameter is the difference in the conduction electron filling on the two sublattices Δn_c. This order parameter changes as the field is applied, because the field does cause motion of the conduction electrons. Even in equilibrium, this order parameter is distinct from the heavy electron order parameter; it starts at 0 at T_c but it generically does not go to 1/2 as $T \to 0$. Indeed, it can even change sign when the system is pumped by an external electric field.

3. DISCUSSION

In our theory for calculation of the PES function, we first iterate the equations of the nonequilibrium DMFT to solve for the Green's function, which depends on two times. In the two-sublattice case, the Green's function is a

2×2 matrix and we need to calculate the response for each sublattice. Two time variables lie on the Kadanoff-Baym-Keldysh contour which are discretized. We extrapolate results for different Δt to $\Delta t \to 0$. We employ the exact sum rules in Eq.(12) as a benchmark for the accuracy of the Green's function. We find that the numerical accuracy varies with the parameters of the pump pulse. In Fig. **??**, we show the results for the zeroth and the first moments for different values of the E_0 of the pump pulse in Eq.(5). For larger fields ($E_0 = 30$), the results of the zeroth and first moments are accurate enough, while for small $E_0 = 1$ they are not. This fact restricts our numerical calculations to large amplitude pump pulses (high fluence).

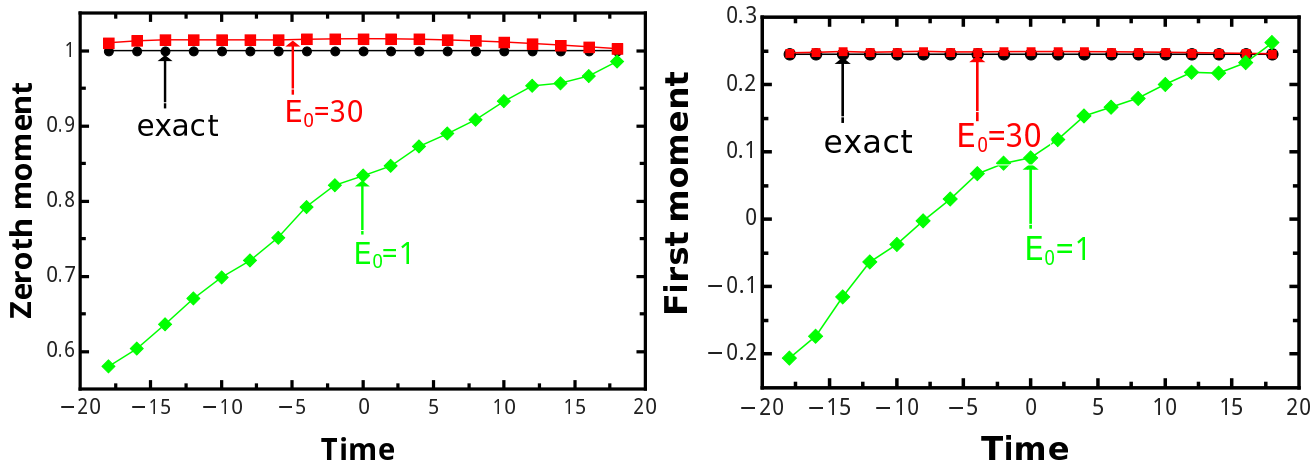

Figure 2. (Color online.) Zeroth (left panel) and first (right panel) moments for the retarded Green's function for different values of E_0 (other parameters are $t_0 = -10$, $\omega_p = 2$, and $\sigma_p = 3$). The Coulomb interaction is $U = 0.5$ ($T_c = 0.0336$) and the temperature is $T = 0.0178$ with $\Delta n_f = 0.49$. The black circles correspond to the exact value of the respective moment; the red squares are for $E_0 = 30$ and the green diamonds are for $E_0 = 1$. The results for the larger field are much more accurate than for the smaller field. These results employed a quadratic extrapolation with the time discretizations $\Delta t = 0.066, 0.05, 0.033$. One needs smaller time discretizations to achieve accurate results for lower field amplitudes.

In Fig. 3, we show the results for the PES response function (with the large field pump pulse $E_0 = 30$) for different widths σ_b of the probe pulse. In these results, the maximum of the probe pulse occurs at the same time as the maximum of the pump pulse ($t_0' = t_0 = -10$). If the width of the probe pulse is small $\sigma_b = 5$ (left panel of Fig. 3), there are single-peak curves for each of the A and B sublattices. The response on the B sublattice is peaked around $-U/2$ (lower band), which arises because there are essentially no electrons available to hop in the upper band under the pump pulse (almost all electrons lie on the A sublattice at this initial temperature $T = 0.0178$). On the A sublattice, the PES response is peaked around $U/2$ (upper zone) because the electrons are excited by the pump pulse into the upper zone. If the width of the probe pulse is large $\sigma_b = 15$ (right panel of Fig. 3), the PES response function shows a many-peak structure. The magnitudes of these peaks are almost three times smaller than in those seen in the previous case with a small probe width but they also are narrower due to the enhanced energy resolution.

It is important to note here that the CDW gap is completely gone in this case. The nonequilibrium excitation of the system dresses the band structure so that the CDW gap disappears. What is amazing, is that it does so despite the fact that the instantaneous eigenvalues of the conduction electrons are unchanged by the Peierls' substitution. While this result may sound bizarre, it occurs because the many-body density of states for a time-dependent system is not given by the density of the instantaneous eigenstates, but is more complex due to the time dependence (essentially because the eigenstate basis changes with time). This also occurs when the order parameter for the heavy electrons is fixed at its equilibrium value and that for the conduction electrons does not equal zero at this point (not shown here).

4. CONCLUSIONS

In this work, we described the general formalism for how to solve for the PES function in an ordered CDW phase. For concreteness we chose to examine the Falicov-Kimball model. We derived analytical expressions for

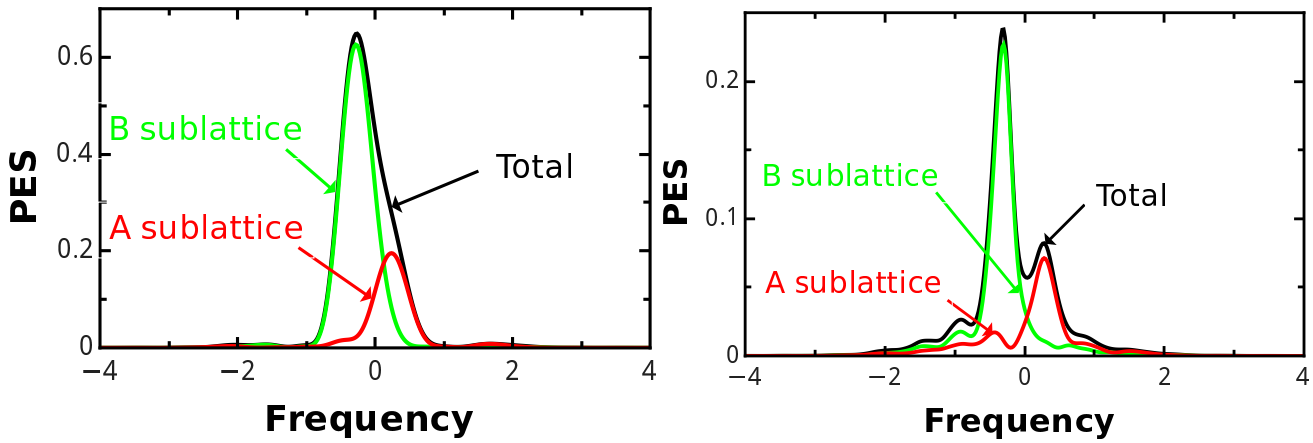

Figure 3. (Color online.) PES function for the interaction $U = 0.5$ ($T_c = 0.0336$) at the temperature $T = 0.0178$ with $\Delta n_f = 0.49$. The pump pulse is $E_0 = 30$ and the maximum is at the time $t_0 = -10$, other parameters are: $\omega_p = 2$, and $\sigma_p = 3$. Probe pulses widths are $\sigma_b = 5$ (left panel) and $\sigma_b = 15$ (right panel). The time of the maximum of the probe pulse is $t_0' = -10$.

the time-dependent lattice Green's functions defined on the Kadanoff-Baym-Keldysh contour. We benchmarked the accuracy of the PES response on the parameters of the pump and probe pulses. We showed that the collapse of the gap in the DOS naturally occurs in these systems when the pump pulse is on. In future work, we will describe other values of the interaction and show other delay times to get a more complete picture of this behavior.

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