Theory of the time-resolved spectral function of high-temperature superconductors with bosonic modes

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We develop a three-temperature model to simulate the time dependence of the electron and phonon temperatures in high-temperature superconductors displaying strong anisotropic electron-phonon coupling. This model not only takes the tight-binding band structure into account but also is valid in the superconducting state. Based on this model, we calculate the time-resolved spectral function via the double-time Green's functions. We find that the dip-hump structure evolves with the time delay. More interestingly, an unusual structure is obtained when the phonons are pumped directly. This finding may serve as a direct evidence for electron-vibration mode coupling.

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I. INTRODUCTION

The physics underlying pairing in high-temperature superconductors (HTSC) remains a topic of current interest. The small isotope effect[1] of optimally doped samples and the very nature of the *d*-wave pairing suggested the importance of a spin-based pairing interaction. Indeed the dip-hump structure of the density of states observed in electron tunneling spectroscopy[2] could be explained with the electronic coupling to spin fluctuations.[3] Due to the improvement of sample quality and momentum resolution, features similar to the electron tunneling spectroscopy have been observed in angle-resolved photoemission spectroscopy (ARPES).[4–13] These features have been equally well explained[14] with the electron-phonon (el-ph) coupling, suggesting that phonon modes that are strongly coupled to electrons may be of relevance. To better understand the physics behind the superconductivity, a good knowledge of the el-ph coupling is essential. Analysis[14] of the spectral function of thermally excited electrons in the cuprates has shown that the out-of-plane and out-of-phase buckling mode strongly couples to the electronic states near the antinodal *M* points in the Brillouin zone while the in-plane breathing mode couples strongly to the electronic states near the *d*-wave nodal points. The signature of the anisotropic el-ph coupling seems to get enhanced in the superconducting state.

However, the usual ARPES only provides the information of the el-ph coupling in thermally populated excitation. Accessing the thermally inaccessible states provides the ability to uncover the signatures of the el-ph coupling, which may not be captured by ARPES. For this reason, time-resolved (TR) ARPES has been developed to offer the capability to capture the single-particle (frequency-domain) and collective (time-domain) information, thus making it possible to directly probe the link between the collective modes and single-particle states. In this setting, either electrons or lattice vibrational modes can be selectively excited with an ultrafast laser pulse. Recent applications of this technique include the studies of transient electronic structure in Mott insulators[15,16] and HTSC (Ref. 17) with optical pump. Furthermore, direct pumping of vibrational mode has also been realized in manganites,[18] though not yet in the cuprates.

Motivated by the thrust of this technique, in this paper, we aim to provide a theoretical underpinning of transient electronic structure for HTSC, in a hope to better understand the nature of phonon modes in these systems. As such, the signature of the el-ph coupling in the cuprates is investigated with the time-resolved spectral function. Our theory consists of two parts. First, we develop a three-temperature model to simulate the time dependence of the electron and phonon temperatures. Then, based on this model, we calculate the time-resolved spectral function up to second order in the coupling constant. The theory is valid in the superconducting state where the quasiparticle excitation is well defined,[3] and may be a good starting point for a better treatment of the non-Fermi-liquid normal state.[19] Our results show a kink structure in the time dependence of the electronic temperature at the superconducting transition temperature. We find that the energy position of the phonon mode in the spectral density is offset by the superconducting energy gap, which is time dependent within the three-temperature model. More interestingly, we obtain another signature of the el-ph coupling when pumping phonons. This signature does not appear when selectively pumping electrons and thus has escaped previous observations. This finding, if confirmed experimentally, will provide a direct evidence for the electron-vibration mode coupling in HTSC.

This paper is organized as follows. In Sec. II, a three-temperature model valid in the superconducting state is presented to simulate the time evolution of the temperatures of electrons, hot phonons, and cold lattice. In Sec. III, we calculate the time-resolved spectral function based on the three-temperature model. The theory is illustrated by applying it to the high-temperature cuprates. Finally conclusions are made in Sec. IV.

II. THREE-TEMPERATURE MODEL

Suppose that the energy transfers within each subsystem (electrons, hot phonons, and cold lattice) are much faster than those between different subsystems. Then a local equilibrium within each subsystem can be reached quickly after pumping. According to the energy conservation, we can

write rate equations for the electron, hot phonon, and lattice temperatures[17] as

$$\frac{\partial T_e}{\partial \tau} = \frac{1}{C_e}\frac{\partial E_e}{\partial \tau} + \frac{P_e}{C_e}, \tag{1}$$

$$\frac{\partial T_{ph}}{\partial \tau} = -\frac{1}{C_{ph}}\frac{\partial E_e}{\partial \tau} + \frac{P_{ph}}{C_{ph}} - \frac{T_{ph}-T_l}{\tau_\beta}, \tag{2}$$

$$\frac{\partial T_l}{\partial \tau} = \left(\frac{C_{ph}}{C_l}\right)\frac{T_{ph}-T_l}{\tau_\beta}, \tag{3}$$

where P_e is the power for pumping electrons, P_{ph} the power for pumping hot phonons, and τ_β the anharmonic decay time of hot phonons. The specific heat of electrons can be calculated from the Boltzmann entropy $S_e=-2k_B\Sigma_{\mathbf{k}}\{[1-f(E_{\mathbf{k}})]\ln[1-f(E_{\mathbf{k}})]+f(E_{\mathbf{k}})\ln f(E_{\mathbf{k}})\}$ by $C_e=T_e\partial S_e/\partial T_e$ while the specific heat of hot phonons for one-vibrational mode can be calculated directly from the phonon energy. The results are given by

$$C_e = \beta_e k_B \sum_{\mathbf{k}} \left[-\frac{\partial f(E_{\mathbf{k}})}{\partial E_{\mathbf{k}}}\right]\left(2E_{\mathbf{k}}^2 + \beta_e \Delta_k \frac{\partial \Delta_k}{\partial \beta_e}\right), \tag{4}$$

$$C_{ph} = \frac{k_B}{4}(\hbar\Omega_0\beta_{ph})^2\left[\coth^2\left(\frac{\hbar\Omega_0\beta_{ph}}{2}\right) - 1\right], \tag{5}$$

respectively, where $\beta=1/k_BT$ and $E_{\mathbf{k}}=\sqrt{\xi_{\mathbf{k}}^2+\Delta_{\mathbf{k}}^2}$ is the superconducting-state quasiparticle energy.

Now we calculate the energy exchange rate $\partial E_e/\partial\tau$ due to el-ph scattering. Let us consider a two-dimensional superconductor exposed to a laser field. The model Hamiltonian for a vibrational mode ν can be written as

$$H = \sum_{\mathbf{k}\sigma}\xi_{\mathbf{k}}c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_{\mathbf{k}}(\Delta_{\mathbf{k}}c_{\mathbf{k}\uparrow}^\dagger c_{-\mathbf{k}\downarrow}^\dagger + \text{H.c.}) + \sum_{\mathbf{q}}\hbar\Omega_{\nu\mathbf{q}} \times\left(b_{\nu\mathbf{q}}^\dagger b_{\nu\mathbf{q}} + \frac{1}{2}\right) + \frac{1}{\sqrt{N_L}}\sum_{\mathbf{kq}\sigma} g_\nu(\mathbf{k},\mathbf{q})c_{\mathbf{k}+\mathbf{q},\sigma}^\dagger c_{\mathbf{k}\sigma}A_{\nu\mathbf{q}} + H_{\text{field}}(\tau), \tag{6}$$

where $c_{\mathbf{k}\sigma}^\dagger$ ($b_{\nu\mathbf{q}}^\dagger$) and $c_{\mathbf{k}\sigma}$ ($b_{\nu\mathbf{q}}$) are the creation and annihilation operators for an electron with momentum $\mathbf{k}$ and spin σ (phonon with momentum $\mathbf{q}$ and vibrational mode ν), $A_{\nu\mathbf{q}}=b_{\nu,-\mathbf{q}}^\dagger+b_{\nu\mathbf{q}}$, $\xi_{\mathbf{k}}$ is the normal-state energy dispersion, μ the chemical potential, $\Delta_{\mathbf{k}}$ the gap function, and g_ν the coupling matrix. A similar Hamiltonian with an explicit form of electron-laser field interaction has been used for a microscopic simulation of optical conductivity.[20] By performing the Bogoliubov-de Gennes transformation,[21] $c_{\mathbf{k}\uparrow}=u_{\mathbf{k}}\alpha_{\mathbf{k}}-v_{\mathbf{k}}\beta_{\mathbf{k}}^\dagger$ and $c_{-\mathbf{k}\downarrow}=u_{\mathbf{k}}\beta_{\mathbf{k}}+v_{\mathbf{k}}\alpha_{\mathbf{k}}^\dagger$, where $u_{\mathbf{k}}^2=(1+\xi_{\mathbf{k}}/E_{\mathbf{k}})/2$ and $v_{\mathbf{k}}^2=(1-\xi_{\mathbf{k}}/E_{\mathbf{k}})/2$, we obtain $E_e=\Sigma_{\mathbf{k}}E_{\mathbf{k}}(\langle\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}}\rangle-\langle\beta_{\mathbf{k}}\beta_{\mathbf{k}}^\dagger\rangle)$. Differentiation of both sides of the expression for E_e with respect to τ and calculating the equations of motion for $\langle\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}}\rangle$ and $\langle\beta_{\mathbf{k}}\beta_{\mathbf{k}}^\dagger\rangle$ to second order in g_ν lead to

$$\frac{\partial E_e}{\partial \tau} = \frac{4\pi}{N_L}\sum_{\mathbf{kq}} g_\nu^2(u_{\mathbf{k}}u_{\mathbf{k}-\mathbf{q}} - v_{\mathbf{k}}v_{\mathbf{k}-\mathbf{q}})^2\delta(E_{\mathbf{k}-\mathbf{q}} - E_{\mathbf{k}} - \Omega_0) \times \Omega_0[e^{(\beta_{ph}-\beta_e)\Omega_0} - 1]f_{\mathbf{k}}(1-f_{\mathbf{k}-\mathbf{q}})N_{\Omega_0}, \tag{7}$$

where $f_{\mathbf{k}}=f(E_{\mathbf{k}})=1/(e^{\beta_e E_{\mathbf{k}}}+1)$ and $N_{\Omega_0}=N(\Omega_0)=1/(e^{\beta_{ph}\Omega_0}-1)$. Here we have specifically set the mode frequency $\Omega_\nu=\Omega_0$.

Equations (1)–(5) and (7) constitute our three-temperature model. The original version of this model was phenomenologically proposed[17] as an extension of the two-temperature model[22] for the normal state. Compared to the original version, the present model has the additional ingredients: (i) it incorporates the detailed band structure, (ii) it is valid for the superconducting state and will essentially reduce to the original version[17] in the normal state, (iii) it includes the anisotropic effect on the el-ph coupling, and (iv) it offers the capability of selectively pumping electron and phonon degrees of freedom.

Next we apply our three-temperature model to simulate the time evolution of the electron, hot phonon, and lattice temperatures for a d-wave superconductor. We use a five-parameter tight-binding model[23] to describe the normal-state energy dispersion. A feature of this dispersion is a flat band with a saddle point at M points. (Unless specified explicitly, energy is measured in units of t and time in units of $\hbar/t$ hereafter. For $t=150$ meV used here, it corresponds to 1740 K in temperature and $\hbar/t$ to 4.4 fs in time.) The d-wave gap function has the form $\Delta_k=\Delta_0(T_e)(\cos k_x-\cos k_y)/2$. The temperature-dependent part is given by[24] $\Delta_0(T_e)=\Delta_{00}\tanh\{(\pi/z)\sqrt{ar(T_c/T_e-1)}\}$, where $z=\Delta_{00}/(k_BT_c)$. In our calculations, we set $\Delta_{00}=0.2$, the critical temperature $T_c=0.06$, the specific-heat jump at T_c is $r=\Delta C_e/C_e\sim 1.43$, and $a=2/3$. Here we take the buckling mode $g_{B_{1g}}$ (Refs. 25–27) as an example to illustrate the theory. To be consistent with experiment,[14,17] we take $g_0=0.4$, $\Omega_0=0.3$, $\tau_\beta=200$, and $C_{ph}/C_l=0.2$. The pump power is a Gaussian pulse $P=P_0e^{-\tau^2/(2\sigma^2)}$, with the full width at half maximum being 2.35 σ. To pump electrons, we set $P_0=0.15$ and $\sigma=1$. Since the energy scale of phonons is much smaller than that of electrons, we set $P_0=0.006$ and $\sigma=100$ for pumping phonons. The convolution product in momentum space is performed via fast Fourier transform with the lattice size 1024×1024 in the Brillouin zone.

Figure 1 displays the temporal evolution of three respective temperatures for (a) pumping electrons and (b) hot phonons. Before pumping, all three types of degrees of freedom (DoF) are in equilibrium, which is set at $T_e=T_{ph}=T_l=0.01$. As shown in Fig. 1(a), when the pump pulse is absorbed by the electrons, the electronic temperature rises steeply around $\tau=0$, and reaches the maximum after a small time delay. It then begins to drop at a time scale determined by the el-ph coupling strength, followed by a slower relaxation. Interestingly, we also observe a small kink in T_e at $T_e=T_c$, due to the Cooper pair breakup, resulting in the dramatic change in the rise rate of T_e, as shown in the inset of Fig. 1(a). This kink was not captured in the early model.[17] Simultaneously, the hot phononic temperature T_{ph} rises smoothly via energy exchange with electrons and then drops

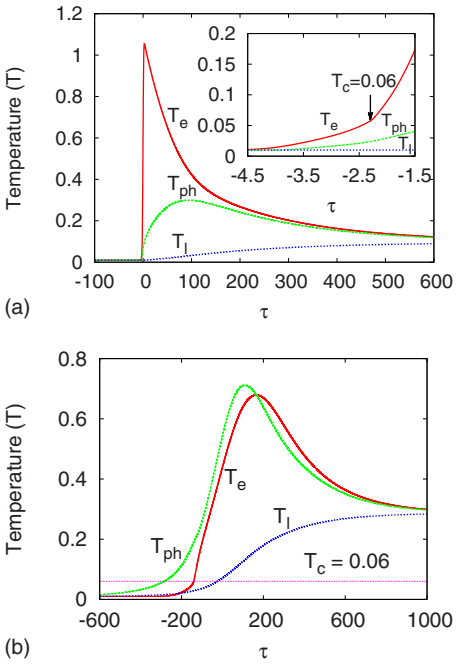

FIG. 1. (Color online) Time evolution of electron (T_e), phonon (T_{ph}), and lattice (T_l) temperatures for selectively exciting (a) electrons and (b) phonons with a laser pulse.

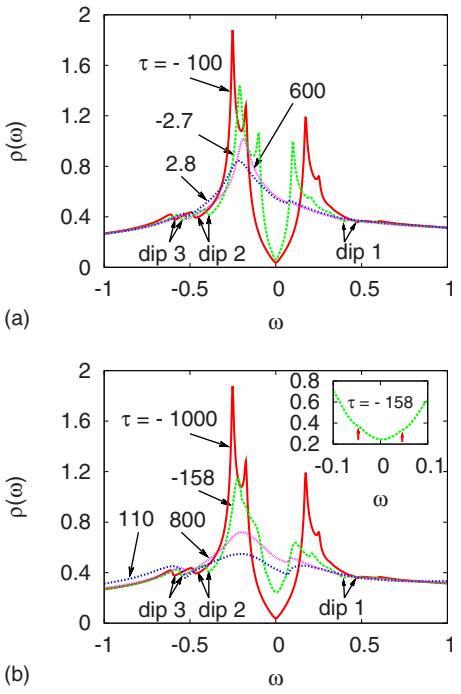

FIG. 2. (Color online) Time evolution of the density of states for selectively exciting (a) electrons and (b) phonons, respectively.

by exchanging energy with the cold lattice via anharmonic cooling, causing the slight increase in T_l. When directly pumping phonons, the time dependence of the temperatures is similarly observed, including the kink in T_e, as shown in Fig. 1(b). Due to the large pumping width, the kink is more easily visible in this case. It is worth pointing out that when pumping electrons, the superconducting energy gap can be suppressed directly, while for pumping phonons it can be suppressed only through the el-ph coupling.

III. TIME-RESOLVED SPECTRAL FUNCTION

The time-resolved spectral function is defined as

$$A(\mathbf{k},\omega) \equiv -\frac{2}{\pi}\mathrm{Im}\,\mathcal{G}_{11}(\mathbf{k},\omega). \tag{8}$$

Here $\mathcal{G}_{11}$ is the one-one component of the retarded Green's function $\hat{\mathcal{G}}(\mathbf{k},\omega)$, which is related to the self-energy by $\hat{\mathcal{G}}^{-1}(\mathbf{k},\omega)=\hat{\mathcal{G}}_0^{-1}(\mathbf{k},\omega)-\hat{\Sigma}(\mathbf{k},\omega)$, with $\hat{\mathcal{G}}_0^{-1}(\mathbf{k},\omega)=\omega\hat{\sigma}_0-\Delta_{\mathbf{k}}\hat{\sigma}_1-\xi_{\mathbf{k}}\hat{\sigma}_3$. $\hat{\sigma}_{0,1,2,3}$ are the unit and Pauli matrices. As is known,[14] in the equilibrium state with $T_e=T_{ph}$, the self-energy can be evaluated more conveniently within the imaginary-time Green's-function approach, in which a key step is to convert the Bose-Einstein distribution to the Fermi distribution, $n_B(i\omega_n \pm E_{\mathbf{k}-\mathbf{q}})=-n_F(\pm E_{\mathbf{k}-\mathbf{q}})$ [with $\omega_n=(2n+1)\pi T$, $T=T_e=T_{ph}$]. For the current situation, the temperatures of electrons and hot phonons are no longer tied to each other and the above conversions are not valid any more. To avoid this restriction, here we apply the double-time Green's-function approach[28] to calculate $\hat{\Sigma}(\mathbf{k},\omega)$. The result is

$$\begin{aligned}\hat{\Sigma}(\mathbf{k},\omega) = \frac{1}{N_L}\sum_{\mathbf{q}} |g_\nu(\mathbf{k},-\mathbf{q})|^2\{&[(\omega-\Omega_0)\Phi_1-(\omega+\Omega_0)\Phi_2 \\ &+\Omega_0(\Phi_3+\Phi_4)]\hat{\sigma}_0+[2E_{\mathbf{k}-\mathbf{q}}(\Phi_1-\Phi_3)+2\Omega_0(\Phi_3 \\ &-\Phi_4)]\hat{\sigma}_1+[\xi_{\mathbf{k}}(\Phi_1-\Phi_2)+(\Omega_0\xi_{\mathbf{k}}/E_{\mathbf{k}})(\Phi_3 \\ &-\Phi_4)]\hat{\sigma}_3\},\end{aligned} \tag{9}$$

where $\Phi_1=N(\Omega_0)/[(\omega-\Omega_0+E_{\mathbf{k}-\mathbf{q}})(\omega-\Omega_0-E_{\mathbf{k}-\mathbf{q}})]$, $\Phi_2=N(-\Omega_0)/[(\omega+\Omega_0+E_{\mathbf{k}-\mathbf{q}})(\omega+\Omega_0-E_{\mathbf{k}-\mathbf{q}})]$, $\Phi_3=f(-E_{\mathbf{k}-\mathbf{q}})/[(\omega+\Omega_0-E_{\mathbf{k}-\mathbf{q}})(\omega-\Omega_0-E_{\mathbf{k}-\mathbf{q}})]$, and $\Phi_4=f(E_{\mathbf{k}-\mathbf{q}})/[(\omega+\Omega_0+E_{\mathbf{k}-\mathbf{q}})(\omega-\Omega_0+E_{\mathbf{k}-\mathbf{q}})]$. In our calculation, a bare $\hat{\mathcal{G}}_0$ is used. This is the approximation suggested in Refs. 14 and 29, where it was found that this self-energy reproduces the main features of the experimental data well for optimally doped sample. Since the coupling constant for the cuprates is small, self-consistency effect on the self-energy is small and is thus neglected in this work. Notice that the dynamics due to the perturbative Hamiltonian (6) enters the spectral function through the three-temperature model.

For simplicity, let us first look at the density of states (DOS), which is defined by $\rho(\omega)=\frac{1}{N_L}\Sigma_{\mathbf{k}}A(\mathbf{k},\omega)$. For direct pumping of electrons, we calculate the DOS at time sequences $\tau=-100$, -2.7, 2.8, and 600, respectively. The results are plotted in Fig. 2(a). At the initial time $\tau=-100$ where $T_e=0.01$ and the system is in the superconducting state [$\Delta_0(T_e)=0.19$], we observe two signatures of the el-ph coupling located at $\pm[\Omega_0+\Delta_0(T_e)]$, as shown in Fig. 2 (dip 1 and dip 2). They are symmetric with respect to $\omega=0$ but dip 2 is much stronger. It is related to the fact that the normal-state Van Hove singularity is located below the Fermi energy and as such the coherent peak at $-\Delta_0(T_e)$ has stronger inten-

sity than that at $\Delta_0(T_e)$. In addition, dip 3 occurs near the characteristic energy $-(E_M+\Omega_0)$, where E_M is the quasiparticle energy at $\mathbf{k}=(\pi,0)$ or equivalent wave-vector points in Brillouin zone. It arises from the Van Hove singularity at $\mathbf{k}=(\pi,0)$. The signature at the energy $E_M+\Omega_0$ is much weaker also because of the Van Hove singularity location in the normal state. (These anomalies in the equilibrium state have been discussed in Ref. 30.) At $\tau=-2.7$ where $T_e=0.041$ and $\Delta_0(T_e)\approx 0.1$, similar behaviors are observed. However, at $\tau=2.8$, $T_e=1.05$ and thus only the signature of the normal-state el-ph coupling (dip 3) can be observed. At $\tau=600$, a similar structure of the normal-state el-ph coupling is observed.

For direct pumping of phonons, we choose different time sequences $\tau=-1000$, -158, 110, and 800 to evaluate the DOS. The results are displayed in Fig. 2(b). From Fig. 2(b), we observe that the dips suggesting the el-ph coupling in both normal and superconducting states becomes stronger than those for selectively pumping electrons, as expected. In particular, we observe two small dips, as shown in the inset of Fig. 2(b), at $\omega=\pm 0.05$ for $\tau=-158$ (at which $T_e=0.041$ and $T_{ph}=0.163$), both of which arise from the poles of $\Sigma_{\mathbf{k}}(\omega)$ at $\omega=\pm[\Omega_0-\Delta_0(T_e)]$. This is because the temperature of hot phonons is higher when pumping phonons than when pumping electrons at the same T_e, and thus the contributions from the terms as weighted by $N(\pm\Omega_0)$ are significantly enhanced. Figure 2(b) clearly shows that, as time elapses from $\tau=-1000$, -158, to 110, the peaks move toward the *zero energy*, while from $\tau=110$ to 800, the locations of these two peaks remain unchanged. This can be understood by considering that, after pumping, T_e rises with the time delay, while $\Delta_0(T_e)$ decreases and rapidly vanishes when T_e reaches T_c.

Finally we numerically evaluate the time-resolved spectral function for the two excitations discussed above at those time sequences chosen for calculating the DOS. In this calculation, we consider cuts along k_x axis in the Brillouin zone at a k_y chosen near the zone boundary. Figure 3 shows the snapshots of the image of the spectral function $A(\mathbf{k},\omega)$ as a function of k_x and ω. From (a_1) and (a_2) in Fig. 3, we observe kinks at $\omega=-[\Delta_0(T_e)+\Omega_0]$ (red lines) and $-(E_M+\Omega_0)$ (black lines) at $\tau=-100$ and -2.7. The red line is shifted in energy as time elapses from $\tau=-100$ to $\tau=-2.7$ because the BCS gap $\Delta_0(T_e)$ drops significantly from 0.19 to 0.1. These kinks correspond to the dip structures discussed in Fig. 2(a). As $\tau>-2.3$, the superconducting gap vanishes, so do the kinks marked with red lines [see (a_3) and (a_4) in Fig. 3]. The kinks marked with black lines persist through the whole time sequence, which can be ascribed to the el-ph coupling signature in the normal state. Therefore, we have every reason to regard these kinks marked with red lines as the signature of the el-ph coupling in the superconducting state. For hot phonon pumping [$(b_1)-(b_4)$ in Fig. 3], a kink structure at $-[\Omega_0-\Delta_0(T_e)]$(marked with magenta line in (b_2) of Fig. 3 is also observed. These observations are consistent with our finding in the DOS.

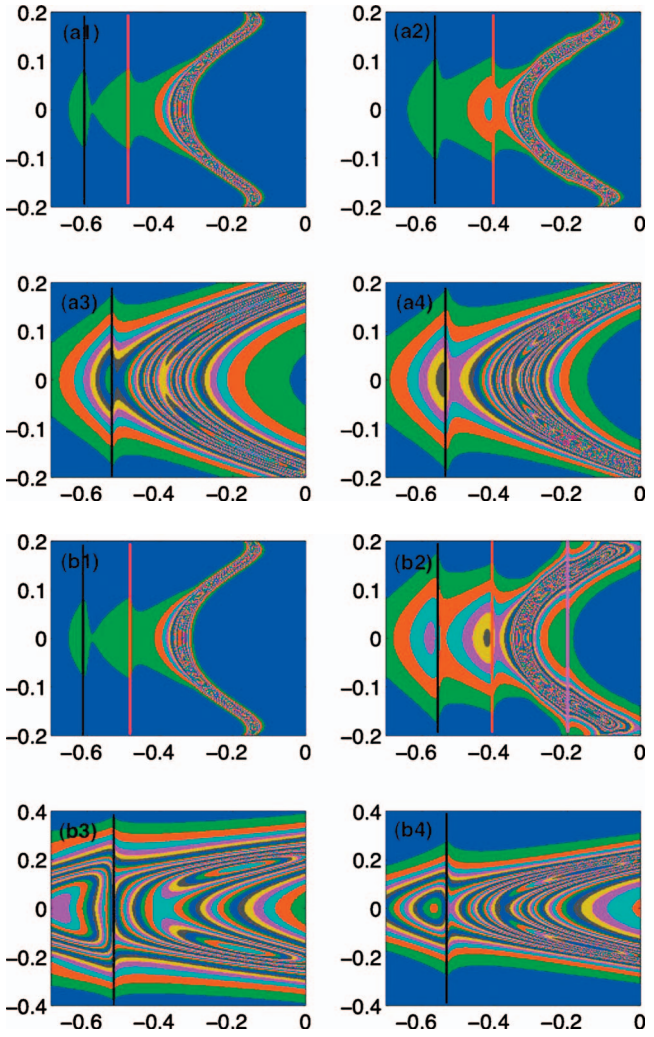

FIG. 3. (Color) Time-resolved electronic spectral function for pumping electrons $[(a_1)-(a_4)$ for $\tau=-100$, -2.7, 2.8, and 600] and phonons $[(b_1)-(b_4)$ for $\tau=-1000$, -158, 110, and 800], respectively. The vertical axis is k_x/π at a fixed $k_y=0.75\pi$ and the horizontal axis is energy. The black, red, and magenta lines represent the energy location, $-(E_M+\Omega_0)$, $-[\Delta_0(T_e)+\Omega_0]$, and $-[\Omega_0-\Delta_0(T_e)]$, corresponding to the dip or hump location discussed in Fig. 2. The red or blue color indicates the maximum or minimum of the spectral function.

IV. CONCLUSION

In conclusion, we have developed a three-temperature model to simulate the time evolution of the temperatures of electrons, hot phonons, and the lattice. Our model takes both quasiparticle excitation and relaxation into account. The theory is valid in the superconducting state. Based on this model, we derive a formula for the time-resolved spectral function, allowing us to investigate the dynamics of the el-ph coupling in HTSC. Our study shows that in the superconducting state, the el-ph coupling signature gets enhanced, suggesting the relevance of the el-ph coupling to the superconductivity. In the viewpoint that the electronic spin fluctuations arise from the strong correlation within the electronic DoF itself, and if one can assume that the spin fluctuations will ride on the electrons and thus its effective temperature will be tied to the electronic temperature, direct pumping of hot phonons will provide a unique way to differentiating the bosonic modes being of electronic or phononic

origin, to which electronic quasiparticles are strongly coupled.

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