<table>
<thead>
<tr>
<th><strong>Title</strong></th>
<th>Theory of ultrafast quasiparticle dynamics in high-temperature superconductors: the dependence on pump fluence</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Author(s)</strong></td>
<td>Tao, Jianmin; Prasankumar, Rohit P.; Chia, Elbert E. M.; Taylor, Antoinette J.; Zhu, Jian-Xin</td>
</tr>
<tr>
<td><strong>Date</strong></td>
<td>2012</td>
</tr>
<tr>
<td><strong>URL</strong></td>
<td><a href="http://hdl.handle.net/10220/9304">http://hdl.handle.net/10220/9304</a></td>
</tr>
<tr>
<td><strong>Rights</strong></td>
<td>© 2012 American Physical Society. This paper was published in Physical Review B and is made available as an electronic reprint (preprint) with permission of American Physical Society. The paper can be found at the following official DOI: [<a href="http://dx.doi.org/10.1103/PhysRevB.85.144302">http://dx.doi.org/10.1103/PhysRevB.85.144302</a>]. One print or electronic copy may be made for personal use only. Systematic or multiple reproduction, distribution to multiple locations via electronic or other means, duplication of any material in this paper for a fee or for commercial purposes, or modification of the content of the paper is prohibited and is subject to penalties under law.</td>
</tr>
</tbody>
</table>
Theory of ultrafast quasiparticle dynamics in high-temperature superconductors:

The dependence on pump fluence

Jianmin Tao,1,∗ Rohit P. Prasankumar,2 Elbert E. M. Chia,3 Antoinette J. Taylor,2 and Jian-Xin Zhu1,1

1Theoretical Division and Center for Nonlinear Studies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
2Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
3Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore SG-637371, Singapore

(Received 16 December 2011; revised manuscript received 7 February 2012; published 6 April 2012)

We present a theory for the time-resolved optical spectroscopy of high-temperature superconductors at high excitation densities with strongly anisotropic electron-phonon coupling. A signature of the strong coupling between the out-of-plane, out-of-phase O-buckling mode (B_{1g}) and electronic states near the antinode is observed as a higher-energy peak in the time-resolved optical conductivity and Raman spectra, while no evidence of strong coupling between the in-plane Cu-O-breathing mode and nodal electronic states is observed. More interestingly, it is observed that under appropriate conditions of pump fluence, this signature exhibits a reentrant behavior with time delay, following the fate of the superconducting condensate.

DOI: 10.1103/PhysRevB.85.144302 PACS number(s): 74.25.nd, 74.72.—h, 71.38.—k, 74.25.Gz

I. INTRODUCTION

Since its discovery in 1986, high-temperature superconductivity in cuprates has been a central topic of study in condensed-matter physics. It is now widely believed that Cooper-pair formation is essential for the superconducting condensate in these systems. However, the nature of the mediator (or glue) responsible for Cooper pairing remains hotly debated. Although the interaction of electrons with lattice vibrations is not likely to solely account for the essential properties of high-Tc superconductors (HTSCs), many probes, including angle-resolved photoemission,1–5 inelastic neutron scattering,6,7 tunneling,7,8 and Raman9 spectroscopies, have revealed that electron-phonon interactions have significant effects on various properties. Complementary to the time-integrated techniques, different ultrafast pump-probe techniques10–15 have been used to disentangle microscopic interactions in HTSCs. These techniques aim to study the recombination of photoexcited quasiparticles and the resulting recovery of the superconducting condensate. In HTSCs, time-resolved (TR) angle-resolved photoemission spectroscopy16 and TR optical reflectivity17 have indicated that the excited quasiparticles preferentially couple to a small number of phonon subsets before decaying through anharmonic coupling to all other lattice vibrations in support of the notion that selective optical-phonon modes give rise to anisotropy of the electron-phonon (el-ph) coupling. In addition, this anisotropy has also been observed in TR electron diffraction,18 while the resonant femtosecond study of both electronic and phononic degrees of freedom suggests strong el-ph coupling19,20.

Despite considerable progress in pump-probe experimental studies, work on microscopic modeling of the influence of el-ph interactions on observables, such as time-dependent optical conductivity or Raman spectra, is very limited. Understanding the nonequilibrium dynamics of quantum many-body systems has, in fact, posed a theoretical challenge. Historically, theoretical attempts to model the time evolution of properties have either used quasiequilibrium models such as T* and μ* models20 to describe nonequilibrium excitations created by a pump-pulse21 or rate-equation approach based on the phenomenological Rothwarf-Taylor model22 to describe the recovery dynamics of the superconducting state. Recently, the time evolution of the optical conductivity has been studied within a microscopic model that treats the excitation and relaxation dynamics on the same footing.23 All these theories are suitable for pump-probe experiments with low excitation fluence wherein the superconducting condensate is merely perturbed but not destroyed. A picture of the dynamics of quasiparticles and the superconducting condensate in the photoinduced phase-transition regime24–26 which is impulsively driven by a high excitation fluence, has as yet been beyond reach.

Here we formulate a theory for the TR optical conductivity and TR Raman scattering in HTSCs in the regime of intermediate-to-high-intensity pump fluence. The theory is aimed to address directly the situation in which the superconducting condensate can be destroyed by a pump pulse. It is based on an effective-temperature model for different subsystems contributing to the response: electrons, hot phonons (i.e., out-of-plane, out-of-phase O-buckling B_{1g} phonons and half-breathing in-plane Cu-O-bond-stretching phonons) that are strongly coupled to electrons, and the cold lattice. The model phenomenologically includes the effect of the pump pulse but addresses in greater depth the electron-hot-phonon coupling based on a microscopic model Hamiltonian for d-wave superconductivity in HTSCs. This microscopic treatment goes beyond previous effective models for the normal state,16,27 allowing us to describe the quasiparticle dynamics in both the normal and superconducting states with the same approach. Within this unified model, the time evolution of the whole set of experimental measurables can be calculated in a streamlined way. Our first test of this approach considered the B_{1g} phonons as the only hot-phonon mode in the calculation of the TR spectral function for a very high pump fluence.28 In the present work, we include the half-breathing phonons as a second hot-phonon mode in the calculation of the TR optical spectroscopy. Importantly, the influence of excitation density on quasiparticle dynamics in HTSCs is also investigated. Our calculations show that,
in the superconducting state, in addition to the peak in the optical conductivity and Raman spectra due to the Drude response, there is another peak at higher frequencies. This high-frequency peak disappears when the system evolves into the normal state but recurs if the superconducting condensate is recovered, suggesting the significance of the superconducting gap in the TR optical properties.

The outline of the paper is as follows. In Sec. II, we lay down the effective-temperature model for a \( d \)-wave superconductor with electronic coupling to both \( B_{1g} \) and half-breathing stretching-phonon modes. The time-dependent effective temperatures for the respective subsystems are evaluated depending on the strength of the pump fluence. With the obtained time dependence of effective temperatures, the time-resolved optical conductivity and Raman spectra and their pump-fluence dependences are presented in Secs. III and IV, respectively. Finally, a conclusion is given in Sec. V.

### II. EFFECTIVE-TEMPERATURE MODEL

Let us consider a two-dimensional superconductor exposed to a laser field. The model Hamiltonian can be written as\(^{23,28}\)

\[
H = \sum_{k\sigma} \xi_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_k (\Delta_k c_{k\sigma}^\dagger c_{-k\sigma} + \text{H.c.}) + \sum_{\nu} \hbar \Omega_{\nu} q \times \left( b_{\nu}^\dagger b_{\nu} + \frac{1}{2} \right) + \frac{1}{\sqrt{N_c}} \sum_{kq\sigma} g_{\nu}(k,q) c_{k+q\sigma}^\dagger c_{k\sigma} A_{\nu q} + H_{\text{local}}(\tau),
\]

where \( c_{k\sigma}^\dagger (b_{\nu}^\dagger) \) and \( c_{k\sigma} (b_{\nu}) \) are the creation and annihilation operators for an electron with momentum \( k \) and spin \( \sigma \) (and for a phonon with momentum \( q \) and vibrational mode \( \nu \), where \( \nu = 1, 2 \), representing the \( B_{1g} \) and half-breathing modes, respectively), \( A_{\nu q} = b_{\nu q}^\dagger \) \( + \) \( b_{\nu q} \), \( \xi_k \) is the normal-state energy dispersion, \( \mu \) is the chemical potential, \( \Delta_k = (\Delta_0/2)(\cos k_x - \cos k_y) \) is the \( d_{x^2-y^2} \)-wave-gap function, \( N_c \) is the total number of lattice sites, and \( g_{\nu} \) is the coupling matrix. Following the procedure sketched in Ref. 28, we arrive at a four-temperature model:

\[
\begin{align*}
\frac{\partial T_e}{\partial \tau} &= \frac{1}{C_e} \sum_{\nu} K_e(T_e, T_{\nu ph}) + \frac{P_e}{C_e}, \\
\frac{\partial T_{\nu ph}}{\partial \tau} &= -\frac{K_e(T_e, T_{\nu ph})}{C_{\nu ph}} - \frac{T_{\nu ph} - T_i}{\tau_{\nu ph}}, \\
\frac{\partial T_i}{\partial \tau} &= \sum_{\nu} \left( \frac{C_{ph,\nu}}{C_i} \right) T_{\nu ph} - T_i \frac{T_{\nu ph} - T_i}{\tau_{\nu ph}}.
\end{align*}
\]

Here, \( K_e \) is the el-ph-coupling kernel, which can be calculated from the model Hamiltonian (1) with the equation-of-motion approach. It is given by

\[
K_e = \frac{4\pi}{N_L} \sum_{kq} g_{\nu}^2 (u_k u_{k-q} - v_k v_{k-q})^2 \delta(\varepsilon_k - \varepsilon_q - \hbar \Omega_{\nu q})
\times \Omega_{\nu q} e^{(\beta_{\nu k} - \beta_{\nu q})/k_B T_e} \Omega_{\nu q},
\]

where the Bogoliubov amplitudes are \( u_k = [(1 + \xi_k/\varepsilon_k)^{1/2}] \) and \( v_k = \text{sgn}(|\Delta_k|)(1 - \xi_k/\varepsilon_k)^{1/2} \) with \( \xi_k = \sqrt{\varepsilon_k^2 + \Delta_k^2} \) being the quasiparticle energy, and the Fermi-Dirac and Bose-Einstein distribution functions are given by \( f_k = f(\varepsilon_k) = 1/(e^{\beta_{\nu k} T_e} + 1) \) and \( N_{\Omega_{\nu q}} = N(\varepsilon_{\Omega_{\nu q}}) = 1/(e^{\beta_{\nu k} T_e} + 1) \) with \( \beta_{\nu k} = 1/k_B T_{\nu ph} \) respectively. In Eq. (2), the specific heat for electrons per unit cell is found to be

\[
C_e = \frac{\beta_k k_B}{N_L} \sum_k \left[ - \frac{\partial f(\varepsilon_k)}{\partial \varepsilon_k} \right] \left( 2 E_k^2 + \beta_k \Delta_k \frac{\partial \Delta_k}{\partial \beta_k} \right),
\]

while that for each hot-phonon mode is given by

\[
C_{\nu ph} = \frac{k_B}{4} \left( \beta \Omega_{\nu ph}^2 \right)^2 \left( \coth^2 \left( \frac{\hbar \Omega_{\nu ph}}{2 T_e} \right) - 1 \right)
\]

in the Einstein-mode approximation \( \Omega_{\nu q} = \Omega_e \). Finally, \( P_e \) is the power intensity (i.e., power per unit cell) for pumping electrons, and \( \tau_{\nu ph} \) is the anharmonic-decay time of each hot-phonon mode.

Throughout this paper, we use a five-parameter tight-binding model\(^{29}\) to describe the normal-state energy dispersion, which is typical of optimally doped Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8-x}\) (Bi-2212):

\[
\xi_k = -2t(\cos k_x + \cos k_y) - 4t'(\cos k_x \cos k_y - \sin k_x \sin k_y - 2t''(\cos k_x + \cos k_y) - 4t'''(\cos k_x \cos k_y) - 4t''''(\cos k_x \cos k_y - \mu).
\]

where the hopping integrals are \( t = 1 \), \( t' = -0.25 \), \( t'' = 0.0872 \), \( t''' = 0.0938 \), \( t'''' = -0.0857 \), and \( \mu = -0.8772 \). The absolute energy of \( t = 150 \) meV. A feature of this dispersion is a flat band with a saddle point at the \( M \) points of the Brillouin zone. The temperature dependence of the \( d \)-wave-gap magnitude is given by\(^{30}\)

\[
\Delta_0(T_e) = \Delta_0 \tanh[(\pi/\tau)(ar(T_e/T_c - 1))]
\]

where \( \Delta_0 = \Delta_0_0/(k_B T_e) \). In our calculations, we set \( \Delta_0_0 = 30 \) meV, the critical temperature \( T_c = 104 \) K (from the setting of \( T_e = 0.06 \) for simplicity), the specific-heat jump at \( T_c \) as \( \Delta C_e/C_e \sim 1.43 \), and \( a = 2/3 \). We take the anisotropic el-ph coupling in the form given in Refs. 3 and 31 with \( \omega_1 = 45 \) meV and \( g_{10}^2 = 90 \) meV and \( \omega_{\tau} = 70 \) meV and \( g_{20}^2 = 120 \) meV with \( \tau_{\nu q} = \tau_{\nu ph} = 880 \) fs and \( C_{\nu ph} = C_{\nu ph,2} = 0.24 \). The pump is represented by a Gaussian pulse \( P = P_0 e^{-t^2/(2\tau_p^2)} \) with a full width at half maximum (FWHM) of 2.35\( \tau_p \). Hereafter, we assume implicitly \( \hbar = 1 \) and set \( \sigma = 4.4 \) fs, giving a FWHM of about 10.34 fs. This value is smaller than the commonly used experimental values of about 30–50 fs but is indeed close to that value of 12 fs used in the recent TR experiment on YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\).\(^{19}\) We believe that this variation will not affect the qualitative physics as will be presented below. We take the number of \( k \) points to be \( 40 \times 40 \) in the Brillouin zone for the temperature evolution and use 256 \( \times 256 \) for the TR optical conductivity and Raman-scattering spectral function. All calculations are done with the system initially in the superconducting state for \( T < T_c \).

Figure 1 shows the time evolution of the effective temperature for each subsystem for a large pump-power intensity \( P_0 = 0.436 \) \( \mu \) W [Fig. 1(a)] and an intermediate value of \( P_0 = 0.022 \) kW [Fig. 1(b)]. These values of pump power correspond to 120 \( \mu \)J/cm\(^2\) and 6 \( \mu \)J/cm\(^2\) of pump fluence in Bi-2212, respectively, assuming a 60-nm optical-absorption depth. Starting from the initial temperature \( T_c = T_{\nu ph} = T_i = \)
17 K, the electron temperature \( T_e \) increases rapidly after photoexcitation, and the superconductor is driven into the normal state while exhibiting a kink structure at \( T_c \). Further energy relaxation is then slowed down with the kink recurring during the cooling stage [see the inset of Fig. 1(a) and 1(b)] for both power intensities. In addition, for \( \tau = -440 \) fs and \(-11\) fs at which the material is superconducting and \( \Delta_0 \approx 30 \) meV, we observe that \( \sigma_T(\omega) \) exhibits a broad peak at about \( \omega = \Delta_0 + \Omega_{ph,1} \). (The specific location may be affected by several factors although \( 2\Delta_0 + \Omega_{ph,1} \) plays a substantial role.) Our observation is consistent with an earlier study of optical conductivity in the thermal-equilibrium state of HTSCs. In contrast, no

and

\[
I_{II}(\mathbf{k}, \omega) = 2i \int d\tau' \text{Tr} \text{Im}[\hat{\hat{F}}(\mathbf{k}, \tau') \hat{\Delta}(\mathbf{k}, \tau')] e^{i\omega \tau'}. \tag{10}
\]

Here, \( \hat{\Delta}(\mathbf{k}, \tau') \) and \( \hat{\hat{F}}(\mathbf{k}, \tau') \) are the Fourier transforms of the TR spectral functions \( \hat{\Delta}(\mathbf{k}, \omega) \) and \( \hat{\hat{F}}(\mathbf{k}, \omega) \), respectively. In the derivation of Eq. (9), we have used the Hilbert transform

\[
\hat{g}(\mathbf{k}, i\omega_n) = \int_{-\infty}^{\infty} d\epsilon \frac{\hat{\Delta}(\mathbf{k}, \epsilon)}{i\omega_n - \epsilon} \tag{11}
\]

with the single-particle spectral function

\[
\hat{\Delta}(\mathbf{k}, \epsilon) = -\frac{1}{\pi} \text{Im}[\hat{g}(\mathbf{k}, i\omega_n \rightarrow \epsilon + i\delta)], \tag{12}
\]

where the Green’s function \( \hat{g} \) and \( \hat{\Delta} \) are 2 \times 2 matrices in the Nambu space. Since this spectral function as calculated with the method of Ref. 28 is a function of the effective electronic temperature, which is time dependent (see the discussion in Sec. II), it is time resolved. Therefore, the optical conductivity and the Raman spectra as discussed in the next section are also time dependent.

Figure 2 shows the time evolution of the real part of the optical conductivity \( \sigma_T(\omega) \) at several selected time delays. From Fig. 2, one can see that at all time delays, the optical conductivity shows the well-known Drude peak at \( \omega = 0 \) due to the nodal quasiparticles for the \( d \)-wave gap symmetry. In addition, for \( \tau = -440 \) fs and \(-11\) fs at which the material is superconducting and \( \Delta_0 \approx 30 \) meV, we observe that \( \sigma_T(\omega) \) exhibits a broad peak at about \( \omega = 2\Delta_0 + \Omega_{ph,1} \). (The specific location may be affected by several factors although \( 2\Delta_0 + \Omega_{ph,1} \) plays a substantial role.) Our observation is consistent with an earlier study of optical conductivity in the thermal-equilibrium state of HTSCs. In contrast, no

III. TIME-RESOLVED OPTICAL CONDUCTIVITY

Within the Kubo formalism, the real part of the TR optical conductivity is given by

\[
\sigma_T(\omega) = -\frac{\epsilon^2 \text{Im}\Pi(\omega)}{\omega}, \tag{8}
\]

where

\[
\text{Im}\Pi(\omega) = \frac{2\pi^2}{N_L} \sum_k \left( \frac{\partial \xi_k}{\partial k_i} \right)^2 I_{II}(\mathbf{k}, \omega) \tag{9}
\]
IV. TIME-RESOLVED RAMAN-SCATTERING SPECTRUM

The time-resolved Raman-scattering intensity is calculated via a simple relation \[^3\] from the imaginary part of the Raman-response function. In the bare-vertex approximation, \[^3\] it is found as

\[
\text{Im} \chi(i \Omega_m \to \omega + i \delta) = -\frac{2 \pi^2}{N_L} \sum_k \gamma_k^2 I_x(k, \omega),
\]

where

\[
I_x(k, \omega) = 2i \int d\tau \text{Tr} \text{Im} \{\hat{\chi} \hat{F}^\dagger(k, \tau) \hat{F}^\ast(k, \tau') \hat{A}(k, \tau')\} e^{i\omega \tau'}. \quad (14)
\]

Here, \( \chi \) is the nonresonant bare Raman vertex given by \( \gamma_k = \chi_{x, \xi} \) with \( \xi \) being the Pauli matrix and \( \gamma_k = \sum_{\alpha, \beta} \gamma^{\alpha} \chi_{\alpha, \beta} \). \( \gamma^{\alpha} \) are the polarization unit vectors of the incident and scattered photons, respectively, and \( \xi_k \) is the electronic-normal-state dispersion of the conduction band.

Figure 3 shows the time evolution of the Raman-scattering spectrum. When the electron-hot-phonon coupling is switched off, the Raman spectrum rises with \( \omega \) and has a large peak at twice the gap, \( \Delta_0 \), at the initial temperature (red solid line in Fig. 3). A small shoulder in the curve around 90 meV arises due to the van Hove singularity. In the presence of the electron-hot-phonon coupling, the superconducting gap function is renormalized, shifting the original \( \Delta_0 \) peak to lower frequencies. Simultaneously, another peak develops at \( \Delta_0 + \Omega_{\text{ph},1} \), but no peak develops at \( \Omega_{\text{ph},2} \) for the same reason as in the case of the optical conductivity (see Fig. 2). After photoexcitation, this double-peak structure evolves into a very broad peak as the system enters the normal state. For large \( P_0 \), this broad peak remains for a few picoseconds. However, for intermediate \( P_0 \), once the superconducting state recovers, the double-peak structure appears again. This result is fully consistent with our calculations of the TR optical conductivity described above.

V. CONCLUSION

We have presented a theory for the time-resolved optical conductivity and Raman spectra, based on the TR spectral function that we have recently formulated for HTSCs. Our calculations show that the signature of the electron-B1g-mode coupling in the TR optical conductivity and Raman spectra is more pronounced than the consequence of the coupling between electrons and the half-breathing mode. This is the result of a concurrence of the anisotropy of the el-ph coupling, band structure, and \( d \)-wave energy gap in HTSCs. Even more interestingly, this signature also shows a reentrant behavior in concurrence with the superconducting condensate, which can be controlled by the pump fluence. The observation of the broad peak in the TR Raman spectra and its reentrant behavior provides direct evidence of the el-ph coupling.

ACKNOWLEDGMENTS

J.-X.Z. thanks A. V. Chubukov, G. L. Dakovski, T. Durakiewicz, F. Marsiglio, and G. Rodriguez for helpful discussions. This work was supported by the National Nuclear Security Administration of the US DOE at LANL under Contract No. DE-AC52-06NA25396, the US DOE Office of Basic Energy Sciences, and the LDRD Program at LANL.


\[^{2}\] Current address: Department of Physics, Tulane University, New Orleans, Louisiana 70118, USA

\[^{3}\] Author to whom correspondence should be addressed: jxzhu@lanl.gov; http://theory.lanl.gov


