Doping dependence of the electron–phonon and electron–spin fluctuation interactions in the high-$T_c$ superconductor Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

E E M Chia$^{1,8}$, D Springer$^1$, S K Nair$^1$, X Q Zou$^1$, S A Cheong$^1$, C Panagopoulos$^{1,2}$, T Tamegai$^3$, H Eisaki$^4$, S Ishida$^5$, S Uchida$^5$, A J Taylor$^6$ and J-X Zhu$^{7,8}$

$^1$ Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore
$^2$ Department of Physics, University of Crete and FORTH, 71003 Heraklion, Greece
$^3$ Department of Applied Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
$^4$ National Institute of Advanced Industrial Science and Technology, Tsukuba 305-8568, Japan
$^5$ Department of Physics, University of Tokyo, Bunkyo-ku, Tokyo 113-0033, Japan
$^6$ Materials Physics and Applications Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
$^7$ Theoretical Division and Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

E-mail: elbertchia@ntu.edu.sg and jxzhu@lanl.gov

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Abstract. Using ultrafast optical techniques, we detect two types of bosons strongly coupled to electrons in the family of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) from the underdoped to overdoped regimes. The different doping dependences of the electron–boson coupling strengths enable us to identify them as phonons and...
spin fluctuations: electron–phonon coupling ($\lambda_{e-ph}$) peaks at optimal doping, while electron–spin fluctuation coupling ($\lambda_{e-st}$) decreases monotonically with doping. This observation is consistent with two facts: (i) superconductivity is in close proximity with antiferromagnetism at low dopings and (ii) a pronounced lattice renormalization effect at larger dopings.

Despite many advances in understanding copper-oxide high-transition-temperature ($T_c$) superconductors, there still exists no universally accepted theoretical explanation. Determining the nature of interaction responsible for the Cooper-pair formation remains one of the grand challenges in modern condensed matter physics. The most probable candidates are lattice vibrations (phonons) [1, 2], spin fluctuation (SF) modes [3] and pairing without invoking glue [4]. For conventional superconductors, structure in the electron tunneling $dI/dV$ characteristics established unambiguously that the attractive pairing interaction was mediated by phonons [5]. For high-$T_c$ superconductors, structure in $dI/dV$ has also been found in many tunneling measurements [6]. More recent scanning tunneling microscopy (STM) experiments revealed an oxygen lattice vibration mode whose energy is anticorrelated with the local gap value on hole-doped Bi-2212 [7], while a bosonic mode of electronic origin was found in the electron-doped Pr$_{0.88}$LaCe$_{0.12}$CuO$_4$ [8]. Together with salient features observed in angle-resolved photoemission spectroscopy (ARPES) [2, 9], these new results raise the fundamental question of whether the bosonic modes are a pairing glue [10] or a signature of an inelastic tunneling channel [11].

The role of the electron–boson interaction in high-$T_c$ superconductors has been studied by different techniques. For example, inelastic neutron scattering tracks the changes in boson energies or dispersions upon entering the superconducting state [12]. Resonant inelastic x-ray scattering (RIXS) found that the integrated inelastic intensity is conserved on doping from the antiferromagnetic insulator to the slightly overdoped (OD) superconductor, while the magnetic spectral weight strongly decreases with hole doping around the antiferromagnetic ordering wave vector $Q_{AF}$, and remains constant everywhere else [13]. However, RIXS does not give a direct measure of the electron–boson coupling strength. ARPES [14] measures the effects of electron–boson interaction on electronic self-energies, and planar junction experiments determine the energy of the bosonic mode [15]. STM measures the local
density of states through the local differential tunneling conductance, and estimates the characteristic boson mode energy from the dip position [7]. However, it does not give the electron–boson coupling strength directly because both the coupling strength and mode energies are encoded in the electron self-energy itself. Time-integrated optical conductivity spectra were inverted to obtain the electron–boson spectral function and hence the electron–boson coupling constant [16–19], but it is difficult to elucidate whether one or more bosons are involved. Time-resolved pump–probe spectroscopy is a powerful technique used to probe the relaxation dynamics of photoexcited quasiparticles in correlated electron systems such as cuprate [20–24], pnictide [25] and actinide [26] superconductors, spin density wave materials [27], charge density wave materials [28] and heavy fermion systems [29, 30]. Its unique contribution lies in its ability to extract the value of the electron–boson coupling strength (λ) directly, via the electron–boson relaxation time, without the need to perform complicated inversion algorithms. This procedure has been experimentally verified on the conventional superconductors [31]. In this paper, we report measurements of time-resolved quasi-particle relaxation of high-quality single crystals of underdoped (UD) to OD Bi$_2$Sr$_2$CaCu$_2$O$_{8+δ}$ (Bi-2212, hole concentration $p = 0.10–0.22$). Our data indicate the coupling of electrons to two bosonic modes: the electron–phonon coupling constant ($\lambda_{e-ph}$) peaks at optimal doping, while the electron–SF coupling constant ($\lambda_{e-sf}$) decreases monotonically with doping.

1. Experiment

The family of the bi-layer cuprate Bi-2212 has been the most intensively studied class of high-$T_c$ superconductors in recent years, due to their (a) extreme cleavability, (b) containing only CuO$_2$ planes and not chains and (c) the possibility of growing samples with a larger range of $T_c$ variations. Single crystals of Bi-2212 were obtained from three groups (two Tokyo and one Tsukuba) grown by the floating zone method with doping control, yielding values of $T_c$ (determined by magnetization data) that depend on the hole doping level ($p$) spanning from the UD ($p = 0.10$, $T_c = 65$ K) to the OD ($p = 0.22$, $T_c = 65$ K) regime. In the optimally doped (OPT) sample, Ca has been doped with Y to obtain the highest $T_c$ of 95 K. Underdoping was achieved using excess Bi atoms substituted for the Sr sites as well as reducing oxygen content. The more OD samples have been doped with Pb to obtain lower values of $T_c$. The values of $p$ were obtained from the $T_c$ values using the parabolic law [32] $T_c/T_c^{max} = 1 - 82.6(p - 0.16)^2$, where $T_c^{max} = 95$ K.

In our experiment, an 80 MHz Ti:sapphire laser produces 45 fs pulses at $\approx 800$ nm (1.55 eV) as a source of both pump and probe pulses. The pump and probe pulses were cross-polarized, with a pump spot diameter of $\sim 60 \mu$m and probe spot diameter of $\sim 30 \mu$m. The reflected probe beam was focused onto an avalanche photodiode detector. The pump beam was modulated at 1 MHz with an acousto-optical modulator to minimize noise. The experiments were performed with an average pump power of 3 mW, giving a pump fluence of $\sim 0.1 \mu$J cm$^{-2}$ and a photoexcited QP density of $\sim 0.02$/unit cell, showing that the system is in the weak perturbation limit. The probe intensity was $\sim 10$ times lower. Resolution is at least 1 part in $10^6$. For all the dopings, probe reflection is specular from the mirror-like sample surfaces, and the same fraction of the incident laser power is absorbed by the sample, i.e. $\sim 90\%$. To avoid any competing relaxation processes from emergent low temperature states (e.g. superconducting, pseudogap, antiferromagnetic or stripe order), all data were taken at room temperature [23], and up to a pump–probe delay of 7 ps.

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Figure 1. Comparison of $\Delta R/R$ versus pump–probe time delay of OD ($T_c = 65$ K), OPT ($T_c = 95$ K) and UD ($T_c = 65$ K) Bi-2212 samples. Notice the difference in peak heights, and the smoothness of the time evolution from the peak into the tail of $\Delta R/R$.

2. Data and analysis

2.1. Three-temperature model

Figure 1 shows the time dependence of the photoinduced change in reflection ($\Delta R/R$) of a UD ($T_c = 65$ K), OPT ($T_c = 95$ K) and OD ($T_c = 65$ K) sample. For all samples, $\Delta R/R$ first shows a rapid rise (of the order of the pump pulse duration) followed by a subsequent decay. Notice two differences in the lineshapes of the samples: (i) the peak value of $\Delta R/R$ is smallest in the UD sample and (ii) the peak evolves into the tail most abruptly in the UD sample.

For time-resolved ARPES data of an OPT Bi-2212 sample, a three-temperature model (3TM) has been successfully used to fit the relaxation dynamics of photoexcited electrons [21]. In the 3TM, photoexcited electrons first transfer their energy to the (‘hot’) phonons that are more strongly coupled to them. These hot phonons then lose their energy to the cold phonons through anharmonic coupling. We thus first attempt to use the 3TM to fit our data, and show in the following that acceptable fits are not possible across the entire doping range.

In conventional superconductors, the relaxation signal occurring on a fast (<1 ps) timescale was assumed to be due to electronic relaxation alone [31]. This is reasonable because, though $\Delta R/R$ should be a linear combination of $\Delta T_e$ and $\Delta T_{lattice}$, $\Delta R$ arising from $\Delta T_{lattice}$ typically occurs on a very slow timescale ($\gg 10$ ps). At very early time delays, only the electronic subsystem (not the lattice and spin subsystems) responds to the pump pulse. Therefore, for the short time delays presented in our work, and since the same amount of incident laser power was absorbed by all the samples, we make the reasonable assumption that the measured reflectivity of the sample is proportional to the electronic temperature, i.e. $\Delta T_e / T_e = \eta \Delta R/R$, with $\eta$ being doping independent.

We cannot find any value of $\eta$ that gives rise to good fits across all dopings. Figures 2 and 3 show the fittings and the accompanying residual analysis. In fact, no choice of proportionality constant will produce an acceptable compromise across the series of samples between fitting the (a) peak electronic temperature and (b) tail out to 7 ps. This suggests that we need cooling.
Figure 2. Nearly optimal doping. The fit shown in the left figure underestimates the peak. At the tail the three temperatures become equal i.e. the electronic system is in equilibrium with the cooling phonon bath (blue dotted line: experimental data multiplied by proportionality constant; black line: fitted electronic temperature; red line: hot phonon temperature; green line: cold phonon temperature). The right figure shows the absolute of the residuals at each point of the curve. The residuals become smaller at later times.

Figure 3. OD sample. If we use the same ratio between reflectivity and temperature as in figure 2 for OD samples we find that the peak is not estimated properly. From the residuals (right) and from the actual fit (left) it can be seen that the tail is not properly captured as well.

channels beyond the one within the 3TM to obtain a good fit to the peak and the tail across all dopings.

2.2. Five-temperature model

To interpret the relaxation phenomena across the whole range of dopings, we need to go beyond the 3TM. We use a model where the relaxation happens in a two-step process (hot/cold modes) like in the 3TM, but with an additional bosonic mode. In this five-temperature model (5TM) there exists a second cooling channel for the hot electrons, that is, via coupling to hot SF.
Figure 4. Pictorial representation of the 5TM, depicting the energy transfers during the relaxation process. Hot electrons relax via two parallel pathways: the phononic and SF channels.

Similar to phonons, hot SF cool via scattering with cold SF. Figure 4 shows a schematic representation of the 5TM, with details in the appendix.

Figures 5(a)–(c) show the 5TM fits of three representative samples. The time evolution of the various subsystems (electron, hot/cold phonons/SFs) depends on the relative strengths of the coupling constants.

After the initial photoexcitation, the electrons heat up, rapidly transferring their energy to other subsystems. Due to the small number of hot phonons ($f_{ph} = 0.05$), equilibration with electrons ($T_{ph}^{hot} = T_e$) is reached within the first picosecond. The value of $\lambda_{e-ph}$ determines how fast the hot phonons respond to the electrons, and characterizes the sharpness of $T_{ph}^{hot}$-peak—from UD to OPT $\lambda_{e-ph}$ increases, resulting in a sharper hot phonon peak; in the OD samples the hot phonons react slower due to a smaller $\lambda_{e-ph}$. Hot SFs ($T_{sf}^{hot}$), on the other hand, equilibrate with electrons within the first 2 ps, due to their larger population ($f_{sf} = 0.50$) compared to hot phonons. The subsequent cooling of the electronic ($T_e$) peak is thus mainly determined by the coupling between electrons and hot SFs. With increasing doping, $\lambda_{e-sf}$ decreases, which is reflected in a less-sharp $T_{sf}^{hot}$-peak. Between 1.5 and 3 ps, cold SFs ($T_{sf}^{cold}$) equilibrate with electrons. Beyond 3 ps (hot and cold) SFs are not capable to decrease $T_e$ any further—further electronic cooling is due to cold phonons only. At times $>7$ ps all five temperatures eventually equilibrate and the tail flattens out. The small shoulder at $\sim 0.5$ ps is an extrinsic effect present in all UD samples, but does not affect the quality of the fits.

The change in peak shapes is well described by our model, and we obtained very good fits for the entire doping range. Figure 6 shows the doping dependence of the respective coupling constants. In the UD region, $\lambda_{e-ph}$ initially increases with doping between $p = 0.098$ and 0.11, then starts to decrease between 0.11 and 0.13 doping, before increasing again to a peak at OPT ($p = 0.16$), and thereafter decreasing with increasing doping in the OD region. On the other hand, $\lambda_{e-sf}$ decreases with increasing doping, with a stronger decrease in the UD than the OD regions.

The monotonic decrease of $\lambda_{e-sf}$ with doping is consistent with the weakening of SFs with increasing doping, resulting in a weaker coupling between electrons and SFs. The doping dependence of $\lambda_{e-sf}$ is similar to that of the magnetic spectral weight around $Q_{AF}$, as seen from the imaginary part of the energy-integrated spin susceptibility in $t$–$J$ model calculations [13]. Note the strong decrease of $\lambda_{e-sf}$ occurs up to $p = 0.13$; thereafter the decrease becomes
Figure 5. (a–c) Gray lines: individual data sets. Open circles = averaged data, multiplied by a proportionality factor. The proportionality factor $\eta = (5.0 \pm 0.1) \times 10^5 \text{K}$ is a fitting parameter constrained to be doping independent, since the same amount of incident laser power is absorbed by all the samples. Solid lines are time evolutions of the fitted temperatures of the various subsystems: electrons (black), hot phonons ($T_{\text{ph}}^{\text{hot}}$, red), cold phonons ($T_{\text{ph}}^{\text{cold}}$, green), hot SFs ($T_{\text{sf}}^{\text{hot}}$, blue) and cold SFs ($T_{\text{sf}}^{\text{cold}}$, magenta). All data, taken at 295 K, are shown here to 3 ps, but were taken and fitted up to 7 ps.

more gradual. This change in behavior at $p = 0.13$ could be related to the formation of stripe ordering [33] at $p = 1/8$. The non-zero value of $\lambda_{e-sf}$, even in our most OD sample ($p = 0.22$), suggests that SFs are present even in the OD regime, in agreement with inelastic neutron scattering studies [34].

For $\lambda_{e-ph}$, its initial increase between 0.098 and 0.11 doping is qualitatively consistent with Bardeen, Cooper and Schrieffer (BCS) theory, where an increased electron–phonon coupling gives rise to an increased $T_c$ and vice versa. Its subsequent decrease between 0.11 and 0.13 ($\sim 1/8$) doping, increase away beyond $p \sim 1/8$, may be due to the presence of stripe order at this doping level [35]. The concurrent decrease of $\lambda_{e-sf}$ in this same ‘stripe’ region implies a suppression in the superconductivity, consistent with a plateau in the superfluid density near this $1/8$ doping [36]. The doping dependence of $\lambda_{e-ph}$ in the OPT-OD region—maximum at OPT, and decrease with overdoping, is also in qualitative agreement with BCS theory, and strong lattice renormalization effects [2, 37].
According to our sensitivity analysis (refer to the appendix), there is another combination of \((f_{ph}, f_{sf})\), i.e. \((0.50, 0.05)\), that produces good fits. This combination would reverse the doping trends of \(\lambda_{e-ph}\) and \(\lambda_{e-sf}\). Since it is \(\lambda_{e-sf}\), and not \(\lambda_{e-ph}\), that should increase in the proximity of the antiferromagnetic region, we believe that the choice \((f_{ph}, f_{sf}) = (0.50, 0.05)\) may not be physically meaningful.

Apart from the best-fit results presented in figure 6, we also systematically investigated the fits over the parameter space. According to Dal Conte [24] \(\eta\) may be a doping dependent quantity. We investigate this scenario by treating \(\eta\) as fitting parameter, and carried out a similar parameter space study. We observe no fundamental change in the doping trends of \(\lambda_{e-ph}\) and \(\lambda_{e-sf}\) and conclude that the established doping trends are robust.

To estimate the fitting error of our parameters we repeat the same fitting process for a large number of experimental data sets. While the actual parameters are obtained by fitting the average of the experimental data, the respective error bars are estimated by fitting all individual data sets separately. We then obtain standard deviations of the distributions of the various fitting parameters to obtain the error bars.

Figures 5(a)–(c) show the original experimental data as gray shaded area behind the averaged curve used to obtain the actual fitting parameters. When we fit the wide range of noisy data we observe that only a narrow range of fitting parameters can give good fits. This suggests that our model parameters react sensitively to changes in the shape of the curve which is an indicator for a tightly constrained model.

3. Additional scenarios

Besides the scenario presented in the main text, we show here the doping dependences of \(\lambda_{e-ph}\) and \(\lambda_{e-sf}\) for three other combinations of hot phonon and hot SF energies (figure 7). The fraction...
Figure 7. Doping dependences of $\lambda_{e-ph}$ and $\lambda_{e-sf}$ corresponding to scenarios (a)–(c). Comparing these three figures with figure 6, we see that the doping dependences of $\lambda_{e-ph}$ and $\lambda_{e-sf}$ are preserved. Only their magnitudes have changed.

Pairs used for these scenarios were obtained from error surface studies as well.

(a) Hot phonon energy = 40 meV [7]. Hot SF energies taken from the energies of the magnetic resonance mode from neutron scattering data [12], and the positions of the peak in the bosonic spectrum from optical spectroscopy data [18, 38].

(b) Hot phonon energy = 70 meV [7]. Hot SF energies taken from the energies of the magnetic resonance mode from neutron scattering data [12], and the positions of the peak in the bosonic spectrum from optical spectroscopy data [18, 38].

(c) Hot phonon energy = 70 meV [7]. Hot SF energies taken from the centroid of the broad background of the bosonic spectrum in optical spectroscopy data [18, 38].

As shown in figure 7, the doping dependence of $\lambda_{e-ph}$ and $\lambda_{e-sf}$ is quite robust against whichever scenario as listed above is used.

4. Concluding remarks

Independent confirmation that electrons couple to multiple bosonic modes comes from frequency-resolved and time-resolved pump–probe data of OPT Bi-2212 [24]. The resulting
spectral function necessitates the electrons to be coupled directly to hot phonons, cold lattice as well as a bosonic mode of electronic origin, which the authors suggested could be SFs or current loops. As mentioned earlier, the fractions used there for the various subsystems are consistent with what we obtained here. Moreover, ARPES data on heavily UD La$_{2-x}$Sr$_x$CuO$_4$ showed fine structure in the electron self-energy, demonstrating the involvement of multiple boson modes in the coupling with electrons [39]. The contribution of our work, besides working on a different class of cuprate superconductors, is that we have obtained the doping dependence of the different electron–boson coupling constants from the UD to the OD regimes.

Our work shows that the 3TM cannot explain the data over the entire doping range. Moreover, our 5TM requires both the electron–phonon and electron–SF interactions to be responsible for the initial fast (~ 100 fs) relaxation. Important confirmation of this comes from the temperature-dependent pump–probe data of our most OD sample ($T_c \sim 65$ K) [40]. Besides observing the opening of a pseudogap at $T^* \sim 100$ K, the fast relaxation time $\tau_{\text{fast}}$ increases with decreasing temperature, before peaking at $T^*$ and decreasing to ~ 100 fs at 30 K. This change in behavior of $\tau_{\text{fast}}$ at $T^*$ is intriguing—it suggests that the electron–SF coupling, in addition to electron–phonon coupling, is involved in the initial fast relaxation of the hot electrons. The peak at $T^*$, and its subsequent decrease below $T^*$, is then due to an increased scattering rate between electrons and SFs as the sample enters the pseudogap phase. This scenario is further confirmed by the temperature-dependence of $\tau_{\text{fast}}$ above $T^*$—a fit to $1/T^n$ yields $n = 1.3$, which disagrees with the behavior predicted for the electron–phonon relaxation time for good ($n = 2$) and poor ($n = 3$) metals [41].

In conclusion, we have taken pump–probe data of a wide doping range of Bi-2212 samples. Our analysis of the relaxation dynamics across the entire doping regime requires the existence of two types of bosonic modes strongly coupled to electrons. The different doping dependences of the electron–boson coupling strengths enables us to identify them as phonons and SFs: electron–phonon coupling ($\lambda_{\text{e-ph}}$) peaks at optimal doping, and electron–SF coupling ($\lambda_{\text{e-sf}}$) decreases monotonically with doping. This observation should give new insight into the mechanism of high-$T_c$ superconductivity in cuprates.

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Appendix. Five-temperature model

The time evolution of the temperatures of the various subsystems: electronic ($T_e$), hot/cold phonon ($T_{ph}^{hot}$/$T_{ph}^{hot}$) and hot/cold SFs ($T_{sf}^{hot}$/$T_{sf}^{hot}$) satisfies the rate equations:

$$\frac{dT_e}{d\tau} = \frac{P}{C_e} - C_1 \left( n_{e}^{ph} - n_{ph}^{hot} \right) \frac{T_e}{T_e} - \frac{3\lambda_{e-sf}(\Omega_{sf}^{hot})^3}{\hbar \pi k_B^2} \left( n_{e}^{sf} - n_{sf}^{hot} \right) \frac{T_e}{T_e},$$

(A.1)
\[ \frac{dT_{ph}^{\text{hot}}}{d\tau} = \frac{C_e}{C_{ph}} C_1 \frac{n_e^{ph} - n_{ph}^{hot}}{T_e} - \frac{T_{ph}^{hot} - T_{ph}^{cold}}{\tau_{ph}}, \]  
(A.2)

\[ \frac{dT_{sf}^{\text{hot}}}{d\tau} = \frac{C_e}{C_{sf}} \frac{3\lambda_{e-sf}\left(\Omega_{sf}^{hot}\right)^3 n_{sf}^{sf} - n_{ph}^{hot}}{\hbar \pi k_B^2} - \frac{T_{sf}^{hot} - T_{sf}^{cold}}{\tau_{sf}}, \]  
(A.3)

\[ \frac{dT_{ph}^{\text{cold}}}{d\tau} = \frac{C_{ph}^{cold}}{\tau_{ph}}, \]  
(A.4)

\[ \frac{dT_{sf}^{\text{cold}}}{d\tau} = \frac{C_{sf}^{cold}}{\tau_{sf}}. \]  
(A.5)

The system is excited by a Gaussian pulse \( P \) with full-width-half-maximum 45 fs and energy density of 0.166 J cm\(^{-3}\). The distribution functions are

\[ n_{ph}^{\text{hot(cold)}} = 1/[e^{\Omega_{ph}^{\text{hot(cold)}}/k_B T_e} - 1], \]  
(A.6)

\[ n_{sf}^{\text{hot(cold)}} = 1/[e^{\Omega_{sf}^{\text{hot(cold)}}/k_B T_e} - 1]. \]  
(A.7)

In (A.1), \( n_{ph}^{\text{hot}} = [e^{\omega_{ph}(T_e)}/k_B T_e} - 1]^{-1} \) are not distribution functions, but are results of performing delta-function energy integrals [42].

The energy of the hot phonon mode \( \Omega_{ph}^{\text{hot}} = 40 \text{ meV} \) corresponds to the out-of-plane out-of-phase oxygen buckling \( B_{1g} \) phonon [7]. Though cuprate samples like Bi-2212 are inhomogeneous both in energy gap and characteristic boson frequency, the spatial average of mode frequency is doping independent [7]. Therefore, we assume \( \Omega_{ph}^{\text{hot}} \) is constant throughout the entire doping regime. Neutron scattering data [43] reveal an acoustic phonon mode at 20 meV. Since recent STM data [7] did not observe any coupling between electrons and this particular acoustic mode, we choose it to be the energy of our cold phonon bath \( \Omega_{ph}^{\text{cold}} \). The hot \( \Omega_{sf}^{\text{hot}} \) and cold \( \Omega_{sf}^{\text{cold}} \) SF energies are taken from optical spectroscopy [38] and are doping dependent: both \( \Omega_{sf}^{\text{hot}} \) and \( \Omega_{sf}^{\text{cold}} \) scale with the centroid position of the broad bosonic background, with the constraint that \( \Omega_{sf}^{\text{hot}} = 41 \text{ meV} \) at OPT.

In Kabanov’s paper [41], the spectral function \( \alpha^2 F(\omega) \propto \omega^2 \) in good metals, while in poor metals \( \alpha^2 F(\omega) \propto \omega \). In the time evolution of the electronic temperature \( T_e \), this effect gives rise to the prefactor \( \left( \frac{3\lambda_{e-ph}(\Omega_{ph}^{hot})^3}{\hbar \pi k_B^2} \right) \) in good metals, and \( \left( \frac{\pi^3 \lambda_{e-ph} k_B T_e^3}{2\hbar} \right) \) in bad metals. This moderation of the electron–phonon interaction, by the change in metallicity with doping, is accounted for in (A.1) and (A.2), by assuming a ‘two-fluid model’, where the parameter \( C_1 \) is the linear combination of these two prefactors [41]:

\[ C_1 = (1 - \alpha) \left( \frac{\pi^3 \lambda_{e-ph} k_B T_e^3}{2\hbar} \right) + \alpha \left( \frac{3\lambda_{e-ph}(\Omega_{ph}^{hot})^3}{\hbar \pi k_B^2} \right), \]  
(A.8)

with the ‘good-metal’ fraction \( \alpha \) given by

\[ \alpha(p) = a + bp. \]  
(A.9)
Figure A.1. Resistivity (at 300 K) versus doping, obtained from [18, 38]. The trend is roughly linear, which justifies the use of relation (A.9).

This linearity of $\alpha$ with doping is justified by the approximate linear dependence, with doping, of the room-temperature resistivity of Bi-2212 [18, 38] (see figure A.1). This good–bad-metal picture, though phenomenological, is consistent with our picture of Bi-2212 becoming more metallic with increasing doping. In the UD region, the hole carriers are so few that the transport is difficult. At high temperatures, the hole carriers are scattered off the randomly distributed spin moments so strongly that the resistivity is much larger, i.e. bad metal. In the OD region, the effective bandwidth is increased and the SFs are suppressed significantly. As such, the resistivity is decreased, i.e. good metal.

The parameters $a$ and $b$ are determined by enforcing that

$$\alpha(0.05) = 0 \quad \text{and} \quad \alpha(0.27) = 1$$

implying that the material is a bad metal at $p = 0.05$ and a good metal at $p = 0.27$. These two end-points correspond to the dopings on the superconducting dome when the superconducting transition temperature $T_c = 0$. Figure 1 of [44] shows that, even at 300 K, the temperature where we took our data, these two end-points are still reasonable.

The specific heat of electrons, hot/cold phonons and hot/cold SFs are, respectively,

$$C_e = \gamma T_e,$$

$$C_{\text{ph}}^\text{hot} = N_{\text{ph}} f_{\text{ph}} \Omega_{\text{ph}}^\text{hot} \frac{\partial n_{\text{ph}}^{\text{hot}}}{\partial T_{\text{ph}}^{\text{hot}}},$$

$$C_{\text{ph}}^\text{cold} = N_{\text{ph}} (1 - f_{\text{ph}}) \Omega_{\text{ph}}^\text{cold} \frac{\partial n_{\text{ph}}^{\text{cold}}}{\partial T_{\text{ph}}^{\text{cold}}},$$

$$C_{\text{sf}}^\text{hot} = N_{\text{sf}} f_{\text{sf}} \Omega_{\text{sf}}^\text{hot} \frac{\partial n_{\text{sf}}^{\text{hot}}}{\partial T_{\text{sf}}^{\text{hot}}},$$

$$C_{\text{sf}}^\text{cold} = N_{\text{sf}} (1 - f_{\text{sf}}) \Omega_{\text{sf}}^\text{cold} \frac{\partial n_{\text{sf}}^{\text{cold}}}{\partial T_{\text{sf}}^{\text{cold}}}.$$
Here the Sommerfeld coefficient $\gamma = 8 \text{ mJ mol}^{-1} \text{K}^{-2}$ is taken from Junod [45]. There is no doping dependence of $\gamma$ for Bi-2212 at room temperature, as seen from specific heat data of Loram et al [46]. The parameter $f_{ph}$ ($f_{sf}$) denotes the fraction of total phonon (SF) modes that are more strongly coupled to the electrons, and $N_{ph}$ ($N_{sf}$) denotes the number of phonon (SF) modes in the irradiated volume.

In unrestricted fits with six fitting variables ($\lambda_{e-ph}, \lambda_{e-sf}, \tau_{e-sf}, \tau_{e-ph}, f_{sf}$ and $f_{ph}$) we found that the parameters $f_{sf}$ and $f_{ph}$ do not show strong doping dependences and may be replaced by constants. Therefore we perform a parameter space study to obtain the combination of values of $f_{ph}$ and $f_{sf}$ that gives the smallest (least-squared) error between the data and fits for all samples. To do that we calculate the fitting error of the 5TM with four fitting variables ($\lambda_{e-sf}, \lambda_{e-ph}, \tau_{e-sf}$ and $\tau_{e-ph}$) at all possible fraction combinations ($f_{sf}, f_{ph} = 0.00 - 0.95$ at 0.05 intervals). This gives us one error surface for each of the differently doped samples. We then average over all of them to obtain the total error surface shown in figure A.2. The fact that the minima (violet) are located along the axes indicates that the two bosonic channels play essentially different roles. If a single bosonic cooling channel, like in the 3TM, were able to fit our data across all dopings, then the regions of minimum error (violet) would run along the diagonal (where $f_{ph} = f_{sf}$), which we do not see here. Note the existence of two symmetrical optimal regions: $(f_{ph}, f_{sf}) = (0.05, 0.50)$ and $(0.50, 0.05)$. We choose the former because it is consistent with the analysis from the four-temperature model (4TM) used to analyze frequency-resolved and time-resolved pump–probe data of OPT Bi-2212 [24], where $(f_{ph}, f_{sf}) = (0.1, 1.0)$—in their model $f_{sf}$ is necessarily 1.0 because there are no cold SFs. Our model also takes into account the doping dependence of the penetration depth of the 800 nm pulse [38]. We assume that the fraction of incident laser power absorbed by the sample is doping independent.

By minimizing the least square error between the experimental data and the relaxation curve obtained from the 5TM, we determine the doping dependence of the coupling constants $\lambda_{e-ph}$ and $\lambda_{e-sf}$.

**Figure A.2.** The normalized least-squared error surface plotted as a function of $f_{ph}$ and $f_{sf}$. Violet regions denote fraction combinations that yield very good fits of 5TM to data. Dark red regions indicate large fitting errors.
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