Role of bosonic modes in the mechanism of high temperature Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ superconductors using ultrafast optical techniques

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Using ultrafast optical techniques, we probe the hole-doping dependence of the electron-boson coupling constant $\lambda$ in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. In the overdoped region, we observe a correlation between $\lambda$ and the superconducting transition temperature $T_c$. Upon performing the McMillan analysis, however, we find that $\lambda$ is too small to explain the high $T_c$'s, and that the Coulomb pseudopotential $\mu^*$ is negative. Our analysis therefore reveals two components in the mechanism of high-$T_c$ superconductivity — a dominant pre-existing pairing interaction, together with a weaker electron-phonon interaction that fine-tunes $T_c$.

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Despite many advances in understanding copper-oxide high-temperature superconductors, there still exists no universally accepted mechanism. 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 [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 transition temperature ($T_c$) 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$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) [8] while a bosonic mode of electronic origin was found in the electron-doped Pr$_{0.88}$LaCe$_{0.12}$CuO$_4$ [9]. Together with salient features observed in angle-resolved photoemission spectroscopy (ARPES) [2, 10, 11], these new results raise the fundamental question of whether the bosonic modes are a pairing glue [12] or a signature of an inelastic tunneling channel [13]. Here, we report a systematic time-resolved pump-probe study on Bi-2212 at various doping levels. It reveals a positive correlation between the quasiparticle relaxation rate and $T_c$ as doping is varied from the optimal toward the overdoped regime, indicating that phonons play a role in the mechanism of high-$T_c$ superconductivity. Our analysis, based on McMillan-type strong coupling theory, shows that (i) the electron-phonon coupling is not sufficiently large to account for the large $T_c$'s of the cuprates, and (ii) the Coulomb pseudopotential is necessarily negative, indicating that a dominant pre-existing pairing interaction is necessary to glue the electrons into pairs with such high $T_c$'s. Candidates of such a pre-existing pairing interaction are electronic coupling to a bosonic mode of electronic origin, or a mechanism without mediators.

The role of the electron-boson interaction (EBI) in high-$T_c$ superconductors were studied by different techniques. For example, inelastic neutron scattering tracks the changes in boson energies or dispersions upon entering the superconducting state. ARPES and time-integrated optical spectroscopy [14, 15] measure the effects of EBI on electronic self-energies, and planar junction experiments determine the energy of the bosonic mode [16]. STM experiments measure the local density of states through the local differential tunneling conductance, where the characteristic boson mode energy is estimated from the peak position in $d^2I/dV^2$ [8]. However, it cannot tell us directly the strength of the electron-boson coupling because all energy is encoded in the electron self-energy itself. Complementary to the above techniques, ultrafast spectroscopy – a temporally-resolved technique, has been used in probing the relaxation dynamics of photoexcited quasiparticles in correlated electron systems [17, 18]. Its unique contribution lies in its ability to extract the value of the electron-boson coupling constant ($\lambda$) directly, without the need to perform complicated inversion algorithms. This procedure has been experimentally verified on the conventional superconductors [19]. Performing time-resolved pump-probe measurements on the same family of cuprates will allow us to determine (i) the magnitude of $\lambda$, and (ii) whether $\lambda$ has any correlation with doping. These will yield crucial information to the role of electron-boson interaction.
in the mechanism of high-$T_c$ superconductivity.

The family of the two-layer cuprate Bi-2212 has been in recent years the most intensively studied class of high-$T_c$ superconductors, 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 $T_c$'s (compared to other cuprates). Single crystals of Bi-2212 were obtained from two groups (Tokyo and AIST) grown by the floating zone method with doping controlled by oxygen depletion, yielding values of $T_c$ (determined by magnetization data) that depends on the hole doping level spanning from the underdoped to the overdoped regime. Due to difficulties in growing high-purity underdoped samples, we present data on only one underdoped sample. OD = overdoped samples. Red lines = best-fit straight line through the OD data points.

In our experiments an 80-MHz repetition rate Ti:sapphire laser produces 45-femtosecond (fs) pulses at approximately 800 nm (1.5 eV) as the source of both pump and probe optical 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 acoustic-optical modulator to minimize noise. The experiments were performed with an average pump power of 3 mW, giving a pump fluence of $\sim$0.1 J/cm$^2$ and a photoexcited quasiparticle density of 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 $10^6$. The fitted values of $\tau$ have a typical error of $\pm$1 %. All measurements are done at room temperature. At this temperature, the electron subsystem reaches a local equilibrium within 20 fs, shorter than our pulse width. Hence the observed quasiparticle relaxation occurs mostly through electron-lattice coupling. Figure 1 shows the time dependence of the photoinduced signal of a typical overdoped Bi-2212 sample. The time evolution of the photoinduced reflection $\Delta R/R$ first shows a rapid rise time (of the order of the pump pulse duration) followed by a subsequent decay. As shown in Fig. 1 the data can be fit better by two exponentials (red line) than a single exponential (blue line). It indicates the quasiparticle relaxation has two components: $\Delta R/R = A + B \exp(-t/\tau_{fast}) + C \exp(-t/\tau_{slow})$. The fast component $\tau_{fast}$ is of the order of 100 fs while the slow component $\tau_{slow}$ is of the order of 650 fs in the optimally-doped to overdoped regimes. Since observation of spin-fluctuation modes in the overdoped Bi-2212 samples has rarely been reported, we ascribe the quasiparticle relaxation in this regime to electron-lattice coupling, not coupling between electrons and spin fluctuations — this is consistent with STM data on the same family of cuprates. Recently a time-resolved photoelectron spectroscopy measurement has been carried out on an optimally doped Bi-2212 sample, where a similar two-stage cooling dynamics was observed at room temperature. Similarly, we interpret the relaxation process as the electrons first transferring energy to the phonons which are more strongly coupled at a characteristic time $\tau_{fast}$ and then continue cooling down via the energy dissipation of these hot phonons by the means of anharmonic decay at a characteristic time $\tau_{slow}$.

FIG. 2: (Color) Doping dependence of the (a) fast relaxation rate ($1/\tau_{fast}$) and (b) slow relaxation rate ($1/\tau_{slow}$). UD = underdoped sample. OD = overdoped samples. Red lines = best-fit straight line through the OD data points.
antiferromagnetic insulator. The existence of a correlation between $1/\tau$ and $T_c$ suggests that phonons play a role in the mechanism of high-$T_c$ superconductivity in the cuprates.

Next, we perform a quantitative analysis by assuming that the quasiparticle relaxation throughout the entire doping regime is due to the electron-phonon interaction. Since, as already mentioned, the transfer of electron energy first occurs through selected modes that are most strongly coupled to electrons, we use $\tau_{\text{fast}}$ to estimate the electron-phonon coupling strength $\lambda$. These strongly coupled phonon modes should be the most relevant in discussing the possible phonon-mediated superconductivity. We consider the out-of-plane out-of-phase oxygen buckling $B_{1g}$ phonon and the half-breathing in-plane copper-oxygen bond stretching phonon with energies of approximately $\Omega = 40$ and 70 meV, respectively. These two types of phonon modes are suggested to be responsible for the dispersion anomalies at the antinodal [10] and nodal [2] directions, and reveal strong line-shape renormalizations with doping and temperature in Raman and neutron scattering measurements [1, 21, 22, 23, 24]. 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. Therefore, we assume $\Omega$ is constant throughout the entire doping regime. $\tau$ is related to $\lambda$ by the Allen relation [25]

$$\frac{1}{\tau} = \frac{3\hbar \langle \omega^2 \rangle}{\pi k_B T_c},$$

where $T_c$ is the electronic temperature (estimated to be 340 K), $\lambda$ ($\omega^2$) is the second moment of the effective electron-phonon coupling strength, $\alpha^2 F(\omega)$. We use the Einstein model for phonons such that $\langle \omega^2 \rangle = \Omega^2$ and $\langle \omega \rangle = \Omega$. Figure 3(a) shows the calculated values of $\lambda$ from Eq. (1), as a function of $T_c$. Since $1/\tau \propto \lambda$, $\lambda$ also correlates with $T_c$. Within the strong-coupling theory, the McMillan [26] formula for the superconducting transition temperature in a $d$-wave superconductor is found to be [27]:

$$T_c = \omega_0 \exp \left[ -\frac{2(1 + \lambda)}{\lambda - \mu^*(1 + \lambda \langle \omega \rangle)/2\omega_0} \right].$$

Here $\mu^*$ is the Coulomb pseudopotential; it is a renormalized quantity and can be very different from the original bare Coulomb repulsion. In this formula, the factor 2 accounts for the $d$-wave nature of superconducting order parameter while the renormalization factor $(1 + \lambda)$ arises from the $s$-wave channel of the electron-phonon coupling. Since the average bosonic mode frequency $\omega_0$ remains unchanged with doping [3], the variation of $T_c$ across the superconducting dome in Bi-2212 is due solely to the interplay between $\lambda$ and $\mu^*$. With the given coupling strength $\lambda$ and $\omega_0 \approx \Omega$, and the experimentally measured $T_c$, one imposes a stringent constraint on $\mu^*$ through Eq. (2). Figure 3(b) shows the calculated values of $\mu^*$ for both phonon modes. The negativity of $\mu^*$ presents two important implications: (i) the electron-phonon interaction alone is not strong enough to cause Bi-2212 to be superconducting at the measured high $T_c$, and (ii) a pre-existing (attractive) pairing interaction is necessary, especially when no bosonic modes of electronic origin (such as spin resonance modes) exist, which is true at least in the deeply overdoped region where they are neither expected nor observed. Figure 3(b) shows a clear positive correlation between $\mu^*$ and $T_c$, with the magnitude of $\mu^*$ reaching a maximum at the optimal doping, which shows that a large pre-existing pairing interaction is needed to produce a large $T_c$. Our data on an almost optimally-doped three-layered cuprate $\mathrm{Th}_2\mathrm{Ba}_2\mathrm{Ca}_2\mathrm{Cu}_3\mathrm{O}_{y}$, with a higher $T_c$ of 115 K (not shown here), revealed even smaller values of $1/\tau_{\text{fast}}$ and $1/\tau_{\text{slow}}$ than the corresponding values of all our Bi-2212 samples, supporting our assertion that electron-phonon coupling alone cannot be the pairing mechanism for high-$T_c$ superconductivity.

In Figure 4 we plot $\lambda$ versus $T_c/\omega_0$, combining the data for Bi-2212 with some conventional ($s$-wave) superconductors, also obtained from pump-probe measurements [19], as well as the recently discovered modeless noncentrosymmetric superconductor $\mathrm{Mg}_{12-x}\delta\mathrm{Ir}_x\mathrm{Bi}_{16}$ [28]. Compared to these conventional superconductors, we notice that data points for Bi-2212 (i) do not follow the same trend, (ii) show a much weaker correlation between $\lambda$ and $T_c/\omega_0$, with a much smaller slope, (iii) have smaller values of $\lambda$, and (iv) have larger values of $T_c/\omega_0$. If the McMillan formula is valid for Bi-2212, then, for a positive $\mu^*$, a large $T_c/\omega_0$ should imply a large $\lambda$, i.e. the Bi-2212 data points should lie on the same trend as the conventional superconductors. However, the values of $\lambda$, directly obtained from $1/\tau$, are too small. For Eq. (2) to still hold, we therefore need a negative $\mu^*$, i.e. we return...
to the same conclusion of the need for a pre-existing pairing interaction.

The central conclusion of this work, is that both a pre-existing pairing interaction and the electron-phonon interaction play a role in the mechanism of high-$T_c$ superconductivity. The pre-existing pairing interaction plays a dominant role, while the electron-phonon coupling, in cooperation with the pre-existing interaction, merely fine-tunes the transition temperature as evidenced by its weak positive correlation with $T_c$. Our finding therefore echoes the hypothesis by Anderson [5] that one cannot neglect the ultimate importance of strong correlation effects to explain the mechanism for high-$T_c$ superconductivity in the cuprates, where the Coulomb repulsion is comparable or even larger than the relevant $d$-orbital bandwidth. It also suggests that, though we can use a convenient effective theory of electronic coupling to bosonic modes to understand many interesting signatures observed in ARPES and tunneling experiments on cuprates, such a theory does not reveal the underpinning mechanism for superconductivity. Theoretically, the spin-fluctuation-mediated pairing mechanism is of electronic origin and should come from the same strong electronic correlation. Recent dynamical cluster calculations [29] have indeed shown that the pairing mechanism in the doped two-dimensional Hubbard model is mediated by the exchange of $S=1$ particle-hole spin fluctuations. So far, whether the extracted coupling between quasiparticles and spin fluctuations is a dominant mechanism or a secondary effect remains hotly debated in a strong electronic correlation model for superconductivity. Experimentally, if there exists a dominant electron-boson (specifically spin fluctuation mode) coupling, it will be very interesting to characterize directly the strength of this coupling of electronic origin.

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1 R. J. McQueeney et al., Phys. Rev. Lett. 82, 628 (1999).
2 A. Lanzara et al., Nature 412, 510 (2001).
3 J. Rossat-Mignod et al., Physica C 185-189, 86 (1991).
4 M. R. Norman et al., Phys. Rev. Lett. 79, 3506 (1997).
5 P. W. Anderson, Science 316, 1705 (2007).
6 W. L. McMillan and J. M. Rowell, in Superconductivity (Dekker, New York, 1969), Vol. 1, p. 469.
7 J. Kirtley, in Handbook of High-Temperature Superconductivity: Theory and Experiment (Springer, New York, 2007).
8 J. Lee et al., Nature 442, 546 (2006).
9 F. C. Niestemska et al., Nature 450, 1058 (2007).
10 T. Cuk et al., Phys. Rev. Lett. 93, 117003 (2004).
11 G.-H. Gweon et al., Nature 430, 187 (2004).
12 A. V. Balatsky and J.-X. Zhu, Phys. Rev. B 74, 094517 (2006).
13 S. Pilgram, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. 97, 117003 (2006).
14 J. P. Carbotte, E. Schachinger, and D. N. Basov, Nature 401, 254 (1999).
15 J. Hwang, T. Timusk, and G. D. Gu, Nature 427, 714 (2004).
16 J. F. Zasadzinski et al., Phys. Rev. Lett. 87, 067005 (2001).
17 R. D. Averitt and A. J. Taylor, J. Phys.: Condens. Matter 14, R1357 (2002).
18 J. Demsar, J. L. Sarrao, and A. J. Taylor, J. Phys.: Condens. Matter 18, R281 (2006).
19 S. D. Brorson et al., Phys. Rev. Lett. 64, 2172 (1990).
20 L. Perfetti et al., Phys. Rev. Lett. 99, 197001 (2007).
21 N. Pyka et al., Phys. Rev. Lett. 70, 1457 (1993).
22 D. Reznik, B. Keimer, F. Dogan, and I. A. Aksay, Phys. Rev. Lett. 75, 2396 (1995).
23 R. J. McQueeney et al., Phys. Rev. Lett. 87, 077001 (2001).
24 S. Sugai et al., Phys. Rev. B 68, 184504 (2003).
25 P. B. Allen, Phys. Rev. Lett. 59, 1460 (1987).
26 W. L. McMillan, Phys. Rev. 167, 331 (1968).
27 J.-X. Zhu, Unpublished.
28 G. Mu, Y. Wang, L. Shan, and H. H. Wen, Phys. Rev. B 76, 064527 (2007).
29 T. A. Maier, M. S. Jarrell, and D. J. Scalapino, Phys. Rev. Lett. 96, 047005 (2006).