SLAC-PUB-13952

A step closer to visualizing the electron–phonon interplay in real time

Y. L. Chen, W. S. Lee, and Z. X. Shen[1]

Departments of Applied Physics and Physics, and SLAC Photon Science, Stanford University, Stanford, CA 94305

The origin of the very high superconducting transition temperature (Tc) in ceramic copper oxide superconductors is one of the greatest mysteries in modern physics. In the superconducting state, electrons form pairs (known as Cooper pairs) and condense into the superfluid state to conduct electric current with zero resistance. For conventional superconductors, it is well established that the 2 electrons in a Cooper pair are "bonded" by lattice vibrations (phonons) (1), whereas in high-Tc superconductors, the "glue" for the Cooper pairs is still under intense discussion. Although the high transition temperature and the unconventional pairing symmetry (d-wave symmetry) have led many researchers to believe that the pairing mechanism results from electron–electron interaction, increasing evidence shows that electron–phonon coupling also significantly influences the low-energy electronic structures (2, 3) and hence may also play an important role in high-Tc superconductivity. In a recent issue of PNAS, Carbone *et al*. (4) use ultrafast electron diffraction, a recently developed experimental technique (5), to attack this problem from a new angle, the dynamics of the electronic relaxation process involving phonons. Their results provide fresh evidence for the strong interplay between electronic and atomic degrees of freedom in high-Tc superconductivity.

In general, ultrafast spectroscopy makes use of the pump-probe method to study the dynamic process in material (see Fig. 1*A1*). In such experiments, one first shoots an ultrafast (typically 10–100 fs) "pumping" pulse at the sample to drive its electronic system out of the equilibrium state. Then after a brief time delay (Δt) of typically tens of femtoseconds to tens of picoseconds, a "probing" pulse of either photons or electrons is sent in to probe the sample's transient state. By varying Δt, one can study the process by which the system relaxes back to the equilibrium state, thus acquiring the related dynamic information. This pump-probe experiment is reminiscent of the standard method been used by bell makers for hundreds of years to judge the quality of their products (hitting a bell then listening to how the sound would fade away),

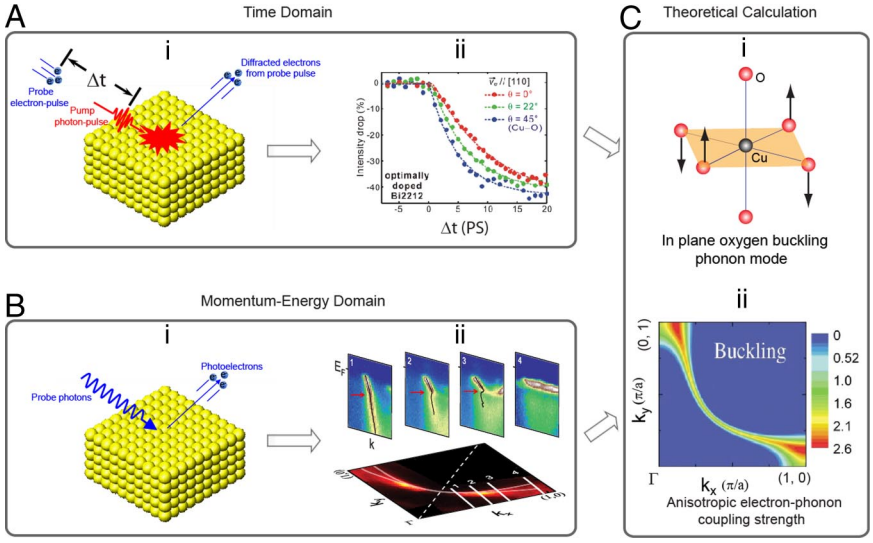

Fig. 1. Relationship of the work by Carbone *et al*. (4) and previous experimental and theoretical research in electron–phonon coupling in high-Tc cuprates. (*A1*) Schematic illustration of the photon-pump, electron-probe experimental method. (*A2*) Results from Carbone *et al*. (4) show that the droping of the electron diffraction spot intensity depends on the relative orientation between the pump photon polarization and the Cu—O bond direction in Bi2212, suggesting an anisotropy electron–phonon coupling within the CuO_2 plane. (*B1*) Schematic illustration of regular photoemission spectroscopy, in which only continuous probing photons are used for generating photoelectrons. (*B2*) Anisotropic electron–phonon coupling seen by ARPES, indicated by different strengths of the band dispersion "kink" at different k-space loci. (*Upper*) Four band dispersions measured at positions 1–4. (*Lower*) The Fermi surface of Bi2212. (*C1*) Simple illustration of the buckling mode motions of oxygen atoms in the CuO_2 plane. (*C2*) Calculated anisotropic electron–phonon coupling strength for in-plane buckling phonon of Bi2212 in k-space. [Reproduced with permission from ref. 10 (Copyright 2004, American Physical Society).] Warmer colors indicate larger coupling strength.

albeit the relevant time scale here is way beyond tens of femtoseconds. Traditionally, ultrafast spectroscopy was carried out to study gas-phase reactions (6), but it has also been applied to study condensed phase systems since the development of reliable solid-state ultrafast lasers approximately a decade ago. In addition, the ability to control pulse width, wavelength, and amplification of the output of Ti:Sapphire lasers has further increased the capability of this experimental method. During the past decade, many ultrafast pump-probe experiments have been carried out in various fields by using different probing methods, such as photo-resistivity (7), fluorescence yield (8), and photoemission (9), and they have revealed much new information complementary to the equilibrium spectroscopy methods used before.

Carbone *et al*. (4) used the photon-pump, electron (diffraction)-probe method. The pumping photon pulse first drives the electrons in the sample into an oscillating mode along its polarization direction. Then during the delay time, these excited electrons can transfer excess energy to the adjacent nuclei and cause crystal lattice vibration (i.e., excitation of phonons) on their way back to the equilibrium state. An ultrashort electron pulse is shot at the sample at various time delays Δt and the diffraction pattern is collected. Because the electron diffraction pattern is directly related to the crystal lattice structure and its motion, this technique provides a natural way to study the electron–

Author contributions: Y.L.C, W.S.L, and Z.X.S wrote the paper.

The authors declare no conflict of interest.

[1]To whom correspondence should be addressed. E-mail: zxshen@stanford.edu.

Work supported in part by US Department of Energy contract DE-AC02-76SF00515.

phonon coupling problem. Furthermore, by adjusting the pump pulse's relative polarization with respect to the Cu—O bond direction, Carbone *et al*. were able to acquire the electron–phonon coupling strength along different directions.

Focusing on the lattice dynamic along the *c* axis, Carbone *et al*. (4) found that the *c*-axis phonons in the optimally-doped $Bi_2Sr_2CaCu_2O_8$ (Bi2212) are coupled to the electrons with different strength along different directions within the CuO_2 plane. The coupling strength reaches its largest value along the 2 Cu—O bond directions and becomes the weakest along the bisector of the angle formed by the 2 Cu—O bonds. As pointed out by Carbone *et al*., these observations agree well with the calculated coupling strength between electrons and the buckling phonons (see Fig. 1 *C1* and *C2*) (10). Furthermore, their observation of this anisotropic electron–phonon coupling also agrees with results from angle-resolved photoemission (ARPES) (2), which measures the equilibrium-state properties of materials. In ARPES measurements, electron–phonon coupling manifests itself as a kink anomaly in the band dispersion and a corresponding sudden broadening in the spectral width. As shown in Fig. 1*B2*), ARPES measurements on Bi2212 indicate that the dispersion kink become more pronounced near the Brilloiun zone boundary (4 in Fig. 1*B2*, along Cu—O bond directions) compared with that near the zone center (1 in Fig. 1*B2*, bisector direction of Cu—O bonds), thus giving the same anisotropic electron–phonon coupling as in ref. 4. This consistency between the measurements in time domain (pump-probe) and energy domain (ARPES) is rather remarkable and suggests that experiments done in the equilibrium (e.g., ARPES) and the nonequilibrium states (e.g., ultrafast pump-probe experiment) can be used to study the same physics and provide complementary information.

In high-Tc superconductors, the "glue" for the Cooper pairs is still under intense discussion.

Unlike the nice agreement in Bi2212, there are, however, discrepancies between Carbone *et al*.'s (4) results and ARPES measurements on $Bi_2Sr_2Ca_2Cu_3O_{10}$ (Bi2223). According to the report by Carbone *et al*., the observed electron–phonon coupling is isotropic in this material, whereas ARPES observation (11) shows a momentum-dependent kink structure in the band dispersion similar to that in Fig. 1*B2*, indicating that similar anisotropic electron–phonon coupling also exists in Bi2223. A possible explanation could be the difference in the sensitivity to material complexity between the two techniques. Unlike Bi2212 where the buckling phonon is active for both CuO_2 planes, Bi2223 has three CuO_2 planes with the buckling phonon inactive for the inner CuO_2 plane. Nevertheless, this discrepancy suggests that there is still much that needs to be learned before we can claim a full understanding of the ultrafast pump-probe measurements that are carried out in the nonequilibrium state, in particular how the outcome should be compared with the equilibrium-state measurements. It would also be very informative to extend Carbone *et al*.'s measurements to more compounds, especially single-layer cuprates and compare the results with existing ARPES measurements. In any case, it is exciting to see that it is now possible to form a complementary physical picture between both equilibrium-state and nonequilibrium-state measurements on complex materials, such as high-Tc cuprates. The work by Carbone *et al*. is another exciting development demonstrating high-Tc research as a driver for technique development and correlation among different measurement modalities.

ACKNOWLEDGMENTS. Our work is supported by the Department of Energy, Office of Basic Energy Science, Division of Materials Science.

1. Bardeen J, Cooper LN, Schrieffer JR (1957) Theory of superconductivity. *Phys Rev* 108:1175–1205.
2. Cuk T, *et al*. (2005) A review of electron-phonon coupling seen in the high-Tc superconductors by angle-resolved photoemission studies (ARPES). *Phys Stat Sol* 242:11–29.
3. Maksimov EG, Kulic ML, Dolgov OV (2008) Bosonic spectral function in HTSC cuprates. Part I: Experimental evidence for strong electron–phonon interaction. e-Print Archive, http://xxx.lanl.gov/abs/cond-mat/0810.3789.
4. Carbone F, Yang D-S, Giannini E, Zewail AH (2008) Direct role of structural dynamics in electron-lattice coupling of superconducting cuprates. *Proc Natl Acad Sci USA* 105:20161–20166.
5. Williamson JC, Cao J, Ihee H, Frey H, Zewail AH (1997) Clocking transient chemical changes by ultrafast electron diffraction. *Nature* 386:159–162.
6. Zewail AH (1994) *Femtochemistry: Ultrafast Dynamics of the Chemical Bond* (World Scientific, Teaneck, NJ).
7. Gedik N, Orenstein J, Liang R, Bonn DA, Hardy WN (2003) Diffusion of nonequilibrium quasiparticles in a cuprate superconductor. *Science* 300:1410–1412.
8. Wang F, Dukovic G, Brus LE, Heinz TF (2004) Time-resolved fluorescence of carbon nanotubes and its implication for radiative lift times. *Phys Rev Lett* 92:177401.
9. Perfetti L, *et al*. (2006) Evolution of the electronics structure of 1T-TaS2 through the insulator-metal transition. *Phys Rev Lett* 97:067402.
10. Devereaux TP, Cuk T, Shen Z-X, Nagaosa N (2004) Anisotropic electron-phonon interaction in the cuprates. *Phys Rev Lett* 93:117004.
11. Sato T, *et al*. (2003) Observation of band renormalization effects in hole-doped high-Tc superconductors. *Phys Rev Lett* 91:157003.