

## Optical Cooper pair breaking spectroscopy of cuprate superconductors

Y. G. Zhao,\* Eric Li, Tom Wu, S. B. Ogale, R. P. Sharma, and T. Venkatesan  
*Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, Maryland 20742*

J. J. Li, W. L. Cao, and C. H. Lee  
*Department of Electrical Engineering, University of Maryland, College Park, Maryland 20742*

H. Sato and M. Naito  
*NTT Basic Research Laboratories, 3-1 Morinosato Wakamiya, Atsugi-shi Kanagawa 243-0198, Japan*  
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The photon energy dependence of the optical Cooper pair breaking rate (CPBR) is studied for compressibly strained  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  (LSCO) films and  $\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$  (YBCZO) thin films, and compared to that in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO). Unlike YBCO, the CPBR for LSCO does not show an obvious photon energy dependence. In YBCZO, the CPBR shows a strong energy dependence similar to YBCO, but with a redshift in the peak position. Analysis of these results strongly favors a physical picture based on electronic phase separation in high- $T_c$  superconductivity.

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The mechanism of high-temperature superconductivity (HTS) remains a mystery to date, although significant understanding has been developed over the years in elucidating the key underlying factors. Various models have been proposed for the understanding of HTS.<sup>1</sup> It is commonly believed that a strong correlation between electrons play a very important role in this system; however, the manner in which such a correlation unfolds as a collective behavior is still not understood. Recently, experimental evidence has accumulated in favor of the occurrence of electronic phase separation (EPS) in such strongly correlated systems; the so-called stripe phase picture being one manifestation of such a scenario.<sup>2</sup> The EPS picture implies an inhomogeneity of both charges and spins in HTS. It is not yet clear whether the EPS or stripes are central to the phenomenon of high- $T_c$  superconductivity.

Based on thermal difference reflectance (TDR) spectroscopy work on cuprates, Little, Collins, and Holcomb<sup>3</sup> concluded that phonons and a high-energy electronic excitation (ranged over 1.6–2.3 eV) are jointly important for pairing in HTS. Stevens *et al.*<sup>4</sup> performed pump-probe measurement on  $\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$  (YBCO) employing excitation by 3-eV photons, with the probe beam detecting the excited state and its relaxation. Their results also showed an absorption peak around 1.5 eV, broadly consistent with the TDR measurement; however, their interpretation of the origin of the peak differs from that of Little, Collins, and Holcomb.<sup>3</sup> In our previous work<sup>5</sup> on electrically characterized optical pair breaking (which differs distinctly from the all-optical measurements by other researchers), we observed a fairly sharp peak in the Cooper pair breaking rate around 1.5 eV for YBCO, confirming a resonance. The existence of such a sharp feature is indeed surprising if one were to think of the superconductor as a uniform conductor. Noting that all these works reflect the importance of states separated in energy by about  $\sim 1.5$  eV we decided to probe the case further by examining other cuprate systems, namely  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  (LSCO) and Zn-doped YBCO. We find that over the energy

range in which YBCO shows resonance, LSCO does not show any obvious photon energy dependence. On the other hand,  $\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$  (YBCZO) does show a strong photon energy dependence, but with a redshift of the peak feature. We argue that it is difficult to reconcile all these data without invoking the electronic phase separation picture for HTS cuprates.

$\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$  thin films were prepared by pulsed laser deposition on (100)LaAlO<sub>3</sub> substrates. The thickness of the films was about 100 nm with  $T_c \sim 58$  K.  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  thin films were prepared by reactive coevaporation (electron beam evaporation) on (001)LaSrAlO<sub>4</sub> substrates following Sato and Naito.<sup>6</sup> The thickness of the films was about 100 nm with  $T_c$  between 40 and 43 K. The films were patterned by standard photolithographic technique to obtain coplanar waveguide devices. The patterning process decreased the  $T_c$  of LSCO films to 34 K. The sketch of the experimental setup and the device (essentially an optically controlled opening switch) can be found in our previous paper.<sup>7</sup> The device was mounted on a cold finger located in a vacuum cryogenic chamber, and biased with a dc current. The device was illuminated with 100-fs pulses from a Ti:sapphire laser system, including an oscillator and a regenerative amplifier with an ability to deliver 5  $\mu\text{J}$  pulse at a repetition rate of 9 kHz. The high peak power and suitable repetition rate allow efficient fast switching without thermal heating problems, as discussed earlier.<sup>5,7</sup> The wavelength of the laser was tunable within the range 760–860 nm (1.63–1.44 eV). When the ultrashort laser pulse illuminated the bridge, transient switch current wave forms were produced instantaneously, resulting in a fast drop of the current flowing through the device. These wave forms were monitored by a fast sampling oscilloscope with a temporal resolution of 20 ps. In the experiment, great care was taken to keep the laser power constant and the beam focused on the superconducting bridge.

Figure 1 shows a typical wave form of the fast optical response for LSCO films. The rise and fall times of the signal are around 40 ps. This wave form is similar to that for

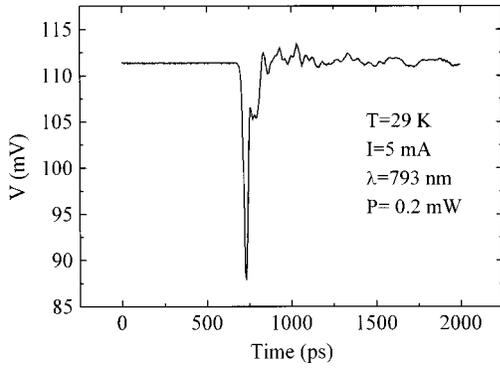


FIG. 1. The wave form of the fast optical transient signal related to the Cooper pair breaking.

YBCO.<sup>5</sup> It has been established that this signal is related to Cooper pair breaking,<sup>8,9</sup> which changes the kinetic inductance of the superconducting waveguide. The amplitude of this signal can be expressed as  $V = IR(\Delta L_{\text{kin}}/\Delta t)/(2\Delta L_{\text{kin}}/\Delta t + 4R)$ ,<sup>5,7</sup> where  $I$  is the bias current,  $R$  is  $50 \Omega$ ,  $\Delta t$  is the pair breaking time,  $L_{\text{kin}}$  is the kinetic inductance of the superconducting bridge, and  $\Delta L_{\text{kin}}/\Delta t = (m^*l/(e^2wdn_s^2))(\Delta n_s/\Delta t)$ . In this formula,  $m^*$ ,  $n_s$ , and  $e$  are the effective mass, the density, and the charge of superconducting carriers, respectively. Parameters  $d$ ,  $l$ , and  $w$  are the thickness, length, and width of the bridge, respectively. Thus, from the amplitude  $V$  measured using a fast oscilloscope, we can obtain  $\Delta L_{\text{kin}}/\Delta t$ . Since  $\Delta L_{\text{kin}}/\Delta t$  is proportional to the pair-breaking rate  $\Delta n_s/\Delta t$ , the temperature and photon energy dependence of the pair-breaking rate can be studied. In such an argument we assume that  $m^*$  is fixed. However, in the stripe phase picture, this aspect may have to be reexamined.

Figure 2 shows the temperature dependence of  $\Delta L_{\text{kin}}/\Delta t$  for LSCO films. The behavior is similar to that in YBCO, and can be explained qualitatively by using a two-fluid model.<sup>5</sup> Note that  $\Delta L_{\text{kin}}/\Delta t$ , as given by  $[ml/(e^2wdn_s^2)](\Delta n_s/\Delta t)$ , is essentially proportional to  $\Delta n_s/\Delta t/n_s^2$ , since  $m, l, e, w$ , and  $d$  are constants. If  $\Delta n_s/\Delta t$  is temperature independent or weakly temperature

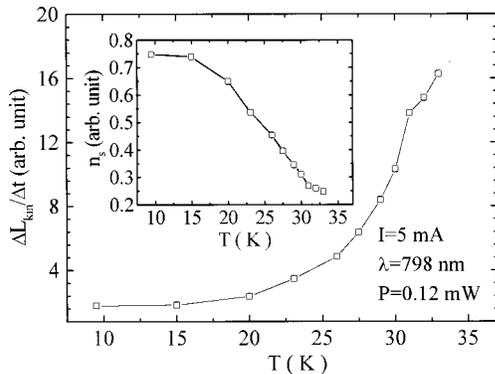


FIG. 2. Temperature dependence of  $\Delta L_{\text{kin}}/\Delta t$  for  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  thin film. The inset shows the square root of  $1/\Delta L_{\text{kin}}/\Delta t$ , which is proportional to  $n$  if we assume  $\Delta n_s/\Delta t$  is temperature independent or weakly temperature dependent.

dependent, the temperature dependence of the amplitude of  $\Delta L_{\text{kin}}/\Delta t$  will be determined by  $1/n_s$ .<sup>2</sup> Since  $n_s$  increases with decreasing temperature,  $\Delta L_{\text{kin}}/\Delta t$  is expected to be reduced rapidly as the temperature decreases. The inset of Fig. 2 shows the square root of  $1/\Delta L_{\text{kin}}/\Delta t$ , which is proportional to  $n_s$  if  $\Delta n_s/\Delta t$  is temperature independent or weakly temperature dependent. The temperature dependence of  $n_s$  shown here is different from the  $n_s(T)$  curve reported by Hardy *et al.*,<sup>10</sup> which shows a linear temperature dependence of  $n_s$  at low temperature, consistent with the  $d$ -wave pairing mechanism. This discrepancy implies that  $\Delta n_s/\Delta t$  has some temperature dependence. Indeed, it has been shown that the charge transfer ( $O2p$  to  $\text{Cu}3d$ ) gap, which is related to the photon absorption, increases with temperature,<sup>11</sup> and the lifetime of the quasiparticles produced by the Cooper pair-breaking process is also expected to change with temperature.<sup>12</sup> If we use the  $n_s(T)$  data obtained from other experiments,<sup>10</sup> it is possible to estimate the  $T$  dependence of  $\Delta n_s/\Delta t$ .

Figure 3(a) gives the photon energy dependence of  $\Delta L_{\text{kin}}/\Delta t$  for LSCO thin films. Unlike YBCO, it does not show any noticeable photon energy dependence. The YBCO data are redrawn in Fig. 3(c) for comparison. This indicates that the resonance of Cooper pair breaking observed in YBCO (Ref. 5) is intrinsic. Figure 3(b) gives the photon energy dependence of  $\Delta L_{\text{kin}}/\Delta t$  for  $\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$ . It shows a dramatic photon energy dependence, as was seen in YBCO. It is clear that the resonance peak shifts to lower energies as compared to that of the resonance peak in YBCO shown in Fig. 3(c).<sup>5</sup>

Now we turn to the analysis of our results. In Ref. 3, the high-energy electronic excitation ( $\sim 1.5$ – $1.7$  eV), which is suggested to be related to the pairing in HTS, is attributed to the energy of the  $d^9-d^{10}L$  charge-transfer excitation associated with the  $\text{CuO}_2$  network which is common to HTS systems. This charge-transfer excitation was also observed in superconducting YBCO by electron-energy-loss spectra.<sup>13</sup> However, the feature we observed in the CPBR spectrum of YBCO near 1.5 eV is considerably narrower (100 meV) than that (500 meV) of the peak in Ref. 3. We argue that the observation of such a sharp resonance is hard to understand for any homogeneous conducting state. On the other hand, the presence of insulating regions in the superconducting state, as is envisaged in the EPS or stripe phase scenario, can lead to narrow absorption features provided that the absorption-induced perturbation of the insulating (antiferromagnetic) state directly couples with the paired hole system and breaks pairs. Interestingly, the insulating  $\text{YBa}_2\text{Cu}_3\text{O}_6$  compound has a charge-transfer excitation peak (from the  $O2p$  to  $\text{Cu}3d$  upper Hubbard band) around 1.7 eV.<sup>14–16</sup> In the small phase-separation length scale anticipated in the EPS or stripe scenario, there could be a renormalization of the energy of this peak, causing its shift to lower energy. To what extent the charge transfer peak for the insulating domains and stripes in superconducting  $\text{YB}_2\text{Cu}_3\text{O}_{7-\delta}$  is different from that of the insulating bulk  $\text{YB}_2\text{Cu}_3\text{O}_6$ , is still an open question, which needs theoretical inputs. In a related context it is useful to point to a recent observation that the screening of phonon modes in high- $T_c$  superconductors is

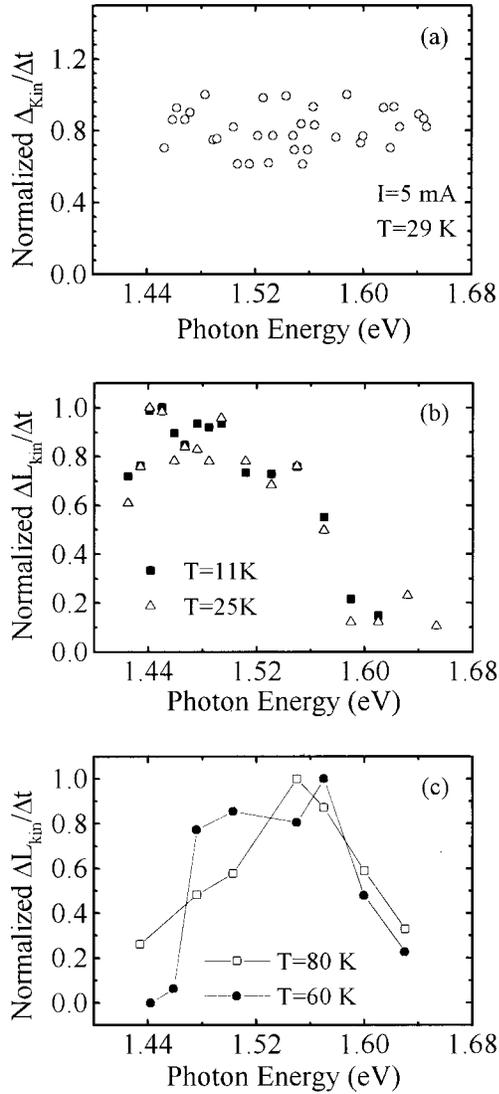


FIG. 3. Photon energy dependence of  $\Delta L_{\text{kin}}/\Delta t$  for (a)  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  (b)  $\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$  and (c)  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin film (see Ref. 5).

poor or totally absent, and the majority of the phonon modes have oscillator strengths similar to those found in the insulating materials.<sup>17</sup> Therefore, it is reasonable to expect that photons are absorbed mainly by the insulating domains and stripes rather than the metallic ones, and that the insulating domains and stripes dominate the optical properties of high- $T_c$  superconductors.

In our experiment we selectively and electrically probe the broken Cooper pairs in an ultrafast measurement. The speed and the concept of our measurement are key to the results we obtain. Kataev *et al.* studied the temperature dependence of the spin fluctuation frequency for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  samples by ESR of Gd spin probes.<sup>18</sup> The spin fluctuation frequency shows strong temperature dependence and changes from  $3 \times 10^{13}$  Hz at 250 K, to about  $10^{10}$  Hz at 5 K. Therefore, the time scale for the spin fluctuation is  $10^{-13}$  s at high temperature and  $10^{-10}$  s at low temperature. In our experiment, the width of the laser pulse is only 100 fs ( $10^{-13}$  s), which is very fast in comparison

with the spin fluctuation time scale. If the latter is considered to represent stripe fluctuations, our measurement would essentially reflect the snapshot picture of charge and spin domains or stripes at a certain time. In contrast, the measurement in Ref. 3 represents a time average. This time-scale difference could be a factor responsible for the different widths obtained in our experiment and the TDR experiment.

As discussed above, a reasonable explanation for the CPBR, in the case of YBCO is the charge-transfer excitation in spatially confined domains and stripes of antiferromagnetic (AF) insulating regions in YBCO. It is possible that a similar excitation in the AF insulating regions of LSCO is out of the photon energy range employed in this work. For example, the charge transfer energy for  $\text{La}_2\text{CuO}_4$  is about 2 eV,<sup>14</sup> which is higher than the charge transfer energy of 1.7 eV for  $\text{YBa}_2\text{Cu}_3\text{O}_6$ .<sup>15,16</sup> Hence, even after renormalization and shift, it may not fall in the range of the measurement. An alternate proposal for the absence of the CPBR in the case of the 214 film could be that both static and dynamic spin and charge stripes are absent in the compressibly strained LSCO thin films.<sup>19</sup> At this stage the existence of the dynamic stripes is still an open issue. Further work is clearly needed to extend the photon energy to both lower- and higher-energy sides, especially close to the 2-eV charge-transfer gap of  $\text{La}_2\text{CuO}_4$ , to verify whether CPBR exists in LSCO, and hence a similar conclusion as for YBCO can be drawn for the 214 case as well.

For Zn-doped  $\text{YB}_2\text{Cu}_3\text{O}_{7-\delta}$ , even though Zn is expected to be in a nonmagnetic  $3d^{10}$  state, its destruction of superconductivity is even stronger than magnetic ion such as Ni.<sup>20,21</sup> It has been found that Zn doping induces a magnetic moment on Cu sites around Zn,<sup>22-24</sup> and that this moment couples strongly with the conduction band at low temperature.<sup>25</sup> Charge localization was reported in Zn-doped  $\text{YB}_2\text{Cu}_3\text{O}_{7-\delta}$ , and has been explained by the destruction of the local AF correlation among Cu spins by Zn.<sup>26</sup> However, a recent NMR result suggests that the AF correlations are enhanced rather than destroyed around Zn.<sup>27</sup> Therefore, another scenario is needed to explain the localization effect. It is also suggested that Zn impurities are surrounded by extended regions whose magnetic properties are strongly modified already far above  $T_c$ , and wherein superconductivity never develops.<sup>28</sup> Superconductivity is then confined to regions far from the Zn impurities. For Zn-doped  $\text{Bi}_2\text{Sr}_2\text{CaCuO}_{8+\delta}$ , a scanning tunneling microscopy study also shows that superconductivity is strongly suppressed within 1.5 nm of the scattering sites.<sup>29</sup> In the stripe phase model, superconductivity is related to the fluctuation of the stripes.<sup>30</sup> It has been suggested that the pinning of the dynamically fluctuating stripes results in a suppression of the superconductivity.<sup>31</sup> In our experiment, Zn doping should not affect the results very much in terms of the time scale, since the stripes are static to the probing light pulse even for the undoped YBCO because of our ultrafast technique. However, Zn doping may affect the charge-transfer gap because of the suggested modification to the bands, which leads to the shift of the CPBR resonance peak to lower energy.

In summary, we have studied the photon energy dependence of the Cooper pair breaking rate (CPBR) for  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  and  $\text{YBa}_2\text{Cu}_{2.92}\text{Zn}_{0.08}\text{O}_{7-\delta}$  thin films, and

compared them with that in YBCO. The strong photon energy dependence of CPBR in YBCO and YBCZO (with a redshift), and its absence in LSCO strongly favor the electronic phase separation (or stripe) picture for cuprates; the absorption responsible for the measured pair breaking is the

charge-transfer excitation in the insulating antiferromagnetic domains confined between charge lines.

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\*Present address: Dept. of Physics, Tsinghua University, Beijing, 100084, P. R. China.

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