Editors' Suggestion

Signatures of Enhanced Superconducting Phase Coherence in Optimally Doped $Bi_2Sr_2Y_{0.08}Ca_{0.92}Cu_2O_{8+\delta}$ Driven by Midinfrared Pulse Excitations

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Optimally doped cuprate are characterized by the presence of superconducting fluctuations in a relatively large temperature region above the critical transition temperature. We reveal here that the effect of thermal disorder, which decreases the condensate phase coherence at equilibrium, can be dynamically contrasted by photoexcitation with ultrashort midinfrared pulses. In particular, our findings reveal that light pulses with photon energy comparable to the amplitude of the superconducting gap and polarized in plane along the copper-copper direction can dynamically enhance the optical response associated with the onset of superconductivity. We propose that this effect can be rationalized by an effective *d*-wave BCS model, which reveals that midinfrared pulses result in a transient increase of the phase coherence.

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Many of the ingredients required for superconductivity in cuprates survive well beyond the region of the phase diagram where the actual macroscopic superconducting phase resides. An example of this hindered superconductivity is represented by the behavior of underdoped cuprates just above the critical temperature (T_c), where some hints indicate that pairing occurs, but the presence of the superconducting phase is inhibited either by a competing charge order or by the local nature of the pair incoherence (global phase incoherence), blocking the formation of a mesoscopic superconducting state [1–5]. Signatures of an incipient superconductivity at temperatures larger than T_c have been revealed also in optimally doped samples, where the superconducting fluctuations survive tens of Kelvin above the actual T_c [6–17]. The relative fragility of the superconducting phase, together with the underlying presence of its ingredients in large portions of the phase diagram, enables the possibility of controlling superconductivity through ultrashort light pulses.

While there is ample evidence that photoexcitation with ultrashort high photon energy pulses melts the superconducting phase under some specific conditions [6,8,9,18,19], it has been shown that the formation of a superconducting phase can be triggered by midinfrared (MIR) excitations in regions of the phase diagram that are not superconducting at equilibrium [20–24]. The possibility of triggering the onset of quantum coherence through MIR excitations could open up new avenues to control quantum states of matter through light. Here we reveal that the time domain response of optimally doped $Bi_2Sr_2Y_{0.08}Ca_{0.92}Cu_2O_{8+\delta}$ (Y-Bi2212) to excitations with photon energies close to the superconducting gap ($2|\Delta| \approx 75$ meV) is highly anisotropic. While excitations with pump polarization along [100] (the copper-oxygen Cu-O axis) lead to a reduction of the superconducting gap, independent of the photon frequency, photoexcitations along [110] (the copper-copper Cu-Cu axis) seem to trigger an increase of phase coherence, which results in an enhancement of the dynamical response associated with the superconducting phase. The enhancement of the response with [110] excitation is explained within a simple BCS framework, with *d*-wave symmetry for the superconducting gap.

A much debated aspect of superconductivity in cuprates is that the onset of the superconducting phase is followed by a change of spectral weight at an energy scale orders of magnitude larger than the superconducting gap. In particular, in Bi2212, the opening of a superconducting gap at about 35–40 meV is accompained by a spectral weight redistribution at frequencies as high as several eV [25–27].

This is visible in time domain studies where the reflectivity in the visible range changes upon a sudden perturbation of the superconducting gap [28–34]. Here we leverage this characteristic and work under the assumption that the spectral response in the visible-near infrared region is intimately related to the superconductor response.

We performed pump-probe measurements Y-Bi2212. At optimal doping it presents a superconducting phase below $T_c = 97$ K, a pseudogap phase between T_c and $T^* \approx 135$ K and an unusual "strange-metal" phase for higher temperatures [35]. The sample has been excited with MIR ultrashort pulses ($h\nu \approx 70$ meV $\approx 2\Delta$, incident fluence $f = 0.09$ mJ cm^{-2}) obtained through difference frequency generation of two infrared pulses coming from a twin optical parametric amplification system. The transient reflectivity induced by the pump is probed by near-infrared (NIR) pulses ($h\nu \approx 1.63$ eV) generated by a noncollinear parametric amplifier system. The time delay between the two pulses can be tuned by changing the probe optical path through a μm-translation stage. Both pump and probe propagation directions are parallel to the c axis (that is, perpendicular to the Cu-O layer) and their polarizations were kept parallel in all measurements. External noise contributions to the transient reflectivity are reduced subtracting a reference signal (which does not interact with the sample) to the probe beam. The acquisition has been performed by a lock-in amplifier. More details on the experimental design can be found in the Supplemental Material [36].

The intensity map shown in Fig. 1(a) represents the relative variation of the reflectivity upon the pump excitation as a function of the time delay between the pump and the probe pulses (horizontal axis) and of the temperature of the sample (vertical axis). We focused our investigation in a temperature range from 80 to 110 K. For $T < T_c$, the reflectivity increases for about 1 ps after the arrival of the pump (at 0 ps) and then it starts decreasing through an exponential decay [brown line in Fig. 1(c)]. The characteristic time of the decay increases with temperature, it is maximum at $T = 97$ K (black line) and decreases at higher temperatures. The observed maximum of the time decay is a precise indicator of the superconducting-pseudgap phase transition [19,37].

The anisotropy of the gap of d-wave superconductors suggests a detailed analysis of the effects of excitations with different pump polarizations [38]. Figures 1(a) and 1(b) show the measured transient reflectivity for two different pulse polarizations: along [100] [the Cu-O axis, see Fig. 1(a)] and [110] [the Cu-Cu direction, see Fig. 1(b)]. We measured the transient reflectivity in both polarizations, using a pump photon energy around the characteristic energy of the system in the superconducting phase, that is $2|\Delta| \approx 75$ meV [39]. A convenient way to visualize the polarization dependence of our measurements is to subtract the two maps in Figs. 1(a) and 1(b), as displayed in the differential map of Fig. 2(a). The difference map reveals a sizable signal around T_c, for time delays from 0 to 2 ps, corresponding to the maximum response in the superconducting phase [red region at about 1 ps in Fig. 2(a)].

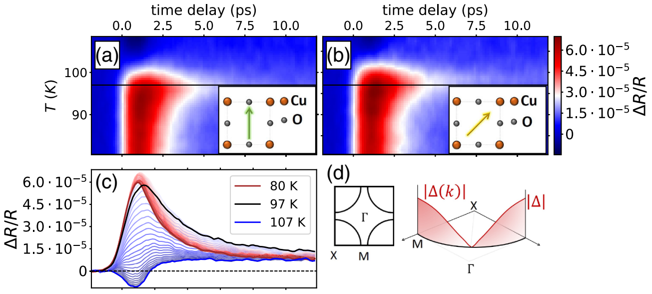

FIG. 1. MIR pump, optical probe measurements. (a) and (b) Reflectivity variation due to an impulsive excitation at time delay 0 ps by a MIR pulse ($h\nu \approx 70$ meV) as a function of the temperature of the sample (vertical axis). The two maps differ in the polarization of the impinging pump, as highlighted by the two insets, representing the direction of the polarization (green and orange arrow) in the Cu-O plane. (c) Time resolved signal at fixed temperatures for Bi2212: the brown line represents the characteristic superconductive signal, while the blue one refers to the pseudogap phase. The transition between the two phases is marked with the black line and is related to the maximum of the time decay. Light colored lines represent intermediate temperatures. (d) Sketch of the first Brillouin zone and of the superconducting d-wave gap amplitude $|\Delta(\mathbf{k})|$ in the reciprocal space. The black curved lines represent the Fermi surface.

The result is confirmed by the visual inspection of the temperature dependence at a fixed time delay (1 ps) for different polarizations of the pump, as shown in the inset of Fig. 2(a).

We observe an increase of response associated with the onset of the superconductivity when the pump is polarized

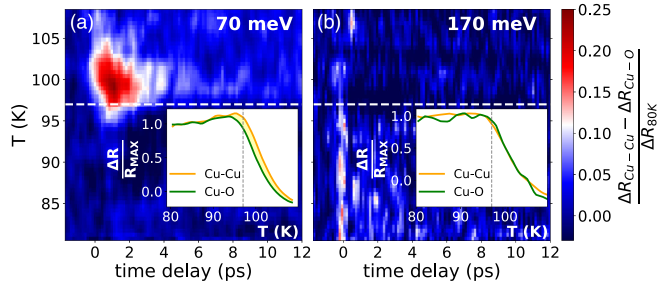

FIG. 2. Wavelength dependent anisotropy. Difference between the transient reflectivities due to Cu-Cu and Cu-O polarized pump in time (horizontal axis) and temperature (vertical axis), induced by excitations with (a) 70 and (b) 170 meV pump photon energies. The dashed lines highlight the critical temperature $T_c = 97$ K. The insets represent the response as a function of temperature at 1 ps time delay for Cu-Cu (orange line) and Cu-O (green line) polarized pump excitations for low and high pump photon energies (a and b, respectively). The gray dashed lines mark T_c. Both in the maps and in the insets the values of the reflectivity have been normalized to the maximum value of the response at 80 K ($\Delta R_{80\,K}$).

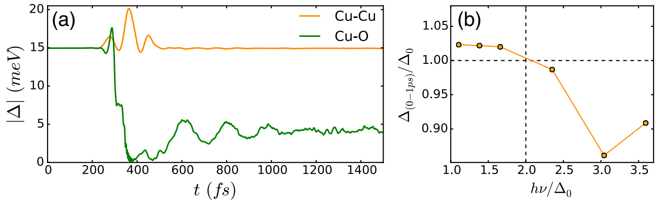

FIG. 3. *d*-wave BCS microscopic model. Results of the microscopic model: (a) Time evolution of the modulus of the superconducting gap ($|\Delta|$) in both Cu-Cu (orange line) and Cu-O (green line) excitation case. The maximum of the pump electric field is reached at about 350 fs. (b) Normalized integral of $|\Delta|$ in the time interval from 0 to 1 ps as a function of the pump photon energy ($\Delta_0 = |\Delta(t = 0)|$) for Cu-Cu polarized pump excitations.

along the Cu-Cu direction both above and below $T_c = 97$ K. We stress that this is an anisotropic response strongly dependent on the photoexcitation wavelength: in Fig. 2(b) we display the differential map retrieved for higher pump photon energy ($h\nu \approx 170$ meV), which reveals no anisotropy at any temperature. This result has been also confirmed by measurements at high pump fluence (see Fig. S4 of the Supplemental Material [36]) demonstrating that the superconducting signal can be increased by a pump polarized along the Cu-Cu axis at long wavelength whereas higher photon energy excitations suppress this effect.

In order to draw a picture of the possible physical scenario emerging from the anisotropic response to low photon energy ultrashort pulses, we implemented a microscopic description based on a generalized BCS Hamiltonian allowing for a *k*-dependent *d*-wave gap (for further details see the Supplemental Material [36]). While it is well known that a simple BCS formalism, disregarding first and foremost the presence of electronic correlations, cannot explain the whole cuprate phenomenology, we will argue here that it accounts well for the nonequilibrium response of the low-energy superconducting gap, at least at a qualitative level. From the described Hamiltonian [see Supplemental Material [36], Eq. (1)], it is possible, through density matrix formalism, to calculate the time evolution of several meaningful quantities (such as the superconducting gap amplitude $|\Delta|$).

The model predicts different behaviors depending on the frequency and the polarization of the pump pulse. In particular, Fig. 3(a) shows that low photon energy excitations with polarization parallel to the Cu-Cu direction are predicted to drive an instantaneous enhancement of the superconducting gap, while a pump polarization rotated 45° induces a dynamical quench of the gap. These results qualitatively rationalize the experimentally observed enhancement of the positive signal associated with the superconducting response, triggered by photoexcitation polarized along the Cu-Cu direction for pump photon energy $h\nu \approx 2|\Delta|$. On the other hand, the collapse of the superconducting signal in the Cu-Cu polarization configuration is predicted for higher pump photon energies, as shown in Fig. 3(b), where we display the transient decreases of $|\Delta|$ as a function of the pump photon energy.

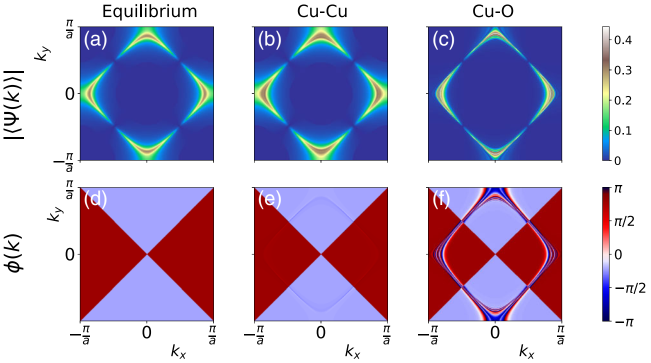

FIG. 4. Time dependent pair operator representation. (a)–(c) Modulus of the expectation value of the pair operator $\hat{\Psi}(\mathbf{k})$ in the reciprocal space in three different situations: (a) No excitation ("Equilibrium"), (b) During a pump excitation polarized along the Cu-Cu and (c) Cu-O direction. Analogously, pictures from (d) to (f) show the phase $\phi(\mathbf{k})$ of $\langle\hat{\Psi}(\mathbf{k})\rangle$, in the same three conditions.

In order to grasp the physical picture that emerges from this microscopic model, we calculated the expectation value of the pair operator $\hat{\Psi}(\mathbf{k})$ (see Supplemental Material [36]).

Figure 4 displays the modulus and the phase of the pair amplitude (in the first Brillouin zone) calculated in three different cases: at equilibrium and during a Cu-Cu and Cu-O low photon energy excitation ($h\nu \approx 2\Delta$). We observe that the value of $|\langle\hat{\Psi}(\mathbf{k})\rangle|$ around the Fermi surface is nearly unperturbed (and actually slightly enhanced) in both excitation schemes [Figs. 4(b) and 4(c)], i.e., that pairing is still present in both cases. On the other hand, the phase reveals a strong anisotropic response [Figs. 4(e) and 4(f)]: the model shows that Cu-O polarized excitations drive intense phase fluctuations, which are responsible for the collapse of $|\Delta|$ shown in Fig. 3(a). Cu-Cu polarized low-photon-energy excitations preserve instead phase coherence and enable an enhanced superconducting dynamical response.

We stress that the calculations were performed at a temperature $T < T_c$, where the amplitude of the superconducting gap $|\Delta|$ has a nonzero equilibrium value. This is an intrinsic limitation of the microscopic BCS model, which does not allow superconducting pairing at temperatures higher than the critical value (T_c). The data instead report a well visible enhancement of the superconducting behavior at temperatures up to ≈10 K above the equilibrium critical temperature T_c [inset of Fig. 2(a)].

In order to extend this effective description to higher temperatures, we propose to run calculations from a modified equilibrium state, whose features are justified in the following, maintaining the BCS framework. Different from standard BCS superconductors, cuprates exhibit signatures of strong superconducting fluctuations at temperatures larger

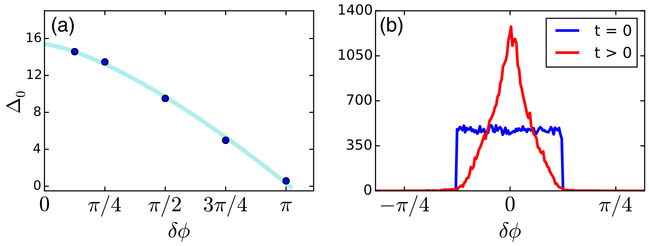

FIG. 5. Extension of the d-wave BCS model above the critical temperature. (a) Values of the superconducting gap at equilibrium $[\Delta_0 = |\Delta|(t = 0)]$ as a function of the amplitude of the phase noise. (b) Distribution of the phase values at equilibrium (blue line) and during the excitation polarized along the Cu-Cu direction (red line) for an initial noise amplitude of $\delta\phi = \pi/8$.

than T_c [7]. In particular, in optimally doped Bi2212, both equilibrium and time-domain techniques revealed superconducting fluctuations up to tens of Kelvin above the critical temperature [6,8–10]. This anomalous feature is commonly taken to imply the presence of Cooper pairs losing phase coherence; i.e., while the mesoscopic coherence vanishes above the transition temperature, pairing remains, together with phase correlations, which are local in space and time [7]. Transport [12,13] and magnetization [11] studies suggest that the local correlations lead to a universal superconducting percolative regime above T_c consistent with a percolation picture of the phase diagram [40]; in particular, a local superconducting gap distribution was able to explain [12] the presence of an effective average gap above T_c in photoemission experiments [41] and the exponential tail exhibited by several observables. Here we observe qualitatively the same signatures of local correlations persisting up to ~10 K above T_c, a temperature range similar to previous studies [12,13,40,41].

In order to explore these effects within the generalized BCS model, we employ the following simple procedure. We proposed a new equilibrium state artificially built by adding a random noise to the phase of the original state pair amplitude $\phi(\mathbf{k})$, while retaining its modulus $(|\langle\hat{\Psi}(\mathbf{k})\rangle|)$. The phase noise introduced in the model leads to a reduction of the gap, as shown in Fig. 5(a), in which the dependence of the gap amplitude on the maximum value for the phase fluctuations $(\delta\phi)$ is plotted. Calculations of the dynamic response, starting from this inhomogeneous (in momentum space) equilibrium state, reveal that Cu-Cu low-photon-energy excitations induce a significant enhancement of phase coherence (and negligible variation in the amplitude) of the pair operator, as highlighted in Fig. 5(b), which depicts a histogram of the phase distribution before (blue line) and during (red line) the photoexcitation (350 fs), for an initial fluctuation of $\delta\phi = \pi/8$ (value chosen for sake of clarity). The plot reveals that the phase distribution, which becomes narrower after the excitation, leads to an enhanced superconducting response. We argue that this scenario rationalizes the enhancement of an out-of-equilibrium superconducting behavior above T_c, which could therefore be associated with a light-driven boost of phase coherence.

The scenario that emerges from our pump-probe experiments reveals the capability to enhance the transient response associated with superconducting fluctuations in cuprates by means of photoexcitations with low-energy photons polarized in the Cu-Cu direction, which is completely suppressed by a pump excitation with polarization parallel to the Cu-O axis. The effective d-wave BCS description of the interaction of the superconductor and pulsed electromagnetic radiation is in qualitative agreement with the experimental results. Moreover it allows us to ascribe the observed dynamical increase of the superconductive response to a light-driven enhancement of phase coherence below and above T_c, where thermodynamic constraints make the superconducting equilibrium state unattainable. On the other hand, it is unclear what will happen in underdoped cuprates, where correlations may play a stronger role. The d-wave BCS model describes well the electrodynamic response of the superconducting phase, but, as discussed, it lacks in describing the effects of strong correlations. Although the discussion of underdoped cuprates is beyond the scope of this work, our conclusions suggest that a field-driven increase of phase coherence may play a role in their response. This opens the intriguing perspective of driving the onset of quantum coherence in these materials by mid-IR excitation.

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