Dynamics of fluctuations in high-temperature superconductors far from equilibrium

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Despite extensive work on high-temperature superconductors, the critical behavior of an incipient condensate has so far been studied under equilibrium conditions. Here, we excite Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ with a femtosecond laser pulse and monitor the subsequent dynamics of the mid-infrared conductivity. The time-resolved THz measurements discriminate the temperature regimes where superconductivity is either coherent, fluctuating or vanishingly small. Above the transition temperature $T_c$, we make the striking observation that the recovery to equilibrium displays a power-law behavior and scaling properties, both for optimally and underdoped superconductors. Our findings provide hints of universality in systems far from equilibrium.

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Many physical properties of high-temperature superconductors cannot be understood in the framework of a mean-field theory. For example, an incipient condensate with short coherence length fluctuates in space and time, leading to precursor effects of the superconducting phase. As a consequence, quantities such as conductivity [1–4], heat capacity [5], diamagnetic susceptibility [6] and the Nernst signal [7] may increase considerably.

The superconducting fluctuations above typically 1.5 eV) are not well-suited for detecting superconductivity. The time-resolved THz measurements discriminate the temperature regimes where superconductivity is either coherent, fluctuating or vanishingly small. Above the transition temperature $T_c$, we make the striking observation that the recovery to equilibrium displays a power-law behavior and scaling properties, both for optimally and underdoped superconductors. Our findings provide hints of universality in systems far from equilibrium.

In this Letter, we address this gap of knowledge. Many physical properties of high-temperature superconductors cannot be understood in the framework of a mean-field theory. For example, an incipient condensate with short coherence length fluctuates in space and time, leading to precursor effects of the superconducting phase. As a consequence, quantities such as conductivity $\sigma(\nu)$, heat capacity $C(\nu)$, diamagnetic susceptibility $\chi(\nu)$ and the Nernst signal $\nu$ may increase considerably when the temperature approaches the critical value $T_c$.

For cuprates, most of these observations have successfully been described by the Ginzburg-Landau (GL) model or quantum-field theories [8–11]. Suitable extensions of these approaches to systems in nonequilibrium states predict critical dynamics and scaling laws [12]. Critical exponents independent of specific material properties may help extend the concept of “universality classes” to systems far from equilibrium. These phenomena should not only occur in condensed-matter systems but also in high-energy physics and ultracold atomic gases [12, 13]. An ideal protocol for the detection of critical dynamics requires that superconducting correlations are monitored just above the transition temperature, where the system is expected to exhibit universal behavior. In order to induce and measure the non-equilibrium, regime ultrafast pump-probe spectroscopy is a very promising approach [14, 21]. Unfortunately, optical probes (photon energy of typically 1.5 eV) are not well-suited for detecting superconducting fluctuations above $T_c$ because of the presence of competing signals [15, 16].

In this Letter, we address this gap of knowledge by ultrafast time-resolved detection of the conductivity in the mid-infrared. Thin films of the superconductor Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (BSCCO) are excited by optical laser pulses and probed by phase-locked electromagnetic pulses with center frequency of 20 THz and a duration of $\sim$ 100 fs [18]. The corresponding photon energy of 80 meV is comparable to the single-particle gap of BSCCO, thereby making the ultrashort THz pulse a highly sensitive probe of superconducting correlation [18, 19, 21, 22]. By these means, we identify a critical scaling behavior and a universal power law in the ultrafast dynamics of the incipient condensate.

Experimental details. Samples used are 150-nm thin films of optimally doped ($T_c = 91$ K) and underdoped ($T_c = 68$ K) BSCCO. The THz spectrometer is driven by laser pulses from a Ti:sapphire laser oscillator (pulse duration of 10 fs, center wavelength of 780 nm, repetition rate of 80 MHz). Part of the laser output is used to excite the sample with an incident fluence of about 16 $\mu$J cm$^{-2}$. We simultaneously measure the THz electric field $E(t,T)$ transmitted through the sample in thermodynamic equilibrium at temperature $T$ and the electric-field change $\Delta E(t,\tau)$ induced by the pump pulse. Here, $t$ denotes the time of the THz pulse with respect to the field maximum around $t = 0$, and $\tau$ is the delay between pump and probe pulse.

Equilibrium case.— Figure 1(a) shows THz field traces $E(t,T)$ after having traversed optimally doped BSCCO films in equilibrium at various temperatures $T$. To extract the conductivity $\sigma(\nu)$ as a function of THz frequency $\nu$, we apply a Fourier transformation and standard Fresnel formulas to the data of Fig. 1(a). The real part of the THz conductivity is shown in Fig. 1(b), and we find that $\text{Re} \sigma(\nu)$ decreases with frequency for $T > T_c = 91$ K whereas it follows the opposite trend in the superconducting phase with $T < T_c$. These results are in excellent agreement with previous reports based on Fourier transform spectroscopy [24] and ellipsometry [25].

Instead of plotting $\text{Im} \sigma$, it is often more instructive to consider a generalized and frequency-dependent Drude-type scattering rate $\Gamma(\nu) = -2\pi\nu \text{Im} \sigma(\nu)/\text{Re} \sigma(\nu)$ [26]. Figure 1(c) shows that $\Gamma(\nu)$ strongly depends on sample temperature for frequencies below 20 THz, whereas less pronounced variations are observed above [25, 26]. Figure 1(d) displays the temperature dependence of $\Gamma$ at a frequency of $\nu^* = 10$ THz. When temperature is lowered from 300 to 100 K, we observe a linear decrease, that turns into an abrupt drop at the phase transition into...
the superconducting phase \((T \sim 90 \text{ K})\).

The physical interpretation of this sudden decrease is straightforward: the emergence of the superconductivity gap \((T \sim T_c)\) reduces the density of electronic states (DOS) in the vicinity of the Fermi energy, thereby reducing the number of scattering channels of the electrons \([27]\). In the fluctuating regime above \(T_c\), the DOS becomes a function of local gap amplitude and of the superconducting coherence length \(\xi\). The latter reflects the typical distance over which the superconducting order parameter maintains its amplitude and phase \([8]\), and it is expected to diverge exactly at the transition temperature. Theoretical models of two-dimensional superconductors \([28]\) predict that the mid-infrared conductivity scales with \(\ln(\xi/\xi_0)\) where \(\xi_0\) is the value of the coherence length at zero temperature. Therefore, THz pulses are an excellent probe of superconducting fluctuations \([3]\).

We note in passing that measuring \(\Gamma\) does not require to detect the full THz waveform. Comparison of Figs. 1(d) and 1(e) shows that the field variation \(\Delta E(T) = E(t^*, 6 \text{ K}) - E(t^*, T)\) in the transmitted THz field at time \(t^* = 50 \text{ fs}\) as a function of \(T\).

FIG. 1: Equilibrium data of optimally doped BSCCO \((T_c = 91 \text{ K})\). (a) Electric field \(E(t, T)\) of the THz waveform transmitted through the BSCCO film as a function of time \(t\). Each sample temperature \(T\) is indicated by a different line color. (b) Real part \(\text{Re} \sigma(\nu)\) of the THz conductivity and (c) electronic scattering rate \(\Gamma(\nu)\) versus frequency \(\nu\) and for various temperatures \(T\). (d) \(\Gamma\) at \(\nu = \nu^* = 10 \text{ THz}\) as a function of temperature \(T\). (e) Change \(\Delta E(T) = E(t^*, 6 \text{ K}) - E(t^*, T)\) in the transmitted THz field at time \(t^* = 50 \text{ fs}\) as a function of \(T\).

FIG. 2: Nonequilibrium data of optimally doped BSCCO \((T_c = 91 \text{ K})\). (a) Pump-induced changes \(\Delta E(\tau) = E(t^*, -1 \text{ ps}) - E(t^*, \tau)\) of the transmitted electric field as a function of pump-probe delay \(\tau\). (b) Signal magnitude \(\Delta E(\nu^*)\) and (b) relaxation time \((\partial \ln \Delta E/\partial \tau)^{-1}\) right after sample excitation \((\tau^* = 50 \text{ fs})\) as a function of temperature (red circles). Blue squares result from a model based on the TDGL equation. The transition temperature \((T_c = 91 \text{ K})\) and the onset of superconducting fluctuations \((T_o = 130 \text{ K})\) are indicated by arrows.
ambient temperatures $T$. As above, we consider the THz transients at $t^* = 50$ fs. Following our observation for the equilibrium case [Figs. 1(d) and 1(e)], it is reasonable to ascribe $\Delta E(\tau)$ to the pump-induced change $\Delta \Gamma$ in the electronic scattering rate. As shown by Fig. 2(a), magnitude and recovery time of $\Delta E(\tau)$ strongly depend on sample temperature. Also, the dynamics deviates substantially from an exponential decay and slows down with increasing pump-probe delay. To better illustrate this behavior, Figs. 2(b) and 2(c), respectively, display the signal magnitude $\Delta E(\tau)$ and decay time $(\partial \ln \Delta E/\partial \tau)^{-1}$ right after excitation at $\tau = \tau^* = 50$ fs as a function of sample temperature $T$.

As seen in Fig. 2(b), $\Delta E$ and, thus, $\Delta \Gamma$ are much larger when $T$ is below the transition temperature. Comparison to Fig. 1(d) indicates that the larger changes arise from the reduction of the superconducting gap. Above $T_c$, we still observe an appreciable $\Delta E$ that above a temperature $T_o$ turns into a relatively flat and linearly decreasing curve. As seen in Fig. 2(c), the initial decay time of the pump-probe signal exhibits an analog behavior: it is large below $T_c$, then drops sharply and turns into a flat curve above $T_o$. We attribute these temperature intervals to the three different regimes: a superconducting phase ($T < T_c$), a fluctuating superconductor ($T_c < T < T_o$) and a metallic phase ($T > T_o$). From Fig. 2, we estimate that $T_c = 130$ K, which is in good agreement with values extracted from measurements of transport [1], specific heat [5], diamagnetism [6] and the Nernst effect [7].

In the temperature region from $T = T_o$ down to the critical temperature $T_c$, the system is governed by an increasing coherence length $\xi$ [8]. The characteristic time $\tau_\xi$ it takes a Cooper pair to cover the coherence length should increase accordingly. The scaling hypothesis suggests that the relaxation at long times should follow a universal power law $\langle 1/\tau_c(T) \rangle ^{-\alpha}$ where $\alpha$ is a critical exponent that only depends on the observable considered, the symmetry of the order parameter and the dimension of the system [14].

Figure 3 shows the key result of this paper: the striking observation that the apparently diverse dynamics of Fig. 2(a) indeed obey a scaling law. As seen in Fig. 3, all decay curves for $T_c < T < T_o$ fall onto one when the time axis $\tau$ is normalized by a suitable time $\tau_c(T)$. More precisely, we find that

$$\Delta E(\tau) = \frac{\Delta E(\tau^*)}{1 + (\tau/\tau_c(T))^\alpha}$$

(1)

and $\alpha = 1.1$ provide a very good description of our experimental data. The inverse scaling factor $\tau_c^{-1}$ is plotted as a function of temperature in the inset of Fig. 3(a). It approximately follows a linear relation $0.45$ ps$^{-1} + (T/T_c - 1)6.5$ ps$^{-1}$. The strong temperature dependence of $\tau_c$ reflects the critical slowing down and the increasing coherence length of superconducting fluctuations while approaching the transition temperature.

**Theoretical model.**—We now simulate our experimental finding using the two-dimensional, time-dependent Ginzburg-Landau (TDGL) model, which is justified for BSCCO due to the strong anisotropy of this layered compound. Temporal variations of the vector potential are neglected due to the large ratio between London penetration depth and coherence length [30, 31]. In this coarse-grained description, the GL time $\tau_{GL}$ is the characteristic response time of the order parameter $\psi(x, \tau)$. The remaining degrees of freedom are assumed to evolve much faster than $\psi$ and merely remain as noise $\eta(x, \tau)$ that affects the dynamics of $\psi$ as a source term. We note that for our experimental conditions, the nonlinear term in the TDGL equation is very small, and the fluctuations can be considered to a very good approximation as a free classic field.

We identify the photoexcited state $\psi(x, \tau^*)$ with a stationary solution of the TDGL equation at the elevated temperature $T + 17$ K. This value is obtained by comparing the transient $\Delta E(\tau)$ of Fig. 2(b) with the equilibrium $\Delta E(T)$ of Fig. 1(e). In contrast, the fast degrees of freedom summarized in $\eta$ are assumed to return to their equilibrium configuration characterized by temperature $T$ directly after excitation (sudden-quench hypothesis). This hypothesis is motivated by the observation that uncondensed electron in the normal phase transfer the pump-deposited excess heat to the lattice within $\sim 100$ fs [22]. We model $\eta$ by white noise with correlation function $\langle \eta(x, \tau)\eta(x', \tau') \rangle = 2S k_B T \delta(x - x') \delta(\tau - \tau')$ where $S$
is the area of the sample film. Finally, for the GL time, we assume \( \tau_{\text{GL}} = (T/T_c - 1)^{-1}0.25 \text{ps} \), consistent with the estimate \( \tau_{\text{GL}} = 48\pi \sigma_{\text{DC}} \lambda^2/c^2 \) based on temperature-dependent measurements of penetration depth \( \lambda \) and normal-state DC conductivity \( \sigma_{\text{DC}} \). From our modeling, we calculate the superconducting coherence length as a function of time \( \tau \) since optical excitation. To enable comparison with the measured \( \Delta E(\tau) \), we exploit the recent theoretical prediction that the mid-infrared transmittance of two-dimensional superconductors scales with \( \ln(\xi/\xi_0) \).

**Discussion.**—As shown by Figs. 2(b), 2(c) and 3, the estimated \( \ln(\xi/\xi_0) \) matches well the magnitude of \( \Delta E \) and the relaxation time for temperatures between \( T_c + 5 \text{ K} \) and \( T_o \). Below \( T_c + 5 \text{ K} \), the TDGL overestimates \( \tau_c \) by 70%. This discrepancy can be explained by an experimental crossover from the sudden-quench regime to an adiabatic regime occurring below \( T_c \) [3,12]. In addition, the experimental curves in Fig. 3 follow a power law whereas the TDGL model predicts an exponential relaxation. This discrepancy might originate from neglected interactions between fluctuations or from an incomplete thermalization of the fast degrees of freedom. Despite these shortcomings, the TDGL provides the correct \( (T - T_c) \)-scaling within the temperature range from \( T_c + 5 \text{ K} \) to \( T_o \).

While we find that the TDGL model provides a partially good description of our experimental data, agreement is less satisfactory for underdoped BSCCO. Figure 4(a) shows the photoinduced change \( \Delta E(T, \tau^*) \) in the transmitted field measured in samples with \( T_c = 68 \text{ K} \). We identify the onset of superconducting fluctuations with the upturn of the signal at \( T_o = 100 \text{ K} \). Note that \( \Delta E(T, \tau^*) \) changes slope at the temperature \( T^* = 200 \text{ K} \). This kink is likely due to the progressive reduction of the DOS known as “pseudogap”. The value of \( T^* \) found here compares well to other experiments on cuprates with similar doping level [55].

The inset shows the relaxation factor \( \tau_c^{-1} \) versus temperature as derived from the experiment (red circles) and a linear fit (solid line).

**FIG. 4: Nonequilibrium data of underdoped BSCCO (\( T_c = 68 \text{ K} \)) and their scaling behavior.** (a) Magnitude of the pump-probe signal \( \Delta E(\tau^*) \) and (b) its relaxation time \( (\partial \ln \Delta E(\tau^*)/\partial \tau)^{-1} \) right after sample excitation \( (\tau^* = 50 \text{ fs}) \) as a function of temperature (red circles). Transition temperature \( T_c = 68 \text{ K} \), onset of superconducting fluctuations \( T_o = 100 \text{ K} \) and pseudogap temperature \( T^* = 200 \text{ K} \) are indicated by arrows. (c) Measured pump-induced changes \( \Delta E(\tau) \) of the transmitted electric field as a function of rescaled pump-probe delay \( \tau/\tau_c(T) \) at temperatures \( T = 68, 78, 88, 98 \text{ and } 108 \text{ K} \) and 130 K. Curves are also normalized to the maximum signal value. The solid black line is a power-law fit [see Eq. (1)]. The inset shows the scaling factor \( \tau_c^{-1} \) versus temperature as derived from the experiment (red circles) and a linear fit (solid line).
direction may identify the parameters and experimental protocols that define universality classes. [23].

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