Superconducting fluctuation probed by the Higgs mode in Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ thin films

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Abstract

Superconducting (SC) fluctuations in cuprate superconductors have been extensively studied to gain a deep insight into the preformed Cooper pairs above the SC transition temperature $T_c$. While the various measurements, such as the terahertz (THz) optical conductivity, Nernst effect and ARPES measurements have provided the signature of the SC fluctuations, the onset temperature of the SC fluctuations depends on the measurement scheme. Here, we shed light on the Higgs mode to investigate the SC fluctuations, as it is the direct fingerprint of SC order parameter and can help elucidate the development of SC phase coherence. We perform the THz pump-optical probe spectroscopy for underdoped and overdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi2212) thin films. We observe oscillatory behavior in the pump-probe signal (THz Kerr signal) that sharply onsets slightly above $T_c$ and is attributed to the emergence of the Higgs mode. We also measure the THz optical conductivity and extracted the superfluid density $N_s$ using the two-fluid model. The onset temperature of the THz Kerr signal shows a good agreement with that of $N_s$ for both thin films, indicating that the Higgs mode emerges when SC phase coherence is established on the THz time scale. We also study five differently doped Bi2212 single crystals where the onset temperature of the Higgs mode is shown to be located 10-30 K above the SC $T_c$ dome.
I. INTRODUCTION

Among the various physical properties in cuprate superconductors, superconducting (SC) fluctuations have attracted intensive interests over decades [1] and been extensively investigated to uncover the Cooper pairing above $T_c$ by various experimental approaches: the terahertz (THz) [2–5] and microwave spectroscopy [6–8], torque magnetometry [9–12], Nernst [13,14], ARPES [15–18] measurements and ultrafast pump-probe spectroscopy [19–22]. Most of these experiments have investigated the onset temperature of the SC fluctuations ($T_{\text{Onset}}$) to understand how the SC coherence emerges from the complex metallic state, yet $T_{\text{Onset}}$ depends on experimental techniques and a unified picture on the onset of SC fluctuations is still lacking. For instance, in the THz and microwave spectroscopy the onset temperature $T_{\text{Onset}}$ has been identified at 10-20 K above $T_c$ in Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi2212) [2,3], La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [4–6] and YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) [8]. On the contrary, the Nernst and ARPES measurements assert much higher temperatures of $T_{\text{Onset}}$, up to 50 K above $T_c$ [13–18]. However, the Nernst signal was shown to be enhanced by the stripe order and its origin remains under debate [23]. Therefore, alternative measurements which are sensitive to the SC order parameter are called for.

To gain deeper insights into the onset of SC fluctuation, we focus on the collective amplitude mode of the SC order parameter, namely the Higgs mode, which directly manifests the development of the SC order parameter [24–26]. For decades, it has been considered that the Higgs mode in superconductors is invisible except for some compounds that exhibit the charge-density wave (CDW) and superconductivity [27–31]. However, with the advances of nonlinear THz spectroscopy [32,33], it has become possible to observe the Higgs mode by pump-probe measurements and THz third-harmonic generation (THG), as demonstrated first in an $s$-wave superconductor NbN [34–36]. In parallel, substantial theoretical progress has been made on the understanding of observability of Higgs mode in the nonlinear THz optical responses, and the importance of paramagnetic coupling of the light-matter interaction has been elucidated [26,37–45].

More recently, the Higgs mode has been observed in the $d$-wave cuprate superconductor Bi2212 by THz pump-optical probe (TPOP) spectroscopy in reflection geometry. Phenomenologically, the measurement scheme resorts to the THz pulse-induced Kerr effect, where the intense THz pulse modifies the optical response of the near-infrared probe pulse. The oscillatory behavior of the optical reflectivity, which follows the squared THz-pump electric field ($E$-field) was identified as a fingerprint of Higgs mode [46]. Subsequently, THG from cuprate superconductors has also been observed in other cuprate superconductors such as LSCO, YBCO and DyBa$_2$Cu$_3$O$_{7-x}$ and interpreted in terms of Higgs mode-mediated THG [43].

In this paper, we investigate the SC fluctuations in underdoped (UD) and overdoped (OD) Bi2212 thin films by the TPOP measurements. The Higgs mode oscillation observed in the pump-probe signal is shown
to emerge at temperatures above $T_c$. The onset temperature of the Higgs mode-amplitude shows a good agreement with that of the superfluid density $N_s$ estimated from the THz transmission spectroscopy for both UD and OD samples. The results are also compared with that of Bi2212 single crystals with different dopings, revealing that the SC fluctuations in Bi2212 extend up to 10-30 K above $T_c$ in a wide range of carrier doping.

II. EXPERIMENT

We performed TPOP measurements on underdoped (UD76, $T_c = 76$ K and thickness of 60 nm) and overdoped (OD67, $T_c = 67$ K and thickness of 160 nm) Bi2212 thin films grown by the sputtering method on MgO substrates. The SC transition temperature $T_c$ was determined by the magnetic susceptibility measurement using SQUID as shown in Appendix A. In Fig. 1(a) we show a schematic of the TPOP spectroscopy. The output of a regenerative amplified Ti:sapphire laser system with the pulse duration of 100 fs, central wavelength of 800 nm, pulse energy of 4 mJ and repetition rate of 1 kHz, was divided into two beams: one for the generation of the intense THz-pump pulse and the other for the optical probe pulse. To generate an intense monocycle THz pulse, we employed the tilted-pulse-front technique with a LiNbO$_3$ crystal [48] combined with the tight focusing method [49]. The THz-pump $E$-field was detected by electro-optic sampling in a 380 $\mu$m GaP (110) crystal placed inside the cryostat. The waveform and power spectrum of the THz-pump pulse are represented in Figs. 1(c) and (d), respectively. The central frequency of the THz-pump $E$-field is located around 0.6 THz = 2.4 meV, which is much smaller than the anti-nodal SC gap energy, $2\Delta_0 > 20$ meV for the present doping levels of the Bi2212 samples [50–52]. We used the peak $E$-field of 130 kV/cm for UD76 and 200 kV/cm for OD67. The TPOP measurements were performed as a function of both the pump and probe polarization angles $\theta_{\text{Pump}}, \theta_{\text{Probe}}$ as defined in Fig. 1(b) to discriminate the Higgs-mode and other contributions (for details of the methodology, see Ref. [46]). Since the oscillatory component of the reflectivity change follows the square of the THz peak $E$-field (Fig. 2(e)), the amplitude of the oscillatory component i.e. the THz Kerr signal can be expressed by the third-order nonlinear susceptibility $\chi^{(3)}$ as [46,53]

$$\frac{\Delta R}{R} (E_{i}^{\text{Probe}}, E_{j}^{\text{Probe}}) = \frac{1}{\varepsilon_0} \frac{\partial R}{\partial \varepsilon_1} \varepsilon_1 \text{Re} \chi^{(3)}_{ijkl} E_{k}^{\text{Pump}} E_{l}^{\text{Pump}},$$

where $\varepsilon_1$ is the real part of the dielectric constant and $E_i$ denotes the $i$-th component of the THz-pump or optical-probe $E$-field. Assuming the tetragonal symmetry for Bi2212, the nonlinear susceptibility can be decomposed into the irreducible representations of $D_{4h}$ point group as [46,54]
\[
\chi^{(3)}(\theta_{\text{Pump}}, \theta_{\text{Probe}}) = \frac{1}{2} (\chi_{A1g}^{(3)}(3) + \chi_{B1g}^{(3)} \cos 2\theta_{\text{Pump}} \cos 2\theta_{\text{Probe}} + \chi_{B2g}^{(3)} \sin 2\theta_{\text{Pump}} \sin 2\theta_{\text{Probe}}).
\]

(2)

Here we defined \(\chi_{A1g}^{(3)} = \chi_{xxxx}^{(3)} + \chi_{xxyy}^{(3)}\), \(\chi_{B1g}^{(3)} = \chi_{xxxx}^{(3)} - \chi_{xxyy}^{(3)}\) and \(\chi_{B2g}^{(3)} = \chi_{xyxy}^{(3)} + \chi_{xyyx}^{(3)}\). In our previous work, mean-field calculations showed that the Higgs mode response should appear only in the isotropic \(A_{1g}\) channel [46]. Figure 2(f) shows the probe polarization dependence of the reflectivity change \(\Delta R/R\) for OD67 at 15 K and demonstrates that \(\Delta R/R\) is dominated by the isotropic \(A_{1g}\) component although a slight angle-dependence is discerned, which is consistent with the results of the Bi2212 single crystals [46]. Therefore, we attribute the observed THz Kerr signal below \(T_c\) to the Higgs mode, and we focus on the \(A_{1g}\) components of \(\Delta R/R\) in the following discussion.

In addition to the TPOP measurement, THz transmission spectroscopy was performed on the same UD76 and OD67 thin films to evaluate the superfluid density, which corresponds to the phase stiffness of the SC order parameter.

III. RESULT

A. THz pump-optical probe spectroscopy

The \(A_{1g}\) component of the THz pulse-induced transient reflectivity change \(\Delta R/R\) for \(\theta_{\text{Pump}} = 45^\circ\) at various temperatures is displayed in Figs. 2(a) for UD76 and (b) for OD67 thin films, respectively. At 30 K, below \(T_c\), \(\Delta R/R\) for both films show oscillatory behaviors which follow the squared THz-pump \(E\)-field \(E_{\text{Pump}}(t)^2\). In addition to the THz Kerr component, \(\Delta R/R\) has a decaying component that survives for as long as 10 ps. For the UD76 thin films at 120 K, above \(T_c\), the signal consists of a weaker THz Kerr component and a decaying signal that switches its sign after 2 ps. At 268 K the decaying signal remains positive for all delays. For the OD67 thin films at 150 K, above \(T_c\), the signal consists of a weaker THz Kerr component and a decaying signal. The overall behaviors are similar to the results obtained in single crystals [46].

Figures 2(c) and (d) display the temperature dependence of the \(A_{1g}\) components of the transient reflectivity change \(\Delta R/R\) for UD76 and OD67, respectively. In both samples \(\Delta R/R\) displays a sharp increase above \(T_c\), indicating the possible presence of finite SC fluctuation above \(T_c\).

As the Higgs mode oscillation is expected to follow \(E_{\text{Pump}}(t)^2\), we can extract the amplitude of the THz Kerr signal from the Fast Fourier transformation (FFT) of \(\Delta R/R\). Figures 3(a) and (b) show the FFT spectrum of \(E_{\text{Pump}}(t)^2\) and the \(A_{1g}\) component of \(\Delta R/R\) for thin films at selected temperatures. The FFT of the \(A_{1g}\) component of \(\Delta R/R\) around 1.5 THz, which corresponds to the peak in the FFT of the squared THz-pump \(E\)-field, increases as the temperature is lowered. The FFT amplitudes for UD76 and OD67 integrated from 1.2 to 2.2 THz \((A_{\text{FFT}})\) are shown in Figs. 3(c) and (d), respectively. With decreasing temperature, the
integrated FFT amplitude $A_{\text{FFT}}$ sharply increases below 100 K in both samples, while it shows a more gradual increase at higher temperatures. The origin of $A_{\text{FFT}}$ at higher temperature will be discussed later in this section.

To quantitatively discuss the temperature dependence of the amplitude of the THz Kerr signal, we should take into account the temperature dependence of the squared THz-pump $E$-field inside the thin film in the analysis of third-order nonlinear susceptibility $\chi^{(3)}$ relevant to the THz Kerr signal. Here, we evaluated the squared THz-pump $E$-field inside the thin film ($B_{\text{FFT}}$) for each temperature by using the optical constants measured by THz transmission spectroscopy. The detailed calculations of $B_{\text{FFT}}$ and $\chi^{(3)}$ are explained in Appendix B. The obtained temperature dependence of $B_{\text{FFT}}$ are shown in Figs. 4(a) and (b). The third-order nonlinear susceptibility $\chi^{(3)}$ is calculated by dividing the integrated FFT amplitude $A_{\text{FFT}}$ by $B_{\text{FFT}}$. Figures 4(c) and (d) show the resulting third-order nonlinear susceptibility $\chi^{(3)}$ as a function of temperature for UD76 and OD67, respectively; it gradually increases as the temperature decreases from 200 K, and shows an upturn at about 20 K above $T_c$. As our previous work has demonstrated [46], we attribute the $A_{1g}$ component of the THz Kerr signal below $T_c$ to the Higgs mode.

In order to determine the onset temperature of the THz Kerr signal ($T_{\text{Kerr}}$) precisely, we take the second derivative of $\chi^{(3)}$ with respect to temperature. In this way we extract the onset temperature of the THz Kerr signal, $T_{\text{Kerr}} = 90$ K for UD76 and $T_{\text{Kerr}} = 80$ K for OD67. One plausible source of the finite $\chi^{(3)}$ response above $T_c$ is SC fluctuations, where the phase coherence among the inhomogeneous Cooper pair islands is established on a pico-second (THz) timescale as probed by the THz and microwave spectroscopy [2–8]. Another candidate is the preformed Cooper pairs without mutual phase coherence as reported by ARPES [15–18] and infrared spectroscopy [55].

In OD67 thin film, the oscillatory behavior appears even at 150 K, which is much higher than the onset temperature for the preformed Cooper pairs as reported by ARPES in similarly doped Bi2212 sample [17]. This excludes the possibility that this weakly temperature dependent signal originates from the preformed Cooper pairs in the OD sample. We note that there is no symmetry argument which forbids a finite third-order susceptibility like the observed THz Kerr signal, even in a normal metal [56–62]. Still the exact origin of this finite THz Kerr signal needs further experimental and theoretical study. While it is outside the scope of this paper to identify the origin of the finite $\chi^{(3)}$ response at high temperatures, the sharp onset of $\chi^{(3)}$ at $T_{\text{Kerr}}$ indicates its relevance to the SC order parameter. Focusing on the origin of the onset of the THz Kerr signal just above $T_c$, we now compare $\chi^{(3)}$ with the superfluid density estimated by the THz transmission spectroscopy.
B. THz transmission spectroscopy

In order to compare the onset of the THz Kerr signal and the SC phase stiffness, we evaluate the superfluid density by the time-domain THz transmission spectroscopy. Figures 5(a)-(d) show the real and imaginary parts of the THz optical conductivity for UD76 and OD67 samples. Below \( T_c \), the imaginary part of the optical conductivity for both thin films exhibit a divergent-like behavior at low frequency, indicating the growth of the superfluid density. The real and imaginary parts of the optical conductivity are reasonably fitted simultaneously by the two-fluid model, which is given by [63]

\[
s_1(\omega) = \frac{\omega_p^2 \tau}{1 + \omega^2 \tau^2} + N_s \delta(\omega),
\]

\[
s_2(\omega) = \frac{\omega_p^2 \tau^2 \omega}{1 + \omega^2 \tau^2} + \frac{N_s}{\omega}.
\]

(3)

Here, the first term is the Drude component and the second term is the SC component, where \( \omega_p \) is the plasma frequency, \( \tau \) is the scattering time and \( N_s \) is the superfluid density. The previous THz optical conductivity measurements and theories showed that the inhomogeneity transfers the spectral weight from the Delta function to the Drude component [64]. While two Drude components were assumed to describe the optical conductivity in Ref. [64], here only one Drude component is assumed to reduce the number of the fitting parameters. The fitting results are shown in Figs. 5(a)-(d) by solid curves and well reproduce the complex optical conductivity of both thin films. The temperature dependence of the three parameters \( N_s \), \( \omega_p \) and \( \tau \) are plotted in Figs. 5(e)-(h). The superfluid density \( N_s \) shown in Figs. 5(e) and (g) sharply increases from above \( T_c \) for both films; \( T_{Ns} = 90 \text{ K} \) for UD76 and \( T_{Ns} = 80 \text{ K} \) for OD67. For UD76 the onset temperature of \( N_s \) is consistent with that reported by the previous THz optical conductivity measurement [2]. The scattering time \( \tau \) of the Drude component plotted in Figs. 5(f) and (h) gradually increases with decreasing temperature, whose behavior is also consistent with the previous report [64]. While we set the plasma frequency \( \omega_p \) as a fitting parameter, it does not show significant temperature dependence for either thin film.

IV. DISCUSSION

In both Bi2212 thin films, \( T_{Kerr} \) coincides with \( T_{Ns} \) within our experimental error bars. This result indicates that the THz Kerr signal above \( T_c \) can be attributed to the Higgs mode from a dynamically fluctuating SC phase on the THz time scale.

We now examine \( T_{Kerr} \) of the Bi2212 single crystals in a wide range of dopings from our previous TPOP results [46]. Since the temperature dependence of the THz \( E \)-field inside the sample does not strongly depend on hole concentration as shown in Figs. 4(a) and (b), we evaluated the temperature dependence of
the third-order nonlinear susceptibility \( \chi^{(3)} \) for single crystals by approximating the temperature dependences of the internal THz \( E \)-field with that of the UD76 thin film (Appendix C). Figure 6 summarizes \( T_{\text{Kerr}} \) for two thin films and five differently doped single crystals. For all samples, the onset temperature of the Higgs mode is located at 10-30 K above \( T_c \). For UD single crystals, \( T_{\text{Kerr}} \) coincides with the onset temperature determined by the previous THz conductivity measurements in Bi2212 thin films [2,3], while it is much lower than the onset temperature reported by the torque magnetometry, Nernst effect and ARPES [9,11,14,17]. Moreover, toward underdoping, \( T_{\text{Kerr}} \) does not show a significant increase, in sharp contrast with the results of ARPES that reported the increase of the onset temperature of the SC gap opening toward UD region.

While our results do not match with the results of the Nernst effect in Bi2212, we note that a recent Nernst measurement claims that the onset temperature is very close to that obtained from the THz spectroscopy in the case of LSCO [65]. In other cuprates such as LSCO and YBCO, the microwave and THz spectroscopy report that the onset temperature of the SC fluctuation is at most 20 K above \( T_c \) in a wide range of dopings [4,5,8]. These tendencies are consistent with our evaluation that the onset of SC phase coherence in Bi2212 is located 10-30K above \( T_c \).

Finally, we address the difference between the onset temperature reported by the ARPES and that of THz spectroscopy. In ARPES measurements, the SC gap opening temperature \( T_{\text{Gap}} \) for Bi2212 is reported to be 120-150 K in the UD region which is substantially higher than \( T_{\text{Kerr}} \) [16–18], indicating the presence of the preformed Cooper pairs above the temperature where the SC phase coherence vanishes. One possible scenario for the difference between \( T_{\text{Gap}} \), \( T_{\text{N_s}} \) and \( T_{\text{Kerr}} \) is that the Cooper pairs start to form locally at \( T_{\text{Gap}} \) while their mutual phase coherence is not established. At \( T_{\text{N_s}} \) and \( T_{\text{Kerr}} \), the phase coherence among the inhomogeneous Cooper pair islands appears, though dynamically fluctuating in a ps (THz) regime, and at \( T_c \), all the Cooper pair islands acquire the static macroscopic phase coherence. This scenario is supported by recent nonlinear conductivity, paraconductivity and torque magnetometry measurements. They provided evidences that the SC phase coherence vanishes rapidly above \( T_c \) in an exponential fashion and the SC phase locking mechanism among the Cooper pair islands was explained by a phenomenological percolation model [12,66,67].

V. CONCLUSION

We have observed the THz Kerr effect in UD and OD Bi2212 thin films using the TPOP spectroscopy. The onset temperature of the THz Kerr signal coincides with that of the superfluid density evaluated by the THz transmission spectroscopy, indicating that the THz Kerr signal above \( T_c \) originates from the Higgs
mode. The comparison with differently doped single crystals reveals that the onset of the SC coherence on a pico-second time scale is located 10-30 K above $T_c$ in a wide range of doping.

We demonstrated that the Higgs mode can probe the dynamics of the SC order parameter sensitively in a pico-second time scale. Consequently, probing of the Higgs mode would lay the foundation to investigate the nonequilibrium phenomena, e.g. for the light-induced superconductivity [68–72]. Moreover, the Higgs mode would be a new probe for the study of dynamical interplay between the SC and other competing or coexisting orders in strongly correlated electron systems.

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**Appendix A: Determination of $T_c$**

Figures 7(a) and (b) show the magnetic moments for UD76 and OD67 Bi2212 thin films measured by SQUID under zero-field cooling (ZFC) and field-cooling (FC). The SC transition temperature $T_c$ is determined by the onset of drop in the magnetic moment. For the Bi2212 single crystals in our previous work [46], $T_c$ is determined by the same manner.

**Appendix B: Calculation of the third-order nonlinear susceptibility**

We evaluated the third-order nonlinear susceptibility by calculating the THz-pump $E$-field inside the thin film as follows. The FFT of the $A_{1g}$ component of $\Delta R/R(t)$ can be described as

$$
\frac{\Delta R_{A_{1g}}}{R} (\omega) = \frac{1}{R \partial \varepsilon_1} \varepsilon_0 \int_0^\infty dt \operatorname{Re} \chi^{(3)}_{A_{1g}} E_{\text{Film}}(t)^2 e^{-i\omega t} = \frac{1}{R \partial \varepsilon_1} \varepsilon_0 \operatorname{Re} \chi^{(3)}_{A_{1g}} B(\omega),
$$

(B1)

where $E_{\text{Film}}(t)$ is the $E$-field inside the thin film in the time-domain and $B(\omega)$ is the FFT of $E_{\text{Film}}(t)^2$. As discussed in our previous work, the third-order nonlinear susceptibility of the Higgs mode does not depend on frequency when the probe photon energy is much higher than the SC gap energy and pump photon energy [46]. Therefore, $\operatorname{Re} \chi^{(3)}_{A_{1g}}$ in Eq. (B1) can be singled out from the integral.

The THz-pump $E$-field inside the film in the time domain $E_{\text{Film}}(t)$ can be calculated by using its FFT $E_{\text{Film}}(\omega)$, which is expressed as [73]
\[
\frac{E_{\text{Film}}(\omega)}{E_{\text{Pump}}(\omega)} = \frac{2}{1 + n_{\text{Film}}(\omega)} e^{i(n_{\text{Sub}}(\omega) - 1)\omega d/c} \left(1 - \frac{n_{\text{Film}}(\omega) - 1}{n_{\text{Film}}(\omega) + 1} e^{2i n_{\text{Sub}}(\omega) \omega d/c} n_{\text{Film}}(\omega) - n_{\text{Sub}}(\omega)\right),
\]

where \(E_{\text{Pump}}(\omega)\) is the incident THz-pump \(E\)-field, \(d\) is the thickness of the thin film, and \(n_{\text{Film}}(\omega)\) and \(n_{\text{Sub}}(\omega)\) are the complex refractive index of the thin film and the substrate, respectively. As the THz-pump FFT intensity \(E_{\text{Pump}}(\omega)^2\) covers from 0.1 to 3 THz (Fig. 1(d)), we can calculate \(E_{\text{Film}}(\omega)\) by using \(n_{\text{Film}}(\omega)\) and \(n_{\text{Sub}}(\omega)\) in the same frequency region. Because the sample size is \(3 \times 3\) mm and not large enough to measure the optical conductivity below 0.4 THz, we used \(n_{\text{Film}}(\omega)\) obtained from the fitting to the complex optical conductivity \(\sigma(\omega)\) with Eq. (3).

By integrating Eq. (B1), the nonlinear susceptibility can be written as

\[
\text{Re} \chi_{A_1g}^{(3)} = \frac{\int_{2\pi*1.2}^{2\pi*2.2} d\omega \frac{\Delta R_{A_1g}}{R}(\omega)}{\int \frac{\Delta R}{R} e^{i\omega t} \int_{2\pi*1.2}^{2\pi*2.2} d\omega B(\omega)} = \frac{A_{\text{FFT}}}{B_{\text{FFT}}},
\]

where \(A_{\text{FFT}}\) is the integrated amplitudes of \(\Delta R/R(\omega)\) from \(\omega/2\pi = 1.2\) to 2.2 THz and shown in Figs. 3(c) and (d). \(B_{\text{FFT}}\) is the integrated amplitudes of \(B(\omega)\) from \(\omega/2\pi = 1.2\) to 2.2 THz and is shown in Figs. 4(a) and (b). Finally, the calculated \(\text{Re} \chi_{A_1g}^{(3)} = \chi^{(3)}\) is plotted as a function of temperature in Figs. 4(c) and (d).

Appendix C: Temperature dependence of the normalized THz Kerr signal for Bi2212 single crystals

Temperature dependence of the third-order nonlinear susceptibility of the THz Kerr signal \(\chi^{(3)}\) for Bi2212 single crystals are summarized in Fig. 8. In all of the single crystals, \(\chi^{(3)}\) increases from 10-20 K above \(T_c\). The onset temperature of the Higgs mode is defined as the temperature where the second derivative of \(\chi^{(3)}\) with respect to temperature exhibits a significant deviation from the smooth normal state signal. In all of the single crystals, \(T_{\text{Kerr}}\) is located 10-30 K above \(T_c\) as summarized in the phase diagram (Fig. 6).
FIG. 1. (a) A geometry for the THz pump-optical probe spectroscopy. The THz-pump pulse and optical-probe pulse propagate collinearly. (b) A schematic illustration of the CuO$_2$ plane. The pump ($\theta_{\text{Pump}}$) and probe ($\theta_{\text{Probe}}$) polarization angles are defined relative to the Cu-O bond ($y$-axis). (c) The waveform of the THz-pump $E$-field. (d) The power spectrum of the THz-pump $E$-field.
FIG. 2. (a), (b) The $A_{1g}$ components of the THz-pump-induced reflectivity change $\Delta R/R$, at selected temperatures for UD76 and OD67, respectively. The upper panels show the waveforms of the squared THz-pump $E$-field. (c), (d) Temperature dependence of the $A_{1g}$ components of the reflectivity change $\Delta R/R$, for UD76 and OD67, respectively. (e) Probe polarization dependence of $\Delta R/R$ at delay time of 1.3 ps for OD67 when the pump polarization is fixed at $\theta_{\text{pump}} = 45^\circ$. (f) The FFT amplitude of $\Delta R/R$ at 15 K integrated from 1.2 to 2.2 THz at $\theta_{\text{pump}} = \theta_{\text{probe}} = 45^\circ$ as a function of the peak THz-pump $E$-field for OD67.
FIG. 3. (a), (b) The FFT amplitude of the $A_{1g}$ component of $\Delta R/R$ at selected temperatures for UD76 and OD67, respectively. The gray curve is the FFT of the squared THz-pump $E$-field ($E_{\text{Pump}}^2$). (c), (d) Temperature dependence of the FFT amplitude of $\Delta R/R$ integrated from 1.2 to 2.2 THz ($A_{\text{FFT}}$) for UD76 and OD67, respectively.
FIG. 4. (a), (b) Temperature dependence of the FFT amplitude of the squared THz-pump $E$-field inside the thin film ($B_{\text{FFT}}$) for UD76 and OD67, respectively. The FFT amplitude is normalized by its value at $T_c$. (c), (d) Temperature dependence of the third-order nonlinear susceptibility of the THz Kerr signal $\chi^{(3)}$ in TPOP measurements (the red curves). The green curves show the second derivative of $\chi^{(3)}$ with respect to temperature. The red vertical bars denote the determined onset temperature of the THz Kerr signals, $T_{\text{Kerr}}$. 
FIG. 5. (a)–(d) Real and imaginary parts of the optical conductivity measured by the THz transmission spectroscopy for UD76 and OD67. The open circles represent the data and the solid lines are the fitting curves by the two-fluid model. In (b) and (d), the SC component (blue) and Drude component (red) at 5 K for UD76 and at 15 K for OD67 are plotted by the dashed curves, respectively. (e)–(h) Temperature dependence of the fitting parameters in Eq. (3) for UD76 and OD67, respectively. (e) and (g) show the temperature dependence of the superfluid density $N_s$. (f) and (h) show the temperature dependence of the plasma frequency $\omega_p$ (left axis) and the scattering time $\tau$ (right axis). Here, $N_s$ and $\omega_p$ are normalized by their value at the lowest temperature. The blue vertical bars denote the determined onset temperature of $N_s$ ($T_N$).
FIG. 6. Doping dependence of the onset temperature of the THz Kerr signal obtained from TPOP experiments and superfluid density evaluated by THz transmission measurements. The hole concentration is determined from $T_c$ using the Presland and Tallon’s equation [74]. The red circles are $T_{Kerr}$ for Bi2212 single crystals that is evaluated from the data in Ref. [42] and the red squares are $T_{Kerr}$ for Bi2212 thin films (this work). The orange diamonds are $T_{Ns}$ for Bi2212 thin films (this work). The data of $T_{Ns}$ for other dopings are adopted from Ref. [3]. The blue triangles are the SC gap opening temperature ($T_{Gap}$) for Bi2212 adopted from Ref. [17,19]. The purple diamonds are the pseudogap opening temperature $T^*$ from Ref. [50].
FIG. 7. (a), (b) The magnetic moment of the UD76 and OD67 Bi2212 thin films, respectively. The black dotted lines denote the determined $T_c$. 
FIG. 8. Temperature dependence of the third-order nonlinear susceptibility of the THz Kerr signal $\chi^{(3)}$ in TPOP measurements for Bi2212 single crystals (the red curves). The green curves show the second derivative of $\chi^{(3)}$ with respect to temperature. The red vertical bars denote the determined onset temperature of the THz Kerr signal $T_{Kerr}$. 
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