論文の内容の要旨

Terahertz nonlinear optical responses in high-temperature cuprate superconductors

(銅酸化物高温超伝導体における テラヘルツ非線形光学応答の研究)

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Ever since the discovery of the high-temperature cuprate superconductors, tremendous researches have been performed to uncover the mechanism of superconductivity. Nevertheless, their unusual physical properties are long-standing mysteries in condensed matter physics.

One of the intriguing phenomena in cuprate superconductors is the superconducting fluctuations. To understand how the Cooper pairs emerge from the complex metallic state, various experiments have been performed to elucidate the onset temperature of the superconducting fluctuations. Nevertheless, the reported onset temperatures depend on experimental techniques, and a unified understanding for the onset of superconducting fluctuations is still lacking. Thus, a novel approach that can sensitively probe the superconducting order parameter has been desired.

Another remarkable phenomenon is the photo-induced superconductivity, recently reported in various types of cuprate superconductors. It has been reported that a Josephson plasma resonance (JPR)-like response emerges in the *c*-axis reflectivity and concomitantly the imaginary part of the optical conductivity $\sigma_2(\omega)$ exhibits the $1/\omega$-like response after the irradiation of an intense laser pulse even at temperatures far above the superconducting critical temperature (T_c), and interpreted as photo-induced superconductivity. However, there remains an ambiguity that in principle, one cannot distinguish the $1/\omega$-response of the superconductivity and the Drude response of the quasiparticle (QP) excitation with a low scattering rate in the measured terahertz (THz) frequency range. Since the photo-induced $1/\omega$-like response in the $\sigma_2(\omega)$ spectrum disappears in a few pico-seconds, it cannot be investigated by resistivity or magnetic susceptibility measurements. Therefore, an

alternative ultrafast probe of the superconducting order parameter has been required.

In this study, we have investigated the superconducting fluctuations and the photo-induced nonequilibrium superconductivity via the THz nonlinear optical responses caused by the collective excitation of the superconducting order parameter in high-T_c cuprate superconductors: the Higgs mode and JPR, which directly manifest the development of the superconducting order parameter within a pico-second time resolution.

First, we have investigated the Higgs-mode response through the THz Kerr effect in $Bi_2Sr_2CaCu_2O_{8+x}$ (BSCCO) thin films utilizing the THz pump-optical probe spectroscopy. In the THz Kerr signal, two onset temperatures are identified. Combining the results of single-crystalline samples, we have found that the lower one (T_1^{ons}) is slightly above T_c, whereas the higher one (T_2^{ons}) is located substantially higher than T_c. T_1^{ons} coincides with that of the superfluid density evaluated from the THz optical conductivity measurements, indicating the the superconducting phase fluctuation on the picosecond time scale evolves from slightly above T_c, while the static superconducting phase coherence develops below T_c. On the other hand, the coincidence between T_2^{ons} and the superconducting gap opening temperature in the previous studies for BSCCO suggests that T_2^{ons} is associated with the preformed Cooper pairs.

Next, we have applied the THz nonlinear optical responses to elucidate the photo-induced nonequilibrium state in cuprate superconductors. To this aim, we have started from the optical pump-THz probe spectroscopy for an underdoped $YBa_2Cu_3O_{6+y}$ (YBCO). We have indeed observed the photo-induced $1/\omega$-like increase in the imaginary part of the c-axis optical conductivity above T_c, consistent with the previous studies. However, the a-axis THz reflectivity is shown to decrease upon the photo-excitation, which is against the interpretation of photo-induced superconductivity.

The observed prompt optical conductivity in YBCO is further examined by the Higgs-mode response by THz pump-optical probe spectroscopy. We have identified the THz-pulse driven Higgs mode and the superconducting QP excitation below T_c in the THz-pump induced optical reflectivity change $\Delta R/R$. When the sample is irradiated with near-infrared (NIR) pump pulse below T_c, the Higgs-mode and QP responses are suppressed, which agrees with the photo-induced destruction of the superconductivity. Above T_c, neither the Higgs mode nor the QP responses are observed in $\Delta R/R$, indicating that it is unlikely to attribute the photo-induced state above T_c to the superconducting phase.

To further examine the photo-induced state above T_c, we have studied the THz third-harmonic generation (THG) caused by the nonlinear c-axis Josephson current. Using a narrowband THz-pulse, we have observed the THG in the reflected THz electric field from YBCO below T_c. Besides, we have performed the NIR pump-THG probe experiments and shown that the THG intensity below T_c decreases after photo-excitation, consistent with the results of the Higgs mode-response. At 100 K, the THG is not identified either in equilibrium or in the photo-induced state.

Therefore, we have concluded that the optically-induced increase in the imaginary part of the c-axis optical conductivity is attributed to the QP excitation but not to the photo-induced superconductivity. Since the temperature range where the characteristic c-axis transient optical conductivity appears coincides with the pseudogap temperature region, the observed nonequilibrium response is most likely attributed to the QP excitation correlated with the pseudogap phase. Even though the photo-induced state is not ascribed to the superconductivity, it is highly intriguing that coherent QPs with the scattering rate as low as a few THz appear under the photo-excitation in the pseudogap phase, considering the incoherent c-axis transport in equilibrium. This result would provide an important clue to understanding the pseudogap phase.

Notably, the THz nonlinear responses demonstrated here would also provide access to the study of the dynamical interplay between the superconductivity and other competing or coexisting orders in unconventional superconductors (beyond the BCS framework) though the observation of the collective modes arising from those orders in the time domain. Furthermore, being an ultrafast probe of the superconducting order parameter, these THz nonlinear responses would lay the foundation to explore the nonequilibrium superconductivity in a wide variety of unconventional superconductors.