論文の内容の要旨

論文題目：Nonlinear terahertz spectroscopy of multiband superconductors

(多バンド超伝導体の非線形テラヘルツ分光)

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The spontaneous symmetry breaking is one of the central concepts in physics. Generally, with the continuous symmetry breaking, two collective modes of an order parameter appear: the massless phase mode called Nambu-Goldstone mode and the massive amplitude mode called Higgs mode. In charged particle systems the phase mode becomes massive and its energy is elevated to higher energy region due to the coupling with the gauge field. This process is called as the Anderson-Higgs mechanism (A-H) mechanism. Superconductor is a typical example that shows the A-H mechanism associated with the $U(1)$ symmetry of the phase of wavefunction.

The Higgs mode in superconductors has long been studied theoretically over decades but its observation has been difficult, because it does not couple directly to the electromagnetic field in the linear response regime. Recently owing to the progress of the intense terahertz (THz) pulse generation technique, it has become possible to unveil the properties of Higgs mode in superconductors.

A remarkable example is the Higgs mode resonance which was found through the observation of third harmonic generation (THG) in an *s*-wave superconductor NbN in the THz frequency range.

So far, the study of collective modes in superconductors through the THz nonlinear response has been limited to the single band superconductor where only one order parameter exists. When there exist two order parameters, two Higgs mode and one massive phase mode called Leggett mode is expected to exist as a low energy collective excitation in charged particle system. The Leggett mode corresponds to the oscillation of the relative phase in which the mass of the mode depends on strength of the interband interaction. The Higgs modes should also interact with each other through the interband coupling. Accordingly, the observation of Higgs modes is expected to provide deeper insights into the multiband superconductors.

While the Leggett mode has been observed in a typical multiband superconductor MgB_2, there is no experimental observation of Higgs mode in multiband superconductors, and the interband coupling effect on the Higgs mode remains unclear. Moreover it has been pointed theoretically that in addition to the Higgs mode, the single particle excitation also contributes to THG, called as change density fluctuation (CDF). It was demonstrated that the contribution of CDF to THG is dominant in clean limit superconductors within the BCS mean field approximation, while it is now revealed that Higgs contribution dominates the THG in the dirty limit superconductor, which was the case of NbN. Then it is also an interesting problem as to whether the Higgs modes dominate THG, or CDF does in other superconductors.

In order to elucidate the lower-energy collective excitations in multiband superconductors, we investigate the nonlinear response of the MgB_2 thin film on MgO substrate, and $FeSe_{0.5}Te_{0.5}$ thin film on CaF_2. Both of which are known as multi band superconductors, while MgB_2 is a relatively weak interband coupling system and $FeSe_{0.5}Te_{0.5}$ is in the opposite strong interband coupling system.

For MgB_2 we first performed the transmission measurement with intense narrow-band THz pulses. We observed a clear third-harmonic generation (THG) associated with superconductivity. Figure 1 (a) indicates the power spectra of the transmitted THz wave, with the center frequency 0.6 THz (2.5 meV) and the peak electric field 7 kV/cm, above (35 K) and below (27 K) the superconducting critical temperature T_c=34 K. The inset shows the electric field (*E*-field) strength dependence of the integrated spectral weight around 1.8 THz. The data are proportional to six power of the *E*-field, which indicates the peak originates from the THG induced by third-order nonlinear process.

We measured the temperature dependence of the THG intensities and extracted the magnitude square of the third-order susceptibility $|\chi^{(3)}|^2$. Figure 1 (b) indicates the calculated temperature dependence of the superconducting gap 2Δ; the black and gray solid lines indicate the temperature dependence of larger gap ($2\Delta_\sigma$) and the smaller gap ($2\Delta_\pi$). The orange, green and blue dashed line show the twice of the incident frequency 2ω for each incident frequency. Figure 1 (b)-(e) plot $|\chi^{(3)}|^2$ normalized by its maximum value. Based on the two band BCS theory, all the CDFs Higgs mode, and Leggett mode are expected to contribute to THG. The resonance condition is given by $2\omega = 2\Delta$ for CDFs and Higgs mode and $2\omega = \omega_L$ for Leggett mode where ω_L is the energy of Leggett mode located between $2\Delta_\pi$ and $2\Delta_\sigma$ in the case of MgB_2. The temperature of $|\chi^{(3)}|^2$ clearly exhibits a resonant peak at $2\omega = 2\Delta_\pi$, which indicates that the THG originates from CDF or Higgs mode associated with the π band. We then considered the band structure of MgB_2 and found that the existence of the Dirac point in π band divergently enhances the THG, whereas this enhancement does not occur for the Higgs mode. Therefore we conclude that the origin of THG is most likely dominated by the CDF of π band.

Next we performed the transient optical conductivity measurement by using THz pump-THz probe spectroscopy with the center pump photon energy ω = 0.8 THz (=3.3 meV) to confirm the origin of THG. We observed a clear spectral weight oscillation of both real and imaginary part of optical conductivity $\sigma_1(\omega)$ and $\sigma_2(\omega)$ with the frequency twice of the incident center frequency 2ω. From this measurement we identified the spectral weight oscillation of σ band, which is plausibly attributed to the σ-band Higgs mode. At low temperature we observed a prominent spectral weight oscillation of π band, which is attributed to CDF.

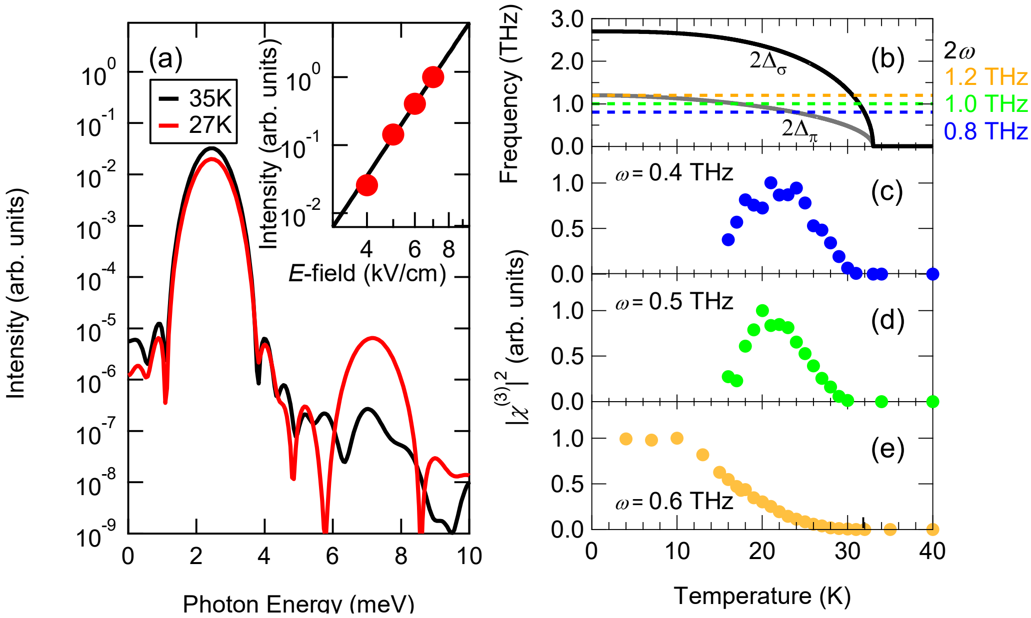

Figure 1 (a) Power spectra of the transmitted THz pulses below and above T_c = 34K with the center frequency of ω = 0.6THz (= 2.5meV). The inset shows the THG intensity as a function of the strength of the incident THz *E*-field. (b) Calculate temperature dependence of $2\Delta_\pi$ (gray line), $2\Delta_\sigma$(black line) and twice of incident frequency 0.8, 1.0, and 1.2 THz (blue, green, and orange dashed lines), respectively. (c)-(e) Temperature dependence of $|\chi^{(3)}|^2$ for the incident frequency of 0.4, 0.5,and 0.6 THz respectively.

For $FeSe_{0.5}Te_{0.5}$ we also performed the THz pump-THz probe spectroscopy with the center photon energy ω = 2.5 meV. We observed a spectral weight oscillation of $\sigma_1(\omega)$ and $\sigma_2(\omega)$ with the frequency twice of the incident center frequency 2ω at 4 K. The pump photon energy ω is between the smaller gap $2\Delta_\Gamma$ and larger gap $2\Delta_M$. Figure 2 (a) and (b) shows the pump THz induced change of the real and imaginary part of the optical conductivity $\delta\sigma_1(\omega)$ and $\delta\sigma_2(\omega)$, respectively. The oscillations are clearly identified.

We evaluate the oscillation of the superfluid density to $\delta\sigma_2(\omega)/\sigma_2^{eq}(\omega)$ where $\sigma_2^{eq}(\omega)$ is the σ_2 in the equilibrium state. Figure 2 (c) shows the photon energy dependence of the oscillatory component of $\delta\sigma_2(\omega)/\sigma_2^{eq}(\omega)$. The oscillation in the low energy region below 2 meV is attributed to that of $2\Delta_\Gamma$, while the oscillation above 6 meV is attributed to that of $2\Delta_M$. This result indicate that the strong interband coupling plays a key role, since in spite of the strong THz excitation with the photon energy larger than $2\Delta_\Gamma$ the smaller gap coherently oscillates without suppressing its magnitude. We also found a π-phase shift of the oscillation between low energy region and high energy region as shown by Fig. 2 (d) which shows the typical dynamics of oscillatory part of $\delta\sigma_2(\omega)/\sigma_2^{eq}(\omega)$ at 1.9 and 8,6 meV. Namely the 2ω-forced oscillation of Higgs mode is induced in the larger gap, and then it induces the oscillation of the smaller gap through the strong interband coupling. To interpret the π-phase shift, we considered the two components Ginzburg-Landau (GL) theory, and obtain the result that qualitatively explain the experimental results in strong coupling limit.

In summary we have investigated the Higgs modes in two-band superconductors in two limiting cases: a relatively weak interband coupling system of MgB_2 and the strong interband coupling system of $FeSe_{0.5}Te_{0.5}$. We revealed the hybridized motion of two Higgs mode in $FeSe_{0.5}Te_{0.5}$ which is qualitatively described by GL theory. The result also suggests that two components can reasonably describes the order parameter dynamics of $FeSe_{0.5}Te_{0.5}$. In MgB_2 on the contrary to the initial expectation THG resonance at π-band is observed due to the existence of Dirac cone in the π-band. The result indicates that the consideration of the band structure is crucial for THG from CDF.

The elucidation of the energy and life time of Higgs modes are important remaining subject. Since they are modulated by the interband interaction, the information of signs or magnitude of it may be resolved by measuring them and comparing microscopic theory. Such a Higgs mode spectroscopy would be promising to gain the insight into interband interaction and order parameter symmetries.

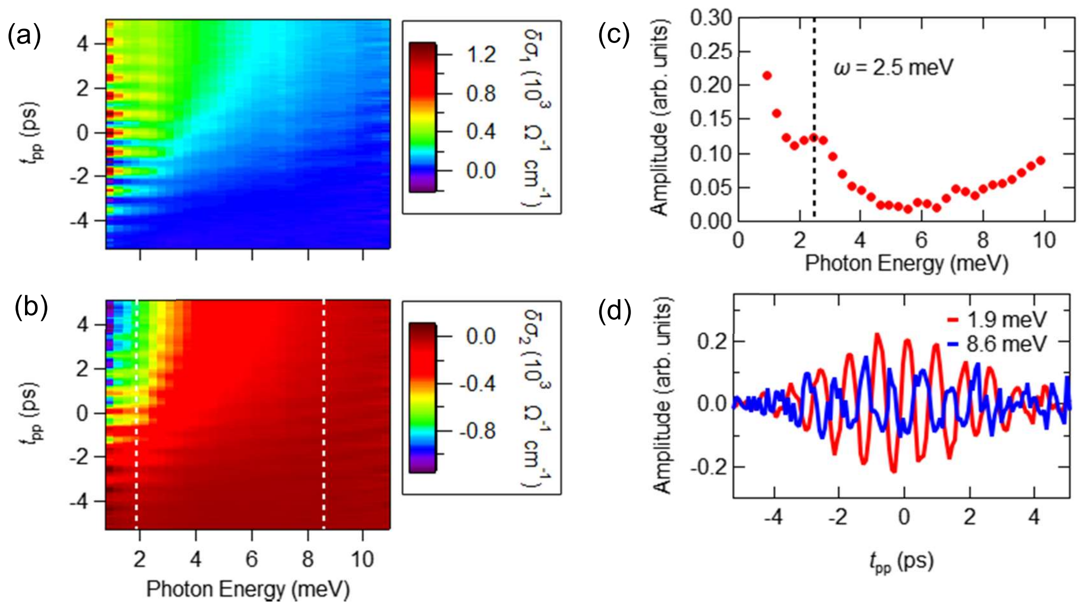

Figure 2 The pump induced change of (a) the real- and (b) imaginary-part optical conductivity spectra. (c) The amplitude of the oscillatory part of $\delta\sigma_2/\sigma_2^{eq}$ as a function of photon energy. (d) The oscillatory components of $\delta\sigma_2/\sigma_2^{eq}$ at 1.9 and 8.6meV as a function of the pump-probe delay time.