Higgs Mode and Terahertz Nonlinear Optics in Superconductors

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Abstract: By using intense terahertz pulses, we investigated the ultrafast coherent light-matter interaction of s-wave superconductors. We observed the Higgs amplitude mode and demonstrated the coherent terahertz nonlinear transient phenomena in superconductors.
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1. Introduction

With the progresses of ultrafast laser spectroscopy technique, it has become possible to study the non-equilibrium dynamics of quantum systems, i.e., atoms, molecules, and condensed matter systems, in a wide range of spectrum with ultrafast temporal resolution. A notable example is the terahertz (THz) technology, which has accelerated the understanding of the low energy electrodynamics in various materials. With the advance of intense THz light sources, nonlinear terahertz optics is currently a nascent field. Since the photon energy of the THz radiation is in the order of 1~10meV, the intense THz pulse can be thought of as a promising light source to investigate the coherent nonlinear transients in various condensed matter systems without exciting phonons which often prevent the emergence of coherent phenomena. In this presentation, we report our recent observation of ultrafast non-equilibrium dynamics of conventional s-wave superconductors induced by intense THz pulse excitation, in particular the Higgs amplitude mode of superconducting order parameter[1].

2. Experiments

Superconductivity is a striking example of the spontaneous symmetry breaking (SSB) phenomena. In general, when a phase transition occurs associated with theSSB, two kinds of collective excitations emerge; the gapless phase mode and the gapped amplitude mode of the complex order parameter, as schematically shown in Fig. 1(a). The latter is also called as the Higgs mode from its analogy to the Higgs bosons in elementary particle physics. The nature of the Higgs amplitude mode in superconductors has been intensively studied theoretically [2-5]. With the framework of the Anderson's pseudo-spin representation (Fig.1(b),(c)), the equation of motion for electrons in superconductor can be effectively described by the Bloch equation of pseudo-spin. Accordingly, it has been shown that the Higgs amplitude mode corresponds to the collective Rabi oscillation of the pseudo-spin [4]. However, the experimental observation of the Higgs mode in superconductors has remained elusive, since the Higgs mode does not couple directly to the electromagnetic field. We overcome this difficulty by adopting the THz pump and THz probe (TPTP) spectroscopy [6], as schematically shown in Fig.2(a). Since the order parameter of superconductor is related to the quasi-particles (QPs) distribution in a self-consistent manner, it can be modulated by coherent excitation of QPs with the THz pump pulse. The Higgs mode oscillation followed by such an intense THz pulse excitation is then monitored by the transmitted THz probe pulse whose photon energy is tuned around the superconducting gap energy.

The sample was the $Nb_{0.8}Ti_{0.2}N$ thin film grown on a fused quartz substrate with the BCS gap energy of 2Δ=0.72 THz (~3.0 meV) at 4 K. The duration of the THz pump pulse is τ_{pump}~1.5 ps, which is shorter than the inverse of the BCS gap energy $\tau_\Delta=\Delta^{-1}$~2.8 ps. The details of our TPTP spectroscopy technique was provided in Ref. [6]. Figure 2(b) shows the temporal evolution of the change of the transmitted THz electric field of the probe pulse at a fixed time point, δE_{probe}, induced by the THz pump pulse as a function of the pump-probe delay time t_{pp}. After the pump THz pulse irradiation, $\delta E_{probe}(t_{pp})$ increases, indicating the reduction of the BCS gap energy. At t_{pp}>2 ps, a damped oscillation is clearly observed. The oscillation frequency obtained from the damped-oscillation fits (solid lines) coincides with the value of asymptotic BCS gap energy, Δ_∞, after the THz excitation, which behavior indicates the character of Higgs amplitude mode.

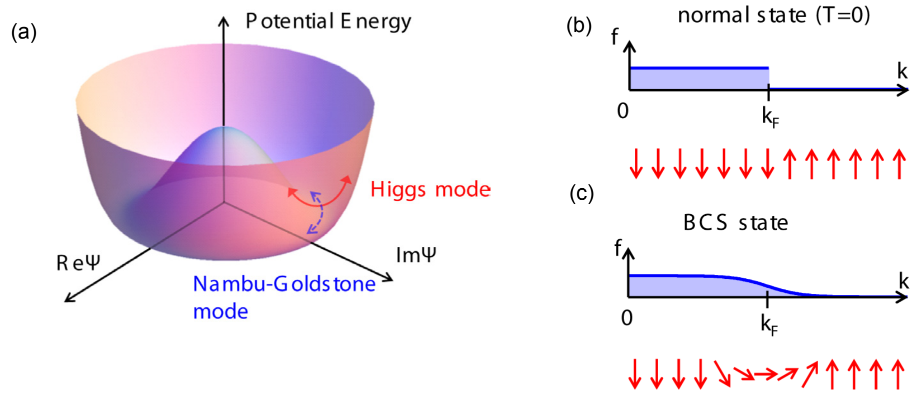

Fig. 1. (a) A schematic picture of the phase mode (blue arrow) and the Higgs amplitude mode (red arrow) in terms of free energy. (b),(c) A schematic picture of normal state and superconducting state represented by the Anderson's pseudo-spins. k_F is the Fermi wave number.

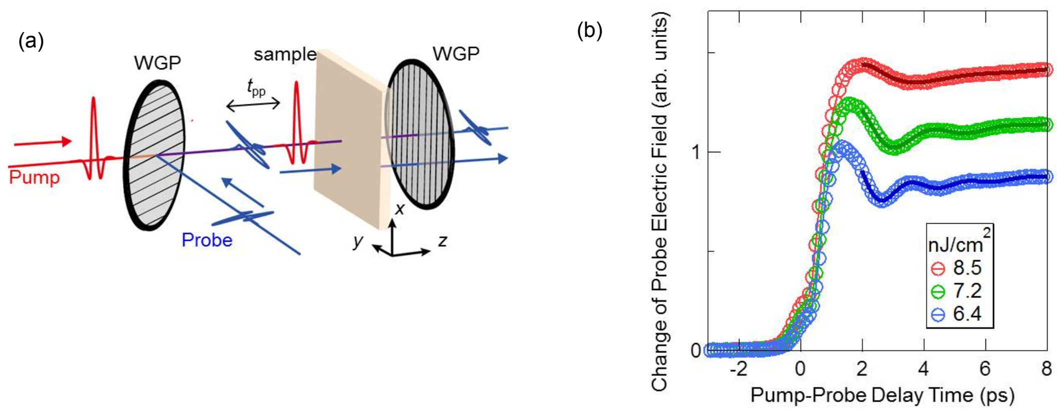

Fig. 2. (a) A schematic picture of the THz pump-THz probe spectroscopy. (b) Experimentally observed temporal evolution of the change of the probe THz electric field at 4 K at the indicated pump intensities. The damped oscillation fits are shown by solid lines.

3. Conclusion

By using the TPTP spectroscopy, we observed for the first time the collective Higgs amplitude mode in a s-wave superconductor. The non-adiabatic excitation condition necessary for the appearance of the Higgs mode is realized by using the monocycle-like intense THz pulse. In the presentation, the nonlinear responses of pseudo-spins arising from the coherent interaction with the THz electric field will also be discussed. Our observation of the Higgs amplitude mode in conventional BCS superconductors opens a new pathway for optical coherent control of the macroscopic quantum states in an ultrafast (~ ps) time scale.

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