

Observation of Higgs Modes in Superconductors by Non-Equilibrium Anti-Stokes Raman Scattering

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Spontaneous symmetry breaking in the Mexican-Hat potential leads to massless phase modes as low-energy excitations. In superconductors, however, coupling between the charged condensate and the gauge field shifts the phase mode to higher energies,[1] leaving the Higgs mode as the dominant low-energy excitation. The Meissner effect reflects a macroscopic quantum condensate where photons gain mass - an analogy to high-energy physics.[2] The Higgs mode was first observed in superconductors by Sooryakumar and Klein via Raman scattering in 1980, and confirmed in 2014.[3,4] Due to weak light coupling, it remained elusive except in NbSe₂, where it couples to a charge density wave (CDW). Over the past two decades, experimental evidence has steadily grown. Notably, Budelmann et al. (2005) and Saichu et al. (2009) observed distinct in-gap features suggestive of collective excitations,[5,6] with later THz studies reinforcing the case for the Higgs mode.[7] After introducing activities in our Raman lab at CFEL, this talk presents Higgs-mode observations in Bi-2212 using Non-Equilibrium Anti-Stokes Raman Scattering (NEARS).[8] We use a Tsunami Ti:Sapphire laser (1.2 ps, 80 MHz, 802 nm) and a 402 nm probe from SHG for the UT-3 spectrometer.[9] NEARS exploits a soft quench of the Mexican-Hat potential to selectively populate Higgs modes of different symmetries,[10] then probes them via anti-Stokes Raman scattering. This leads to a tunable population inversion, characterized by comparing Stokes and anti-Stokes signals. An emergent in-gap anti-Stokes peak (25 meV) grows with fluence, indicating Higgs mode population, while the Stokes side shows suppressed pair-breaking at approx. 60 meV, confirming the superconducting state 3 ps post-pump.

Our results, interpreted via Ginzburg-Landau theory and a BCS weak-coupling model, link Higgs mode energy to Cooper-pair coherence length. Phonon-subtracted susceptibilities match well with microscopic theory, notably avoiding the A1g problem at 3.1 eV photon energy.[11] NEARS thus emerges as a powerful tool for Higgs spectroscopy in quantum condensates, with broad implications for superconductivity research, including light-induced, interface, and topological effects.

References

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