BEAMLINES
U12IR, U10A

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DISTURBING SUPERCONDUCTIVITY WITH LIGHT

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Superconductors can be sent out of equilibrium by using light to break Cooper pairs. Using a pump-probe setup, we studied the time-dependent behavior of thus depleted superconductivity. Laser pulses were utilized to break pairs and synchrotron far-infrared (a.k.a. terahertz) pulses measured the spectra as a function of time in the sub-nanosecond scale. This measurement allowed us to determine how electrons recombine into pairs, bringing the system back to equilibrium. The direct observation of the superconducting gap diminution, expected for an excited superconductor, was measured for the first time.

The Bardeen, Cooper, and Schrieffer (BCS) theory of superconductivity showed that, in a classical superconductor, zero electrical resistance is a consequence of electrons pairing themselves. These "Cooper pairs" have a typical binding energy (2Δ) that leads to a gap for unpaired electrons at the Fermi energy. However, Cooper pairs can be broken into two electrons if they are given an energy larger than the gap. One of such possible processes is the absorption of a photon of energy greater than 2Δ. As Cooper pairs are energetically more favorable than electrons, the electrons eventually reform the pair. The full relaxation process is a bit more complex, as shown in **Figure 1**. In equilibrium, all the electrons in a superconductor should form pairs below the Fermi level (**Figure 1A**). The absorption of a photon will excite pairs across the gap and produce electrons above the Fermi energy (**Figure 1B**). We then reach a situation where the "unstable" excited electrons want to recombine into a pair. Energy conservation imposes that the energy gain occurring when electrons form a pair must be emitted, usually as a crystal lattice vibration (phonon). Nevertheless, phonons can also break pairs; therefore, the emitted phonon is able to produce two new excited electrons. This subtle competition between electrons willing to recombine and phonons trying to break pairs only ends because, eventually, phonons become unavailable by either relaxing to very low energies or simply escaping the material. The whole process of relaxation is schematized in **Figure 1C**.

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A collaboration between the University of Florida and the NSLS built a pump-probe setup to study systems out of equilibrium. The facility consists of a Ti:Sapphire pulsed laser synchronized with the electron bunches of the VUV-IR ring. We utilized the laser pulses to break pairs (pump) in several conventional superconductors, including Pb, Nb, and NbN, and the far-infrared light from beamlines U12IR and U10A to probe the changes in the optical response of the superconductor, which strongly depends on the number of Cooper pairs. By delaying the laser pulses with respect to the synchrotron bunches, we can obtain the time-dependent optical response for the specimen.

Figure 2 shows the integrated far-infrared transmission for several out-of-equilibrium superconductors as a function of time and temperature. At zero delay, both the laser and synchrotron pulses arrive at the sample at the same time and the changes in the optical transmission are maximum. As we push the synchrotron pulses further away from the laser, we allow enough time for some pairs to recombine and the optical transmission to recover its equilibrium value. Finally, we obtained the measurement of the non-equilibrium infrared spectrum (**Figure 3**), which is the first direct determination of the superconducting gap shrinkage expected for superconductors with excess unpaired electrons.

This technique is now being used to analyze more complex superconductors such as the cuprate high critical-temperature materials and the “two-superconductors-in-one” material MgB_2.

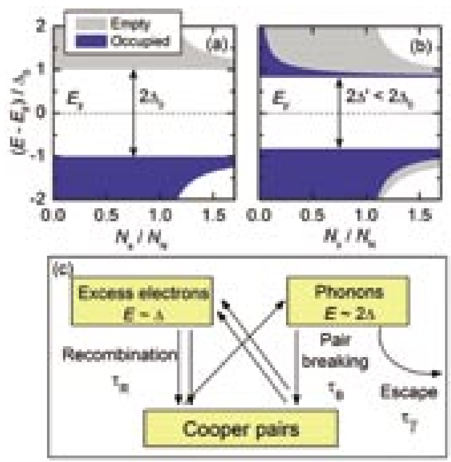

Figure 1. Density of states for a BCS superconductor (A) in equilibrium and (B) where pairs were broken into electrons. Panel (C) shows schematically the processes involved when the system relaxes back to equilibrium. Each process (recombination, pair breaking, and phonon escape) is characterized by a different time scale.

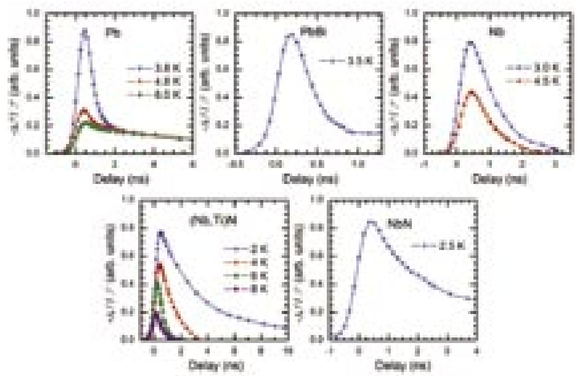

Figure 2. Relative variation in the far-infrared transmission of several classical superconductors as a function of temperature and time.

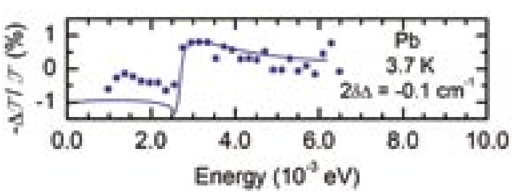

Figure 3. Non-equilibrium far-infrared spectrum for superconducting lead (dots) and expected response from BCS theory (line). The jump in the curve is a direct measurement of the superconducting gap shrinkage expected for a superconductor with excess unpaired electrons.