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Photons and magnetic fields: The use of synchrotron sources to study pairbreaking in superconductors

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Abstract. Pair-breaking effects in metallic superconductors have been studied by both linear and nonlinear spectroscopy using infrared synchrotron radiation. The measurements were performed at the National Synchrotron Light Source, Brookhaven National Laboratory, in magnetic fields up to 10 T. The optical conductivity of thin-film superconductors in applied magnetic fields has been estimated from the results of far-infrared transmission and reflection measurements. The combined measurements have been analyzed to give the real and imaginary parts of the conductivity. In turn, these quantities allow the magnetic-field dependence of the superconducting energy gap, pairbreaking parameter, and superfluid density to be estimated. Photons also may break Cooper pairs. Pump-probe studies of excess quasiparticle relaxation in the these superconducting films show a relaxation rate proportional to the excess quasiparticle number density, as expected for bimolecular recombination driven by a large excess quasiparticle population. Application of a magnetic field parallel to the sample surface is found to slow significantly the quasiparticle recombination process.

1. Introduction

Optical spectroscopy is a widely used technique for the study of the electronic properties of solids, with the interesting photon energy range extending at least from the far-infrared/terahertz range (sub-meV) to the ultraviolet (several eV). These experiments allow studies of physics as diverse as the energy gaps of metallic superconductors, spin excitations in magnets, free-carrier dynamics in metals and semiconductors, interband excitations, the vibrational modes of crystals, and spectral weight transfers associated with metal-insulator or superconducting phase transitions.

The great majority of optical studies are linear, equilibrium experiments. The only deviations from the ground state are caused by thermal excitations. In contrast, this paper describes studies in a rather different situation: infrared studies of dynamics of systems away from equilibrium, in some cases far from equilibrium. We investigate nonequilibrium states of matter through pump-probe far-infrared spectroscopy. A wide range of materials can be studied using this time-resolved method, which makes use of a key feature of synchrotron infrared radiation: the electrons in the synchrotron ring exist in tight bunches traveling around the ring, making the synchrotron light occur as pulses, emitted each time a bunch passes the aperture of the beamline. The length of the bunch sets the duration of the pulse, typically in the sub-nanosecond range. Although perhaps the least exploited aspect of synchrotron radiation, this time structure makes the synchrotron able to do a class of very important experiments that cannot be done in any other way: broadband infrared time-resolved or pump-probe spectroscopy.[1,2]

Our experimental setup uses a short-pulsed near-infrared laser as pump and the very broadband synchrotron infrared radiation as probe. Synchronization between laser pulses and infrared pulses gives the ability to study transient behavior on timescales from ∼300 ps to ∼100 ns. Spectrometers attached to the beamline permit spectroscopy of these transient events in the far-infrared to near infrared ranges.

2. Linear spectroscopy

2.1. Synchrotron infrared radiation

Our measurements used the far-infrared photons from beamline U4IR in the VUV synchrotron storage ring of the National Synchrotron Light Source. The spectra were collected using a Bruker IFS 66-v/S spectrometer and a high sensitivity, large area, B-doped Si composite bolometer operating at 1.8 K; cooled filters limited the upper frequency to 110 cm^{-1}. The samples were mounted in a ^{4}He Oxford cryostat equipped with a 10 Tesla superconducting magnet; the minimum temperature is 1.6 K.

The high brightness of the synchrotron radiation allowed transmission and reflection measurements to be made in the cramped space of the high field region after carrying it many meters from the synchrotron to the interferometer, on to the magnet, and eventually to the detector.

2.2. Magnetic-field-induced pair breaking in superconducting $Nb_{0.5}Ti_{0.5}N$

We measured the complex optical conductivity of a superconducting thin-film of $Nb_{0.5}Ti_{0.5}N$ in an external magnetic field. The field was applied parallel to the film surface and the conductivity extracted from far-infrared transmission and reflection measurements. The real part shows the superconducting gap, which we observe to be suppressed by the applied magnetic field. We compare our results with the pair-breaking theory of Abrikosov and Gor'kov[3] and confirm directly the theory's validity for the optical conductivity.

The normalized optical conductivities at 0, 5, and 10 T are shown in panels (a)–(c) of Fig. 1. A weak interference fringe in both the transmission and reflection measurements results in the excess σ_2/σ_N over the 40 to 80 cm^{-1} range. The solid lines are fits to the data using the pair-breaking theory as extended by Skalski *et al.*[4] to calculate σ_1/σ_N at 0 K. Panel (d) shows the fitted σ_1/σ_N at six different fields. The fits used the pair-breaking theory as extended by Skalski *et al.*[4] to calculate σ_1/σ_N at 0 K. Clearly, the absorption

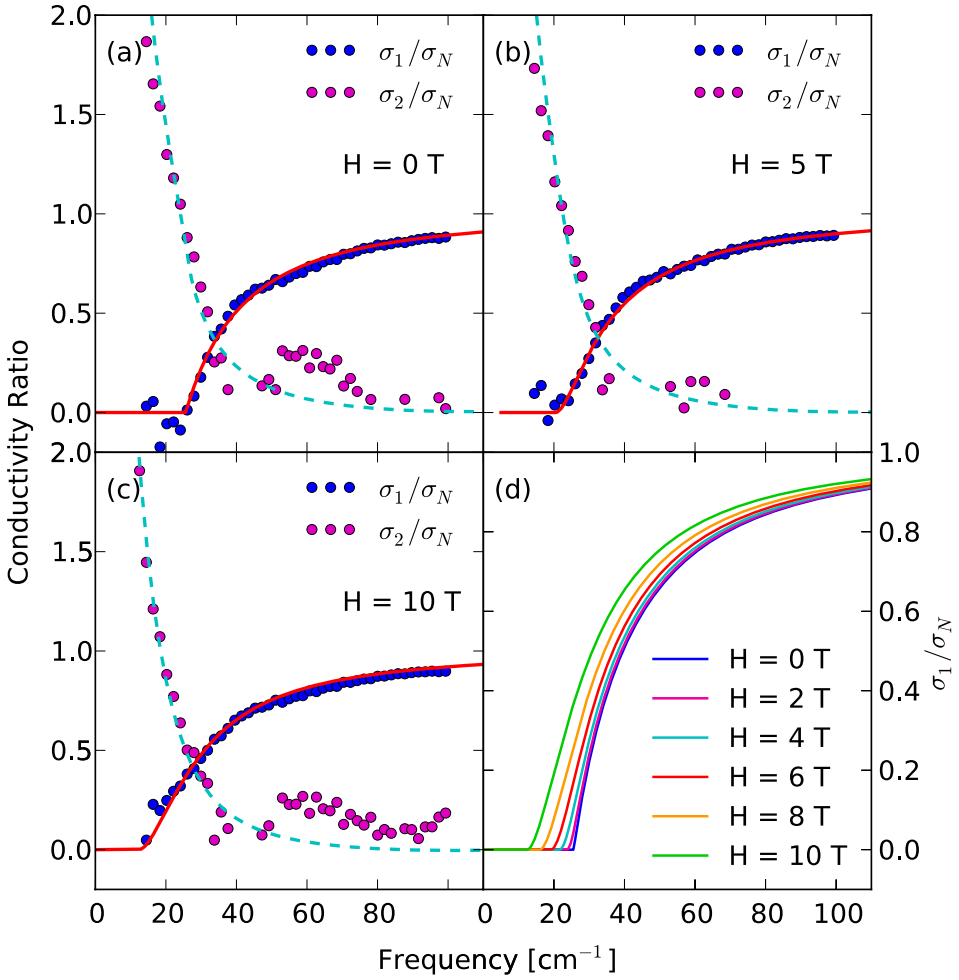

Fig. 1.(a)–(c): The real and imaginary parts of the $T = 3$ K superconducting state optical conductivity (normalized to the normal state conductivity) at three different applied magnetic fields. The solid lines are fits to σ_1/σ_N using the pair-breaking theory and the dashed lines show σ_2/σ_N from a Kramers-Kronig transform of the real part. (d): The fitted σ_1/σ_N at six fields.

edge moves to lower energy as the field increases. The field-induced pair breaking also smears out the gap-edge singularity in the quasiparticle density of states[4], so that the initial rise of σ_1 becomes less abrupt for increasing fields, as can be seen by comparing the 0 T and the 10 T results in Fig. 1. Our analysis[5] finds that the suppression of both the superconducting order parameter and the spectroscopic gap is in good accord with the pair-breaking theory.[3]

3. Pump-probe spectroscopy

3.1. Experimental methods

Figure 2 shows a schematic diagram of the pump-probe facility.[2] The sample is first irradiated with a short, relatively high-power, "pump" pulse, creating a nonequilibrium photoexcited state. These excitations, their interactions, and their dynamics modify the frequency-dependent optical properties of the material; these changes are detected using a "probe" pulse, which irradiates the material with a known time delay after the pump

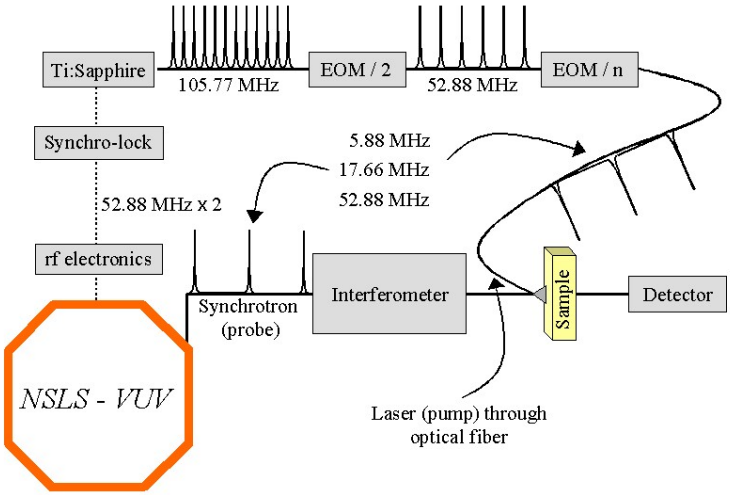

Fig. 2. Apparatus for time-resolved pump-probe measurements.

pulse. The evolution of these photoinduced changes in optical properties can be followed by varying the time delay. Beamline U4IR was used for pump-probe spectroscopy.

A major issue in any pump-probe spectroscopy is minimization of thermal effects due to the pump laser. If the experiment is done by measuring with the laser on for a while and then measuring with the laser off for a while, it is extremely difficult to ensure that the sample temperatures are identical for the two phases of the measurement. Because photoinduced effects are rather small, small temperature changes, hotter when illuminated and cooler when in the dark can mimic the nonequilibrium effects that one wishes to observe. Consequently in our experiment, the laser is *always* on. We inject a small audio-frequency modulation into the laser/synchrotron locking apparatus, causing the arrival time of the laser pulses to dither back and forth relative to the infrared pulse

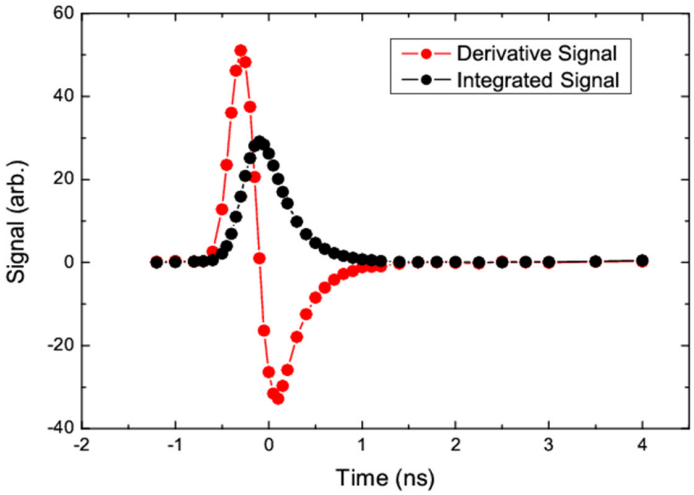

Fig. 3. The red curve shows the measured derivative signal and the black curve the integrated photoinduced absorption. The probe beam in this case is broadband infrared synchrotron radiation, with energy over 10–80 cm^{-1}, peaked near the energy gap of the NbTiN film.

arrival. Just as when field modulations are used in an NMR or EPR experiment, this dither gives us a signal which is the derivative of the photoinduced response of the sample. Integration gives us the desired photoinduced signal, uncontaminated with thermal effects. Figure 3 shows the measured signal and its integral.

The photoinduced signal (at low fluence) is a convolution of the exponential relaxation of the photoinduced carriers and the near-Gaussian time dependence of the probe beam. We can fit to this behavior to extract the recombination time in the sample.

3.2. Quasiparticle dynamics

When a superconductor is subjected to a pulse of above-bandgap electromagnetic radiation, excess quasiparticles are created. Their number and how they recombine back into Cooper pairs are of fundamental importance. Calculations based on a phonon emission process,[6–8] found a rather fast recombination rate. However, Rothwarf and Taylor[7] showed that the measured (or effective) lifetime was longer than the "bare" or intrinsic recombination time due to a phonon-bottleneck effect.

Figure 4 shows a cartoon picture of the process. The laser photon creates a pair of very hot quasiparticles. These relax rapidly by interaction with other charged particles and by emission of high-energy phonons, breaking other Cooper pairs during this time. In picoseconds, much faster than the measurement capability of the synchrotron probe,[9] the quasiparticles relax to the gap edge. Here, each quasiparticle must find a partner of opposite spin to recombine into a Cooper pair. The recombination releases the pair binding energy in the form of a 2Δ phonon. However, this phonon can (and will) break another Cooper pair, creating two gap-edge quasiparticles. A stalemate occurs. Only if the phonon escapes from the superconductor or decays anharmonically can the excess quasiparticle density be relaxed. It is this bottleneck that we have studied.

Experiments on photoinduced pairbreaking include the work of Testardi,[10] who

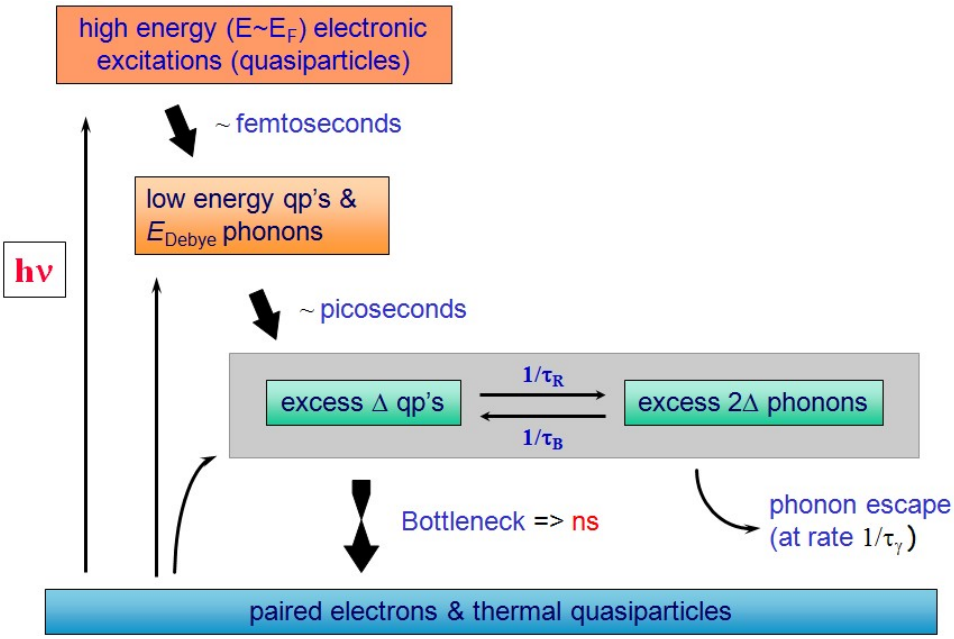

Fig. 4. Cartoon of the pairbreaking and recombination process, showing the bottleneck for the gap-edge quasiparticles and 2Δ phonons.

showed that light of enough intensity could destroy the superconductivity in a process not attributable to lattice heating. Parker and Williams[11] and Schuller and Gray[12] used this phenomenon to measure pair recombination rates in illuminated tunnel junctions. Sai-Halasz *et al.*[13] used a pulsed laser to create the excess number of quasiparticles and analyzed the changes in the microwave reflectivity. Their data support the theory of Owen and Scalapino[14] for the low-fluence regime, where the number of quasiparticles created by the photon absorption is much smaller than the number of unpaired quasiparticles produced by finite sample temperature Johnson[15] was the first to use fast lasers and electronics to look at voltage transients with time resolution in the range of 100 ps. Measurements of quasiparticle recombination using synchrotron radiation include the work of Carr *et al.*[16] on Pb and Lobo *et al.*[17] on a variety of superconductors.

3.3. Pump-probe results

We used pump-probe spectroscopy to measure quasiparticle recombination dynamics in metallic superconductors ($Nb_{0.5}Ti_{0.5}N$ and NbN thin films) in an external magnetic field. The field was applied parallel to the film surface in order that vortex dynamics not play a role in the measurements. Figure 5 shows an example of our results. The photoinduced signal—proportional to the excess quasiparticle density; $S(t) \propto N_{ex}(t)$—is shown versus the delay time between laser pump and far-infrared probe. Note the semilog presentation. The left panel shows the relaxation at low laser fluence whereas the right panel shows the behavior at high fluence. The low fluence data are close to a simple exponential; the high fluence data are not. At high fluences, the recombination is fast, and then slows later in the process, as the excess density becomes smaller. Note that the signal is followed over two orders of magnitude in its strength.

The effect of field is to decrease the rate in both low and high fluence situations. The recovery takes longer when the field strength is increased. We can consider two

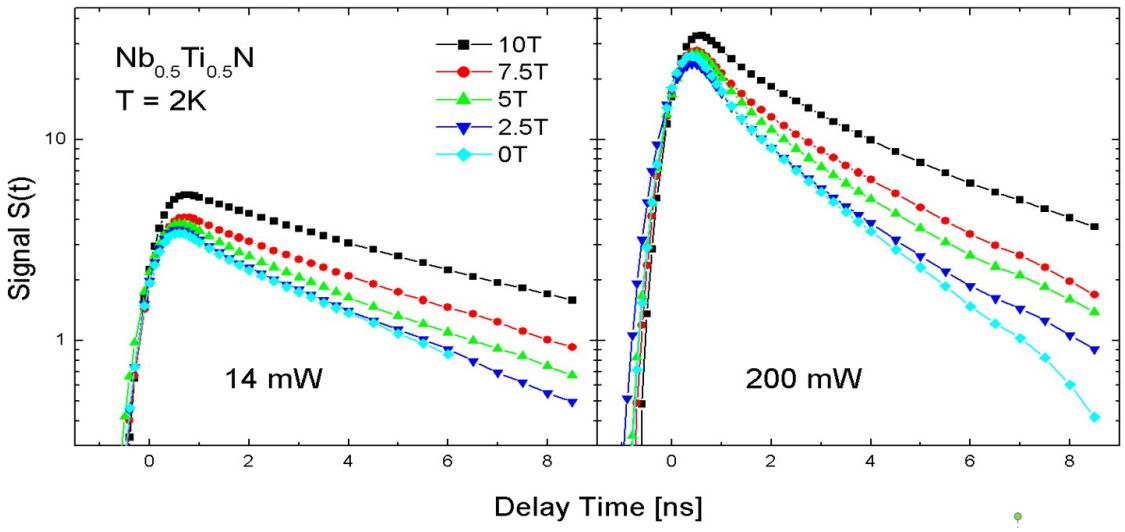

Fig. 5. The photoinduced signal is shown on a logarithmic scale versus the delay time between laser pump and far-infrared probe. The left panel shows the signal for low laser fluence and the right panel for higher fluence. The measurements were made with the sample immersed in 2 K liquid Helium and at applied magnetic fields from 0 to 10 T.

effects to understand this result. In one, the magnetic field produces a paramagnetic spin polarization, making it difficult for a majority-spin quasiparticle to find an opposite-spin partner to pair up with, thus slowing the recombination. Although attractive, the presence of spin-orbit coupling reduces the importance of this effect. The second effect is finite pair lifetime, observed in the linear spectroscopy in field (Fig. 2). We can therefore understand the recombination within the same pairbreaking theory[3] used to explain the linear spectroscopy.

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