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Time-resolved magnetospectroscopy of quasiparticle dynamics in superconducting $Nb_{0.5}Ti_{0.5}N$

X. Xi [a,*], J. Hwang [a,b], H. Zhang [a], C.J. Stanton [a], D.H. Reitze [a], D.B. Tanner [a], G.L. Carr [c]

[a] Department of Physics, University of Florida, Gainesville, FL 32611, USA
[b] Department of Physics, Pusan National University, Busan 609-735, Republic of Korea
[c] National Synchrotron Light Source, Brookhaven National Laboratory, Upton, NY 11973, USA

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ABSTRACT

Pump–probe spectroscopy has been used to study quasiparticle dynamics in a superconducting $Nb_{0.5}Ti_{0.5}N$ thin film subject to a parallel magnetic field. The broadband, time-resolved, photo-induced far-infrared transmission $S(t)$ was measured and used to extract a time-dependent effective recombination rate $\tau_{\rm eff}^{-1}(t)$. We found that $\tau_{\rm eff}^{-1}(t)$ decreases with increasing field. The rate $\tau_{\rm eff}^{-1}(t)$ is found to scale with $S(t)$ at high laser fluence.

1. Introduction

The pump-probe technique has become a powerful tool for the study of excitations in superconductors. A pulsed laser generates excess quasiparticle excitations by breaking Cooper pairs. These excitations relax rapidly to the superconducting gap edge and then, more slowly, recombine with partners of opposite spin, releasing energy as phonons. The most commonly used model is the phenomenological theory of Rothwarf and Taylor [1] that considers both the bimolecular nature of the recombination process as well as the "phonon bottleneck" that results when recombination phonons re-break pairs. In general, the relaxation time is governed by conditions such as temperature and the density of excess quasiparticles. In this paper, we study the quasiparticle dynamics as a function of laser fluence and static magnetic field. The influence of a magnetic field on a superconducting thin film has been discussed in terms either of a Zeeman splitting of the quasiparticle states [2] or as a pair-breaking effect similar to that caused by magnetic impurities [3].

2. Experimental

The sample, $a \sim 10$ nm thick $Nb_{0.5}Ti_{0.5}N$ film on a 0.5 mm thick quartz substrate, is a type II superconductor with $T_c \approx 12$ K and optical gap $2\Delta_0 \approx 28.5\ \rm cm^{-1}$. H_{c2} is estimated to be 15 T from magnetic susceptibility measurements. An Oxford superconducting magnet provides fields up to 10 T and temperatures down to 2 K. In our time-resolved study, we pump the sample with near-infrared Ti:sapphire laser pulses (∼1 ps rms) and probe with far-infrared pulses (down to 150 ps rms) produced as synchrotron radiation at beamline U4IR of the National Synchrotron Light Source (Brookhaven Nat'l Lab).

To improve our sensitivity, we employ a differential measurement technique where the arrival time of the laser pump pulse is sinusoidally dithered a small amount dt with respect to the infrared probe pulse. The resulting change in the far-infrared detector signal dS/dt is then measured using a lock-in amplifier. Integrating over time, we obtain the broadband far-infrared transmission $S(t)$, which is a measure of the excess quasiparticle density. Fig. 1 shows the field dependence of $S(t)$ at low and high laser fluences. Simple exponential behavior is seen at low fluence, but more complex decays are seen at high fluence: initially fast, but then slowing as the density of excess quasiparticles diminishes.

3. Results and discussion

In our analysis, we work with an instantaneous effective recombination rate, defined as

$$\tau_{\rm eff}^{-1}(t) = -\frac{1}{S(t)}\frac{dS}{dt}(t) \qquad (1)$$

* Corresponding author.
E-mail address: xiaoxiang@phys.ufl.edu (X. Xi).

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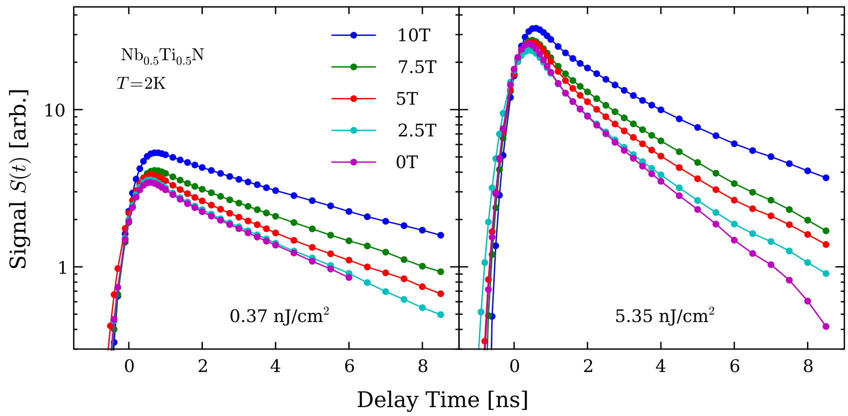

Fig. 1. Semi-log plots of excess quasiparticle signal at different magnetic fields for weak (left) and strong (right) laser fluences. The laser pulse was focused on a 6 mm diameter spot.

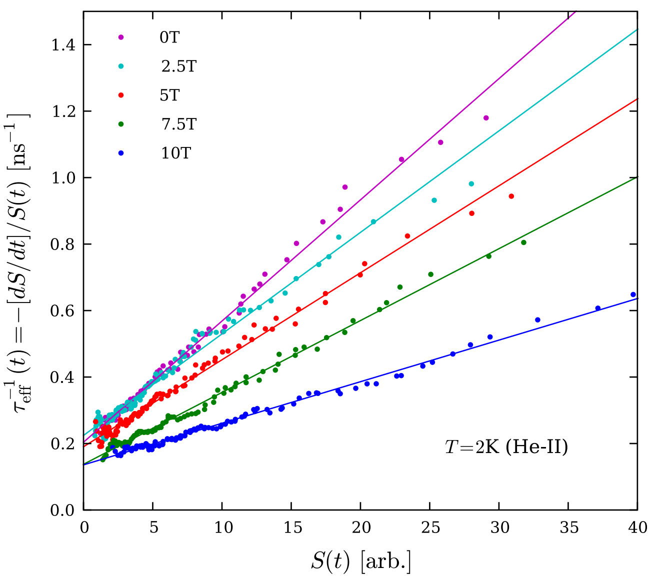

Fig. 2. The instantaneous relaxation rate shows a linear dependence on signal at each field. The data shown include fluences from 0.37 to 10.70 nJ/cm^2.

We find that the rate $\tau_{\rm eff}^{-1}(t)$ increases linearly with $S(t)$ (see Fig. 2) as expected for a bimolecular type of recombination. Moreover, the rate $\tau_{\rm eff}^{-1}(t)$ for a given $S(t)$ decreases with increasing field. This new result suggests that the spin polarization of the excess quasiparticles may be limiting the effective recombination rate. To address this, we extend the recombination rate expressions to include spin-polarized quasiparticle populations. In this model, the total number of quasiparticles N consists of the thermal number $N_{\rm th}$ and the excess number $N_{\rm ex}$ generated by laser pulses: $N = N_{\rm th} + N_{\rm ex}$. For a bi-molecular recombination of quasiparticles with opposite spin we expect that,

$$\frac{dN}{dt} = \frac{dN^{\uparrow}}{dt} + \frac{dN^{\downarrow}}{dt} = -2R(N^{\uparrow}\cdot N^{\downarrow} - N^{\uparrow}_{\rm th}\cdot N^{\downarrow}_{\rm th}) \tag{2}$$

where $N^{\uparrow}$ and $N^{\downarrow}$ denote the spin-up and spin-down populations, and R is the effective recombination rate including phonon bottleneck effects. Noting that the photo-induced signal S is directly proportional to the excess quasiparticle number, we can write $S = C\cdot N_{\rm ex}$. Defining the spin-up and spin-down population fractions as $N^{\uparrow} = P^{\uparrow}\cdot N$ and $N^{\downarrow} = P^{\downarrow}\cdot N$ we can re-write Eq. (2) as

$$-\frac{1}{S}\frac{dS}{dt} = 2\frac{R}{C}\cdot(P^{\uparrow}\cdot P^{\downarrow})\cdot[S + 2S_{\rm th}] \tag{3}$$

where $S_{\rm th} = C\cdot N_{\rm th}$. Eq. (3) has the form of straight line that accounts for the measured relaxation rate (shown in Fig. 2) if the slope $2\frac{R}{C}\cdot(P^{\uparrow}\cdot P^{\downarrow})$ is a function of applied field. Note that the product $P^{\uparrow}\cdot P^{\downarrow}$ is maximum when $P^{\uparrow} = P^{\downarrow} = \frac{1}{2}$ (no net polarization) and approaches zero as the polarization increases.

While the degree of spin polarization is implicitly field-dependent, the effective recombination rate R is determined by factors such as the energy gap Δ and transition temperature T_c [4], both of which are also affected by an applied field. Which factor is more important in determining the field-dependence depends on whether the sample is in the paramagnetic limit or the pair-breaking limit [5]. Measurements of the energy gap as a function of field (presently underway) will hopefully resolve this question.

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