Impact of damping on superconducting gap oscillations induced by intense Terahertz pulses

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We investigate the interplay between gap oscillations and damping in the dynamics of superconductors taken out of equilibrium by strong optical pulses with sub-gap Terahertz frequencies. A semi-phenomenological formalism is developed to include the damping within the electronic subsystem that arises from effects beyond BCS, such as interactions between Bogoliubov quasiparticles and decay of the Higgs mode. Such processes are conveniently expressed as $T_1$ and $T_2$ times in the standard pseudospin language for superconductors. Comparing with data on NbN that we report here, we argue that the superconducting dynamics in the picosecond time scale, after the pump is turned off, is governed by the $T_2$ process.

Introduction. – The coherent control of non-equilibrium states of interacting quantum matter promises far-reaching capabilities by turning on (or off) desired electronic material properties. A particular focus in this field has been the manipulation of superconductivity by non-equilibrium probes. While earlier works showed that microwave pulses could be used to enhance the superconducting transition temperature $T_c$ of thin superconducting films [1, 2], recent advances in ultrafast pump-and-probe techniques opened the possibility of investigating superconductivity in the pico- and femto-second timescales by coherent light pulses [3, 4]. Such coherent pulses have been employed to manipulate the electronic and lattice properties of quantum materials, resulting in transient behaviors that are consistent with the onset of non-equilibrium superconductivity above $T_c$ [5–7]. Alternatively, coherent pulses have also been employed to assess the coherent dynamics of the superconducting state [3, 4, 8–13].

To maintain coherence and avoid excess heating, it is advantageous to apply pulses at energies below twice the superconducting gap $2\Delta$, where quasi-particle (Bogoliubov) excitations are absent. As the superconducting gap energy scale lies in the Terahertz (THz) regime, this requires the application of intense and coherent sub-gap THz light pulses [14]. In Ref. [3], a monocycle intense THz pulse was applied to a thin film of the conventional $s$-wave superconductor NbN, reporting coherent oscillations of the superconducting gap with frequency $2\Delta$.

Such oscillations arise naturally from the solution of the time-dependent BCS (Bardeen-Cooper-Schrieffer) equation [15–22], which can be conveniently recast in terms of Anderson pseudospins [23] precessing around a pseudo magnetic field that is changed by the optical pulse. While this coherent evolution describes qualitatively well the behavior of the system in a restricted time window, there is also damping present in the system, which is absent in this BCS approach.

Here, we develop a semi-phenomenological model that captures not only the coherent evolution of the gap function, in the picosecond time scale, but also damping effects in the time scale of tens to hundreds of picoseconds. Since this time scale precedes the thermalization with the lattice, the relevant relaxation processes arise within the electronic subsystem from effects not captured by BCS. These include interactions between Bogoliubov quasiparticles and the coupling between the Higgs (amplitude) mode and the continuum. In the pseudospin notation, we identify two types of relaxation process: the longitudinal relaxation $T_1$, corresponding to relaxation of quasiparticles, and the transverse relaxation $T_2$, corresponding to relaxation of the gap.

We apply this formalism to elucidate the dynamics of superconducting NbN, which was measured at very low temperatures using intense THz fields with sub-gap spectra. Our data reveals the gap oscillating at a frequency corresponding to twice the pump frequency. When the pump is turned off, however, the gap oscillations quickly disappear, and the amplitude of the gap continues to be suppressed. Such a behavior is at odds with the nonequilibrium dynamics given by the time-dependent BCS equation, where the gap displays coherent oscillation with very slow collisionless relaxation [15]. We show instead that this behavior is well captured by our semi-phenomenological model, and arises from a dominant $T_2$ relaxation process whose time scale is of the same order as the duration of the pump.

Experimental results. – The data was acquired using an intense THz pump, weak THz probe ultrafast spectroscopy setup. A Ti-Sapphire amplifier was used to generate pulses of energy 3 mJ, duration 40 fs, 1 KHz repetition rate, and 800 nm center wavelength. The pulses were split into three paths: pump, probe and sampling. The intense THz pump pulses were generated by the tilted-pulse-front phase matching through a 1.3% MgO doped LiNbO$_3$ crystal. The weak THz probe pulses, generated by optical rectification, were detected by free space electro-optic sampling through a 1mm thick (110) ZnTe
Figs. 1 (A)-(B) show the behavior of the real and imaginary parts of the optical conductivity, \( \sigma_1(\omega) \) and \( \sigma_2(\omega) \), respectively. In equilibrium (gray curves), the onset of superconductivity below \( T_c \approx 13.4 \) K is signaled by the opening of a gap \( 2\Delta \approx 4.2 \) meV in \( \sigma_1(\omega) \), and by a \( 1/\omega \) dependence of \( \sigma_2(\omega) \) at low frequencies. The post-pump state (red curve) exhibits larger values of \( \sigma_1(\omega) \) within the \( 2\Delta \) range, and slightly reduced values of \( \sigma_2(\omega) \), presumably due to the THz-induced quench of the SC condensate [8].

To extract the ultrafast dynamics of the gap function, we measure the change in the transmitted field \( \Delta E/E \), which was shown in Ref. [4] to faithfully reflect the transient behavior of \( \Delta(t) \). Fig. 1 (C) shows the ultrafast time evolution of \( \Delta E/E \propto 1 - |\Delta(t)|/\Delta_0 \), with \( \Delta_0 \equiv \Delta(t=0) \), well inside the superconducting state (blue curve, at \( T = 4 \) K), superimposed with the applied pump pulse (red curve). Interestingly, we find oscillations on \( \Delta(t) \) only while the pump pulse is on. After it is turned off, the oscillations disappear quickly, but \( \Delta(t) \) continues to decrease on the time scale of tens of picoseconds. The Fourier decomposition of \( \Delta E/E \) (not shown) indeed demonstrates that the gap oscillations do not scale with the gap function, unlike reported for shorter monocyte pulses [3], but instead correspond to twice the pump frequency [4].

To model and elucidate these experimental results, we need to consider relaxation processes beyond the standard coherent time evolution predicted in BCS theory. Within BCS, the quench dynamics of \( \Delta(t) \) can display three different behaviors [15, 25–27]: (i) overdamped decay of \( \Delta(t) \rightarrow 0 \) (phase I); (ii) underdamped oscillations with frequency \( 2\Delta_\infty \) that decay algebraically \( \propto t^{-1/2} \) towards a finite asymptotic value \( \Delta(t) \rightarrow \Delta_\infty \) (phase II); and (iii) persistent undamped oscillations (phase III).

In contrast to these predictions, our experimental observation is that although the gap oscillations are rapidly damped out, the gap remains finite after the pump pulse is off (see Fig. 1(C)). Moreover, it continues to show a slow decay between 10 ps and 20 ps, a behavior that presumably persists into the time scale of hundreds of picoseconds. The gap eventually returns to its initial equilibrium value on even longer nanosecond time scales via equilibration with phonons. This regime is not discussed in this paper.

To explain this discrepancy, one must include damping within the electronic subsystem. Before discussing possible microscopic mechanisms for damping, we employ a phenomenological approach that is best expressed within the pseudospin description of the BCS model. The stan-
The Hamiltonian then takes the simple form
\[
\frac{|\Delta|^2}{V_0} \sum_{\mathbf{k}} (\Delta_{\mathbf{k}+\mathbf{t}} + \Delta_{\mathbf{k}-\mathbf{t}} + \text{h.c.}) + |\Delta|^2 
\]

Here we consider the square-lattice dispersion \( \varepsilon_{\mathbf{k}} = -2J(\cos k_x + \cos k_y) \), and \( \xi_\mathbf{k} = \varepsilon_{\mathbf{k}} - \mu \), with chemical potential \( \mu = -1.18J \) corresponding to quarter-filling, and electron charge \( e_0 \). The superconducting order parameter obeys the self-consistent equation \( \Delta = -V_0 \sum_{\mathbf{k}} (c_{\mathbf{k}+\mathbf{t}} c_{\mathbf{k}-\mathbf{t}}) \), where \( V_0 < 0 \) denotes an attractive interaction. For the calculations in this paper, we set \( V_0 = -3J \) and the Debye frequency \( \omega_D = J/2 \), yielding \( \Delta_0 = 0.08J \) and \( T_c = 0.048J \).

The vector potential \( \mathbf{A}(t) \) is related to the electric field of the pump via \( \mathbf{E}_{\text{pump}} = -\frac{\partial}{\partial t} \mathbf{A} \). In our experiment, it takes the form \( \mathbf{A}(t) = A_0 \theta(-t) \theta(\tau - t) \mathbf{E}_{\text{pump}} e^{-(t-\tau)^2/2\sigma^2} \cos(\omega_{\text{pump}} t) \) with center frequency \( \omega_{\text{pump}}, \) temporal width \( \sigma \), linear polarization vector \( \mathbf{e}_{\text{pump}} \), and duration \( \tau \). For the calculations in Fig. 1(D) and 2, which refer to our experiments on NbN, we consider a long pulse with \( \tau = 10\pi/\Delta_0 \), \( \sigma = \tau/5 \), \( A_0 = \sqrt{0.75\Delta_0} \) and \( \omega_{\text{pump}} = 1.41\Delta_0 \), corresponding to a sub-gap frequency. To compare with previous experiments involving short pulses, such as Ref. [3], in Fig. 3 we consider a Gaussian-shaped short pulse \( \mathbf{A}(t) = A_0 \theta(-t) \theta(\tau - t) \mathbf{E}_{\text{pump}} e^{-(t-\tau)^2/2\sigma^2} \) with \( \tau = 5/\Delta_0 \), \( A_0 = \sqrt{1.5}\Delta_0 \).

To describe the gap dynamics, it is convenient to use Anderson pseudospins \( \mathbf{S}_\mathbf{k} = \psi_{\mathbf{k}}^\dagger \vec{\tau} \psi_{\mathbf{k}} \), with Nambu spinor \( \psi_{\mathbf{k}} = (c_{\mathbf{k}+\mathbf{t}}, c_{\mathbf{k}-\mathbf{t}}^\dagger)^T \) and Pauli matrices \( \vec{\tau} \). The Hamiltonian then takes the simple form \( H_{\text{BCS}} = -\sum_{\mathbf{k}} \mathbf{B}_\mathbf{k} \cdot \mathbf{S}_\mathbf{k} + \frac{|\Delta|^2}{V_0}, \) with a pseudo magnetic field \( \mathbf{B}_\mathbf{k} = 2(\Delta' - \Delta'', -\xi_\mathbf{k} + e_0 A_\mathbf{k} \mathbf{t}) \), where \( \Delta = \Delta' + i\Delta'', \) and \( \xi_\mathbf{k} + e_0 A_\mathbf{k} \mathbf{t} \). Importantly, the magnetic field depends itself on the state of the pseudospins via \( \Delta = -V_0 \sum_{\mathbf{k}} \langle \mathbf{S}_\mathbf{k} \rangle \).

In equilibrium, all spins are aligned with the field direction and their expectation value is given by \( \langle \mathbf{S}_\mathbf{k} \rangle = \frac{1}{2} \hat{s}_{\mathbf{k},\text{eq}} \tanh(\frac{\mu}{2T_c}) \). Here, \( T_c \) denotes the initial temperature, \( E_k = \sqrt{|\Delta|^2 + \xi_\mathbf{k}} \) is the Bogoliubov quasiparticle dispersion, and \( \hat{s}_{\mathbf{k},\text{eq}} = (\cos \phi \sin \theta_\mathbf{k}, -\sin \phi \sin \theta_\mathbf{k}, -\cos \phi) \) is a unit vector denoting the direction of the pseudospins. The polar angle is determined by the ratios \( \sin \theta_\mathbf{k} = |\Delta|/(2E_k) \) and \( \cos \theta_\mathbf{k} = \xi_\mathbf{k}/(2E_k) \), whereas \( \phi \) is the phase \( \Delta = |\Delta|e^{i\phi} \).

The pump pulse \( \mathbf{A}(t) \) changes the band dispersion, which in turn changes the \( z \)-component of the pseudo magnetic field \( B_k \). Within BCS, the spins precess around the new \( B_k \) according to \( \frac{d}{dt} \hat{s}_{\mathbf{k},\text{eq}} = \langle \mathbf{S}_\mathbf{k} \rangle \times \mathbf{B}_\mathbf{k} \). Importantly, the pseudospin dynamics is immediately fed back into the magnetic field via the gap equation. Due to parity symmetry, only even-order terms of \( \mathbf{A}(t) \) appear [23, 28], and the oscillation frequency of the gap during the pump is a multiple of \( 2\omega_{\text{pump}} \).

By setting \( 2\omega_{\text{pump}} \approx 2\Delta_0 \), the pump pulse resonantly drives the coherent \( 2\Delta \) gap oscillations after the pump pulse is turned off (i.e. \( t > \tau \)), similarly to interaction quenches [29]. However, as none of the quench dynamics predicted by time-dependent solutions of the BCS Hamiltonian (phases I-III described above) is observed experimentally in NbN (see Fig. 1), we go beyond this description and include phenomenologically damping in the pseudospin equations of motion. The microscopic origin of these terms will be discussed below. In analogy with the general problem of spin precession, we introduce longitudinal \( (T_1) \) and transverse \( (T_2) \) relaxation rates:

\[
\frac{d\langle S_{\mathbf{k}} \rangle}{dt} = \langle S_{\mathbf{k}} \rangle \times \mathbf{B}_\mathbf{k} - \frac{\langle S_{\mathbf{k}} \rangle \cdot \hat{s}_{\mathbf{k},\text{eq}} - \langle |\langle S_{\mathbf{k}} \rangle | \rangle}{T_1} \hat{s}_{\parallel,\mathbf{k}} - \sum_{i=1}^{2} \frac{\langle S_{\mathbf{k}} \rangle \cdot \hat{s}_{\perp,\mathbf{k}}^i - \langle |\langle S_{\mathbf{k}} \rangle | \rangle}{T_2} \hat{s}_{\perp,\mathbf{k}}^i \tag{2}
\]

Here, \( \langle S_{\mathbf{k}} \rangle[T_1(t)] = \frac{1}{2} \hat{s}_{\perp,\mathbf{k}}^+ [T_1(t)] \tanh(\frac{\Delta^2 - \Delta'^2}{2T_1(t)}) \) is the thermalized pseudospin configuration at time \( t \) at an effective temperature \( T_1 \). The two vectors \( \hat{s}_{\perp,\mathbf{k}}^i \) span the plane perpendicular to the equilibrium pseudospin direction \( \hat{s}_{\parallel,\mathbf{k}} \). Physically, the time scale \( T_1 \) is related to a re-

FIG. 2. (A) The time evolution of the internal energy of the electronic subsystem for \( T_1 = T_2 = \infty \) (dashed) and \( T_1 = 2T_2 = 1.5\tau \) (red) arising from the energy deposited by the pump. The energy is normalized by \( N_f \Delta_0 \), where \( N_f \) is the density of states at the Fermi level. (B) The effective temperature after the pump is turned off, \( T_f \equiv T^* (\tau) \), normalized by \( T_c \), as a function of the pump intensity for various \( T_1 \) and \( T_2 \). For finite \( T_1, T_2 \), the system will relax to the normal state once \( T_f > T_c \), which leads to an increased energy absorption (as indicated by the change of slope of \( T_f \) when crossing the red dashed line).
distribution of the quasiparticles, whereas the time scale \( T_2 \) is related to the relaxation of the gap to the thermalized value \( \Delta_* \).

To compute \( \hat{\delta}_k \) and the effective temperature \( T_* \), we consider that all the energy deposited in the electronic subsystem by the pump is converted into a change in the internal energy \( \mathcal{E}(t) = \langle H_{\text{BCS}}(t) \rangle_{A=0} - \langle H_{\text{BCS}} \rangle_i \) (see also Ref. [30]). Here, the expectation value is calculated in the time-evolved BCS state according to Eq. (2) and \( \langle H_{\text{BCS}} \rangle_i \) is the initial ground state energy. From \( \mathcal{E}(t) \), we extract both \( T^* \) and \( \Delta^* \), which are themselves function of time while the pump is turned on. Once the pump is turned off, energy is no longer deposited in the electronic subsystem, and thus \( T^*(t > \tau) = T^*(\tau) \equiv T_f \). Fig. 2(A) shows \( \mathcal{E}(t) \) for different values of \( T_1 \) and \( T_2 \). The parameters used are the same as in Fig. 1(D). Clearly, the effects of \( T_1 \) and \( T_2 \) kick in when the pump is weak, as the relaxation processes redistribute the energy within the electronic subsystem. In Fig. 2(B), we show how the “final” temperature \( T^*(\tau) \equiv T_f \) depends on the pump fluence. As expected, for sufficiently strong pumps, the superconducting state can be completely melted by heating.

Using this semi-phenomenological approach, we can capture, as shown in Fig. 1(D), the experimentally-observed gap dynamics of NbN shown in Fig. 1(C). In this calculation, we set \( T_1 = 2T_2 = 1.5\tau \). In contrast to the case with no damping, \( T_1 = T_2 = \infty \) (Fig. 1(D)), we find that the oscillations of \( |\Delta(t)| \) are quickly suppressed after the pulse is turned off, and that a continuous and slow increase of \( 1 - |\Delta(t)|/\Delta_0 \) takes place over the time scale of tens of picoseconds. This characteristic behavior has also been recently observed in ultraclean samples of Nb$_3$Sn, with a larger post-pump suppression of the gap [31].

To correctly capture the experimental observations, it is crucial to restrict \( T_2 \) to the time scale of the order of the pump duration. To further elucidate how \( T_1 \) and \( T_2 \) affect the time-evolution of \( |\Delta(t)| \), in Fig. 3 we explore different parameter regimes. In order to highlight the effects of \( T_1 \) and \( T_2 \), and also to make connection with experiments using short pulses [3], we consider a short Gaussian-shaped pulse of duration \( \tau = 5/\Delta_0 \). As expected, when \( T_{1,2} \gg \tau \), the behavior of \( |\Delta(t)| \) is essentially the same as of the system without damping (Fig. 3A). As \( T_{1,2} \) decrease, the damping increases and the gap oscillations become noticeably damped for \( T_{1,2} \) of the same order as the pump duration \( \tau \). To disentangle the contributions of \( T_{1,2} \), we show \( |\Delta(t)|/\Delta_0 \) for fixed \( T_2 \) (\( T_1 \)) and changing \( T_1 \) (\( T_2 \)) in panel B (C). It is evident that the oscillatory behavior of \( |\Delta(t)| \) is much more sensitive on the transverse relaxation \( T_3 \) than on the longitudinal relaxation \( T_1 \), which only affects weakly the asymptotic value of the gap.

We therefore conclude that our experimental observations using long pulses suggest a dominant \( T_2 \) process in NbN. It is interesting to note that signatures of damping were also present in previous experiments on the same material but using short pulses [3]. Although oscillations were observed in that case after the pump was off, their decay was reported to be much stronger than the polynomial \( 1/\sqrt{t} \) decay predicted by the coherent BCS dynamics. Comparison with our results in Fig. 3A reveals that this effect may be explained by the same damping processes revealed in our experiment.

Although \( T_1 \) and \( T_2 \) are phenomenological quantities, it is important to discuss their possible microscopic origins. As we explained above, \( T_{1,2} \) processes arise within the electronic subsystem, before equilibration with the lattice. Because the BCS Hamiltonian is integrable [16, 25, 32], any damping must arise from non-BCS effects. Residual interactions between the Bogoliubov quasiparticles, which are neglected in the mean-field BCS approach, could provide a mechanism for quasiparticle relaxation, which affects \( T_1 \). Moreover, the Higgs (amplitude) mode excited resonantly by the laser pump disperses into the quasiparticle continuum [29, 33]. As a re-
result, one expects damping of the amplitude mode, which should affect the $T_2$ process.

**Conclusions.**—In this paper, we established a semi-phenomenological framework that allows us to incorporate damping in the picosecond time-evolution of the gap function of an $s$-wave superconductor subject to an intense THz pulse. In the pseudospin language, damping arises from a longitudinal process $T_1$ (related to quasiparticle relaxation) and from a transverse process $T_2$ (related to relaxation of the gap). Our experimental results reveal that, in NbN, for large-amplitude long pump pulses, the picosecond evolution of the gap function is different than that expected for coherent BCS-like dynamics. Instead, we showed that the experimental behavior is consistent with a dominant $T_2$ process that arises within the electronic subsystem, and that has the same time scale as the duration of the pump. Future application of this approach to different superconductors will allow one to distinguish the type of relaxation processes dominant in each system.

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