Nonequilibrium dynamics and coherent control of BCS superconductors driven by ultrashort THz pulses

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Abstract. The nonequilibrium dynamics of BCS superconductors after excitation with short THz pulses slightly above the superconducting gap is studied theoretically. Calculated pump-probe spectra show how the gap is influenced by this optical excitation. For ultrashort pump pulses a nonadiabatic regime emerges, in which the BCS order parameter exhibits oscillations. These oscillations do not show up in pump-probe spectra. However they may be made visible by using a coherent control type excitation with two phase-locked pump pulses.

1. Introduction
Superconductors have been studied since many years using optical techniques such as absorption or pump-probe spectroscopy [1; 2]. Absorption spectra of conventional superconductors under linear response conditions have successfully been calculated on the basis of the BCS theory [3; 4]. Here we apply this theory to the analysis of the nonequilibrium dynamics induced by the excitation with strong THz pulses. Our calculations are based on the mean-field BCS theory extended by a coupling to the electromagnetic field. By numerically solving the equations of motion both for the normal and the anomalous density matrix elements we thereby obtain information on the temporal evolution of microscopic variables like quasiparticle occupations and coherences and at the same time we can analyze how the microscopic dynamics is reflected in observable quantities like the absorption spectrum detected by a delayed probe pulse [5].

2. Model system
We use the standard BCS model in mean-field approximation with the Hamiltonian given by

\[ H_{BCS} = \sum_{k,\sigma} \varepsilon_k c_{k,\sigma}^\dagger c_{k,\sigma} - \sum_k \left( \Delta c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger + \Delta^* c_{-k\downarrow}^\dagger c_{k\uparrow}^\dagger \right), \]

where \( c_{k,\sigma}^\dagger \) and \( c_{k,\sigma} \) are electron creation and annihilation operators, \( \hbar \mathbf{k} \) and \( \sigma \) denote momentum and spin and \( \varepsilon_k \) is the dispersion relation for quasi-free electrons with effective mass \( m \). The BCS order parameter \( \Delta \) is defined by \( \Delta = \sum_k W_k (c_{-k\downarrow}^\dagger c_{k\uparrow}^\dagger) \). The BCS interaction only affects electrons close to the Fermi level and its strength \( W_k \) is assumed to be constant for these electrons. Under thermal equilibrium conditions \( \Delta \) is real and the energy gap is given by...
2\Delta$, thus \( \Delta \) can be deduced from the experiment. Under nonequilibrium conditions the order parameter becomes a dynamical variable and is in general complex.

For a fixed value of \( \Delta \) the BCS Hamiltonian can be diagonalized by a Bogoliubov transformation [6] introducing new fermionic quasiparticle operators \( a_k^\dagger \) and \( a_k \) according to \( a_k^\dagger = u_k c_k^\dagger - v_k c_{-k} \) and \( a_k = u_k c_k + v_k c_{-k}^\dagger \). The parameters \( u_k \) and \( v_k \) depend on \( \Delta \) and the Hamiltonian (1) then simply reads

\[
H_{\text{BCS}} = \sum_{k, \sigma} E_k a_k^\dagger a_k + \text{const} \quad \text{with} \quad E_k = \sqrt{|\Delta|^2 + (\varepsilon_k - E_F)^2},
\]

\( E_F \) being the Fermi energy. As the transformation depends on \( \Delta \), which as a dynamical variable changes in time, this does not mean that the problem has been rewritten into an easily-solvable diagonal one. However it provides us with a way to better understand the microscopic state of the system [6]. In the electron basis even the ground state is quite complicated and excited states cannot easily be analyzed. In contrast, in the quasiparticle picture the ground state is just the vacuum state and an excitation of the superconductor can be understood as the creation of a pair of quasiparticles. The order parameter in this picture is given by

\[
\Delta = \sum_k W_k [u_k^2 v_k (1 - \langle a^\dagger_{k\downarrow} a_{k\uparrow} \rangle - \langle a^\dagger_{k\uparrow} a_{k\downarrow} \rangle)] + u_k^2 \langle a^\dagger_{k\downarrow} a_{k\uparrow} \rangle - v_k^2 \langle a^\dagger_{k\uparrow} a_{k\downarrow} \rangle].
\]

We immediately see that quasiparticle occupations (normal density matrices) lead to a reduction of the order parameter. In addition, however, the order parameter is also affected by quasiparticle coherences (anomalous density matrices).

To describe the pulse-induced dynamics the BCS Hamiltonian has to be complemented by the coupling to the electromagnetic field, which is done in the standard way for a vector potential in Coulomb gauge [5]. Due to the mean-field approximation we then obtain a closed set of equations of motion for the dynamical variables \( \langle a_{k\sigma}^\dagger a^\dagger_{k'\sigma'} \rangle \) and \( \langle a_{k\sigma} a^\dagger_{k'\sigma'} \rangle \); the equations and further details can be found in Ref. [5].

3. Pulse-induced gap reduction

Based on this model simulations have been performed using material parameters for superconducting aluminum [7], but the effects do not depend qualitatively on the parameters used [5; 8]. Similar calculations have also been made for a model with d-wave symmetry of the order parameter typical for high-\( T_c \) superconductors, and they also yield qualitatively similar results [9]. The THz pulses are of Gaussian shape and centered around 0.4 meV, which is slightly above the gap energy of 0.35 meV.

The pump pulse modifies the order parameter, as can be seen in Fig. 1(a). Here we have plotted \( \lim_{k \to \infty} 2|\Delta(t)| = 2\Delta_{\infty} \) versus the integrated intensity of pump pulses of different full width at half maximum (FWHM). Figure 1(b) shows absorption spectra (real part of the complex conductivity) for a weak probe pulse. The pump-probe delay has been taken to be sufficiently long to avoid a temporal overlap of the pulses. The spectra belong to different pump-pulse widths and intensities as indicated in the plot. The circled numbers link the excitation conditions to the corresponding points in part (a). In all spectra, we observe a well-defined energy gap terminated by a sharp peak. As can be seen by comparing with part (a), the gap visible in the absorption spectra agrees with \( 2\Delta_{\infty} \), which therefore can indeed be interpreted as the energy gap, although the superconductor is not in an equilibrium state. The sharp peak in the spectra is due to a van-Hove singularity in the the density of states at the band edge.

In the quasiparticle picture, a pump pulse creates quasiparticles which results in the reduction of the energy gap. For sufficiently low pulse intensities the number of excited quasiparticles is expected to increase linearly with the intensity. Indeed, the intensity dependence in Fig. 1(a)
starts linearly for all different pulse widths, but the slopes are different. This is because for the shortest pulse the spectral width is so large that a considerable amount of the intensity is below the gap and therefore cannot create quasiparticles. Since for a given value of the integrated intensity the peak intensity for this pulse is quite high compared to the longer pulses, we observe a small quadratic term in the intensity dependence resulting from two-photon processes. The 50 ps pulse, on the other hand, creates quasiparticles with a rather sharp energy distribution. Thus for higher intensities the quasiparticle creation saturates because of Pauli blocking leading to a flattening of the curve. In the pump-probe spectrum corresponding to this pulse Pauli blocking is seen as a hole-burning: the absorption is slightly reduced just below the pump energy of 0.4 meV.

4. The nonadiabatic regime
While for longer pulses the order parameter drops monotonically from its equilibrium value to the final value $\Delta_\infty$, for short pump pulses we observe an oscillation in the time dependence of $|\Delta(t)|$ as shown in Fig. 2(a). The reason is that a long pulse adiabatically creates a quasiparticle occupation corresponding to the instantaneous value of the gap. For short pulses instead a nonadiabatic regime is reached where in addition quasiparticle coherences are excited [8; 10]. If, in the quasiparticle basis corresponding to the instantaneous value of $\Delta$, only an occupation exists, the state is stationary because the Hamiltonian is diagonal. But if quasiparticle coherences exist as well, the state is no longer stationary and the oscillation appears. For an unperturbed mean-field BCS system (i.e., without the coupling to the electromagnetic field) analytical solutions for the temporal evolution after preparation of a nonequilibrium initial state has been found [11]. Here the order parameter takes the form

$$|\Delta(t)| = \Delta_\infty + a t^{-1/2} \cos(2\Delta_\infty t / \hbar + \phi),$$  \hspace{1cm} (4)

where $a$ and $\phi$ depend on the initial conditions. This formula fits very well to our numerical results for the time after the pump pulse.

Surprisingly, however, it turns out that the oscillation of $|\Delta|$ does not show up in pump-probe spectra. The energy gap visible in the absorption spectrum of the probe pulse is independent of the pump-probe delay time and agrees with $2\Delta_\infty$. The reason is that the probe-induced current extends over many periods of the oscillation; the oscillation hence averages out. This leads to the question of how to detect this oscillation in measurable quantities. We have found that this is possible by application of two phase-locked pump pulses [8]: The reaction of the superconductor
Figure 2. (a) Time dependence of $2|\Delta|$ after excitation with a 3 ps pump pulse with different intensities. (b) Long-time value $2\Delta_{\infty}$ after two pump pulses as a function of the time delay between the pulses. Pulse 1 has the intensity as in (a), pulse 2 always the intensity $I_0$.

to the second pump pulse depends on the instantaneous value of $\Delta$ at the time when the second pulse hits the system. The instantaneous value of $\Delta$ after the first pulse thus reflects itself in the final value $\Delta_{\infty}$ after the second pulse, which can be detected by a subsequent probe pulse. This is demonstrated in Fig. 2(b), which shows the the long-time value $\Delta_{\infty}$ after a second pulse versus the delay between the two pump pulses. Here the intensity of the first pump pulse is the same as for the corresponding curve in Fig. 2(a), whereas the second pump pulse in all cases has the same intensity $I_0$. The oscillation of $|\Delta(t)|$ now is clearly visible in $\Delta_{\infty}$ and thus can be measured by a subsequent probe pulse.

5. Conclusion
We have presented numerical simulations of a BCS superconductor driven by short pulses with frequencies slightly above the energy gap. The optical excitation lowers the energy gap, which can be observed in the absorption spectrum of a delayed probe pulse. The fast oscillation of the order parameter caused by short pump pulses, however, is not visible with this technique. Instead it may be revealed in the probe spectra by using a coherent control-type excitation with two phase-locked pump pulses.

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