

Coherent dynamics and pump-probe spectra of BCS superconductors

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The density matrix formalism is employed to calculate pump-probe spectra of BCS superconductors in the coherent regime. Two dynamical regimes, one adiabatic and one nonadiabatic, can be clearly distinguished. In the adiabatic regime, the modulus of the BCS order parameter can be identified with half the gap in the probe spectra. In the nonadiabatic regime, the order parameter oscillates in real time, whereas the gap observed in the spectra reflects its temporal average. The transition between these regimes is accompanied by a qualitative change of the intensity dependence of the dynamical gap renormalization. Furthermore, a hole-burning effect occurs if the spectral shape of the pump pulse is sufficiently sharp. A probe pulse preceding the pump pulse leads to spectral oscillations, and both the gap before and after the pump pulse are visible in the probe spectra.

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I. INTRODUCTION

The nonequilibrium dynamics of superconductors has been of active interest for a long time. In recent years, in particular, the ultrafast response to optical excitations has been studied experimentally.¹⁻⁴ Most profoundly understood from a theoretical viewpoint are BCS superconductors. Traditional approaches for the nonequilibrium dynamics of the latter systems are based either on the time-dependent Ginzburg-Landau theory^{5,6} (TDGL) or the Boltzmann equation^{6,7} (BE). The TDGL is a phenomenological description where the dynamical problem is reduced to tracing a single variable, the order parameter $\Delta(t)$. This procedure is applicable when a local thermal equilibrium compatible with the instantaneous value of $\Delta(t)$ is established on a time scale much shorter than the typical variation of $\Delta(t)$. This requirement restricts the validity of the TDGL to only a few limiting cases. The BE approach goes beyond the TDGL by keeping all quasiparticle distributions as dynamical variables, but coherences are left out. It usually also involves a gradient expansion for the spatial and momentum dependences of the distribution functions and is, thus, applicable only when the latter variations are slow.

Recently, BCS-type systems have been analyzed on short time scales for situations far away from thermal equilibrium. Here, a nonadiabatic regime is reached where it is no longer sufficient to follow only the dynamics of quasiparticle distribution functions. In particular, it is necessary to fully account for the time evolution of anomalous expectation values, i.e., coherences between different quasiparticle states. Indeed, a number of recent papers have been devoted to the derivation of explicit solutions to initial-value problems within the framework of the mean-field approximation.⁸⁻¹¹ The initial-value problems are meant to model a system immediately after a sudden change of parameters, e.g., a fermionic alkali gas where the strength of the pairing interaction can be abruptly readjusted by switching external magnetic fields.

In this paper, we explore the nonadiabatic regime for conventional metallic superconductors driven by ultrashort laser pulses. As the action of the laser is not restricted to a single point in time, we cannot apply the solution to the initial-value problem; we rather use the density matrix formalism to

model the coherent collisionless response of the system. Such a description has been worked out many years ago for the linear response.¹² Here, we extend this description to the nonlinear regime. This approach allows us to fully account for the dynamics of all quasiparticle occupations and coherences within the limits of the mean-field theory.

Driving a system into the regime of nonadiabatic coherent dynamics by ultrafast laser excitations has been routinely done for many systems. This holds not only for systems with long decoherence times like, e.g., atomic systems, but also for systems such as semiconductors with decoherence times on the subpicosecond time scale.¹³ For BCS superconductors, however, such scenarios are widely unexplored, even though typical quasiparticle lifetimes are of the order of nanoseconds.^{14,15} Resonant studies of BCS systems in the coherent regime require sources generating pulses much shorter than these times with carrier frequencies of the order of the superconductor gap, that is, in the terahertz scale. Only recently, ultrashort terahertz sources have become available^{16,17} and, thus, paved the way toward the nonadiabatic regime.

Compared to other materials like semiconductors, such studies for BCS-type systems are interesting because the BCS model has a number of distinct features such as a ground state exhibiting a coherent superposition of states with different numbers of electrons. In view of these unique properties, also the coherent dynamics within the BCS model should be characteristically different from other systems. It is the goal of the present paper to provide a model study revealing the special dynamical behavior of a BCS superconductor driven by ultrafast pulses in the coherent regime. In a first part, we study the reaction of the superconductor to a single laser pulse. We show that by reducing the pulse length, a transition takes place from an adiabatic to a nonadiabatic regime, where the order parameter oscillates and a description only in terms of quasiparticle occupations becomes insufficient. In addition, we calculate pump-probe spectra which are directly observable quantities. Interestingly, the oscillations of the order parameter that occur in the nonadiabatic regime are not reflected by corresponding shifts of the pump-probe spectra. From the pump-probe spectra, the dynamical gap renormalization can be extracted as a function of the pulse intensity. We find that the transition to

the nonadiabatic regime is accompanied by a change in the intensity dependence of the shift. The intensity dependence also deviates characteristically from corresponding results for the dynamical Stark shift in atomic or semiconductor systems.^{18–21}

II. THEORY

A. Mean-field approximation of the BCS model

We begin with the BCS-Hamiltonian^{22,23}

$$H_{\text{BCS}} = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}} c_{\mathbf{k}, \sigma}^{\dagger} c_{\mathbf{k}, \sigma} - W_0 \sum_{\mathbf{k}, \mathbf{k}' \in \mathcal{W}} c_{\mathbf{k}' \uparrow}^{\dagger} c_{-\mathbf{k}' \downarrow}^{\dagger} c_{-\mathbf{k} \downarrow} c_{\mathbf{k} \uparrow}, \quad (1)$$

where $c_{\mathbf{k}, \sigma}^{\dagger}$ and $c_{\mathbf{k}, \sigma}$ are the creation and annihilation operators for electrons with momentum $\hbar \mathbf{k}$ and spin σ , $\varepsilon_{\mathbf{k}} = \hbar^2 \mathbf{k}^2 / 2m - E_F$, m is the effective mass, and E_F is the Fermi energy. The second sum describes the attractive interaction induced by electron-phonon coupling; as usual, the retardation is neglected so that the interaction becomes instantaneous. It is restricted to electrons whose energies deviate from E_F by no more than $\hbar \omega_D$ (ω_D is the Debye frequency), so \mathcal{W} is the set of all k vectors with $|\varepsilon_{\mathbf{k}}| \leq \hbar \omega_D$. We have also adopted the standard BCS assumption that pairing only occurs between electrons with opposite momentum and spin orientation, because in a translationally invariant system these are the dominant terms for the formation of the BCS ground state.^{24,25} W_0 is a positive number that determines the strength of this interaction.

We now apply a mean-field (MF) approximation to the interaction part of the Hamiltonian:²⁶ $ab \approx \langle a \rangle b + a \langle b \rangle - \langle a \rangle \langle b \rangle$, with $a = c_{\mathbf{k}' \uparrow}^{\dagger} c_{-\mathbf{k}' \downarrow}^{\dagger}$ and $b = c_{-\mathbf{k} \downarrow} c_{\mathbf{k} \uparrow}$. Neglecting a constant, this yields

$$H_{\text{MF}} = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}} c_{\mathbf{k}, \sigma}^{\dagger} c_{\mathbf{k}, \sigma} - \Delta \sum_{\mathbf{k} \in \mathcal{W}} c_{\mathbf{k} \uparrow}^{\dagger} c_{-\mathbf{k} \downarrow}^{\dagger} - \Delta^* \sum_{\mathbf{k} \in \mathcal{W}} c_{-\mathbf{k} \downarrow} c_{\mathbf{k} \uparrow}, \quad (2)$$

where $\Delta = W_0 \sum_{\mathbf{k} \in \mathcal{W}} \langle c_{-\mathbf{k} \downarrow} c_{\mathbf{k} \uparrow} \rangle$ is the (complex) mean-field parameter.

We can diagonalize H_{MF} for a fixed Δ with the aid of a Bogoliubov transformation.^{23,27} The order parameter Δ can change with time, so we will call the value for which the transformation is done Δ_1 in order to distinguish it more easily from the current value of the order parameter. This transformation is usually done with $\Delta_1 = \Delta_0$, where Δ_0 is the order parameter in the BCS ground state, but we will later on need to diagonalize H_{MF} with an arbitrary complex order parameter. The transformation is defined by two constants $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$, which are allowed to be complex in order to account for a complex Δ_1 , and by

$$\begin{aligned} \alpha_{\mathbf{k}} &= u_{\mathbf{k}} c_{\mathbf{k} \uparrow} - v_{\mathbf{k}} c_{-\mathbf{k} \downarrow}^{\dagger}, & \alpha_{\mathbf{k}}^{\dagger} &= u_{\mathbf{k}}^* c_{\mathbf{k} \uparrow}^{\dagger} - v_{\mathbf{k}}^* c_{-\mathbf{k} \downarrow}, \\ \beta_{\mathbf{k}} &= u_{\mathbf{k}} c_{-\mathbf{k} \downarrow} + v_{\mathbf{k}} c_{\mathbf{k} \uparrow}^{\dagger}, & \beta_{\mathbf{k}}^{\dagger} &= u_{\mathbf{k}}^* c_{-\mathbf{k} \downarrow}^{\dagger} + v_{\mathbf{k}}^* c_{\mathbf{k} \uparrow}. \end{aligned} \quad (3)$$

This transformation has the important property that as long as the condition $|u_{\mathbf{k}}|^2 + |v_{\mathbf{k}}|^2 = 1$ holds, the new quasiparticle operators $\alpha_{\mathbf{k}}$, $\alpha_{\mathbf{k}}^{\dagger}$, $\beta_{\mathbf{k}}$, and $\beta_{\mathbf{k}}^{\dagger}$ satisfy the fermionic commutation relations just like $c_{\mathbf{k} \uparrow}$, $c_{\mathbf{k} \uparrow}^{\dagger}$, $c_{\mathbf{k} \downarrow}$, and $c_{\mathbf{k} \downarrow}^{\dagger}$. The following

choice of $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ satisfies this condition and, furthermore, diagonalizes H_{MF} :

$$u_{\mathbf{k}} = 0, \quad v_{\mathbf{k}} = 1 \quad \text{if } \varepsilon_{\mathbf{k}} < -\hbar \omega_D$$

$$u_{\mathbf{k}} = 1, \quad v_{\mathbf{k}} = 0 \quad \text{if } \varepsilon_{\mathbf{k}} > \hbar \omega_D$$

$$\left\{ \begin{array}{l} u_{\mathbf{k}} = \sqrt{\frac{1}{2} \left(1 + \frac{\varepsilon_{\mathbf{k}}}{E_{\mathbf{k}}} \right)} \\ v_{\mathbf{k}} = \frac{\Delta_1}{|\Delta_1|} \sqrt{\frac{1}{2} \left(1 - \frac{\varepsilon_{\mathbf{k}}}{E_{\mathbf{k}}} \right)} \end{array} \right\} \quad \text{if } |\varepsilon_{\mathbf{k}}| \leq \hbar \omega_D, \quad (4)$$

with

$$E_{\mathbf{k}} = \sqrt{\varepsilon_{\mathbf{k}}^2 + |\Delta_1|^2}. \quad (5)$$

This choice is not mandatory; in particular, a phase parameter remains free, which we have used to ensure that $u_{\mathbf{k}}$ is always real, whereas $v_{\mathbf{k}}$ is complex whenever Δ_1 is complex. For $\Delta_1 = \Delta_0$, the vacuum state defined by $\alpha_{\mathbf{k}}|0\rangle = \beta_{\mathbf{k}}|0\rangle = 0$ is the BCS ground state.

For $\Delta_1 = \Delta$, this transformation turns H_{MF} into the diagonal form (again, an additive constant is being ignored)

$$H_{\text{MF}} = \sum_{\mathbf{k} \in \mathcal{W}} |\varepsilon_{\mathbf{k}}| (\alpha_{\mathbf{k}}^{\dagger} \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k}}) + \sum_{\mathbf{k} \in \mathcal{W}} E_{\mathbf{k}} (\alpha_{\mathbf{k}}^{\dagger} \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k}}). \quad (6)$$

However, Δ changes with time, and we have to take this into account by one of the following means: First, we can take our quasiparticle basis to be time dependent and include in our equations of motions additional terms which arise through such a time-dependent transformation.²⁸ Second, we can keep the transformation parameter Δ_1 at a fixed value and take into account that H_{MF} is no longer diagonal. Both options are equivalent; here, we will stick to the second one. For an arbitrary value of Δ , H_{MF} reads

$$\begin{aligned} H_{\text{MF}} &= \sum_{\mathbf{k} \in \mathcal{W}} |\varepsilon_{\mathbf{k}}| (\alpha_{\mathbf{k}}^{\dagger} \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k}}) + \sum_{\mathbf{k} \in \mathcal{W}} [R_{\mathbf{k}} (\alpha_{\mathbf{k}}^{\dagger} \alpha_{\mathbf{k}} + \beta_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k}}) \\ &\quad + C_{\mathbf{k}}^* \alpha_{\mathbf{k}}^{\dagger} \beta_{\mathbf{k}}^{\dagger} + C_{\mathbf{k}} \beta_{\mathbf{k}} \alpha_{\mathbf{k}}], \end{aligned} \quad (7a)$$

with

$$R_{\mathbf{k}} = \frac{\varepsilon_{\mathbf{k}}^2 + \text{Re}(\Delta^* \Delta_1)}{E_{\mathbf{k}}} \quad (7b)$$

and

$$C_{\mathbf{k}} = \Delta_1^* \left\{ \frac{\varepsilon_{\mathbf{k}}}{E_{\mathbf{k}}} \left[1 - \text{Re} \left(\frac{\Delta}{\Delta_1} \right) \right] + i \text{Im} \left(\frac{\Delta}{\Delta_1} \right) \right\}, \quad (7c)$$

which, of course, is reduced to the above diagonal form [Eq. (6)] if Δ_1 is equal to Δ .

B. Interaction with an electromagnetic field

The superconductor is exposed to a laser field with wave vector $\mathbf{q}_0 = q_0 \mathbf{e}_x$ and angular frequency $\omega_0 = q_0 c$, which in the Coulomb gauge is described by a transverse vector potential

$$\mathbf{A}_q(t) = \mathbf{A}_0 \exp \left[- \left(\frac{2\sqrt{\ln 2} t}{\tau} \right)^2 \right] (\delta_{q, \mathbf{q}_0} e^{-i\omega_0 t} + \delta_{q, -\mathbf{q}_0} e^{i\omega_0 t}), \quad (8)$$

with amplitude $\mathbf{A}_0 = A_0 \mathbf{e}_y$ and full width at half maximum (FWHM) τ .

This vector potential affects the electrons through the Hamiltonian

$$H_{\text{em}} = \sum_{\mathbf{k}, \mathbf{q}, \sigma} \frac{1}{2m} \left[e\hbar(2\mathbf{k} + \mathbf{q}) \cdot \mathbf{A}_q + e^2 \sum_{\mathbf{q}_i} \mathbf{A}_{\mathbf{q}-\mathbf{q}_i} \cdot \mathbf{A}_{\mathbf{q}_i} \right] c_{\mathbf{k}+\mathbf{q}, \sigma}^\dagger c_{\mathbf{k}, \sigma}. \quad (9)$$

Using Eq. (3) again, this becomes

$$H_{\text{em}} = H_{\text{em}}^{(1)} + H_{\text{em}}^{(2)} + \text{complex number}, \quad (10a)$$

$$\begin{aligned} H_{\text{em}}^{(1)} = & \frac{e\hbar}{2m} \sum_{\mathbf{k}, \mathbf{q}} (2\mathbf{k} + \mathbf{q}) \cdot \mathbf{A}_q [(u_{\mathbf{k}}^* u_{\mathbf{k}+\mathbf{q}} + v_{\mathbf{k}}^* v_{\mathbf{k}+\mathbf{q}}) \alpha_{\mathbf{k}+\mathbf{q}}^\dagger \alpha_{\mathbf{k}} \\ & - (u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{q}}^* + v_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}}^*) \beta_{\mathbf{k}}^\dagger \beta_{\mathbf{k}+\mathbf{q}} + (u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} - u_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}}) \alpha_{\mathbf{k}+\mathbf{q}}^\dagger \beta_{\mathbf{k}}^\dagger \\ & + (u_{\mathbf{k}}^* v_{\mathbf{k}+\mathbf{q}}^* - u_{\mathbf{k}+\mathbf{q}}^* v_{\mathbf{k}}^*) \beta_{\mathbf{k}+\mathbf{q}} \alpha_{\mathbf{k}}], \end{aligned} \quad (10b)$$

$$\begin{aligned} H_{\text{em}}^{(2)} = & \frac{e^2}{2m} \sum_{\mathbf{k}, \mathbf{q}} \sum_{\mathbf{q}_i} \mathbf{A}_{\mathbf{q}-\mathbf{q}_i} \cdot \mathbf{A}_{\mathbf{q}_i} [(u_{\mathbf{k}}^* u_{\mathbf{k}+\mathbf{q}} - v_{\mathbf{k}}^* v_{\mathbf{k}+\mathbf{q}}) \alpha_{\mathbf{k}+\mathbf{q}}^\dagger \alpha_{\mathbf{k}} \\ & + (u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{q}}^* - v_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}}^*) \beta_{\mathbf{k}}^\dagger \beta_{\mathbf{k}+\mathbf{q}} + (u_{\mathbf{k}+\mathbf{q}} v_{\mathbf{k}} + u_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}}) \alpha_{\mathbf{k}+\mathbf{q}}^\dagger \beta_{\mathbf{k}}^\dagger \\ & + (u_{\mathbf{k}}^* v_{\mathbf{k}+\mathbf{q}}^* + u_{\mathbf{k}+\mathbf{q}}^* v_{\mathbf{k}}^*) \beta_{\mathbf{k}+\mathbf{q}} \alpha_{\mathbf{k}}]. \end{aligned} \quad (10c)$$

We are interested in pump-probe spectra, so we need to calculate the electric current density $\mathbf{j}(\mathbf{r}_0, t)$. The spatial Fourier transform of its Fock space operator is

$$\hat{\mathbf{j}}_{\mathbf{q}_0} = \hat{\mathbf{j}}_{\mathbf{q}_0}^{(1)} + \hat{\mathbf{j}}_{\mathbf{q}_0}^{(2)}, \quad (11a)$$

$$\hat{\mathbf{j}}_{\mathbf{q}_0}^{(1)} = \frac{-e\hbar}{2mV} \sum_{\mathbf{k}, \sigma} (2\mathbf{k} + \mathbf{q}_0) c_{\mathbf{k}, \sigma}^\dagger c_{\mathbf{k}+\mathbf{q}_0, \sigma}, \quad (11b)$$

$$\hat{\mathbf{j}}_{\mathbf{q}_0}^{(2)} = -\frac{e^2}{mV} \sum_{\mathbf{k}, \mathbf{q}, \sigma} \mathbf{A}_{\mathbf{q}_0-\mathbf{q}} c_{\mathbf{k}, \sigma}^\dagger c_{\mathbf{k}+\mathbf{q}, \sigma}, \quad (11c)$$

where V is the normalization volume. When calculating the linear spectrum of our probe pulse, we neglect $\hat{\mathbf{j}}_{\mathbf{q}_0}^{(2)}$ as it only entails an offset in the imaginary part $\omega \text{Im} \sigma$ of the spectrum.²⁴

Finally, we transform $\hat{\mathbf{j}}_{\mathbf{q}_0}^{(1)}$ according to Eq. (3):

$$\begin{aligned} \hat{\mathbf{j}}_{\mathbf{q}_0}^{(1)} = & \frac{-e\hbar}{2mV} \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{q}_0) [(u_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}_0} - u_{\mathbf{k}+\mathbf{q}_0} v_{\mathbf{k}}) \alpha_{\mathbf{k}}^\dagger \beta_{\mathbf{k}+\mathbf{q}_0}^\dagger \\ & + (u_{\mathbf{k}+\mathbf{q}_0}^* v_{\mathbf{k}}^* - u_{\mathbf{k}}^* v_{\mathbf{k}+\mathbf{q}_0}^*) \beta_{\mathbf{k}} \alpha_{\mathbf{k}+\mathbf{q}_0} + (u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{q}_0}^* \\ & + v_{\mathbf{k}} v_{\mathbf{k}+\mathbf{q}_0}^*) \alpha_{\mathbf{k}+\mathbf{q}_0}^\dagger \alpha_{\mathbf{k}} - (u_{\mathbf{k}}^* u_{\mathbf{k}+\mathbf{q}_0} + v_{\mathbf{k}}^* v_{\mathbf{k}+\mathbf{q}_0}) \beta_{\mathbf{k}+\mathbf{q}_0}^\dagger \beta_{\mathbf{k}}]. \end{aligned} \quad (12)$$

C. Calculating the superconductor's response

To determine $\hat{\mathbf{j}}_{\mathbf{q}_0}^{(1)}$, it obviously suffices to know the following expectation values:

$$\begin{aligned} & \langle \alpha_{\mathbf{k}}^\dagger \beta_{\mathbf{k}+\mathbf{q}}^\dagger \rangle, \quad \langle \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}+\mathbf{q}} \rangle, \\ & \langle \beta_{\mathbf{k}} \alpha_{\mathbf{k}+\mathbf{q}} \rangle, \quad \langle \beta_{\mathbf{k}+\mathbf{q}}^\dagger \beta_{\mathbf{k}} \rangle. \end{aligned} \quad (13)$$

Starting from Heisenberg's equation of motion with the Hamiltonian $H = H_{\text{MF}} + H_{\text{em}}$ [Eqs. (7a) and (10)], we obtain a set of differential equations for these variables. As an example, the equation of motion for the first variable is shown in the Appendix [Eq. (A1)]. The others have a similar structure. We observe that the equations contain no other expectation values than those in Eq. (13). This means that we have found a closed set of differential equations; that we do not get an infinite hierarchy of equations is a virtue of the mean-field approximation.

There is still an infinite number of \mathbf{k} 's and \mathbf{q} 's. However by looking closely at the equations of motion, we can see that there is no interaction with the electromagnetic field for every term with $\mathbf{k}, \mathbf{k}+\mathbf{q} \notin \mathcal{W}$, as long as we start from the BCS ground state; so we will impose the restriction $\mathbf{k}, \mathbf{k}+\mathbf{q} \in \mathcal{W}$.³⁴

As we can see from Eq. (A1), only expectation values with indices $(\mathbf{k}, \mathbf{k}+n\mathbf{q}_0)$ with an integer n have to be considered. We can further reduce the number of tuples $(\mathbf{k}, \mathbf{k}+n\mathbf{q}_0)$ we have to calculate because $H_{\text{em}}^{(1)}$ couples only tuples with $n_1 - n_2 \equiv \Delta n = \pm 1$ and $H_{\text{em}}^{(2)}$ couples only those with $\Delta n \in \{-2, 0, 2\}$. New excitations can only arise for $n = \pm 1$ through $H_{\text{em}}^{(1)}$ and for $n = -2, 0$, and 2 through $H_{\text{em}}^{(2)}$. Now $H_{\text{em}}^{(1)}$ is proportional to \mathbf{A}_0 and $H_{\text{em}}^{(2)}$ is proportional to \mathbf{A}_0^2 . This means that if \mathbf{A}_0 is small enough, we may ignore all tuples with $n > N$. We have found that it suffices to set $N=4$, which we have done in all the calculations presented herein.

For simplicity, we have computed these equations in two and not in three dimensions. Two-dimensional calculations may be of interest on their own, but they should also provide us with a good approximation for the three-dimensional superconductor for the following reason: As we can see from Eqs. (8) and (10), the laser field couples only indices $(\mathbf{k}, \mathbf{k}+n\mathbf{q}_0)$ and $(\mathbf{k}', \mathbf{k}'+n'\mathbf{q}_0)$ with $\mathbf{k}-\mathbf{k}'=j\mathbf{q}_0$ with an integer j . So, leaving the second index aside for a moment, we have one-dimensional subspaces of the k space, wherein the matrix elements are strongly coupled, but two different subspaces are completely independent of each other except for the weak indirect coupling through the mean-field parameter Δ , which is, of course, dependent on all of these subspaces and vice versa affects all of them. As all of these subspaces are similar to each other, it seems like a good approximation to choose a very coarse discretization in the directions perpendicular to \mathbf{q}_0 or even to reduce the number of dimensions. So we have made our calculations in two dimensions using the rather coarse discretization of 32 points in the direction perpendicular to \mathbf{q}_0 . Using 128 discretization points instead produces very similar results indeed, and even using only one dimension leads to the same qualitative behavior, as will be seen later.

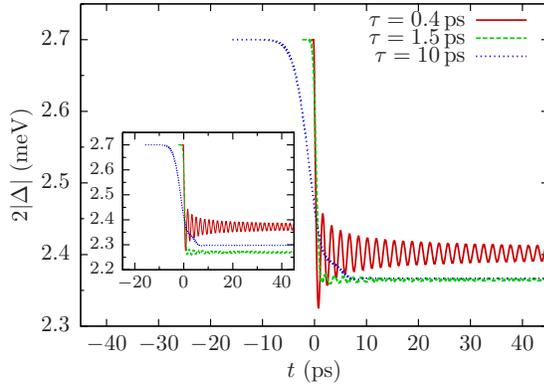


FIG. 1. (Color online) Time dependence of the order parameter $|\Delta|$. All pump pulses are centered at $t=0$ and have the same energy. Note that we have plotted $2|\Delta|$ as this is the value that corresponds to the gap energy. The inset shows the results of the one-dimensional calculation.

III. RESULTS

For our numerical computation we use the following parameters which are motivated by the experimental values for lead.²⁹ $2\Delta_0=2.7$ meV (Δ_0 is the order parameter for the BCS ground state and, therefore, half the energy gap), $\hbar\omega_D=8.3$ meV, $E_F=9.47$ eV, and $m=1.9m_0$ with the free electron mass m_0 . The pump and probe pulse frequencies are $\omega_0=4.50\times 10^{12}$ s⁻¹ and $\omega_p=3.80\times 10^{12}$ s⁻¹, so $\hbar\omega_0=2.96$ meV is greater and $\hbar\omega_p=2.50$ meV is less than the energy gap $2\Delta_0$.

The calculations start from the BCS ground state at zero temperature. Pump and probe pulses are Gaussian in time, but are cut off at a thousandth of their maximum amplitude. The pulses do not overlap.

A. Effects of the pump pulse

Figure 1 shows twice the modulus of the order parameter $|\Delta|$ as a function of time for different widths of the pump pulse. As a consequence of the action of the pump pulse, $|\Delta|$ decreases. We observe an adiabatic and a nonadiabatic regime: If the pulse is sufficiently wide ($\tau=10$ ps), $|\Delta|$ remains constant after the pump pulse has ended. However, for short pump pulses, $|\Delta|$ continues to oscillate even when the pump pulse has been switched off long ago. The amplitude of this oscillation increases with decreasing pulse width. The decrease of $|\Delta|$ is due to the fact that the laser pulse populates the expectation values $\langle\alpha_k^\dagger\alpha_k\rangle$ and $\langle\beta_k^\dagger\beta_k\rangle$, just as an increase in temperature does.

If we change into the instantaneous Bogoliubov space $[\Delta_1=\Delta(t)]$ for any given time t after the pump pulse has expired, the time derivative of these expectation values is zero as H_{MF} is diagonal. In the adiabatic regime, the expectation value $\langle\alpha_k^\dagger\beta_k^\dagger\rangle$ now is almost zero, so $|\Delta|$ stays constant when the pump pulse has subsided. This means that the new state can be described by a quasiparticle distribution without coherences, although the quasiparticles are not the same as in the ground state.

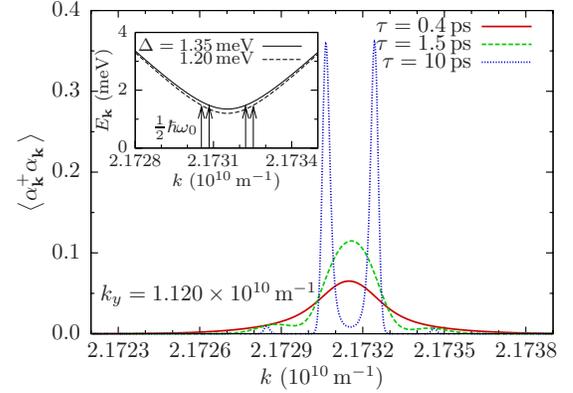


FIG. 2. (Color online) Occupations $\langle\alpha_k^\dagger\alpha_k\rangle$ with $\Delta_1=\Delta(t)$ immediately after the pump pulses. The inset shows how a change in $|\Delta|$ affects the k positions of excitations with a fixed energy.

The short pulse ($\tau=0.4$ ps) lies beyond the adiabatic regime. Setting $\Delta_1=\Delta(t)$ again, the expectation value $\langle\alpha_k^\dagger\beta_k^\dagger\rangle$ is not zero, so a description that takes into account only the quasiparticle distribution is no longer possible. Its time derivative is not zero; as $\langle\alpha_k^\dagger\beta_k^\dagger\rangle$ changes, it affects Δ , and then H_{MF} is no longer diagonal, so $\langle\alpha_k^\dagger\alpha_k\rangle$ and $\langle\beta_k^\dagger\beta_k\rangle$ will also change. This gives rise to the oscillation we observe for the short pulses.

As soon as the pump pulse is switched off, we can regard the time evolution as an initial-value problem as it is discussed in Ref. 11. There it is shown that one possible solution for the time dependence of the modulus of the order parameter is an oscillation with frequency $\omega=2\Delta_\infty/\hbar$ decaying with $1/\sqrt{\Delta_\infty t/\hbar}$, where Δ_∞ is the value $|\Delta(t)|$ asymptotically reaches. Our results are in very good agreement with this prediction.

In Fig. 2, we have plotted the expectation value $\langle\alpha_k^\dagger\alpha_k\rangle$ for a fixed time immediately after the pump pulse. $\langle\beta_k^\dagger\beta_k\rangle$ looks exactly the same. The longest pulse with $\tau=10$ ps creates a population with two pronounced maxima. The positions of these maxima correspond approximately to half the pump pulse energy, $\frac{1}{2}\hbar\omega_0$, because every excitation consists of two Bogoliubovian quasiparticles. The maxima are broadened both by the energy uncertainty of the pump pulse and by the fact that $|\Delta|$ changes while the pump pulse is active. In the inset of Fig. 2, we have sketched how a change in $|\Delta|$ affects the quasiparticle energies and, thereby, shifts the positions of the excitation maxima. The short pulses' energy spectrum is too broad to create such sharp maxima, instead we get a broad population centered at the Fermi wave number k_F . There are also two secondary maxima (e.g., at $k \approx 2.17284 \times 10^{10}$ m⁻¹) which are best visible for the longest pump pulse. These are created by two-photon processes, as can be seen from the fact that they are rather small and that their k values belong to the full energy of the pump pulse.

Performing the same calculation in one dimension on one of the one-dimensional subspaces with $k_y=1.120 \times 10^{10}$ m⁻¹, which is roughly $\frac{1}{2}k_F$, yields quasiparticle distributions which are very similar to those presented above. However, there are some differences in the time evolution of the order

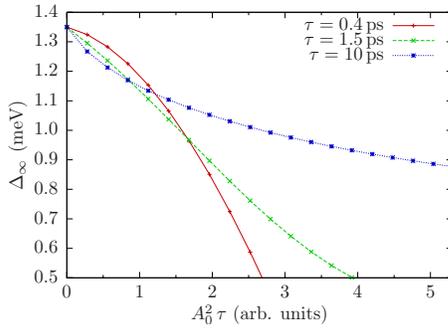


FIG. 3. (Color online) Intensity dependence of the shift in $|\Delta|$. Δ_∞ is the asymptotic value of $|\Delta|$ for long times.

parameter, as can be seen in the inset of Fig. 1. In particular, the shift in the energy gap $2|\Delta|$ is slightly different. Nonetheless, the qualitative behavior is the same. The probe spectra, which will be discussed in the next section, again are virtually independent of whether the number of dimensions is 1 or 2, although, of course, they reflect the changes in the values of $|\Delta|$.

Figure 3 shows the intensity dependence of the shift in $|\Delta|$. The data for this figure have been calculated in only one dimension; as discussed previously, this should be a good approximation for higher dimensional systems. For the longer pulses, the shift starts linearly but then flattens. The flattening occurs because the pump pulses create rather sharp maxima in the quasiparticle distribution, so due to Pauli blocking it gets increasingly difficult to create new quasiparticles with higher intensities. A linear shift is also observed in semiconductors and atoms, e.g., in the dynamical Stark effect.^{18–21} For the shortest pump pulse, the shift is quadratic with a small linear term. This is because shorter pulses need higher intensities to yield a noticeable shift and, with these intensities, the quadratic term of the shift dominates.

B. Probe spectra

In Fig. 4, the probe spectra are shown. The pump pulse precedes the probe pulse, and the pulses do not overlap. The probe pulse has a FWHM of 0.25 ps and is calculated in linear approximation. A Hann window is applied to $j^{(1)}(t)$ prior to Fourier transforming it,³⁰ and the spectrum itself is calculated via $\sigma(\omega) = j^{(1)}(\omega) / E(\omega) = j^{(1)}(\omega) / [i\omega A(\omega)]$.

The first thing we notice about our probe spectra is that both real and imaginary parts have a sharp edge at the same frequency, and the real part is zero below this frequency. This is the energy gap and we expect it to end at $2|\Delta|$, which is correct as long as $|\Delta|$ does not oscillate. If it does oscillate, the edge is located at the average value of $2|\Delta|$; the spectrum does not oscillate itself, i.e., it is independent of the exact timing of the probe pulse, even though the probe pulse width is shorter than one period of the oscillation. So this oscillation cannot be perceived by means of a simple probe spectrum.

The real part of the spectra for the longer pulses additionally exhibits a small dip (cf. inset of Fig. 4). Its corresponding energy is two times the quasiparticle energy of the popu-

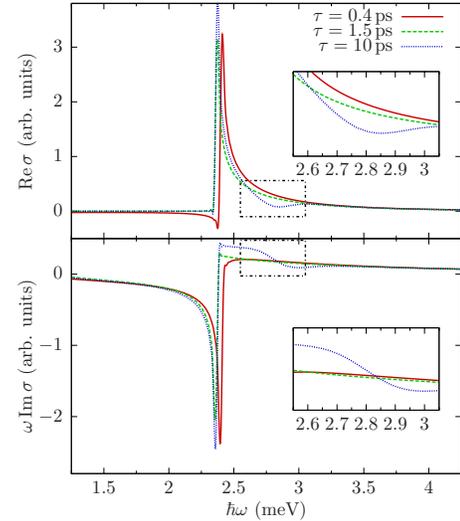


FIG. 4. (Color online) Real and imaginary parts of the probe spectra. The insets magnify the dashed boxes.

lation maxima. This means that the sharp population maxima which the pump pulse has created now inhibit the absorption through Pauli blocking, leading to this hole-burning effect.

Figure 5 shows the spectra for a negative time delay $\delta t = -25$ ps, which means that the probe pulse precedes the pump pulse. We observe a spectral oscillation whose period $\delta\omega = 2\pi / (-\delta t)$ is inversely proportional to the delay δt . Such spectral oscillations are well known in semiconductors where the same relation between $\delta\omega$ and δt applies.^{31–33} In contrast to typical semiconductor results, both the energy gap before and after the pump pulse, indicated by dashed lines, can be seen in the spectra as sharp features. Physically, under these excitation conditions, the pump pulse modifies the dynamics of the current induced by the preceding probe pulse. The main oscillation frequency of the current, which is the gap

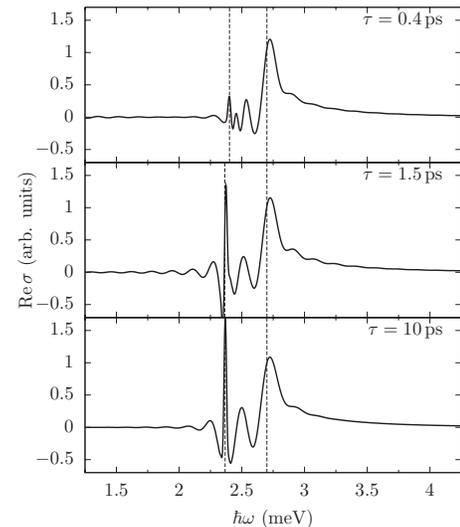


FIG. 5. Real part of probe spectra with negative time delay $\delta t = -25$ ps between pump and probe pulses. The dashed lines show the energy gap before and after the pump pulse.

frequency, changes rather abruptly from the equilibrium value to the reduced value determined by the pump pulse.

IV. CONCLUSION

We have calculated the coherent dynamics of a BCS superconductor using the density matrix formalism. Short pump pulses put the system into a coherent superposition state which can no longer be described by its quasiparticle distribution alone. In this nonadiabatic regime, the modulus of the order parameter performs a damped oscillation. In addition, the intensity dependence of the shift in $|\Delta|$ is very different from the one in the adiabatic regime.

The reaction to the pump pulse may be measured with the aid of pump-probe spectroscopy. One, thereby, gains information about maxima in the quasiparticle distribution and about the modulus of the order parameter, but its oscillation remains invisible. If the probe pulse precedes the pump pulse, spectral oscillations occur and both the gap before and

after the pump pulse are visible in the spectra as sharp features.

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APPENDIX: EQUATIONS OF MOTION

We show the equation of motion for only one of the four expectation values in Eq. (13); the remaining ones are similar. We have used the fact that \mathbf{A}_q is of a special form [see Eq. (8)] and that all u_k are real. R_k and C_k are the same as in Eq. (7a). As discussed previously, we have limited ourselves to the subset of momentum space called \mathcal{W} , where the attractive interaction occurs. The term independent of $\mathbf{A}_q(t)$ is, therefore, valid for $\mathbf{k}, \mathbf{k}+\mathbf{q} \in \mathcal{W}$ only.

$$\begin{aligned}
i\hbar \frac{d}{dt} \langle \alpha_k^\dagger \beta_{k+q}^\dagger \rangle = & -(R_k + R_{k+q}) \langle \alpha_k^\dagger \beta_{k+q}^\dagger \rangle + C_{k+q}^* \langle \alpha_k^\dagger \alpha_{k+q} \rangle + C_k^* (\langle \beta_{k+q}^\dagger \beta_k \rangle - \delta_{q,0}) + \frac{e\hbar}{2m} \sum_{q'=\pm q_0} 2\mathbf{k} \cdot \mathbf{A}_{q'}(t) \\
& \times [-(u_k u_{k+q'} + v_k^* v_{k+q'}) \langle \alpha_{k+q'}^\dagger \beta_{k+q'}^\dagger \rangle + (u_{k+q-q'} u_{k+q} + v_{k+q-q'} v_{k+q}^*) \langle \alpha_k^\dagger \beta_{k+q-q'}^\dagger \rangle \\
& + (u_{k+q-q'} v_{k+q}^* - u_{k+q} v_{k+q-q'}^*) \langle \alpha_k^\dagger \alpha_{k+q-q'} \rangle + (u_k v_{k+q}^* - u_{k+q} v_k^*) (\langle \beta_{k+q}^\dagger \beta_{k+q'} \rangle - \delta_{q,q'})] \\
& + \frac{e^2}{2m} \sum_{q'=0, \pm 2q_0} \left(\sum_{q_i=\pm q_0} \mathbf{A}_{q'-q_i}(t) \cdot \mathbf{A}_{q_i}(t) \right) [-(u_k u_{k+q'} - v_k^* v_{k+q'}) \langle \alpha_{k+q'}^\dagger \beta_{k+q'}^\dagger \rangle \\
& - (u_{k+q} u_{k+q-q'} - v_{k+q}^* v_{k+q-q'}^*) \langle \alpha_k^\dagger \beta_{k+q-q'}^\dagger \rangle + (u_{k+q} v_{k+q-q'}^* + u_{k+q-q'} v_{k+q}^*) \langle \alpha_k^\dagger \alpha_{k+q-q'} \rangle \\
& + (u_k v_{k+q'}^* + u_{k+q} v_k^*) (\langle \beta_{k+q}^\dagger \beta_{k+q'} \rangle - \delta_{q,q'})]. \tag{A1}
\end{aligned}$$

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