Coherent control of the gap dynamics of BCS superconductors in the nonadiabatic regime

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By exciting a BCS superconductor with ultrashort pulses in the frequency range of the superconducting gap a nonadiabatic regime can be reached. In this regime the modulus of the order parameter oscillates in time. Although its average value can be identified with the gap in the absorption spectrum, the oscillation itself remains invisible in pump-probe spectra. In this Brief Report we demonstrate that by employing a coherent control-type scheme of excitation by two phase-locked pump pulses this oscillation can be unveiled in the probe spectrum. We find that the reaction of the superconductor to a second pump pulse depends strongly on its instantaneous state at the time of the impact of the second pulse. Based on numerical calculations performed on the mean-field level it is shown that by varying the delay time between the two pump pulses the transient oscillation of the order parameter can be transformed into an oscillation of its long-time value, which shows up in the absorption spectrum of a subsequent weak probe pulse as an oscillation of the gap when plotted as a function of the delay time.

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On short time scales in situations far away from equilibrium a new regime emerges in BCS-type superconductors, showing fascinating effects such as a rapidly oscillating order parameter.[1–4] This nonadiabatic regime is inaccessible by means of the approximations commonly utilized to calculate the dynamics of superconductors, e.g., the Ginzburg-Landau theory or Boltzmann equation approach.[5] This is due to the fact that in terms of Bogoliubovian quasiparticles, the state of a superconductor in the nonadiabatic regime cannot be described by quasiparticle occupations alone but instead constitutes a coherent superposition state. It is, however, possible to calculate the dynamics in mean-field BCS theory without any further approximations. An exact solution has been found to the initial-value problem imposed by a BCS system which is abruptly put into a nonequilibrium state and from there on develops without any further perturbations.[2,6,7] This solution is applicable to, e.g., fermionic alkali gases, where a nonequilibrium state can be generated by almost instantaneously modifying the BCS pairing interaction. In metallic superconductors such a way of reaching a nonequilibrium state is apparently not feasible. Here it is a common technique to excite the superconductor into a nonequilibrium state by using a short pulse and then analyzing this state with a probe pulse. Such pump-probe experiments have been performed both for conventional and high-T_C superconductors (see, e.g., Refs. 8–11). However, calculations of pump-probe signals of a BCS superconductor have shown that even if such oscillations in the order parameter are excited by a sufficiently short pump pulse, they are invisible in the absorption spectrum of a subsequent weak probe pulse.[4] Essentially, the probe-induced polarization averages over the transient oscillations and thus, independent of the delay time between pump and probe pulse, only the long-time or average value of the gap is seen in the spectra. In this Brief Report we will show that by extending the standard pump-probe scenario to a coherent control-type setup with two phase-locked pump pulses followed by a weak probe pulse, the oscillations can be made visible in the probe spectra because by varying the delay time between the two pump pulses the second pulse translates the transient oscillations of the order parameter into oscillations of the long-time value of this quantity, which then shows up as a gap in the spectra which oscillates as a function of the control delay time.

The optically driven dynamics of a BCS superconductor may be calculated using the density-matrix formalism, as we have done in Ref. 4. In that paper we have studied the dynamics of the superconductor after excitation with a short pulse, and we have calculated pump-probe spectra which reflect the response of the system to a subsequent weak probe pulse. Typically these spectra have the overall shape of the well-known thermal equilibrium spectra as derived for clean superconductors from the BCS Hamiltonian by linear-response theory[12] or for superconductors with impurity scattering for sufficiently high values of the collision time.[13] However, the pump pulse leads to a reduction in the gap energy seen in the spectra and, for sufficiently long pump pulses, to a hole-burning effect due to the pump-generated quasiparticles. Related calculations involving a model of a *d*-wave superconductor have been carried out to describe the relaxation dynamics of high-T_C superconductors.[14] The absorption spectra obtained in these calculations are comparable with our results and indeed they show the same features, i.e., a strong peak revealing the superconducting gap and a hole-burning effect.

In Ref. 4 we have shown that the pulse-induced dynamics in the superconductor strongly depends on the duration of the exciting pulse. For sufficiently long pulses the modulus of the order parameter $|\Delta|$ adiabatically reaches its new steady-state value determined by the distribution of the generated quasiparticles. In this case $2|\Delta|$ can be identified with the superconducting gap which can be measured by the absorption of a subsequent probe pulse. On the other hand, when the superconductor is excited by a sufficiently short laser pulse a nonadiabatic regime is reached. This regime is characterized by a fast oscillation of the order parameter after the pump pulse has finished. We have shown that the precise form of this oscillation found numerically for the optically driven superconductor after the pulse indeed agrees with one

of the scenarios discussed in Ref. 2 on the basis of the exact solution for an initial value problem, i.e.,

$$|\Delta(t)| = \Delta_\infty \left[1 + a \frac{\cos(2\Delta_\infty t/\hbar + \phi)}{\sqrt{\Delta_\infty (t - t_0)/\hbar}} \right], \tag{1}$$

where Δ_∞ is the value $|\Delta(t)|$ asymptotically reaches and a, ϕ, and t_0 depend on the excitation conditions.

In our previous calculations we have found that when going from long to short pump pulses the intensity dependence of the gap parameter Δ_∞ changes, the oscillation of the gap parameter in the nonadiabatic regime, however, fails to be perceived in pump-probe spectra.[4] Although an energy gap is clearly visible in the absorption spectra, it does not change when varying the pump-probe delay time. Instead, it always stays in the same place regardless of the delay time. The gap measured in the spectra corresponds to the temporal average of $2|\Delta(t)|$ or, in other words, it agrees with $2\Delta_\infty$.

In this Brief Report we discuss an approach to unveil the oscillation of $|\Delta|$. Instead of using a single pump pulse we will consider here an excitation by a pair of phase-locked pump pulses. Such coherent control-type techniques employing two-pulse excitations have been widely used for the study of coherent aspects in the ultrafast dynamics of semiconductors such as the coherent control of exciton density[15] and spin.[16] More important for our present purpose, they have also proven to be well suited to analyze and control a variety of phenomena involving pump-induced oscillations in the carrier dynamics such as quantum beats in coupled quantum wells[17] and the resulting terahertz emission from these structures,[18] quantum beats between heavy hole and light hole excitons[15,19] and phonon quantum beats.[19–21] In those studies it has been found that a second phase-locked pulse has a strong influence on the oscillations and that this influence can be made visible, e.g., in pump-probe or four-wave-mixing signals employing a third pulse. Here we will show that also in the present case of the nonadiabatic dynamics of the order parameter the reaction of the superconductor to a second pump pulse is highly susceptible to the instantaneous state of the superconductor and in particular to the instantaneous value of $|\Delta|$. Hence, by making use of a two-pulse coherent control technique, the oscillation of the order parameter may be observed in measurable quantities.

Our calculations are based on the same model discussed in Ref. 4. We start from the standard BCS Hamiltonian in mean-field approximation coupled to an external electromagnetic field, $H = H_{\mathrm{BCS}} + H_{\mathrm{em}}$, where

$$H_{\mathrm{BCS}} = \sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}} c^+_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} - \Delta \sum_{\mathbf{k}\in\mathcal{W}} c^+_{\mathbf{k}\uparrow} c^+_{-\mathbf{k}\downarrow} - \Delta^* \sum_{\mathbf{k}\in\mathcal{W}} c_{-\mathbf{k}\downarrow} c_{\mathbf{k}\uparrow} \tag{2}$$

and

$$H_{\mathrm{em}} = \sum_{\mathbf{k},\mathbf{q},\sigma} \frac{1}{2m} \left[e\hbar(2\mathbf{k} + \mathbf{q}) \cdot \mathbf{A}_{\mathbf{q}}(t) + e^2 \sum_{\mathbf{q}'} \mathbf{A}_{\mathbf{q}-\mathbf{q}'}(t) \cdot \mathbf{A}_{\mathbf{q}'}(t) \right] c^+_{\mathbf{k}+\mathbf{q},\sigma} c_{\mathbf{k},\sigma}. \tag{3}$$

Here, $c^+_{\mathbf{k},\sigma}$ and $c_{\mathbf{k},\sigma}$ denote 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. $\Delta = W_0 \Sigma_{\mathbf{k}\in\mathcal{W}} \langle c_{-\mathbf{k}\downarrow} c_{\mathbf{k}\uparrow} \rangle$ is the (complex) order parameter, where W_0 is a positive number that determines the strength of the phonon-mediated attractive interaction and $\mathcal{W}$ is the set of all k vectors with $|\varepsilon_{\mathbf{k}}| \leq \hbar\omega_D$, with ω_D being the Debye frequency. The electromagnetic field is treated in the Coulomb gauge and given by the transverse vector potential $\mathbf{A}_{\mathbf{q}}(t)$ with wave vector $\mathbf{q}$. All pulses have a Gaussian time dependence. For the calculations a Bogoliubov transformation is performed and the resulting equations of motion for the density-matrix elements of the quasiparticles are solved numerically. All results presented in this Brief Report have been calculated using parameters reflecting the experimental values for lead. We have used quasi-one-dimensional calculations, which have been shown to provide a good approximation for two- and three-dimensional systems. Further details of the calculations can be found in Ref. 4.

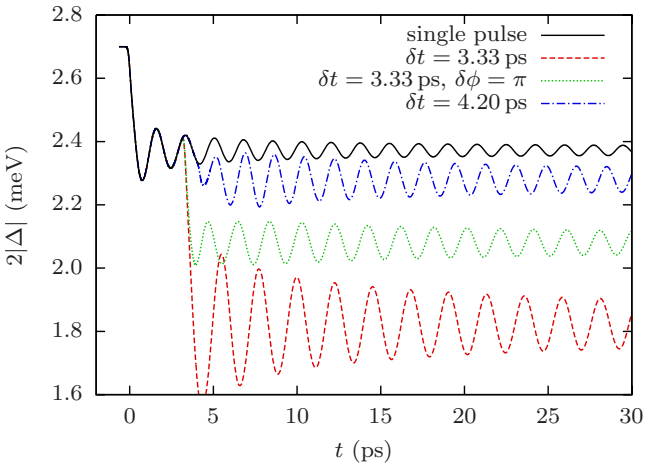

FIG. 1. (Color online) Time dependence of the modulus of the order parameter for a single pump pulse (solid line) and two phase-locked pump pulses with different delay times and relative phases (other lines).

Figure 1 shows the time dependence of the modulus of the order parameter for different excitation conditions. A single pump pulse with a full width at half maximum of 0.4 ps and a central energy slightly above the gap energy produces a damped oscillation (solid line) which after the pulse can be described by Eq. (1) by fitting suitable values of a, ϕ, and t_0. The other lines show the cases where a second pump pulse identical to the first one has been applied. The peak of the second pulse is at the time when the solid line has a maximum (dashed and dotted lines) or a minimum (dashed-dotted line). For the dashed and dashed-dotted lines, the second pulse has the same phase as the first pulse, and for the dotted line the phase is shifted by π. Evidently, both the time delay and the relative phase of the two pump pulses have a strong influence on the dynamics of the order parameter. They do not only modify the amplitude and frequency of the oscillations, but they also affect the long-time value Δ_∞.

From our previous studies[4] we know that in the case of a single pump pulse $2\Delta_\infty$ reflects itself as the superconducting gap in the absorption spectrum of a subsequent probe pulse. That this also holds in the present case can be seen in Fig. 2,

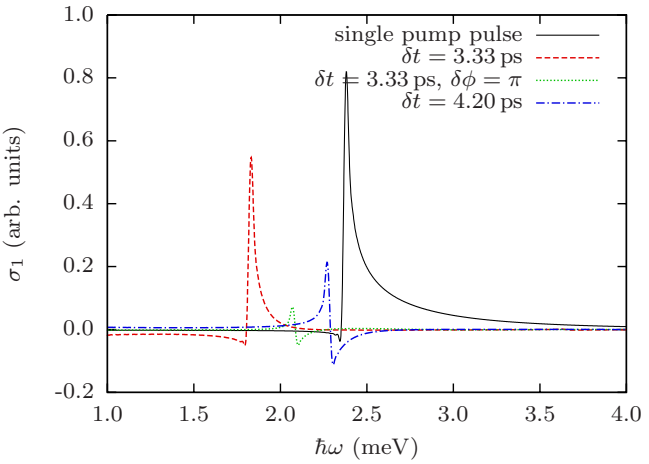

FIG. 2. (Color online) Absorption spectra of a probe pulse after the excitation with a single pump pulse (solid line) and with two phase-locked pump pulses with different delay times and phases (other lines). The delay time between the last pump pulse and the probe pulse is always 1.42 ps.

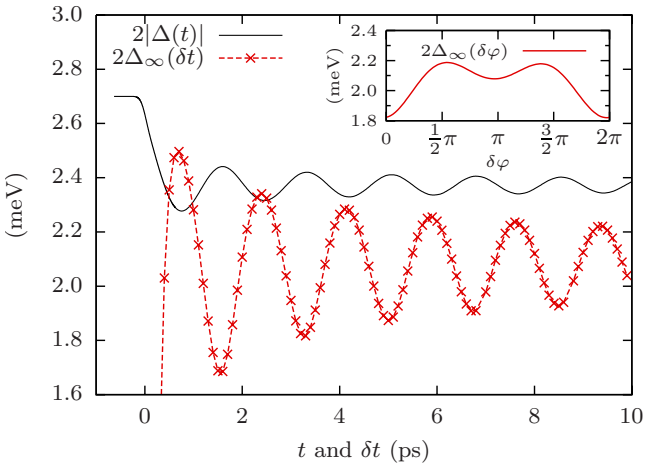

FIG. 3. (Color online) Order parameter $2|\Delta|$ as a function of the time for the case of excitation by a single pump pulse (solid line) and $2\Delta_\infty$ after a second pulse with varying delay time and fixed phase $\delta\varphi=0$ (crosses). The dotted line connects the crosses and is meant as a guide for the eyes. In the inset the dependence on the phase difference between the two pulses is shown for a fixed delay time of 3.33 ps.

where the real part σ_1 of the complex conductivity is shown for the four curves discussed above. At the value of $2\Delta_\infty$ all spectra possess a distinctive peak. It therefore seems reasonable that the oscillation of the order parameter after the first pump pulse may be made visible by measuring Δ_∞ after a second pump pulse for varying delay times between these pump pulses.

From Fig. 2 we notice that the detailed shape of σ_1 may be different for different parameters. In fact, it turns out that the shape depends on the probe delay time. This is because in the response of a time-dependent system in general there is no clear separation between absorptive features in the real part and dispersive features in the imaginary part of the conductivity such as in the case of thermal equilibrium spectra. Instead, both real and imaginary parts may contain absorptive and dispersive contributions, the relative weight between those contributions depending on the detailed dynamical state of the system at the arrival time of the probe pulse. The important point, however, is the fact that the position of the peak does not depend on the probe delay time and indeed agrees with $2\Delta_\infty$. To be specific, for the data shown we have set the delay between the second pump pulse and the probe pulse to a fixed value of 1.42 ps.

In Fig. 3 this approach to measure the oscillation is simulated. As above, the solid line shows the oscillation after a single pulse. Every cross marks the value of $2\Delta_\infty$ (determined as the temporal average $2|\Delta|$) after a second pulse which hits the system after the delay time indicated by the x coordinate. The phase of the second pulse is the same as for the first pulse. The oscillation of $|\Delta|$ in the single-pulse solution is clearly reproduced by Δ_∞ in the two-pulse case although it is swapped upside down and slightly shifted. The mirroring effect is also visible in Fig. 1, where the second pulse hitting at a maximum of the oscillation produces a lower average value than the pulse hitting at a minimum.

Typically the value of Δ_∞ after the second pulse is smaller than its value after the first pulse because the second pulse creates additional quasiparticles. Interestingly, however, Fig. 3 reveals that for short delay times (within the first oscillation period) it is even possible to enhance the value of Δ_∞ by a second pulse, which means that here we have a coherent destruction of quasiparticles that have been generated by the first pulse. For longer times such a coherent destruction is no more possible due to the broad energy distribution of the quasiparticles (see, e.g., Fig. 2 in Ref. 4) which means that these quasiparticles quickly run out of phase.

In the inset of Fig. 3, the dependence on the phase of the second pulse is shown for a fixed delay time of $\delta t = 3.33$ ps. This dependence varies for different delay times, as the dependencies on phase and on delay time interlock with each other. However, for any fixed value of the phase the delay time dependence of Δ_∞ reproduces the real-time oscillations in $|\Delta|$.

We have recently become aware of a paper discussing the stability of states with an oscillating order parameter against parametric excitation of spatial fluctuations.[22] As our model implies an essentially homogeneous system, such fluctuations are not accounted for. However, these inhomogeneities need some time to develop; the linear stability analysis provided in Ref. 22 for the case of undamped oscillations of the order parameter shows that the maximum growth rate of the fluctuations is much smaller than the frequency of the oscillations. For damped oscillations as in our case it can be expected that the growth rate is even smaller. In our scenario we only require that the homogeneous oscillatory solution created by the first pump pulse persists up to the second pump pulse, which thus should be satisfied in the range of delay times shown in Fig. 3.

In conclusion, we have simulated the response of a BCS superconductor to a sequence of two phase-locked pump pulses with varying delay time and phase difference. By an excitation with sufficiently short pulses the superconductor is driven into a nonadiabatic regime exhibiting a temporally oscillating order parameter. While after a single-pulse exci-

tation these oscillations remain invisible in pump-probe spectra as a function of the pump-probe delay, we have found that after a two-pulse excitation they can be unveiled by extracting the superconducting gap from such spectra as a function of the delay time between the two pump pulses. In addition we have shown how the response is affected by the relative phase between the two pump pulses. Our results therefore demonstrate that two-pulse coherent control experiments with a pump-probe-type detection scheme are indeed well suited to observe temporal oscillations in the order parameter of a BCS superconductor which is driven in the nonadiabatic regime.

Most of our calculations have been carried out on a grid managed by software from the Condor Project by the University of Wisconsin-Madison.

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