Rapid Communications

Collective modes in pumped unconventional superconductors with competing ground states

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Motivated by the recent development of terahertz pump-probe experiments, we investigate the short-time dynamics in superconductors with multiple attractive pairing channels. Studying a single-band square lattice model with a spin-spin interaction as an example, we find the signatures of collective excitations of the pairing symmetries (known as Bardasis-Schrieffer modes) as well as the order parameter amplitude (Higgs mode) in the short-time dynamics of the spectral gap and quasiparticle distribution after an excitation by a pump pulse. We show that the polarization and intensity of the pulse can be used to control the symmetry of the nonequilibrium state as well as frequencies and relative intensities of the contributions of different collective modes. We find particularly strong signatures of the Bardasis-Schrieffer mode in the dynamics of the quasiparticle distribution function. Our work shows the potential of modern ultrafast experiments to address the collective excitations in unconventional superconductors and highlights the importance of subdominant interactions for the nonequilibrium dynamics in these systems.

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Recently, ultrafast pump-probe techniques became a powerful tool to probe the temporal evolution of symmetry broken states and relaxation in conventional and unconventional superconductors [1–12]. An intense pulse couples nonlinearly to the Cooper pairs of the superconductor and, as was argued theoretically, should lead to a coherent excitation of the Higgs amplitude mode, i.e., $|\Delta(t)|$ performs a damped oscillation with frequency $\omega_H = 2|\Delta(\infty)|$ [13–24]. Nonlinear terahertz spectroscopy has enabled the observation of the Higgs mode in conventional superconductors in the form of a free or forced oscillation and the resulting third-harmonic generation [4,8,11]. Interestingly, this technique has been also recently applied to the unconventional superconductors such as high-T_c cuprates with the d-wave symmetry of the superconducting gap [12,25] where some additional oscillations have been reported [25].

In contrast to conventional superconductors, where the pairing is driven by the attractive electron-phonon interaction, the pairing interaction in unconventional superconductors is most likely of a repulsive nature. To overcome the net repulsion among the quasiparticles, the superconducting gap has to change its sign across different parts of the Fermi surface, which typically yields the superconducting gap of a lower symmetry than an isotropic s-wave. For example, it is generally known that the antiferromagnetic spin fluctuations peaking near the wave vector $\mathbf{Q}_{\mathrm{AF}} = (\pi,\pi)$ within a single-band model on a square lattice give rise to a $d_{x^2-y^2}$-wave symmetry of the superconducting gap, yet states having other symmetries, such as strongly anisotropic sign-changing (extended) s-wave symmetry and the d_{xy} symmetry, are closely competing. As a result, the temporal dynamics of single-band unconventional superconductors might be significantly richer than that of the conventional ones [15,16,19,22].

In this Rapid Communication we analyze the short-time dynamics in a single-band unconventional superconductor with multiple competing pairing symmetries. In particular, we consider a single-band model of fermions on a square lattice interacting via a spin-spin interaction. The interaction can be decoupled into various pairing channels with different symmetry. Varying the band filling we find two competing even-parity superconducting states forming a typical phase diagram of an unconventional superconductor where different ground states can be accessed by doping. Studying the system driven out of equilibrium by a laser pulse, we show how the collective signatures of symmetries different from a given ground-state symmetry, known as Bardasis-Schrieffer modes [26–29] in the context of an s-wave ground state in the equilibrium, evolve as a function of doping and the polarization direction. Depending on the polarization direction of the incoming light the tetragonal symmetry is broken, which necessarily leads to a mixing of s- and d-wave symmetries in nonequilibrium. Furthermore, we show that the particle distribution acquires an additional $d_{x^2-y^2}$ character and oscillates dominantly with the Bardasis-Schrieffer mode frequency, which may be observed in time-resolved angle-resolved photoemission spectroscopy (tr-ARPES) experiments. Our study highlights the important role of subdominant pairing states in the short-time dynamics of unconventional superconductors and we identify the signatures of the resulting collective modes that can be observed in future pump-probe experiments.

Our starting point is a model of fermions interacting via a spin-spin interaction,

$$H = -t\sum_{\langle ij\rangle,\sigma} c^\dagger_{i\sigma}c_{j\sigma} + J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j, \tag{1}$$

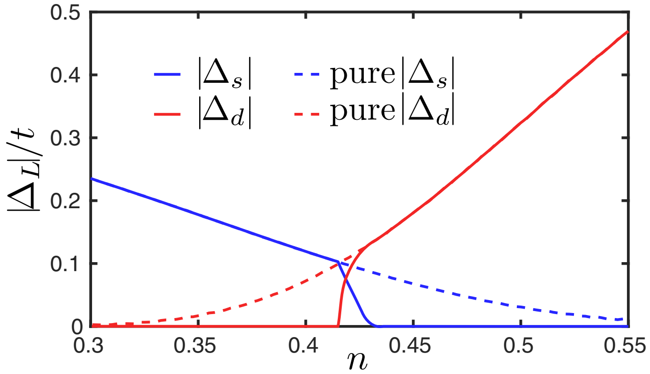

FIG. 1. Phase diagram of $H_{\rm MF}$ at $T = 0$ for the fillings where the $d_{x^2-y^2}$-wave order parameter closely competes with that of the extended s-wave symmetry. The solid lines refer to the actual ground-state values for Δ_s and $\Delta_{d_{x^2-y^2}}$, while the dashed lines show the behavior of a pure s-wave or pure $d_{x^2-y^2}$-wave solution, ignoring the possibility of coexistence.

where $c^{(\dagger)}_{i,\sigma}$ are fermionic annihilation (creation) operators on site i and spin σ, t is the hopping integral between nearest neighbors, and $S^\alpha_i = \frac{1}{2}\sum_{s,s'} c^\dagger_{is}\sigma^\alpha_{ss'}c_{is'}$ are the spin-1/2 operators. This model was considered before in the context of cuprates, pnictides, and heavy fermion systems [30]. In the momentum space the tight-binding energy dispersion is given by $\xi_{\mathbf{k}} = -2t[\cos(k_x) + \cos(k_y)] - \mu$. The spin-spin interaction can also be transformed into momentum space and decoupled into a number of superconducting channels,

$$J_{\mathbf{k},\mathbf{k}'} \equiv V_s\gamma_{\mathbf{k},s}\gamma_{\mathbf{k}',s} + V_{d_{x^2-y^2}}\gamma_{\mathbf{k},d_{x^2-y^2}}\gamma_{\mathbf{k}',d_{x^2-y^2}} + V_{p_x}\gamma_{\mathbf{k},p_x}\gamma_{\mathbf{k}',p_x} + V_{p_y}\gamma_{\mathbf{k},p_y}\gamma_{\mathbf{k}',p_y}, \tag{2}$$

where $V_s = V_{d_{x^2-y^2}} = -3J/2$ is the even-parity spin-singlet interaction with $\gamma_{\mathbf{k},s} = [\cos(k_x) + \cos(k_y)]/2$ and $\gamma_{\mathbf{k},d_{x^2-y^2}} = [\cos(k_x) - \cos(k_y)]/2$ form factors, respectively. The remnant components are repulsive odd-parity spin-triplet coupling constants $V_{p_x} = V_{p_y} = J/2$ with the p_x- and p_y-wave form factors $\gamma_{\mathbf{k},p_{x(y)}} = \sin(k_{x(y)})$. As we show below, the light pulse we consider does not couple the odd- and even-parity channels. As we focus on the regime, where the ground state has even parity, the odd- and even-parity solutions cannot mix both in and out of equilibrium. In the mean-field approximation the Hamiltonian reduces to

$$H_{\rm MF} \simeq \sum_{\mathbf{k}\sigma}\xi_{\mathbf{k}}c^\dagger_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma} + \sum_{\mathbf{k},l}[\Delta_l\gamma_{\mathbf{k},l}c^\dagger_{\mathbf{k}\uparrow}c^\dagger_{-\mathbf{k}\downarrow} + \text{H.c.}], \tag{3}$$

where $l = s, d_{x^2-y^2}$ and the $\Delta_l = -V_l\sum_{\mathbf{k}}\gamma_{\mathbf{k},l}\langle c_{-\mathbf{k}\downarrow}c_{\mathbf{k}\uparrow}\rangle$ are the s- and $d_{x^2-y^2}$-wave component of the total superconducting order parameter $\Delta_{\mathbf{k}} = \Delta_s\gamma_{\mathbf{k},s} + \Delta_{d_{x^2-y^2}}\gamma_{\mathbf{k},d_{x^2-y^2}}$. In what follows, we work at $T = 0$. Minimizing the energy, we obtain the equilibrium values of the superconducting order parameters Δ_s, $\Delta_{d_{x^2-y^2}}$ as a function of n, shown in Fig. 1. We concentrate on the region of the phase diagram where s-wave and d-wave symmetries are neighbors, but avoid the regime of coexistence of both order parameters, i.e., a possible $s + id$ state, as this state (and the similar case of $d + id$ state) and its nonequilibrium dynamics were discussed previously [27,31,32]. Instead, we focus on pairing fluctuations of the subdominant symmetry close to the transition points. To simplify further calculations, we introduce the Anderson pseudospin notation [33]

$$\mathbf{s}_{\mathbf{k}} = \frac{1}{2}(c^\dagger_{\mathbf{k}\uparrow}, c_{-\mathbf{k}\downarrow})\boldsymbol{\sigma}\begin{pmatrix} c_{\mathbf{k}\uparrow} \\ c^\dagger_{-\mathbf{k}\downarrow}\end{pmatrix}, \tag{4}$$

where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)^T$ are the Pauli matrices. Using this vector, one can recast the Hamiltonian $H = \sum_{\mathbf{k}}\mathbf{B}_{\mathbf{k}}\cdot\mathbf{s}_{\mathbf{k}}$, which has the form of a set of (pseudo)spins $\mathbf{s}_{\mathbf{k}}$ coupled to a (pseudo)magnetic field $\mathbf{B}_{\mathbf{k}} = (2\Delta'_{\mathbf{k}}, 2\Delta''_{\mathbf{k}}, 2\xi_{\mathbf{k}})^T$, with the notation $\Delta_{\mathbf{k}} = \Delta'_{\mathbf{k}} - i\Delta''_{\mathbf{k}}$. At zero temperature the thermal expectation values of the pseudospin components are given by $\langle s^x_{\mathbf{k}}\rangle = -\frac{\Delta'_{\mathbf{k}}}{2E_{\mathbf{k}}}$, $\langle s^y_{\mathbf{k}}\rangle = -\frac{\Delta''_{\mathbf{k}}}{2E_{\mathbf{k}}}$, and $\langle s^z_{\mathbf{k}}\rangle = -\frac{\xi_{\mathbf{k}}}{2E_{\mathbf{k}}}$ with the quasiparticle energy dispersion $E_{\mathbf{k}} = \sqrt{\xi^2_{\mathbf{k}} + |\Delta_{\mathbf{k}}|^2}$.

To investigate the collective modes in our model, we study the equations of motion for the pseudospin expectation values $\mathbf{s}_{\mathbf{k}}$ that have the form of Bloch equations,

$$\frac{d}{dt}\langle\mathbf{s}_{\mathbf{k}}\rangle = \mathbf{B}_{\mathbf{k}}\times\langle\mathbf{s}_{\mathbf{k}}\rangle. \tag{5}$$

This equation, together with the self-consistency equation for the superconducting gaps $\Delta_{s,d}(t) = -\sum_{\mathbf{k}}V_{s,d}\gamma_{\mathbf{k},s,d}[\langle s^x_{\mathbf{k}}\rangle(t) - i\langle s^y_{\mathbf{k}}\rangle(t)]$, yields a closed set of coupled differential equations, which defines the temporal evolution of all relevant quantities. To drive the system out of equilibrium we model the electric field of a laser pulse by including a time-dependent vector potential $\mathbf{A}(t)$ via the Peierls substitution. This results in $\mathbf{B}_{\mathbf{k}} = (2\Delta'_{\mathbf{k}}, 2\Delta''_{\mathbf{k}}, \xi_{\mathbf{k}+\frac{e}{c}\mathbf{A}} + \xi_{\mathbf{k}-\frac{e}{c}\mathbf{A}})^T$ [34].

To consider the possibility of exciting order parameter symmetries, different from the ground-state one, it is instructive to split the equations of motion in Eq. (5) into different symmetry channels. As the pulse temporarily breaks C_4-rotational symmetry, different symmetry representations may mix. In particular, we can decompose the pseudomagnetic field into all even-parity irreducible representation for the tetragonal D_{4h} symmetry as follows, $\mathbf{B}_{\mathbf{k}} = \mathbf{B}_{\mathbf{k},s} + \mathbf{B}_{\mathbf{k},d_{x^2-y^2}} + \mathbf{B}_{\mathbf{k},d_{xy}} + \mathbf{B}_{\mathbf{k},g_{xy(x^2-y^2)}}$, where we define $\mathbf{B}_{\mathbf{k},l} = (2\Delta'_l\gamma_{\mathbf{k},l}, 2\Delta''_l\gamma_{\mathbf{k},l}, \xi_{\mathbf{k},\mathbf{A},l})^T$, where we have introduced the symmetrized notations for $B^z_{\mathbf{k}} = \xi_{\mathbf{k}+\frac{e}{c}\mathbf{A}} + \xi_{\mathbf{k}-\frac{e}{c}\mathbf{A}} \equiv \xi_{\mathbf{k},\mathbf{A},s} + \xi_{\mathbf{k},\mathbf{A},d_{x^2-y^2}} + \xi_{\mathbf{k},\mathbf{A},d_{xy}} + \xi_{\mathbf{k},\mathbf{A},g_{xy(x^2-y^2)}}$. There is no odd-parity component of the pseudomagnetic field $\mathbf{B}_{\mathbf{k}}$, as can be seen immediately from the definition above.

Let us now discuss the symmetry mixing in nonequilibrium. Denoting the ground-state symmetry as l_0, one finds that immediately after the perturbation $\frac{d}{dt}\langle s^x_{\mathbf{k},l'}\rangle = 0$ and $\frac{d}{dt}\langle s^y_{\mathbf{k},l'}\rangle = B^z_{\mathbf{k},l}\langle s^x_{\mathbf{k},l_0}\rangle$, where $l' = l\otimes l_0$ is the symmetry of a product of functions with symmetries l and l_0. Therefore, a finite l-symmetry component of the $\mathbf{B}_{\mathbf{k}}$ field is needed to induce pairing correlations of symmetry l'. Moreover, the self-consistency equations imply that inducing an out-of equilibrium order parameter $\Delta_{l'}$ additionally requires a nonzero $V_{l'}$, making the consideration of the subdominant pairing interaction crucial.

In our case, the pseudospin expectation values $\langle\mathbf{s}_{\mathbf{k}}\rangle$ at $t = 0$ are the equilibrium values, with s- or $d_{x^2-y^2}$-wave symmetry. Consequently, one requires $B^z_{\mathbf{k},d_{x^2-y^2}} = \xi_{\mathbf{k},\mathbf{A},d_{x^2-y^2}}$ to be finite at nonzero t to induce a finite $d_{x^2-y^2}$-wave order parameter in the s-wave ground state and vice versa. In particular, a finite $\xi_{\mathbf{k},d_{x^2-y^2}} \sim \cos(A_x) - \cos(A_y)$ [34] can be induced by a vector potential $\mathbf{A}$ with polarization at $\phi \approx 0$, where ϕ is the polar angle in momentum space. Importantly, the effect manifestly depends on the polarization of $\mathbf{A}$, suggesting a possibility to control the induced order parameter symmetry in nonequilibrium. As $\xi_{\mathbf{k},\mathbf{A},d_{xy}}$, $\xi_{\mathbf{k},\mathbf{A},g_{xy(x^2-y^2)}}$ and the odd-parity components are identically zero, only s and $d_{x^2-y^2}$ components remain in Eq. (5), leading to

$$\frac{d}{dt}\langle \mathbf{s}_{\mathbf{k},s}\rangle = \mathbf{B}_{\mathbf{k},s} \times \langle \mathbf{s}_{\mathbf{k},s}\rangle + \mathbf{B}_{\mathbf{k},d_{x^2-y^2}} \times \langle \mathbf{s}_{\mathbf{k},d_{x^2-y^2}}\rangle,$$
$$\frac{d}{dt}\langle \mathbf{s}_{\mathbf{k},d_{x^2-y^2}}\rangle = \mathbf{B}_{\mathbf{k},d_{x^2-y^2}} \times \langle \mathbf{s}_{\mathbf{k},s}\rangle + \mathbf{B}_{\mathbf{k},s} \times \langle \mathbf{s}_{\mathbf{k},d_{x^2-y^2}}\rangle. \quad (6)$$

We note that this result does depend on the absence of spatial variations of $\mathbf{A}$. However, for THz light used in the experiments [35], the wavelength is of the order $\sim$100 μm, which is much larger than, e.g., a superconducting coherence length that rarely exceeds $\sim$100 nm (as estimated by the upper critical field H_{c2} [36–38]), justifying the assumption. Yet spatial variations and its pulse polarization were discussed in Ref. [39] for a pure d-wave superconductor.

We discuss now the short-time dynamics described by Eqs. (6). We integrate the equations numerically using the Runge-Kutta method with a momentum grid of 513×513 points for fixed $V_s = V_d = -0.4t$ varying n and the polarization of $\mathbf{A}(t)$. Let us consider first the situation of an extended s-wave ground-state symmetry for $\Delta_\mathbf{k}$, i.e., $\Delta_s \neq 0$ and $\Delta_{d_{x^2-y^2}} = 0$ at $n = 0.411$. The transition into an $s + id_{x^2-y^2}$ state occurs at the filling $n \approx 0.415$. We start the analysis by driving the model out of equilibrium with a vector potential in the k_x direction. The vector potential $\mathbf{A}$ is for simplicity chosen as $\mathbf{A} = \mathbf{A}_0\theta(t-\tau)\theta(2\tau - t)$ simulating two pulses at $t = \tau$ and $t = 2\tau$. Here, we choose $\frac{e}{c}|\mathbf{A}_0| = 0.01$ and $\tau\Delta = 0.05$. Typically, a Gaussian pulse shape is used in similar approaches [19], with a half duration $t_{1/2} \ll \Delta^{-1}$ to work in the nonadiabatic regime. Our choice of the pulse is equivalent to this approach in the nonadiabatic limit, but we found it to be advantageous for computations. The results of the numerical calculations are shown in Fig. 2. As one can see, the vector potential (pump pulse) induces a nonzero $d_{x^2-y^2}$-wave order parameter $\Delta_{d_{x^2-y^2}}$ in the extended s-wave ground state, which shows undamped oscillations at a frequency ω_{BS}. Note that $|\Delta_{d_{x^2-y^2}}|$ shown in Fig. 2 oscillates at twice the frequency of $\Delta_{d_{x^2-y^2}}$ due to the absence of sign changes in the former. According to above considerations a finite $\Delta_{d_{x^2-y^2}}$ can only be induced if $\xi_{\mathbf{k},\mathbf{A},d_{x^2-y^2}}$ is finite for some period of time. Thus by applying the pulse along the diagonal $k_x = k_y$, for which $\xi_{\mathbf{k},d_{x^2-y^2}}$ remains zero, there is no induced oscillation of the $d_{x^2-y^2}$-wave order parameter. In both cases the s-wave order parameter oscillates with its Higgs mode $\omega_H = 2\Delta_{s,\max}$, where $\Delta_{s,\max} = \max\{\Delta_s\gamma_\mathbf{k}|\mathbf{k} \in \mathrm{FS}\}$ is the maximum gap size on the Fermi surface. The frequency ω_{BS} is smaller than ω_H due to a residual attractive interaction and therefore undamped, while ω_H shows weak damping similar to the one expected [13,15] for isotropic s-wave states due to coupling to the quasiparticle continuum. This similarity is due to the s-wave solution being highly isotropic at the dopings we consider. Indeed, for an anisotropic gap (see the d-wave case in the Supplemental Material [34]) damping can be expected

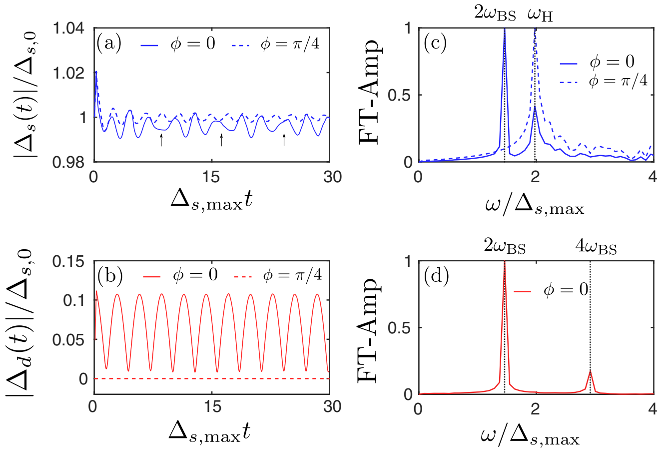

FIG. 2. Calculated evolution of the superconducting gap amplitudes for an s-wave ground state and an applied light pulse with polarization at an angle $\phi = 0$ (solid line) and an angle $\phi = \pi/4$ (dashed line) to the k_x axis. (a) and (b) show the short-time dynamics of the s- and $d_{x^2-y^2}$-wave order parameter, respectively. To make the existence of a second frequency in (a) clear, arrows illustrate the beating pattern. (c) and (d) refer to the Fourier transform of $|\Delta_s|$ and $|\Delta_{d_{x^2-y^2}}(t)|$, respectively. As $|\Delta_{d_{x^2-y^2}}(t)|$ remains 0 for $\phi = \pi/4$, no Fourier transform of this case is shown in (d).

to be stronger. We note that in our BCS approach we do not take into account other sources of dissipation, such as the electron-phonon coupling. However, thermalization effects due to coupling to the acoustic phonons occur at a much longer timescale of approximately 100 ps [40] compared to the typical timescale of the superconductivity, $\hbar/\Delta \sim 1$ ps. In particular, in modern THz experiments the quasiparticle lifetime due to phonons may reach nanoseconds, due to the absence of high-energy phonons created by the pulse [4,8]. Furthermore, quasiparticle interactions beyond BCS theory may play a role at high pump fluences yet these effects do not seem to alter the qualitative picture that results from a BCS-like approach [41]. Nonetheless, some effects of increasing fluence can be already observed within the BCS approach. In Fig. 3 we show the fluence dependence of ω_{BS} and ω_H. It can

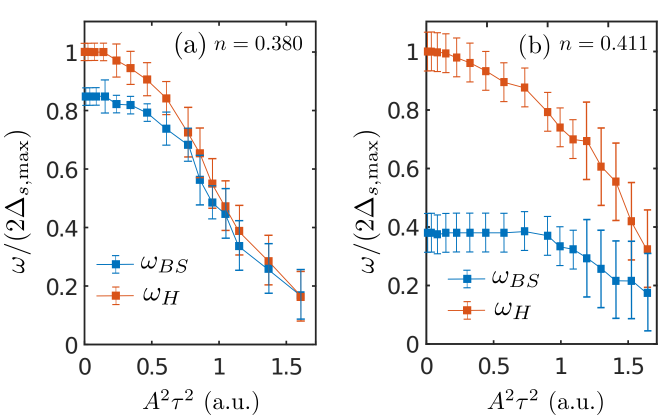

FIG. 3. Fluence dependence of the Bardasis-Schrieffer and the Higgs mode in the s-wave ground state for the filling (a) $n = 0.38$ and (b) $n = 0.411$, away (a) and close (b) to the phase transition to the $s + id_{x^2-y^2}$ ground state, respectively.

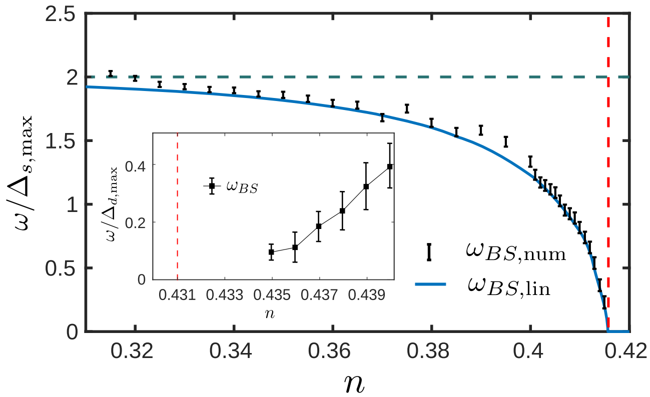

FIG. 4. Frequency of the Bardasis-Schrieffer mode for different band fillings close to the transition point between extended s-wave and $d_{x^2-y^2}$-wave ground states at $n \approx 0.415$. The numerical results (squares) and the results of the linearized equations (solid blue curve) are in good agreement.

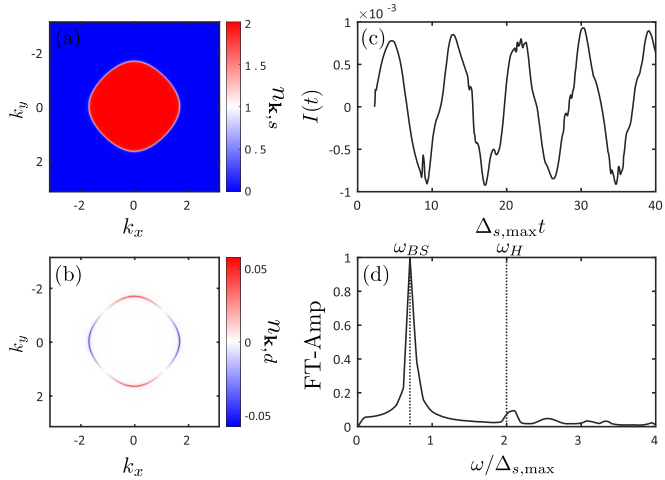

FIG. 5. Oscillation of the quasiparticle distribution, projected into fully symmetric s-wave (a) and $d_{x^2-y^2}$-wave (b) parts. The oscillation of the k_x-integrated $d_{x^2-y^2}$-wave part in (c) shows a signature of the Bardasis-Schrieffer mode and of the Higgs mode as shown in its Fourier transform (d).

be seen that in the case where ω_{BS} and ω_H are well separated [Fig. 3(b)], ω_{BS} is not directly affected by the light pulse, unlike ω_H. However, once the Higgs mode frequency becomes close to ω_{BS}, both start being suppressed by the increasing fluence and eventually become indistinguishable. Note that instead of using a pulse one could also by hand set a finite $\Delta_{d_{x^2-y^2}} \neq 0$ as the initial value, as it was shown above that the pulse also generates a finite $d_{x^2-y^2}$-wave component. We find that the quench scenario yields results equivalent to Fig. 2.

To verify whether the oscillations at ω_{BS} are due to the existence of the Bardasis-Schrieffer mode in the subdominant pairing channel, we analyze Eq. (5) in the linear regime (i.e., weak driving) and compare the frequencies of the resulting modes to the ones observed in the nonequilibrium (see Fig. 2). In particular, we linearize Eq. (5) around the equilibrium state at $T = 0$ and perform a subsequent Fourier transform $-i\omega\delta\mathbf{s}_{\mathbf{k}} = \mathbf{B}^{\mathrm{eq}}_{\mathbf{k}} \times \delta\mathbf{s}_{\mathbf{k}} + \delta\mathbf{B}_{\mathbf{k}} \times \mathbf{s}^{\mathrm{eq}}_{\mathbf{k}}$. A numerical solution of the homogeneous part of the resulting equations [34] gives three types of solutions: (i) $\omega = 0$ mode for the ground-state pairing symmetry independent of doping, (ii) $\omega = 2\Delta_{l,\max}$ modes for the both types of the ground states $l = s$ and $l = d_{x^2-y^2}$ (Higgs mode), and (iii) the Bardasis-Schrieffer mode, whose frequency depends on the proximity to the phase boundary between extended s-wave and $d_{x^2-y^2}$-wave symmetries in the equilibrium. We show the frequency of the Bardasis-Schrieffer mode in the s-wave ground state in Fig. 4 together with the results of the numerical solution of the full nonlinear equation [Eq. (5)] as a function of doping. For values of n far away from the transition this mode merges into the Higgs mode, while close to the phase transition it softens as is expected, signaling the transition to the new ground state. The results of the linearized calculation of ω_{BS} are in good agreement with the results for the full nonlinear dynamics.

We also considered the excitation of the Bardasis-Schrieffer mode in the $d_{x^2-y^2}$-wave ground state, which should also exist once the s-wave ground state is close enough. As the $d_{x^2-y^2}$-wave gap is nodal, all excited modes, including the Higgs mode, are strongly damped. However, we still see a clear signature of a mode at $\omega < 2\Delta_{d,\max} = \omega_H$. Due to the finite-size effects and the limited accuracy of the numerical integration, the error bar is quite broad. Therefore, we only show the frequency of this mode close to the $s + id$ transition in the inset of Fig. 4. To obtain these frequencies we can again equivalently use both a pulse or a quench, for example, if one sets $\Delta_s = 0.1\Delta_{d_{x^2-y^2}}$. Note that for better numerical accuracy, all frequencies in the inset of Fig. 4 are obtained via a quench.

The presence of the signatures of the Bardasis-Schrieffer mode in the order parameter dynamics raises the possibility of an enhanced third-harmonic generation when the pump frequency roughly matches ω_{BS}. Unlike the Higgs mode, where the issues of the quasiparticle contribution are still under debate [11,22,42–45], the signature of a Bardasis-Schrieffer mode would be definitive as $\omega_{\mathrm{BS}} < 2\Delta_0$, below the edge of the quasiparticle continuum, at least for fully gapped s-wave superconductors. At the same time, the amplitude oscillation of the order parameter (Higgs oscillation) was also predicted to show up in the tr-ARPES experiments [46]. The quasiparticle distribution function that can be measured in these experiments can be addressed in our model. In particular, the quasiparticle distribution function is determined by the $s^z_{\mathbf{k}}$ component of the pseudospin $s^z_{\mathbf{k}} = \frac{1}{2}(\langle c^\dagger_{\mathbf{k}\uparrow}c_{\mathbf{k}\uparrow}\rangle + \langle c^\dagger_{\mathbf{k}\downarrow}c_{\mathbf{k}\downarrow}\rangle - 1)$. In equilibrium this quantity is equal to $-\xi_{\mathbf{k}}/(2E_{\mathbf{k}})$ and thus is C_4 symmetric. Due to the perturbation via an electric field, the tetragonal symmetry is temporarily broken down to C_2 symmetry and $s^z_{\mathbf{k}}$ develops a finite $d_{x^2-y^2}$-wave component $s^{z,d}_{\mathbf{k}}$, i.e., the quasiparticle distribution along the x and y axis becomes asymmetric. Therefore, one expects that the nonequilibrium particle distribution acquires the information on ω_{BS} and ω_H. To investigate this in detail we define the quantity $I(t) = \int_{-\pi}^{\pi} dk_x[2s^{z,d}_{\mathbf{k}=(k_x,0)}(t)]$, which describes the integrated $d_{x^2-y^2}$-wave component of the quasiparticle distribution along the $k_y = 0$ cut and is equivalent to the number of particles with $k_y = 0$. In Fig. 5 we show $I(t)$ for the same parameters as in Fig. 2 for $\mathbf{A}$ oriented along the x axis. One can readily see that it oscillates mostly with $\omega = \omega_{\mathrm{BS}}$, where we find the amplitude of the oscillations increases with an increasing pump strength. The integration along the $k_x = 0$ cut leads to similar results but shifted by a phase $\varphi = \pi$.

To conclude we analyze the short-time dynamics in a single-band unconventional superconductor with multiple competing pairing symmetries. Driving the system out of equilibrium with a light pulse (modeled as a time-dependent vector potential), we show how the collective signatures of symmetries different from a given ground-state symmetry, known as Bardasis-Schrieffer modes [26–29] in the context of the s-wave ground state in the equilibrium, evolve as a function of doping and the polarization direction. Depending on the polarization direction of the incoming light the tetragonal symmetry is broken, which necessarily induces a coupling of s- and d-wave symmetries in a nonequilibrium. Furthermore, we show that the particle distribution acquires an additional $d_{x^2-y^2}$ character, due to coupling to the vector potential and that this quantity shows a dominant signature of the Bardasis-Schrieffer mode frequency, which might be observed in time-resolved ARPES experiments. Therefore we conclude that taking the subdominant pairing channels and the corresponding interactions into account is important, when discussing the polarization-dependent excitation of unconventional superconductors. Finally, we find that apart from quenching the order parameter, the pump pulse can be used to selectively probe excitations and control the nonequilibrium state of the system by employing the light polarization. In addition, stronger pulses lead to a coupling between Higgs and Bardasis-Schrieffer modes, which affects the frequency position of both.

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