Calculation of an Enhanced A_{1g} Symmetry Mode Induced by Higgs Oscillations in the Raman Spectrum of High-Temperature Cuprate Superconductors

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In superconductors the Anderson-Higgs mechanism allows for the existence of a collective amplitude (Higgs) mode which can couple to eV light mainly in a nonlinear Raman-like process. The experimental nonequilibrium results on isotropic superconductors have been explained going beyond the BCS theory including the Higgs mode. Furthermore, in anisotropic d-wave superconductors strong interaction effects with other modes are expected. Here we calculate the Raman contribution of the Higgs mode from a new perspective, including many-body Higgs oscillations effects and their consequences in conventional, spontaneous Raman spectroscopy. Our results suggest a significant contribution to the intensity of the A_{1g} symmetry Raman spectrum in d-wave superconductors. In order to test our theory, we predict the presence of measurable characteristic oscillations in THz quench-optical probe time-dependent reflectivity experiments.

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Introduction.—Collective excitations of superconductors in nonequilibrium are a new emerging field. The most studied collective modes in superconductors, among others, are the amplitude (AM, so-called Higgs mode) and phase (Goldstone) modes [1,2]. Yet, while the Higgs is a collective oscillation having a frequency of twice the superconducting order parameter Δ, i.e., $\omega_{\mathrm{Higgs}} = 2\Delta$, the phase mode is shifted to the plasma frequency at higher energies due to the Anderson-Higgs mechanism [1]. Since the Higgs mode does not carry a dipole moment, it is well known that a linear activation by light is almost impossible [3,4], such that the relevant process is the nonlinear Raman effect. In ultrafast nonequilibrium experiments the Higgs mode has been uncovered both in quench probes [5] and in periodically driven setups [6–8], respectively, where the coupling of light to the charges of the superconductor can be described by a quadratic Raman-like process. In this context, the third harmonic generation has been a widely studied example of this nonlinear optical effect [9–11].

In the 1990s conventional, steady-state Raman spectroscopy on superconductors turned out to be an effective experimental technique to provide evidence via polarization dependence for d-wave Cooper pairing [12–17]. In high-T_c cuprates with D_{4h} crystal symmetry the three most important Raman symmetries (A_{1g}, B_{1g}, B_{2g}) could be explained by a d-wave order parameter [17–21]. However, in *all* spectra of cuprates there is still an open question concerning the A_{1g} peak intensity [21,22]. In conventional Raman scattering theory, due to Coulomb screening of quasiparticles (QPs) the response in A_{1g} symmetry is much smaller than the experimental observations in cuprates [16,21], which is of the same order or larger than the B_{1g} peak [17–19]. This is the so-called long-standing A_{1g} problem [21]. Over the past three decades, various attempts have been made to reconcile theory with experiments without arriving at conclusive results: these include, but are not limited to, the use of higher harmonics [18,19], magnon resonance [23,24], screening effects [25], neutron magnetic resonance [26], and the interaction of the AM with the η mode, a spin-singlet excitation which can appear below 2Δ in d-wave superconductors [27]. However, none of these approaches could solve the A_{1g} problem quantitatively, namely, providing the correct intensity and peak position.

Recently, Puviani *et al.* [28] have shown that both equilibrium and nonequilibrium activation of the Higgs oscillations in superconductors correspond to the same physical nonlinear Raman process. The conventional equilibrium Raman response of s-wave superconductors should be small for symmetry arguments, with the exception of the presence of competing orders, i.e., superconductivity (SC) and charge density wave (CDW) [29,30]. Eventually, recent terahertz pump-optical probe (TPOP) experiments have provided hints of the presence of the Higgs mode through transient reflectivity change in the d-wave superconductor Bi2212 [31,32]. Despite this, no effective time-dependent oscillation detection, nor a consistent theoretical calculation have been provided for d-wave superconductors to support this.

In this Letter, we go beyond the present theoretical background on the amplitude mode in superconductors, including many-body effects of Higgs oscillations on quasiparticles: when light interacts with the charges of the condensate creating an electron-hole pair, these particles undergo the so-called Andreev scattering which leads to creation of pairs of electrons or holes which add to the existing condensate and eventually produce the Higgs [Fig. 1(a)]. However, in addition to this production mechanism, which is generally subdominant, the quasiparticle excitation generated by light can lead to dominant many-body Higgs oscillations due to a mechanism of interaction between Cooper pairs (CPs) generated by Andreev scattering [Fig. 1(b)], which has been neglected so far. Theoretically, this is done through a systematic diagrammatic approach, adding vertex corrections in the amplitude channel alongside with the usual random phase approximation (RPA) summation for the AM propagator.

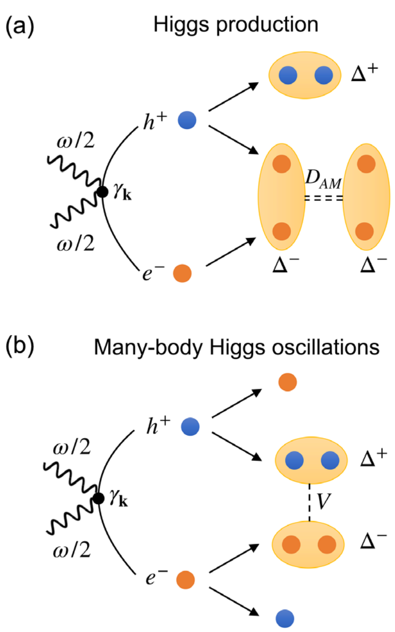

FIG. 1. Higgs production and many-body Higgs oscillation mechanisms. (a) Raman two-photon interaction (vertex function $\gamma_{\mathbf{k}}$) of light with the condensate leading to the creation of an electron-hole pair ($e^- - h^+$): when the hole (electron) undergoes Andreev scattering, it produces a Cooper pair and an electron (hole), and the latter forms a pair with the other electron (hole) thus producing a Higgs. (b) Light creates an electron-hole pair: both particles undergo Andreev scattering and produce Cooper pairs which interact via many-body Higgs oscillations [36].

This calculation gives rise to an effective hybridization of the amplitude and charge degrees of freedom, namely, many-body effects of Higgs and quasiparticles, resulting now in a new strong contribution of the Higgs mode. This approach is able to show that there is a significant signature of the Higgs mode in the calculated Raman spectra of d-wave superconductors, as suggested in Refs. [33,34], generalizing the phenomenological coupling used by Zeyher and Greco in Ref. [35]. In our theory a qualitative and quantitative agreement with the Raman experimental data is possible. Simply speaking, similar to phonons which can be renormalized to polarons due to strong electron-phonon interactions, the Raman-active Higgs mode can be strongly renormalized due to quasiparticles to form a Cooper pair polaron, resulting in a dressed Higgs mode with strong intensity in A_{1g} polarization. Note that the conclusions of our work can be generalized to other clean systems (e.g., SC + CDW, multiband superconductors) where the Higgs peak is shifted from the quasiparticle continuum and the symmetry allows for a many-body response.

In order to further test our theory, we suggest that using an ultrashort THz pump in TPOP experiments our theory would explain the relative intensity of the transient reflectivity change measured for the A_{1g} and B_{1g} symmetries, and it predicts the possibility to detect the characteristic time-dependent oscillations due to the amplitude mode contribution in both of these symmetries.

Calculation of the amplitude mode.—To describe a d-wave superconducting system, we consider a generalized version of the BCS theory, namely, the Hamiltonian: $H = H_{\mathrm{el}} + H_p + H_C$. It is the sum of the electronic tight-binding Hamiltonian H_{el}, a momentum-dependent attractive pairing interaction H_p, and a long-range Coulomb repulsive energy H_C, respectively. The pairing term responsible for the AM is given by

$$H_p = \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} V(\mathbf{k},\mathbf{k}')\hat{P}^{\dagger}_{\mathbf{k},\mathbf{q}}\hat{P}_{\mathbf{k}',\mathbf{q}}, \tag{1}$$

with the pair creation operator $\hat{P}^{\dagger}_{\mathbf{k},\mathbf{q}} = \hat{c}^{\dagger}_{\mathbf{k}+\mathbf{q},\uparrow}\hat{c}^{\dagger}_{-\mathbf{k},\downarrow}$, and the pairing interaction function $V(\mathbf{k},\mathbf{k}')$ [9,10]. For d-wave superconductors we can factorize the pairing interaction term $V(\mathbf{k},\mathbf{k}') = -Vf_{\mathbf{k}}f_{\mathbf{k}'}$, where $f_{\mathbf{k}} = f^{x^2-y^2}_{\mathbf{k}}$ is the $d_{x^2-y^2}$-wave form factor, V being the pairing strength derived from the self-consistent gap equation. The Hamiltonian $H_{\mathrm{el}} + H_{\mathrm{P}}$ can be treated at mean-field level defining the order parameter $\Delta_{\mathbf{k}} = \Delta_{\max}f_{\mathbf{k}}$, giving rise to a mean-field Hamiltonian for unconventional superconductors.

The bare susceptibility response of the condensate coupling quadratically to light is given by

$$\chi_{\gamma\gamma}(\mathbf{q},\omega) = \sum_{\mathbf{k}'}\gamma^{i}_{\mathbf{k}'}\gamma^{s}_{\mathbf{k}'}\frac{1}{\beta}\sum_{i\nu_m}\mathrm{Tr}[G_{\mathbf{k}',m}\tau_3 G^{+}_{\mathbf{k}',m}\tau_3], \tag{2}$$

where $\tau_{i=1,2,3}$ are the Pauli matrices, τ_0 the identity matrix, $\gamma^i_{\mathbf{k}}(\gamma^s_{\mathbf{k}})$ the interaction vertex for the incident (scattered) light for a given symmetry, $G_{\mathbf{k}',m} \equiv G(\mathbf{k}', i\nu_m)$ and $G^+_{\mathbf{k}',m} \equiv G(\mathbf{k}'+\mathbf{q}, i\nu_m + i\omega)$ the matrices of the Matsubara Green's functions in Nambu-Gor'kov space. The coupling between the light and the amplitude mode occurs indirectly via the charge-amplitude susceptibility, $\chi_{\gamma f}(\mathbf{q},\omega)$. Then, the AM propagator can be calculated with the RPA summation as in Ref. [10]

$$D_{\mathrm{AM}}(\mathbf{q},\omega) = -[2/V + \chi_{ff}(\mathbf{q},\omega)]^{-1}. \tag{3}$$

Here we have introduced the susceptibility $\chi_{ff}(\mathbf{q},\omega)$ with vertices $f_{\mathbf{k}}\tau_1 - f_{\mathbf{k}'}\tau_1$ in the amplitude channel. This allows us to calculate the total Raman susceptibility including both quasiparticles [Eq. (2)] and amplitude mode as

$$\tilde{\chi}(\omega) = \tilde{\chi}_{\gamma\gamma}(\omega) - \frac{\tilde{\chi}^2_{\gamma f}(\omega)}{2/V + \tilde{\chi}_{ff}(\omega)}, \tag{4}$$

where we used the susceptibilities $\tilde{\chi}$ which include charge fluctuations due to the long-range Coulomb interaction between quasiparticles [36]. We want to stress here that for a given value of the order parameter $\Delta_{\max}$ we calculate self-consistently within the gap equation the value of pairing strength V.

Note that here we have considered only the diamagnetic susceptibility, since the paramagnetic contributions are vanishing in clean superconductors and in visible light experiments, as well as coupling to the spin channel (resonance peak) is negligible [36].

Many-body Higgs oscillations.—As mentioned earlier, we now want to go beyond the RPA summation for the amplitude mode propagator, including many-body dynamic Higgs interactions between CPs generated by electrons or holes via Andreev scattering [Fig. 1(b)], mixing the amplitude and charge channels. To do that, we defined a dressed light-condensate interaction vertex through the ladder diagram summation

$$\Gamma(\mathbf{k},\omega) = \gamma_{\mathbf{k}}\tau_3 - \frac{V}{2} f_{\mathbf{k}} \sum_{\mathbf{k}'} f_{\mathbf{k}'} \frac{1}{\beta} \sum_{i\nu_m} [\tau_1 G_{\mathbf{k}',m} \Gamma(\mathbf{k}',\omega) \times G^+_{\mathbf{k}',m}\tau_1]. \tag{5}$$

Analogously, for the $f_{\mathbf{k}}\tau_1$ vertex in the amplitude channel we derived the self-consistent sum

$$\Lambda(\mathbf{k},\omega) = \tau_1 - \frac{V}{2} f_{\mathbf{k}} \sum_{\mathbf{k}'} f_{\mathbf{k}'} \frac{1}{\beta} \sum_{i\nu_m} [\tau_1 G_{\mathbf{k}',m} \Lambda(\mathbf{k}',\omega) \times G^+_{\mathbf{k}',m}\tau_1]. \tag{6}$$

Substituting the bare vertices with the dressed ones including the many-body effects of the Higgs mode, the Raman response in Eq. (4) can be replaced with the full many-body expression

$$\tilde{\chi}_{\mathrm{full}}(\omega) = \tilde{\chi}_{\Gamma\Gamma}(\omega) - \frac{\tilde{\chi}^2_{\Gamma\Lambda}(\omega)}{2/V + \tilde{\chi}_{\Lambda\Lambda}(\omega)}, \tag{7}$$

where the susceptibilities $\tilde{\chi}$ include charge fluctuations [10,16].

Steady-state Raman spectroscopy.—The intensity measured in Raman experiments is given by the photon scattering differential cross section, which is proportional to the imaginary part of the susceptibility, namely, $\partial^2\sigma/\partial\omega\partial\Omega \propto -\tilde{\chi}''(\mathbf{q},\omega)$. In order to solve the set of equations for the dressed vertices and the susceptibilities, we considered the lowest terms in a Fermi-surface harmonic expansion of the Raman vertices, restricting

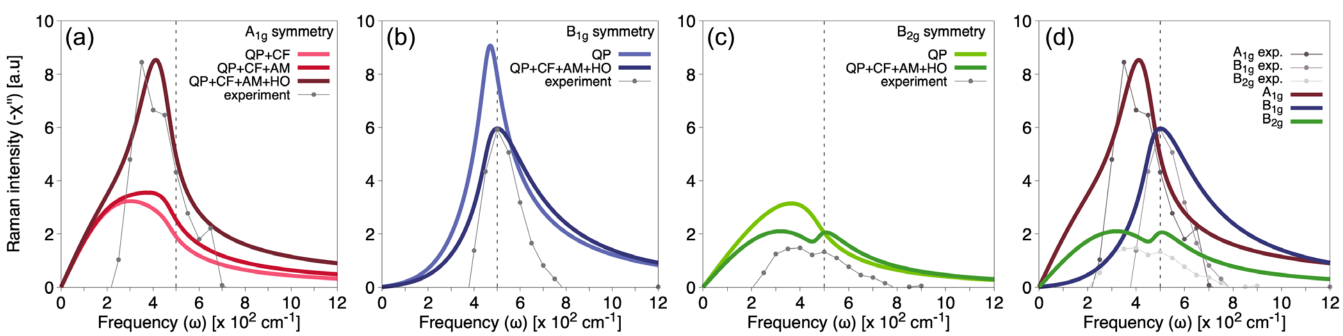

FIG. 2. Raman spectra of Bi2212. Raman spectra contributions of various symmetries of a D_{4h} crystal: comparison between different theoretical contributions and experimental results (represented by dots: the lines connecting them are a guide for the eyes) for Bi2212 [36]. The dashed vertical lines are placed at $\omega = 2\Delta_{\max}$, tuned with the B_{1g} peak position. (a) A_{1g} symmetry: the light red line is the quasiparticle (QP) + charge fluctuations (CF) contribution, the intermediate one includes the amplitude mode (AM), the darker line includes also the many-body effects of the Higgs oscillations (HO). (b) B_{1g} spectrum intensity due to QP (light blue) and adding the CF, AM, and HO contributions (the QP + CF + AM plot would coincide with QP). (c) B_{2g} Raman spectrum response of QP (light green) and including CF and the full Higgs contribution (the QP + CF + AM plot would coincide with QP). (d) The responses for all the symmetries are placed together.

ourselves to $\mathbf{q} = 0$ for the light and to the clean limit of superconductor, in agreement with previous studies [18,19]. Using the same optimized parameters fixed for all the Raman symmetries investigated, we additionally checked systematics in order to confirm the stability of the results [36]. In Fig. 2(a) we show the effect of the bare AM and the many-body dressing contribution in the Raman spectra for different symmetries of Bi2212 (a d-wave superconductor with $2\Delta_{\max} \sim 550\ \mathrm{cm}^{-1}$): on the one hand, for the A_{1g} response the dressed AM calculated within the RPA approximation using Eq. (4) is negligible, while the full many-body Higgs oscillations result in a contribution which is more intense and peaked. On the other hand, for the B_{1g} and B_{2g} symmetries the charge fluctuations and Higgs oscillations screen the Raman response, reducing its intensity. In particular, the B_{1g} spectrum becomes broader in frequency and the peak gets shifted to higher frequencies, while the B_{2g} acquires a shoulder peaked at around $\omega \sim 2\Delta_{\max}$.

These many-body Higgs effects describe the experimental Raman on Bi2212 in a much improved fashion, thus contributing to the long-standing problem regarding the A_{1g} peak intensity [18,19]. As we have concluded before, the A_{1g} response provided by QPs is much lower than the experimental one, and the bare AM contribution is negligible. However, considering many-body effects of the Higgs oscillations, the B_{1g} [Fig. 2(b)] and B_{2g} [Fig. 2(c)] spectra get screened, while the A_{1g} response becomes enhanced and peaks now at a frequency $\omega \sim 400\ \mathrm{cm}^{-1}$, in quite good agreement with the experimental result. Note that a simultaneous and quantitative understanding of all relevant polarizations is indeed possible employing the *same* set of many-body parameters [36] as shown in Fig. 2(d). This contributes to explain the long-standing A_{1g} problem concerning the peak intensity in optimally doped Bi2212 as formulated in the literature [17,18,21].

Ultrafast Raman-like optics.—As discussed earlier, the same Higgs oscillations should be seen in nonequilibrium in a time-dependent experiment. While, in principle, it is possible to measure Fig. 2(d) time dependently in an ultrafast experiment, it is much easier to detect the Higgs oscillations in the transient reflectivity change $\Delta R(t)/R$. It has been widely demonstrated that the coupling of the light to the collective AM can happen only in a nonlinear regime with a Raman-like interaction, thus new evidence should be looked for in nonlinear optical effects, such as the Kerr signal in pump-probe experiments. To this extent, it is useful to look at the time-dependent reflectivity change in nonequilibrium superconductors, which is related to the equilibrium Raman susceptibility aforementioned and easily accessible in experiments. The transient reflectivity change due to a pump electric pulse $E_{\mathrm{pump}}(t)$ is given by [43]

$$\frac{\Delta R}{R}(t) \propto \int_{-\infty}^{t} \chi'^{(3)}(t-t') E_{\mathrm{pump}}^2(t') dt', \tag{8}$$

where $\chi'^{(3)}(t-t')$ is the real part of the third order susceptibility in real time, t being the time at which the reflectivity change is detected, $t' < t$ the previous time at which the pump pulse interacts with the condensate. Recently, Katsumi *et al.* [31,32] demonstrated that the relative intensity between the transient reflectivity change projected onto the symmetries A_{1g} and B_{1g} in a TPOP experiment on Bi2212 cannot be explained only with the quasiparticle density fluctuations contribution, deducing that the Higgs should play an important role. However, since they pumped the system in the fully adiabatic regime ($\omega_{\mathrm{pump}} \ll \Delta_{\max}, \Delta t_{\mathrm{pump}} \sim 4$ ps), they were not able to probe any Higgs oscillation in the time-dependent response.

We performed the calculation of the theoretically expected time-dependent transient reflectivity change on a d-wave superconductor, using Eq. (8) together with the third-order susceptibility containing the full many-body Higgs contribution according to Eq. (7). Considering the same pump regime of the experiments, we have been able to get a good agreement of the relative intensities for the A_{1g} and B_{1g} symmetries [36]. In addition to this, we repeated the numerical calculation with an ultrashort THz pump (duration $\Delta t_{\mathrm{pump}} \sim 0.1$ ps, frequency $\omega_{\mathrm{pump}} = 5.57\ \mathrm{THz} \lesssim \Delta_{\max}$) in order to simulate a quench experiment: the result is shown in Fig. 3. Including the full Higgs

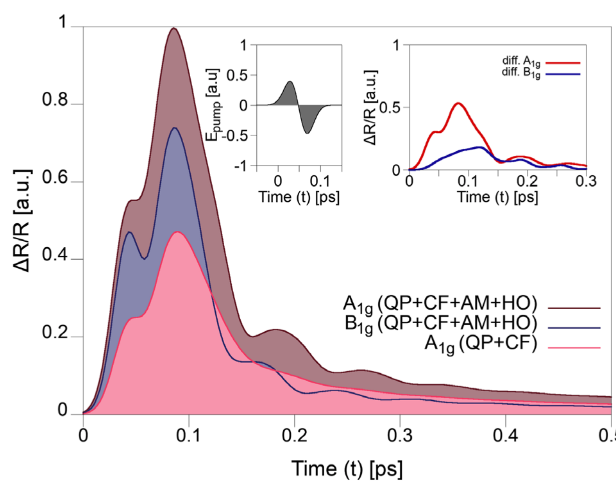

FIG. 3. Transient reflectivity. Normalized transient reflectivity change $\Delta R(t)/R$ after a quench, obtained applying an electric pump pulse (E_{pump}, shown in the left inset) with duration $\Delta t_{\mathrm{pump}} \sim 0.1$ ps and frequency $\omega_{\mathrm{pump}} = 5.57$ THz. The dark red and dark blue curves represent the normalized transient reflectivity of the A_{1g} and B_{1g} symmetries, respectively, calculated including the full many-body Higgs contribution. The light red curve, instead, shows only the QP and CF response in the A_{1g} symmetry. The top right inset shows the differential A_{1g} (red line) and B_{1g} (blue line) signal due to the Higgs contribution.

contribution to the third-order susceptibility, oscillations with a frequency $\omega = \omega_{\mathrm{Higgs}} = 12.5$ THz $\lesssim 2\Delta_{\max}$ appear in the decaying region of $\Delta R(t)/R$, while a lower intensity and no oscillations are present considering only the QPs and charge fluctuations' contributions. Performing such a time-resolved experiment would provide clear evidence of the presence of the Higgs mode and the effects of many-body contributions in d-wave superconductors: indeed, they are responsible for enhancing the A_{1g} symmetry response and, at the same time, for the screening effect on the B_{1g}.

Conclusion and outlook.—For the first time, we provide calculations of Higgs oscillations for d-wave superconductors including many-body contributions originating from the interaction with quasiparticles (e.g., *broken* Cooper pairs), resulting in a renormalized, dressed Higgs mode. We treat the full vertex corrections within a diagrammatic approach, which is capable to describe both nonequilibrium and conventional steady-state Raman experiments, respectively. In particular, we are able to provide a contribution to the long-standing A_{1g} problem in Raman spectra of d-wave superconductors [18,19] in Fig. 2(d). Our conclusions are not based on any specific scenario for cuprates. Furthermore, our results are also unchanged if we take into account that Higgs oscillations can be dressed by phonons as well. However, these contributions to the Raman spectra are minor [36]. Furthermore, we explain the nonequilibrium response and the correct magnitude of the transient reflectivity change in recent THz pump-optical probe experiments [31]. Our theory can simply be tested by a smoking gun experiment: we propose an experimental setup using ultrashort THz pump pulse in a nonadiabatic regime to measure the reflectivity in d-wave superconductors, in order to quench the condensate and thus directly observe the time-dependent oscillation characteristics of the Higgs mode.

It is expected that the resonance peak obtained with inelastic neutron scattering can play a role in the A_{1g} problem because of the energy matching with the A_{1g} Raman response. However, it is interesting to note that there is no linear coupling of the spin channel to the Higgs mode or to the Raman vertex: as shown by Venturini *et al.* in [23], only by including spin fluctuations at higher order it is possible to match the energy position of the A_{1g} peak, but without substantially affecting its intensity.

We also point out that so far it has been possible to clearly detect the Higgs contribution only in the presence of competing orders such as in $2H$-$NbSe_2$, namely, superconductivity and charge density wave. The latter provides the excited phonons which couple to the superconducting AM [44]. This mechanism allows us to shift the Higgs A_{1g} peak to energies lower than 2Δ, becoming thus unambiguously detectable [44–47]. Similarly, a Higgs response has been reported in the E_{2g} Raman response of pressure-induced $2H$-TaS_2 [30], a transition metal dichalcogenide with cohexisting SC and CDW. Our findings provide a new route for calculations of many-body effects of the Higgs mode while interacting with various other modes in superconductors [45,48]. We believe that an extension of our model to these systems can provide further insights and guide future experiments on unconventional superconductors and systems with coexisting phases.

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