Current-assisted Raman activation of the Higgs mode in superconductors

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The Higgs mode in superconductors is a scalar mode without electric or magnetic dipole moment. Thus, it is commonly believed that its excitation is restricted to a nonlinear two-photon Raman process. However, recent efforts have shown that a linear excitation in the presence of a supercurrent is possible, resulting in a new resonant enhancement at \( \Omega = 2\Delta \) with the driving light frequency \( \Omega \) and the energy of the Higgs mode \( 2\Delta \). This is in contrast to the usual \( 2\Omega = 2\Delta \) resonance condition found in nonlinear third-harmonic generation experiments. In this communication, we show that such a linear excitation can still be described as an effective Raman two-photon process, with one photon at \( \omega = 2\Delta \) and one virtual photon at \( \omega = 0 \) which represents the dc supercurrent.

At the same time we demonstrate that a straightforward infrared activation with a single photon excitation is negligible. Moreover, our generalized theory provides an explanation for how the excitation of the Higgs mode in both THz quench and drive experiments can be understood within a conventional difference-frequency generation or sum-frequency generation process, respectively. In such a picture, the observed new resonance condition \( \Omega = 2\Delta \) is just a special case.

Introduction. Light excitation of collective modes in condensed matter physics is typically realized due to an infrared or Raman coupling of light to the system. This corresponds to a one- or two-photon process, i.e., a linear or a nonlinear coupling. If the mode does not have a dipole moment, a linear activation is forbidden, such that the only allowed process is due to the nonlinear Raman effect. This is true for the Higgs mode in superconductors, which is a collective oscillation of the amplitude of the order parameter \( \psi \). Its observation so far was realized either in a quench-probe or a periodic driven setup, where in both cases the coupling of light to the superconductor can be described by a quadratic nonlinear effective Raman process. Due to this nonlinear coupling, light of frequency \( \Omega \) drives the system effectively with a frequency of \( 2\Omega \) leading to enforced \( 2\Omega \) oscillations of the order parameter. A tuning of the effective driving frequency to the energy of the Higgs mode at \( 2\Delta \) leads to an enhancement of the oscillations and a resonance peak in the spectrum at \( 2\Omega = 2\Delta \). This second-harmonic component in the order parameter oscillation translates into third-harmonic generation (THG) in emission and the resonance is observable in the emitted THG signal.

Recent studies have shown that a linear coupling of light to the condensate is possible in the presence of a supercurrent. Hereby, the current provides an additional momentum component, such that the scalar Higgs mode can be excited with a single photon. With this, the previously forbidden linear driving with the frequency \( \Omega \) is now possible and enforces the order parameter to oscillate at just \( \Omega \). As a result, the first-harmonic oscillation of the order parameter shows a new resonance condition at \( \Omega = 2\Delta \). In an emission experiment, second-harmonic generation (SHG) would occur and the new resonance is then observable in the emitted SHG signal. Furthermore, the effect is also visible in the linear response and indeed, an enhancement in the optical conductivity was observed in the superconductor NbN, which was driven by a dc supercurrent. The question arises, how this linear excitation can be understood on a microscopic level. Is the Higgs mode excited by a single photon and thus becomes infrared active in the presence of a supercurrent?

In this communication, we argue that this coupling should not be understood as an infrared activation of the Higgs mode in a single photon process but rather as a current-assisted Raman process, where the excitation is still realized in a two-photon process. Hereby, one photon is at the energy \( \omega = \Omega \) and the other photon is a virtual photon with \( \omega = 0 \) representing the dc supercurrent. We find that a single photon infrared activation is negligible compared to such an effective Raman process. The interpretation of the excitation process is more general and allows to describe the usual nonlinear two-photon activation of the Higgs mode in quench or drive experiments with a sum-frequency generation (SFG) or difference-frequency generation (DFG) scheme. Within this picture, the current-assisted excitation just represents a special case with one photon energy set to zero.

Current-assisted Raman. To describe the coupling of light to current-carrying superconductors, we start with a phenomenological description using a Lagrangian of Ginzburg-Landau type. For the complex superconducting order parameter \( \psi(r, t) \) coupled to a gauge field \( A_\mu = (\phi, A(t)) \) where we choose \( \phi = 0 \), the time- and space-dependent Lagrangian reads

\[
\mathcal{L} = (D_\mu \psi)^*(D^\mu \psi) - V(\psi) - \frac{1}{4} F_{\mu\nu} F^{\mu\nu},
\]

where the potential \( V(\psi) = \alpha |\psi|^2 + \frac{\beta}{2} |\psi|^4 \) has the shape...
of a Mexican hat in the superconducting state with $\alpha < 0$. The gauge covariant derivative reads $D_\mu = \partial_\mu + ieA_\mu$ with the effective charge $e$ and the electromagnetic field tensor $F_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$. We are interested in small fluctuations around the groundstate value $|\psi_0| = \sqrt{-\alpha/\beta}$, thus, we use an ansatz $\psi(r,t) = (\psi_0 + H(r,t))e^{i\theta(r,t)}$ to describe amplitude (Higgs) fluctuations $H$ and phase (Goldstone) fluctuations $\theta$. We assume that the superconductor is driven by a light field $A(t) = A_0e^{i\Omega t}$ and that it simultaneously carries a constant dc supercurrent $j$, such that the gauge field can be written as $A(t) = A(t) + j/t^2$. Neglecting higher orders and constant terms, the resulting Lagrangian for the fluctuations reads as

$$\mathcal{L} = \left(\partial_\mu H - ie \left( A_\mu + \frac{1}{e} \partial_\mu \theta \right) \right) (\psi_0 + H) \times \left( \partial^\mu H + ie \left( A^\mu + \frac{1}{e} \partial^\mu \theta \right) \right) + 2\alpha H^2 - \frac{1}{4} F_{\mu\nu} F^{\mu\nu}.$$

The gauge freedom of the Lagrangian allows to perform a gauge transformation $\psi \rightarrow \psi e^\chi$ and $A_\mu \rightarrow A_\mu - \frac{e}{i} \partial_\mu \chi$, where we choose the unitary gauge $\chi = -\theta$. With this particular choice, we remove the Goldstone mode, whereas a mass term $e^2\psi_0^2 A_\mu A^\mu$ for the gauge field arises. This corresponds to the Anderson-Higgs mechanism, which is unaffected by the presence of the dc supercurrent. The Lagrangian then reads

$$\mathcal{L} = \left(\partial_\mu H\right)\left(\partial^\mu H\right) + 2\alpha H^2 - \frac{1}{4} F_{\mu\nu} F^{\mu\nu} + e^2\psi_0^2 A_\mu A^\mu + 2e^2\psi_0 A_\mu A^\mu H.$$  

(3)

The term $\mathcal{L}_{int} = 2e^2\psi_0 A_\mu A^\mu H$ describes the coupling between the gauge field and Higgs and reads explicitly

$$\mathcal{L}_{int} = 2e^2\psi_0 (\mathcal{A}^2 H + \frac{2}{e^2} j A H + \frac{1}{e^4} j^2 H).$$  

(4)

We observe that for finite $j$ a linear coupling term arises. The equation of motion for the amplitude fluctuations $H$ at $q = 0$ reads

$$\partial_t^2 H = 2\alpha H + e^2\psi_0 A^2 + 2e^2\psi_0 j A + \frac{1}{e^2} \psi_0 j^2,$$

(5)

and the stationary solution is easily obtained as

$$H = \frac{\psi_0 j^2}{e^2\omega_H^2} \left( \frac{2e^2\psi_0 A_0}{\Omega^2 - \omega_H^2} e^{i\Omega t} - \frac{e^2\psi_0 A_0^2}{4\Omega^2 - \omega_H^2} e^{2i\Omega t}. \right)$$

(6)

where the energy of the Higgs mode $\omega_H^2 = -2\alpha$ is obtained for $A_0 = 0$. From the microscopic theory it is known that $\omega_H = 2\Delta^1$. For $j = 0$, only the nonlinear driven oscillations exist with $\omega = 2\Omega$ resonating at $2\Omega = 2\Delta$. For $j \neq 0$, a linear coupling is possible resulting in an oscillation with $\omega = \Omega$ resonating at $\Omega = 2\Delta$. Using similar arguments, this result of linear coupling was first derived in$^3$. In general, one could have included an additional nonrelativistic Gross-Pitaevskii-like term $\propto \psi^* D_0 \psi$ in addition to the relativistic Klein-Gordon-like description of the dynamics. However, it is known that in the nonrelativistic case, there is no distinct Higgs mode as amplitude and phase channel are coupled$^2$. Despite the fact that a current would support a particle-hole breaking term, its contribution cannot be large. Otherwise, a resonance at $\omega_H$ is not explainable.

The phenomenological description does not give insight, how the linear coupling on a microscopic level is possible. At first glance, an infrared activation described by the diagram in Fig. 1(a) seems reasonable, where light couples linearly to the electron bubble and the current provides additional momentum $Q$ within an effective vertex interaction. We calculate the current-current correlation function using this diagram to obtain the response of the system $R(Q,\Omega) \propto Q A|\chi_{jj}(Q,\Omega)|^2$  

Figure 1. Feynman diagrams describing excitation of Higgs mode in current-carrying state with condensate momentum $Q$ and light frequency $\Omega$. a) Single-photon infrared excitation with $j_k \propto \partial_k e_k$. b) Effective Raman excitation with $\gamma_k \propto \partial_k^2 e_k$. Wiggly, solid and double dashed lines represent photon, electron and Higgs propagator.

The susceptibility $\chi_{jj}^H(q,\Omega)$ is given by

$$\chi_{jj}^H(q,\Omega) = H(q,\Omega) \chi_{jj}^2(q,\Omega) = -\frac{\chi_{jj}^2(q,\Omega)}{2/V + \chi_{11}(q,\Omega)}$$

(7)

with the pairing interaction $V$ and the Higgs propagator $H(q,\Omega) = -(2/V + \chi_{11}(q,\Omega))^{-1}$. The susceptibility $\chi_{jj}(q,\Omega)$ reads after analytic continuation $i\omega_n \rightarrow \Omega$ of the expression

$$\chi_{jj}(q,i\omega_n) = \sum_k j_k f_k \int d\omega_1 d\omega_2 \frac{\Delta_k \omega_2 + \Delta_{k+q} \omega_1}{2E_k E_{k+q}} \times \frac{n_F(\omega_1) - n_F(\omega_2)}{\omega_1 - \omega_2 + i\omega_n} \times [\delta(\omega_1 - E_k) - \delta(\omega_1 - E_k)] \times [\delta(\omega_2 - E_{k+q}) - \delta(\omega_2 + E_{k+q})],$$

(8)

with $j_k = \partial_k e_k$, electron dispersion $e_k$, energy gap $\Delta_k$, gap symmetry $f_k$, quasiparticle energy $E_k = \sqrt{\Delta_k^2 + e_k^2}$. 


and Fermi function \( n_F \). Details of the calculation can be found in the supplemental material. In the limit \( q \to 0 \), the term \( \Delta_k(\omega_1 + \omega_2) \) is nonzero only when \( \omega_1 = \omega_2 = E_k \), but in this case, \( n_F(E_k) - n_F(\omega_k) = 0 \), thus, \( \chi_{ij}(q = 0, \Omega) = 0 \) is identically zero. For finite but small \( q = Q \), the contribution of this diagram is still small. For very large \( Q \), the contribution gets comparable to the Raman process, which we will discuss next. However, it is no longer peaked at \( \Omega = 2\Delta \), but is shifted to higher energies reflecting the quadratic dispersion of the Higgs mode. This is shown in more detail in the supplemental material. As no such shift is observed experimentally, the value of \( Q \) must be small. Using parameters from the experiment in \(^{10}\), we estimate the value of the supercurrent induced momentum

\[
Q = \frac{m^*}{\hbar} v_s a_0 = \frac{m^*}{e^* n_s} J a_0 \approx 3.7 \times 10^{-4}
\]  

(9)

with Cooper pair mass \( m^* = 2m_e \) and charge \( e^* = 2e \), superfluid density \( n_s = 5.4 \times 10^{20} \text{cm}^{-3} \), current density \( J = 3.7 \text{MA/cm}^2 \) and lattice constant \( a_0 = 10^{-8} \text{cm} \). Thus, the contribution from the infrared diagram in the current-carrying state is negligible.

If such an infrared coupling has difficulties to explain a resonance at \( \Omega = 2\Delta \), one has to think about a different process. We let us guide by a microscopic description using BCS theory. The BCS Hamiltonian within Anderson pseudospin formulation \(^{14}\) can be written as

\[
H = \sum_k b_k \sigma_k
\]  

(10)

with the definition of the pseudospin \( \sigma_k = \frac{1}{2} \Psi_k \tau \Psi_k \), where \( \Psi_k = (c^{\dagger}_{k\uparrow}, -c^{\dagger}_{k\downarrow}) \) is the Nambu-Gorkov spinor and \( \tau \) the vector of Pauli matrices. The pseudomagnetic field \( b_k \) for a real gap \( \Delta_k = \Delta f_k \), driving amplitude \( A(t) = A_0 \sin(\omega t) \) and current induced momentum \( Q \) reads

\[
b_k(t) = \left( -2\Delta \right) \sin(\omega t), \quad 0, \quad -q + \epsilon_k \epsilon_{tA}(t) + q + \epsilon_k \epsilon_{tQ}(t) \right) .
\]  

Neglecting the imaginary part of the gap, which is unimportant for our discussion, the gap equation expressed in the pseudospin picture reads

\[
\Delta(t) = V \sum_k f_k (\sigma_k^\dagger) (t)
\]  

(11)

where the function \( f_k \) describes the symmetry of the gap and \( V \) is the pairing interaction. We expand the \( z \)-component of the pseudomagnetic field in powers of \( A \) and obtain

\[
b_k \approx 2\epsilon_k + \sum_{ij} \partial^2_{ij} \epsilon_k \left( e^2 A_i A_j + 2e A_i Q_j + Q_i Q_j \right) .
\]  

(12)

Hereby, the first order term vanishes due to parity, corresponding to the vanishing of the infrared diagram for \( q \to 0 \). However, similar to the phenomenological description shown before, for finite \( Q \), a new term linear in \( A \) arises. We proceed by evaluating the linearized Bloch

\[
\frac{\partial \sigma_k}{\partial t} = b_k \times \sigma_k \text{ for small deviations } \delta \Delta(t) \text{ from the equilibrium value } \Delta .
\]  

(13)

with the solution

\[
\delta \Delta(t) = \delta \Delta_A(t) + \delta \Delta_{AQ}(t) + \delta \Delta_{AQ}(t) \cos(2\Omega t)
\]  

(14a)

\[
\delta \Delta_{AQ}(t) \propto \frac{4e^2 A_0^2 \Omega \sin(2\Omega t)}{\int \frac{d\varphi}{f^2} \sqrt{\Delta^2 f^2 - \Omega^2} \sin^{-1} \left( \frac{\Omega}{\Delta} \right)}
\]  

(14b)

(14b)

The first term \( \delta \Delta_A(t) \) describes the \( 2\Omega \) oscillations of the order parameter resonating at \( \Omega = 2\Delta \) which are induced by the usual quadratic coupling. The second term \( \delta \Delta_{AQ}(t) \) describes the previously forbidden \( \Omega \) oscillations of the order parameter with a new resonance at \( \Omega = 2\Delta \), which is only present for finite \( Q \). The amplitudes of both terms are shown exemplary in Fig. 2 for \( s \)-wave symmetry with \( f(\varphi) = 1 \). An evaluation for \( d \)-wave can be found in the supplemental material. The same qualitative behavior compared to the phenomenological description is observed.

Yet, the microscopic description contains further details, which allows to understand the excitation process on a diagrammatic level. From the expansion in Eq. (12), we see that both the linear and quadratic couplings are proportional to the second derivative of the dispersion. In addition, the coupling occurs in the \( z \)-component of the pseudomagnetic field, i.e. the \( \tau_3 \) channel, reflecting a Raman coupling. Thus, we might describe both processes, the quadratic and linear coupling, with an effective Raman vertex as shown in Fig. 1(b). For the quadratic
Two-photon current-assisted Raman process with Ω shown in Fig. 3(a). The Higgs mode is excited via the scheme of impulsive stimulated Raman scattering of the superconducting order parameter. That process follows pulse quenches the Mexican hat potential of the complex boson operator due to the superfluid condensate enabling the Higgs excitation in the superfluid phase of unconventional superconductors. This mechanism also has an analogy to the Higgs excitation in the superfluid phase of supercold bosons. Here, a nonzero expectation value of a boson operator due to the superfluid condensate enables a linear coupling to the vector potential similar to the finite Q of the supercurrent. Thus, such a current-assisted Raman diagram can explain the Ω = 2∆ resonance in the current-carrying state.

**SFG and DFG for Higgs mode.** With this insight, we can summarize and classify the possible excitation schemes of Higgs modes as sketched in Fig. 3. In the impulsive excitation of a Higgs mode, a short intense THz pulse quenches the Mexican hat potential of the complex superconducting order parameter. That process follows the scheme of impulsive stimulated Raman scattering shown in Fig. 3(a). The Higgs mode is excited via the difference-frequency of the photons Ω1 and Ω2 that stem from the same pulse. The required frequencies are within the bandwidth of the ultrashort broadband THz pulse. Experimentally, the free Higgs oscillations observed in NbN are excited this way.

Instead of a difference-frequency process between the incoming photons, it is also possible to excite the Higgs mode via a sum-frequency process to excite Raman active modes as shown in Fig. 3(b). The 2Ω oscillations of the driven Higgs mode in NbN, Nb3Sn, and in cuprates as well as the driven Leggett mode in MgB2 can be understood in this way. In these cases, the two photons Ω1 = Ω2 = Ω have the same frequency leading to the experimentally observed second harmonic generation and the 2Ω = 2∆ resonance condition. This two-photon Raman process is described by the ∝ A1A2 term in Eq. (12). In contrary, an infrared excitation is a one photon absorption process by coupling to a dipolar moment shown in Fig. 3(c). However, as we have discussed in this communication, the current-driven superconductor does not change the character of the Higgs mode. The current rather gives rise to a new ∝ AQj term in Eq. (12) that describe an effective Raman two-photon excitation. In addition to the photon Ω1 = Ω, the current takes the role of a photon with Ω2 = 0. As such, the current-driven case can be understood in analogy with the SFG or DFG Raman processes, leading to Ω1 ± Ω2 = Ω and the new Ω = 2∆ resonance condition shown in Fig. 3(d).

This picture properly describes the experimental observation of the appearance of the Higgs mode in the linear THz spectrum of NbN in the presence of a dc current. Moreover, it explains the appearance of the Ω oscillation in the THz driven Nb3Sn in addition to the 2Ω terms and the higher order interference terms. In this experiment, the strong THz field dynamically drives a supercurrent giving rise to the current-assisted Raman excitation of Higgs oscillations in addition to the real two-photon process.

In summary, we provide an alternative explanation for the recent experimental conclusion that in the presence of a supercurrent the Higgs mode becomes infrared active. Our theory shows that in this case the activation of the Higgs mode is an effective Raman process (SFG or DFG), where one of the photons is a virtual photon at ω = 0. On the other hand we demonstrate that an infrared activation is negligible. Our results are not restricted to conventional s-wave superconductors and thus, will guide further current-assisted experiments also on unconventional superconductors.

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Supplemental material:
Current-assisted Raman activation of the Higgs mode in superconductors
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I. EVALUATION OF THE INFRARED DIAGRAM

The response of the diagram in Fig. 1(a) from the main text is proportional to

\[ R(Q, \Omega) \propto QA |\chi_{ji}^1(Q, \Omega)|. \]  

(1)

In particular, the susceptibility for the current-current interaction vertices (each with interaction term \( j_k \tau_0 \)) can be expressed as

\[ \chi_{ji}^1(Q, \Omega) = H(Q, \Omega) \chi_{ji}^2(Q, \Omega), \]

(2)

with the dressed Higgs propagator calculated by means of a RPA summation\(^1\) as \( H(Q, \Omega) = -(2/V + \chi_{11}(Q, \Omega))^{-1} \), where \( \chi_{11}(Q, \Omega) \) is the \( \tau_1 - \tau_1 \) bubble susceptibility and \( V \) the pairing strength. Therefore, we can rewrite the expression in (2) as

\[ \chi_{ji}^1(Q, \Omega) = -\frac{\chi_{ji}^2(Q, \Omega)}{2/V + \chi_{11}(Q, \Omega)}. \]

(3)

The bubble susceptibilities appearing in this expression are calculated as

\[ \chi_{11}(Q, \omega_n) = \sum_k f_k^2 \sum_{\nu_m} \frac{1}{\beta} \sum_{\nu_m} \text{Tr} \left[ G(k, \nu_m) \tau_1 G(k + Q, \nu_m + i\omega_n) \tau_1 \right], \]

(4)

\[ \chi_{jj}(Q, \omega_n) = \sum_k f_k j_k \sum_{\nu_m} \frac{1}{\beta} \sum_{\nu_m} \text{Tr} \left[ G(k, \nu_m) \tau_0 G(k + Q, \nu_m + i\omega_n) \tau_1 \right], \]

(5)

where \( f_k \) is the gap symmetry form factor, \( j_k \) is the gradient of the energy projected onto the direction of the light polarization, \( \tau_i \) with \( i = 1, 2, 3 \) are the Pauli matrices and \( \tau_0 \) is the identity matrix. Moreover, we use \( \nu_m \) for the fermion frequency and \( \omega_n \) for the boson frequency, where the Nambu Green’s function with Matsubara frequencies \( \nu_m \) is given by

\[ G(k, \nu_m) = \frac{1}{(\nu_m)^2 - E_k^2} \begin{pmatrix} \nu_m + \epsilon_k & \Delta_k \\ \Delta_k & \nu_m - \epsilon_k \end{pmatrix}. \]

(6)

Solving analytically the Matsubara summation we get the forms

\[ \chi_{11}(Q, \omega_n) = \sum_k f_k^2 \int d\omega_1 d\omega_2 \frac{\Delta_k \Delta_{k+q} + \omega_1 \omega_2 - \epsilon_k \epsilon_{k+q} n_F(\omega_1) - n_F(\omega_2)}{2E_k E_{k+q}} \omega_1 - \omega_2 + i\omega_n \]

\[ \times [\delta(\omega_1 - E_k) - \delta(\omega_1 + E_k)] [\delta(\omega_2 - E_{k+q}) - \delta(\omega_2 + E_{k+q})], \]

(7)

\[ \chi_{jj}(Q, \omega_n) = \sum_k j_k f_k \int d\omega_1 d\omega_2 \frac{\Delta_k \omega_2 + \Delta_{k+q} \omega_1 n_F(\omega_1) - n_F(\omega_2)}{2E_k E_{k+q}} \omega_1 - \omega_2 + i\omega_n \]

\[ \times [\delta(\omega_1 - E_k) - \delta(\omega_1 + E_k)] [\delta(\omega_2 - E_{k+q}) - \delta(\omega_2 + E_{k+q})]. \]

(8)

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Figure 1. Evaluation of the Higgs susceptibility using the infrared diagram in Fig. 1(a) of the main text compared to the Raman-like diagram in Fig. 1(b). For small $q$, the infrared diagram is negligible. For increasing values of $q$, the diagram gets finite but shifted to higher energies, reflecting the dispersion of the Higgs mode.

The susceptibilities in (3) are obtained with the analytic continuation $i\omega_n \to \Omega + i\delta$ from these expressions. In the limit $q \to 0$, the susceptibility $\chi_{33}(q = 0, \Omega) = 0$ vanishes. An evaluation for increasing $q$ is shown in Fig. 1 compared to the contribution from the Raman-like diagram (see Sec. II). For small $q$, the infrared contribution is still negligible. For large $q$, the contribution gets comparable to the Raman-like contribution, however is no longer peaked at $\Omega = 2\Delta$ but shifted to higher energies. This reflects the dispersion of the Higgs mode for finite $q$. Thus, the infrared diagram cannot explain a resonance at $\Omega = 2\Delta$ as either its contribution is negligible for small $q$ or not peaked at $\Omega = 2\Delta$ for large $q$.

II. EVALUATION OF THE RAMAN-LIKE DIAGRAM

The susceptibility for the diagram in Fig. 1(b) of the main text is given by

$$\chi^{H}_{\gamma\gamma}(q, \Omega) = H(q, \Omega)\chi_{11}^{2}(q, \Omega),$$

with

$$\chi_{11}(q, i\omega_n) = \sum_k f_k^\gamma \frac{1}{\beta} \sum_{m} \text{Tr} \left[ G(k, i\nu_m) \tau_3 G(k + q, i\nu_m + i\omega_n) \tau_1 \right].$$

Solving analytically the Matsubara summation, we get the form

$$\chi_{\gamma\gamma}(q, i\omega_n) = \sum_k \gamma_k f_k \int_{-\infty}^{+\infty} \frac{\epsilon_k \Delta_{k+q} + \epsilon_{k+q} \Delta_k}{2E_k E_{k+q}} \frac{n_F(\omega_1) - n_F(\omega_2)}{\omega_1 - \omega_2 + i\omega_n} \times [\delta(\omega_1 - E_k) - \delta(\omega_1 + E_k)] [\delta(\omega_2 - E_{k+q}) - \delta(\omega_2 + E_{k+q})].$$

A calculation of the susceptibility in real frequency spectrum for varying values of $q$ is shown in Fig. 1. An evaluation in real frequency spectrum and in the limit $q \to 0$ yields

$$\chi^{H}_{\gamma\gamma}(\Omega) = -2 \sum_k \frac{\gamma_k f_k^2 \epsilon_k \Delta}{E_k(4E_k^2 - 1\beta^2)} \frac{\epsilon_k \Delta_{k+q}}{2E_k E_{k+q}}.$$  

This expression is always nonzero. We can compare the susceptibility with the solution of $\delta \Delta$ from the pseudospin analysis in the next section. Inserting Eq. (31) with just the $\gamma_k^A(s)$ term, written as $\gamma_k^A(s) = \partial_{\epsilon_k}^2 \epsilon_k e^2 A(s)^2$, into the
gap equation, we obtain for $s = i\Omega$
\[
\delta \Delta(\Omega) = V \sum_k \frac{2c_k^2 f_k^2}{E_k(4E_k^2 - \Omega^2)} \delta \Delta(\Omega) - V \sum_k \frac{\partial^2 c_k f_k^2 \epsilon_k \Delta e^2 A(\Omega)^2}{E_k(4E_k^2 - \Omega^2)}. \tag{13}
\]
Solving for $\delta \Delta$ yields just the result (12). The linear pseudospin analysis is therefore equivalent to the Raman-like diagram.

III. PSEUDOSPIN ANALYSIS

We use the BCS Hamiltonian and the time-dependent gap equation $\Delta_k(t) = \Delta(t) f_k$ in the Anderson pseudospin formalism

\[
H = \sum_k b_k \mathbf{\sigma}_k, \quad \Delta(t) = V \sum_k f_k \langle \sigma_k^z \rangle(t), \tag{14}
\]
where the pseudospins are defined as $\mathbf{\sigma}_k = \frac{1}{2} \Psi_k^\dagger \mathbf{\tau} \Psi_k$, with the Nambu-Gorkov spinor $\Psi_k^\dagger = \left( \epsilon_{k\uparrow}, c_{-k\uparrow} \right)$ and $\mathbf{\tau}$ the vector of Pauli matrices. The pairing strength is given by $V$ and the gap symmetry is described by the function $f_k$. Please note that in equilibrium the gap is assumed to be real and we neglect also the induced imaginary part in the time-evolution as its dynamics simply follows the driving without additional features and is therefore unimportant for the following discussion. In equilibrium, all the pseudospins are aligned parallel to the pseudomagnetic field, such that the expectation values for $T = 0$ read

\[
\langle \sigma_k^x \rangle = \frac{\Delta f_k}{2E_k}, \quad \langle \sigma_k^y \rangle = 0, \quad \langle \sigma_k^z \rangle = -\frac{\epsilon_k}{2E_k}. \tag{15}
\]
We use the following ansatz to describe the time-evolution of the pseudospins for small perturbations

\[
\langle \sigma_k^x \rangle(t) = \langle \sigma_k^x \rangle + x_k(t), \quad \langle \sigma_k^y \rangle(t) = \langle \sigma_k^y \rangle + y_k(t), \quad \langle \sigma_k^z \rangle(t) = \langle \sigma_k^z \rangle + z_k(t), \tag{16}
\]
and

\[
\Delta(t) = \Delta + \delta \Delta(t) \tag{17}
\]
with $x_k(t), y_k(t), z_k(t), \delta \Delta(t) \ll 1$. The coupling to the light field $A(t) = A_0 \sin(\Omega t)$ and the supercurrent induced momentum $Q$ is incorporated with the usual minimal coupling $\epsilon_k \rightarrow \epsilon_k - c A(t) - Q$, such that the pseudomagnetic field reads

\[
b_k^\dagger(t) = \left( -2\Delta_k(t), 0, \epsilon_k - c A(t) - Q + \epsilon_{k+c} A(t) + Q \right). \tag{18}
\]
The $z$-component of the pseudomagnetic field can be expanded in powers of $A_0$

\[
b_k^\dagger(t) = 2\epsilon_k + \gamma_k^A(t) + \gamma_k^{AQ}(t) + \gamma_k^{Q^2} + \mathcal{O}(A_0^3), \tag{19}
\]
with

\[
\gamma_k^A(t) = e^2 \sum_{ij} \partial^2_{ij} \epsilon_k A_i(t) A_j(t), \quad \gamma_k^{AQ}(t) = 2e \sum_{ij} \partial^2_{ij} \epsilon_k A_i(t) Q_j, \quad \gamma_k^{Q^2} = \sum_{ij} \partial^2_{ij} \epsilon_k Q_i Q_j. \tag{20}
\]
The term $\sum_i \xi_i \epsilon_k$ vanishes due to particle-hole symmetry. For isotropic band dispersion and under summation over all momentum space, we simplify the derivation of the dispersion by expanding in powers of the dispersion

\[
\gamma_k^A(t) \approx e^2 A_0^2(t)(\alpha_0 + \alpha_1 \epsilon_k), \quad \gamma_k^{AQ}(t) \approx 2eA_0(t)Q(\alpha_0 + \alpha_1 \epsilon_k), \quad \gamma_k^{Q^2} \approx Q^2(\alpha_0 + \alpha_1 \epsilon_k), \tag{21}
\]
where $\alpha_i$ are expansion coefficients. The Bloch equations $\partial_t \langle \mathbf{\sigma}_k \rangle(t) = b_k(t) \times \langle \mathbf{\sigma}_k \rangle(t)$, neglecting higher orders in the deviations, read

\[
\dot{x}_k(t) = -2\epsilon_k y_k(t), \tag{22}
\]
\[
\dot{y}_k(t) = 2\epsilon_k x_k(t) + 2\Delta f_k z_k(t) + \frac{f_k}{E_k} \left( \frac{\Delta}{2} \left( \gamma_k^A(t) + \gamma_k^{AQ}(t) + \gamma_k^{Q^2} \right) - \epsilon_k \delta \Delta(t) \right), \tag{23}
\]
\[
\dot{z}_k(t) = -2\Delta f_k y_k(t). \tag{24}
\]
We apply a Laplace transform from time \( t \) to complex frequency \( s \) to obtain algebraic equations

\[
\begin{align*}
sx_k(s) &= -2\epsilon_k y_k(s), \\
sy_k(s) &= 2\epsilon_k x_k(s) + 2\Delta f_k z_k(s) + \frac{f_k}{E_k} \left( \frac{\Delta}{2} \left( \gamma_k^{A^2}(s) + \gamma_k^{AQ}(s) + \gamma_k^{Q^2}(s) \right) - \epsilon_k \delta \Delta(s) \right), \\
sz_k(s) &= -2\Delta f_k y_k(s),
\end{align*}
\]

where

\[
\begin{align*}
\gamma_k^{A^2}(s) &= e^2 A_0^2 (\alpha_0 + \alpha_1 \epsilon_k) \frac{2\Omega^2}{s(4\Omega^2 + s^2)}, \\
\gamma_k^{AQ}(s) &= 2e A_0 Q (\alpha_0 + \alpha_1 \epsilon_k) \frac{\Omega}{\Omega^2 + s^2}, \\
\gamma_k^{Q^2}(s) &= Q^2 (\alpha_0 + \alpha_1 \epsilon_k) \frac{1}{s}.
\end{align*}
\]

The solution for \( x_k(t) \) reads

\[
x_k(s) = \frac{\epsilon_k f_k \left( 2\epsilon_k \delta \Delta(s) - \Delta \left( \gamma_k^{A^2}(s) + \gamma_k^{AQ}(s) + \gamma_k^{Q^2}(s) \right) \right)}{E_k (4E_k^2 + s^2)}.
\]

Substituting this expression into the gap equation and solving for \( \delta \Delta(s) \), one finds

\[
\delta \Delta(s) = \frac{1}{2} \alpha_1 \Delta \left( e^2 A_0^2 \frac{2\Omega^2}{s(4\Omega^2 + s^2)} + 2e A_0 Q \frac{\Omega}{\Omega^2 + s^2} + Q^2 \frac{1}{s} \right) \left( 1 - \frac{1}{\lambda \int d\varphi f^2 (4\Delta^2 f^2 + s^2) F(s, \varphi)} \right),
\]

where

\[
F(s, \varphi) = \int_{-\epsilon_c}^{\epsilon_c} \frac{1}{2E (4E^2 + s^2)} d\epsilon = \frac{1}{s \sqrt{4\Delta^2 f^2 + s^2}} \sinh^{-1} \left( \frac{s}{2\Delta |f|} \right).
\]

Here, we replace the momentum sum with an integral

\[
\sum\limits_\mathcal{V} k \rightarrow \lambda \int_{-\epsilon_c}^{\epsilon_c} d\epsilon \int_0^{2\pi} d\varphi
\]

assuming \( \epsilon_k = \epsilon(|k|) \) and \( f_k = f(\varphi) \) and using \( \lambda = V D(\epsilon_F) \), where the density of states \( D(\epsilon_F) \) is assumed to be constant near the Fermi energy. There are three contributions which determine the dynamics of the gap, resulting from the terms \( \gamma_k^{A^2}(t) \), \( \gamma_k^{AQ}(t) \), \( \gamma_k^{Q^2}(t) \). Thus we write

\[
\delta \Delta(s) = \delta \Delta_{A^2}(s) + \delta \Delta_{AQ}(s) + \delta \Delta_{Q^2}(s).
\]

The solution in time domain is obtained by an inverse Laplace transform, where the Bromwich integral

\[
\delta \Delta(t) = \frac{1}{2\pi i} \int_{\gamma - i\infty}^{\gamma + i\infty} e^{s} \delta \Delta(s) ds
\]

has to be evaluated for \( \gamma \in \mathbb{R} \) larger than any poles of the integrand. The integration can be extended into a closed loop contour integral in the complex plane as shown in Fig. 2. The contribution in the long-time limit consisting of the driven oscillations at which we are interested in, can be obtained by evaluating the residues for the poles. For \( \delta \Delta_{A^2}(s) \) we have

\[
\text{Res}_0 (e^{s} \delta \Delta_{A^2}(s)) = \frac{1}{4} \alpha_1 \Delta e^2 A_0^2 \left( 1 - \frac{1}{\lambda \int d\varphi f^2} \right),
\]

\[
\text{Res}_{\pm 2i\Omega} (e^{s} \delta \Delta_{A^2}(s)) = \frac{1}{8} \alpha_1 \Delta e^2 A_0^2 \left( e^{\pm 2i\Omega} \frac{\Omega e^{\pm 2i\Omega}}{\lambda \int d\varphi f^2 \sqrt{\Delta^2 f^2 - \Omega^2} \sin^{-1} (\frac{\Omega}{\Delta |f|})} \right).
\]
Figure 2. Bromwich integral from Eq. (36) extended into a closed loop contour integral in the complex plane. There is a continuous line of branch points at \( s = \pm 2i\Delta f \) (depending on \( f \)) and poles at \( s = 0, \pm 2i\Omega \) for \( \delta \Delta_{A2} \), at \( s = 0 \) for \( \delta \Delta_{AQ} \), and at \( s = 0 \) for \( \delta \Delta_{Q2} \).

An evaluation of the residues for the poles in \( \delta \Delta_{AQ}(s) \) yields

\[
\text{Res}_{\pm i\Omega}(e^{s:\delta \Delta_{AQ}(s)}) = \frac{1}{2} \alpha_{1} \Delta e A_{0} Q \left( \frac{\pm i e^{\pm i\Omega}}{\frac{i \Omega e^{\pm i\Omega}}{\lambda \int d\varphi f^{2} \sqrt{4\Delta^{2} f^{2} - \Omega^{2} \sin^{-1}\left( \frac{\Omega}{2 \Delta |f|} \right)}}} \right). \quad (39)
\]

The expression \( \delta \Delta_{Q2}(s) \) just adds an offset, which is not interesting for our discussion. Combining the expressions, we find

\[
\delta \Delta_{A2}(t) = \alpha_{1} \Delta e^{2} A_{0}^{2} \left( \frac{1}{2} \sin(\Omega t)^{2} - \frac{1}{4 \lambda \int d\varphi f^{2}} - \frac{\Omega \cos(2\Omega t)}{4 \lambda \int d\varphi f^{2} \sqrt{\Delta^{2} f^{2} - \Omega^{2} \sin^{-1}\left( \frac{\Omega}{2 \Delta |f|} \right)}} \right), \quad (40)
\]

\[
\delta \Delta_{AQ}(t) = \alpha_{1} \Delta e A_{0} Q \left( \sin(\Omega t) + \frac{\Omega \sin(\Omega t)}{\lambda \int d\varphi f^{2} \sqrt{4\Delta^{2} f^{2} - \Omega^{2} \sin^{-1}\left( \frac{\Omega}{2 \Delta |f|} \right)}} \right). \quad (41)
\]

As shown in the main text in Fig. 2, the \( A^{2} \) driving leads to a \( 2\Omega \) oscillation of the order parameter, which resonates at \( 2\Omega = 2\Delta \), while the \( AQ \) driving term leads to a \( \Omega \) oscillation, resonating at \( \Omega = 2\Delta \). An evaluation for \( d \)-wave symmetry with \( f(\varphi) = \cos(2\varphi) \) is shown in Fig. 3.

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Figure 3. Amplitudes of the induced gap oscillations from Eq. (14) of the main text for $s$- and $d$-wave superconductor with $f(\varphi) = 1$ and $f(\varphi) = \cos(2\varphi)$. Resonances of the driving light with the Higgs mode appear at $2\Omega = 2\Delta$ without and at $\Omega = 2\Delta$ with supercurrent.