Incoherent Excitation of Coherent Higgs Oscillations in Superconductors

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We investigate theoretically the excitation of Higgs oscillations of the order parameter in superconductors by incoherent short pulses of light with frequency much larger than the superconducting gap. The excitation amplitude of the Higgs mode is controlled by a single parameter which is determined by the total number of quasiparticles excited by the pulse. This fact can be traced back to the universality of the shape of the light-induced quasiparticle cascade at energy below the Debye frequency and above the gap.

The dynamics of the superconducting state, described by BCS theory, has been a subject of research for a long time1–10. There are two important time scales characterizing the dynamics of the system: the quasiparticle energy relaxation time \( \tau_{in}(\epsilon) \) and \( 1/\Delta \), where \( \epsilon \) is the quasiparticle energy and \( \Delta \) is the value of the order parameter. If the temperature \( T \) is not too close to the transition, the quasiparticle energy relaxation rate is much smaller than the superconducting energy gap:

\[
\frac{1}{\tau_{in}(T)} \ll \Delta. \tag{1}
\]

In this case, for processes with frequency \( \omega \ll \Delta \), the density of states is a local function of time and the low frequency dynamics of the superconductor is described by a Boltzmann kinetic equation for the quasiparticle distribution function \( n(\epsilon, t) \) and a self-consistent equation for \( \Delta(t) \) (see, e.g., 11).

At frequency \( \omega \approx \Delta \), the Boltzmann approach fails. Rather, for time \( t \ll \tau_{in} \), the dynamics are governed by non-dissipative equations which conserve both the entropy and the total energy. As a result, the system exhibits coherent oscillations of the order parameter1, which in the linearized regime decay slowly

\[
\delta \Delta(t) = B \frac{\cos(\omega_H t + \phi)}{\Delta_0^2 t^{3/2}}. \tag{2}
\]

Here \( \omega_H = 2\Delta_0 \) is the Higgs frequency, \( \Delta_0 \) is the equilibrium gap, and the parameters \( B \) and \( \phi \) depend on the initial conditions.

Since the Higgs mode is scalar, it cannot couple linearly to electromagnetic fields directly. Rather, several excitation mechanisms have been studied: via combined dynamics of the Higgs mode with charge density wave oscillations12,13, linear excitation by coherent THz electromagnetic waves in the presence of DC super-current14,15, and nonlinear coherent excitation3,16 using high intensity THz light.

In this work, we discuss excitation of the Higgs mode by incoherent short light pulses with duration \( \tau_{in} \ll 1/\Delta_0 \) and frequency \( \Omega_0 \gg \Delta_0 \). The physical picture of the mechanism is the following: the pulse creates non-equilibrium quasiparticles with characteristic energy \( \epsilon \gg \Delta_0 \). Initially, these quasiparticles are not effective at exciting the Higgs mode because they have high energy and the relaxation rate \( 1/\tau_{in} \) of their distribution is much faster than \( \Delta_0 \). As their energy decreases due to various inelastic processes, the relaxation rate decreases as well. When the typical energy \( \epsilon \) is smaller than the Debye energy \( \Omega_D \) but still much larger than \( \Delta_0 \), the relaxation is controlled by phonon emission with rate\(^{17} \)

\[
\tau_{in}^{-1}(\epsilon) = \gamma \epsilon^3 \tag{3}
\]

where \( \gamma = \alpha/\Omega_D^3 \), and \( \alpha \) is a coefficient of order one, just as in a normal metal.

The optimal coupling between the quasiparticle cascade and the Higgs mode is achieved when the rate of change of the quasiparticle distribution function is of order \( \omega_H \). This provides an estimate of the characteristic energy at this stage of the relaxation process,

\[
\epsilon^* \equiv \left(2\Delta_0 \Omega_D^2/\alpha\right)^{1/3} \tag{4}
\]

It is important that \( \epsilon^* \gg \Delta_0 \): at such high energy, superconducting correlations are negligible, and we may approximate \( n(\epsilon, t) \) with the solution of the Boltzmann equation for a normal metal:

\[
\frac{\partial n(\epsilon, t)}{\partial t} = \gamma \int \frac{d\epsilon'}{(\epsilon - \epsilon')^2} n(\epsilon', t) - \frac{1}{3} \epsilon^3 n(\epsilon, t) \tag{5}
\]

Here, we assume the intensity of the exciting pulse is small and neglect terms nonlinear in \( n \) as well as stimulated emission of phonons.

It is furthermore possible to neglect phonon reabsorption during the cascade down to \( \epsilon^* \), because the phonon reabsorption rate is much smaller than the inelastic electron relaxation rate \( 1/\tau_{ph} \ll 1/\tau_{in} \) in the energy interval \( [\epsilon^*, 2\Delta_0] \). Indeed, the phonon reabsorption rate is \( 1/\tau_{ph} \approx \omega c/v_F \) at \( \Delta_0 \ll 1 \) and \( 1/\tau_{ph} \approx \omega^2 \tau \) at \( q l \gg 1 \). Here \( \omega \) and \( q \) are the frequency and the wave vector of phonons, \( c \) and \( v_F \) are the speed of sound and the Fermi velocity, \( l \) and \( \tau \) are the electron mean free path and mean free time. The desired inequality follows from the fact that \( c \ll v_F \).

At \( \epsilon \ll \Omega_0 \ll \Omega_D \) the dynamics described by equation (5) has no scales, and the distribution function approaches a scaling form

\[
n(\epsilon, t) = C \Omega_D (\gamma t)^{1/3} N \left( \left[ \frac{t}{\tau_{in}(\epsilon)} \right]^{1/3} \right) \tag{6}
\]

where \( N(u) \) is a scaling function satisfying the equation

\[
u N''(u) + (1 + u^3) N(u) = 3 \int du' (u - u')^2 N(u') \tag{7}
\]
Green functions are independent of the direction of $\mathbf{p}$ and can be parametrized by $\xi$ alone, $G^K_{\alpha\beta}(t; \mathbf{p}, \omega) = G^K_{\alpha\beta}(t; \xi, \omega)$.

We generalize the equations obtained in Ref.\(^1\) for $g$ and $f$ taking into account inelastic electron-phonon processes in the first order of perturbation theory,

\begin{align}
\frac{i\partial_t \delta g + 2\Delta_0 \Re[\delta f]}{\partial_t} &= I_{11} \quad (10a) \\
\left(i\partial_t - 2\xi\right) \delta f + 2g_{eq} \delta \Delta + 2\Delta_0 \delta g &= I_{12} \quad (10b) \\
\delta \Delta &= -\frac{\lambda_{\text{BCS}}}{2} \int d\xi \Im[\delta f] \quad (10c)
\end{align}

where $\lambda_{\text{BCS}}$ is the dimensionless BCS coupling, and

\begin{align}
I_{11} &= 2\gamma \text{sign}(\xi) \left(\frac{1}{3} \xi^3 n(\epsilon, t) \right) \\
&\quad - \int_{\epsilon}^{\infty} d\epsilon' \left( \epsilon - \epsilon' \right)^2 n(\epsilon', t) \quad (11a)
\end{align}

\begin{align}
I_{12} &= 2\gamma \Delta_0 \left( n(\epsilon, t) \right) \int_{\epsilon - \Delta}^{\epsilon} d\epsilon' \left( \epsilon - \epsilon' \right)^2 \left( \frac{1}{\epsilon} - \frac{1}{\epsilon'} \right) \\
&\quad - \int_{\epsilon - \Delta}^{\epsilon} d\epsilon' \left( \epsilon - \epsilon' \right)^2 \left( \frac{1}{\epsilon} - \frac{1}{\epsilon'} \right) n(\epsilon', t) \quad (11b)
\end{align}

Eqs. (10) and (11) describe the linearized dynamics of small deviations around the equilibrium solution

\begin{align}
f_{eq} &= -i\frac{\Delta_0}{\epsilon} \left( 1 - 2n_F(T) \right) \quad g_{eq} = -i\frac{\xi}{\epsilon} \left( 1 - 2n_F(T) \right) \quad (12)
\end{align}

where $n_F(T) = (1 + \exp(\epsilon/T))^{-1}$ is the equilibrium Fermi distribution and $\epsilon = \sqrt{\xi^2 + \Delta_0^2}$. Eq. (11) is valid for energies $\epsilon \ll \Delta$, where we need not distinguish the quasiparticle energy from $|\xi|$. We also take into consideration that, for optical excitation, the quasiparticle distribution $n$ is even in $\xi$ and follows the metallic cascade governed by Eq. (6).

We sketch details of the derivation of Eqs. (10) and (11) in the Appendix. As we are interested in the case $T \ll \Delta_0$, we approximate $1 - 2n_F(T) \approx 1$. Furthermore, for optical excitation, Re $[\delta f]$ is odd in $\xi$, while Im $[\delta f]$ and $\delta g$ are even in $\xi$ at all times.

As the excitation of $\Delta(t)$ is controlled by energies $\epsilon \sim \epsilon^* \gg \Delta$, we can substitute the solution for the quasiparticle cascade in the normal metal (Eq. (6)) into the RHS of Eq. (11). In Fig. 2 we present an example trace of $\Delta(t)$ with these excitation conditions.

Taking the Fourier transform of Eq. (10), solving for $\delta f$ in Eqs. (10a,b) and substituting the result into Eq. (10c), we obtain

\begin{align}
\delta \Delta(\omega) &= \frac{1}{\omega F(\omega)} \left( \frac{4\Delta_0}{\omega^2 - 4\Delta_0^2} \left( \frac{\xi I_{11}(\xi, \omega)}{\omega^2 - 4\Delta_0^2} - \frac{I_{12}(\xi, \omega)}{\omega^2 - 4\Delta_0^2} \right) \right) \quad (13)
\end{align}

where the angle brackets are shorthand for

\begin{align}
\langle \ldots \rangle &\equiv \frac{\lambda_{\text{BCS}}}{2} \int_{-\Omega_D}^{\Omega_D} d\xi \ldots \quad (14)
\end{align}
The semicircle $R$ on the real axis at $\omega = \pm 2\Delta_0$, which is equivalent to the integral along the contour $L$ in Fig. 3. Using the Cauchy residue theorem we have

$$\delta \Delta(t) = i \text{Res}(\delta \Delta(\omega), 0) + \int_{\omega_1} d\omega \frac{e^{-i\omega t} \delta \Delta(\omega)}{2\pi}$$

(17)

We focus on the second term in Eq. (17) which describes the oscillating part of $\Delta(t)$. The first term describes the mean value of $\Delta(t)$ at late time, which is determined physically by quasiparticle processes at energies of order the gap $\Delta$ where our perturbative expressions for $I_{11}$ and $I_{12}$ are no longer valid.

Close to the branch points of $F(\omega)$, we have

$$\left\langle \frac{4\Delta_0 \xi I_{11}(\xi, \omega)}{\omega^2 - 4\epsilon^2} \right\rangle \approx i\pi \frac{\Gamma(1/3)}{6} \text{sign} \text{Re} \omega$$

(18)

where $C$ is the normalization constant in Eq. (6) and

$$a(\omega) = -\frac{\pi}{\Gamma(-1/3)} + i\frac{\Gamma(1/3)}{6} \text{sign} \text{Re} \omega$$

(19)

The dependence on the sign of $\omega$ is due to $I_{11}$ being the Fourier transform of a real function.

It turns out that the term including $I_{12}$ gives a subleading contribution at long times – the amplitude decays as $1/t$, which is negligible compared to $1/\sqrt{t}$ as $t \to \infty$. Taking the inverse Fourier transform of Eq. (13), we arrive at Eq. (2) where the values of the parameters $B, \phi$ are

$$B = \left( \frac{2}{\pi} \right)^{3/2} \frac{\pi|a| N(0)}{\nu(0) \Omega_D} \frac{\Omega_D}{e^*} \quad \phi = \pi/4 + \arg(a)$$

(20)

where $a = -\pi/\Gamma(-1/3) + i\Gamma(1/3)/6$ so that $|a| \approx 0.893$. With our choice of normalization, $N(0) \approx 1.44$.

Thus, we arrive at the conclusion that the excitation amplitude of the Higgs mode is controlled by a single parameter which is determined by the total number of quasiparticles excited by a short light pulse with duration $\tau_{im} < 1/\Delta$. This fact can be traced back to the universality of the shape of the quasiparticle cascade $n(\epsilon, t)$ at energy $\epsilon \sim e^*$.

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The encode the effect of inelastic processes on the dynamics: Keldysh technique\textsuperscript{19} in the spatially uniform case. These were derived in Ref.\textsuperscript{1} ignoring the quasiparticle relaxation processes, which we include to leading order in perturbation theory.

We work with the fermionic $G$ and phononic $D$ Green functions in the uniform case,

\begin{align}
G_{\alpha\beta}(p; t_1, t_2) &\equiv -i \langle T_C \psi_\alpha(p, t_1) \psi_\beta^\dagger(p, t_2) \rangle \\
D_{ij}(p; t_1, t_2) &\equiv -i \langle T_C \phi(p, t_1) \phi(-p, t_2) \rangle
\end{align}

where $T_C$ is the contour ordering symbol, $i, j$ are Keldysh indices, and $\alpha, \beta$ are Nambu indices. Below, after the Keldysh rotation, we denote the retarded, advanced and Keldysh components of the Green functions $G^R, G^A, G^K$ (and similarly for $D, \Sigma$) explicitly.

The equations of motion follow from the matrix Dyson equation. The Keldysh components yield,

\begin{align}
i \partial_t G_{11}^K + \Delta G_{12}^{K*} + G_{12}^K \Delta^* &= I_{11} \tag{A3a} \\
i \partial_t G_{12}^K - 2\xi G_{12}^K - \Delta G_{11}^{K*} + G_{11}^K \Delta &= I_{12} \tag{A3b}
\end{align}

where $\xi = p^2/(2m) - \epsilon_F$ and

\[ \Delta = \frac{1}{2} (\Sigma_{12}^R + \Sigma_{12}^A) \tag{A4} \]

is the order parameter. Equation (A3) corresponds to Eq. (16) of Ref\textsuperscript{4}. The RHS, $I_{11}$ and $I_{12}$, are formally given by

\begin{align}
I_{11} &= \Sigma_{11}^K C_{11}^A - G_{11}^R \Sigma_{11}^K - \Sigma_{11}^R G_{11}^K - G_{11}^R \Sigma_{11}^A + \Sigma_{12}^K G_{12}^{A*} + G_{12}^R \Sigma_{12}^K - \Sigma_{12}^R G_{12}^{A*} + \Sigma_{12}^R - \Sigma_{12}^A - \frac{\Sigma_{12}^R - \Sigma_{12}^A}{2} \tag{A5a} \\
I_{12} &= \Sigma_{11}^K C_{12}^A - G_{12}^R \Sigma_{12}^K - \Sigma_{12}^K G_{11}^{A*} - G_{11}^R \Sigma_{12}^K - \Sigma_{11}^R G_{12}^{A*} + G_{12}^R \Sigma_{11}^K - \Sigma_{11}^R G_{12}^{A*} + \Sigma_{12}^R - \Sigma_{12}^A - \frac{\Sigma_{12}^R - \Sigma_{12}^A}{2} \tag{A5b}
\end{align}

These encode the effect of inelastic processes on the dynamics: $I_{11}$ yields the Boltzmann collision integral in the metal when $\Delta \to 0$ ($I_{12} \to 0$ in this limit).

All operator products in Eq. (A3) and (A5) are space-time contractions. Also, $\partial_\tau$ denotes the derivative with respect to average time after changing time coordinates to $t = (t_1 + t_2)/2$ and $\tau = t_1 - t_2$.

We compute the self-energy $\Sigma$ to leading order in the self-consistent Born approximation\textsuperscript{20} which gives us

\[ \Sigma_{ij}^{\alpha\beta}(x, y) = i(-1)^{\alpha+\beta+i+i+j} C_{\alpha\beta}(x, y) D^{ij}(x, y) \tag{A6} \]
\[ G_{11}^R(\xi; \omega) = (G_{11}^A(\xi, \omega))^* = \frac{u_\xi^2}{\omega - \varepsilon + i0^+} + \frac{v_\xi^2}{\omega + \varepsilon + i0^+} \]  

(A7a)

\[ G_{11}^K(\xi; \omega, t) = -2\pi i (1 - 2n(\xi, t)) \left( u_\xi^2 \delta(\omega - \varepsilon) - v_\xi^2 \delta(\omega + \varepsilon) \right) \]  

(A7b)

\[ G_{12}^R(\xi; \omega) = (G_{12}^A(\xi, \omega))^* = -\frac{u_\xi v_\xi}{\omega + \varepsilon + i0^+} + \frac{u_\xi v_\xi}{\omega - \varepsilon + i0^+} \]  

(A7c)

\[ G_{12}^K(\xi; \omega, t) = -2\pi i u_\xi v_\xi (1 - 2n(\xi, t)) \left( \delta(\omega + \varepsilon) + \delta(\omega - \varepsilon) \right) \]  

(A7d)

\[ D^R(k; \omega) = (D^A(k; \omega))^* = |m_k|^2 \frac{\omega_k}{(\omega + i0^+)^2 - \omega_k^2} \]  

(A7e)

\[ D^K(k; \omega, t) = -i\pi |m_k|^2 \left( \delta(\omega + \omega_k) + \delta(\omega - \omega_k) \right) \]  

(A7f)

where

\[ u_\xi^2 = \frac{1}{2} \left( 1 + \frac{\xi}{\varepsilon} \right) \quad v_\xi^2 = \frac{1}{2} \left( 1 - \frac{\xi}{\varepsilon} \right) \]  

(A8)

with \( \varepsilon = \sqrt{\xi^2 + \Delta_0^2} \). In these expressions, \( |m_k|^2 \) is the squared electron–phonon matrix element (which is linear in \( k \)), and \( \omega_k = ck \) is the phonon dispersion. Finally, linearizing the resulting equations, we obtain Eq. (10).