Higgs Amplification in Non-Equilibrium $K_3C_{60}$

Daniel Podolsky
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With:
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Background: Collective modes of a superconductor

Anderson-Higgs mechanism:
- Goldstone mode “eaten” by gauge field, gapped to plasma frequency
- Leads to Meisner effect

Higgs mode:
- Fluctuations in order parameter amplitude.
- Gapped excitation.
- Not charged.
Motivation: Non-Equilibrium Superconductivity

Higgs Oscillations in Nb$_{1-x}$Ti$_x$N

PRL 111, 057002 (2013)

Photo-induced superconductivity in K$_3$C$_{60}$

Nature 530, 461 (2016)
Background: Higgs Oscillations in Nb$_{1-x}$Ti$_x$N

Higgs mode is not charged → doesn’t couple linearly to light

Transmission through thin film of Nb$_{1-x}$Ti$_x$N, following pump pulse

Pump Energy (nJ/cm$^2$) vs. $t_{pp}$ (ps)

PRL 111, 057002 (2013)
Background: Photoinduced superconductivity in $K_3C_{60}$

M. Mitrano et al., Nature 530, 461 (2016)

Equilibrium: $T=25$ K and $T=10$ K

$\sigma_1(\omega)$ ($\Omega^{-1}$ cm$^{-1}$) vs Energy (meV)

$\sigma_2(\omega)$ ($\Omega^{-1}$ cm$^{-1}$) vs Energy (meV)
Background: photoinduced superconductivity in K$_3$C$_{60}$

Response — Equilibrium vs Photoexcited:

M. Mitrano et al., Nature 530, 461 (2016)
Photoinduced superconductivity — Possible mechanism

Parametric enhancement of electron-phonon interaction

Part I: Higgs Amplification in $K_3C_{60}$
Can we suggest a new probe for light-induced SC?

Challenge: very short-lived state (~10 ps)
→ only optical measurements available

Idea: A sudden quench into SC state gives rise to large Higgs oscillations

Are there signatures of such oscillations in the optical conductivity?
Higgs-photon coupling

Coupling between superfluid stiffness and light:

\[ \mathcal{H}_{\text{diamagnetic}} = \frac{e^2 \hbar^2}{2mc^2} \rho_s A^2 \]
\[ \rho_s = \rho_{s,0} + \delta \rho \cos(\omega_H t) \]

Upon quantization, this yields:

\[ A \sim \sum_\omega (a_\omega + a_\omega^\dagger) \]

\[ \mathcal{H}_{\text{Higgs/photons}} \sim \sum_{\omega_1+\omega_2=\omega_H} (\delta \rho a_{\omega_1}^\dagger a_{\omega_2}^\dagger + \text{h.c.}) \]

Photons in bulk are gapped to plasma frequency (Anderson-Higgs mechanism).

Near the surface they remain gapless

\( \rightarrow \) Coupling is important for reflection problem
Photon pair creation

$$\mathcal{H}_{\text{Higgs/photons}} \sim \sum_{\omega_1 + \omega_2 = \omega_H} (\delta \rho a^\dagger_{\omega_1} a^\dagger_{\omega_2} + \text{h.c.})$$

**incoming**

$\omega_s$

$N$ photons

$\omega_H = \omega_s + \omega_i$

**outgoing**

idler $\omega_i$

1 photon

signal $\omega_s$

$N + 1$ photons
“Nonlinear optics” involving Higgs mode

\[ \mathcal{H}_{\text{Higgs/photons}} \sim \sum_{\omega_1 + \omega_2 = \omega_H} (\delta \rho a^\dagger_{\omega_1} a^\dagger_{\omega_2} + \text{h.c.}) \]

Three-wave mixing (one Higgs mode and two photons) and parametric resonance. Frequency \( \omega_1 \) is selected by the probe beam due to Bose enhancement factor. In the language of parametric resonance, \( \omega_1 \) is a signal wave and \( \omega_2 \) is an idler wave.
Electrodynamics of SC with excited Higgs mode

Maxwell’s equations to analyze reflection:

\[ \nabla \times \mathbf{B} - \frac{\epsilon}{c^2} \frac{\partial \mathbf{E}}{\partial t} = \mu_0 \mathbf{j} \]

\[ \nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = 0 \]

Response of the medium is determined by the superfluid density \( \Lambda \)

London equation:

\[ \mathbf{j} = \Lambda(t) \left( \frac{\hbar}{2e} \nabla \theta - \mathbf{A} \right) \]

\[ \Lambda(t) = \Lambda_s + 2\Lambda_m \cos(\omega_H t) \]

Solve the Fresnel-Floquet reflection problem
Floquet modes inside medium

Evanescent waves inside medium: \( D \in \{E, B, j\} \)

\[
D = (D_1 e^{-i\omega_1 t} + D_2^* e^{i\omega_2 t}) e^{\kappa z} + \text{c.c.}
\]

London equation mixes frequencies

\[
\omega_1 + \omega_2 = \omega_H
\]

Matrix equation for \( E \):

\[
\begin{pmatrix}
\kappa^2 c^2 + \omega_1^2 \epsilon_1 & \Lambda_m \omega_1 / \omega_2 \\
\Lambda_m \omega_2 / \omega_1 & \kappa^2 c^2 + \omega_2^2 \epsilon_2
\end{pmatrix}
\begin{pmatrix}
E_1 \\
E_2^*
\end{pmatrix} = 0
\]

Solution:

\[
\kappa_\pm^2 c^2 = -\frac{\omega_1^2 \epsilon_1 + \omega_2^2 \epsilon_2}{2} \pm \frac{1}{2} \sqrt{(\omega_1^2 \epsilon_1 - \omega_2^2 \epsilon_2)^2 + 4\Lambda_m^2}
\]

\[
\begin{align*}
\epsilon_1 &\equiv \epsilon(\omega_1) \\
\epsilon_2 &\equiv \epsilon(-\omega_2)
\end{align*}
\]
Fresnel-Floquet reflection problem

Boundary conditions imposed independently on each frequency:

\[ B_1^i + B_1^r = B_{1,+}^t + B_{1,-}^t \]
\[ E_1^i + E_1^r = E_{1,+}^t + E_{1,-}^t \]
\[ B_2^{r*} = B_{2,+}^{t*} + B_{2,-}^{t*} \]
\[ E_2^{r*} = E_{2,+}^{t*} + E_{2,-}^{t*} \]
Results — Normal incidence

Effect is weak because light doesn’t penetrate deeply into the sample:

\[ R_{\text{max}} = 1 + \frac{1}{4} \frac{\Lambda_m^2}{\Lambda_s^2} \frac{\epsilon_{\text{out}}}{\epsilon_{\text{in}}} \frac{\omega_H^2}{\omega_p^2} \]

Two ways to enhance: shallow incidence or small plasma frequency
Shallow incidence

Reflectivity of signal

\[ \omega_H = 0.4\omega_p \]

Intensity of idler mode

\[ \omega_H = 0.4\omega_p \]

Blue \( \omega_1=0.1\omega_p \), orange \( \omega_1=0.2\omega_p \), green \( \omega_1=0.3\omega_p \).

Dependence on polarization: \( p \)-polarization gives stronger effect

There is an analogue of total internal reflection: for very shallow angle, idler is not allowed by conservation laws. Large signal right before the transition.
Small plasma frequency

Reflectivity of signal

Intensity of idler mode

Normal incidence, $\omega_H = 1.3 \omega_p$

Light penetrates into bulk of SC
$\rightarrow$ Reflectivity very sensitive to dissipation
Higgs amplification in $K_3C_{60}$

Low electron density (3 per $C_{60}$ molecule)
Weak hopping between $C_{60}$ molecules

$\rightarrow$ Small plasma frequency $\omega_p = 72$meV

Photo-induced superconductivity has onset at $\sim 120$ K

$\rightarrow$ Estimate for Higgs frequency $\omega_H \approx 2\Delta = 30$meV

In order to induce Higgs oscillations, need quench faster than

$$\tau^* = \frac{h}{\omega_H} \approx 300 \text{ fs}$$
Experiment — Pulse width dependence

(a) Slow Quench (1.8 ps)

(b) Prompt Quench (0.1 ps)

$\tau \gg \tau^*$

$\tau \ll \tau^*$
Comparing model to experiment

Maximum amplification (6%) at 10 meV

This suggests that Higgs frequency is somewhat higher than 20 meV

Good match to data for

$\omega_H = 24 \text{ meV}$
In this paper, we have introduced the phenomenon of light-induced superconductivity. We have shown that, by illuminating a superconductor with pump pulses, one can excite a Higgs amplification and provided strong evidence for it in experiments on the light-induced superconducting state.

The measured optical conductivity of the unmodulated superconductor with parameters fixed by the onset of superconductivity, shows the complex effective optical conductivity more strongly. Note that, in order to preserve the shorter pulses, which can occur since the pump drives the system nonlinearly. Therefore, even though the total Higgs amplification and provided strong evidence for it in experiments on the light-induced superconducting state.

The Lorentzians represent the broad mid-infrared optical conductivity of the superconductor with parameters explored in Sec. IV, which one of the photons matches the incident photons. In addition to its interest as fundamental physics, the energy of the excited Higgs mode is converted to outgoing light with two frequencies, which interacts directly with the superconducting state.

The measured optical conductivity of the superconductor is shown in Fig. 8, which corresponds to the reflectivity, are effects on the reflectivity. There are only two parameters we allow to be fixed: the Drude peak we replace by a Lorentzian, see Sec. V. The corresponding reflectivity is given by the complex conductivity, and the amplitude of the reflectivity is larger for longer pulses, which can occur since the pump drives the system nonlinearly. Therefore, even though the total Higgs amplification and provided strong evidence for it in experiments on the light-induced superconducting state.

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Amplification is robust to Higgs decay

Higgs oscillations decay (into quasiparticle pairs or vacuum photons)

This results in oscillations with a broadened spectral weight

To leading order in the amplitude of oscillations, amplification is an integral over this spectral weight

Idler’s frequency broadened, making it more difficult to observe
Transient superconductivity without superconductivity

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FIG. 2. (a) Sketch of incident pump pulse leading to spontaneous polarization in the active layer. (b) Sketch of the \( j-E \) characteristic for a \( \sigma < 0 \) state. (c) Entropy and charge density profiles inside the active layer; notice the thin surface charge accumulation on the external boundary of the layer. (d) Incident and reflected probe waves at an angle \( \theta \); the reflection occurs in the \( x-z \) plane and the magnetic field is along the \( y \) axis for TM polarization.

FIG. 3. (Color online) (a) Plot of the real (solid line) and imaginary (dashed line) part of \( Y(\omega) \). (b) Theoretical \( R(\theta) \) of \( K_3C_60 \) at \( \omega = 6 \text{ meV} = 1.44 \text{ THz} \) in equilibrium (red) and non-equilibrium (blue); notice the marked dependence on the angle and the presence of a region where \( R > 1 \). Plots are for the parameters of Fig. 1a.
Further experimental checks?

Idler beam?

Angular dependence?

Direct observation of Higgs oscillations?
Higgs amplifier in other types of system?

Electrodynamics of CDW with excited Higgs mode

In the frequency range between the pinning frequency and the quasiparticle gap frequency, dynamics of the incommensurate CDW materials can also be described by the London equation

\[
\frac{dj}{dt} = \Lambda E(t)
\]

Gruner, Density waves in solids
Frontiers in Physics
Part II: Phonon laser
(adventures in the complex plane)
Motivation: Parametric amplification of optical phonons

SiC — Bulk insulator with IR-active optical phonon at \( \omega_p = 24 \text{ THz} \)

Fig. 3. (A) Crystal structure of SiC, polytype 4H (space group \( C_{6v}^4-P6_3mc \)). Si atoms in blue, C atoms in red. (B) Eigenvectors of the infrared active mode excited by the pump pulse (\( E_u \) symmetry). (C) Reflectivity at equilibrium associated to the driven mode [data from literature (24)].
Phonon coupled to EM field

Potential for phonon and electric field:

\[ V = \frac{\Omega_{\text{TO}}^2}{2} Q^2 - \left( \frac{\epsilon_0(\epsilon_\infty - 1)}{2} + \beta Q^2 \right) E^2 - (\eta + \alpha Q^2)Q \cdot E, \]

Phonon EOM:

\[ \ddot{Q} + \Gamma \dot{Q} = - \frac{\partial V}{\partial Q} = - \frac{\Omega_{\text{TO}}^2}{2} Q + \eta E + 2\beta E^2 Q + 3\alpha Q^2 E \]

Maxwell equations:

\[ \frac{1}{\mu_0} \nabla \times B - \partial_t (\epsilon_0 E + P) = 0 \]

\[ \nabla \times E + \partial_t B = 0 \]

Polarization:

\[ P = - \frac{\partial V}{\partial E} = \epsilon_0(\epsilon_\infty - 1)E + (\eta + \alpha Q^2)Q + 2\beta Q^2 E \]

Combined:

\[ - \nabla^2 E + \frac{1}{c^2} \partial_t^2 \left( \frac{\epsilon_\infty E}{\epsilon_0} + \frac{\eta}{\epsilon_0} Q + \frac{\alpha}{\epsilon_0} Q^3 + 2\frac{\beta}{\epsilon_0} Q^2 E \right) = 0 \]
Polaritons

Linearized equations of motion:

\[ \begin{align*}
\ddot{Q} + \Gamma \dot{Q} + \Omega_{\text{TO}}^2 Q &= \eta E \\
- \nabla^2 E + \frac{1}{c^2} \partial_t^2 \left( \epsilon_\infty E + \frac{\eta}{\epsilon_0} Q \right) &= 0
\end{align*} \]

\[ Q(t) = Q_p \cos(\omega_p t) \]
Frequency mixing

When polariton is excited, the non-linear equations of motion

\[ \ddot{Q} + \Gamma \dot{Q} = -\Omega_{TO}^2 Q + \eta E + 2\beta E^2 Q + 3\alpha Q^2 E \]

\[-\nabla^2 E + \frac{1}{c^2} \partial_t^2 \left( \epsilon_\infty E + \frac{\eta}{\epsilon_0} Q + \frac{\alpha}{\epsilon_0} Q^3 + \frac{2\beta}{\epsilon_0} Q^2 E \right) = 0. \]

mix frequencies: \( \omega_1 + \omega_2 = 2\omega_p \)

\[ Q = Q_p \cos(\omega_p t) + \left( Q_1 e^{-i\omega_1 t} + Q_2^* e^{i\omega_2 t} \right) e^{ikz} \]

\[ E = E_p \cos(\omega_p t) + \left( E_1 e^{-i\omega_1 t} + E_2^* e^{i\omega_2 t} \right) e^{ikz} \]

polariton, strongly driven by pump

weak fields, from probe
Results: Reflectivity of semi-infinite medium

Discontinuities!!!
Finite slab

\[ a \equiv r_{21}^2 e^{2ikL} \]

\[ r = r_{12} + t_{12}t_{21}r_{21}e^{2ikL}(1 + a + a^2 + ...) \]

\[ = r_{12} + \frac{t_{12}t_{21}r_{12}e^{2ikL}}{1 - a} \]

Pole in reflectivity for

\[ a = r_{21}^2 e^{2ikL} = 1 \quad k = k(\omega) \]
Finite slab

Thin

Thick

A line of poles gives a jump discontinuity

$$r = r_{12} + \frac{t_{12}t_{21}r_{12}e^{2ikL}}{1 - a}$$

Pole in reflectivity for

$$a = r_{21}^2 e^{2ikL} = 1 \quad k = k(\omega)$$

For thick slab, condition becomes:

$$\text{Im } k = 0 \quad \text{Re } k = \frac{2\pi n}{L}$$
Physical process responsible for lasing

\[ \omega_{\text{upper}}(k) + \omega_{\text{lower}}(-k) = 2\omega_{\text{upper}}(0) \]

Process conserves energy and momentum “Phase matching”

Polariton must be driven strongly enough.

Is this useful?
Conclusions and Outlook (Higgs amplifier)

Possible realization of “non-linear optics” involving collective modes

- Higgs amplification in K3C60
- Phonon lasing in SiC

These effects can be used as a probe of the collective modes themselves

Further experimental checks:

- Idler beam?
- Angular dependence?

Microscopic calculations of Higgs amplification

Do these effects have practical applications? How tunable are they?
Drude and polaronic peaks

\[ \sigma_{\text{eq}}(\omega) = \frac{\Lambda_{s,\text{eq}}}{\varepsilon_0} \frac{1}{\gamma_D - i\omega} + \sum_{n=1}^{3} \frac{B_n \omega}{i(\omega^2 - \Omega_n^2) + \gamma_n \omega} \]
Fit to experiment with adjusted polaronic peak

**Real part**

- Blue curve: Fit to a sum of Drude and polaronic peaks.
- Orange and green curves: Contributions to \( \sigma \).
- Dots – measured conductivity of the normal state.
- Remaining parameters have the following rough parametrization for \( \tilde{\sigma} \) (meV):
  - \( \omega_g = 23 \) meV (gap-like scale for the onset of the polaronic peak).
  - \( \omega_0 = 8 \) meV (parameter that broadens this onset; and \( \omega_0 \) is a gap-like scale for the onset of the polaronic peak).
  - \( \omega_{\text{e.f.f.}} = 1 \) (effective optical phonon).
  - \( \omega_{\text{P}} \) is a parameter that controls the deviation of \( \tilde{\sigma} \).

**Imaginary part**

- 2000 ps
- 1000 ps
- 100 ps

The resulting complex conductivity and the reflectivity are plotted in Fig. 9 for each pump pulse width: 100 ps, 2000 ps, and 1000 ps.
Higgs Oscillations from THz pump

Fig. 1 (color online). (a) A schematic picture of the phase and amplitude modes represented by the arrows in azimuthal and radial directions, respectively, on the effective potential in the plane of complex order parameter indicated by the vertical line in (e) for sample A.

(b) WGP: a wire grid polarizer. WGP: a wire grid polarizer.

(c) Temperature dependence of the real-part optical conductivity spectra in sample C without the pump. The solid curves are calculated by the Mattis-Bardeen model. (d) Temperature dependence of the probe THz electric field (arb. units) for various pump intensities: 5 ps, 10 ps, 20 ps, and 30 ps.

(d) Temperature dependence of the BCS gap energy at the fixed delay time of gate delay time defined by FWHM of the pump THz pulse.

Note that, this correspondence between the gap energy and the delay time of the probe to the pump THz pulse, we recorded the probe THz field, which is in excellent agreement with the theoretical predictions.

(e) The waveform of the probe THz field was detected by the tilted-optic (EO) sampling of the transmitted probe THz pulse. The intense pump THz pulse was generated by the tilted-optic (EO) sampling of the transmitted probe THz pulse.

(f) The BCS gap energies at 4 K, 8 K, and 14 K, and film thickness 0.5 mm are shown in Fig. 1(c). The BCS gap energies at 4 K, 8 K, and 14 K, and film thickness 0.5 mm are shown in Fig. 1(c).
### Phenomenological model for conductivity

\[
\tilde{\sigma}_1(\omega) = \frac{A}{(1 + e^{(E_g - \omega)/\delta})^2}
\]

\[
\tilde{\sigma}_2(\omega) = \frac{\Lambda_s}{\epsilon_0 \omega} \frac{1 + (\omega/E_e)^4}{1 + (\omega/E_s)^2}
\]

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<th>(E_g)</th>
<th>(\delta)</th>
<th>(\Lambda_s)</th>
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<td>12.5</td>
<td>27</td>
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</tr>
</tbody>
</table>
Higgs oscillations

\[ E_{e-ph} = g \sum_q (a_q + a_q^\dagger) c_{k+q,\sigma}^\dagger c_{k\sigma} \]

FIG. 9. Dynamics of the order parameter \( \Delta^S(t) \) after the quantum quench from \( g_{\text{start}} \) equilibrium state to \( g = 4J \) at \( t = 0 \). The dashed line denotes the static point \( g_{\text{start}} = g \).
Higgs oscillations

FIG. 10. The Fourier spectra of post quench dynamics in reference to the frequency $\omega_{\text{dyn}}$, for different starting states $g_{\text{start}}$ along the horizontal axis and different quenched interactions (a) $g = 3J$, (b) $g = 4J$ and (c) $g = 5J$. The black arrows denote the corresponding static point, while the blue crosses mark the asymptotic frequency at the static limit.

FIG. 11. The comparison of asymptotical dynamics frequencies (blue dots) extracted in the Fourier spectra of Fig.10, to the corresponding static point, while the black arrows denote $\omega_{\text{dyn}}$. The red line represents $2\Delta^5$.
Higgs oscillations

\[ H_{\text{drive}} = I(t) \sum_{q} x_q x_{-q} \]

\[ x_q = \frac{1}{\sqrt{2}} (a_q + a^\dagger_{-q}) \]

FIG. 12. Pumped induced dynamics of the order parameter \( \Delta^S \) for various pump frequencies \( \Omega \)s with (a) \( A_0 = 0.2 \) and (b) \( A_0 = 0.05 \). The black curve sketches the pump pulse, while the dashed line denotes the Higgs frequency \( \omega_H = 2\Delta^S \) at this system.

FIG. 13. Fourier spectra of dynamics induced by pump strength \( A_0 = 0.01 \sim 0.15 \). The left and right panels show two pump frequencies \( \Omega = 2J \) and \( \Omega = 2.5J \). The double-end arrow denotes twice of the order parameter.
What do these two have in common?

Superconductivity          Weak force

Answer: The Anderson-Higgs mechanism
Reasons for bosons to be massless

1. Spontaneous symmetry breaking \( \Rightarrow \) Goldstone’s theorem

2. Gauge invariance

In superconductor, both phenomena exist, yet all excitations are gapped. Why?

Anderson: Goldstone boson and gauge field mix. Both become massive, yielding plasmon and Meissner effect.
Anderson-Higgs mechanism vs Higgs boson

Anderson-Higgs mechanism:
If symmetry is gauged, Goldstone mode is eaten up to give massive vector field

Higgs boson:
- Fluctuations in order parameter amplitude.
- Massive, whether symmetry is gauged or not.
- Not charged.
Anderson-Higgs mechanism vs Higgs boson

Gauged U(1) theory:

\[ \mathcal{L} = \frac{1}{2} |(\partial_\mu + ieA_\mu) \psi|^2 - m^2 (\psi^* \psi - 1)^2 - \frac{1}{4} F_{\mu\nu} F^{\mu\nu} \]

Amplitude and phase:

\[ \psi = (1 + \eta) e^{i\xi} \]

Gauge invariance:

\[ \psi \rightarrow e^{-i\xi} \psi, \quad \tilde{A}_\mu \equiv A_\mu - e^{-1} \partial_\mu \xi \]

Then:

\[ \mathcal{L} = \frac{1}{2} (\partial_\mu \eta)^2 + m^2 (\eta^2 + 2\eta^3 + 2\eta^4) + e^2 (1 + \eta)^2 \tilde{A}_\mu \tilde{A}^\mu - \frac{1}{4} \tilde{F}_{\mu\nu} \tilde{F}^{\mu\nu} \]

★ There is no \( \xi \) particle!
★ Photon becomes massive triplet \( \tilde{A}_\mu \)
★ The Higgs boson (\( \eta \)) has mass, \( m_H = \sqrt{2}m \), independent of \( e \)