

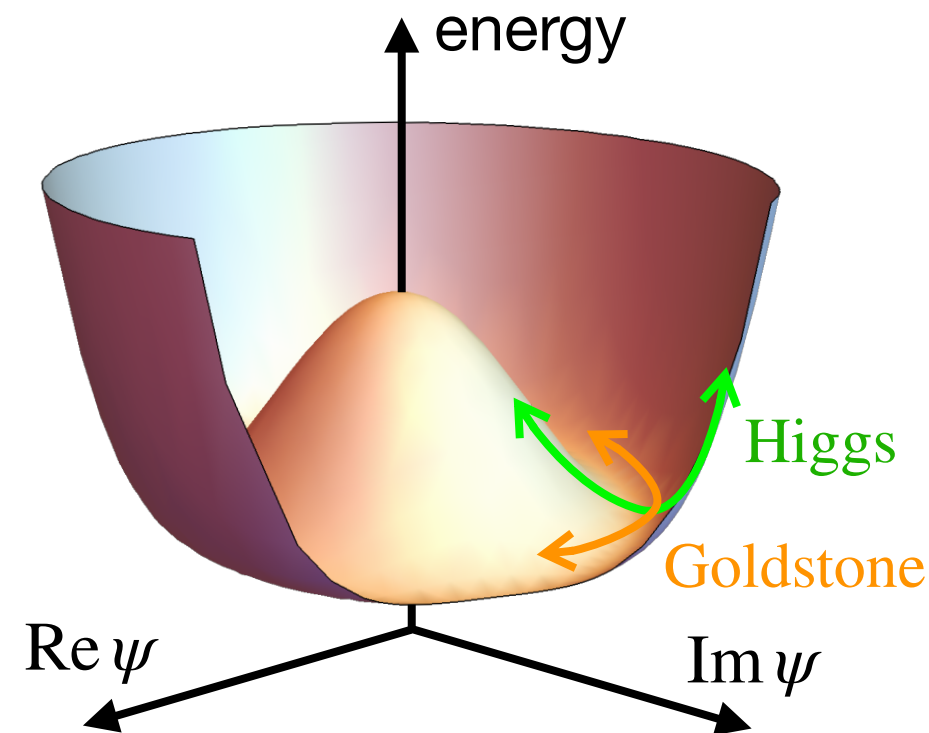
Higgs Amplification in Non-Equilibrium K_3C_{60}

Daniel Podolsky
Nonequilibrium Superconductivity Workshop
January 2020

With:

Eugene Demler, Misha Lukin, Bert Halperin, Yao Wang, Sho Sugiura (Harvard)
Andrea Cavalleri, Michelle Buzzi, Gregor Jotsu (Max Planck, Hamburg)
Ignacio Cirac, Tao Shi (Max Planck, Garching)

Background: Collective modes of a superconductor



Anderson-Higgs mechanism:

- Goldstone mode "eaten" by gauge field, gapped to plasma frequency
- Leads to Meissner effect

Higgs mode:

- Fluctuations in order parameter amplitude.
- Gapped excitation.
- Not charged.

Motivation: Non-Equilibrium Superconductivity

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

PRL 111, 057002 (2013)

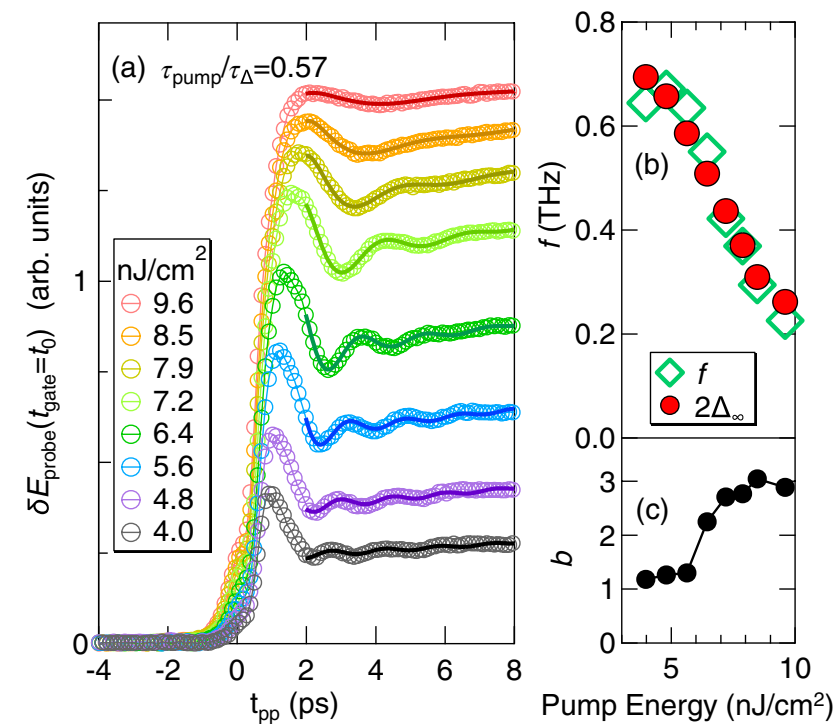
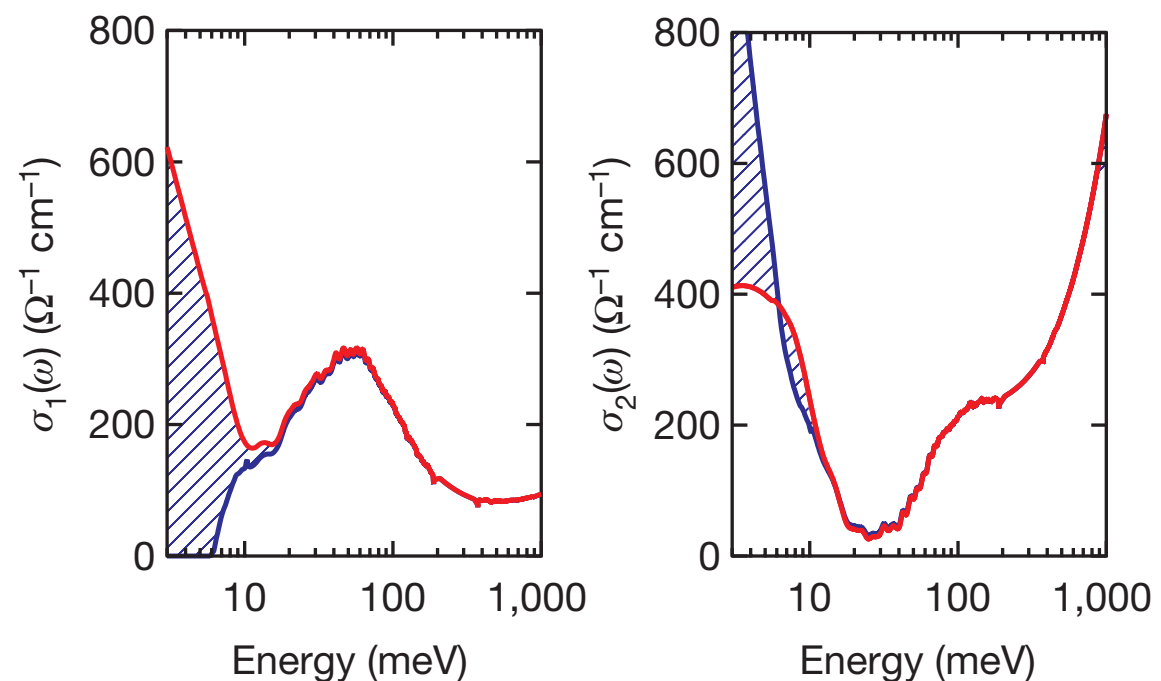


Photo-induced superconductivity in K_3C_{60}

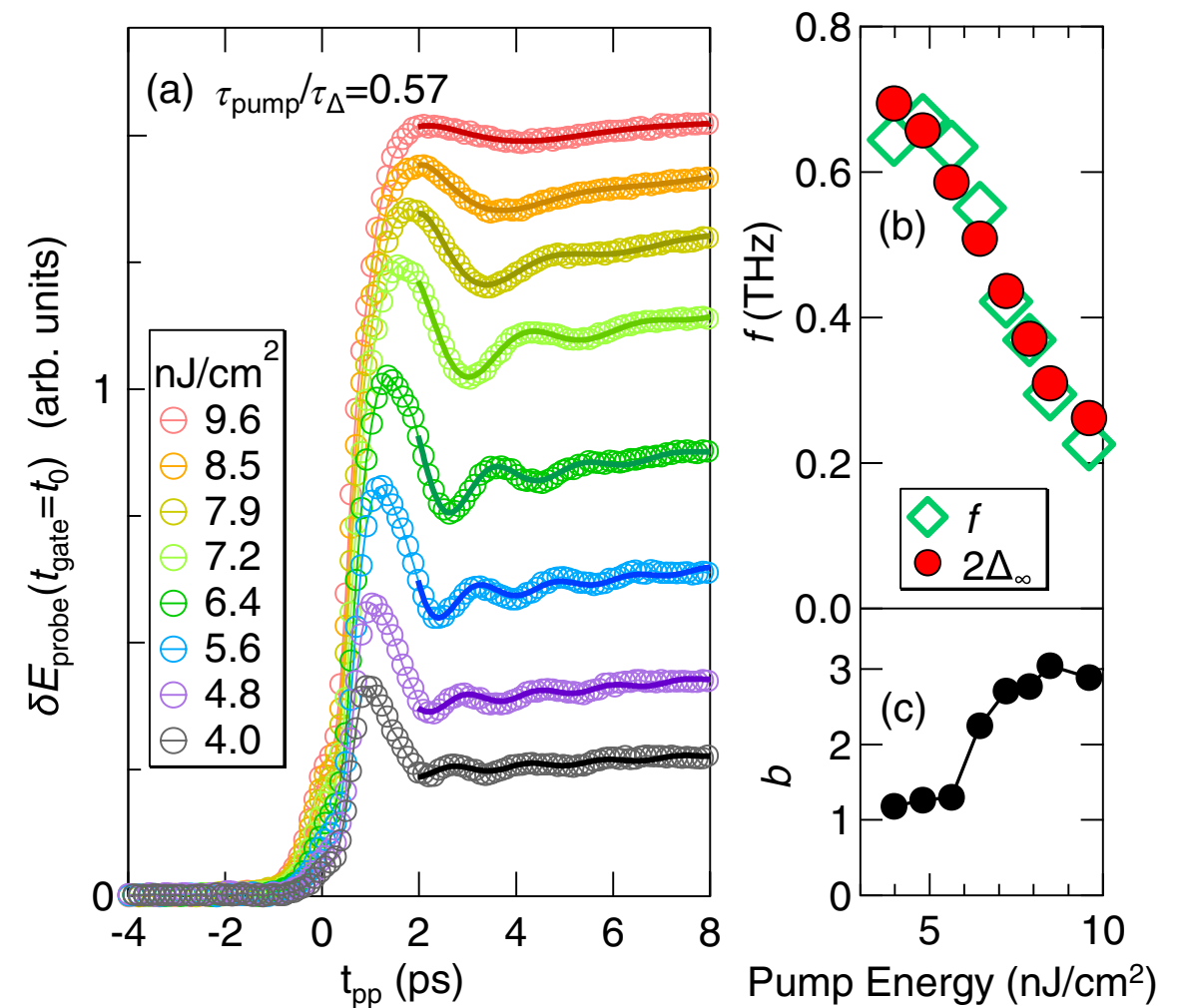
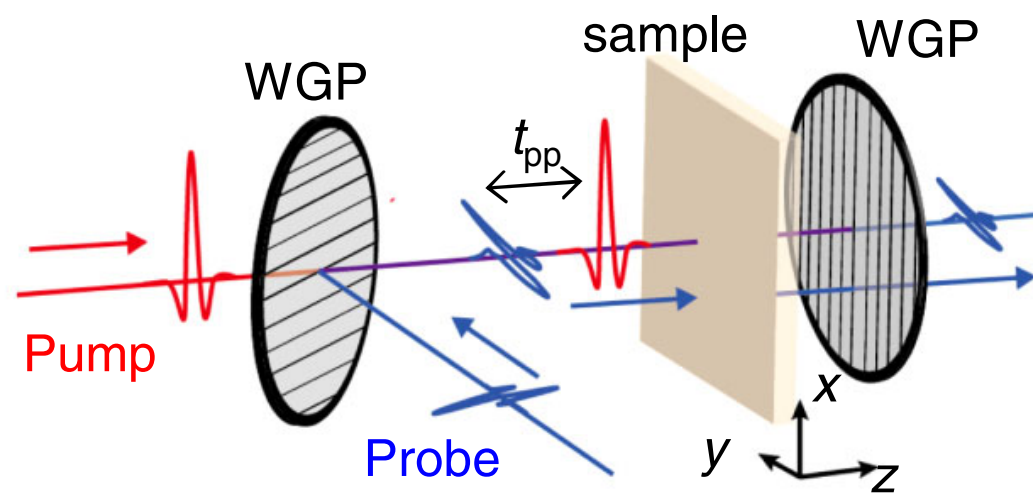
Nature 530, 461 (2016)



Background: Higgs Oscillations in $\text{Nb}_{1-x}\text{Ti}_x\text{N}$

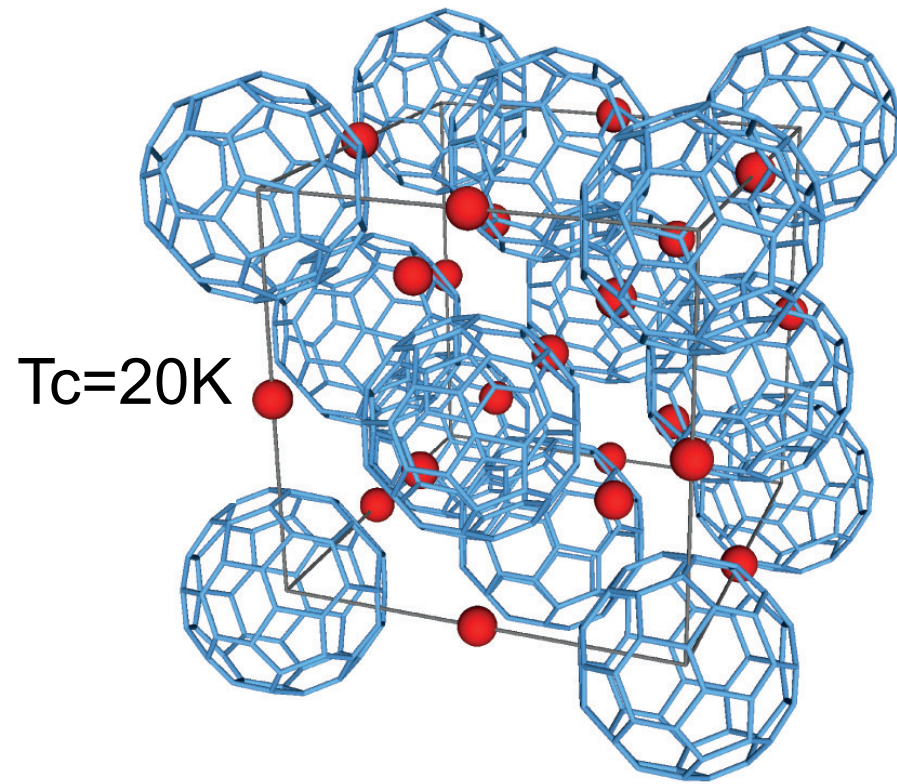
Higgs mode is not charged $\rightarrow$ doesn't couple linearly to light

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

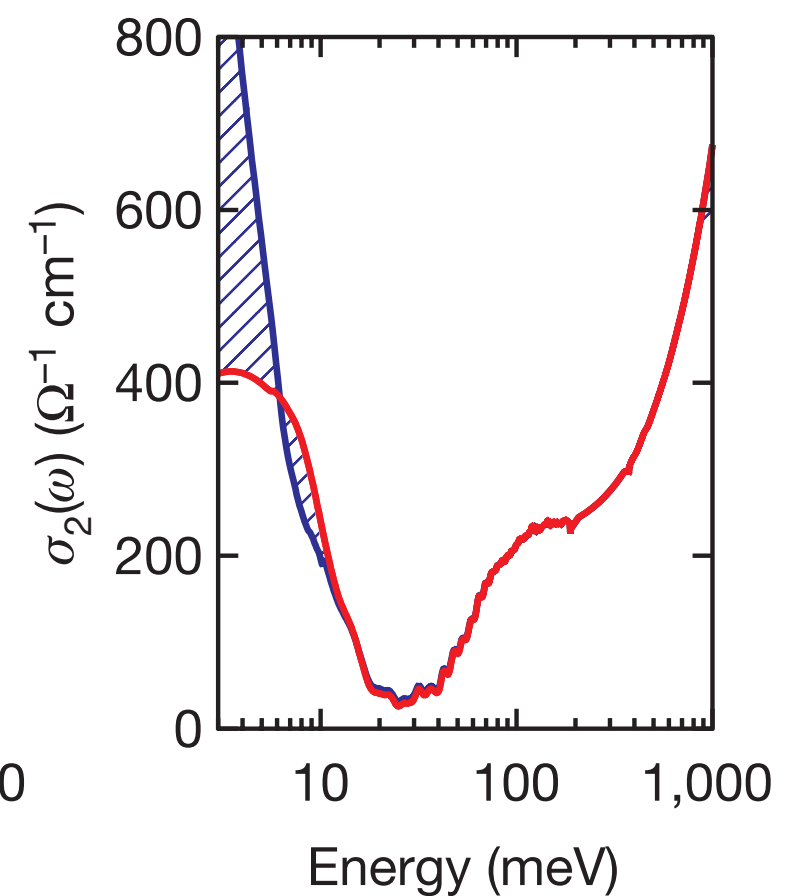
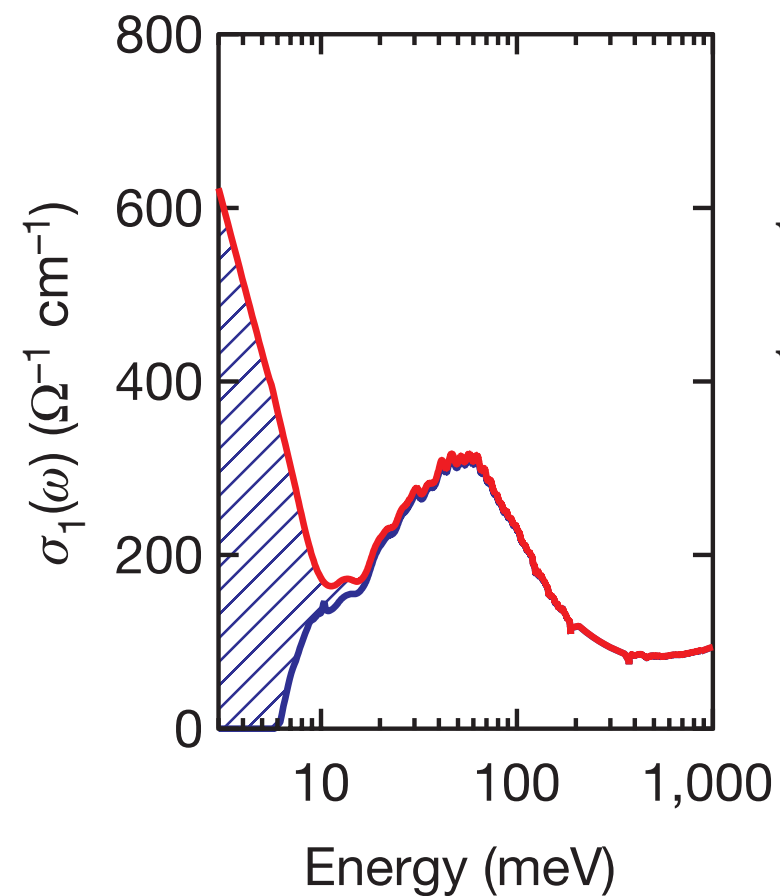


Background: Photoinduced superconductivity in K_3C_{60}

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



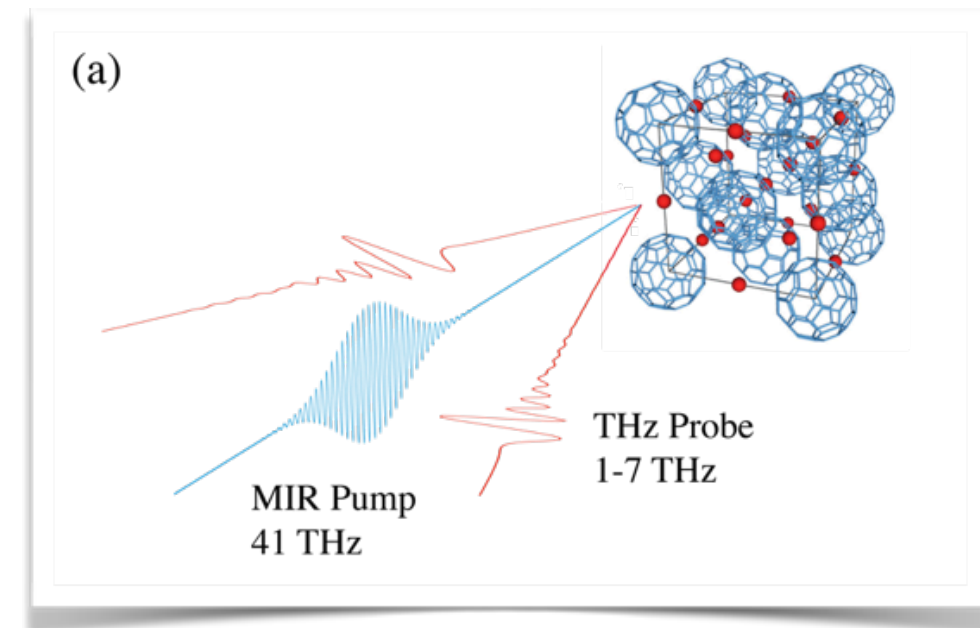
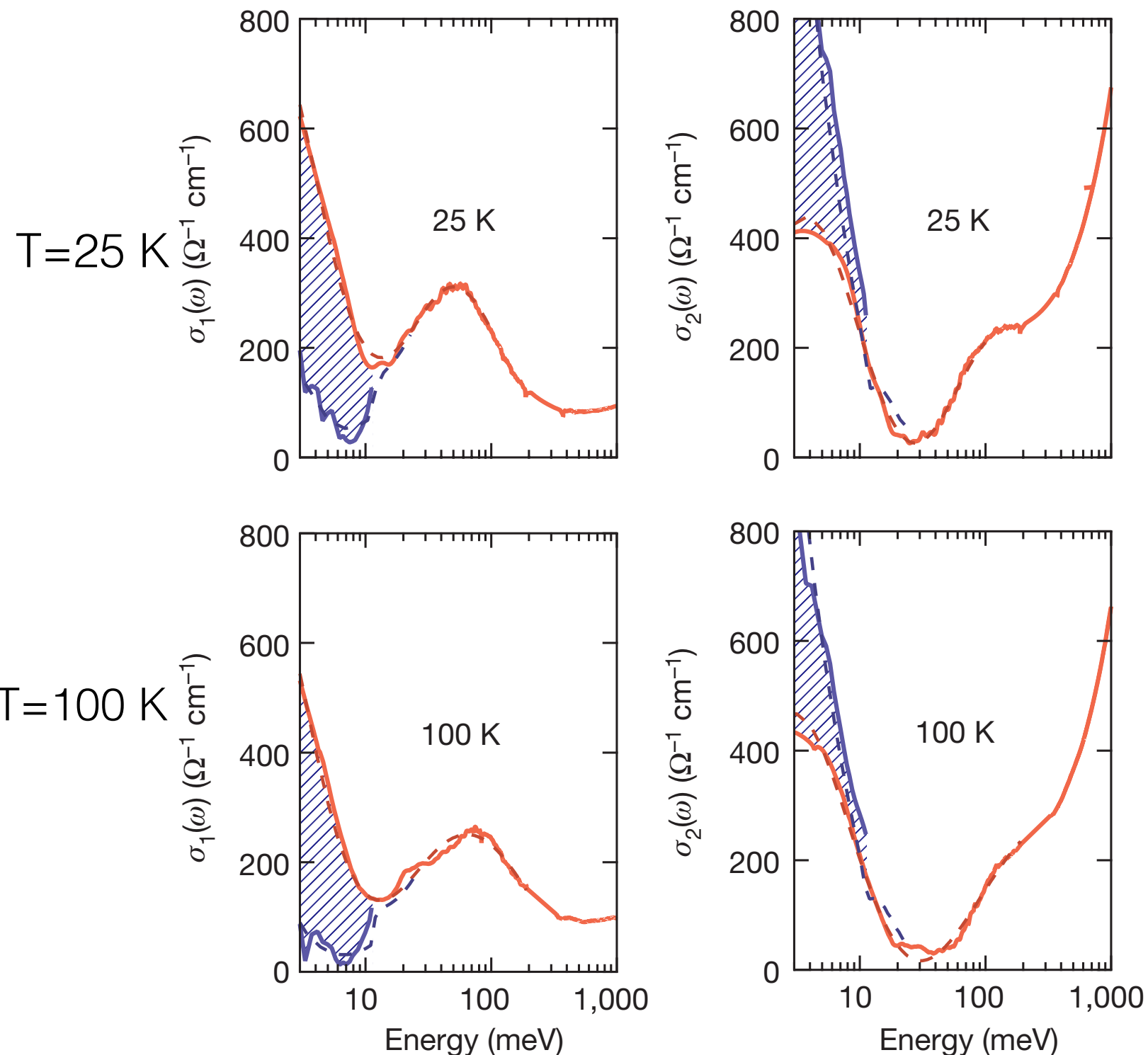
Equilibrium: $T=25$ K and $T=10$ K



Background: photoinduced superconductivity in K_3C_{60}

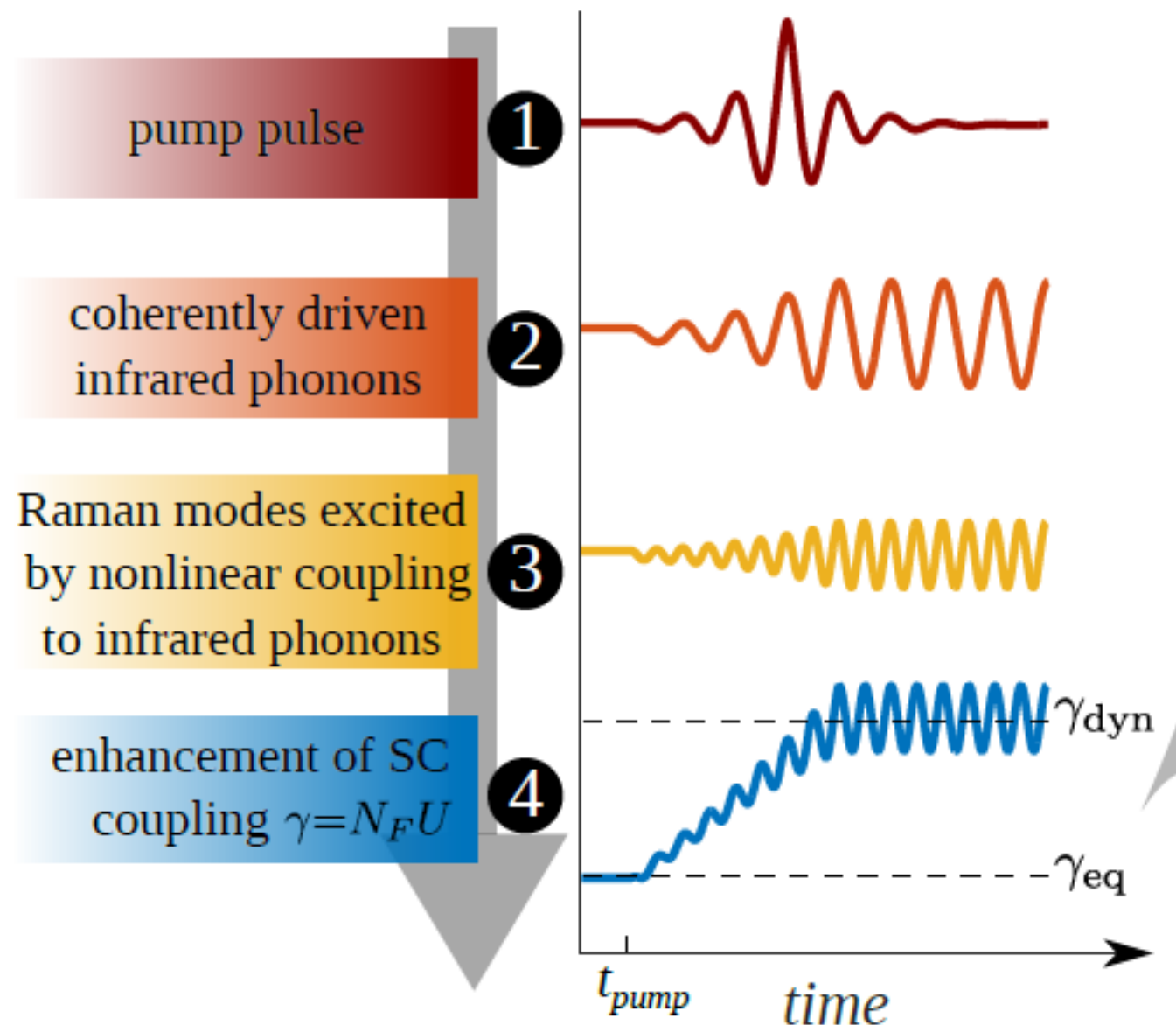
Response — **Equilibrium** vs **Photoexcited**:

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



Photoinduced superconductivity — Possible mechanism

Parametric enhancement of electron-phonon interaction



M. Babadi, M. Knap, G. Refael, I. Martin, E. Demler, Phys. Rev. B 94, 214504 (2016)

M. Babadi, M. Knap, G. Refael, I. Martin, E. Demler, Phys. Rev. B 96, 014512 (2017)

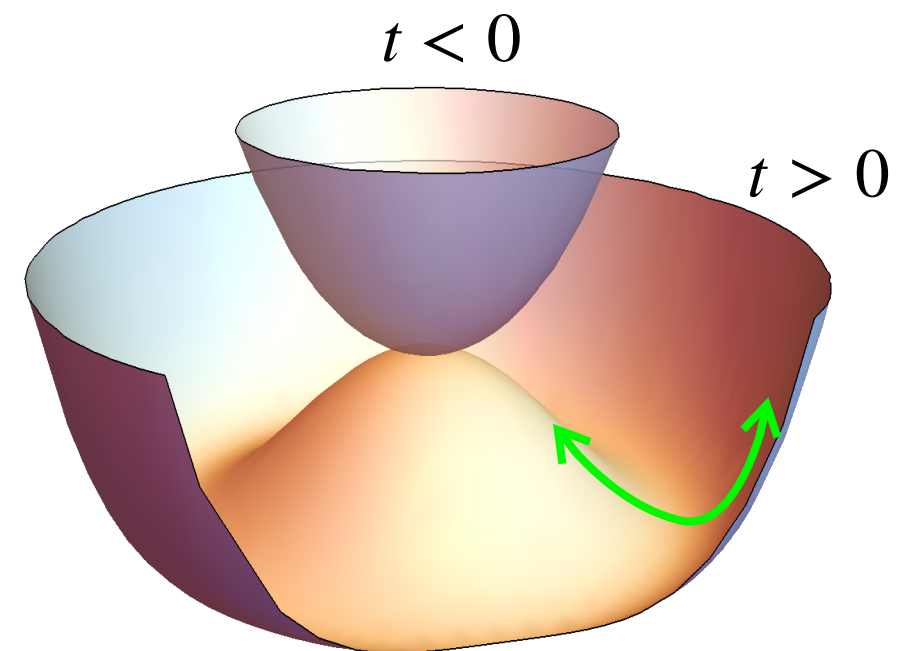
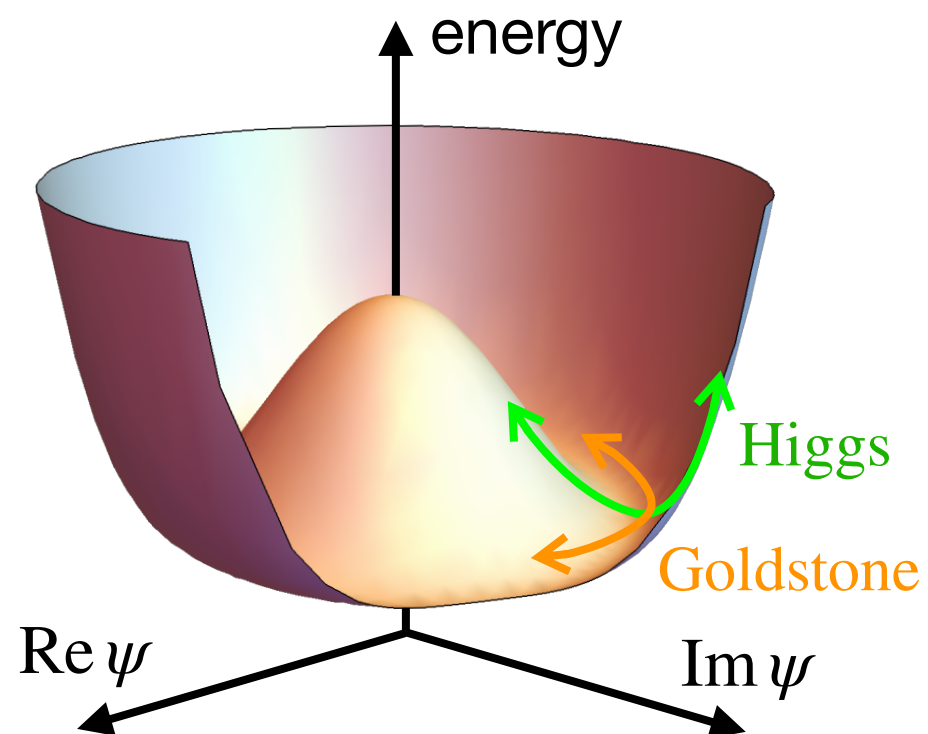
Part I: Higgs Amplification in K_3C_{60}

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:

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

Upon quantization, this yields:

$$\mathcal{H}_{\text{Higgs/photons}} \sim \sum_{\omega_1 + \omega_2 = \omega_H} (\delta\rho a_{\omega_1}^\dagger a_{\omega_2}^\dagger + \text{h.c.}) \qquad \mathbf{A} \sim \sum_{\omega} (a_{\omega} + a_{\omega}^\dagger)$$

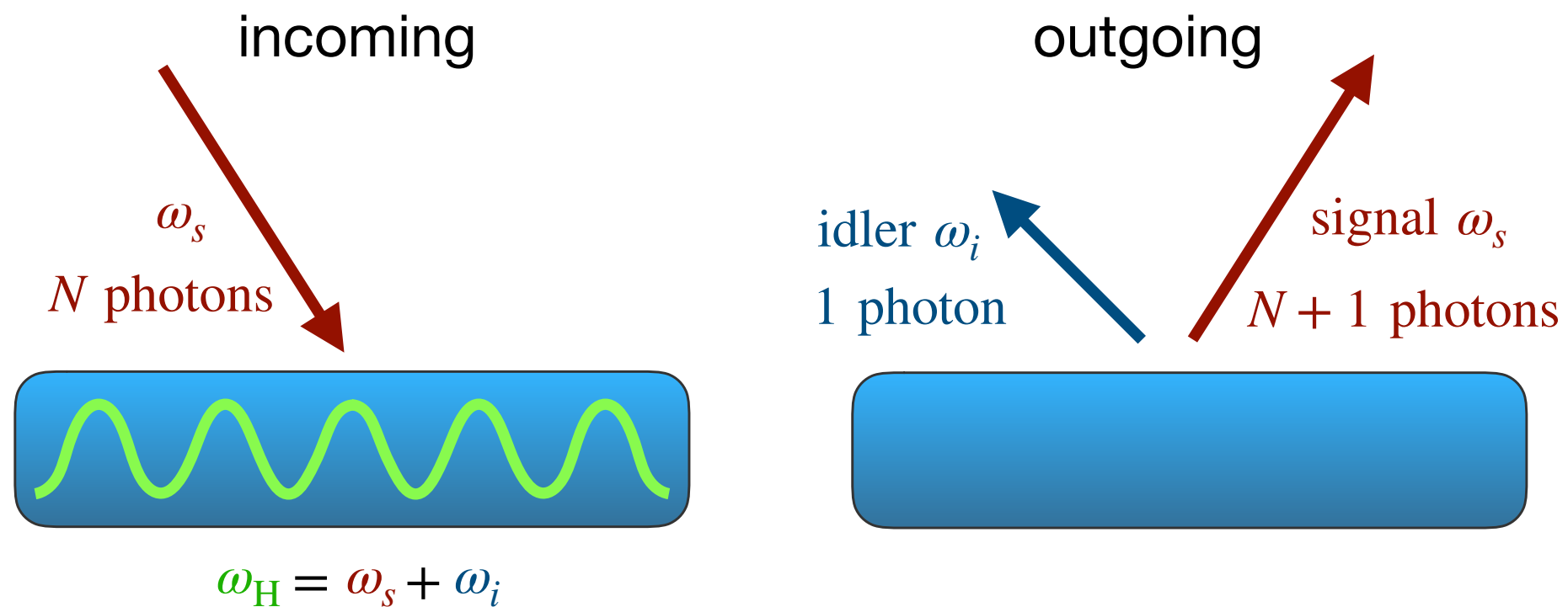
Photons in bulk are gapped to plasma frequency (Anderson-Higgs mechanism).

Near the surface they remain gapless

→ Coupling is important for reflection problem

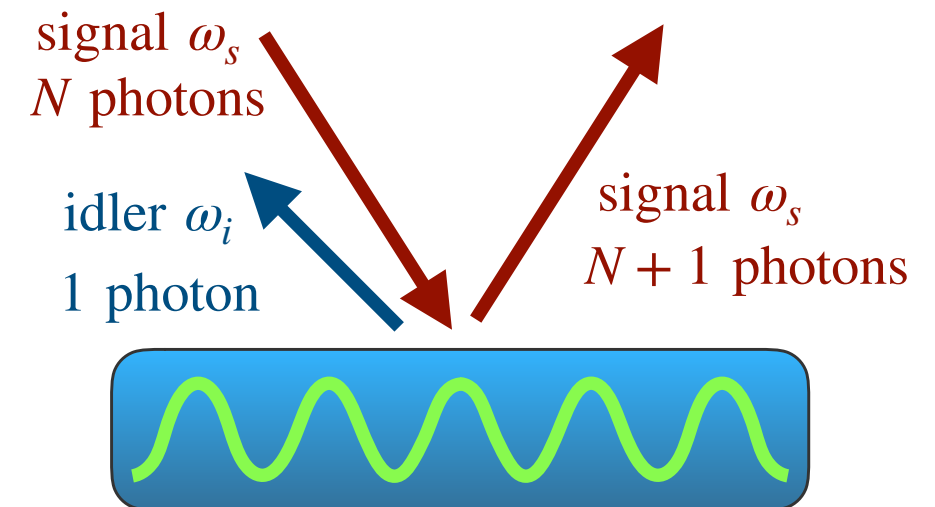
Photon pair creation

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

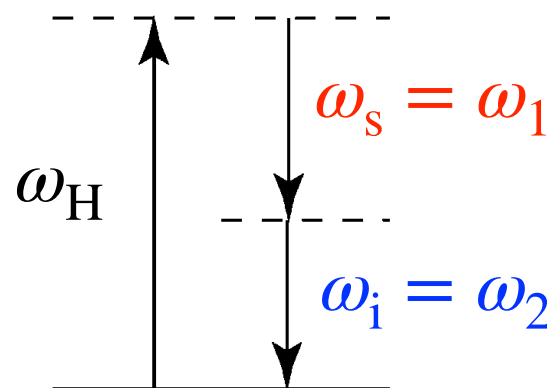


“Nonlinear optics” involving Higgs mode

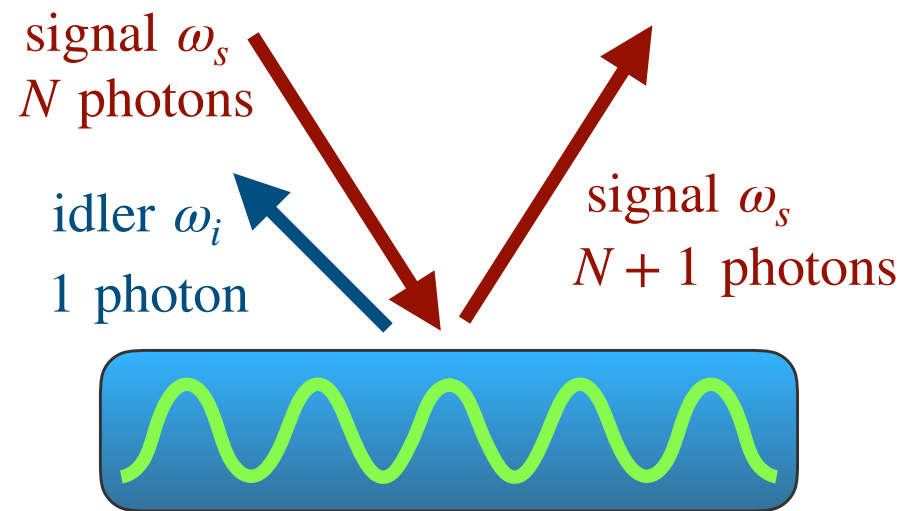
$$\mathcal{H}_{\text{Higgs/photons}} \sim \sum_{\omega_1 + \omega_2 = \omega_H} (\delta\rho a_{\omega_1}^\dagger a_{\omega_2}^\dagger + \text{h.c.})$$



Three-wave mixing (one Higgs mode and two photons) and parametric resonance. Frequency ω_1 is selected by the probe beam due to Bose enhancement factor. In the language of parametric resonance, ω_1 is a **signal wave** and ω_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 Λ

London equation:

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

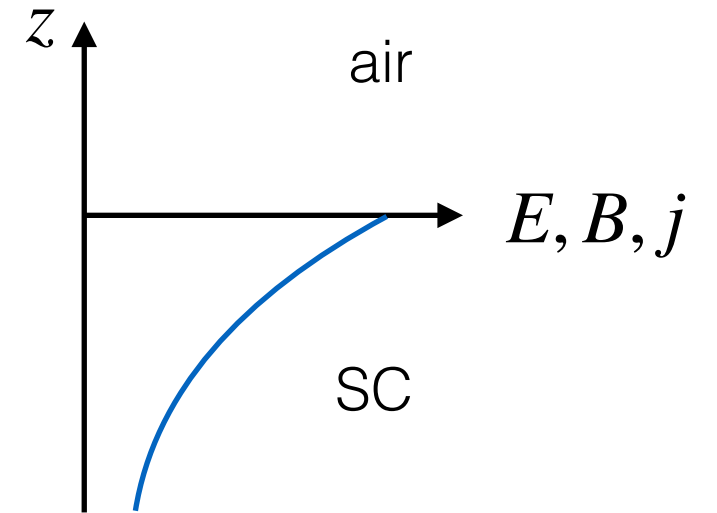
$$\Lambda(t) = \Lambda_s + 2\Lambda_m \cos(\omega_H t)$$

Solve the Fresnel-Floquet reflection problem

Floquet modes inside medium

Evanescent waves inside medium: $\mathbf{D} \in \{\mathbf{E}, \mathbf{B}, \mathbf{j}\}$

$$\mathbf{D} = \left(\mathbf{D}_1 e^{-i\omega_1 t} + \mathbf{D}_2^* e^{i\omega_2 t} \right) e^{\kappa z} + \text{c.c.}$$



London equation mixes frequencies

$$\omega_1 + \omega_2 = \omega_H$$

$$\begin{cases} \mathbf{j}_1 = \Lambda_s \frac{\mathbf{E}_1}{-i\omega_1} + \Lambda_m \frac{\mathbf{E}_2^*}{i\omega_2} \\ \mathbf{j}_2 = \Lambda_s \frac{\mathbf{E}_2^*}{i\omega_2} + \Lambda_m \frac{\mathbf{E}_1}{-i\omega_1} \end{cases}$$

Matrix equation for $\mathbf{E}$:

$$\begin{pmatrix} \kappa^2 c^2 + \omega_1^2 \epsilon_1 & \Lambda_m \omega_1 / \omega_2 \\ \Lambda_m \omega_2 / \omega_1 & \kappa^2 c^2 + \omega_2^2 \epsilon_{\bar{2}} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2^* \end{pmatrix} = 0 \quad \begin{cases} \epsilon_1 \equiv \epsilon(\omega_1) \\ \epsilon_{\bar{2}} \equiv \epsilon(-\omega_2) \end{cases}$$

Solution:
$$\kappa_{\pm}^2 c^2 = -\frac{\omega_1^2 \epsilon_1 + \omega_2^2 \epsilon_{\bar{2}}}{2} \pm \frac{1}{2} \sqrt{(\omega_1^2 \epsilon_1 - \omega_2^2 \epsilon_{\bar{2}})^2 + 4\Lambda_m^2}$$

Fresnel-Floquet reflection problem

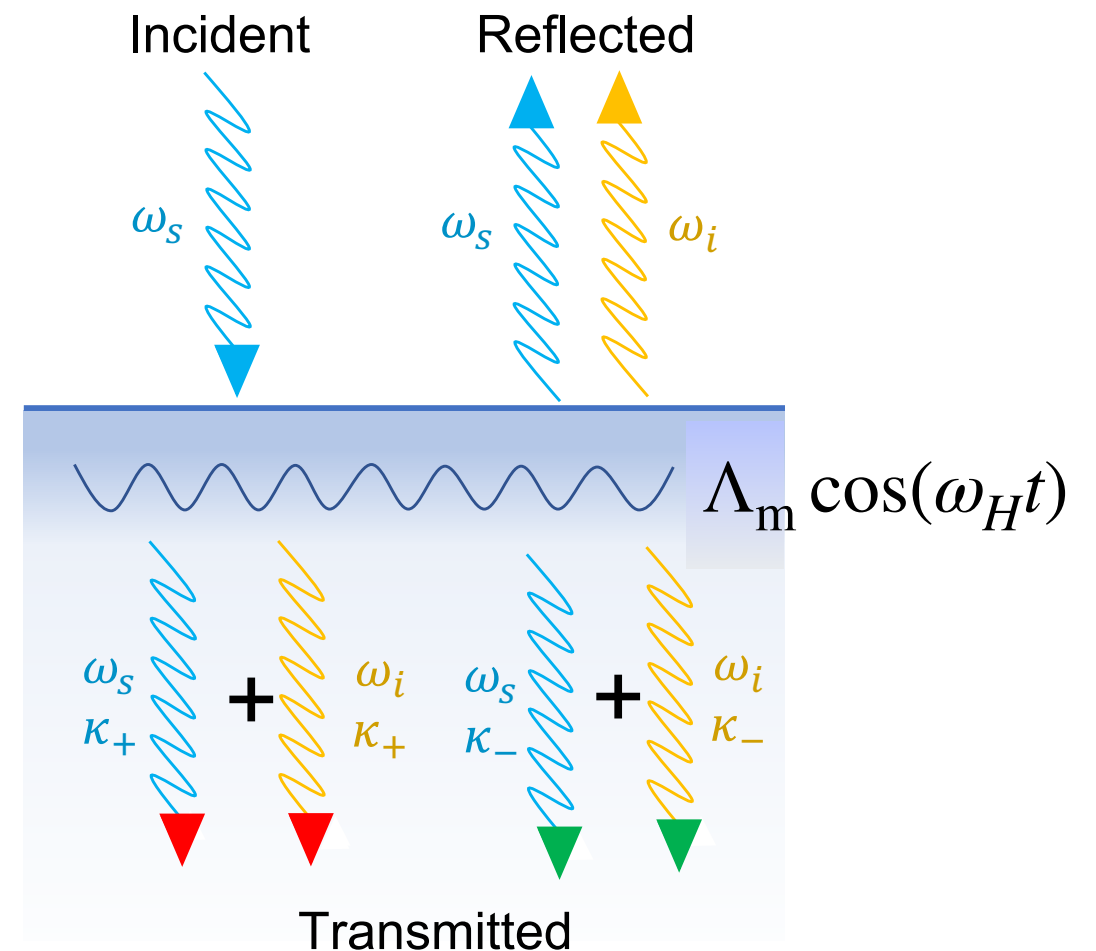
Boundary conditions imposed independently on each frequency:

$$B_1^i + B_1^r = B_{1,+}^t + B_{1,-}^t$$

$$E_1^i + E_1^r = E_{1,+}^t + E_{1,-}^t$$

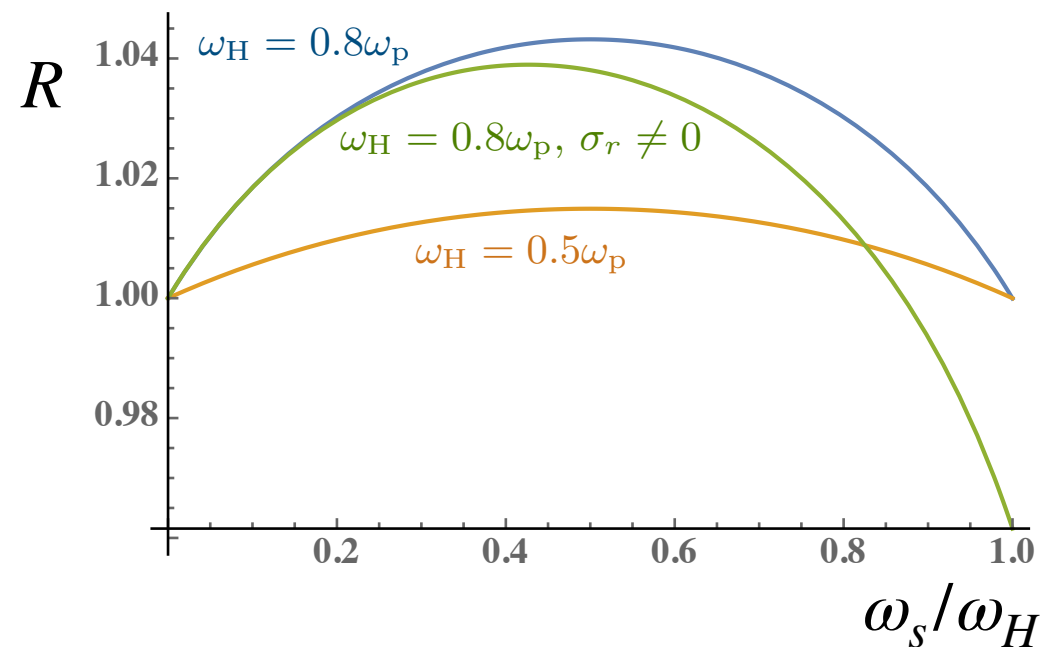
$$B_2^{r*} = B_{2,+}^{t*} + B_{2,-}^{t*}$$

$$E_2^{r*} = E_{2,+}^{t*} + E_{2,-}^{t*}$$

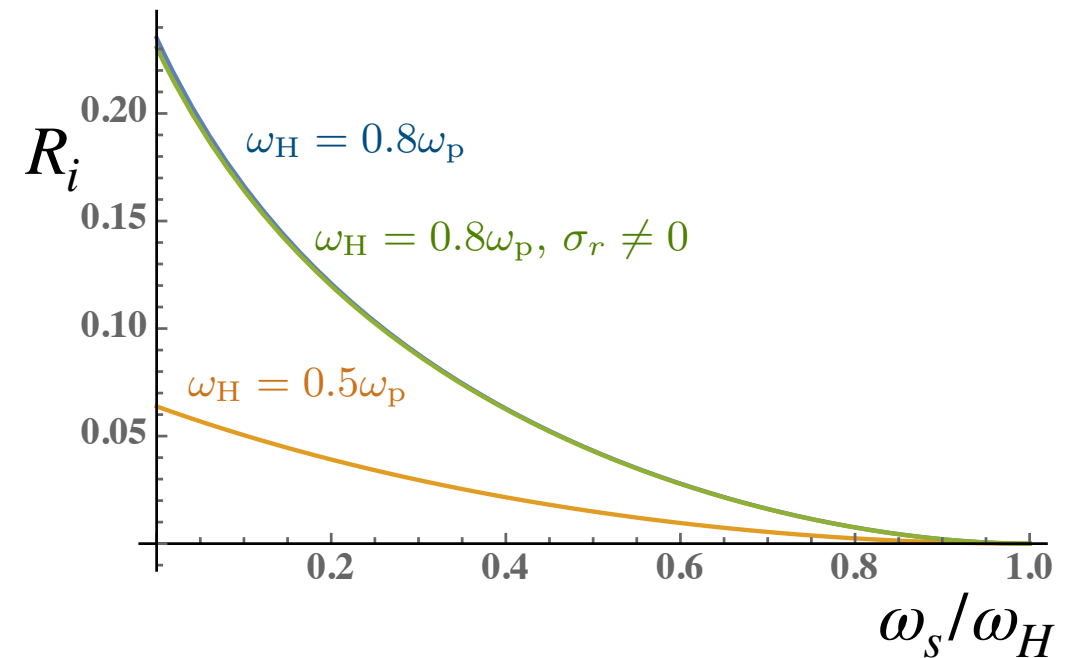


Results — Normal incidence

Reflectivity of signal



Intensity of idler mode

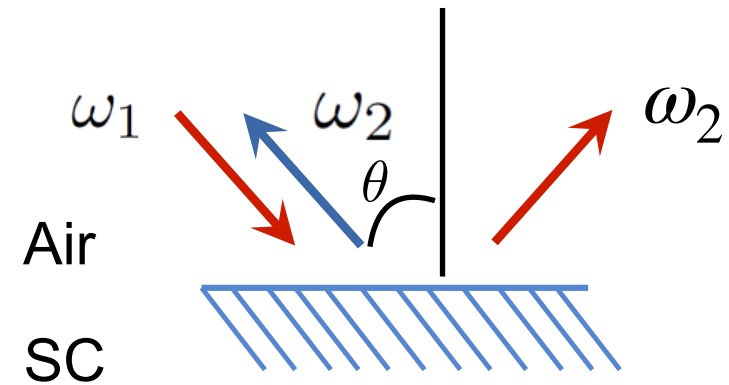


Effect is weak because light doesn't penetrate deeply into the sample:

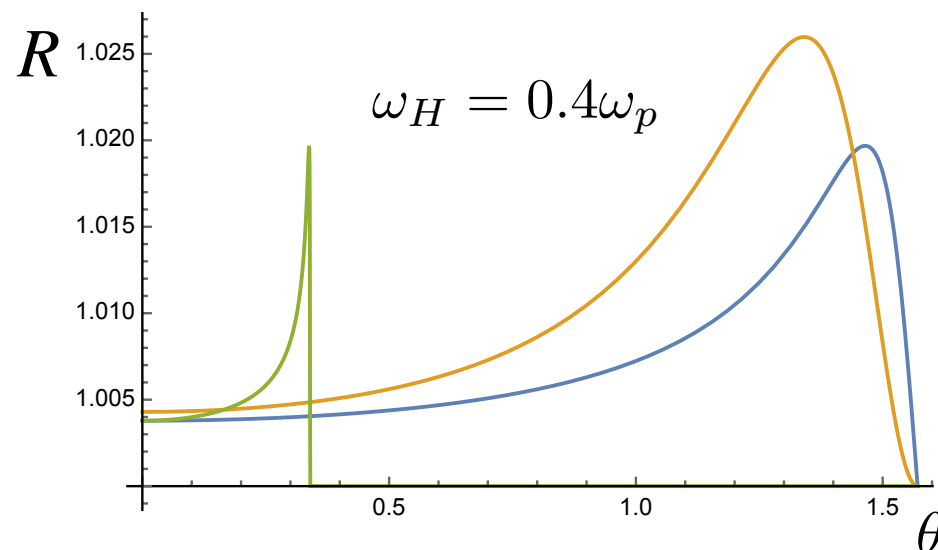
$$R_{\max} = 1 + \frac{1}{4} \frac{\Lambda_m^2}{\Lambda_s^2} \frac{\epsilon_{\text{out}} \omega_H^2}{\epsilon_{\text{in}} \omega_p^2}$$

Two ways to enhance: shallow incidence or small plasma frequency

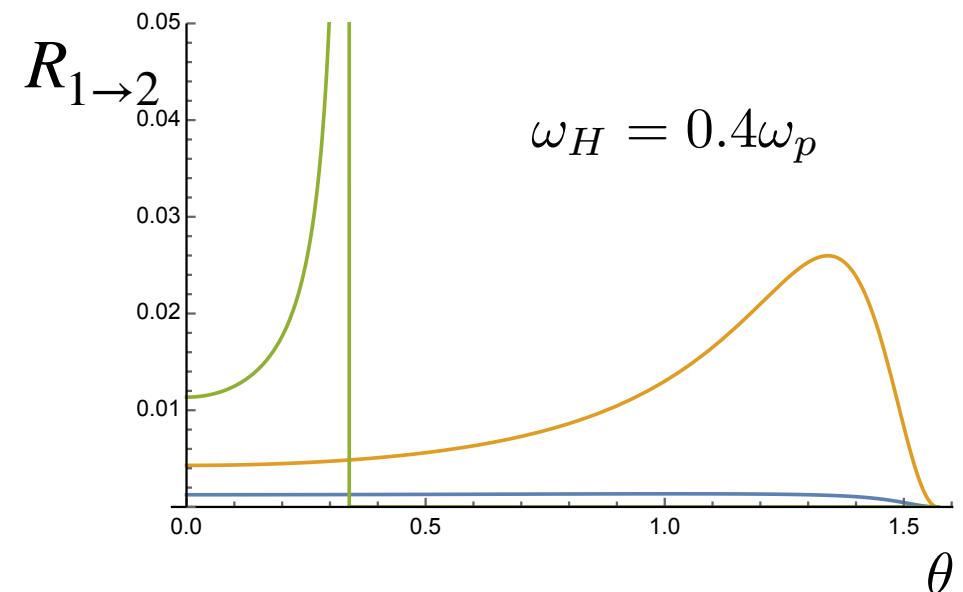
Shallow incidence



Reflectivity of signal



Intensity of idler mode



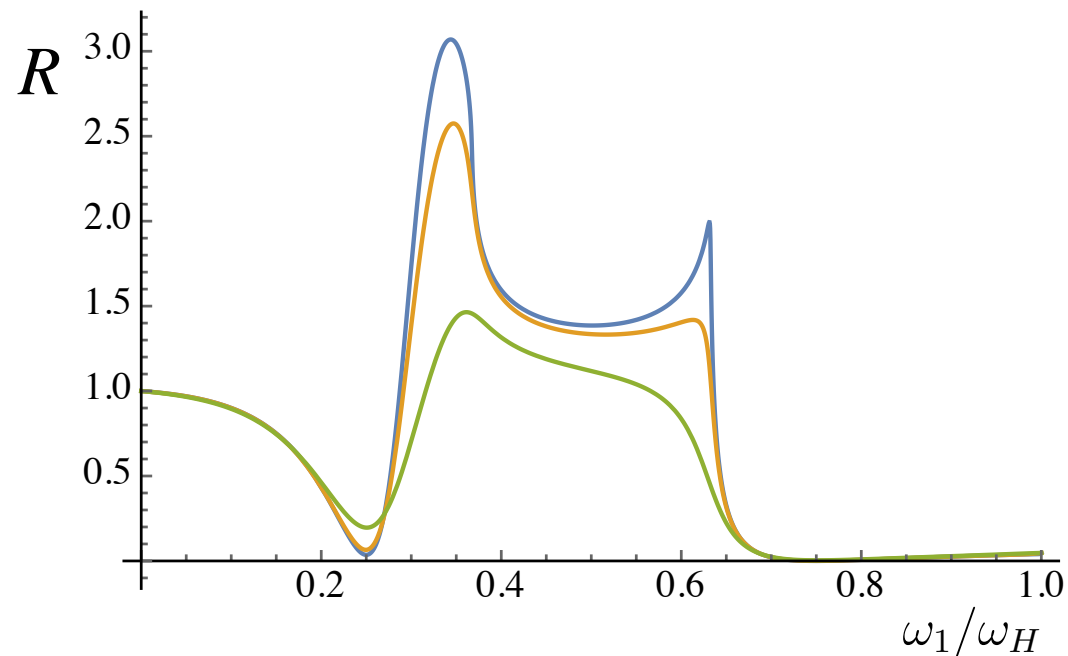
Blue $\omega_1 = 0.1 \omega_p$, orange $\omega_1 = 0.2 \omega_p$, green $\omega_1 = 0.3 \omega_p$.

Dependence on polarization: p-polarization gives stronger effect

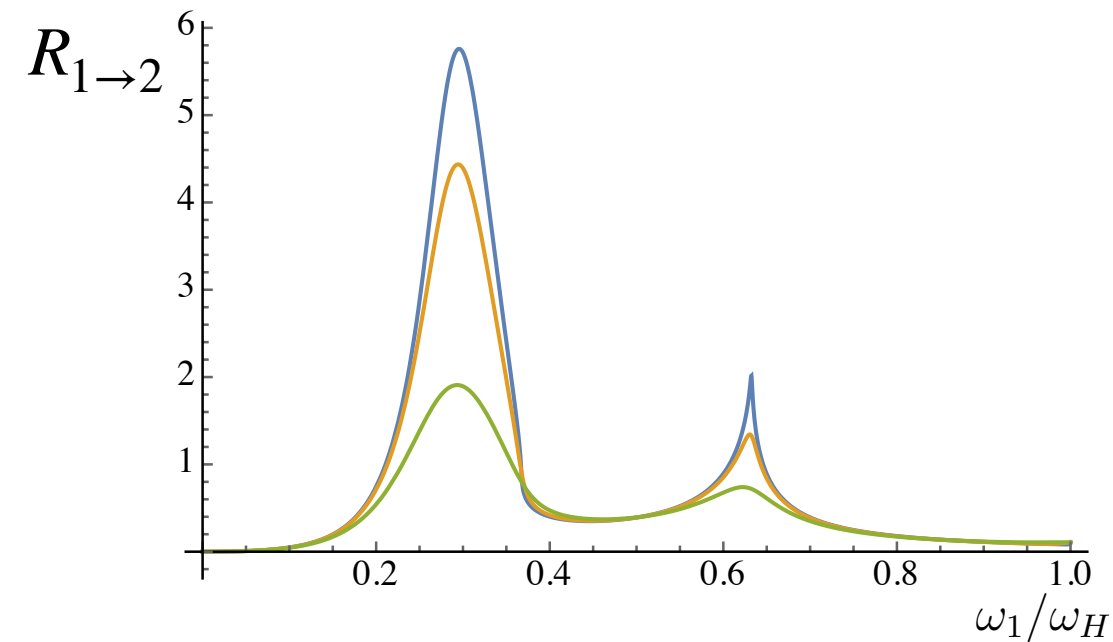
There is an analogue of total internal reflection: for very shallow angle, idler is not allowed by conservation laws. Large signal right before the transition.

Small plasma frequency

Reflectivity of signal



Intensity of idler mode



Normal incidence, $\omega_H = 1.3 \omega_p$

Light penetrates into bulk of SC

→ 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

→ Small plasma frequency $\omega_p = 72\text{meV}$

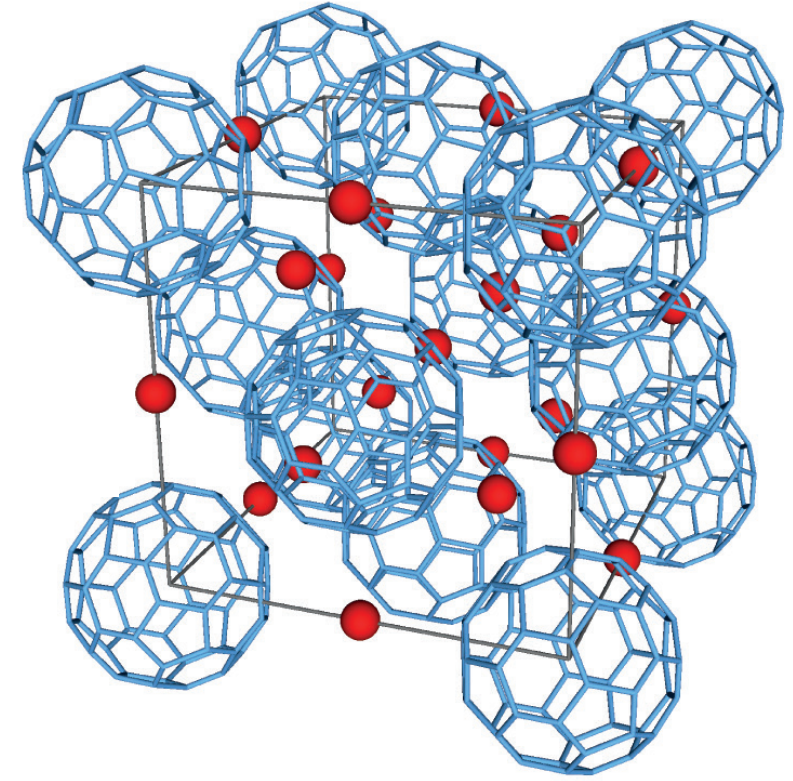


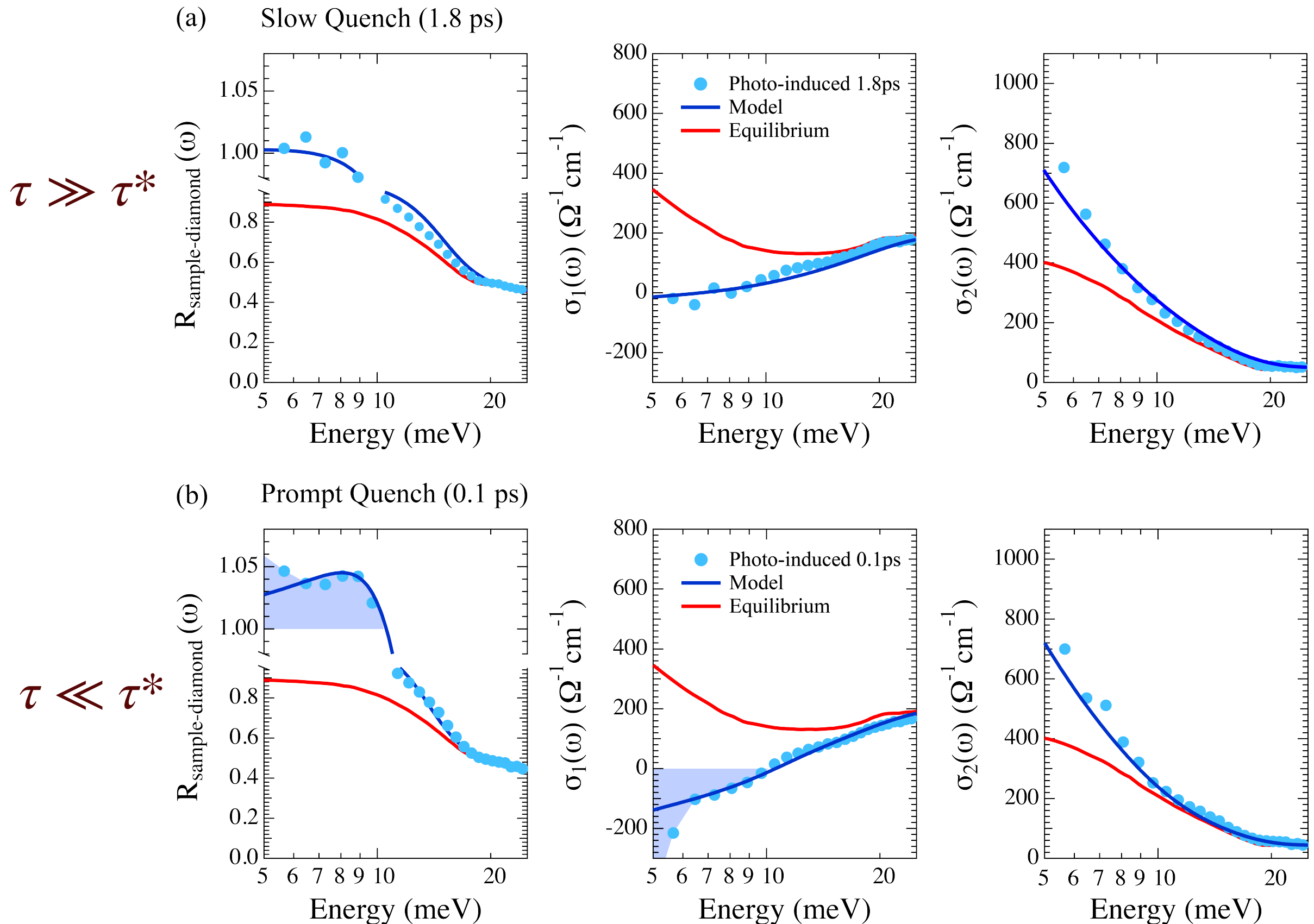
Photo-induced superconductivity has onset at $\sim 120\text{ K}$

→ Estimate for Higgs frequency $\omega_H \approx 2\Delta = 30\text{meV}$

In order to induce Higgs oscillations, need quench faster than

$$\tau^* = \frac{\hbar}{\omega_H} \approx 300\text{ fs}$$

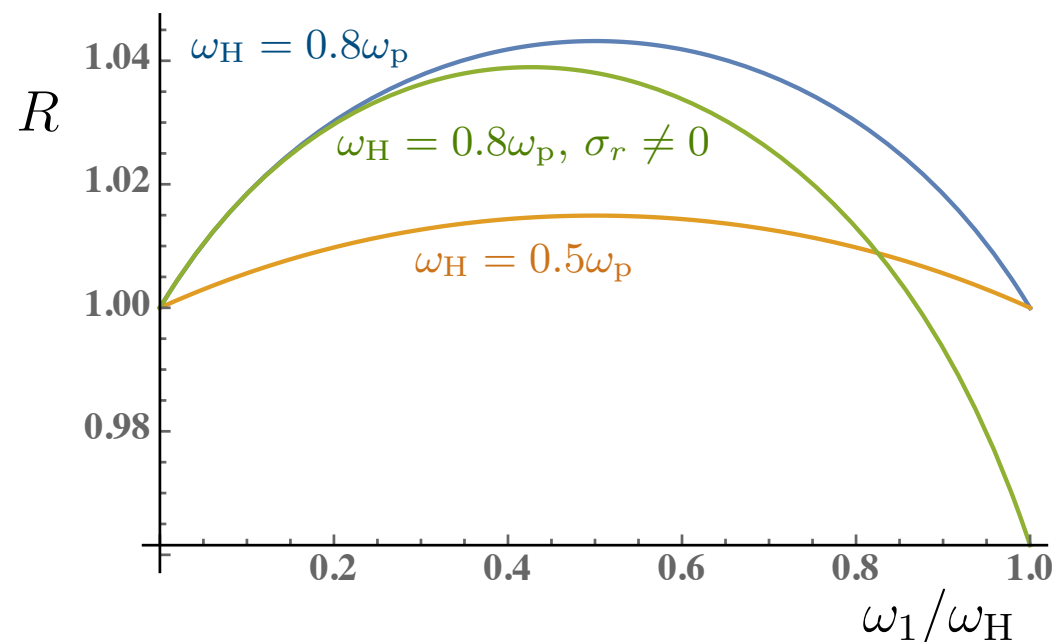
Experiment — Pulse width dependence



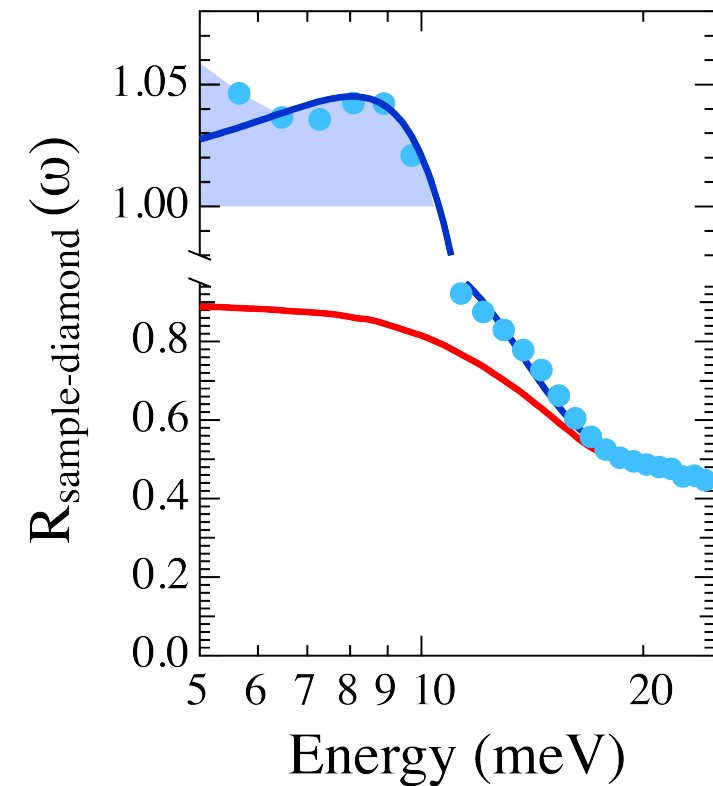
Comparing model to experiment

Maximum amplification (6%) at 10 meV

This suggests that Higgs frequency is somewhat higher than 20 meV



(b) Prompt Quench (0.1 ps)

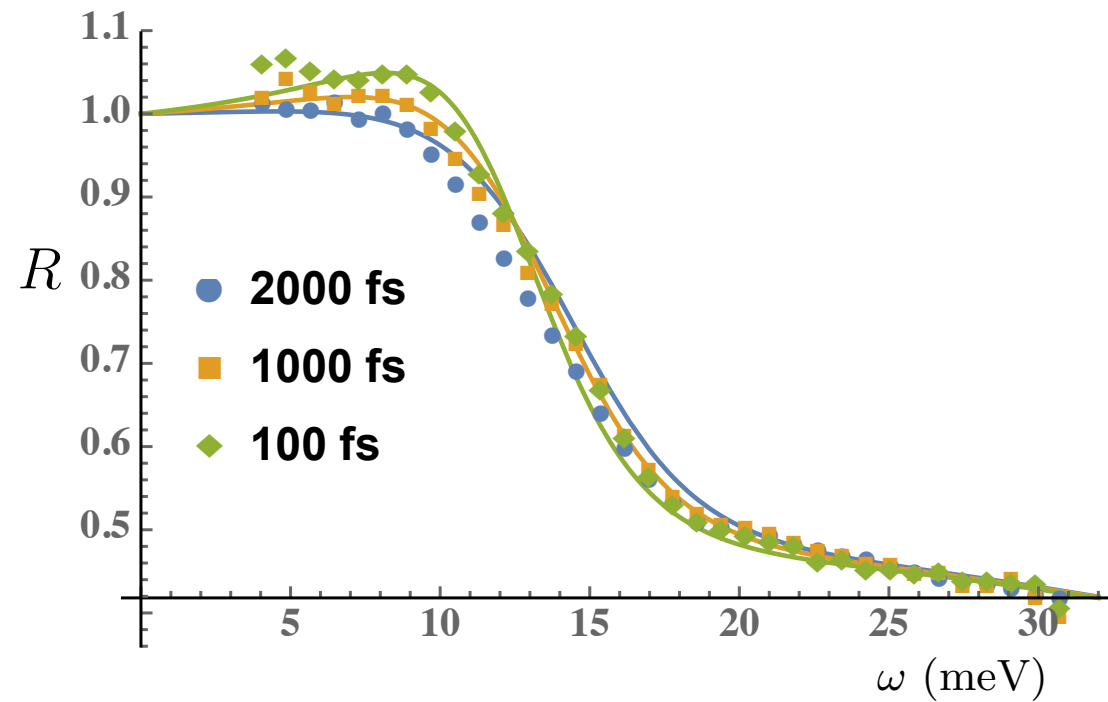


Good match to data for

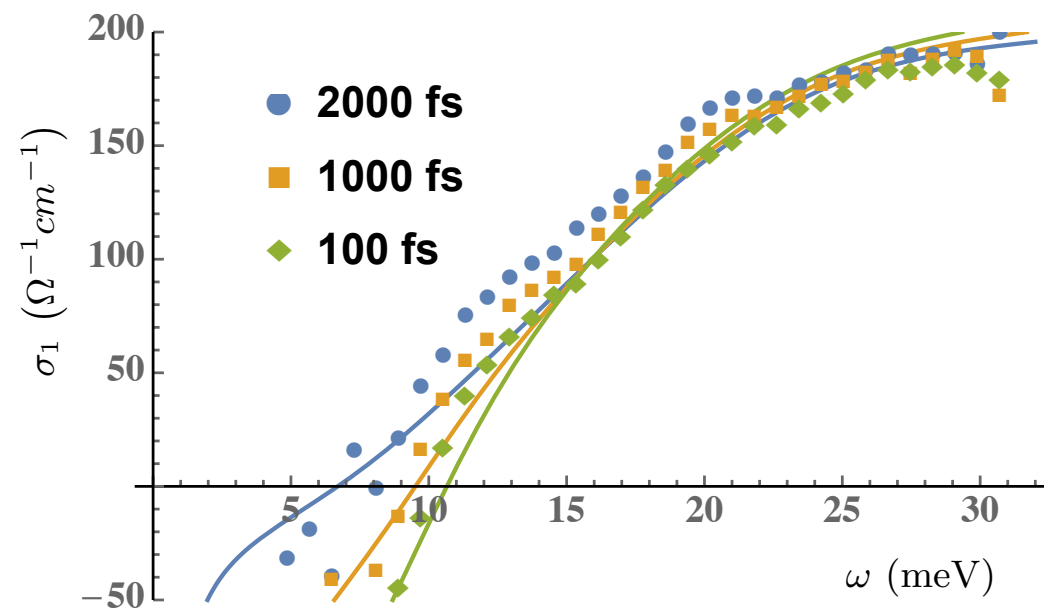
$$\omega_H = 24 \text{ meV}$$

Fit to experiment

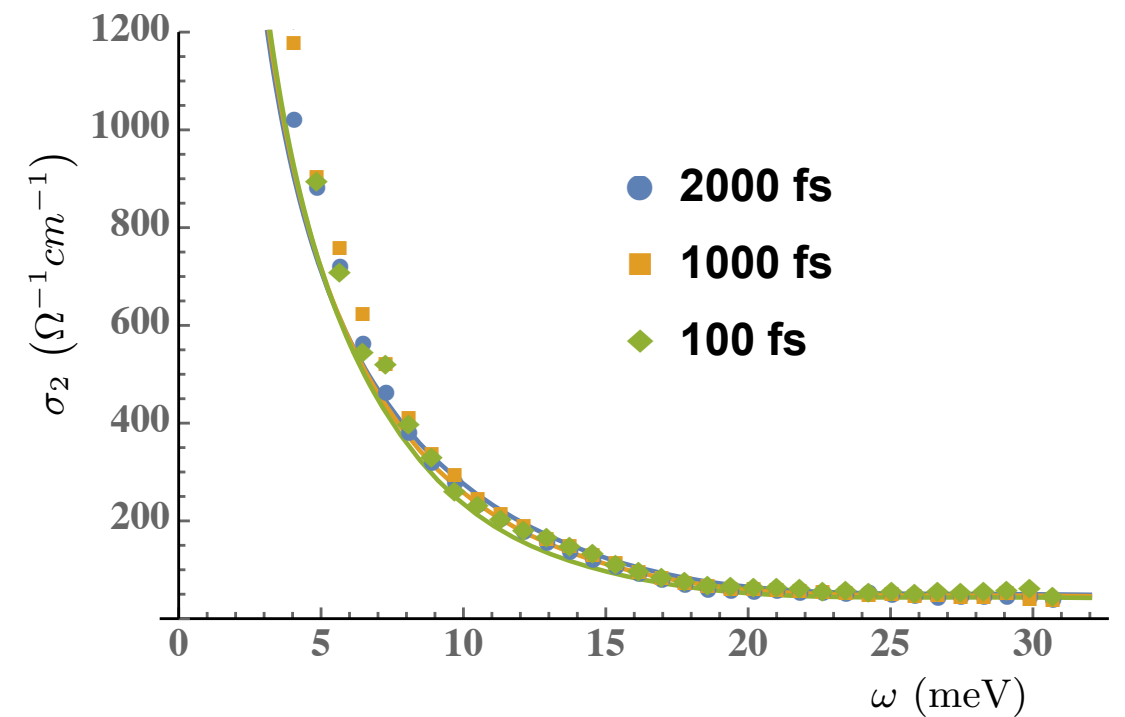
$$\omega_H = 24 \text{ meV}$$



τ (fs)	$\Lambda_s/\Lambda_{s,\text{eq}}$	Λ_m/Λ_s
2000	1.05	0.2
1000	1.11	0.37
100	1.17	0.47



(a) Real part

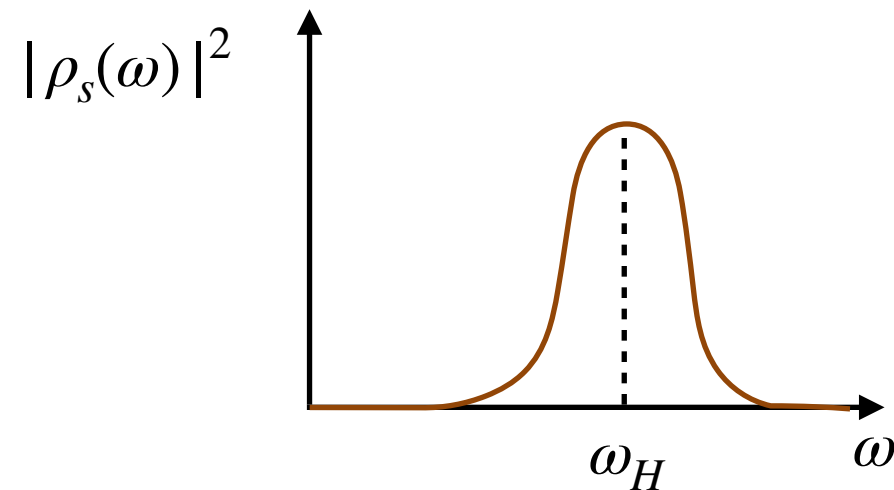


(b) Imaginary part

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

Giuliano Chiriacò,¹ Andrew J. Millis,^{1,2} and Igor L. Aleiner¹

¹*Department of Physics, Columbia University, New York, New York 10027, USA*

²*Center for Computational Quantum Physics, The Flatiron Institute, New York, New York 10010, USA*



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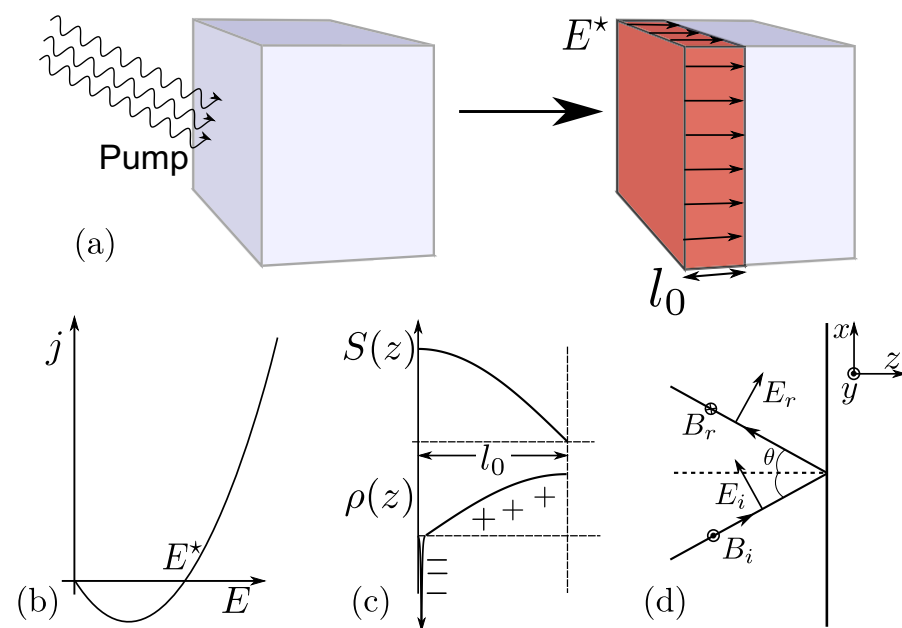


FIG. 2. (a) Sketch of incident pump pulse leading to spontaneous polarization in the active layer. (b) Sketch of the j - E characteristic for a $\sigma < 0$ state. (c) Entropy and charge density profiles inside the active layer; notice the thin surface charge accumulation on the external boundary of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

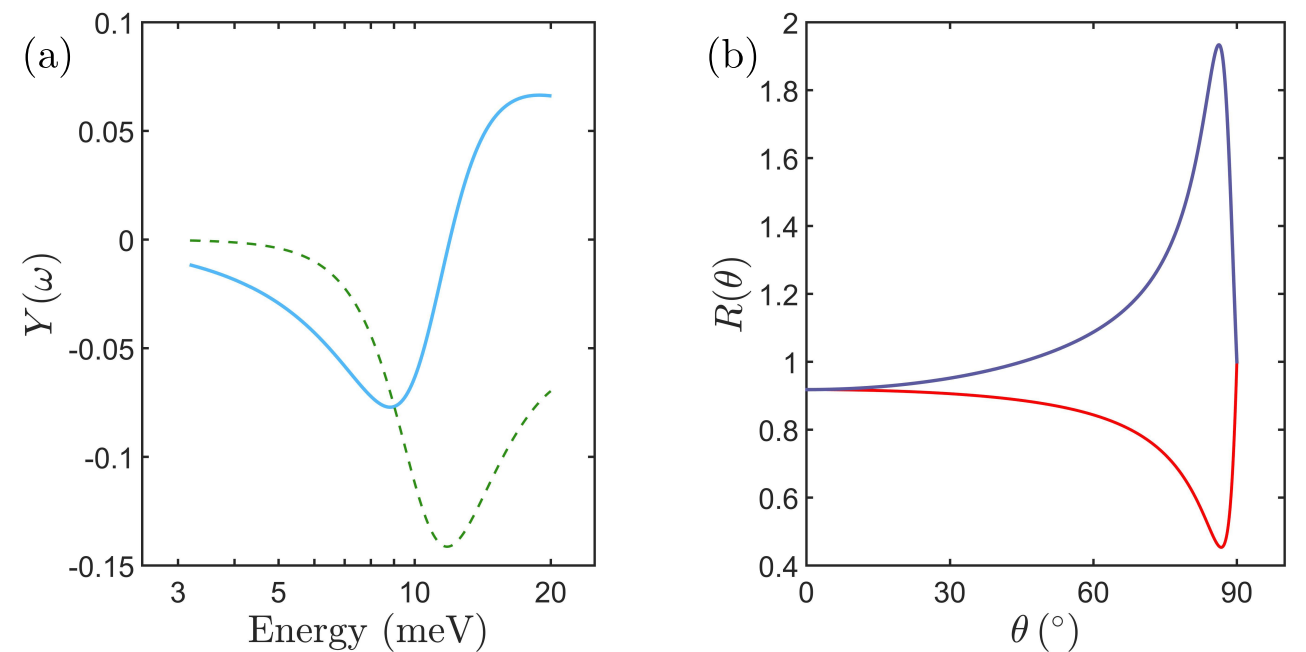
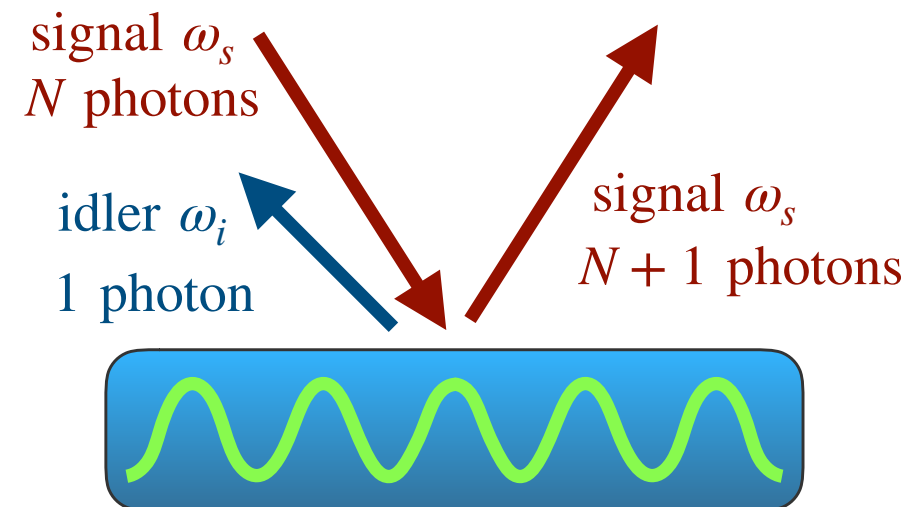


FIG. 3. (Color online) (a) Plot of the real (solid line) and imaginary (dashed line) part of $Y(\omega)$. (b) Theoretical $R(\theta)$ of K_3C_{60} at $\omega = 6 \text{ meV} = 1.44 \text{ THz}$ in equilibrium (red) and non-equilibrium (blue); notice the marked dependence on the angle and the presence of a region where $R > 1$. Plots are for the parameters of Fig. 1a.

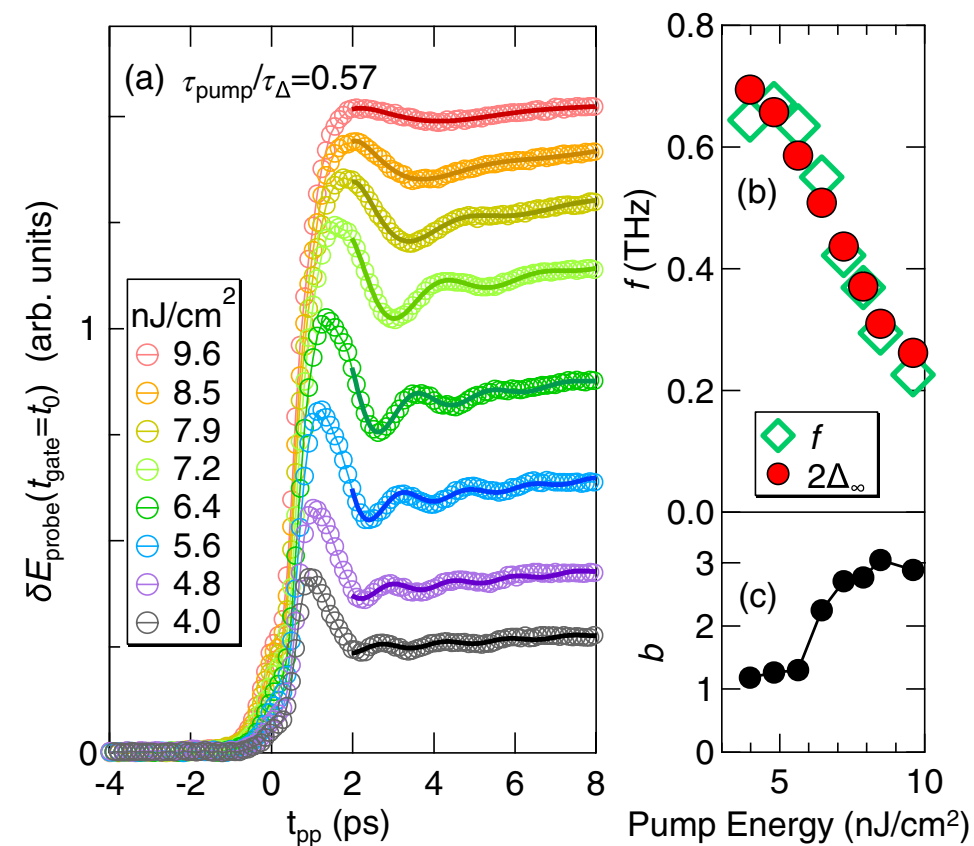
Further experimental checks?

Idler beam?



Angular dependence?

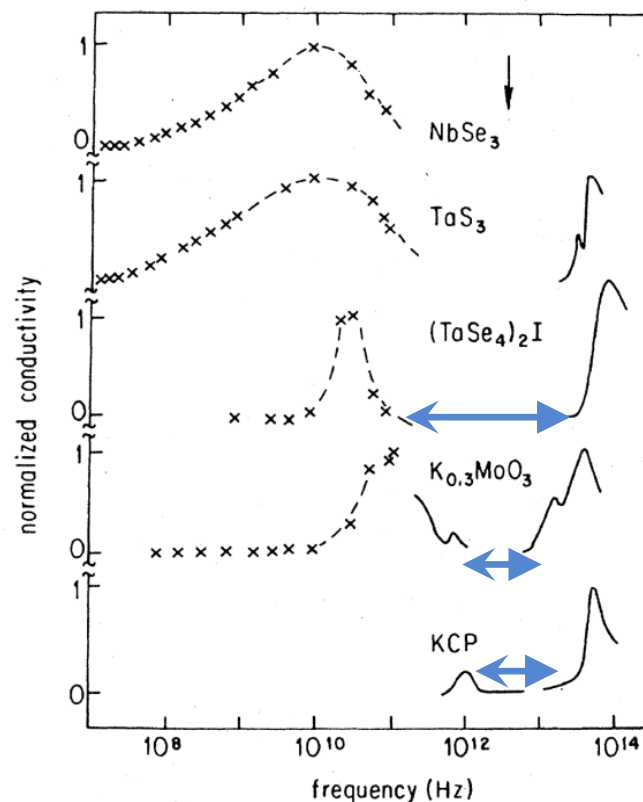
Direct observation of Higgs oscillations?



Higgs amplifier in other types of system?

Electrodynamics of CDW with excited Higgs mode

Gruner, Rev Mod Phys (1988)



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 $\omega_p = 24$ THz

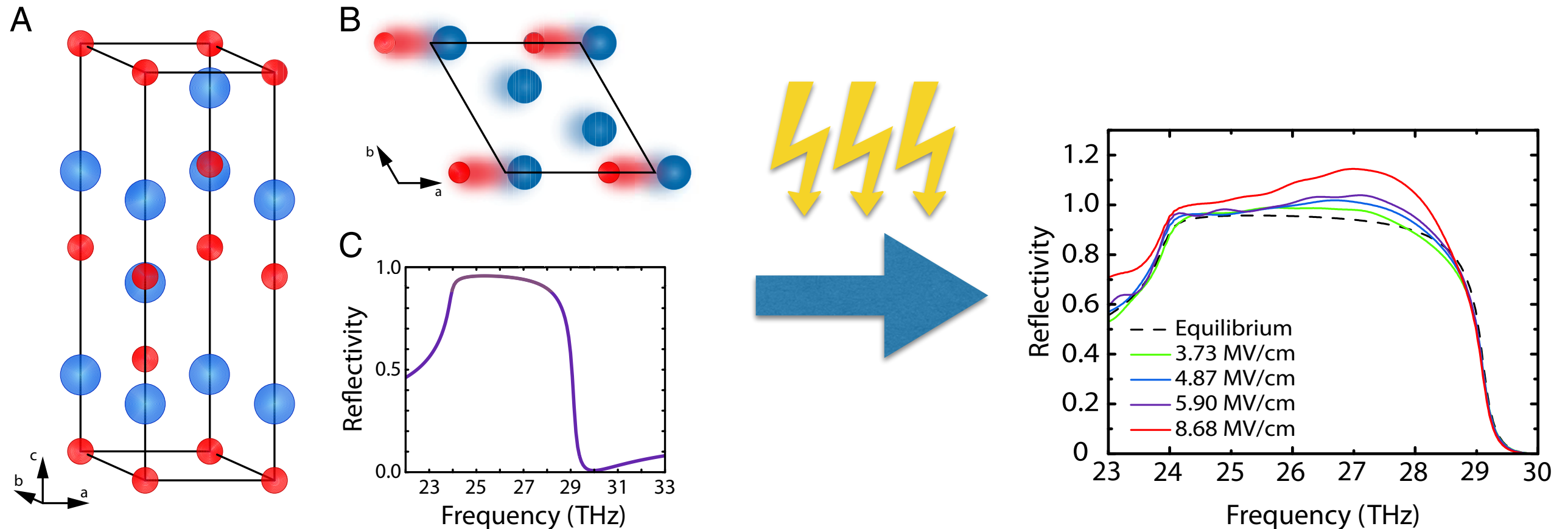


Fig. 3. (A) Crystal structure of SiC, polytype 4H (space group C_{6v}^4 -P6₃mc). Si atoms in blue, C atoms in red. (B) Eigenvectors of the infrared active mode excited by the pump pulse (E_u symmetry). (C) Reflectivity at equilibrium associated to the driven mode [data from literature (24)].

Parametric amplification of optical phonons

A. Cartella^{a,1}, T. F. Nova^{a,b}, M. Fechner^a, R. Merlin^c, and A. Cavalleri^{a,b,d}

PNAS | November 27, 2018 | vol. 115 | no. 48 | 12149

Phonon coupled to EM field

Potential for phonon and electric field:

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

Phonon EOM:

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

Maxwell equations:

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

$$\nabla \times \mathbf{E} + \partial_t \mathbf{B} = 0$$

Polarization:

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

Combined:

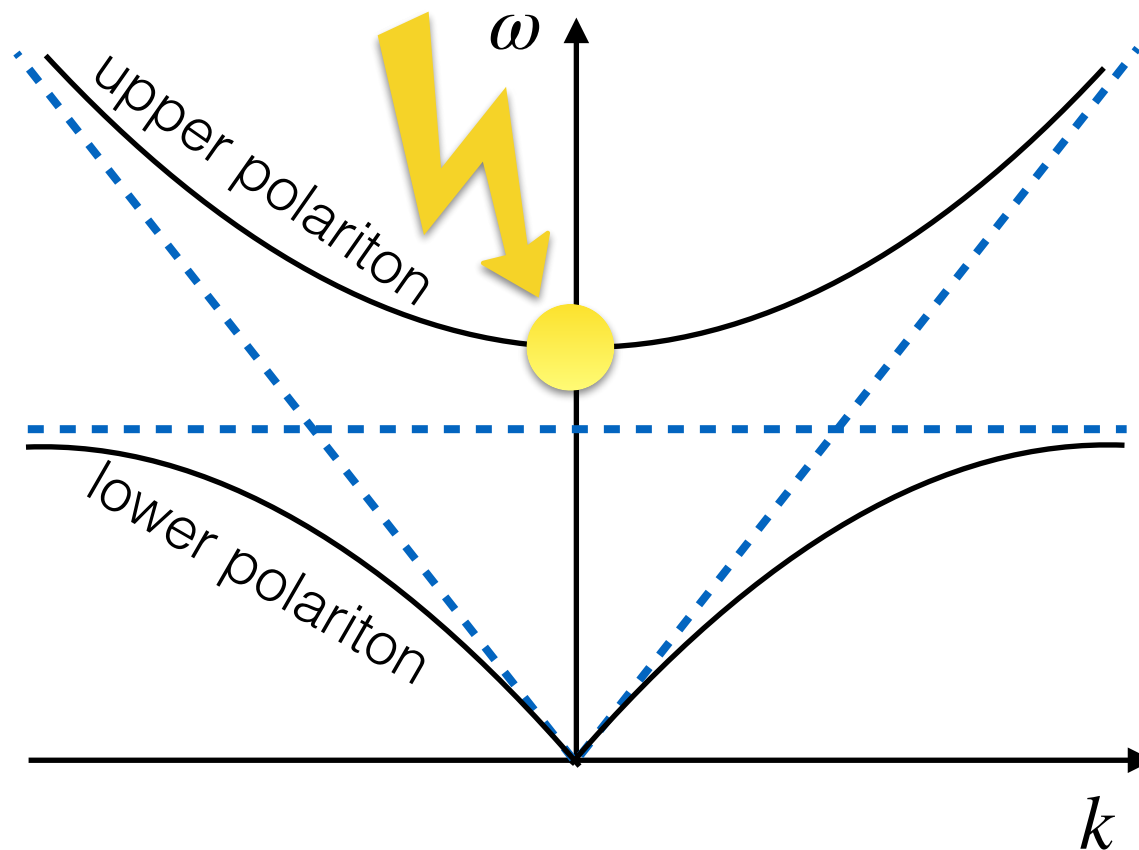
$$-\nabla^2 \mathbf{E} + \frac{1}{c^2} \partial_t^2 \left(\epsilon_\infty \mathbf{E} + \frac{\eta}{\epsilon_0} \mathbf{Q} + \frac{\alpha}{\epsilon_0} \mathbf{Q}^3 + 2 \frac{\beta}{\epsilon_0} \mathbf{Q}^2 \mathbf{E} \right) = 0$$

Polaritons

Linearized equations of motion:

$$\ddot{\mathbf{Q}} + \Gamma \dot{\mathbf{Q}} + \Omega_{\text{TO}}^2 \mathbf{Q} = \eta \mathbf{E}$$

$$-\nabla^2 \mathbf{E} + \frac{1}{c^2} \partial_t^2 \left(\epsilon_{\infty} \mathbf{E} + \frac{\eta}{\epsilon_0} \mathbf{Q} \right) = 0$$



$$\mathbf{Q}(t) = \mathbf{Q}_p \cos(\omega_p t)$$

Frequency mixing

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

$$\ddot{\mathbf{Q}} + \Gamma \dot{\mathbf{Q}} = -\Omega_{\text{TO}}^2 \mathbf{Q} + \eta \mathbf{E} + 2\beta \mathbf{E}^2 \mathbf{Q} + 3\alpha \mathbf{Q}^2 \mathbf{E}$$
$$-\nabla^2 \mathbf{E} + \frac{1}{c^2} \partial_t^2 \left(\epsilon_\infty \mathbf{E} + \frac{\eta}{\epsilon_0} \mathbf{Q} + \frac{\alpha}{\epsilon_0} \mathbf{Q}^3 + 2\frac{\beta}{\epsilon_0} \mathbf{Q}^2 \mathbf{E} \right) = 0.$$

mix frequencies: $\omega_1 + \omega_2 = 2\omega_p$

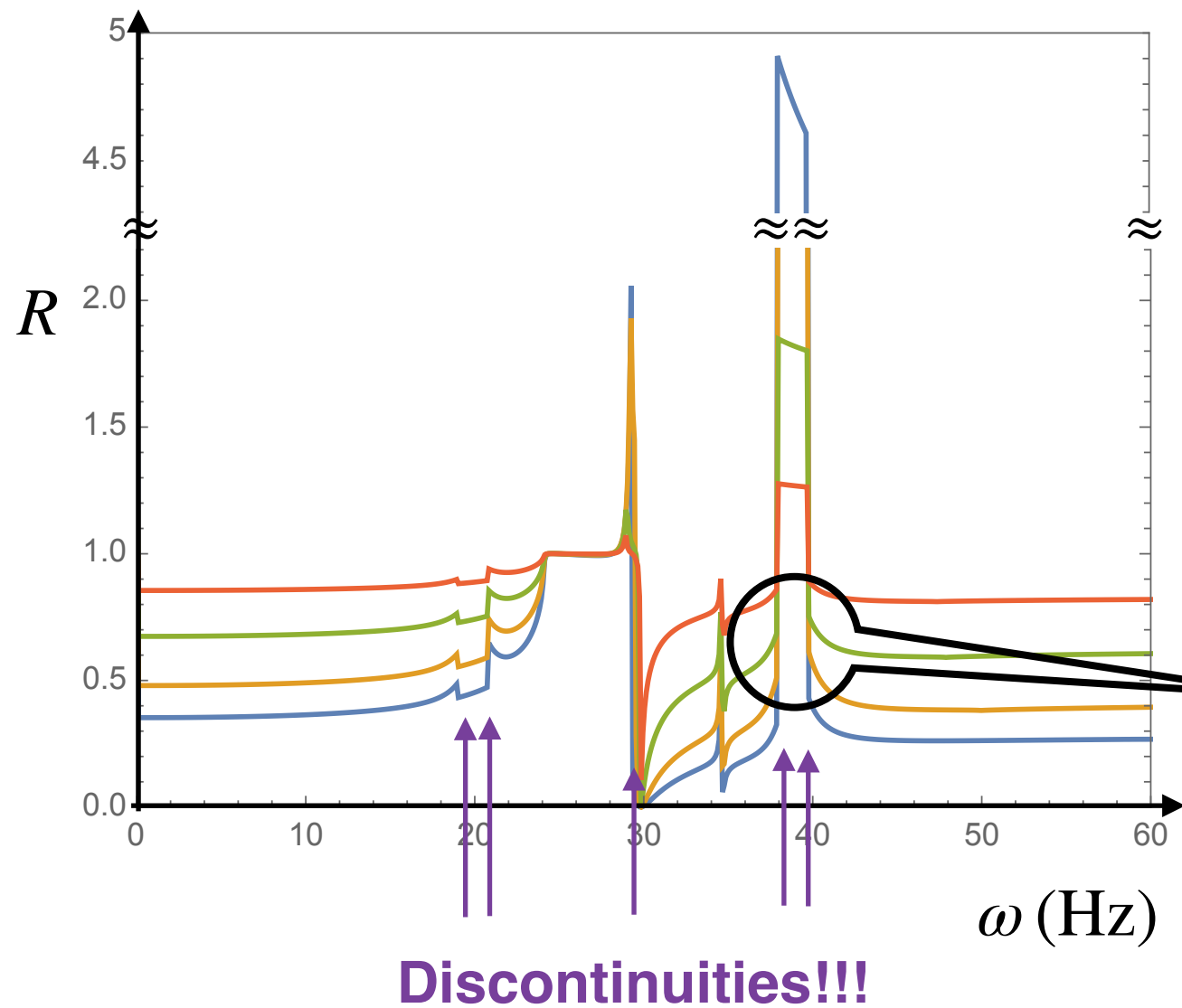
$$\mathbf{Q} = \mathbf{Q}_p \cos(\omega_p t) + \left(\mathbf{Q}_1 e^{-i\omega_1 t} + \mathbf{Q}_2^* e^{i\omega_2 t} \right) e^{ikz}$$

$$\mathbf{E} = \mathbf{E}_p \cos(\omega_p t) + \left(\mathbf{E}_1 e^{-i\omega_1 t} + \mathbf{E}_2^* e^{i\omega_2 t} \right) e^{ikz}$$

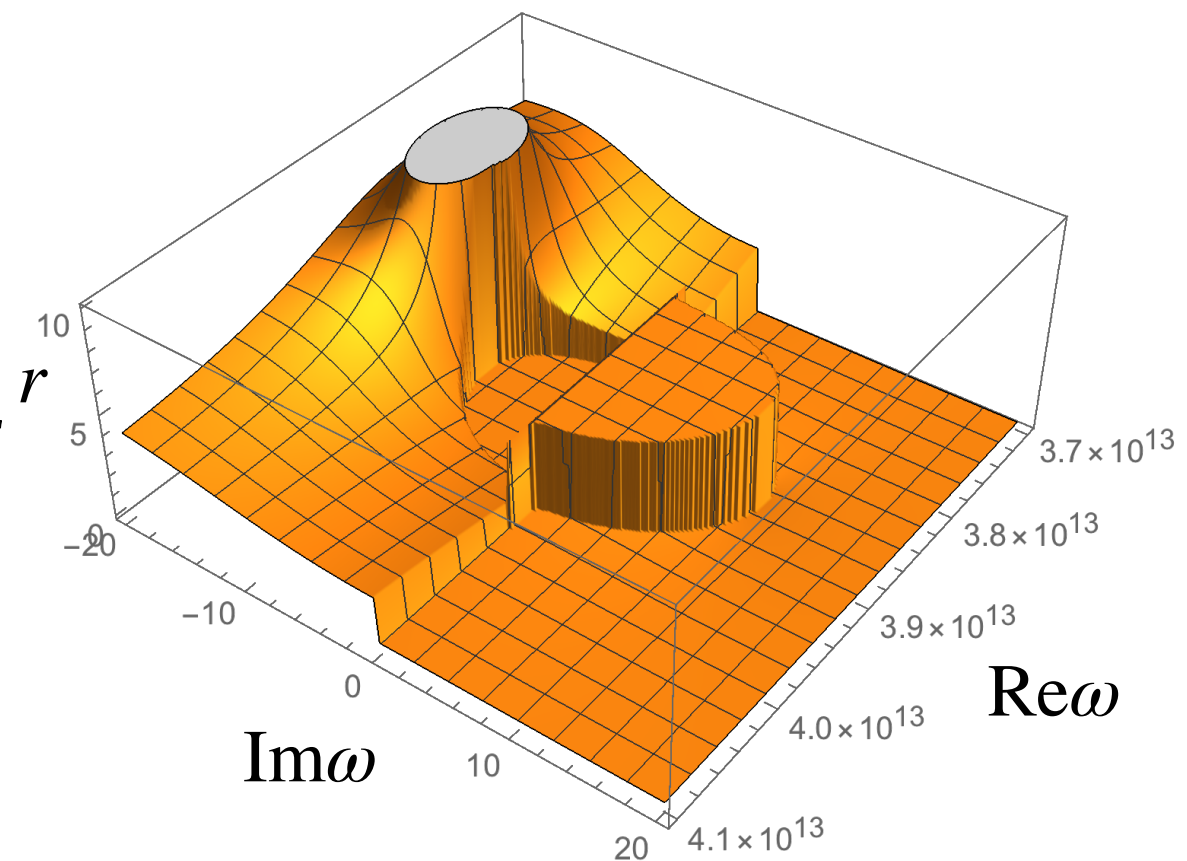

polariton,
strongly driven by pump


weak fields, from
probe

Results: Reflectivity of semi-infinite medium

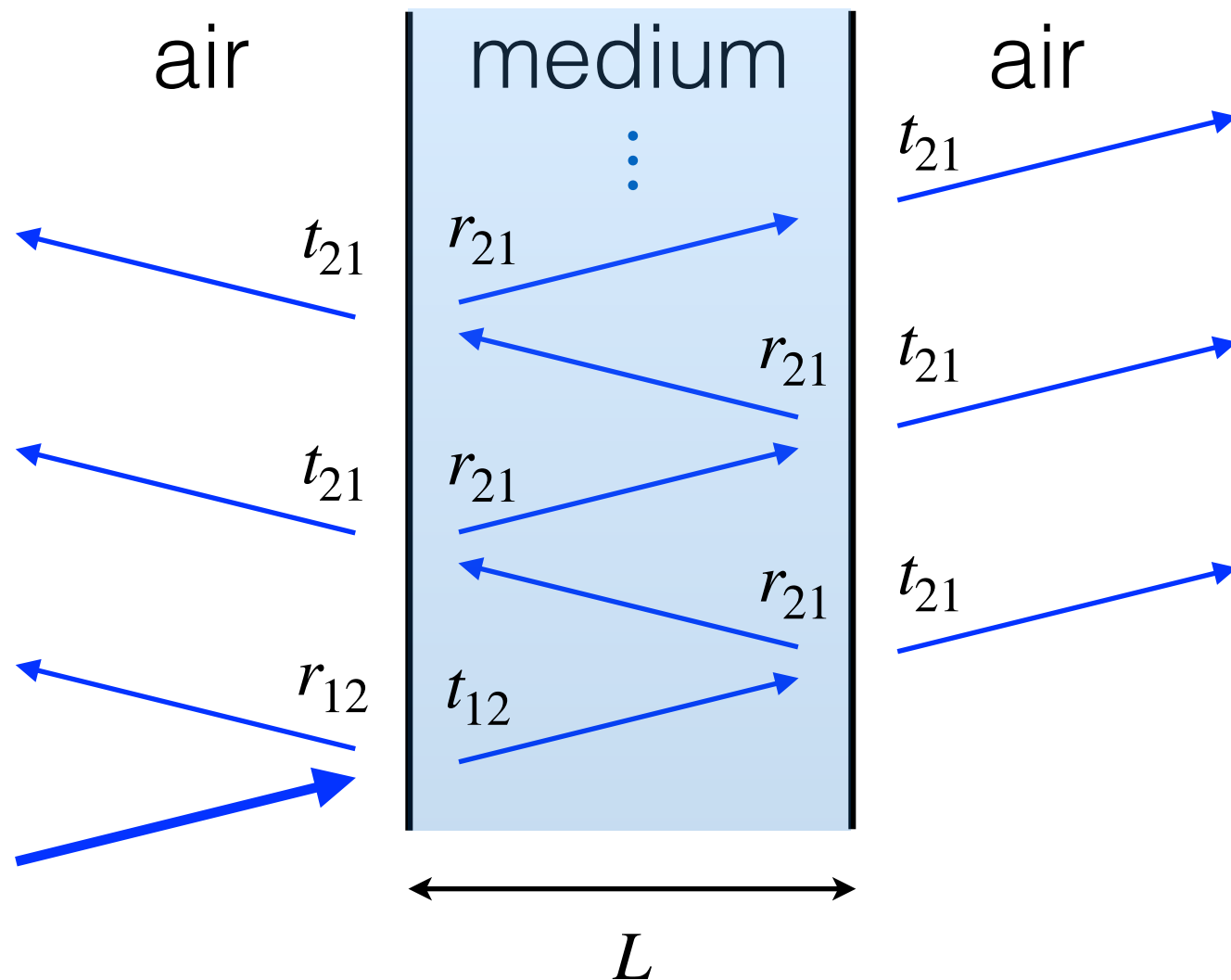


Complex frequency plane:



step discontinuity in upper half plane
→ acausal response! (= lasing)

Finite slab



$$a \equiv r_{21}^2 e^{2ikL}$$

$$r = r_{12} + t_{12}t_{21}r_{21}e^{2ikL} (1 + a + a^2 + \dots)$$

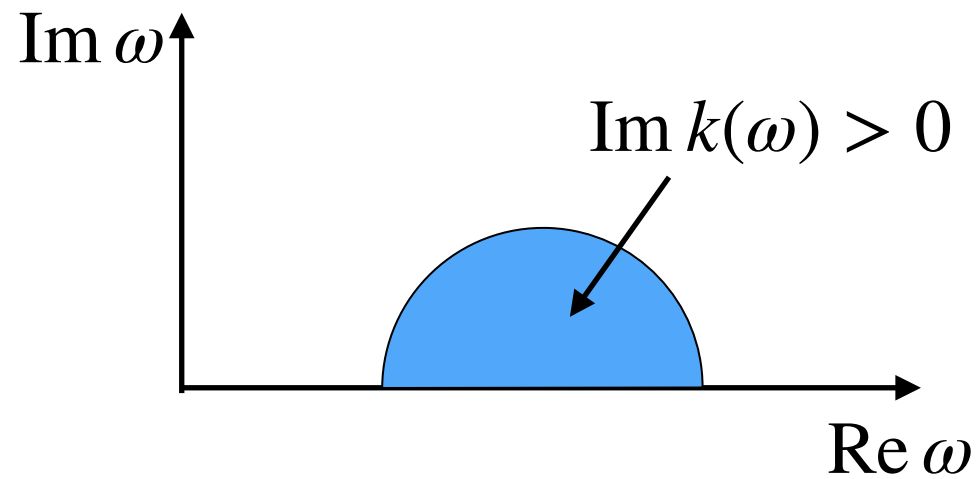
$$= r_{12} + \frac{t_{12}t_{21}r_{12}e^{2ikL}}{1 - a}$$

Pole in reflectivity for

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

Finite slab

Thin



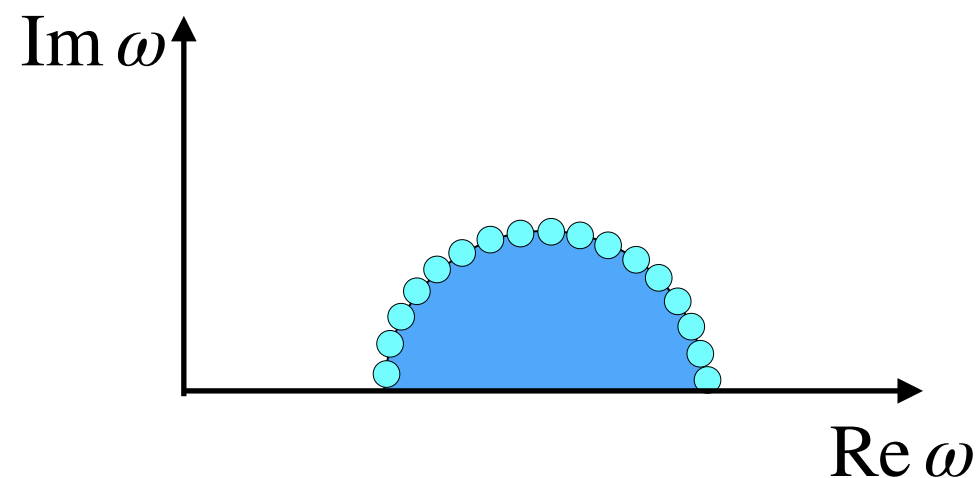
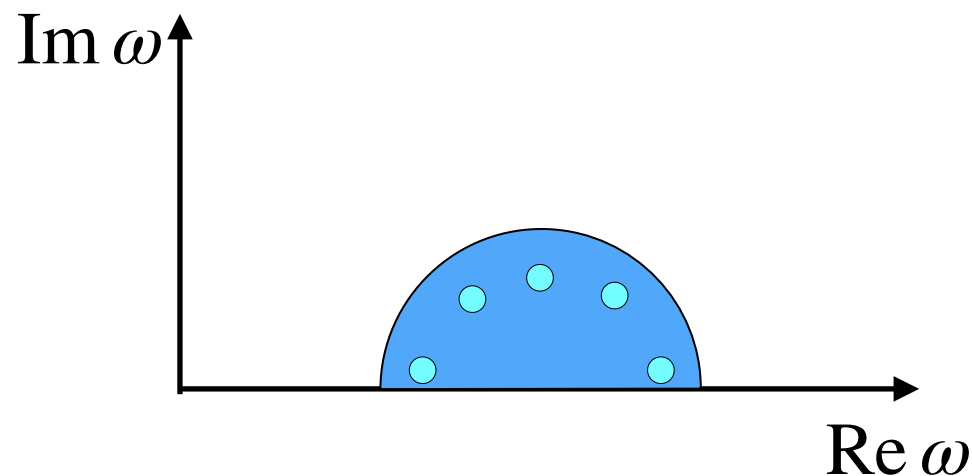
$$r = r_{12} + \frac{t_{12}t_{21}r_{12}e^{2ikL}}{1 - a}$$

Pole in reflectivity for

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

For thick slab, condition becomes:

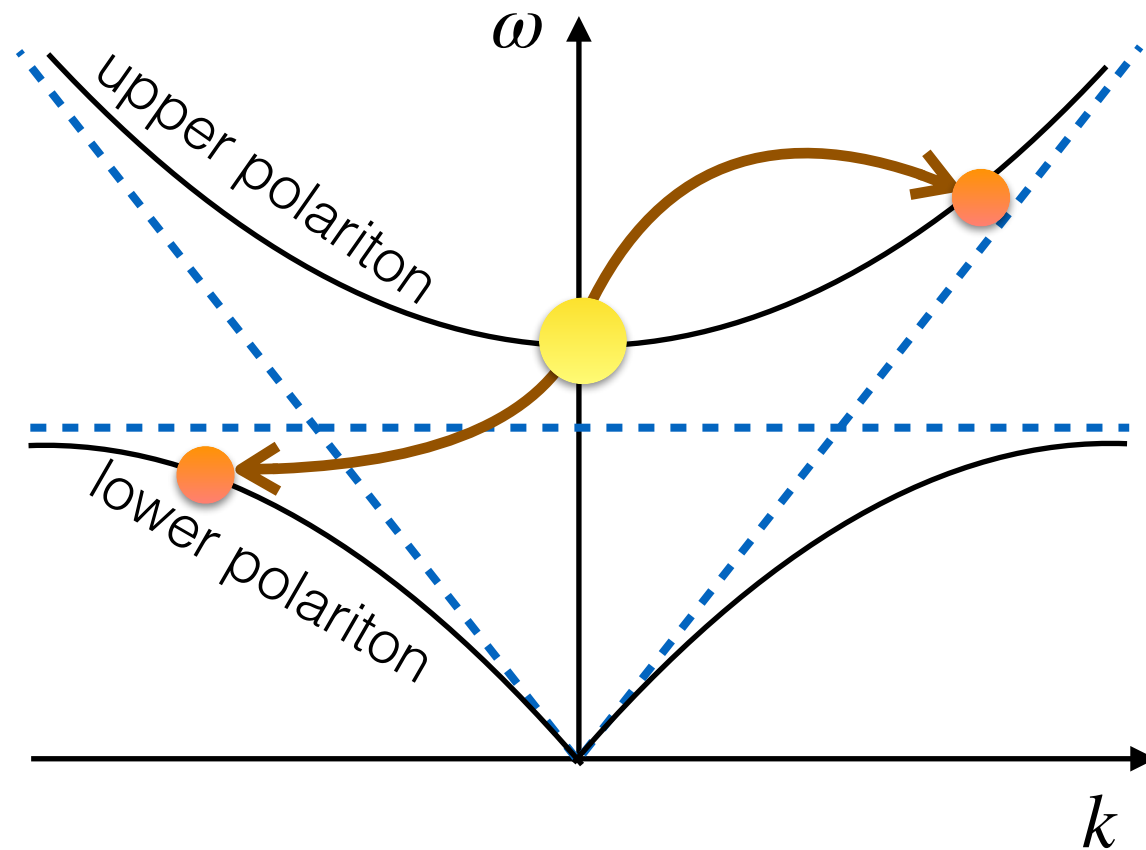
$$\text{Im } k = 0 \quad \text{Re } k = \frac{2\pi n}{L}$$



A line of poles gives a jump discontinuity

Thick

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

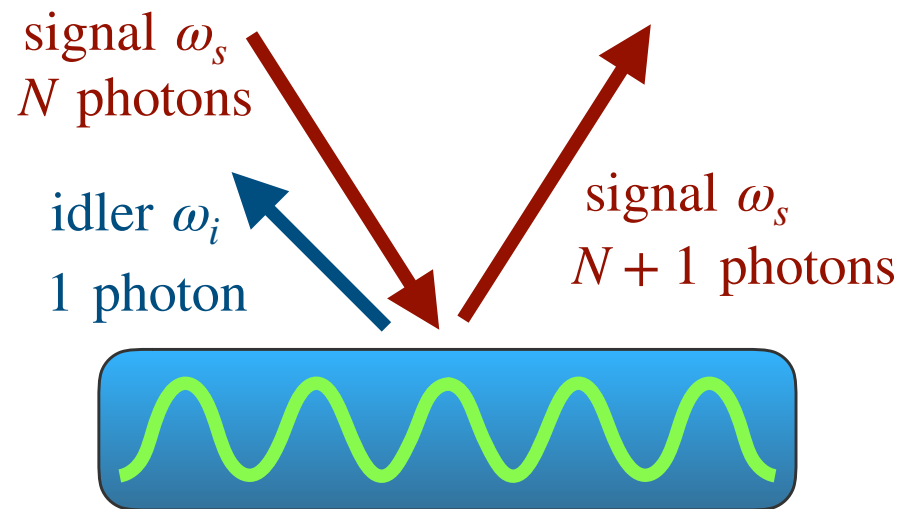
- Phonon lasing in SiC

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

Further experimental checks:

Idler beam?

Angular dependence?

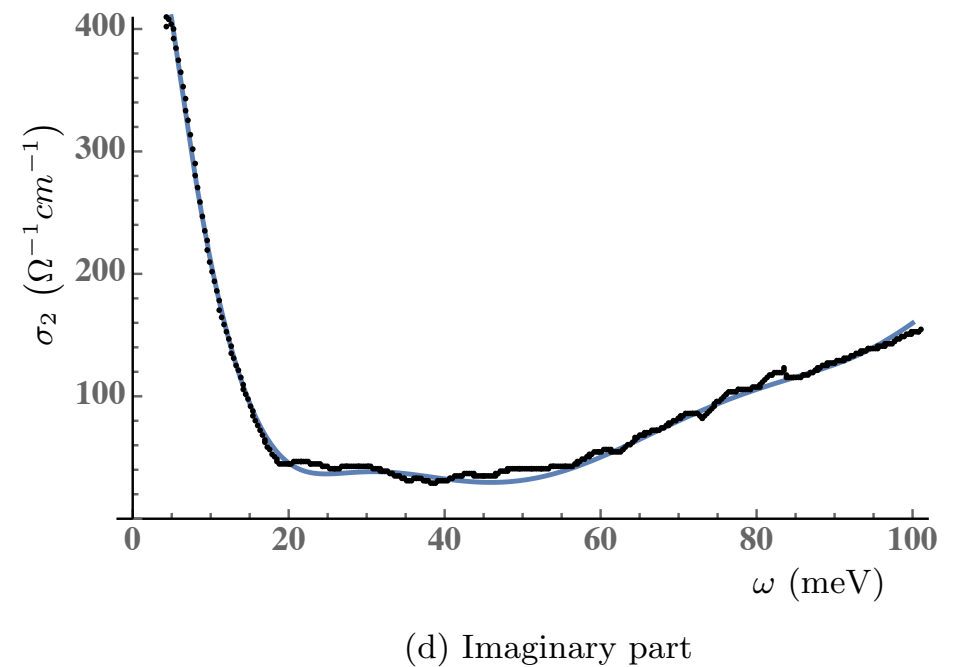
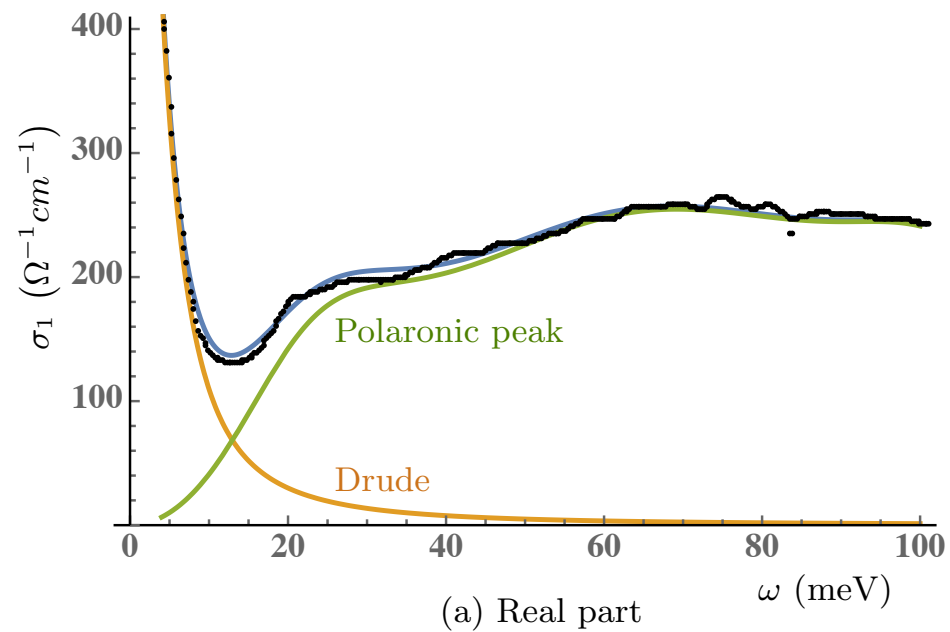


Microscopic calculations of Higgs amplification

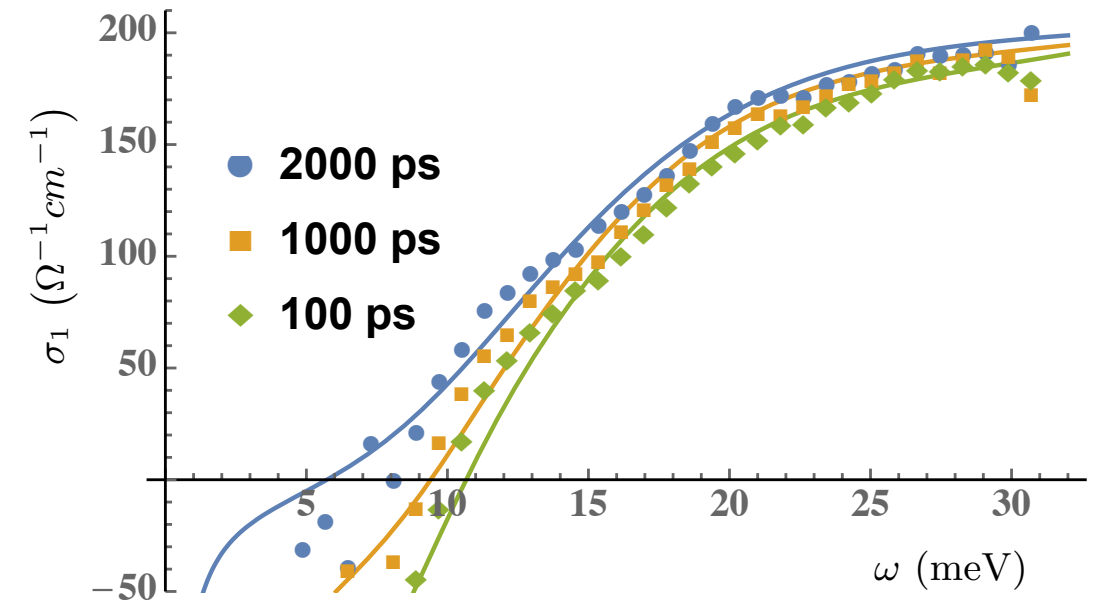
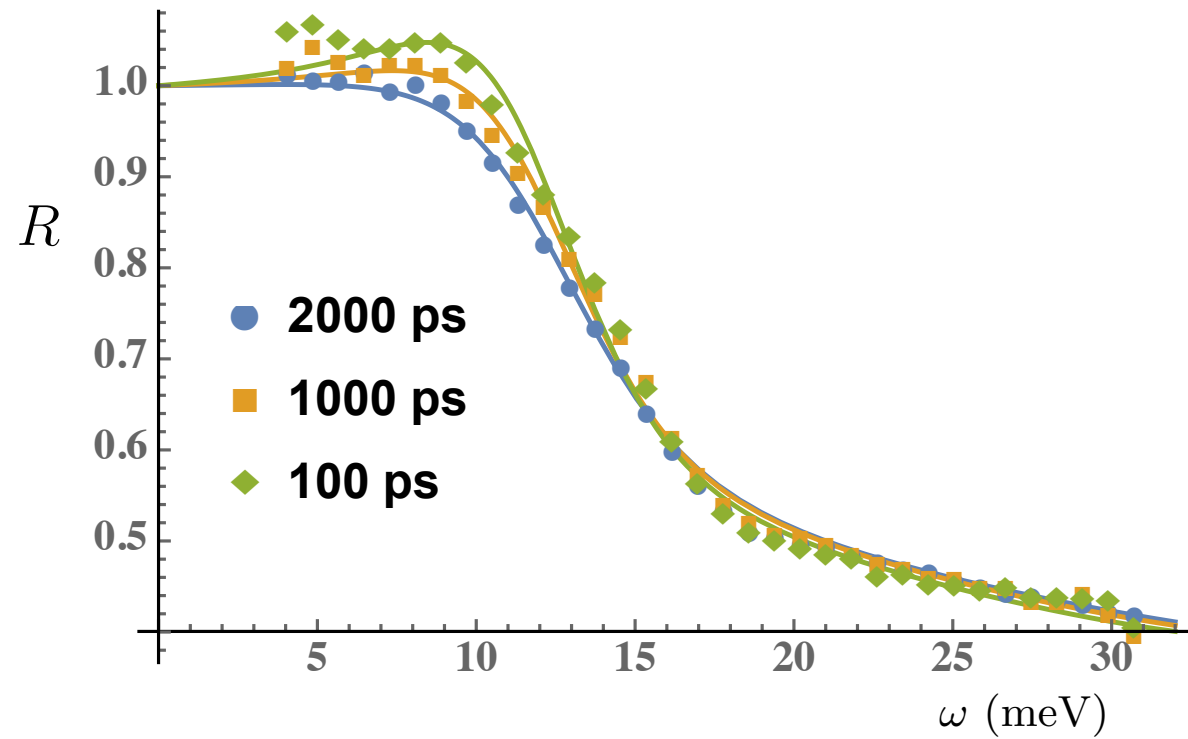
Do these effects have practical applications? How tunable are they?

Drude and polaronic peaks

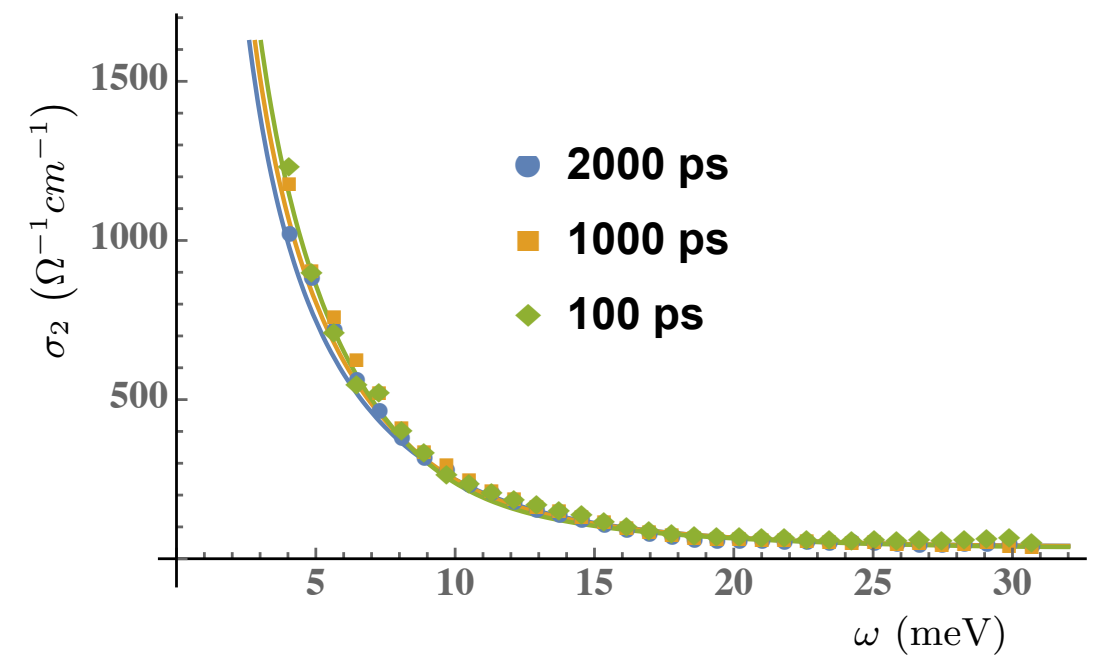
$$\sigma_{\text{eq}}(\omega) = \frac{\Lambda_{\text{s,eq}}}{\epsilon_0} \frac{1}{\gamma_{\text{D}} - i\omega} + \sum_{n=1}^3 \frac{B_n \omega}{i(\omega^2 - \Omega_n^2) + \gamma_n \omega}$$



Fit to experiment with adjusted polaronic peak

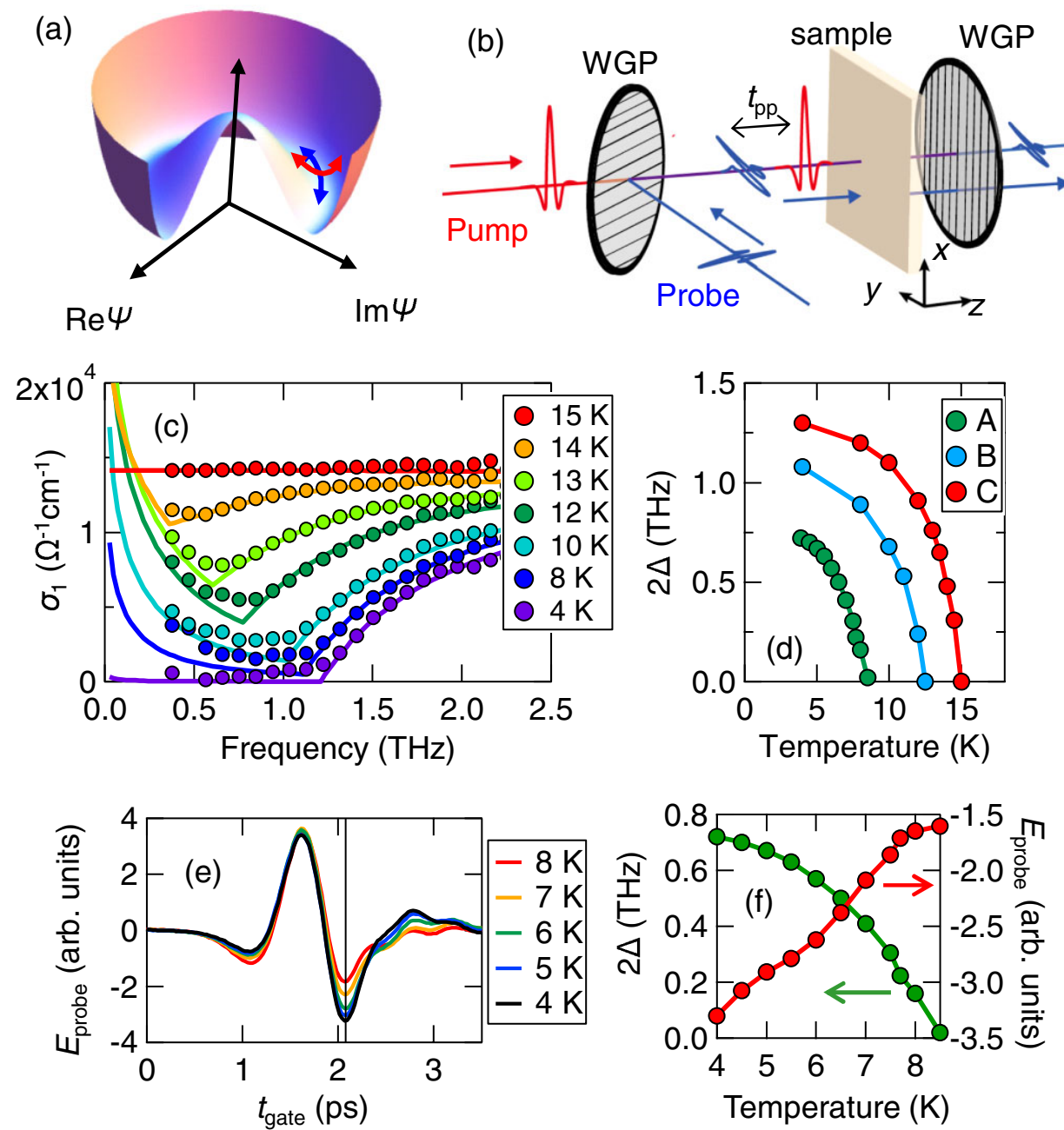


(a) Real part



(b) Imaginary part

Higgs Oscillations from THz pump



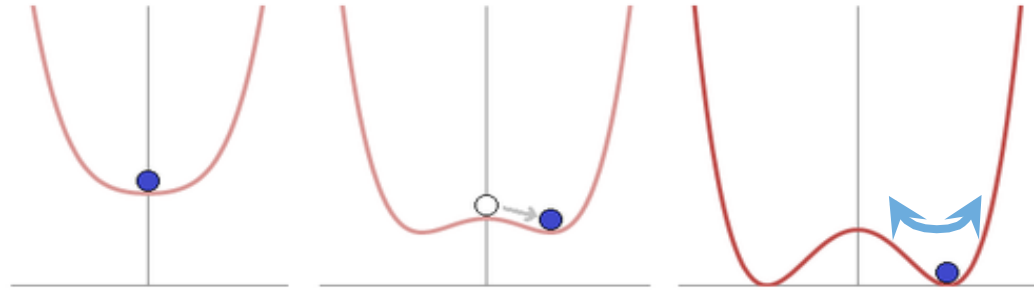
Phenomenological model for conductivity

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

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

τ (fs)	A	E_g	δ	Λ_s	E_s	E_e	Λ_m/Λ_s
2000	200	10.5	4.5	33.5	11.5	27	0.15
1000	190	11.0	4.0	37.2	12.0	27	0.28
100	180	11.5	4.0	40.9	12.5	27	0.37

Higgs oscillations



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

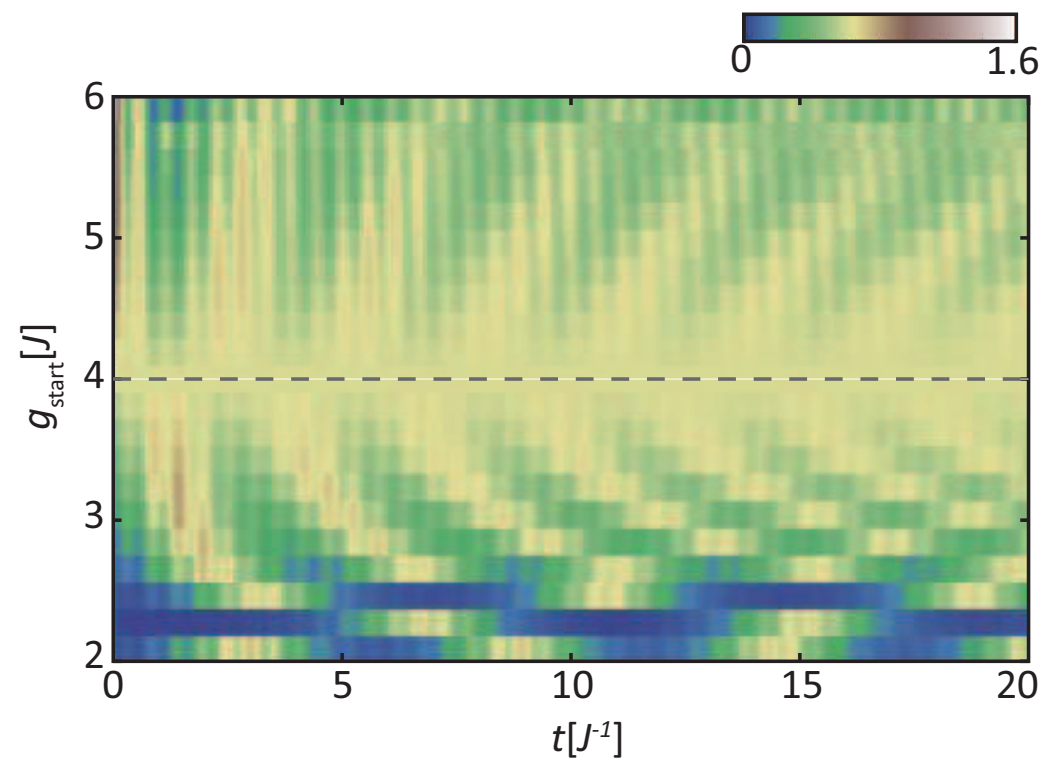


FIG. 9. Dynamics of the order parameter $\Delta^S(t)$ after the quantum quench from g_{start} equilibrium state to $g = 4J$ at $t = 0$. The dashed line denotes the static point $g_{\text{start}} = g$.

Higgs oscillations

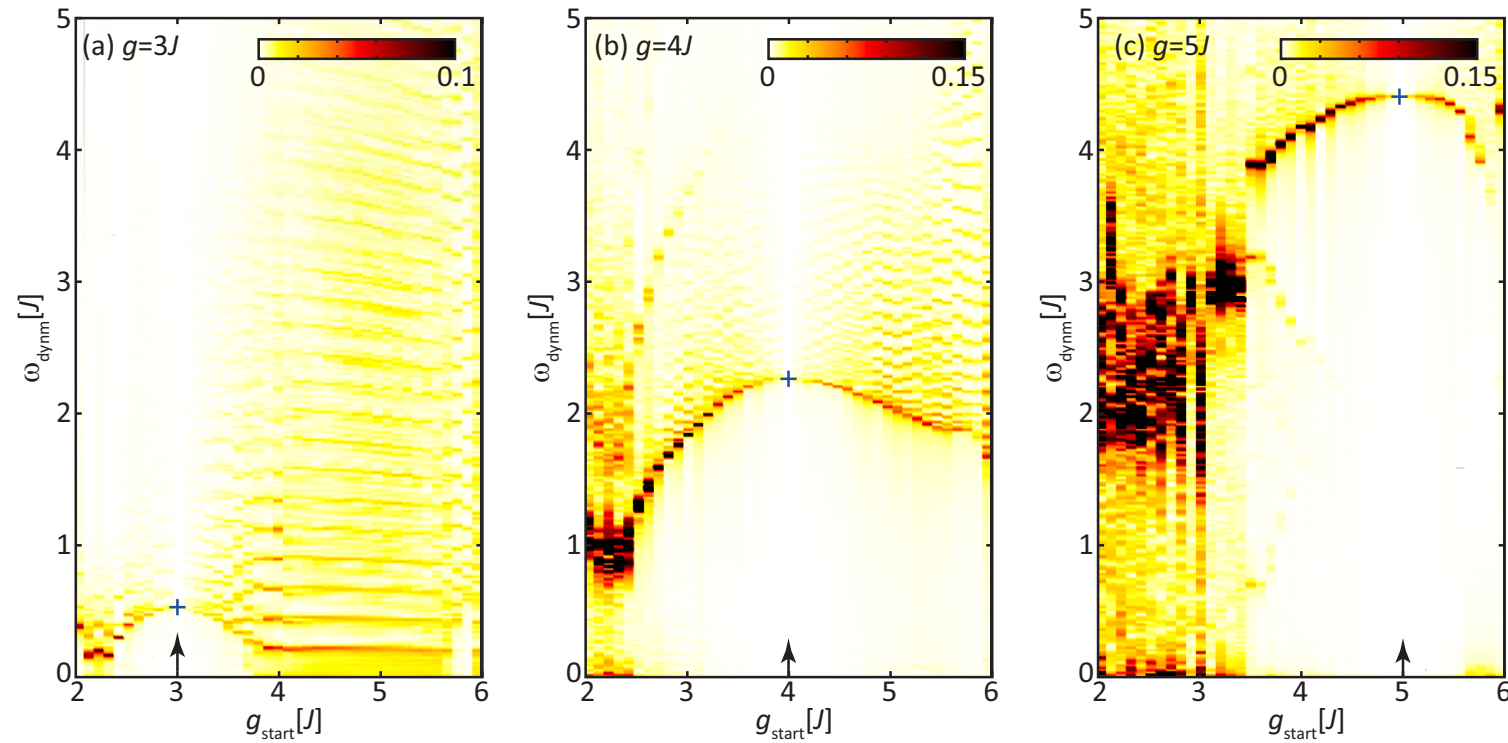
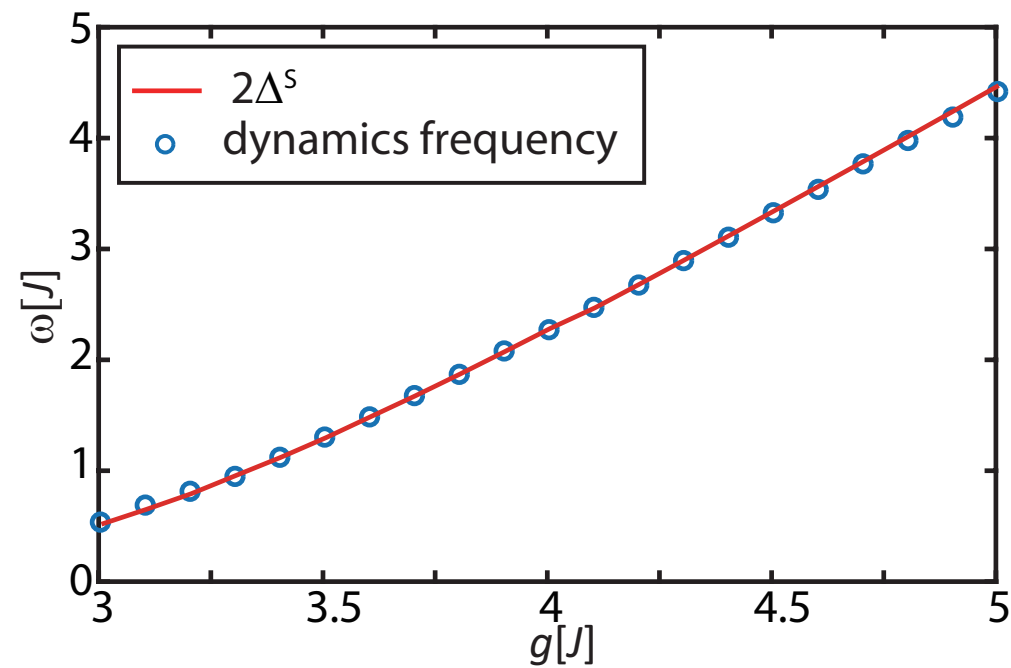


FIG. 10. The Fourier spectra of post quench dynamics in reference to the frequency ω_{dynam} , for different starting states g_{start} along the horizontal axis and different quenched interactions (a) $g = 3J$, (b) $g = 4J$ and (c) $g = 5J$. The black arrows denote the corresponding static point, while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



Higgs oscillations

$$H_{\text{drive}} = I(t) \sum_q x_q x_{-q}$$

$$x_q = \frac{1}{\sqrt{2}} (a_q + a_{-q}^\dagger)$$

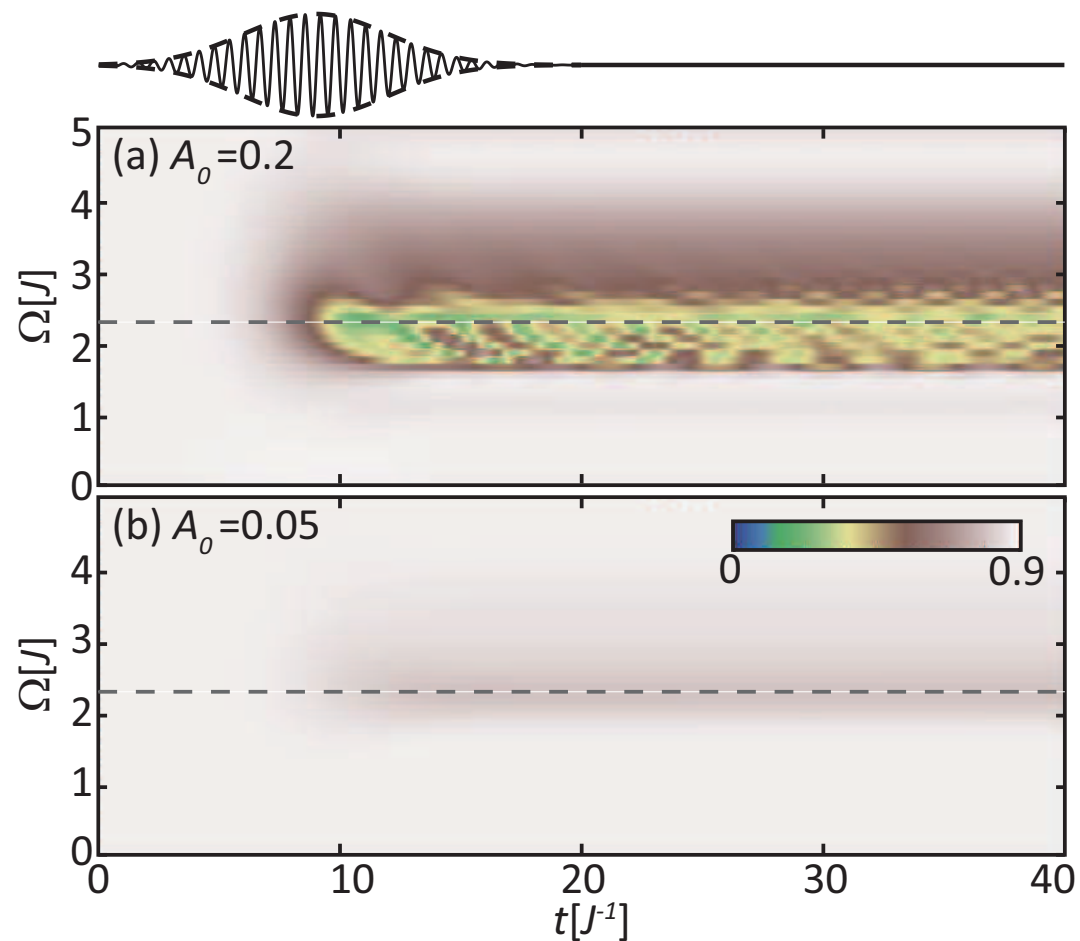


FIG. 12. Pumped induced dynamics of the order parameter Δ^S for various pump frequencies Ω s with (a) $A_0 = 0.2$ and (b) $A_0 = 0.05$. The black curve sketches the pump pulse, while the dashed line denotes the Higgs frequency $\omega_H = 2\Delta^S$ at this system.

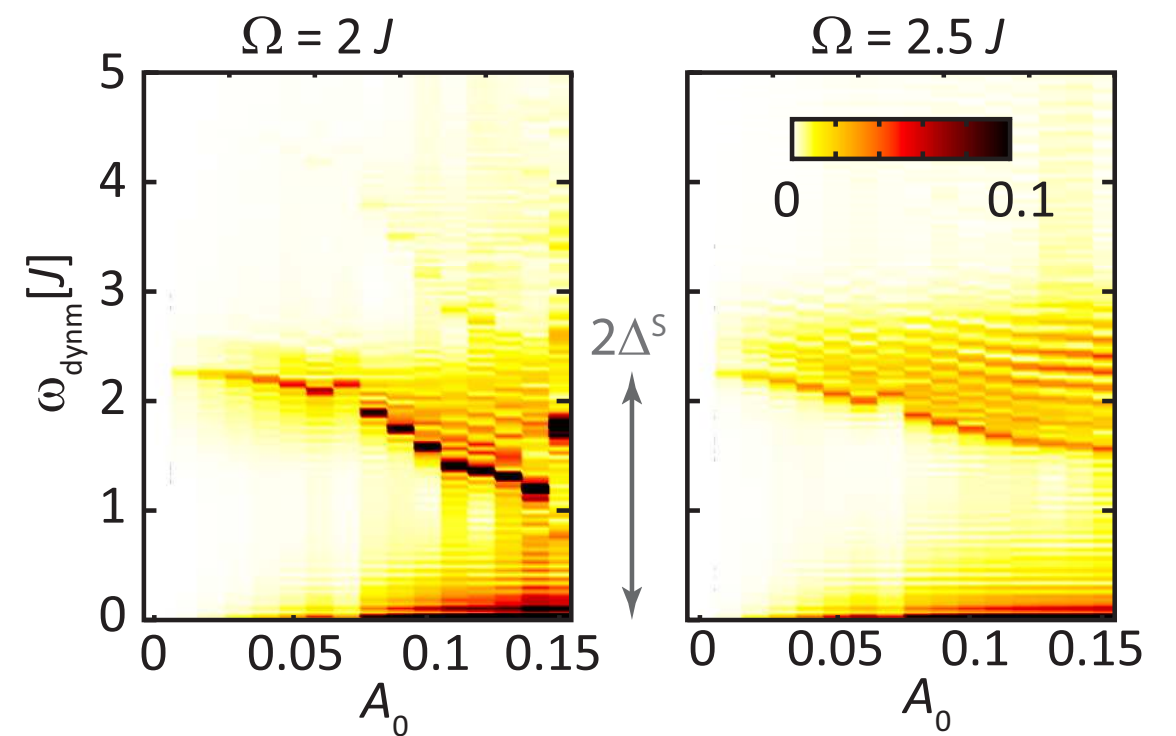
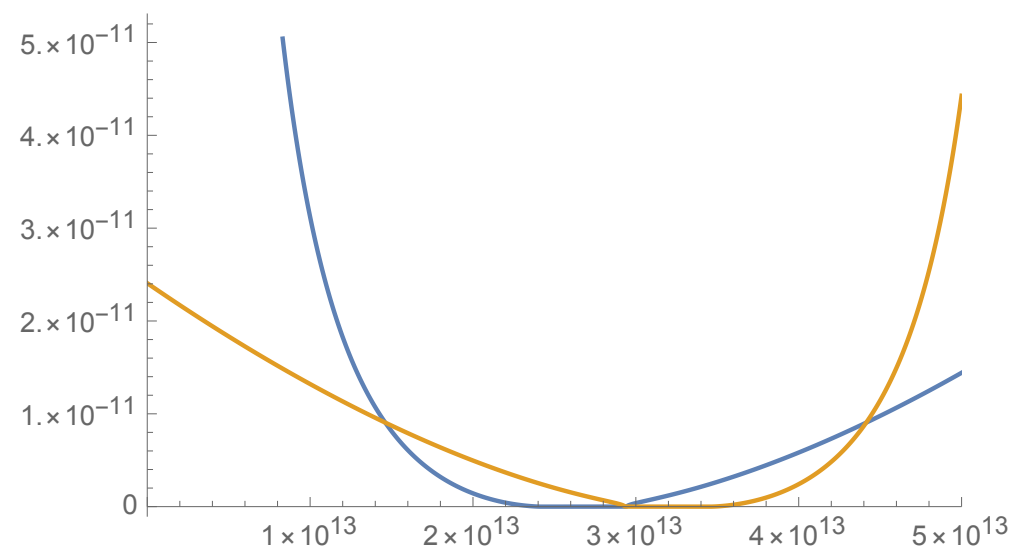
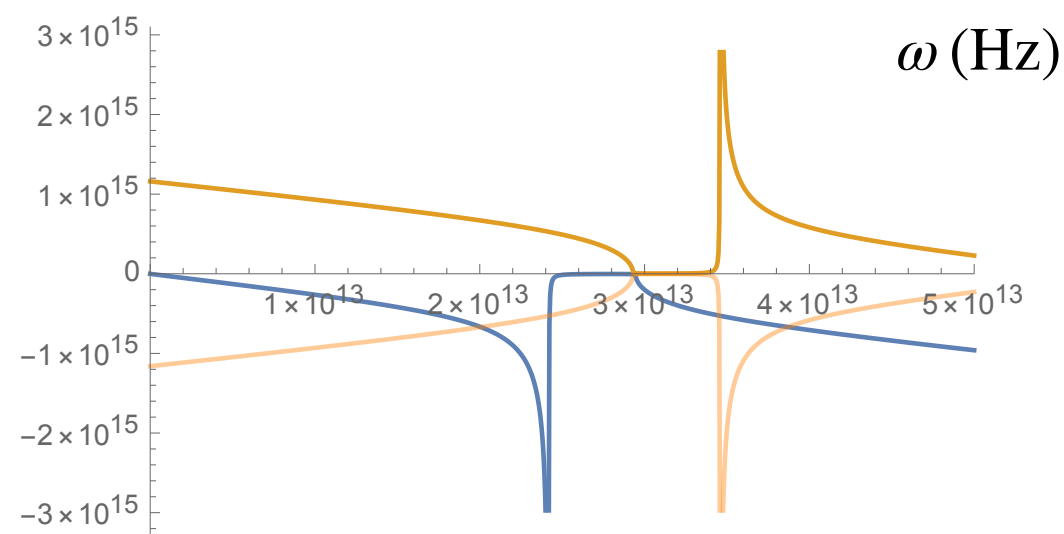
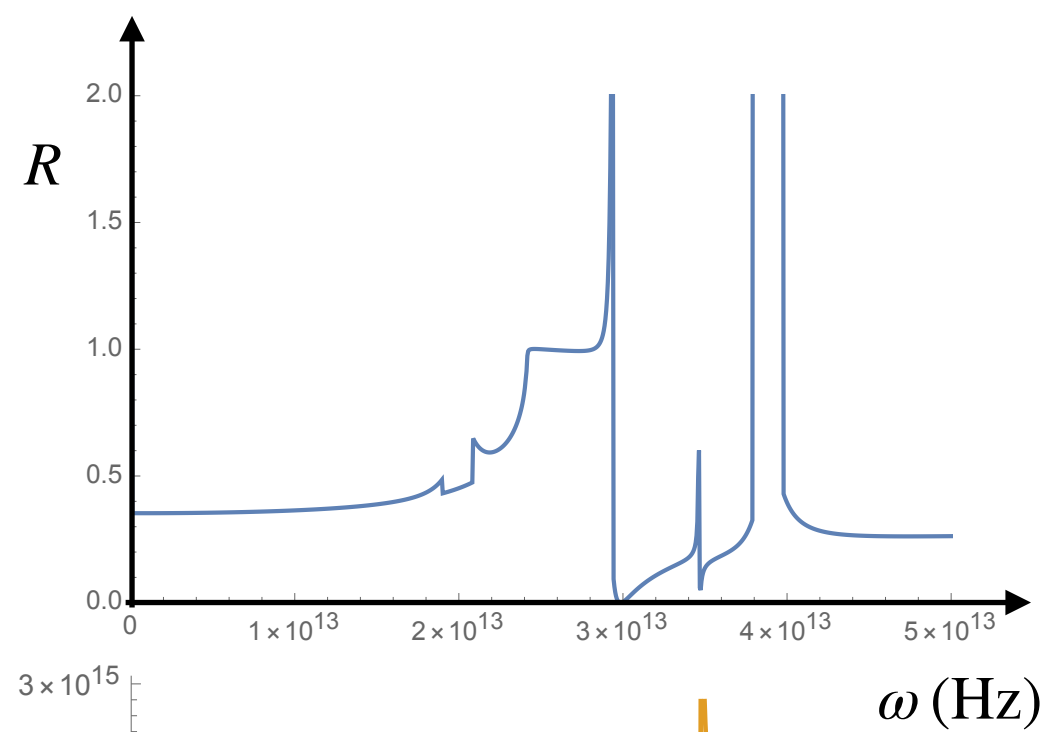
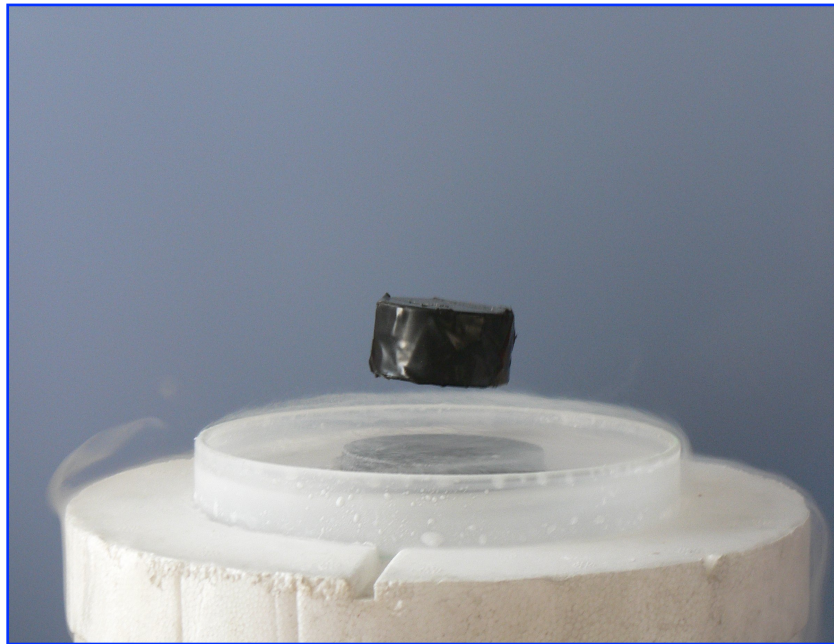


FIG. 13. Fourier spectra of dynamics induced by pump strength $A_0 = 0.01 \sim 0.15$. The left and right panels show two pump frequencies $\Omega = 2J$ and $\Omega = 2.5J$. The double-end arrow denotes twice of the order parameter.

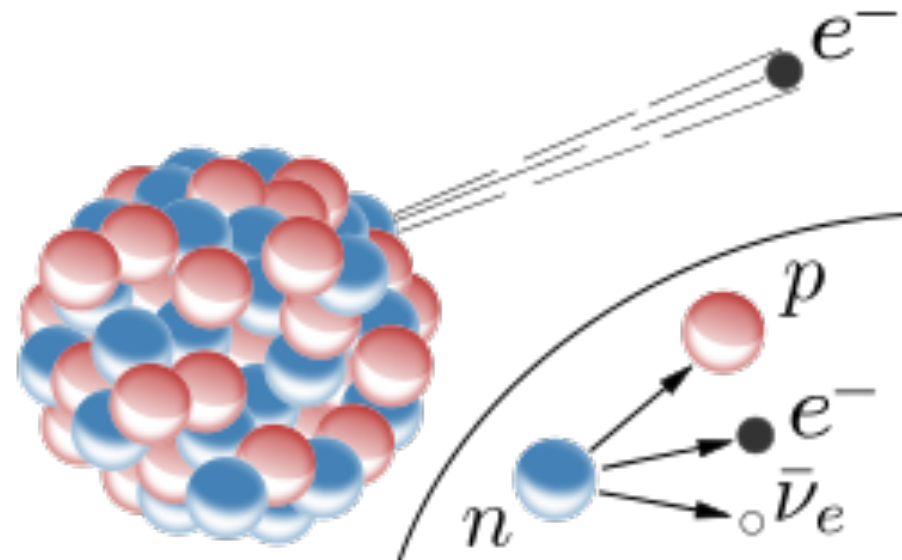


What do these two have in common?

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?

PHYSICAL REVIEW

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Plasmons, Gauge Invariance, and Mass

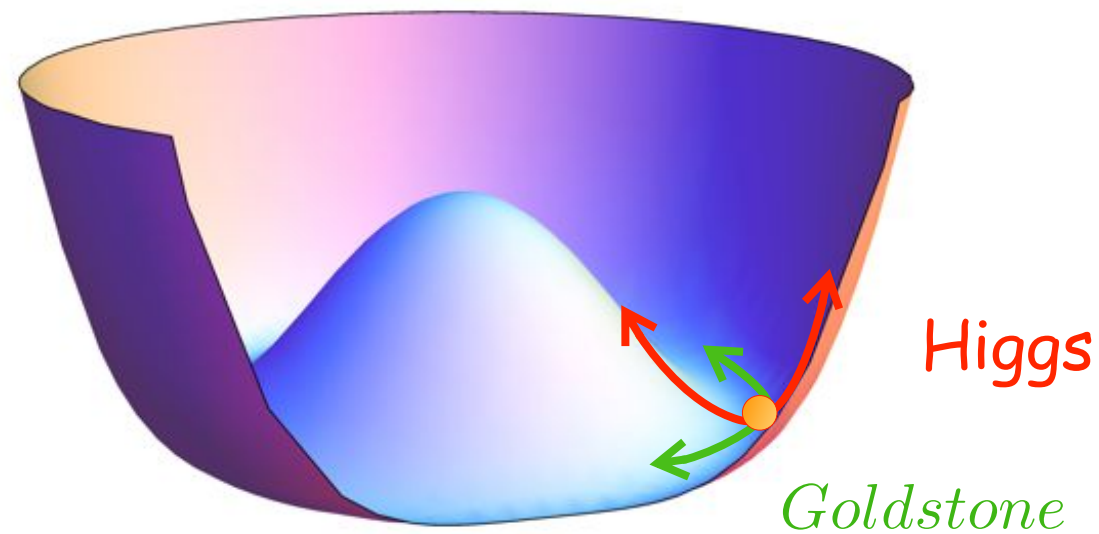
P. W. ANDERSON

Bell Telephone Laboratories, Murray Hill, New Jersey

(Received 8 November 1962)

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

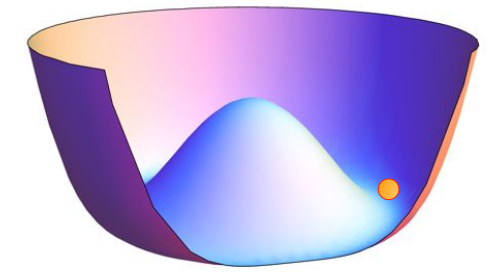
Higgs boson:

- 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: $\psi \rightarrow e^{-i\xi} \psi$, $\tilde{A}_\mu \equiv A_\mu - e^{-1} \partial_\mu \xi$

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 ξ particle !
- ★ Photon becomes massive triplet $\tilde{A}_\mu$
- ★ The Higgs boson (η) has mass, $m_H = \sqrt{2}m$, independent of e