Sapienza Università di Roma

Istituto dei Sistemi Complessi

OPTICAL RESPONSE OF JOSEPHSON-LIKE PLASMA WAVES IN SUPERCONDUCTORS

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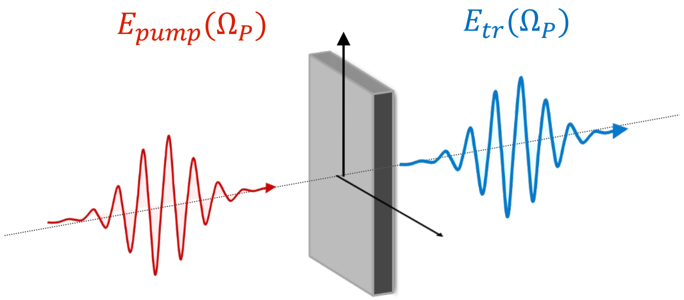
Spectroscopy of superconductors at strong THz fields

"Natural" non-linearity of (bulk) Josephson plasmons

$$H[\theta] = -J_s \sum_j \cos(\theta_{j+1} - \theta_j)$$

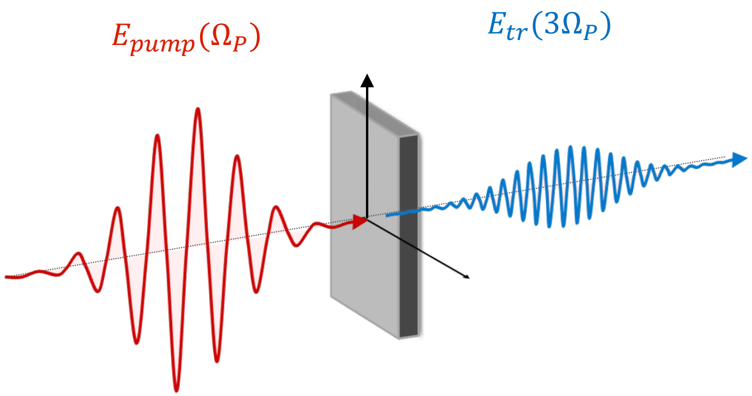
"Less trivial" non-linearity within a microscopic picture
(fermionic vs bosonic, clean vs disordered, …)

Non-linear response in strong THz fields: Third-Harmonic Generation (THG)



Linear response: *weak* perturbation, one measures the **optical-active** excitations of the system

$$J^L \sim Z\,\chi\,E$$



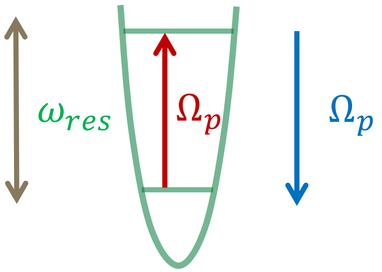
Non-linear response: *strong* perturbation, one can access **Raman-like** excitations of the system

$$J^{NL} \sim E\,R\,KE^2$$

Example: phonon mode Q coupled to light

$$H_{int} = -ZQ_{IR}E - RQ_RE^2$$

Non-linear response in strong THz fields: Third-Harmonic Generation (THG)



Enhanced when $\Omega_P \approx \omega_{res}$
$\chi(\omega)$ singular at ω_{res}

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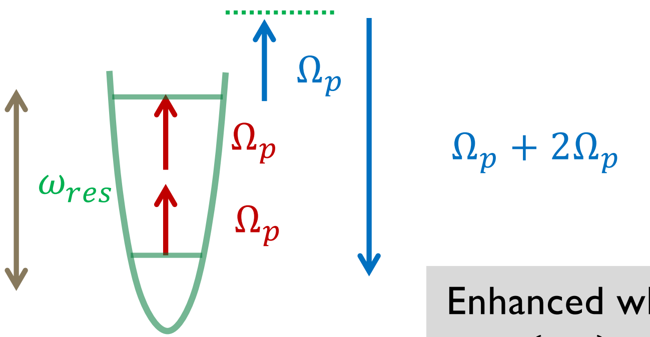


Enhanced when $2\Omega_P \approx \omega_{res}$
$K(2\omega)$ singular at ω_{res}

Non-linear response: *strong* perturbation, one can access **Raman-like** excitations of the system

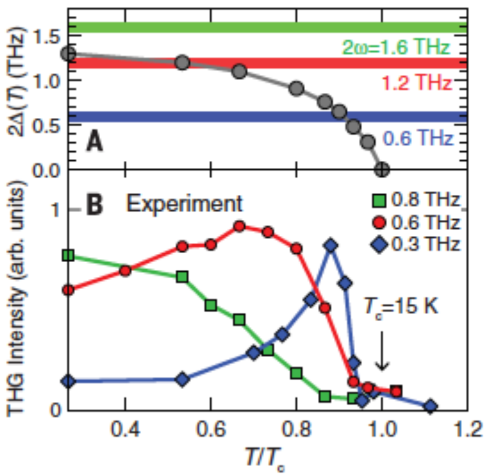
$$J^{NL} \sim E\, R\, K E^2$$

Example: phonon mode Q coupled to light

$$H_{int} = -ZQ_{IR}E - RQ_R E^2$$

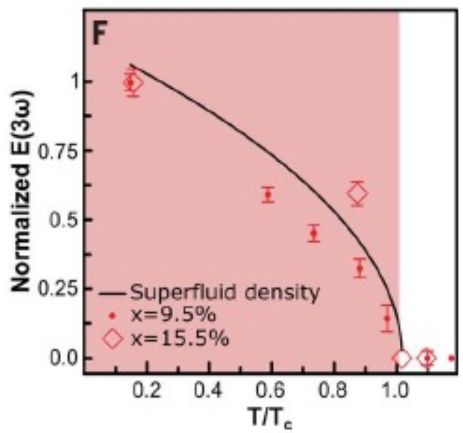
Superconductors show large THG

- Several experiments in conventional superconductors, like NbN and MgB2, and unconventional superconductors, like cuprates and pnictides



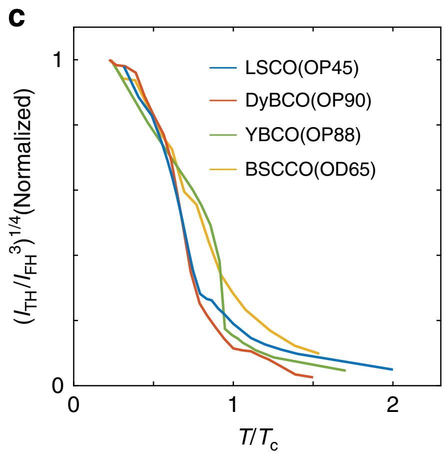
R. Matsunaga et al.,
Science 345, 1145 (2014)

NbN



S. Rajasekaran et al.
Science 359, 575 (2018)

$La_{2-x}Ba_xSrCuO_4$
z-axis polarization



H. Chu et al.
Nat. Comm. 11, 1793 (2020)

Cuprates
CuO_2 plane polarization

Superconductors show large THG

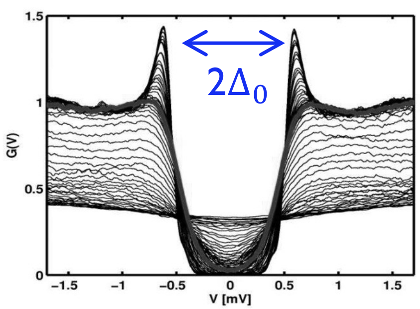
- Several experiments in conventional superconductors, like NbN and MgB_2, and unconventional superconductors, like cuprates and pnictides

- Below Tc the quasiparticle spectrum changes due to gap opening, new collective modes emerge, connected to the amplitude (Higgs) and phase (Plasmon) of the order parameter

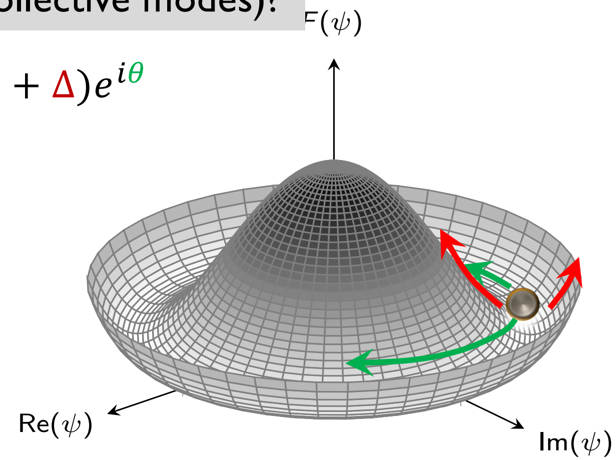
Why non-linearity should be so strong in superconductors?

Can we really separate fermionic effects (quasiparticles) from bosonic ones (collective modes)?

$$\langle c_\uparrow c_\downarrow \rangle = \psi = (\Delta_0 + \Delta) e^{i\theta}$$



$$E_k = \sqrt{\xi_k^2 + \Delta_0^2}$$



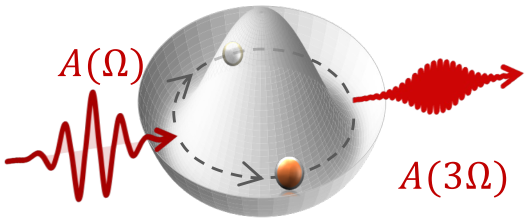
Non linearity of Josephson plasmons

- In a superconductor the e.m. field couples to the phase degrees of fredom

- *Interacting* phase-only model: a non-linear coupling to light emerges naturally

$A \to 2eA/c$ $\quad H[\theta] = -J_s \sum_j \cos(\theta_{j+1} - \theta_j) \Rightarrow H[\varphi, A] = -J_s \sum_j \cos(\varphi_j - A)$ $\quad \varphi_j = \theta_{j+1} - \theta_j$

Effective coarse-grained model on the scale $\sim\xi_0$



- Currents depends non-linearly on the gauge-invariant phase

$$J = J_s \sin(\varphi - A)$$

- Linear response: usual superfluid behavior

$$J^L = -J_s A \qquad J_s = \frac{(\hbar/2e)^2}{L_K}$$

- Non-linear response: contribution of exchange of phase fluctuations, i.e. plasmons

$$J^{NL} \sim A J_s^2 \langle \varphi^2 \varphi^2 \rangle A^2 = A K A^2$$

N.B. Here the *same* coupling J_s controls both linear and non-linear response

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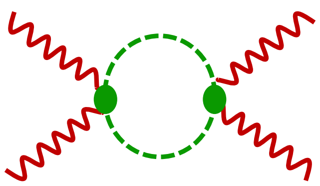
- One needs to add quantum effects: for a simple non-dispersive plasmon

$$S[\varphi, A] = \dot{\varphi}^2 - \omega_J^2 \varphi^2 - J_s \varphi^2 A^2 \qquad \omega_J^2 = 4\pi e^2 J_s$$

$$S^{(4)}[A] = J_s^2 A^2 \langle \varphi^2 \varphi^2 \rangle A^2$$

$$J^{NL} \equiv -\frac{\partial S^{(4)}}{\partial A} \sim A J_s^2 \langle \varphi^2 \varphi^2 \rangle A^2 = AKA^2$$



$$K(\omega) \propto \frac{J_s^2}{\omega_J} \frac{\coth(\beta \omega_J)}{4\omega_J^2 - \omega^2} \qquad I^{THG} \propto |K(2\Omega_p)|^2$$

Kernel resonant at $2\boldsymbol{\omega_J}$ gives enhanced THG at $\boldsymbol{\Omega_p} \approx \boldsymbol{\omega_J}$

Non linearity of Josphson plasmons

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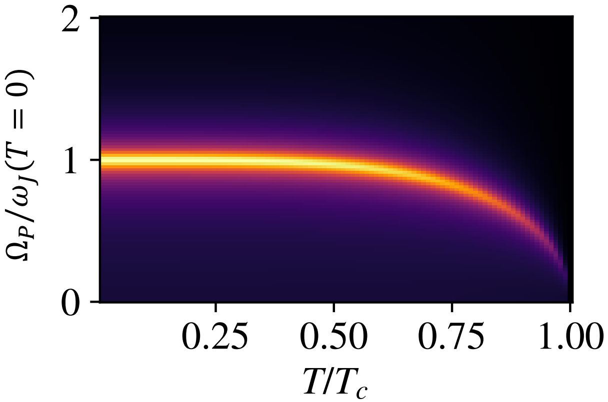
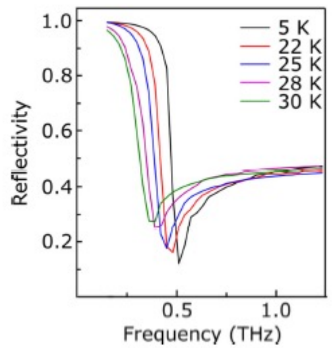
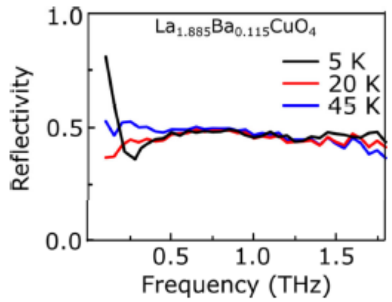
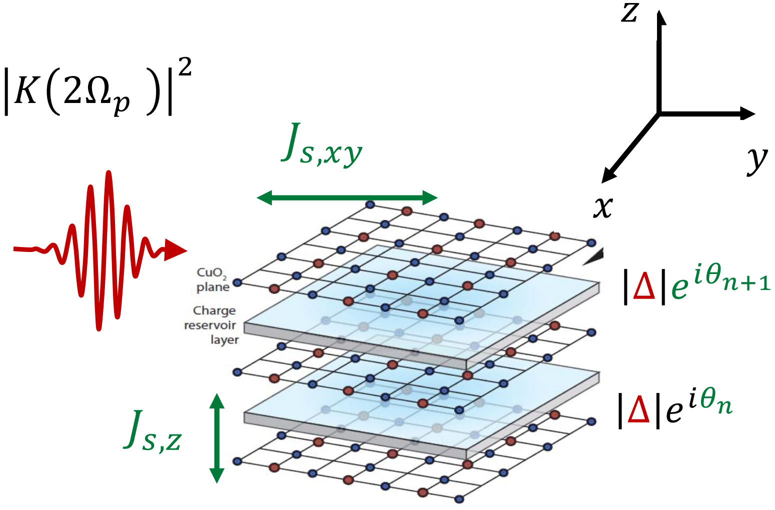




$\omega_{J,z}^2(T) = 4\pi e^2 J_{s,z}(T)$





La-based single-layer superconductors

$\omega_{J,z}^2 = 4\pi e^2 J_{s,z}$

Soft out-of-plane plasmon

F. Gabriele, M. Udina and L. B., Nat. Comm. 12, 752 (2021)
J. Fiore et al, preprint 2023

Non linearity of Josphson plasmons

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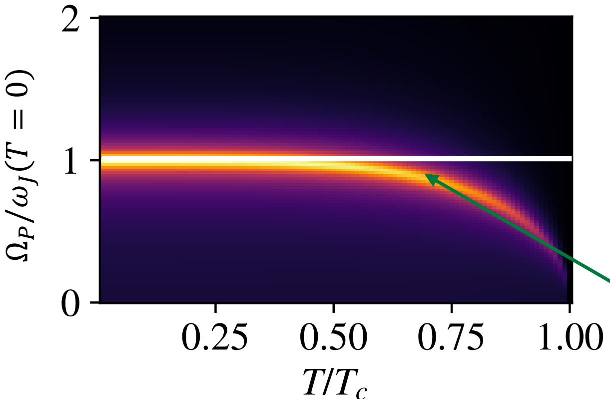

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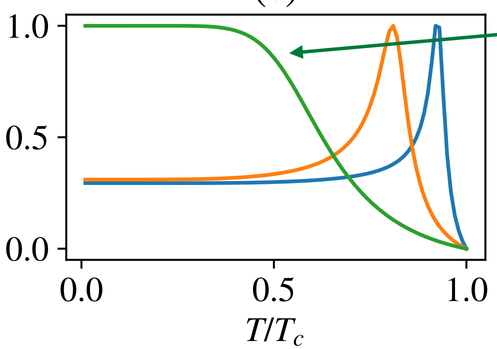

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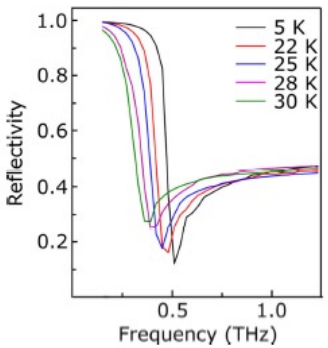
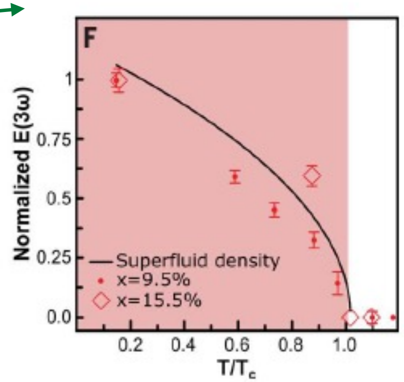
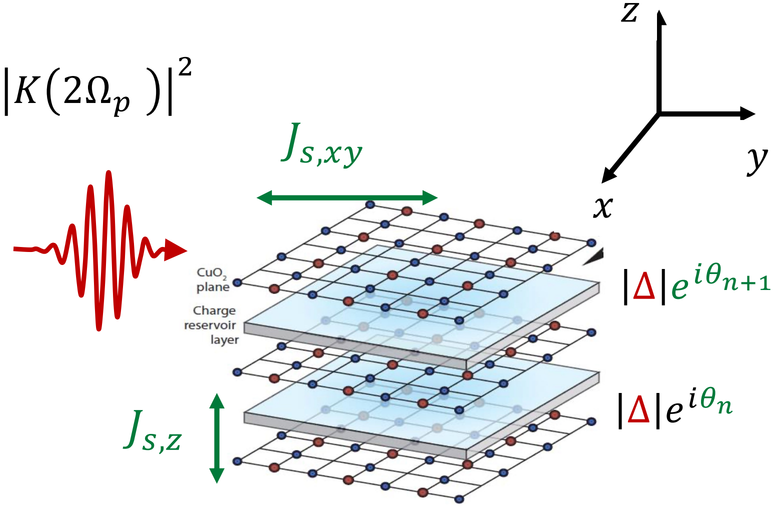




S. Rajasekaran et al.
Science 359, 575 (2018)



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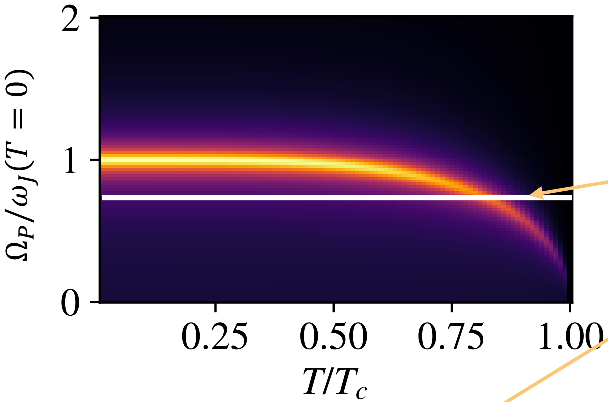

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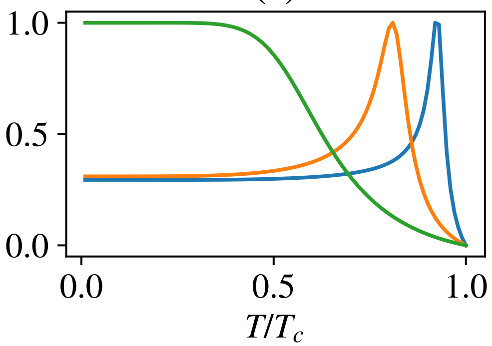

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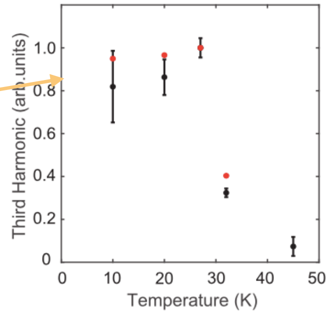
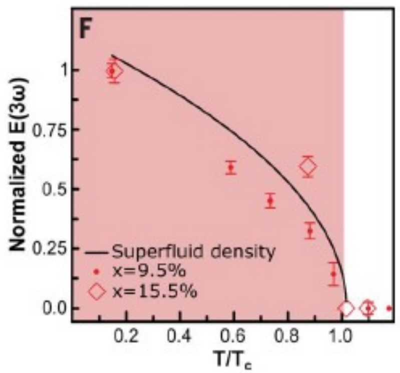


K. Kai et al., PRB 107, L140504 (2023)



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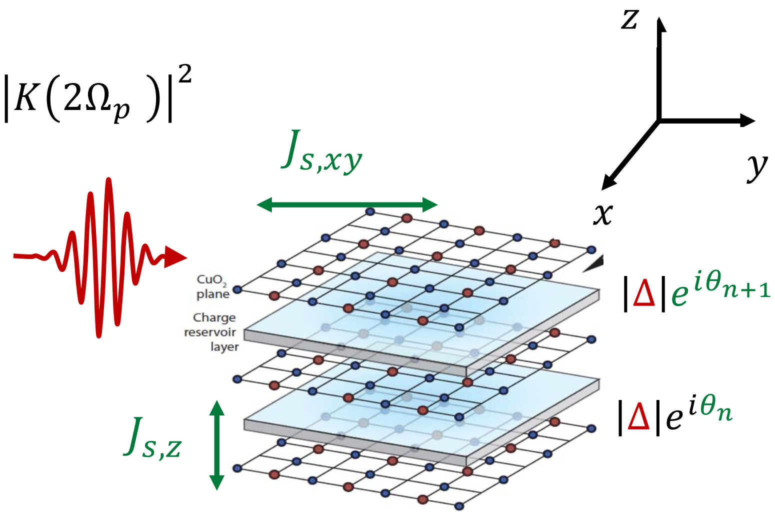

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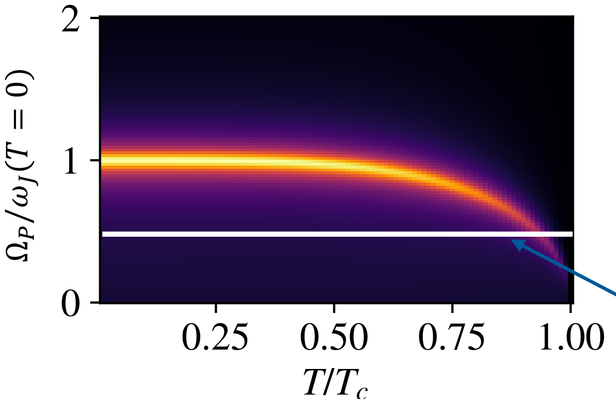
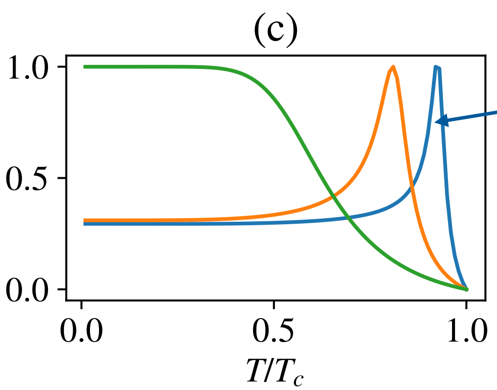
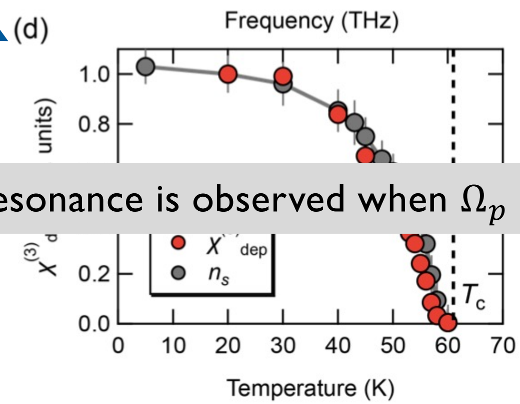
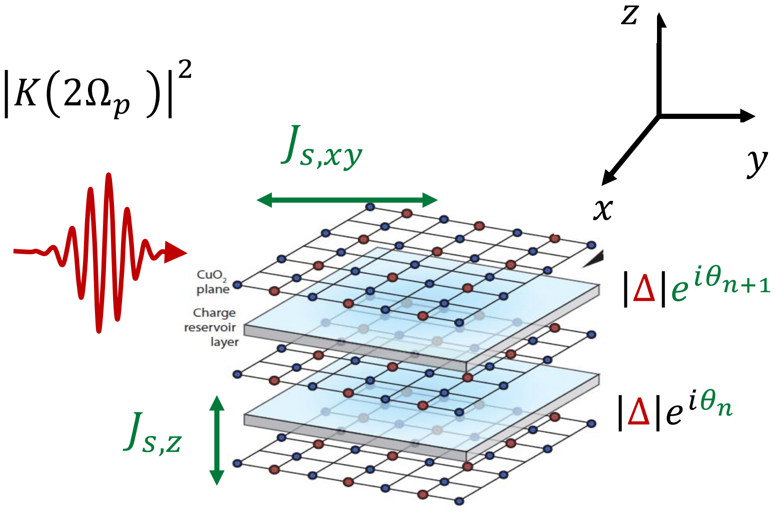








Y-based two-layers superconductor

No resonance is observed when $\Omega_p = \omega_{J,z}(T)$

$\omega_{J,z}^2 = 4\pi e^2 J_{s,z}$

Soft out-of-plane plasmon

F. Gabriele, M. Udina and L. B., Nat. Comm. 12, 752 (2

J. Fiore et al, preprint 2023

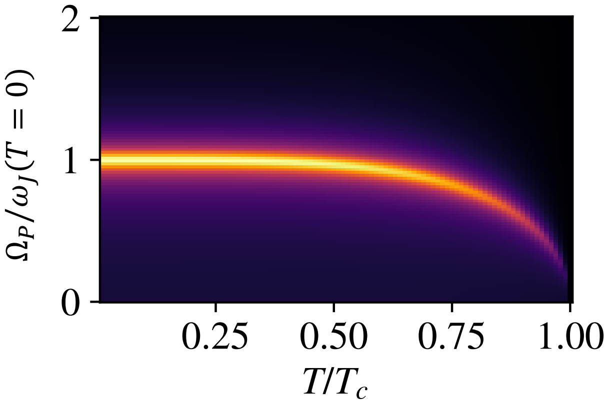
Why the result should change with the number of layers?

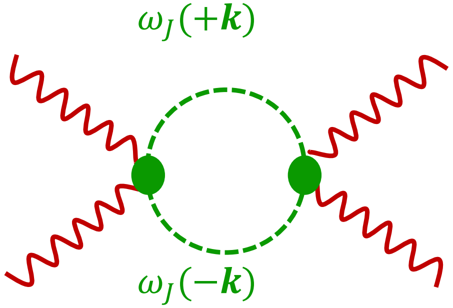
Non linearity of Josphson plasmons

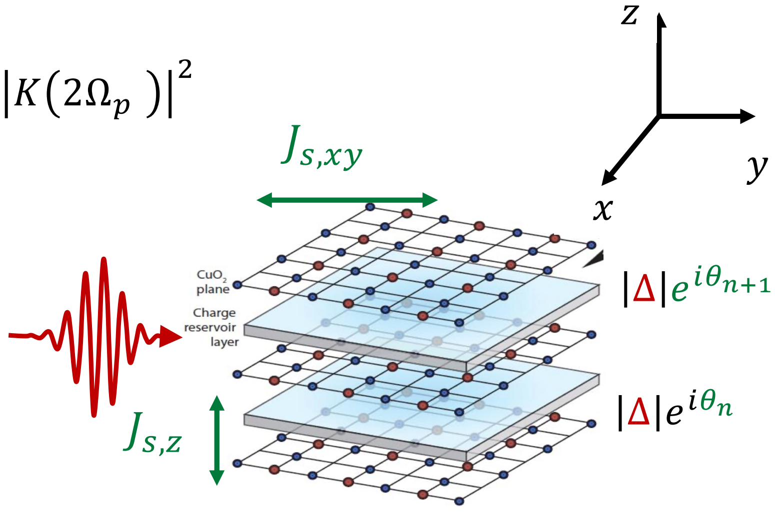
$J^{NL} \sim A J_s^2 \langle \varphi^2 \varphi^2 \rangle A^2 = A K A^2$

$$K(\omega) \propto \frac{J_s^2}{\omega_J} \frac{\coth(\beta\omega_J)}{4\omega_J^2 - \omega^2}$$

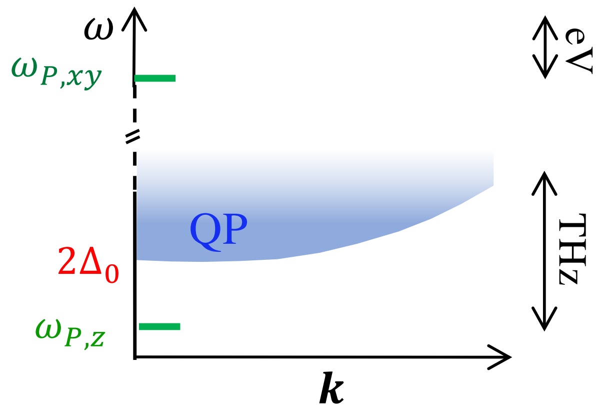
$I^{THG} \propto |K(2\Omega_p)|^2$



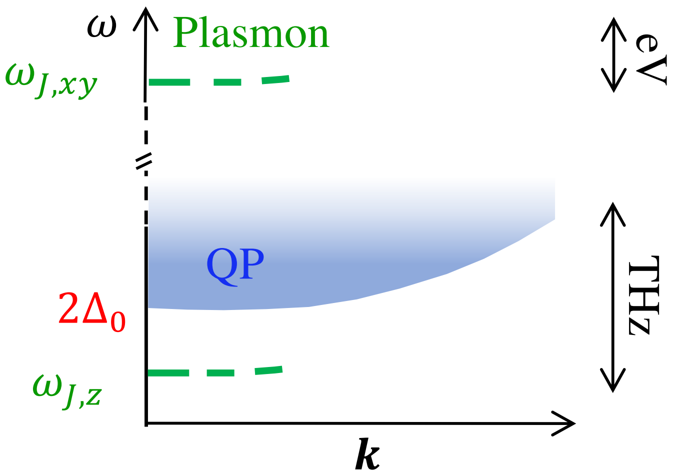




We have in mind excitation of two plasmons with opposite and **arbitrary momenta k**
We should account for **dispersion** of plasmons in a layered system, where the soft out-of-plane plasmon mixes with the large in-plane one



Plasma waves in *layered* superconductors



$$\omega_{J,xy}^2 = 4\pi e^2 J_{s,xy} \qquad k_z = 0, k_{xy} \to 0$$

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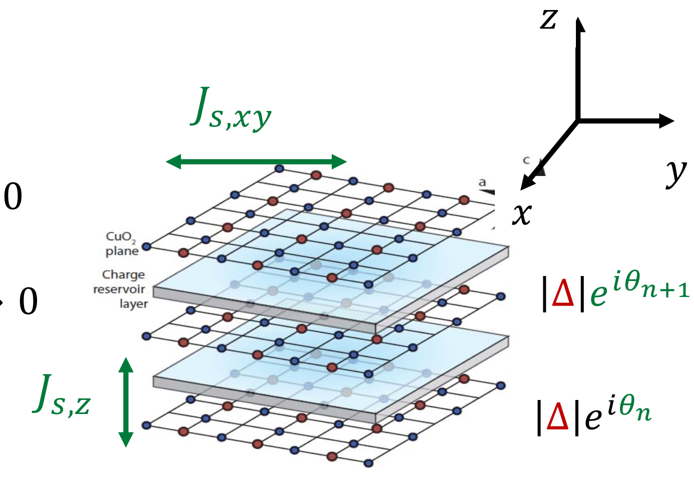
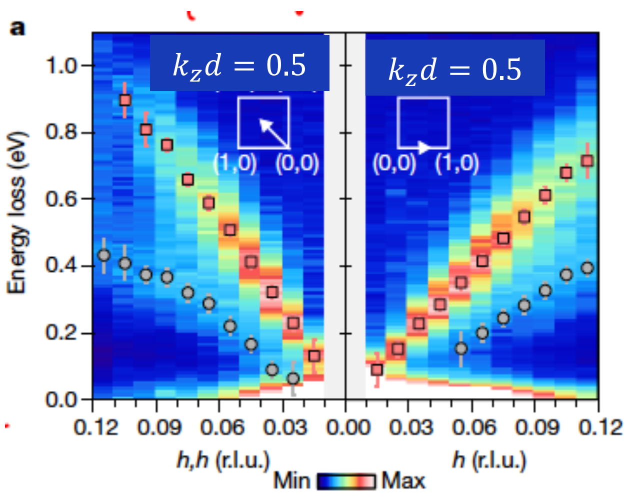


Stiffness anisotropy reflects in plasma-waves anisotropy

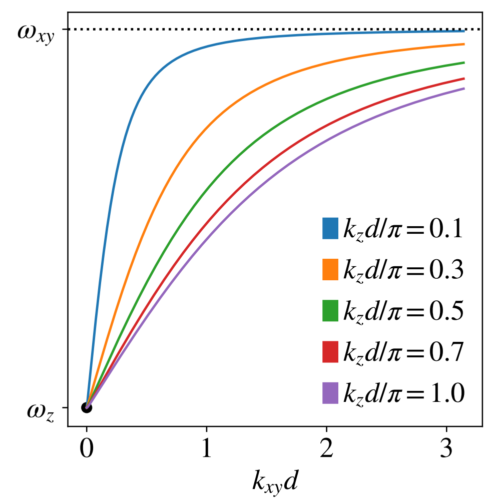


M. Hepting et al., Nature 563, 374 (2018)

$$\omega_L^2 = \omega_{J,xy}^2 \frac{k_{xy}^2}{k^2} + \omega_{J,z}^2 \frac{k_z^2}{k^2}$$

N.B. This expression fails as $k \to 0$ ...

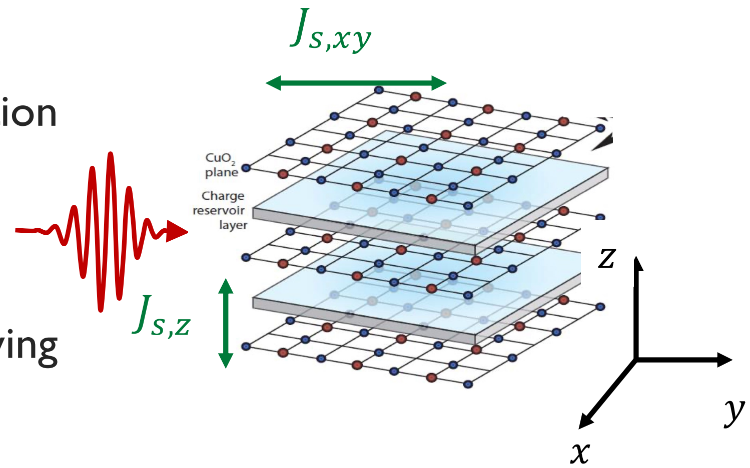
F. Gabriele, C. Castellani an L.B PRR 4, 023112 (2022)
F. Gabriele, C. Castellani and L.B., preprint 2023



Plasmon dispersion and polarization effects

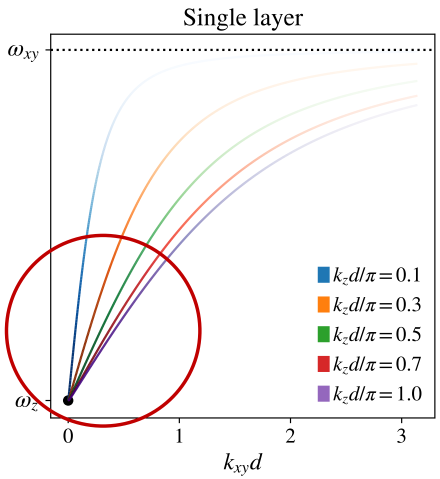
- Light polarized along c couples to phase gradient along the same direction

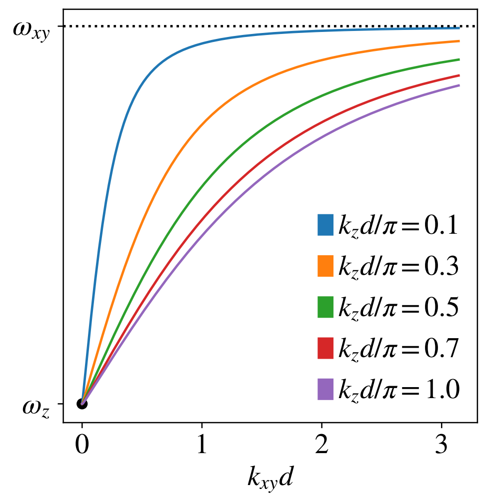
$$H[\theta] = -\sum_{xy,z} J_{s,z}\cos(\theta_{r+z} - \theta_r - A_{ext}) + J_{s,xy}\cos(\theta_{r+xy} - \theta_r)$$



- Polarization projects out the anisotropic 3D dispersion at small k_z, leaving most of the spectral weight around ω_z

$$K(\omega) \sim J_z^2 \sum_{\boldsymbol{k}} \frac{k_z^4}{|\boldsymbol{k}|^4} \frac{\coth(\beta\omega_L(\boldsymbol{k}))}{\omega_L(\boldsymbol{k})(4\omega_L^2(\boldsymbol{k}) - (\omega + i\gamma)^2)}$$





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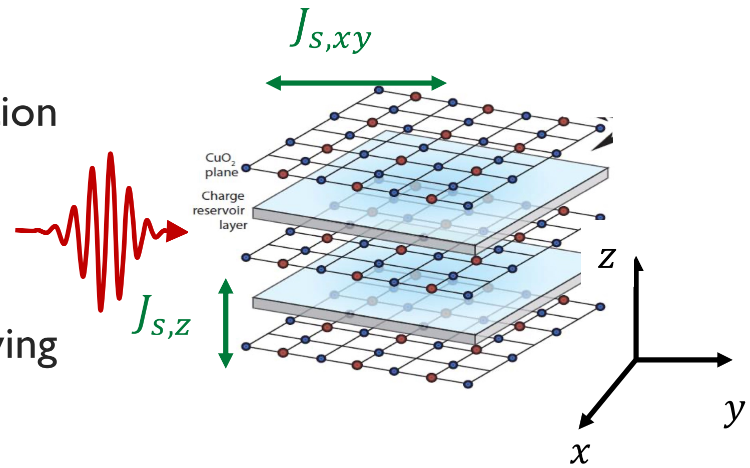

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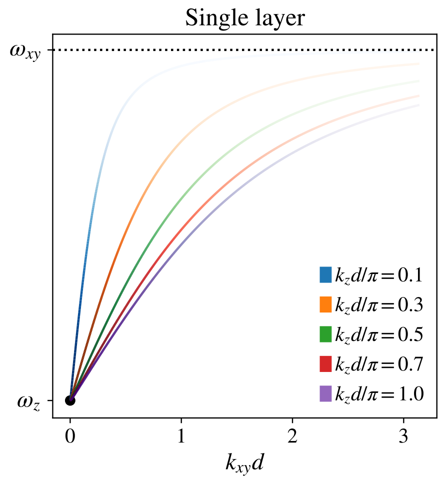
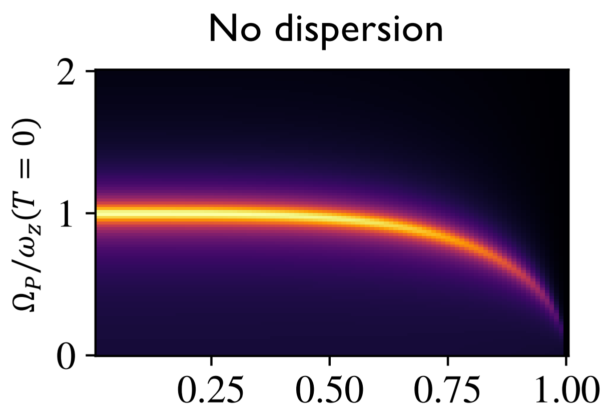
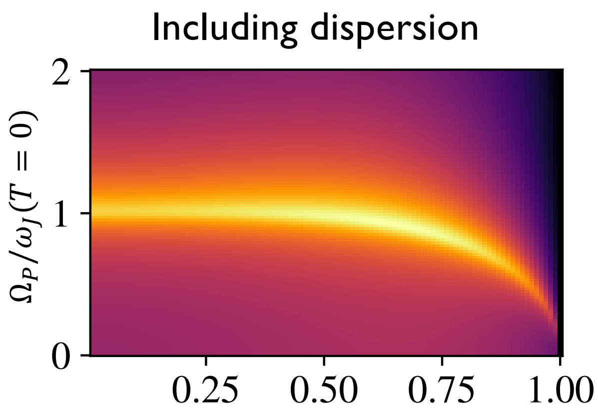


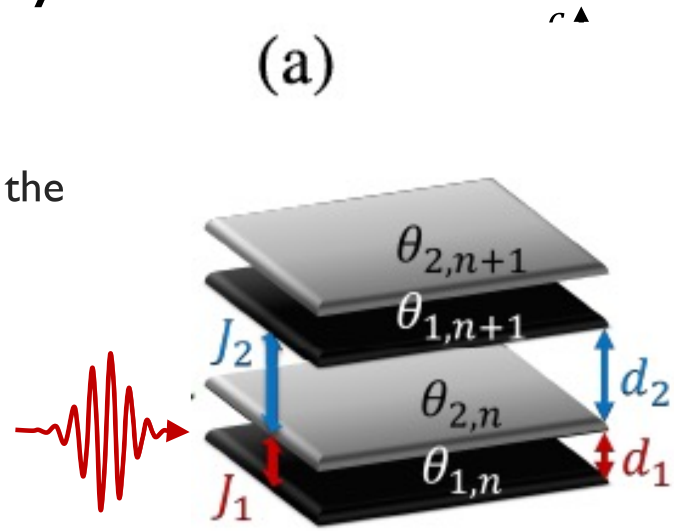


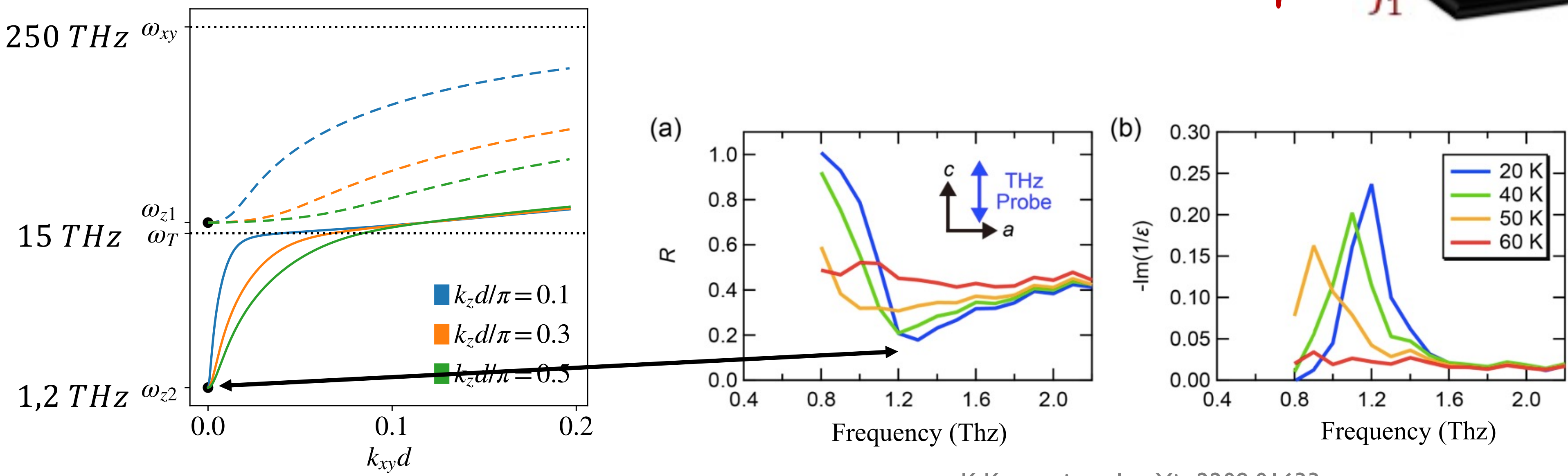


In the single-layer case the response remains peaked at ω_z

Plasmon dispersion and polarization effects: bilayer case

- In the bilayer case two Josephson plasmons emerge
- The reflectivity edge at q=0 and the superfluid stiffess are dominated by the lower one ω_{z2}

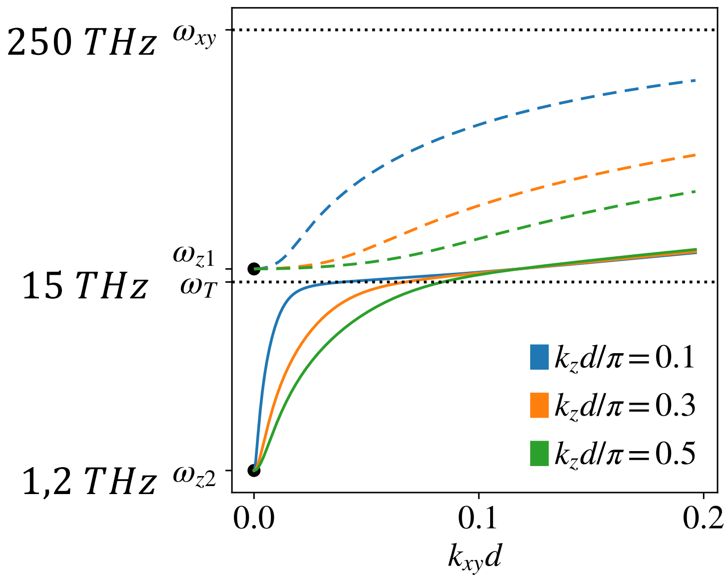


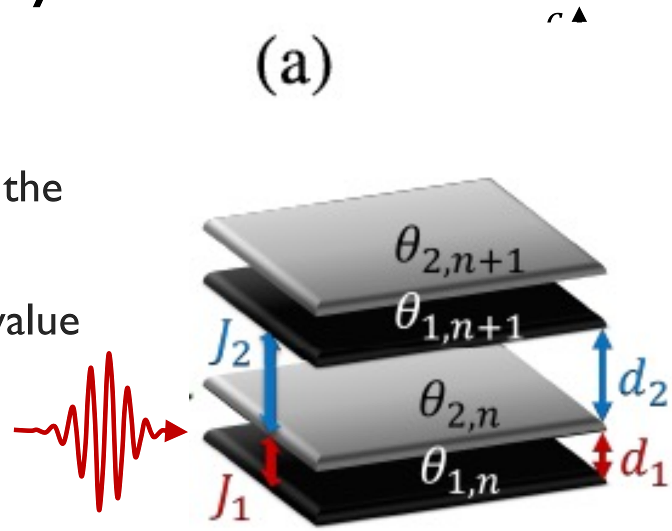


K.Katsumi et al. arXiv:2209.01633

Plasmon dispersion and polarization effects: bilayer case

- In the bilayer case two Josephson plasmons emerge
- The reflectivity edge at q=0 and the superfluid stiffess are dominated by the lower one ω_{z2}
- The dispersion moves the energy of the soft plasmon towards a larger value ω_T as momentum increases





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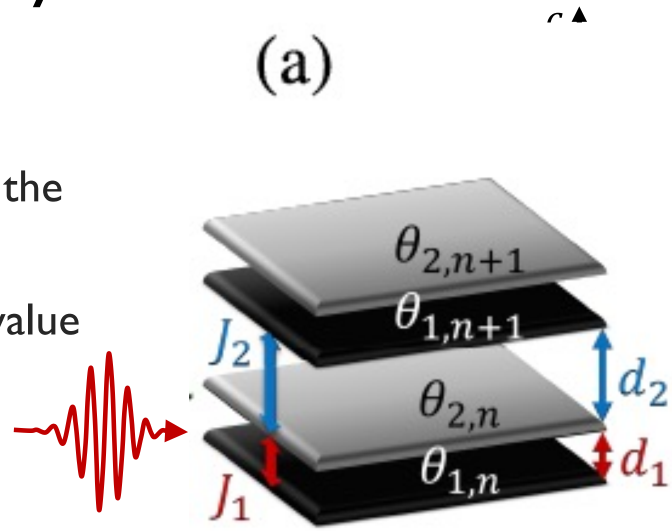

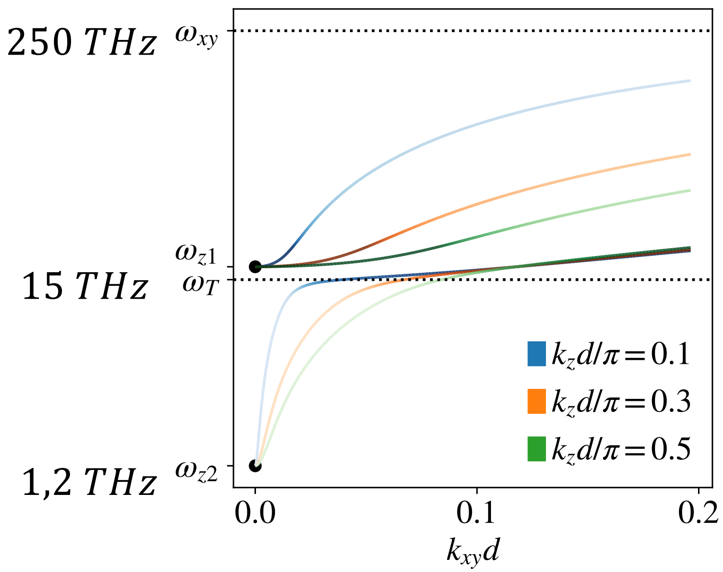

$$K_{\pm}(\Omega) = \sum_{\sigma\sigma',\mathbf{k}} W_{\sigma\sigma'} \frac{\omega_\sigma \pm \omega_{\sigma'}}{4\omega_\sigma \omega_{\sigma'}} \frac{\coth\left(\frac{\beta\omega_\sigma}{2}\right) \pm \coth\left(\frac{\beta\omega_{\sigma'}}{2}\right)}{(\omega_\sigma \pm \omega_{\sigma'})^2 - (\Omega + i\delta)^2},$$

Projection due to polarization of the incoming light moves the spectral weight towards ω_T

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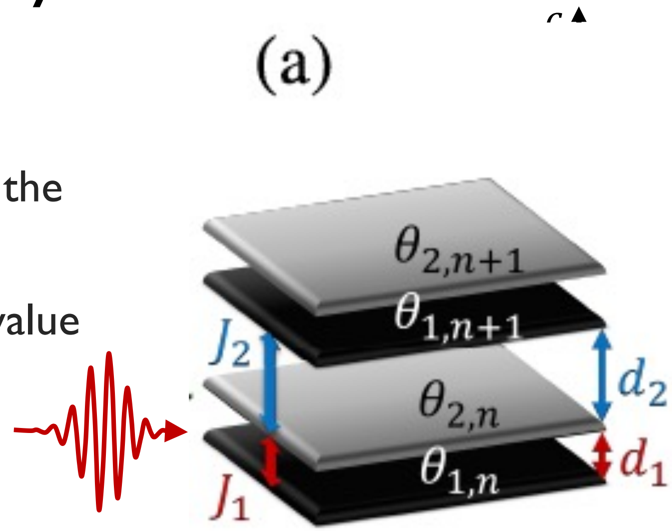

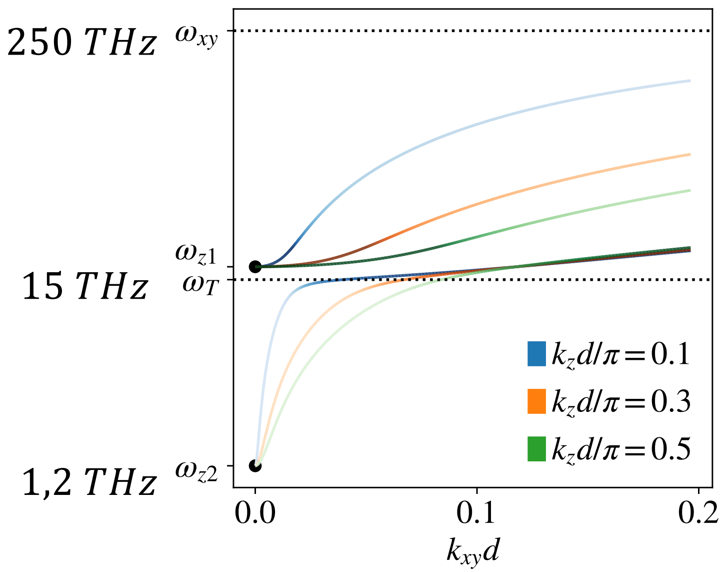

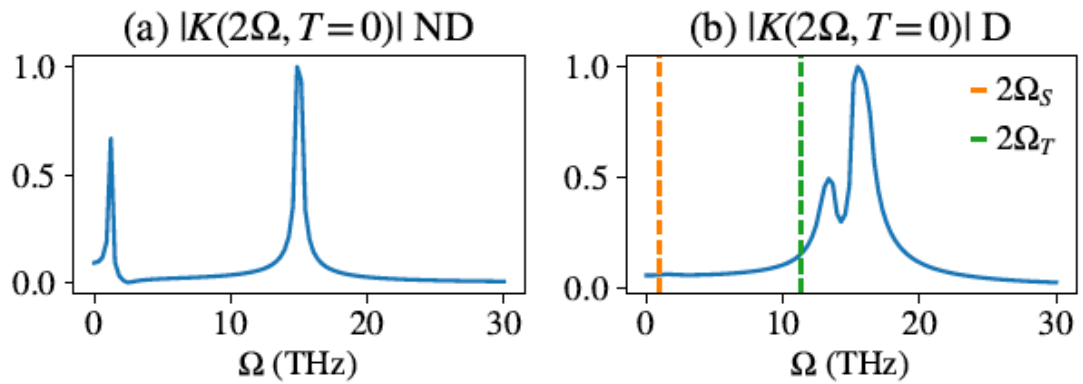

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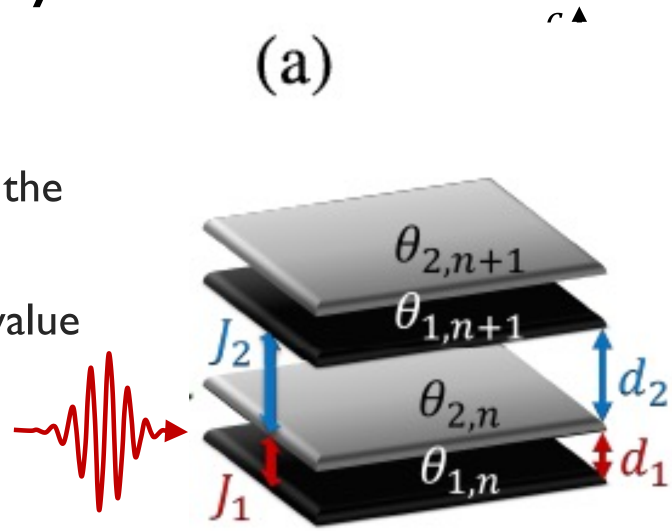
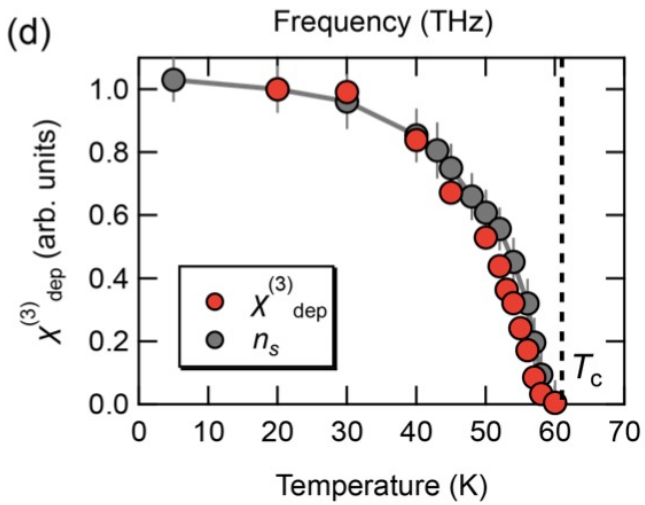
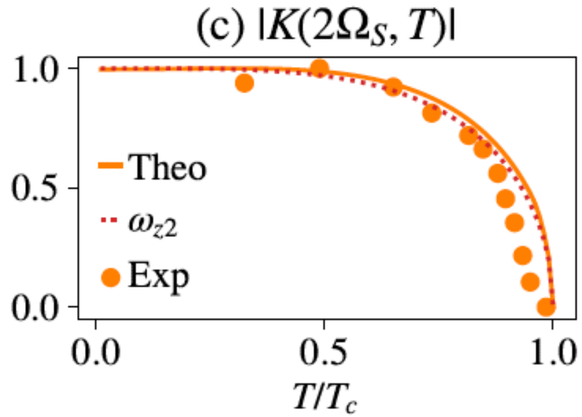




K.Katsumi et al. arXiv:2209.01633

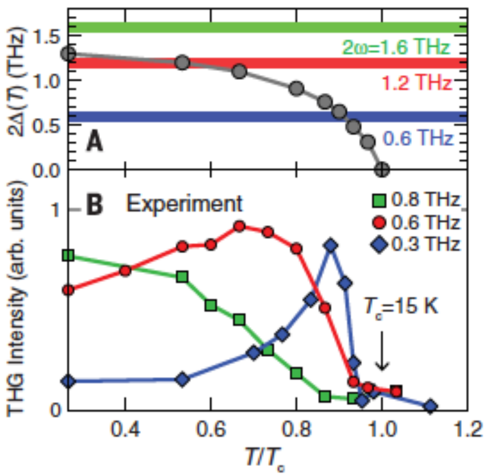


J. Fiore et al, preprint 2023

No resonance is observed when $\Omega_p = \omega_{z2}(T)$

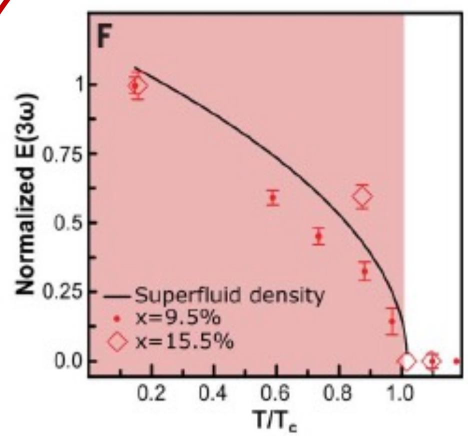
Superconductors show large THG

- Several experiments in conventional superconductors, like NbN and MgB2, and unconventional superconductors, like cuprates and pnictides



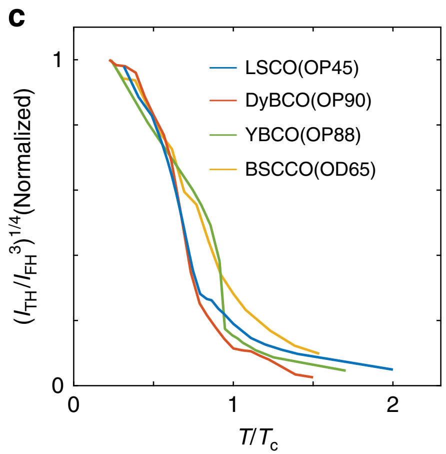
R. Matsunaga et al., Science 345, 1145 (2014)

NbN



S. Rajasekaran et al. Science 359, 575 (2018)

$La_{2-x}Ba_xSrCuO_4$
z-axis polarization



H. Chu et al. Nat. Comm. 11, 1793 (2020)

Cuprates
CuO_2 plane polarization

Non linearity of Josephson plasmons within *microscopic* models

$$H[\theta] = -J_s \sum_j \cos(\theta_{j+1} - \theta_j) \qquad J = J_s \sin(\varphi - A)$$

There is a single parameter J_s

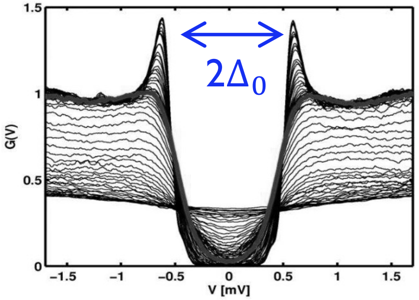
At microscopic level: gauge field couples to electrons. One has bare response of quasiparticles, and effective coupling functions for collective excitations

$$H(\vec{A}) = \sum_k \frac{1}{2m}\left(\vec{k} - \frac{e}{c}\vec{A}\right)^2 c_k^+ c_k + H_{int}$$

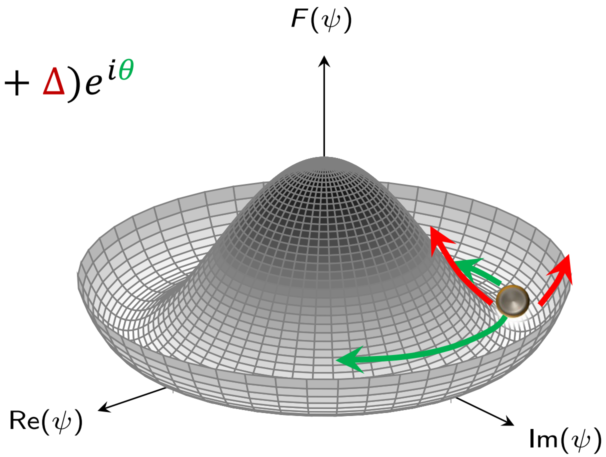
$$J^{NL} \sim AKA^2 \quad K = K_{BCS} + K_{Higgs} + K_{plasmon}$$

T.. Cea, C. Castellani and L.B.. PRB 93, 180507(R) (2016)
M. Udina, T. Cea and L.B. PRB 100, 165131 (2019)

$$E_k = \sqrt{\xi_k^2 + \Delta_0^2}$$



$$\langle c_\uparrow c_\downarrow \rangle = \psi = (\Delta_0 + \Delta)e^{i\theta}$$



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Even *weak disorder* makes BCS response very strong

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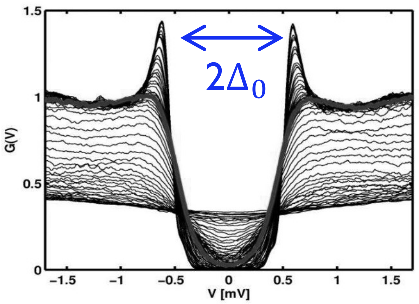

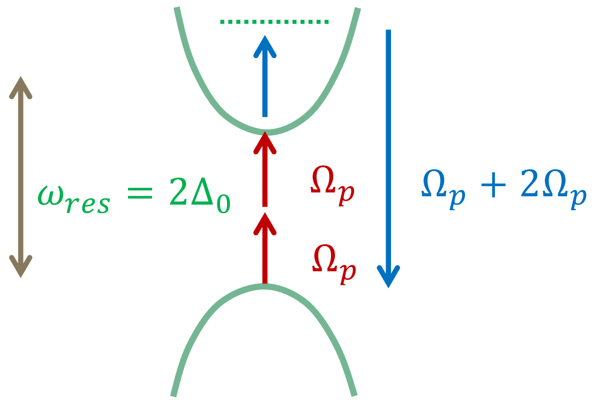

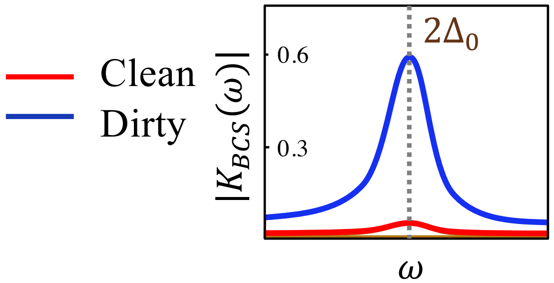

M. Silaev, PRB 99, 224511 (2019)

Y. Murotani and R. Shimano, PRB 97, 094516 (2019)

N.Tsuji and N. Nomura, PRR2, 043029 (2020)

G. Seibold, L.B. et al. PRB 103, 014512 (2021)

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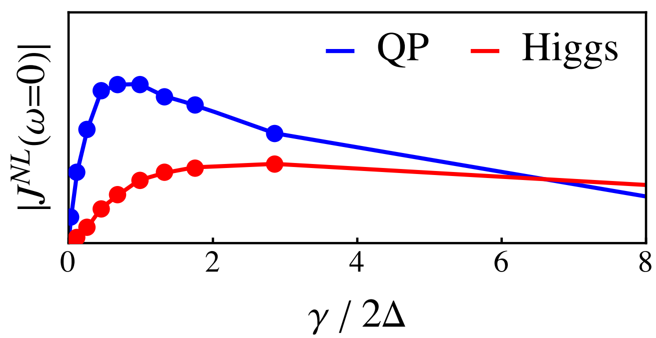
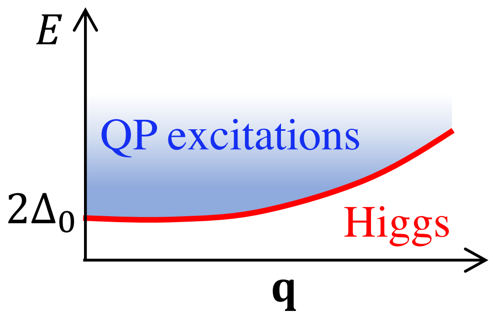
Strong disorder can also increase the Higgs response mediated by electrons (but still difficult to distinguish since they occur at the same energy…)





M. Silaev, PRB 99, 224511 (2019)
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We can expect that the ***effective coupling*** of two-plasmon excitations depends on disorder, temperature, frequency, etc.

$$H[\theta] \sim \frac{J_s}{2}(\nabla\theta)^2 + \boldsymbol{J_{eff}}(\mathbf{T}/\boldsymbol{\Delta},\ \boldsymbol{\Gamma}/\boldsymbol{\Delta},\ \boldsymbol{\omega}/\Delta)(\nabla\theta)^4 + \cdots$$

Conventional NbN at very large disorder, near SIT

$$H[\theta] = -J_s \sum_j \cos(\theta_{j+1} - \theta_j)$$

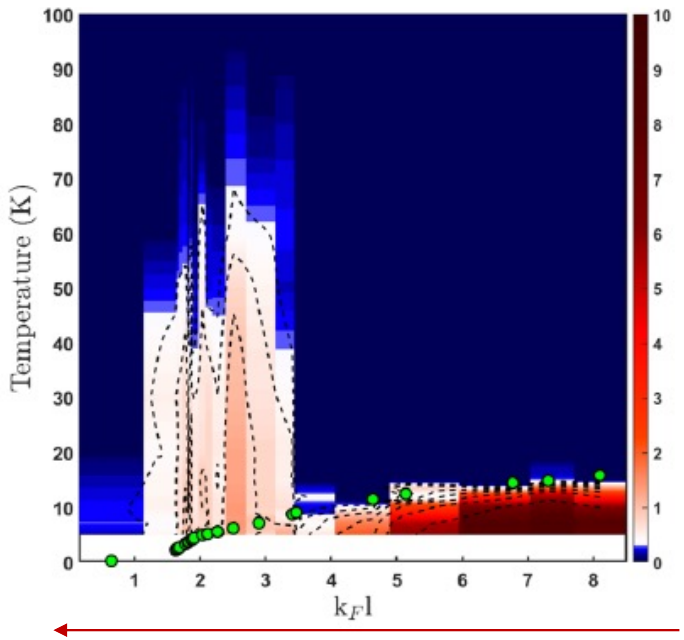
$$J = J_s \sin(\varphi - A)$$

There is a single parameter $J_s(T)$
When $J_s(T)$ goes to zero one should not see any THG

Non-linear response survives in a wide range of T above T_c, spectral range moves well above 2Δ

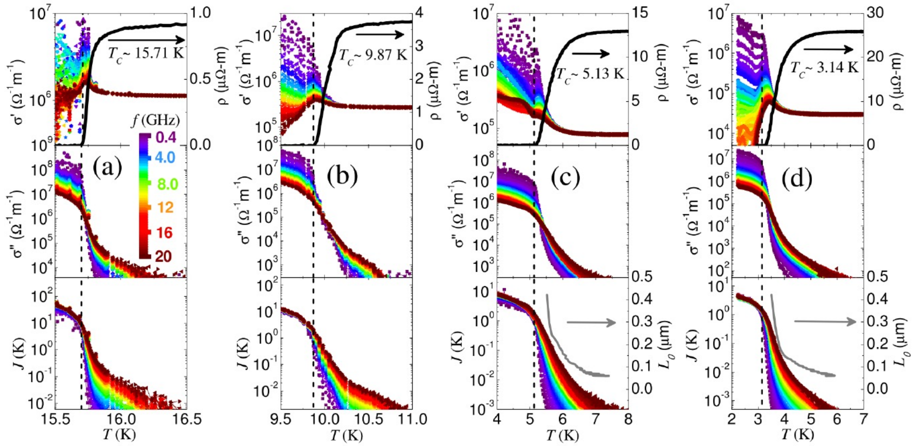
NbN samples

D. Chaudhuri et al. arXiv:2204.04203



Increasing disorder

Conventional NbN at very large disorder, near SIT



Increasing disorder

M. Mondal, L.B. … P. raychaudhuri, Sci. Rep. 3, 1357 (2013)

$$H[\theta] \sim \frac{J_s}{2}(\nabla\theta)^2 + J_{eff}(\mathrm{T}/\Delta,\ \Gamma/\Delta, \omega/\Delta)(\nabla\theta)^4 + \cdots$$

$$I^{THG}(T) \sim J_{eff}(\mathrm{T}/\Delta,\ \Gamma/\Delta, \Omega_P/\Delta)$$

THz pump frequency Ω_P much larger than T_c
The fluctuating stiffness can remain large above T_c

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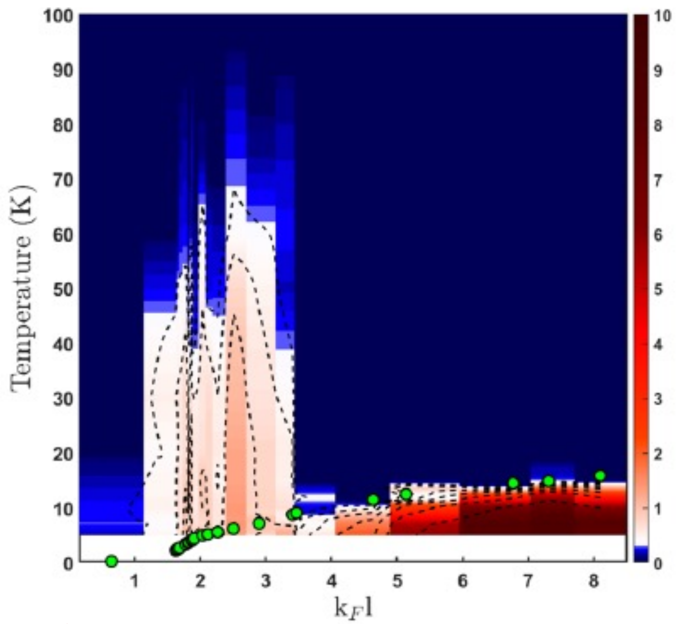

Increasing disorder

Conclusions

Effective Josephson models seems to reproduce well features connected to soft out-of-plane plasmons

Why non-linearity should be so strong in superconductors?

Can we really separate fermionic effects (quasiparticles) from bosonic ones (collective modes)?

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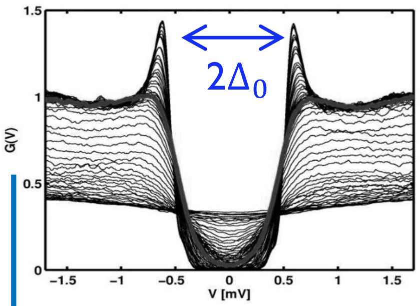
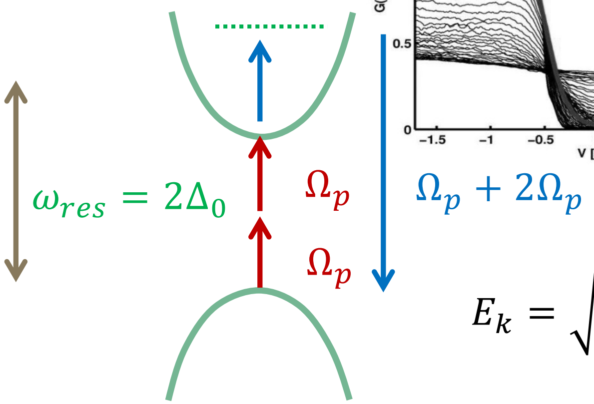
Modification of the *fermionic* spectrum makes available excitations at q=0 in the Thz range





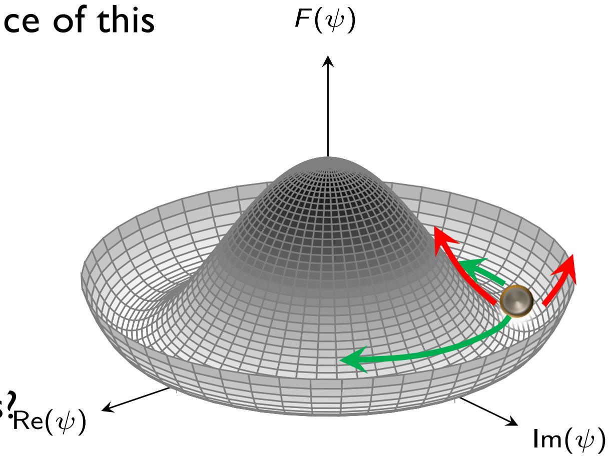
$$E_k = \sqrt{\xi_k^2 + \Delta_0^2}$$

This effect manifests *at all orders* in the response to the applied e.m. fiels

$$\langle c_\uparrow c_\downarrow \rangle = \psi = (\Delta_0 + \Delta) e^{i\theta}$$

Non-linear plasmons are a consequence of this

$$H[\theta] = -J_s \sum_j \cos(\theta_{j+1} - \theta_j)$$



What about effective Josephson models for real junctions?