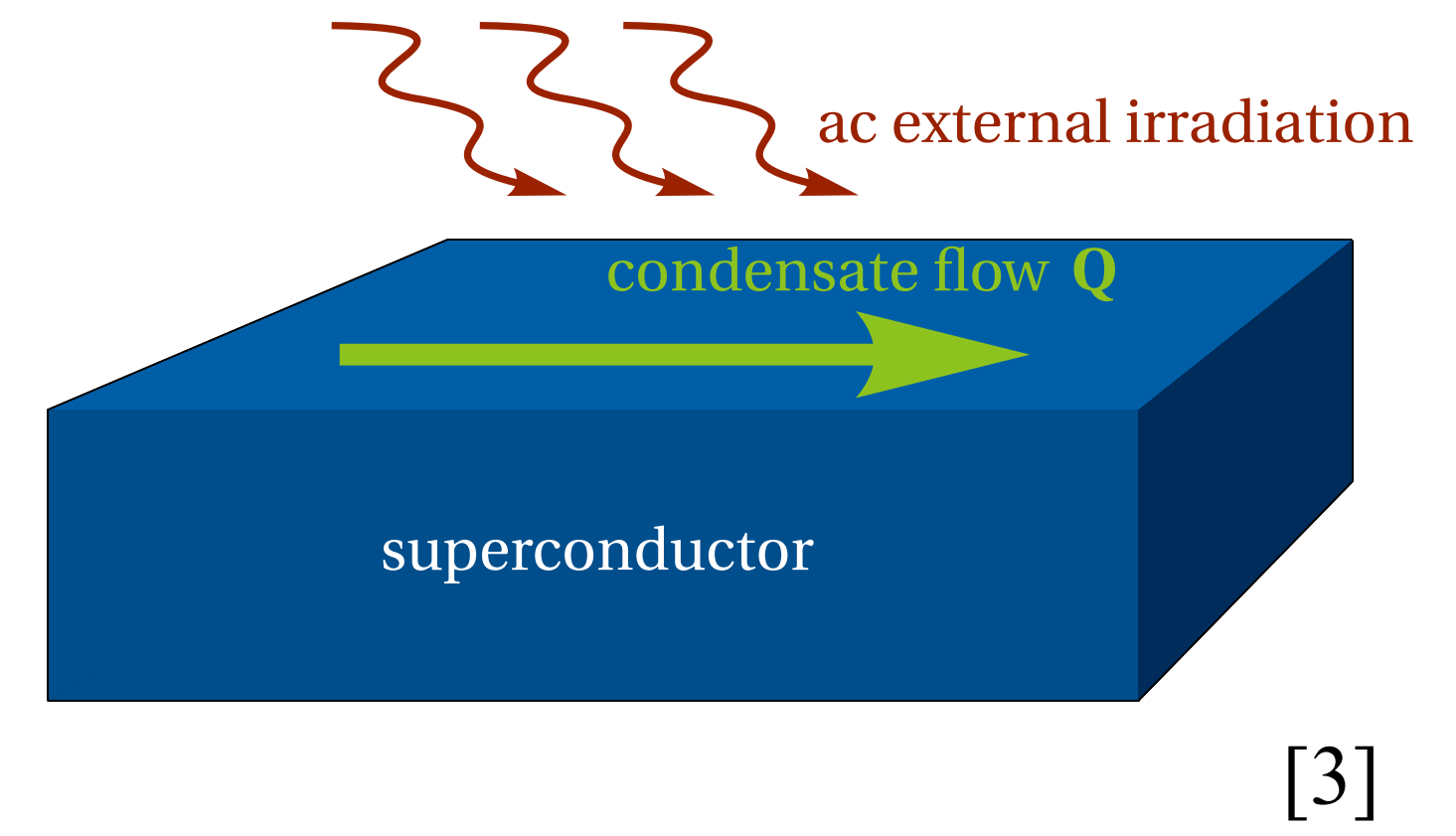


Introduction

We develop a theory of the nonlinear optical responses in superconducting systems in the presence of a dc supercurrent. The optical transitions between particle-hole pair bands across the superconducting gap are allowed in clean superconductors as the inversion-symmetry-breaking by supercurrent. Vertex correction is included in optical conductivity to maintain the $U(1)$ gauge symmetry in the mean-field formalism. We show two pronounced current dependent peaks in the second-order nonlinear optical conductivity $\sigma^{(2)}(\omega)$ at $2\hbar\omega = 2\Delta$ and $\hbar\omega = 2\Delta$. Our theory predicts the current induced peak in $\text{Im}[\sigma^{(2)}(\omega)]$ is with the same order of magnitude as the recent experimental observation of second-harmonic generation in NbN[2]. The supercurrent induced nonlinear optical spectroscopy provides a valuable toolbox to explore novel superconductors.



[3]

Formalism

Self-consistent Hartree-Fock

The BCS mean field theory can be understood as a self-consistent Hartree-Fock (SCHF) approximation:

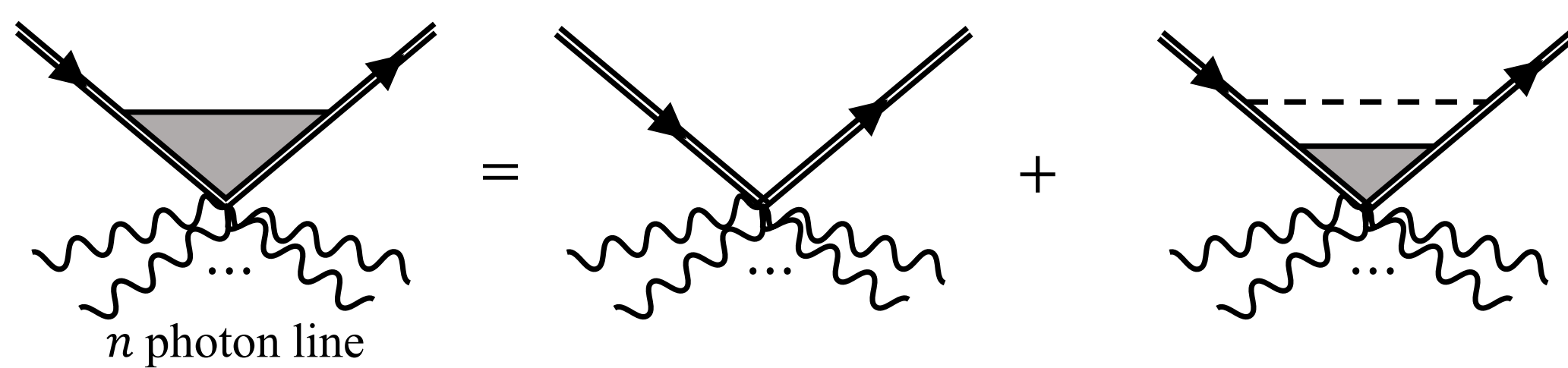
$$H_{int} = \sum_{k,k'} V_{k,k'} [\Psi_k^\dagger \tau_3 \Psi_{k'}] [\Psi_{k'}^\dagger \tau_3 \Psi_k]$$

$$\Sigma_F(k_0, \mathbf{k}) = \frac{1}{\beta} \sum_{k'_0} \int \frac{d^2 \mathbf{k}'}{(2\pi)^2} V_{k-k', k} \tau_3 G(k_0 - k'_0, \mathbf{k} - \mathbf{k}') \tau_3$$

$$\text{Diagrammatic representation of the self-consistent Hartree-Fock approximation showing a fermion line with a dashed loop representing the self-energy correction.$$

Vertex correction

The gauge invariance should be maintained when calculating optical responses. [4-6]



$$\Gamma(p+q, p) = \gamma(p+q, p) + \frac{1}{\beta} \sum_{k_0} \int \frac{d^2 \mathbf{k}}{(2\pi)^2} V_{k,p}$$

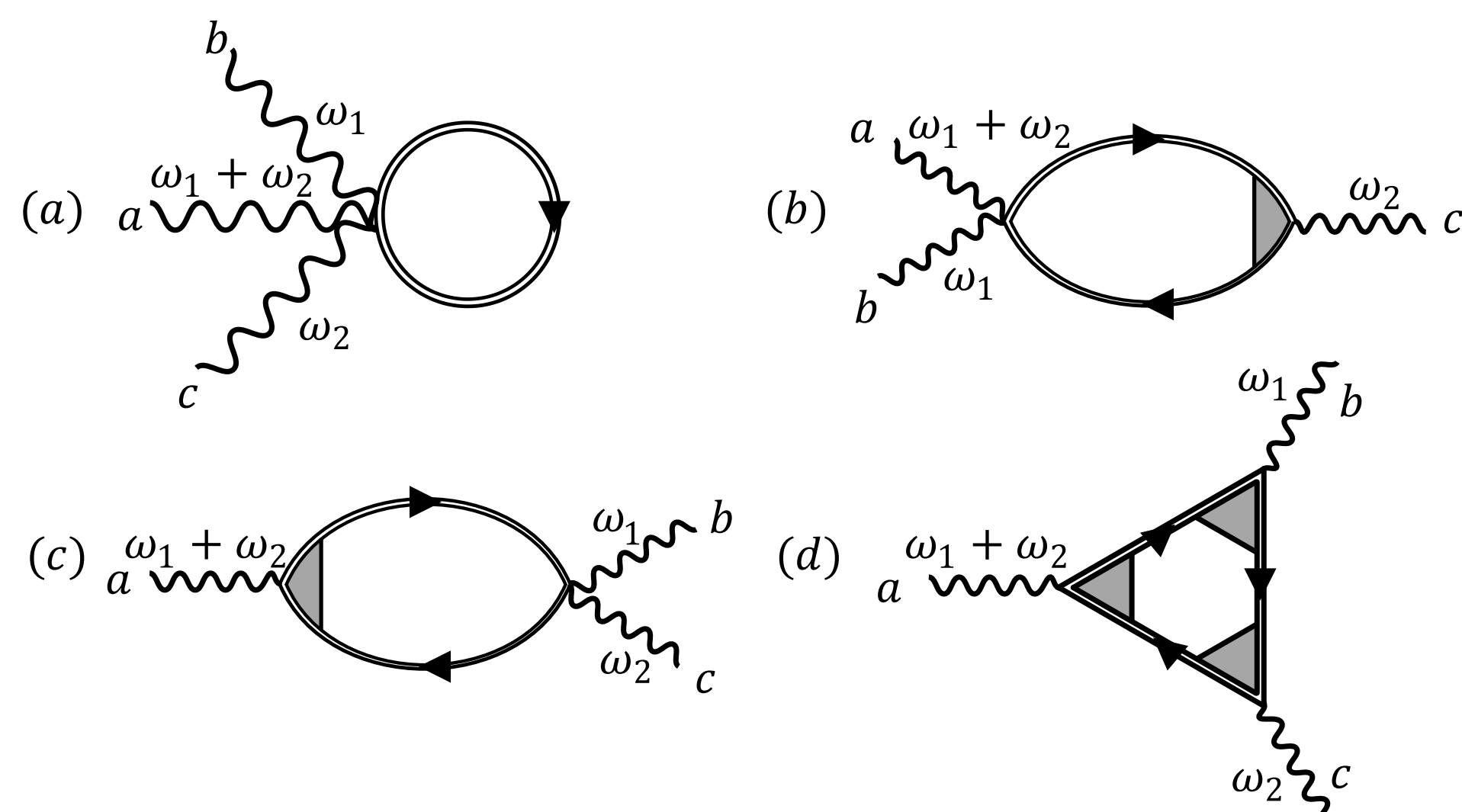
$$[\tau_3 G(k_0 + q_0, \mathbf{k} + \mathbf{q}) \Gamma(k+q, k) G(k_0, \mathbf{k}) \tau_3]$$

Gauge-invariant nonlinear response

Second-order optical conductivity:

$$j_a^{(2)} = \sigma_{abc}^{(2)}(\omega_1 + \omega_2, \omega_1, \omega_2) E_b(\omega_1) E_c(\omega_2)$$

Feynman diagrams[7]:



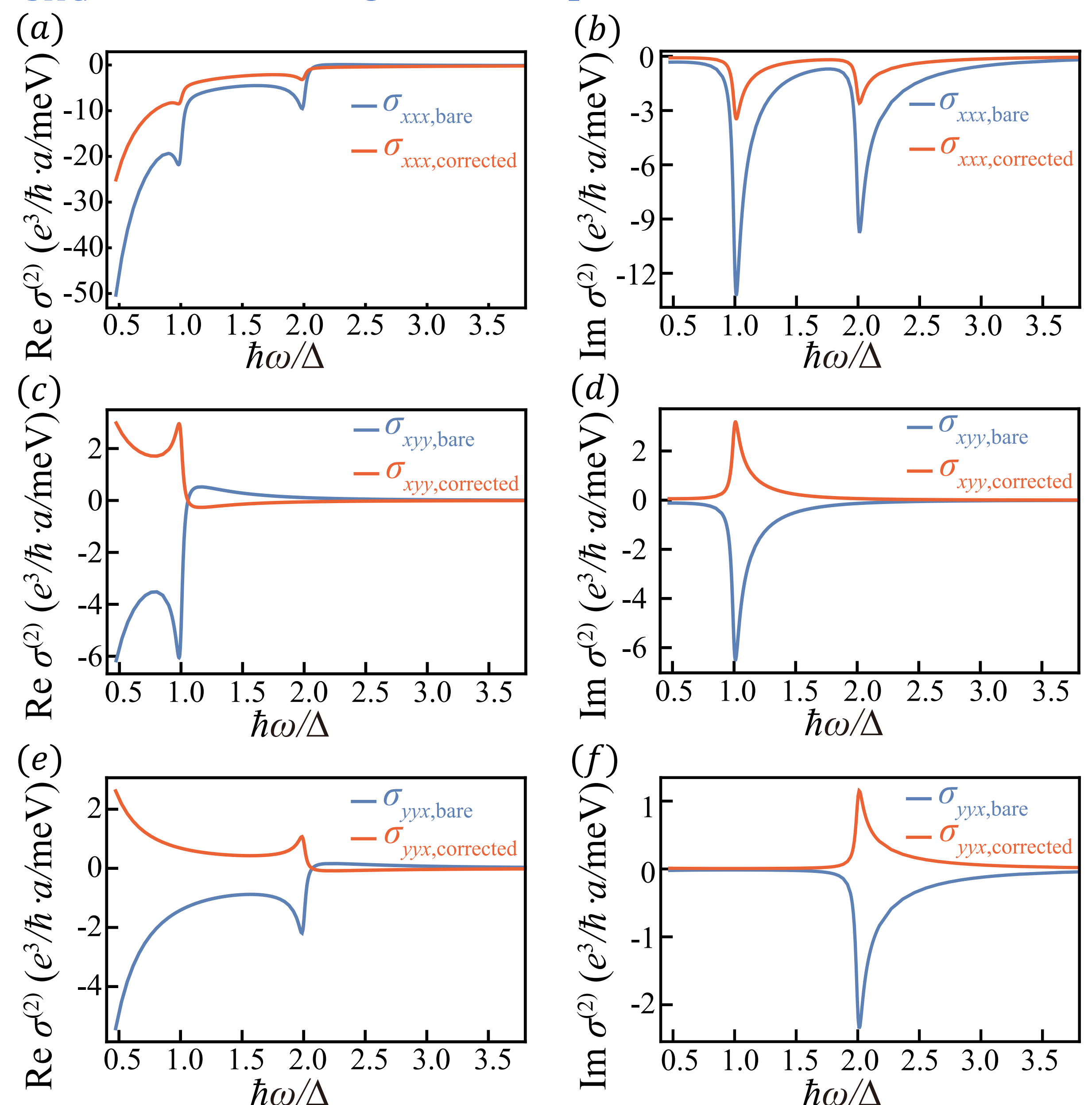
Two processes:

second-harmonic generation (SHG): $\sigma_{abc}^{(2)}(2\omega, \omega, \omega)$

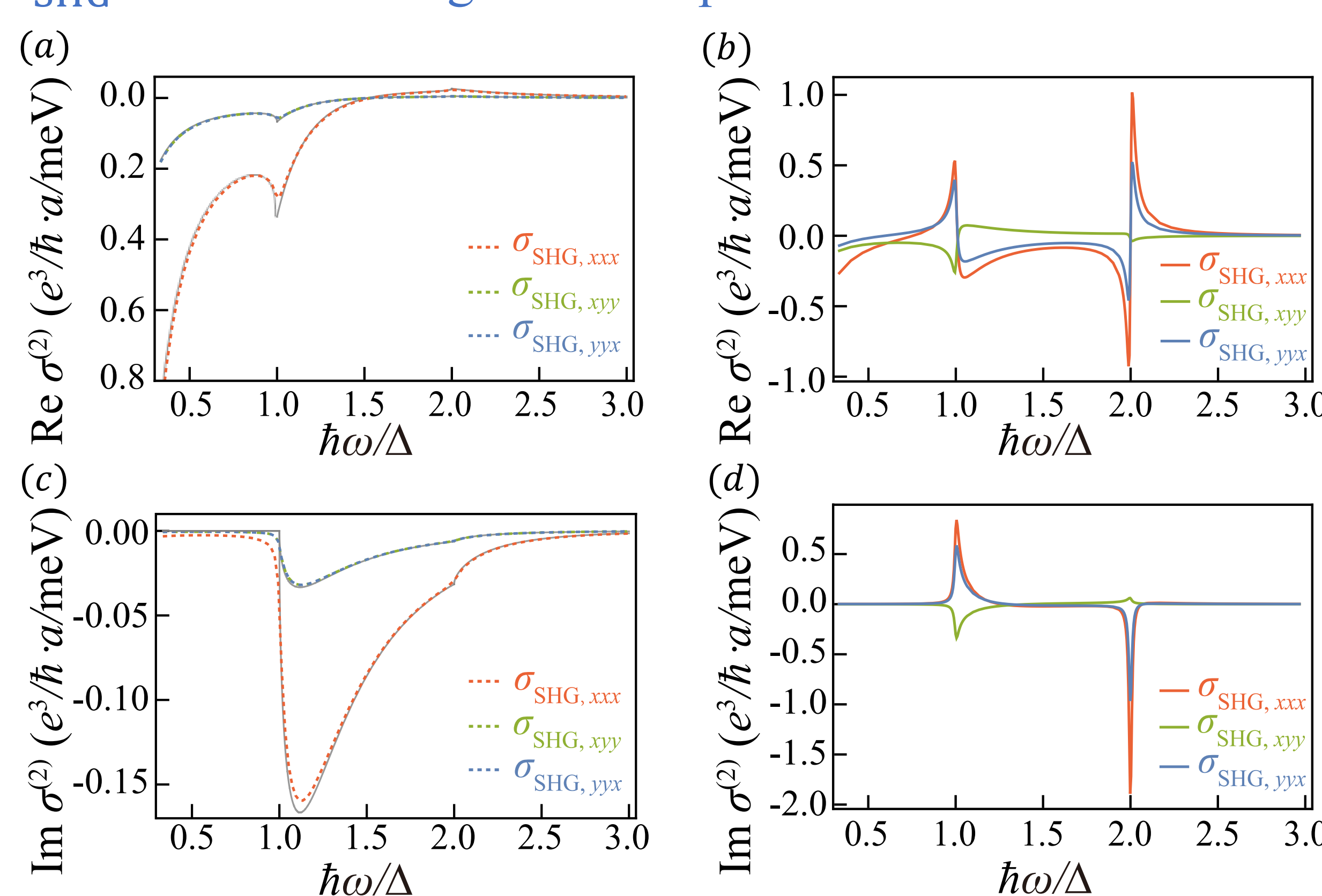
photocurrent effect (PC): $\sigma_{abc}^{(2)}(0, \omega, -\omega)$

Results

σ_{SHG} for s-wave single-band superconductor



σ_{SHG} for d-wave single-band superconductor



For more results (e.g. PC), see [1].

References

[1] Phys. Rev. B **108**, 224516 (2023).

[2] Phys. Rev. Lett. **125**, 097004 (2020).

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[4] Phys. Rev. **117**, 648–663 (1960).

[5] Phys. Rev. B **95**, 014506 (2017).

[6] Phys. Rev. B **106**, L220504 (2022).

[7] Phys. Rev. B **99**, 045121 (2019).