

Nonlinear microwave response of clean superconducting films

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We develop an explicitly gauge-invariant semiclassical approach to investigate the nonlinear response of superconductors to monochromatic terahertz radiation. We demonstrate that in clean superconductors charge conservation forbids nonlinear response to a uniform field. We apply our approach to quasi-two-dimensional films and obtain an explicit expression for the photoinduced current. We find that the photoinduced current exhibits a strong dependence on polarization and the incidence angle of the radiation. Our predictions may be directly verified in experiments with quasi-two-dimensional superconducting films.

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I. INTRODUCTION

Recently, the response of superconductors to an electromagnetic field received a surge of attention following the advances in nonlinear terahertz (THz) spectroscopy [1]. Sufficiently intensive and coherent THz radiation allows us to explore the nonlinear response in a characteristic range of frequencies of the order of the superconducting gap Δ_0 . Of particular interest are frequencies near $2\Delta_0$, where direct quasiparticle excitation becomes possible.

Superconductors are known to host a number of collective modes associated with variations of the complex superconducting order parameter $\Delta = |\Delta|e^{i\varphi}$ (see [2]). The phase degree of freedom φ is coupled to the electromagnetic field and is responsible for the Meissner effect, as well as for the phenomenon of dissipationless current. Associated with phase is the superconducting plasma mode whose properties vary significantly depending on the temperature and the effective dimensionality of the sample. This mode has been extensively studied [2–4] as it significantly affects the linear response of superconductors. Another mode, usually referred to as the Higgs mode or Schmid mode [5], is associated with variations of the absolute value of the order parameter and has a characteristic frequency $\omega = 2\Delta_0$. Unlike phase variations, in BCS superconductors this mode is decoupled from the electromagnetic field in the linear order, which has long prevented its direct observation.

In the last decade, the Higgs mode has received extensive attention in the context of experiments [6,7] in which excitation of this mode serves as a possible explanation of the observed features of response at characteristic frequency $2\Delta_0$. A significant number of theoretical works which use different techniques have since been dedicated to the problem of the nonlinear response of superconductors, especially to the third harmonic generation [8–11]. However, these works attribute the observed effects to different excitations: while some claim the Higgs mode produces the largest contribution [9,10], others find the contribution of density fluctuations to be dominant [8,11].

One of the challenges posed by the problem of nonlinear response is the gauge invariance. Within the standard diagram-

matic approach, obtaining gauge-invariant response kernels requires accuracy even in the first order [12]. In this paper, we aim to formulate a physically transparent approach in which gauge invariance manifests itself outright and is identically satisfied in all orders of perturbation theory. We also apply our approach to calculate photoinduced current in quasi-two-dimensional superconducting films.

The structure of this paper is as follows. In the following section we define the system under consideration. Then we specify the general formalism used in our analysis and formulate our approach. In the following sections we employ our approach to analyze the nonlinear response of a quasi-two-dimensional film to microwave radiation. Finally, in the last section we discuss our findings and compare them to the results of other authors.

II. SYSTEM UNDER CONSIDERATION

Below we will consider a system depicted in Fig. 1 – a large superconducting film of constant thickness d exposed to monochromatic radiation of frequency Ω . We will be interested in frequencies of order Δ_0 , which usually lie in the THz range. We assume that the effects of size quantization may be neglected at the energy scale of Δ_0 , which usually holds for samples with d of the order of several decades of nanometers or larger. Effectively, this condition ensures that the electron dynamics inside the film may be considered three-dimensional. On the other hand, the dynamics of the electric current and order parameter may be considered two-dimensional if d is smaller than the London penetration depth λ_L and the coherence length ξ , which usually are about 100 nm to a few hundred nanometers. In this limit magnetic field can freely penetrate the film, and due to the large wavelength of radiation at these frequencies, all perturbations may be set constant along the transverse coordinate. However, dependence on coordinates in the plane of the film is allowed. The in-plane coordinate dependence of the electromagnetic field is controlled by the incidence angle θ and is periodic with the wave vector $q = \Omega \cos \theta / c$. We will also assume our

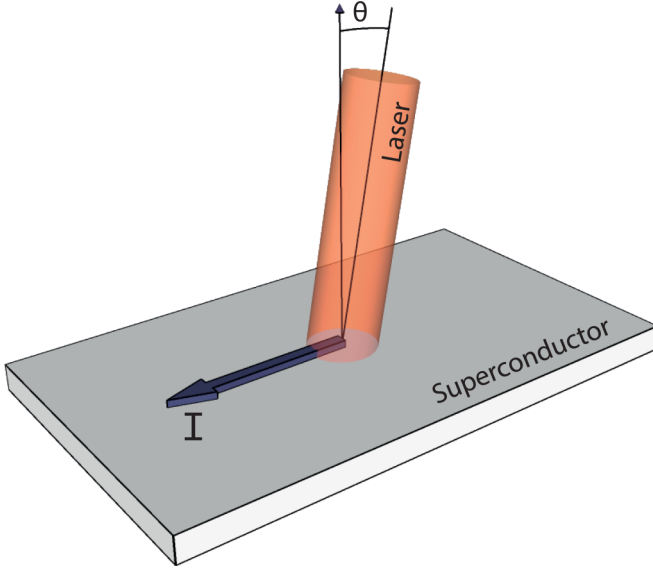


FIG. 1. Schematic depiction of the system under consideration. A large quasi-two-dimensional superconducting film subject to a monochromatic electromagnetic plane wave falling at an incidence angle θ .

system remains in thermodynamic equilibrium at a temperature T below and not too close to the critical temperature T_c .

III. GENERAL FORMALISM

The dynamics of an interacting BCS superconductor is described in terms of the complex superconducting order parameter field $\Delta = (\Delta_0 + \delta\Delta)e^{i\varphi}$ and electromagnetic field introduced via scalar and vector potentials V and A . In order to highlight gauge invariance, we perform a standard unitary transformation [13,14] which leaves the order parameter real while incorporating its phase into electromagnetic potentials, introducing gauge-invariant fields $\Phi = V + \dot{\varphi}/2e$ and $\mathcal{A} = A - c\nabla\varphi/2e$.

For our problem, we chose to use the Keldysh technique [15]. Therefore, for every field X we introduce components X_F and X_B living on the forward and backward branches of the time contour, respectively. For convenience, we also perform Keldysh rotation to classical and quantum components $X_{cl,q} = (X_F \pm X_B)/2$. In order to somewhat compactify our notation, we also introduce matrices $\check{X} = \tau_0 X_{cl} + \tau_1 X_q$, where $\tau_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$ and $\tau_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ act in Keldysh space.

Within the Keldysh technique, the dynamics of a BCS superconductor interacting with electromagnetic field is described by means of an effective action [15]

$$iS = iS_{\text{EM}} + iS_{\Delta} + \text{tr} \ln \check{G}^{-1}, \quad (1)$$

where

$$\begin{aligned} S_{\text{EM}} = & 4 \int \frac{d\omega}{2\pi} \frac{d^3k}{(2\pi)^3} \left\{ [A_{cl}(\omega, k) - A_0(\omega, k)] \right. \\ & \times \frac{\omega^2/c^2 - k^2}{8\pi} A_q(-\omega, -k) + [V_{cl}(\omega, k) - V_0(\omega, k)] \\ & \times \left. \frac{k^2}{8\pi} V_q(-\omega, -k) \right\} \end{aligned} \quad (2)$$

is the action of the free electromagnetic field both inside and outside of the superconductor, with A_0 and V_0 describing the applied (laser) field. We assume the vector potential satisfies the Coulomb gauge condition $\nabla A = 0$. The next part,

$$S_{\Delta} = -\frac{4}{g} \int dx dt \Delta_{cl}(x, t) \Delta_q(x, t), \quad (3)$$

is the Hubbard-Stratonovich term for the order parameter, with g describing the strength of the attractive BCS interaction. The last term, $\text{tr} \ln \check{G}^{-1}$, incorporates the contribution of electrons. The inverse electron Green's function operator \check{G}^{-1} has a matrix structure in the tensor product of Nambu and Keldysh spaces (which we occasionally denote by indices K and N over matrices if there is a need to clarify where they act), depends on collective fields, and is given by

$$\begin{aligned} \check{G}^{-1} &= \check{\mathbf{I}}^K \otimes \begin{pmatrix} i\partial_t - \xi_{p-\xi} \mathcal{A} - e\check{\Phi} & \check{\Delta} \\ -\check{\Delta} & -i\partial_t - \xi_{p+\xi} \mathcal{A} - e\check{\Phi} \end{pmatrix}^N \\ &= \check{G}_0^{-1} - \check{X}, \end{aligned} \quad (4)$$

where

$$\check{G}_0 = \begin{pmatrix} G_0^R & G_0^K \\ 0 & G_0^A \end{pmatrix}^K, \quad (5)$$

$$G_0^{R,A}(\omega, p) = \frac{1}{(\omega \pm i0)^2 - (\Delta_0^2 + \xi_p^2)} \begin{pmatrix} \omega + \xi_p & \Delta_0 \\ -\Delta_0 & -\omega + \xi_p \end{pmatrix}^N, \quad (6)$$

$$G_0^K(\omega, p) = G_0^R(\omega, p)F(\omega) - F(\omega)G_0^A(\omega, p) \quad (7)$$

is the bare (unperturbed) equilibrium Green's function of a BCS superconductor with $\xi_p = (-i\nabla)^2/2m - \mu$, Δ_0 is the saddle point (BCS) value of the modulus of the order parameter, and $F(\epsilon) = \tanh \frac{\epsilon}{2T}$ is the equilibrium electron distribution function, while

$$\begin{aligned} \check{X} &= \left(e\check{\Phi} + \frac{e^2}{2mc^2} \mathcal{A}^2 \right) \sigma_0 - \delta\check{\Delta} i\sigma_2 - \frac{e}{2mc} \{\vec{p}, \vec{\mathcal{A}}\} \sigma_3 \\ &= \sigma_0 \check{X}_0 + i\sigma_2 \check{X}_2 + \sigma_3 \check{X}_3 \end{aligned} \quad (8)$$

includes perturbation terms. Matrices σ_a are Pauli sigma matrices with $\sigma_0 = \hat{1}$ which act in Nambu space. Finally, a full operator trace and multiplication operations involve both matrix multiplication and integration over internal space-time arguments. We will also use partial trace operations $\text{tr}_{K,N}$ which include only tracing out matrix indices in Keldysh and Nambu spaces, respectively. One can see that the presented theory depends only on gauge-invariant potentials Φ and \mathcal{A} ; therefore, the gauge invariance is identically satisfied.

Average values of X are given by the functional integral

$$\langle X \rangle = \int [DA DV D\Delta D\Delta^*] X_{cl} e^{iS} \quad (9)$$

performed over both classical and quantum components of fields. The average electric current and charge density can be found as

$$\begin{aligned} \langle \rho(x, t) \rangle &= \left\langle \frac{i}{2} \frac{\delta}{\delta V_q(x, t)} \text{tr} \ln \check{G}^{-1} \right\rangle - \rho_0^e \\ &= \left\langle -\frac{ie}{2} \text{tr}_N (G^K)(x, x, t, t) \right\rangle - \rho_0^e, \end{aligned} \quad (10)$$

$$\begin{aligned}
\langle j(x, t) \rangle &= \left\langle -\frac{ic}{2} \frac{\delta}{\delta A_q(x, t)} \text{tr} \ln \check{G}^{-1} \right\rangle \\
&= \left\langle -\frac{ie}{2m} \text{tr}_N (\sigma_3 \vec{p} G^K)(x, x, t, t) \right. \\
&\quad \left. - \frac{e}{mc} \mathcal{A}_{cl}(x, t) (\rho(x, t) + \rho_0^e) \right\rangle. \quad (11)
\end{aligned}$$

Here \check{G} is the inverse operator to \check{G}^{-1} given by (4) and itself depends on fields, while $\rho_0^e = \frac{ie}{2} \text{tr}_N G_0^K(x, x, t, t)$ is the bare unperturbed electron charge density. Note that in (10) we need to subtract ρ_0^e due to the presence of the static ionic background, which exactly cancels the average electron density.

IV. SEMICLASSICAL APPROACH

Our problem as formulated above remains too complicated for analytical solution. Usually, one expands action up to the second order in collective fields [13,14] and treats subsequent nonlinear terms perturbatively. Alternatively, one can treat this problem semiclassically and expand the action only in quantum components of collective fields. Within this approach, the full nonlinearity of the classical dynamics is captured exactly, while quantum fluctuations are considered to be small [15,16]. It is sufficient for our purposes to expand the action up to the first order in quantum components of all fields (this approximation will be justified below). Then integration over quantum components yields

$$\langle X \rangle = \int [DA_{cl} DV_{cl} D\Delta_{cl} D\Delta_{cl}^*] X_{cl} \delta^f \left(\frac{\delta S}{\delta X_q} \right) \Big|_{X_q=0}. \quad (12)$$

All classical components assume values determined by equations of motion, while quantum components remain zero. Taking the variation of the action with respect to $V_q, A_q, \delta\Delta_q$, and φ_q consecutively, we arrive at the following set of equations:

$$\frac{-\nabla^2}{4\pi} (V - V_0) = \rho, \quad (13)$$

$$\frac{-\nabla^2 + \partial_t^2/c^2}{4\pi} (A - A_0) = \frac{1}{c} j, \quad (14)$$

$$\Delta = -\frac{ig}{4} \text{tr}_N (i\sigma_2 G^K)(x, x, t, t), \quad (15)$$

$$0 = \dot{\rho} + \vec{\nabla} \cdot \vec{j}, \quad (16)$$

with ρ and j given by Eqs. (10) and (11). The first two are essentially Maxwell equations with electron density and current as sources. The third equation is the dynamic self-consistency equation. The fourth one is the continuity equation. It represents charge conservation and is needed to ensure gauge invariance of response functions. This set of equations together with

$$\check{G}^{-1}[X] \check{G}[X] = (\check{G}_0^{-1} - \check{X}) \check{G}[X] = \check{I} \quad (17)$$

completely describes the dynamics of a superconductor in our approximation. Let us notice that Eq. (17) incorporates two independent equations. The retarded and advanced blocks

describe changes in spectral properties of the electron system due to radiation, while the Keldysh component accounts for the electron distribution function and may be used to derive a collisionless kinetic equation, similar to one for ordinary plasma [15]. Omission of higher orders of quantum components in action is a valid approximation under the assumption that we can neglect quantum and thermal fluctuations. For typical laser intensities it is, indeed, justified. Nevertheless, the quantum nature of electron motion is incorporated into the theory in a manner similar to how it is incorporated within the commonly used self-consistent Born approximation which can be reproduced using this approach. One should also note that this approximation misses the collision integral. However, dissipation is still present due to the Landau damping. Another justification for the omission of the collision integral in the context of nonlinear response is the fact that superconductors exhibit little heating. Let us finally note that the resulting set of collisionless equations is analogous to Vlasov equations for plasma accompanied by the dynamic self-consistency equation and the continuity equation [15].

In what follows we will consider electric field to be completely screened by the substrate and thus use a simplified version of Eq. (13) in the form

$$C(V - V_0) = \rho, \quad (18)$$

where C is the capacitance of the film per unit area [17,18].

V. PERTURBATIVE EXPANSION

Equations (14)–(18) are nonlinear. Therefore, in order to solve them, one generally needs to employ numeric methods. Luckily, the nonlinear response is usually sufficiently weak and can be treated perturbatively in external field A_0 . We will seek the solution of Eqs. (14)–(18) while taking A_0 as the expansion parameter. For the Green's function we have

$$\check{G} = \check{G}_0 + \check{G}_0 \check{X} \check{G}_0 + \check{G}_0 \check{X} \check{G}_0 \check{X} \check{G}_0 + \dots \quad (19)$$

As X has a matrix structure in Nambu space and since we need only $G^K(x, x, t, t)$, it is convenient to introduce kernels

$$\begin{aligned}
&\chi_{a,a_1,\dots,a_n}^{\Omega,\dots,\Omega_n}(p;q) \\
&= \frac{i}{2} \int \frac{d\omega}{2\pi} \text{tr} [\tau_1 \hat{\sigma}_a \check{G}_0(Q + Q_1 + \dots + Q_n) \\
&\quad \times \hat{\sigma}_{a_1} \check{G}_0(Q + Q_2 + \dots + Q_n) \times \dots \times \hat{\sigma}_{a_n} \check{G}_0(Q)]. \quad (20)
\end{aligned}$$

Here Q includes both ω and p , and $Q_i = (\Omega_i, q_i)$ stands for frequencies and momenta of perturbations. We assume q_i lies in the two-dimensional plane of the film and p is three-dimensional. The integral over p is not performed at this stage as X depends on the momentum through $\{p, \mathcal{A}\}$. With use of these kernels the perturbative expansion of the right-hand

sides of Eqs. (14)–(16) and (18) is given by

$$\rho(Q) = -e \sum_p \sum_{n=1}^{\infty} \int \left[\prod_{i=1}^n \frac{d^3 Q_i}{(2\pi)^3} \right] (2\pi)^3 \delta[Q - (Q_1 + \dots + Q_n)] \chi_{0a_1 \dots a_n}^{\Omega_1 \dots \Omega_n}(p; q) X_{a_1}(Q_1) \dots X_{a_n}(Q_n), \quad (21)$$

$$\begin{aligned} \vec{j}(Q) = & -\frac{e}{m} \sum_p \sum_{n=1}^{\infty} \int \left[\prod_{i=1}^n \frac{d^3 Q_i}{(2\pi)^3} \right] (2\pi)^3 \delta[Q - (Q_1 + \dots + Q_n)] \left(\vec{p} + \frac{\vec{q}_1 + \dots + \vec{q}_n}{2} \right) \chi_{3a_1 \dots a_n}^{\Omega_1 \dots \Omega_n}(p; q) X_{a_1}(Q_1) \dots X_{a_n}(Q_n) \\ & - \frac{e}{mc} \int \frac{d^3 Q'}{(2\pi)^3} \vec{A}(Q - Q') [\rho(Q') + (2\pi)^3 \delta(Q') \rho_0^e], \end{aligned} \quad (22)$$

$$\Delta(Q) = -\frac{g}{2} \sum_p \sum_{n=0}^{\infty} \int \left[\prod_{i=1}^n \frac{d^3 Q_i}{(2\pi)^3} \right] (2\pi)^3 \delta[Q - (Q_1 + \dots + Q_n)] \chi_{2a_1 \dots a_n}^{\Omega_1 \dots \Omega_n}(p; q) X_{a_1}(Q_1) \dots X_{a_n}(Q_n). \quad (23)$$

Evaluation of kernels $\chi(p, q)$ with subsequent summation over momentum is a demanding task, especially for a dirty superconductor. Luckily, for our purposes it is not needed as we are interested in frequencies ranging from almost zero to several Δ . At these frequencies typical light wavelengths are of the order of 10 μm or (usually) larger. Hence, conditions $q \ll p_F$ and $q \ll 1/l_{el}$, where p_F is the Fermi momentum and l_{el} is the electron mean free path, are well satisfied. Therefore, we can expand all kernels in momenta q_i and consider only zeroth- and first-order (if needed) terms which are expressed in terms of kernels $\chi_{a \dots a_n}(p)$ defined as $\chi_{a \dots a_n}(p; q_1 = 0, \dots, q_n = 0)$. This expansion is performed explicitly in the Appendix.

Subsequent calculations can be further simplified if we take into account the Ward identity

$$\begin{aligned} \hat{\sigma}_0 \check{G}_0(\omega, p) - \check{G}_0(\omega + \Omega, p) \hat{\sigma}_0 \\ = \check{G}_0(\omega + \Omega, p) (\Omega \hat{\sigma}_3) \check{G}_0(\omega, p). \end{aligned} \quad (24)$$

It allows us to express kernels $\chi(p)$ of order n with index 3 through kernels of order $n - 1$ via

$$\begin{aligned} \chi_{a_0 a_1 \dots a_n}^{\Omega_1 \dots \Omega_i \dots \Omega_n} \\ = \left(\chi_{a_0 a_1 \dots a_{i-1}, a_{i+1} \dots a_n}^{\Omega_1 \dots \Omega_{i-1} + \Omega_i, \Omega_{i+1} \dots \Omega_n} - \chi_{a_0 a_1 \dots a_{i-1}, a_{i+1} \dots a_n}^{\Omega_1 \dots \Omega_{i-1}, \Omega_i + \Omega_{i+1}, \dots \Omega_n} \right) / \Omega_i. \end{aligned} \quad (25)$$

From this it immediately follows that kernels $\chi_{33 \dots 3a_3 \dots 33}(p)$, where all indices, except maybe one, are equal to 3, vanish identically. It is important to stress that such a claim is valid only in the limit of $q \rightarrow 0$ and $\Omega \neq 0$ since some of these kernels exhibit noninterchangeability of limits $q \rightarrow 0$ and $\Omega \rightarrow 0$. This fact ensures the diamagnetic nature of current in the system, which will be shown to have a dramatic effect on the response.

It is also important to distinguish kernels that vanish in the presence of electron-hole (EH) symmetry. It is straightforward to verify (see the Appendix) that kernels with an even number of indices 0 are EH symmetric, while those with an odd number are EH asymmetric.

VI. UNIFORM CASE

Now we are ready to investigate the nonlinear response itself starting with the simplest uniform case, that is, when the

falling plane wave is perpendicular to the film. Our observable of interest is electric current j . From rotational symmetry considerations the current includes only odd orders in A_0 , while ρ , $\delta\Delta$, and φ have only even-order terms. As the incident wave is uniform along the film, all the perturbations should also be uniform. Now we take into account the continuity equation and immediately get $\rho(x, t) = 0$.

The solution of Eqs. (13)–(17) is now found trivially with the result

$$\vec{j} = -\frac{e}{mc} \vec{A} \rho_0^e. \quad (26)$$

For the third-order response we must carefully investigate all potentially contributing kernels in Eq. (23). Such an analysis is carried out in the Appendix. The result shows the absence of a third-order response in the uniform case as a result of the fact that the current in our system is purely diamagnetic due to the Ward identity (25). A more thorough investigation shows that in the clean case this result should hold at all orders as a consequence of the identity

$$[\hat{j}_{q=0}, \hat{H}] = 0, \quad (27)$$

which holds for uniform fields $\vec{A}(x, t) = \vec{A}(t)$. Therefore, within the adopted model with a parabolic electron spectrum the nonlinear response occurs only for space-dependent perturbations (see, e.g., [8, 9] and references therein) or, equivalently, when the incident wave is not normal to the plane of the film. In real materials with disorder the identity (27) no longer holds, and the optical response of the film may become nontrivial (see [19]). In a real experiment, the third harmonic generation may also occur at the boundary of the sample for various reasons, mainly due to the presence of defects. However, theoretical analysis of boundary effects lies beyond the scope of the present work.

VII. NONUNIFORM CASE: FIRST-ORDER RESPONSE

Now we will turn to the case of a nonuniform incident field which corresponds to the situation when the electromagnetic wave is not perpendicular to the film. Let us first investigate the first-order response within our approach. The first-order

expressions for ρ , j , and Δ are given by

$$\begin{aligned}\rho^{(1)}(q) &= -e \sum_p \left[\Phi^q \chi_{00}^\Omega(p; q) e - \chi_{02}^\Omega(p; q) \delta \Delta^q \right. \\ &\quad \left. - \frac{\chi_{03}^\Omega(p; q) e \mathcal{A}_{a_1}^q}{cm} \left(p_{a_1} + \frac{q_{a_1}}{2} \right) \right], \\ j_a^{(1)}(q) &= -\frac{\rho_0^e e \mathcal{A}_a^q}{cm} - \frac{e}{m} \sum_p \left(p_a + \frac{q_a}{2} \right) \left[\Phi^q \chi_{30}^\Omega(p; q) e \right. \\ &\quad \left. - \chi_{32}^\Omega(p; q) \delta \Delta^q - \frac{\chi_{33}^\Omega(p; q) e \mathcal{A}_{a_1}^q}{cm} \left(p_{a_1} + \frac{q_{a_1}}{2} \right) \right], \\ \Delta^{(1)}(q) &= -\frac{g}{2} \sum_p \left[\Phi^q \chi_{20}^\Omega(p; q) e - \chi_{22}^\Omega(p; q) \delta \Delta^q \right. \\ &\quad \left. - \frac{\chi_{23}^\Omega(p; q) e \mathcal{A}_{a_1}^q}{cm} \left(p_{a_1} + \frac{q_{a_1}}{2} \right) \right].\end{aligned}\quad (28)$$

A major simplification can be made if we take into account that $v_F \ll c$, which leads to $q \ll \Omega/v_F$ (in addition to the aforementioned condition $q \ll p_f, 1/l_{el}$). Being interested in frequencies in the microwave range, we see that all the kernels can be expanded in q up to the lowest necessary order for which relatively simple expressions are available (see the Appendix). Inclusion of terms of higher order in q produces an additional small parameter $\propto v_F/c$. We will also consider electromagnetic field generated by currents inside the film to be small compared to the incident laser field. This is justified as long as the film is thinner than the London penetration depth. This assumption allows us to set $A = A_0$ and completely discard the Maxwell equation (14).

Taking into account these considerations, we now insert the expansion (28) into Eqs. (15), (16), and (18) and find V and φ . Inserting these solutions back into Eq. (28) yields the linear response current. In the simplest case of electron-hole symmetric material we get

$$\vec{j}^q = -\frac{\rho_0^e e}{mc} \left(\vec{A}^q - \frac{\vec{q}(\vec{q}\vec{A}^q)}{q^2 - g(\Omega)\frac{\Omega^2}{v_0^2}} \right), \quad (29)$$

where

$$v_0^2 = \frac{d\rho_0 e}{mC} \sim v_F^2 \frac{1}{\varepsilon} \left(\frac{d}{a_0} \right)^2 \quad (30)$$

is the plasma mode velocity [4,18] and

$$g(\Omega) = \frac{\sum_p \chi_{00}^\Omega(p) e^2 d}{\sum_p \chi_{00}^\Omega(p) e^2 d + C} \approx 1. \quad (31)$$

Equation (29) represents the first-order result for the electric current. The contribution of the Higgs mode is small due to both electron-hole asymmetry and the smallness of q and is not included in (29). However, even in electron-hole asym-

metric materials the Higgs mode does not change much, with its only effect being a renormalization of the plasma mode parameters. The expression for the current contains a perfect diamagnetic response term and a contribution from the plasma mode, which has a pole at $\Omega \approx v_0 q$. If the plasma mode velocity satisfies $v_0 \ll c$, then we arrive at $\vec{j}^{(1)} \approx -\frac{\rho_0^e e}{mc} (\vec{A} - \frac{\vec{q}(\vec{q}\vec{A})}{q^2})$. Note that the expression in parentheses is not equal to A^\perp since here A and q are projections of the corresponding three-dimensional wave amplitude and wave vector on the plane of the film.

VIII. NONUNIFORM CASE: PHOTOINDUCED CURRENT

Now let us proceed to the second order of the perturbative expansion. As $A(x) = A_q e^{iqx} + A_q^* e^{-iqx}$, in the second order we naturally get a current response at wave vectors $\pm 2q, 0$. Here we will consider specifically a stationary and uniform component of the current. In the regime of fluctuating superconductivity (at $T > T_c$) the effect was studied in [20]. Normally, in bulk crystals with inversion symmetry the second-order response is forbidden. In a thin-film geometry, this is no longer the case as there exists a unique direction normal to the plane of the film. The current is allowed to flow only along the plane of the film and is then affected only by the in-plane components of A , which change with a spatial period defined by the in-plane wave vector q . The projection of A is not orthogonal to q , and one may easily obtain a nonzero second-order current response. The appearance of the second-order response in such systems may be thought of as a boundary effect and may happen not only in low-dimensional geometry but also at the boundary of a bulk sample.

After a series of simplifications including decomposition in q and angular integration, the resulting expression in the second order assumes the form

$$j_a = -\frac{e}{cm} (\mathcal{A}_a^q \rho^{-q} + \mathcal{A}_a^{-q} \rho^q) + \text{subleading terms}. \quad (32)$$

Subleading terms result from the $n = 2$ expansion of (23) and are smaller by a parameter $\propto (v_F/c)^2 (\Omega/\Delta)^2$ (for details see the Appendix), unless the frequency of the incident wave is not too close to 2Δ , where resonant excitation of the collective modes takes place. Omission of higher-order terms is no longer possible for $\Omega - 2\Delta \sim \Delta (\frac{v_F}{c})^2$, where their contribution is of the same order. However, since all kernels become singular in this limit, the response itself becomes nonlinear. In this regime Eqs. (14)–(18) can no longer be treated perturbatively and require alternative solution methods.

Collecting the answer using the already obtained first-order solution, for the stationary current density we get

$$\vec{j} = -\frac{eC}{mc^2 d} g(\Omega) \frac{(\vec{q}\vec{A}_q)\Omega}{q^2 - \frac{\Omega^2}{v_0^2} g(\Omega)} \left(\vec{A}_{-q} - \frac{\vec{q}(\vec{q}\vec{A}_{-q})}{q^2 - \frac{\Omega^2}{v_0^2} g(-\Omega)} \right). \quad (33)$$

In sufficiently thin films where $v_0 \ll c$ this answer reduces to the simple form $j = \frac{\rho_0 e^2}{(mc)^2 \Omega} (\vec{q}\vec{A}_q)\vec{A}_{-q}$, which is valid not too close to $\Omega = 2\Delta$. It is interesting that this answer is almost identical to the standard result for two-dimensional systems [21], with the main difference being the dependence on light polarization.

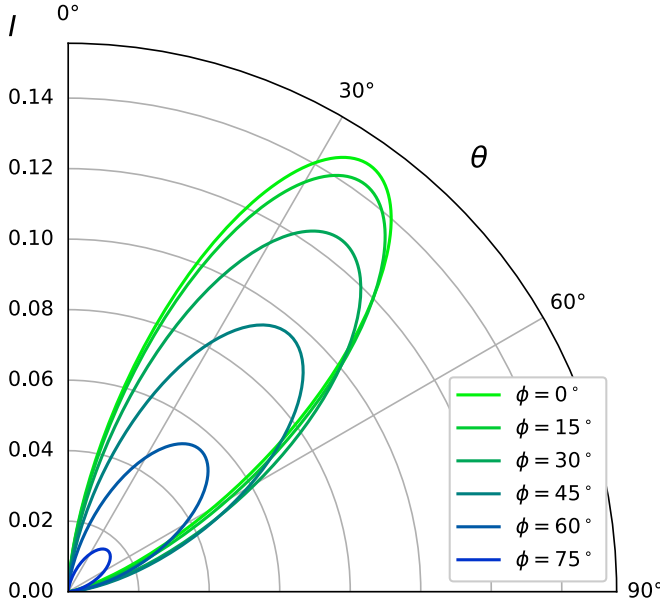


FIG. 2. Dependence of the second-order photoinduced current I on the incidence angle θ for different angles ϕ in the limit $c \rightarrow \infty$ [see Eq. (34)]. At $\phi = 0$ the incident wave is transverse magnetically (TM) polarized, while at $\phi = \pi/2$ the polarization is transverse electric (TE).

The current strongly depends both on the incidence angle of the light wave and on its polarization. The answer becomes zero in the limits of the wave being both perpendicular and parallel to the plane of the superconducting film. It is also zero if the polarization of the wave is transverse electric (TE), for which $\vec{A} \perp \vec{q}$. Therefore, the largest values of the shift current are obtained for TM polarization at intermediate incident angles θ . Let us now explore the full dependence of the photoinduced current on the angle and polarization assuming that polarization components are given by $A^{\text{TM}} = A \cos \phi$ and $A^{\text{TE}} = A \sin \phi$. Taking into account that \vec{q} is the projection of the full momentum of the incident wave, in the limit $v_0/c \rightarrow 0$ we get

$$|j|^2 \propto \cos^2 \phi \sin^2 \theta \cos^2 \theta (\sin^2 \phi + \cos^2 \phi \cos^2 \theta) \quad (34)$$

(see Fig. 2). For a fixed linear polarization characterized by ϕ the maximal value of the current is obtained at the incidence angle determined by $\cos^2 \theta = \frac{1+\cos^2 \phi}{2+\cos^2 \phi}$.

It is important to stress that the contribution of the Higgs mode to the second-order response is suppressed by both approximate electron-hole symmetry and the ratio v_F/c and may safely be omitted in our situation.

IX. EFFECTIVE NONLINEAR ACTION

In the previous sections we identified leading first- and second-order terms in the perturbative expression for the current. Instead, it may be more convenient to carry out a perturbative expansion directly in the action (1) where dominant terms may be determined using the same arguments as before. An effective action sufficient to reproduce the obtained results

for the current is given by

$$S_{\text{eff}} = 2d \int \frac{d^3 k}{(2\pi)^3} \left(-\tilde{\Phi}_{cl}(k) K_{\Phi\Phi}(k) \tilde{\Phi}_q(-k) - \frac{\rho_0^e}{mc^2} \mathcal{A}_{cl}(k) \mathcal{A}_q(-k) + \delta \Delta_{cl}(k) K_{\Delta\Delta}(\Omega) \delta \Delta_q(-k) + \delta \Delta_{cl}(k) K_{\Delta\Phi}(\Omega) \tilde{\Phi}_q(-k) + \delta \Delta_{cl}(-k) K_{\Delta\Phi}(-\Omega) \tilde{\Phi}_q(k) \right) + 2 \int dt d^2 x C V_{cl} V_q - 2 \int dt \int d^3 x \frac{H_{cl} H_q}{4\pi}, \quad (35)$$

with k encompassing both Ω and q , the new potential $\tilde{\Phi}$ given by $\tilde{\Phi}_{F,B} = \Phi_{F,B} + \frac{e}{2mc^2} \mathcal{A}_{F,B}^2$, $\tilde{\Phi}_{cl,q} = (\tilde{\Phi}_F \pm \tilde{\Phi}_B)/2$, and the kernels being

$$K_{\Phi\Phi}(\Omega) = e \sum_p \chi_{00}^\Omega(p), \quad K_{\Delta\Phi}(\Omega) = \sum_p \chi_{20}^\Omega(p),$$

$$K_{\Delta\Delta}(\Omega) = -\frac{4}{g} + \sum_p \chi_{22}^\Omega(p).$$

Nonlinearity in Eq. (35) is introduced through $\tilde{\Phi}$, which contains \mathcal{A}^2 . It may be shown that all other nonlinear terms (up to third order) only generate smaller corrections to the final expression for the current as they contain higher orders of q . Within our approximations, this action provides a complete classical description of our system generating all equations of motion. It can be used to evaluate other nonlinear effects, such as generation of the second and third harmonics. However, one should take into account that the action (35) contains terms relevant specifically for current response, while different types of response may be more sensitive to other terms.

Comparing the obtained action (35) with [8], we find that the authors derived a quite similar effective action for uniform perturbations. Their action taken in the limit of parabolic electron dispersion coincides with ours if we identify “density fluctuations” ρ in [8] with V , take $q \rightarrow 0$, and consider screening in the substrate in this limit negligible (i.e., set $C = 0$). The differences originate both from considering different geometries and from the fact that in our case of parabolic dispersion uniform response is forbidden, so the phase gradient must be accounted for. As our analysis shows, the variable φ is always crucial for obtaining gauge-invariant response functions.

It is also necessary to mention that the effective action (35) is a simple nonlinear extension of the Gaussian effective action obtained in [13,14]. In these works, all the linear response kernels were evaluated in the dirty limit for arbitrary frequencies, which serves as a good starting point for further investigation of nonlinear microwave response in the dirty limit.

Finally, the effective action (35) can be easily modified in order to incorporate quantum and fluctuation effects. For this purpose it is sufficient to introduce terms $\propto X_q X_q$, while the coefficients may be restored using the fluctuation-dissipation theorem.

X. RESULTS AND DISCUSSION

In this paper we investigated the nonlinear response of a thin superconducting film in electromagnetic field. We developed an approach based on equations of motion for the collective fields in the spirit of self-consistent Vlasov equations used to describe the dynamics of interacting plasma. The resulting set of equations (18) and (14)–(16), along with (17), governs the dynamics of the electromagnetic field and order parameter and has a clear physical interpretation and includes the Poisson equation, the self-consistency equation, the continuity equation, and the equation for the Green's function. In contrast to the more widely used diagrammatic approach in which in order to achieve gauge invariance one needs to carefully sum various diagrammatic contributions [12], within our approach the charge conservation manifests itself right away: the equation of motion for the superconducting phase coincides with the continuity equation for electric charge.

Charge conservation is particularly important in the context of the electromagnetic response of a superconductor due to the diamagnetic nature of the superconducting current. At least for pure superconductors, as long as the external field varies slowly in space ($q \ll \Omega/v_F$), the paramagnetic response is very weak and vanishes identically for uniform fields in several of the lowest orders [see Eq. (25) and the paragraph below]. Therefore, the current is given by the simple expression $\vec{j} = -\frac{e}{mc}\rho(\vec{A} - \frac{c}{2e}\vec{\nabla}\varphi)$. In a uniform field, $\varphi(x)$ is uniform in space, while the density assumes its equilibrium value due to the charge conservation. Hence, only the linear term $\vec{j} = -\frac{e}{mc}\rho_0\vec{A}$ survives reproducing a well-known classical answer [22].

It is instructive to compare this conclusion to the result of [8], in which the authors studied third harmonic generation in bulk superconductors. They found that the third-order current is determined predominantly by the diamagnetic term with a (usually) small correction produced by the amplitude mode, in agreement with our results. A closer examination of the response kernels obtained in [8] shows that after renormalization by phase degrees of freedom they obtained a correction which cancels the kernels out exactly in the case of parabolic electron dispersion, leaving the third harmonic equal to zero. This again highlights the role of the phase mode.

The crucial role of scalar fields φ and V in the context of the nonlinear response was also emphasized in [11], in which the authors showed how their dynamics cancels out the density response from other channels. The authors also studied the problem of third harmonic generation. According to their results, the main contribution to the third-order response (at least, in the absence of voltage applied to the film) comes from the term $S_{AA\Delta} \propto K_{AA\Delta}^{\Omega\Omega} \mathcal{A}^2(\Omega)\delta\Delta(-2\Omega)$, which generates current $j(3\Omega) \propto A(\Omega) \frac{[K_{AA\Delta}^{\Omega\Omega}(2\Omega)]^2}{K_{AA\Delta}} \mathcal{A}^2(\Omega)$. They claimed it persists in the limit of $q \rightarrow 0$ and provided an expression for the necessary kernel in the limit $\lim_{\Omega \rightarrow 0} K_{AA\Delta}$, which remains finite. We find the latter statement to be erroneous since the identities (24) and (25) leave this kernel equal to zero at $q = 0$. One may obtain a nonzero expression for this kernel in the opposite limit $\lim_{q \rightarrow 0} \lim_{\Omega \rightarrow 0}$, which is relevant in the context of the response to nonuniform static fields but not in our situation.

Such behavior is an example of a well-known nonanalyticity which many response kernels exhibit at $q, \Omega \rightarrow 0$, with the most common example being the Lindhard function for the polarization operator [23,24]. In the context of third harmonic generation in superconductors, we find again that in our situation the evidence for the dominance of the Higgs mode is insufficient.

Our results demonstrate that in clean isotropic superconducting films second- and third-order responses to a uniform microwave field vanish. This implies strong dependence of the response on the incidence angle of the microwave, as well as on its polarization. The absence of a response at $q = 0$ introduces the necessity to expand the response kernels in momenta, which in turn makes the resulting current small by the additional parameter v_F/c . This additional smallness provides an edge to thin films as they allow an angle-dependent nonlinear response already in the second order. In our analysis, we focus our attention on the shift current—a stationary, or, in practice, slowly varying, component of the photoinduced current, for which we derive the explicit analytical expression given in Eq. (33). It is proportional to the total carrier density and shows weak dependence on other parameters of the superconducting material or the substrate, as long as the superconducting film is properly insulated and the velocity of the plasma mode remains much smaller than c . The frequency dependence is simple, with $|j| \propto I/\Omega^2$ outside of a narrow range of frequencies near $\Omega = 2\Delta$, where the current displays nonanalyticity in all orders while a perturbative calculation becomes impossible. In this case Eqs. (13)–(17) must be solved using alternative methods, for example, numerical ones. It should be stressed that the resonance at $\Omega = 2\Delta$ occurs in our analysis due to the excitation of the plasma mode, while the contribution of the Higgs mode is negligibly small.

It is important to discuss the range of validity of our results. It is based on two premises: the diamagnetic nature of the current in superconductors and the conservation of electric charge. The first one is a consequence of the identity (24), which holds for pure superconductors and leaves zero all the paramagnetic response kernels in the limit $q \rightarrow 0$, $\Omega \neq 0$ at any temperature. In disordered systems such a simple relation no longer exists, and a paramagnetic response may arise. Moreover, several studies claim the paramagnetic response of higher orders is much stronger in the presence of disorder [9,10]. Thus, the disordered case requires separate treatment and will be worked out elsewhere.

The charge conservation is another issue which should be addressed here as it requires the film to be completely insulated from other electron reservoirs. It might be especially difficult to achieve in DC measurements, which usually require electrodes connected to the film and involve larger timescales. If charge is allowed to be transferred between the film and the environment, then the charge conservation for the film alone no longer holds, and the response at $q = 0$ becomes possible, although in this case it becomes largely dependent on the environment and cannot be considered a property of the superconductor alone, but rather that of the whole setup.

Finally, even if the film is sufficiently pure and is properly insulated, it may still demonstrate an angle-independent

third-order response due to the boundary effects. As the bulk third-order response is relatively weak due to its dependence on the angle, the boundary might hide it in a given sample. Therefore, a film of sufficiently large area may be required. Within our approach, it is possible to account for boundary effects due to the fact that the effective action (35) includes only spatial derivatives of first order. In order to do this, boundary conditions on charge and current must be imposed.

Our findings may be tested in experiments with quasi-two-dimensional films.

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APPENDIX

1. Kernels

Evaluating integrals in Eq. (20) at $q = 0$, we obtain

$$\sum_p \chi_{00}^{\Omega}(p) = \frac{2v_0}{y\sqrt{1-y^2}} \arctan \frac{y}{\sqrt{1-y^2}}, \quad (\text{A1})$$

$$\sum_p \chi_{003}^{\Omega}(p) = \frac{v_0}{2\Delta y} \left(-1 + \frac{\sqrt{1-y^2}}{y} \arctan \frac{y}{\sqrt{1-y^2}} \right), \quad (\text{A2})$$

$$\sum_p \chi_{22}^{\Omega}(p) = (y^2 - 1) \sum_p \chi_{00}^{\Omega}(p), \quad (\text{A3})$$

where $T \rightarrow 0$ and $y = (\Omega + i0)/2\Delta$.

2. Spectral decomposition

Any retarded or advanced propagator $f^{R,A}$ can be expressed via the Lehman representation

$$f^{R,A}(\omega) = \int_{-\infty}^{\infty} dz \frac{J_f(z)}{\omega \pm i0 - z}, \quad (\text{A4})$$

where

$$J_f(\omega) = \frac{i}{2\pi} [f^R(\omega) - f^A(\omega)]. \quad (\text{A5})$$

For the bare equilibrium Green's function of a BCS superconductor

$$\hat{J}_p(\omega) = J_p(\omega) \cdot \hat{M}_p^{(N)}(\omega), \quad (\text{A6})$$

with

$$J_p(\omega) = \frac{1}{2\varepsilon_p} [\delta(\omega - \varepsilon_p) - \delta(\omega + \varepsilon_p)], \quad (\text{A7})$$

$$\hat{M}_p(\omega) = (\xi_p \hat{\sigma}_0 + \Delta i \hat{\sigma}_2 + \omega \hat{\sigma}_3)^N. \quad (\text{A8})$$

Accordingly,

$$\begin{aligned} \check{G}_0(\omega, p) &= \int_{-\infty}^{\infty} dz \left(\frac{1}{\omega + i0 - z} - \frac{-2\pi i \delta(\omega - z) F_\omega}{\frac{1}{\omega - i0 - z}} \right)^K \\ &\times J_p(z) \hat{M}_p(z) \end{aligned} \quad (\text{A9})$$

$$= \begin{pmatrix} S^R(\omega) & [S^R(\omega) - S^A(\omega)] F_\omega \\ 0 & S^A(\omega) \end{pmatrix}^K \otimes \hat{M}_p(\omega), \quad (\text{A10})$$

$$S^{R,A}(\omega) = \frac{1}{(\omega \pm i0)^2 - \varepsilon_p^2}. \quad (\text{A11})$$

Spectral decomposition can be used in the regularization of kernels or to analyze their nontrivial limits. Separation of the Nambu space structure in matrix \hat{M} is useful for determining some properties of kernels.

3. EH symmetry of kernels $\chi(p)$

Let us consider how kernel $\chi_{aa_1 \dots a_n}(p)$ changes upon the substitution $\xi_p \rightarrow -\xi_p$. We notice that the kernel's symmetry is determined by the symmetry of the trace

$$\text{Tr} = \text{tr}_N [\sigma_a \hat{M}(\omega + \dots) \sigma_{a_1} \hat{M}(\dots) \dots \sigma_{a_n} \hat{M}(\omega)]. \quad (\text{A12})$$

Another observation is $\hat{M}(\xi \rightarrow -\xi) = -\sigma_1 \hat{M} \sigma_1$. Inserting this relation into the just mentioned trace Tr and performing a cyclic permutation, we get the same expression multiplied by $(-1)^{n_0}$, where n_0 is the number of indices 0 in the kernel.

4. Momentum expansion of kernels $\chi(p; q)$

Let us use the identity

$$\frac{\partial G_0(\omega, p)}{\partial p} = -G_0(\omega, p) \frac{\partial G_0^{-1}(\omega, p)}{\partial p} G_0(\omega, p). \quad (\text{A13})$$

Therefore,

$$G_0(\omega, p + q) \approx G_0(\omega, p) - \vec{v} \vec{q} G_0(\omega, p) G_0(\omega, p). \quad (\text{A14})$$

Inserting the last equality in the definition of kernels $\chi(p; q)$, we have

$$\begin{aligned} \chi_{a, a_1, \dots, a_n}^{\Omega_1, \dots, \Omega_n}(p; q_1, \dots, q_n) &= \chi_{a, a_1, \dots, a_n}^{\Omega_1, \dots, \Omega_n}(p) \\ &\approx -\vec{v}(\vec{q}_1 + \dots + \vec{q}_n) \chi_{a, 0, a_1, \dots, a_n}^{0, \Omega_1, \dots, \Omega_n}(p) \\ &\quad - \vec{v}(\vec{q}_2 + \dots + \vec{q}_n) \chi_{a, a_1, 0, a_2, \dots, a_n}^{\Omega_1, 0, \Omega_2, \dots, \Omega_n}(p) \\ &\quad - \dots - \vec{v} \vec{q}_n \chi_{a, a_1, \dots, a_{n-1}, 0, a_n}^{\Omega_1, \dots, \Omega_{n-1}, 0, \Omega_n}(p). \end{aligned} \quad (\text{A15})$$

In the case of extreme necessity such a decomposition may be carried out at higher orders.

5. Third-order response in the uniform case

In the uniform case, due to the charge conservation, the diamagnetic contribution to the current is given by (26). Hence, we need to consider only paramagnetic contributions. Symmetry considerations show that from all the collective fields only \mathcal{A} obtains first-order corrections. From the third-order expansion of the current (23) only a single term with χ_{3333} remains relevant. However, the Ward identity (25) leaves it identically zero. From the second-order expansion angular integration leaves terms containing kernels with two indices 3, such as χ_{303} , which again vanish by virtue of the Ward identity (25). In the first order all paramagnetic terms are trivially zero.

As a result, the current does not acquire second- and third-order corrections in the uniform case as a result of the charge

conservation and the Ward identity (24), which cancels all paramagnetic contributions. Therefore, Eq. (26) is correct at least up to fifth order.

6. Subleading terms of photoinduced current

The subleading terms of the photoinduced current originate from the $n = 2$ order of Eq. (23). They vanish at $q = 0$ and therefore require decomposition in q . As the result, for them we have

$$j_a^{\text{subl}} = -\frac{e}{m} \sum_p p^2 \left(\frac{\Phi^{-q} \chi_{3002}^{-\Omega 0 \Omega} \delta \Delta^q e}{3m} q_a - \frac{\Phi^{-q} \chi_{3200}^{\Omega 0 -\Omega} \delta \Delta^q e}{3m} q_a \right. \\ - \frac{\Phi^q \chi_{3002}^{\Omega 0 -\Omega} \delta \Delta^{-q} e}{3m} q_a + \frac{\Phi^q \chi_{3200}^{-\Omega 0 \Omega} \delta \Delta^{-q} e}{3m} q_a \\ + \frac{\Phi^{-q} \chi_{3003}^{-\Omega 0 \Omega} e^2 \mathcal{A}_{a_2}^q q_{a_2} q_a}{6cm^2} + \frac{\Phi^{-q} \chi_{3300}^{\Omega 0 -\Omega} e^2 \mathcal{A}_{a_1}^q q_{a_1} q_a}{6cm^2} \\ + \frac{\Phi^q \chi_{3003}^{\Omega 0 -\Omega} e^2 \mathcal{A}_{a_2}^{-q} q_{a_2} q_a}{6cm^2} + \frac{\Phi^q \chi_{3300}^{-\Omega 0 \Omega} e^2 \mathcal{A}_{a_1}^{-q} q_{a_1} q_a}{6cm^2} \\ - \frac{\chi_{3203}^{-\Omega 0 \Omega} \delta \Delta^{-q} e \mathcal{A}_{a_2}^q q_{a_2} q_a}{6cm^2} - \frac{\chi_{3203}^{\Omega 0 -\Omega} \delta \Delta^q e \mathcal{A}_{a_2}^{-q} q_{a_2} q_a}{6cm^2} \\ \left. - \frac{\chi_{3302}^{-\Omega 0 \Omega} \delta \Delta^q e \mathcal{A}_{a_1}^{-q} q_{a_1} q_a}{6cm^2} - \frac{\chi_{3302}^{\Omega 0 -\Omega} \delta \Delta^{-q} e \mathcal{A}_{a_1}^q q_{a_1} q_a}{6cm^2} \right). \quad (\text{A16})$$

They all contain kernels of high order and are proportional to q^3 . Compared to the diamagnetic contribution $\propto \rho \mathcal{A}$, they are small. Therefore, the photoinduced current is predominantly determined by diamagnetic terms associated with charge density variations.

Let us investigate the range of validity of the above statement. For this purpose we need to compare similar terms from Eq. (32) with those from Eq. (A16). For this purpose we need

explicit expressions for the relevant kernels. For $(\Phi \mathcal{A})$ terms at $T \rightarrow 0$ the relevant kernels read

$$\chi_{00}^{\Omega}(p) = -\frac{4\Delta^2}{\varepsilon_p(\Omega_1 - 2\varepsilon_p)(\Omega + 2\varepsilon_p)}, \quad (\text{A17})$$

$$\chi_{3300}^{\Omega 0 -\Omega}(p) = \chi_{3003}^{-\Omega 0 \Omega} = \frac{\Delta^2(\Omega^2 + 4\varepsilon_p^2)}{\varepsilon_p^3(\Omega - 2\varepsilon_p)^2(\Omega + 2\varepsilon_p)^2}. \quad (\text{A18})$$

At $\Omega \rightarrow 0$ we have

$$\sum_p \chi_{00}(p) = 2\nu_0, \quad (\text{A19})$$

$$\sum_p p^2 [\chi_{3300}^{\Omega 0 -\Omega}(p) + \chi_{3003}^{-\Omega 0 \Omega}(p)] \approx \frac{8}{3} \frac{\nu_0 p_F^2}{\Delta^2}. \quad (\text{A20})$$

Then the leading contribution $j_{\Phi \mathcal{A}}^{\text{lead}}$ and the subleading term $j_{\Phi \mathcal{A}}^{\text{subl}}$ proportional to $\Phi^{-q} \mathcal{A}^q$ are given by

$$\tilde{j}_{\Phi \mathcal{A}}^{\text{lead}} = \frac{e^3}{mc} 2\nu_0 \Phi^{-q} \tilde{\mathcal{A}}^q, \quad (\text{A21})$$

$$\tilde{j}_{\Phi \mathcal{A}}^{\text{subl}} = -\frac{e^3}{6m^3 c} \frac{8}{3} \frac{\nu_0 p_F^2}{\Delta^2} \Phi^{-q} (\tilde{\mathcal{A}}^q \tilde{q}) \tilde{q}. \quad (\text{A22})$$

It is now straightforward to verify that the subleading contribution is, indeed, small by the parameter $\propto (\nu_F/c)^2 (\Omega/\Delta)^2$.

The situation changes for frequencies Ω close to 2Δ , where kernels cease to be analytic functions of Ω . For $(\Omega - 2\Delta)/\Delta = \varepsilon \ll 1$ in the leading order we have

$$\sum_p \chi_{00}(p) \approx \frac{\nu_0 \pi}{\sqrt{\varepsilon}}, \quad (\text{A23})$$

$$\sum_p p^2 (\chi_{3300}^{\Omega 0 -\Omega}(p) + \chi_{3003}^{-\Omega 0 \Omega}(p)) \approx \frac{\nu_0 \pi p_F^2}{4\Delta^2 \varepsilon^{3/2}}. \quad (\text{A24})$$

Now we have $j_{\Phi \mathcal{A}}^{\text{subl}}/j_{\Phi \mathcal{A}}^{\text{lead}} \sim \frac{1}{12\varepsilon} (\frac{\nu_F}{c})^2$. Hence, omission of higher-order kernels is unjustified in a narrow range of frequencies $\Omega - 2\Delta \sim \Delta (\frac{\nu_F}{c})^2$. We also need to point out that in this range the response should become highly nonlinear due to the more singular behavior of higher-order kernels.

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