Nonlinear response of diffusive superconductors to ac electromagnetic fields

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Motivated by the recent experimental progress in studying conventional and unconventional superconductors in a pump-probe setup, we perform a comprehensive theoretical analysis of the nonlinear response of a diffusive BCS conventional superconductor to the action of an alternating electromagnetic field using a generalized Usadel equation. We analyze the response up to the second order of the perturbation in the amplitude of the vector potential $\vec{A}$, the superconducting order parameter Δ, and in the third order for the current $\vec{j}$. On the basis of this approach, we derive general expressions for the retarded (advanced) Green's functions, as well as the Keldysh function for an arbitrary number of harmonics of the incident field. Most importantly, we analyze the set of physical observables in a nonequilibrium superconductor, such as frequency and the temperature dependencies of the zero harmonic $\delta(\Delta)_0$ (Eliashberg effect), the second harmonic $\delta(\Delta)_{2\Omega}$, as well as the third harmonic for the electric current $j(3\Omega)$ under the action of monochromatic irradiation. For the same set of parameters, we also analyze the behavior of the reflectivity and the down-conversion intensity of a thin superconducting film, discussed recently in the context of parametric amplification of superconductivity. We derive these quantities microscopically and show the connection of the down-conversion intensity to the third-harmonic generation currents induced by the amplitude mode and the direct action of the electric field on the charge carriers.

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I. INTRODUCTION

The study of nonstationary properties of superconductors started soon after the formulation of the BCS theory of superconductivity [1] with the calculation of the ac conductance of superconductors [2,3]. Later, Gor'kov and Eliashberg [4] investigated nonlinear ac properties on the basis of generalized Ginzburg-Landau equations. The ac conductance in a current-carrying superconductors was studied in Refs. [5–7] and more recently in Refs. [8–11] using a more microscopic approach.

For obvious reasons, one of the most interesting problems is to study the spectrum of collective modes of the phase φ and amplitude mode $\delta\Delta$ of the superconducting order parameter Δ. In the absence of a microscopic theory, these modes were discussed by Anderson [12] and independently by Bogoliubov [13]. In particular, Anderson came to the conclusion that phase variations in space and time (phase mode) $\varphi(t,x)$ lead to perturbations of the electric charge and, therefore, they cannot exist in metals until the frequency ω exceeds the plasma frequency $\omega_{\rm pl}$, which is much higher than Δ. Remarkably, this idea of Anderson's motivated Higgs in his prediction of a new type of boson (Higgs boson) in particle physics [14–16].

Contrary to that, the amplitude mode (AM) is electrically neutral and can be excited in the absence of an additional interaction, for example the Coulomb interaction. The AM in BCS superconductors corresponds to perturbations of the quasiparticle distribution function $n(\xi_p)$ symmetric with respect to a variable ξ_p, here $\xi_p = v_F(p - p_F)$. The phase mode is caused by a branch-imbalance, i.e., by a deviation from zero of a part of the function $n(\xi_p)$ asymmetric in variable ξ_p [17–19]. The amplitude mode is confusingly called the Higgs mode, although logically the Higgs mode would correspond instead to the plasma mode. This fact has also been highlighted in several articles [20,21].

Theoretically, the evolution of $\delta\Delta(t)$ after a sudden perturbation (quench) was studied in Ref. [22] in a way similar to the problem of the electric field E evolution in a collisionless plasma considered by Landau [23]. Landau showed that an initial perturbation of the electric field E in plasma oscillates in time with plasma frequency $\omega_{\rm pl}$ and attenuates *exponentially* even in the absence of collisions. The same problem in a BCS superconductor leads to a different result. In the absence of inelastic collisions, homogeneous perturbations of the order parameter $\delta\Delta(t)$ in superconductors oscillate with a frequency of 2Δ, and also decay in time as follows:

$$\delta\Delta(t) \sim \delta\Delta_{\rm exc}\frac{\cos(2\Delta t + \theta_0)}{\sqrt{2\Delta t}} + \delta\Delta(\infty), \tag{1}$$

where $\delta\Delta(\infty)/\Delta_0 \neq \delta\Delta(0)$, i.e., $\delta\Delta(t)$ approaches not the initial value $\delta\Delta(0)$ but some steady (smaller) value. Here, the order parameter Δ is expressed as an integral over momenta p or energies ϵ of the distribution function [the condensate wave function $f(\epsilon)$]. The decay rate in Eq. (1) stems from the branch points of singularities in the integral from the function $f(\epsilon)$, while the exponential attenuation of E in plasma is due to the pole of the function $n(\xi_p)$. The temporal behavior of $\delta\Delta(t)$ stemming from the branch point at $\epsilon = 2\Delta$ was discussed by different methods for various systems not only in Refs. [22,24–30] for BCS s-wave superconductors, but also for neutral fermionic superfluids [31–34] or the BCS-BEC (Bose-Einstein condensate) crossover regime [35–41], as well as in proximitized structures [42,43].

The analogy between collisionless superconductors and plasma turned out to be even deeper; both systems belong to the class of fully integrable systems (in the absence of collisions). In these systems, information about oscillations is conserved in electrons and is lost in the order parameter $\delta\Delta(t)$ (in superconductors) or in the electric field E (in plasma). Such a memory may lead to echo effects [44]. The analogy with plasma was noted already in Refs. [22,45]. In particular, it was emphasized in Ref. [45] that contrary to $\delta\Delta(t)$ the condensate wave function $f(\epsilon, t)$ experiences nondecaying oscillations. Further development of plasma theory has shown that undamped plasma waves can also be excited in plasma if the initial conditions are chosen appropriately [44], and similar ideas have been further developed for superconductors [25,26,31,46] and superfluids [33,35–40].

Experimental observation of the $\delta\Delta(t)$ dynamics is a rather difficult task since inelastic scattering times are short ($\tau_{\text{in}} \sim 10^{-9}-10^{-12}$ s) and in the linear approximation the amplitude mode does not directly couple with the electromagnetic field. Historically, the amplitude mode was first revealed in the Raman scattering in a charge-density-wave superconductor $NbSe_2$ in 1981 [47,48] (see also recent works and references therein [49]). In addition, early studies of superconducting properties under continuous irradiation with subgap energies using microwaves already showed surprising results that superconductivity could be observed above the equilibrium critical temperature [50–52]. These observations were explained by Eliashberg, who showed that in the vicinity of the critical temperature, the effect is mainly related to a nonequilibrium distribution of quasiparticles induced by the microwave field [53,54].

Furthermore, recent advances in the technical development of THz spectroscopy enabled the construction of THz-pump THz-probe spectroscopy in a conventional s-wave superconductor such as NbN and $Nb_{1-x}Ti_xN$ thin films, which reveal novel experimental observations. For example, transient gap enhancement, suggesting an additional Eliashberg effect present at temperatures close to T_c, was observed in NbN [55,56].

Further studies concerned the observation of the collective modes followed [57,58]. The idea of this experimental setup is that a single-cycle (pump) pulse with an energy on the order of Δ excites the amplitude mode in the superconducting condensate nonadiabatically. This leads to an amplitude oscillation of the order parameter Δ with the frequency $\Omega_h = 2\Delta$. These oscillations can be detected with a second, weaker pulse that comes with a delay δt and probes these dynamics. The transmitted electric fields can be measured using electro-optic probing [58]. The probe pulse is applied with an orthogonal polarization to the pump pulse. Thus, the pump pulse can be filtered out from the probe pulse using a wire grid polarizer [59].

While a nonadiabatic quench with a single-cycle pulse on the picosecond scale is an experimental challenge, one can consider driving the system periodically with a multicycle pulse of frequency Ω. Since the amplitude mode couples to light in the second order, it is driven at twice the driving frequency 2Ω. The driven oscillation can then be measured as an induced current. Since the electromagnetic vector potential oscillates with the driving frequency $\mathbf{A}(t) \sim e^{i\Omega t}$ and the amplitude mode oscillates with twice the driving frequency $\delta\Delta_H(t) \sim e^{2i\Omega t}$, one obtains a total current, which oscillates with three times the driving frequency $\mathbf{j}(t) \sim e^{3i\Omega t}$ [46,60]. This generation of a third harmonic is a process, which only occurs inside the superconducting state at $T < T_c$ and should be resonantly enhanced once 2Ω activates the amplitude mode at $2\Omega = \Omega_H = 2\Delta$.

In Ref. [58], the first successful measurement of the third-harmonic generation in the conventional s-wave superconductor $Nb_{1-x}Ti_xN$ was reported. However, soon after it was realized that the activation of the amplitude mode is not the only process measured in the current produced by the third-harmonic generation. Since the frequency of the amplitude mode is given at $\Omega_H = 2\Delta$, it coincides with the onset of the particle-hole generation and with the energy needed to break the Cooper pairs. In particular, Ref. [61] has shown that in the clean BCS superconductor, the contribution to the third-harmonic generation current caused by a direct action of an ac electric field (sometimes called charge density fluctuations, which is not completely adequate) is three orders of magnitude larger than that from the amplitude mode. Subsequent studies analyzed how impurities, realistic electron-phonon coupling, and strong-coupling features affect this ratio [62–68]. Obviously, this shows that research on the detection of the collective modes and their behavior in nonequilibrium in conventional and unconventional superconductors due to a driving field is still ongoing. Furthermore, several works were devoted to the study of possibilities to detect the amplitude mode from the nonlinear response of superconductor irradiated terahertz electromagnetic fields. One of these methods is to measure the ac conductance of a current-carrying superconductor $\sigma(\Omega)$ [8–10]. In this case, the electromagnetic field is coupled to the AM, and the ac conductance $\sigma(\Omega)$ contains a term related to the excited amplitude mode (Higgs mode) [8].

If a superconductor is suddenly brought out of equilibrium and the order parameter $\delta\Delta(t)$ oscillates, as follows from Eq. (1), the behavior of the system resembles the behavior of a generator with a resonant frequency of 2Δ. It is therefore quite natural to suppose that this system might be used as a parametric oscillator. The authors of Refs. [69,70], on the basis of a phenomenological model, analyzed the possibility of parametric amplification, but microscopic analysis is still lacking. Most importantly, the idea of parametric amplification has been used among others to explain the highly interesting observation in high-T_c cuprates [71–76], K_3C_{60} [69,77–80], and certain organic superconductors [69,81]. In particular, the intense far-infrared optical pulses have been shown to create nonequilibrium coherent states with optical properties that are consistent with the transient photoinduced superconductivity phenomenon, yet the nature of this state is still debated [82–84].

In this manuscript, we calculate within a microscopic quasiclassical description the response of a superconductor $\delta\Delta(t)$ in the second order to the action of a monochromatic irradiation or of many signals with different Ω following the experimental setup, presented in Fig. 1. In particular, we consider the superconducting sample, which is continuously irradiated by the monochromatic THz pulse $\vec{E}_{\text{THZ}}[\Omega \sim \Delta(T)]$. In the second order of the field intensity $|Q_\Omega| = eE_\Omega/\Omega$, we obtain general formulas for the quasiclassical Green's function $f(\epsilon, t)$. From these we find the variation $\delta\bar{\Delta}$ averaged in time (Eliashberg effect), the variation $\delta\Delta_{2\Omega}$ at the

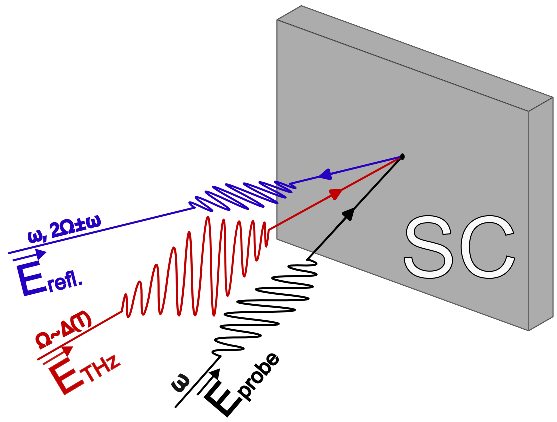

FIG. 1. Schematic representation of the system under consideration. We consider the superconducting sample, which is continuously irradiated by the monochromatic THz pulse $\vec{E}_{\rm THZ}[\Omega \sim \Delta(T)]$. At the same time, the probe field is applied with orthogonal polarization $\vec{E}_{\rm probe}(\omega)$. The reflected electromagnetic field $\vec{E}_{\rm refl}$ is measured either for the probe field frequency, ω, or at $2\Omega \pm \omega$ to reveal the parametric effects.

doubled frequency of a monochromatic ac field. We also calculate the third harmonic of the current $I_{3\Omega}$ and the response I_p on the action of an ac field with a frequency Ω_p. Here we assume that the probe field is applied with orthogonal polarization $\vec{E}_{\rm probe}[\omega < 2\Delta(T)]$. The reflected electromagnetic field $\vec{E}_{\rm refl}$ is measured either for the probe field frequency, ω, or at $2\Omega \pm \omega$ to reveal the parametric gain (amplification) conditions and a change in the ac conductance in the presence of a pump irradiation. Note, unlike the recent work [85], we do not study a transient behavior of the superconductor irradiated by ac pulses, and we assume that ac irradiation is continuous in time.

II. GENERAL EQUATIONS

To model a realistic experimental situation, we consider diffusive superconductors ($\tau_{\rm el}T_c \ll 1$). To describe nonstationary and nonlinear response of an s-wave conventional superconductor, we will use a well-established theory for matrix quasiclassical Green's functions $\hat{g}$ [86–90] (for review, see also [91–93]). The equation for the retarded $g^{R(A)}(t,t')$ function has the form [we dropped the superindex $R(A)$]

$$i(\hat{\tau}_3\partial_t\hat{g} + \partial_{t'}\hat{g}\hat{\tau}_3) + [\hat{\Delta},\hat{g}] - iD\partial(\hat{g}(\partial\hat{g})) = 0, \quad (2)$$

where $[\hat{\Delta},\hat{g}] = \hat{\Delta}(t)\hat{g}(t,t') - \hat{g}(t,t')\hat{\Delta}(t')$. In the presence of a gauge-invariant "momentum" $\mathbf{Q} = \nabla\chi/2 - (e/c)\mathbf{A}$, related to the condensate velocity $v_S = Q/m$, Eq. (2) acquires the form

$$i(\hat{\tau}_3\partial_t\hat{g} + \partial_{t'}\hat{g}\hat{\tau}_3) + [\hat{\Delta},\hat{g}] = -iD[\hat{Q}\hat{g}\hat{Q},\hat{g}]. \quad (3)$$

Here $\hat{Q} = Q\hat{\tau}_3$ and

$$[Q\hat{\tau}_3\hat{g}Q\hat{\tau}_3,\hat{g}] = \int dt_1\{\hat{Q}(t)\hat{g}(t,t_1)\hat{Q}(t_1)\hat{g}(t_1,t') - \hat{g}(t,t_1)\hat{Q}(t_1)\hat{g}(t_1,t')\hat{Q}(t')\}. \quad (4)$$

In the considered spatially uniform case, one can choose the phase χ equal to zero so that the electric fields $\mathbf{E}$ is related to $\mathbf{Q}$ via $\mathbf{E} = -(1/c)\partial_t\mathbf{A} = \partial_t\mathbf{Q}/e$. We consider the case when momentum $Q(t)$ consists of a sum of periodic functions

$$Q(t) = \sum_{\pm(\nu,\mu)}[Q_\nu\exp(i\Omega_\nu t) + Q_\mu\exp(i\Omega_\mu t)] \quad (5)$$

with arbitrary frequencies $\Omega_{\nu,\mu}$. Because $Q(t)$ is a real function, we require that $Q_{\nu,\mu} = Q_{-\nu,-\mu}$. In particular, the frequency Ω_ν can be equal to Ω_μ: $\Omega_\nu = \Omega_\mu = \Omega$ (monochromatic irradiation). For the Fourier components $\hat{g}_{\epsilon,\epsilon'} = \int dtdt'\hat{g}(t,t')\exp(i\epsilon t - i\epsilon' t')$, Eq. (3) becomes (see, for example, [8,19,64])

$$\epsilon\hat{\tau}_3\hat{g} - \hat{g}\hat{\tau}_3\epsilon' + [\hat{\Delta},\hat{g}]_{\epsilon,\epsilon'} = -iD[Q\hat{\tau}_3\hat{g}Q\hat{\tau}_3,\hat{g}]_{\epsilon,\epsilon'}. \quad (6)$$

The right-hand side can be written as

$$iD[Q\hat{\tau}_3\hat{g}Q\hat{\tau}_3,\hat{g}]_{\epsilon,\epsilon'} = iD\sum_{\nu\mu}\{\hat{Q}_\nu\hat{g}_{\epsilon+\Omega_\nu}\hat{Q}_\mu\hat{g}_{\epsilon'} - \hat{g}_\epsilon\hat{Q}_\nu\hat{g}_{\epsilon'-\Omega_\mu}\hat{Q}_\mu\}2\pi\delta(\epsilon_- + \Omega_{\nu+\mu}) \quad (7)$$

with $\epsilon_- = \epsilon - \epsilon'$ and $\Omega_{\nu+\mu} \equiv \Omega_\nu + \Omega_\mu$.

Consider first the unperturbed ground state. The retarded (advanced) Green's functions in the ground state have a standard form,

$$\hat{g}_0^{R(A)} = g(\epsilon)\hat{\tau}_3 + i\hat{\tau}_2 f(\epsilon)|^{R(A)}, \quad (8)$$

$$g_0^{R(A)}(\epsilon) = (\epsilon \pm i\gamma)/\zeta^{R(A)}, \quad f(\epsilon) = \Delta/\zeta^{R(A)}, \quad (9)$$

where

$$\zeta^{R(A)}(\epsilon) = \begin{cases} \pm\mathrm{sgn}(\epsilon)\sqrt{(\epsilon \pm i\gamma)^2 - \Delta^2}, & |\epsilon| > \Delta, \\ i\sqrt{\Delta^2 - \epsilon^2}, & |\epsilon| < \Delta. \end{cases} \quad (10)$$

Here γ is a phenomenological damping coefficient introduced by Dynes *et al.* [94,95] which is assumed to be small ($\gamma \ll \Delta$). One can see that $\zeta^A(\epsilon) = \zeta^R(-\epsilon)$ and $\zeta^A(\epsilon) = -[\zeta^R(\epsilon)]^*$. At the next step, we consider the corrections to Δ_0 and to $\hat{g}_0^{R(A)}$ due to ac perturbations.

III. ACTION OF THE ac FIELDS

In a nonequilibrium or nonstationary case, the system is described by a matrix $\check{g}$ whose elements are the retarded (advanced), $\hat{g}^{R(A)}$, and the so-called Keldysh matrix function $\hat{g}^K$. The supermatrix Green's function $\check{g}$ is defined as

$$\check{g} = \begin{Bmatrix} \hat{g}^R, \hat{g}^K \\ 0, \hat{g}^A \end{Bmatrix}, \quad (11)$$

where $\hat{g}^{R(A)}$ are the retarded (advanced) Green's functions and the Keldysh Green's function $\hat{g}^K$ [96]. The latter matrix Green's function is expressed in terms of the matrix distribution function $\hat{n}$ [89–93],

$$\hat{g}^K = \hat{g}^R\hat{n} - \hat{n}\hat{g}^A, \quad (12)$$

and the matrix $\check{g}$ obeys the normalization condition

$$\check{g}\cdot\check{g} = \check{1}. \quad (13)$$

The correction $\delta\check{g}(\epsilon,\epsilon') = \check{g}(\epsilon,\epsilon') - \check{g}_0(\epsilon)2\pi\delta(\epsilon-\epsilon')$ satisfies the linearized Eq. (6), which acquires the form

$$(\zeta_\epsilon \check{g}_\epsilon)\delta\check{g} - \delta\check{g}(\check{g}_{\epsilon'}\zeta_{\epsilon'}) = \sum_{\nu,\mu}[\check{R}_\Delta(\epsilon,\epsilon') + \check{R}_Q(\epsilon,\epsilon')]2\pi\delta(\epsilon_- + \Omega_{\nu+\mu}), \quad (14)$$

where the right-hand side contains only the Green's functions in the ground state, and the matrix elements of the 4×4 matrices $\check{g}_\epsilon$ are $\hat{g}^{R(A)}_\epsilon = \hat{g}^{R(A}_0(\epsilon)$, $\hat{g}^K_0 = [\hat{g}^R_0(\epsilon) - \hat{g}^A_0(\epsilon)]t_\epsilon$ with $t_\epsilon = \tanh(\epsilon\beta)$ and $\beta = (2T)^{-1}$. The supermatrix $(\zeta_\epsilon\check{g}_\epsilon)$ is given by

$$(\zeta_\epsilon\check{g}_\epsilon) = \begin{Bmatrix} \zeta^R_\epsilon \hat{g}^R_\epsilon & 0 \\ 0 & \zeta^A_\epsilon \hat{g}^A_\epsilon \end{Bmatrix}. \quad (15)$$

The matrices $\check{R}_\Delta(\epsilon,\epsilon')$, $\check{R}_Q(\epsilon,\epsilon')$ are defined as

$$\check{R}_\Delta(\epsilon,\epsilon') = \check{g}_\epsilon\delta\check{\Delta}_\Omega - \delta\check{\Delta}_\Omega\check{g}_{\epsilon'}, \quad (16)$$

$$\check{R}_Q(\epsilon,\epsilon') = iD\{\check{g}_\epsilon\check{Q}_\nu\check{g}_{\epsilon+\Omega_\nu}\check{Q}_\mu - \check{Q}_\nu\check{g}_{\epsilon+\Omega_\nu}\check{Q}_\mu\check{g}_{\epsilon'}\}, \quad (17)$$

where $\check{Q}_\nu = Q_\nu\check{\tau}_3$ with

$$\check{\tau}_i = \begin{pmatrix} \hat{\tau}_i & 0 \\ 0 & \hat{\tau}_i \end{pmatrix}. \quad (18)$$

The order parameter $\check{\Delta}(t) = \Delta(t)i\check{\tau}_2$ is related to the matrix $\hat{g}^K$ via

$$\Delta(t) = \lambda \mathrm{Tr}\int \frac{d\epsilon d\epsilon'}{(2\pi)^2}(-i\hat{\tau}_2)\hat{g}^K(\epsilon,\epsilon')\exp(-i\epsilon_- t). \quad (19)$$

Let us consider first the retarded (advanced) function. The normalization condition, Eq. (13), yields

$$(\hat{g}_\epsilon\delta\hat{g} + \delta\hat{g}\hat{g}_{\epsilon'})^{R(A)} = 0. \quad (20)$$

Using Eq. (20), we obtain from Eq. (14)

$$\delta\hat{g}^{R(A)}(\epsilon,\epsilon') = \sum_{\nu,\mu}\left\{\frac{\hat{\varrho}_\Delta + \hat{\varrho}_Q}{C_{\epsilon,\epsilon'}}\right\}^{R(A)} 2\pi\delta(\epsilon_- + \Omega_{\nu+\mu}), \quad (21)$$

where $\hat{\varrho} = \hat{g}_\epsilon\hat{R}$ and $C^{R(A)}_{\epsilon,\epsilon'} = (\zeta_\epsilon + \zeta_{\epsilon'})^{R(A)}$. The correction $\delta\hat{g}^K$ to the Keldysh function can be represented as a sum of regular and anomalous parts [4],

$$\delta\hat{g}^K = \delta\hat{g}^{\mathrm{reg}} + \delta\hat{g}^{\mathrm{an}}. \quad (22)$$

The regular part is defined as

$$\delta\hat{g}^{\mathrm{reg}} = \delta\hat{g}^R t_{\epsilon'} - t_\epsilon\delta\hat{g}^A, \quad (23)$$

with the matrices $\delta\hat{g}^{R(A)}$ given by Eq. (21). When studying the response of a superconductor to ac radiation, the authors of many works used the Matsubara frequency representation. The transition to real energies is carried out with the help of an analytical continuation. In our method, we do not use the Matsubara representation and the analytic continuation.

The key point of this approach is to split the Keldysh Green's function $\hat{g}^K$ into a regular $\hat{g}^{\mathrm{reg}}$ and an anomalous $\hat{g}^{\mathrm{an}}$ part [4]. The method of analytical continuation was used, for example, in a recent paper [64] (see also Kopnin's book [93] and references therein), while the former was applied in Refs. [8,19]. For the anomalous Green's function $\delta\hat{g}^{\mathrm{an}}$ we find (see the Appendix)

$$\delta\hat{g}^{\mathrm{an}} = \sum_{\nu,\mu}\frac{\hat{\varrho}^{\mathrm{an}}_\Delta + \hat{\varrho}^{\mathrm{an}}_Q}{C^{\mathrm{an}}_{\epsilon,\epsilon'}}2\pi\delta(\epsilon_- + \Omega_{\nu+\mu}) \quad (24)$$

with $C^{\mathrm{an}}_{\epsilon,\epsilon'} = \zeta^R_\epsilon + \zeta^A_{\epsilon'}$. The matrices $\hat{\varrho}^{\mathrm{an}}_\Delta = \hat{g}^R\hat{R}^{\mathrm{an}}_\Delta$ and $\hat{\varrho}^{\mathrm{an}}_Q = \hat{g}^R\hat{R}^{\mathrm{an}}_Q$ are defined as follows:

$$\hat{\varrho}^{\mathrm{an}}_\Delta = \left(\hat{g}^R_\epsilon\delta\hat{\Delta}_\Omega\hat{g}^A_{\epsilon'} - \delta\hat{\Delta}_\Omega\right)(t_{\epsilon'} - t_\epsilon), \quad (25)$$

$$\begin{aligned}\hat{\varrho}^{\mathrm{an}}_Q = {}& iD\left[\hat{g}^R_\epsilon\hat{Q}_\nu\hat{g}^R_{\epsilon+\Omega_\nu}\hat{Q}_\mu\hat{g}^A_{\epsilon'} - \hat{Q}_\nu\hat{g}^R_{\epsilon+\Omega_\nu}\hat{Q}_\mu\right](t_{\epsilon'} - t_{\epsilon+\Omega_\nu}) \\ &- iD\left[\hat{g}^R_\epsilon\hat{Q}_\nu\hat{g}^A_{\epsilon+\Omega_\nu}\hat{Q}_\mu\hat{g}^A_{\epsilon'} - \hat{Q}_\nu\hat{g}^A_{\epsilon+\Omega_\nu}\hat{Q}_\mu\right](t_\epsilon - t_{\epsilon+\Omega_\nu}),\end{aligned} \quad (26)$$

where $\epsilon' = \epsilon + \Omega_{\nu+\mu}$. The matrices $g^{R(A)}_\epsilon = g^{R(A)}_0(\epsilon)$ are the Green's functions in the ground state [see Eqs. (8) and (9)].

Equations (22)–(25) represent one of the main results of our derivation. Using these equations together with the self-consistency equation (19), one can now find the function $\delta\hat{\Delta}(t)$ in the second order in $Q_{\nu,\mu}$. The component $\delta\Delta_{\nu\mu}(t)$ at the frequency $\Omega_{\nu+\mu}$ is equal to

$$\begin{aligned}\delta\Delta_{\nu+\mu}(t) = {}& \tfrac{1}{2}[\delta\Delta(\Omega_{\nu+\mu})\exp(i\Omega_{\nu+\mu}t) \\ &+ \delta\Delta(-\Omega_{\nu+\mu})\exp(-i\Omega_{\nu+\mu}t)].\end{aligned} \quad (27)$$

As to the regular part δg^{reg}, the integral in Eq. (19) can be transformed into a sum over Matsubara frequencies [$\epsilon \to i\epsilon_n$, $\epsilon_n = \pi T(2n+1)$] because the retarded (advanced) Green's functions $\delta g^{R(A)}$ are analytical functions in the upper (lower) half-plane, and the singular points of the regular part δg^{reg} are determined only by poles of the functions $t_\epsilon = \tanh(\epsilon\beta)$ and $t_{\epsilon'} = \tanh(\epsilon'\beta)$, i.e., $\epsilon' = i\epsilon_n + \Omega_{\nu+\mu}$ and $\epsilon = i\epsilon_n$ with $\epsilon_n = \pi T(2n+1)$. The integral in Eq. (19) from the anomalous part cannot be reduced to a sum over Matsubara frequencies and should be calculated explicitly.

IV. MONOCHROMATIC IRRADIATION

In the case of a monochromatic ac field, it is of interest to calculate the Fourier components of the Δ variations: $\delta\Delta_0$ and $\delta\Delta_{2\Omega}$. This means that the following terms should be extracted from the sum over frequencies $\Omega_{\nu,\mu}$:

($\mathcal{A}$) $\Omega_\nu \equiv \Omega = -\Omega_\mu$, so that $\Omega_{\nu+\mu} \equiv \Omega_\nu + \Omega_\mu = 0$; and

($\mathcal{B}$) $\Omega_\nu = \Omega = \Omega_\mu$, so that $\Omega_{\nu+\mu} \equiv \Omega_\nu + \Omega_\mu = 2\Omega$.

Also the zero Fourier component $\delta\Delta_0$ describes the time-averaged change of $\delta\Delta(t)$ under the influence of an electromagnetic radiation. The component $\delta\Delta_{2\Omega}$ is the magnitude of the second harmonic of the amplitude mode excited by the irradiation $Q(t) = Q_\Omega\cos(\Omega t)$. It contributes to the third harmonic of the current.

Consider first the case $\mathcal{A}$: ($\Omega_{\nu+\mu} = 0$). In this case, the most interesting quantity is a time-averaged variation of the order parameter, $\langle\delta\Delta(t)\rangle_t = \delta\Delta_0$, which we discuss in detail below.

A. Eliashberg effect

In particular, Eliashberg showed that a microwave irradiation under certain conditions can enhance the order parameter

Δ as well as the critical temperature T_c [53]. This effect as well as the enhancement of the critical current was studied in more detail in subsequent works [54,97–100]. Stability of the nonequilibrium state with an enhanced $\Delta = \Delta_{\rm eq} + \delta\Delta_0$ was investigated in Ref. [101]. Recently, the authors of Refs. [102,103] analyzed, in particular, the region of the enhanced Δ and of the enhanced critical current j_c and plotted this region in the plane (T, Ω). The predicted effect was observed experimentally, although it was not found to be as strong as expected (see the review by Klapwijk and Visser [104] and references therein).

Since in the considered case $\Omega_{\nu+\mu} = 0$ we have $\epsilon = \epsilon'$ and the regular part is given by Eq. (23) with $t_\epsilon \equiv \tanh(\epsilon\beta) = t_{\epsilon'}$,

$$\delta\hat{g}^{\rm reg} = t_\epsilon(\delta\hat{g}^R - \delta\hat{g}^A), \tag{28}$$

where the matrices $\delta\hat{g}^{R(A)}$ are given by Eq. (21) with $\epsilon = \epsilon'$. The matrices $\delta\hat{g}^{R(A)}$ can be represented in the form

$$\delta\hat{g}^{R(A)} = \frac{\pi}{\zeta_\epsilon}[\hat{\tau}_3(\delta g_\Delta + \delta g_Q) + i\hat{\tau}_2(\delta f_\Delta + \delta f_Q)]^{R(A)}\delta(\epsilon_-). \tag{29}$$

Here the coefficients A and B are [for clarity, indices $R(A)$ are omitted]

$$\delta g_\Delta = \delta\Delta B_{\epsilon,\epsilon}, \delta f_\Delta = \delta\Delta A^{(+)}_{\epsilon,\epsilon}, \tag{30}$$

$$\delta g_Q = iDQ^2(A^{(-)}_{\epsilon,\epsilon} g_{\epsilon+\Omega} - B_{\epsilon,\epsilon} f_{\epsilon+\Omega}), \tag{31}$$

$$\delta f_Q = -iDQ^2(A^{(+)}_{\epsilon,\epsilon} f_{\epsilon+\Omega} + B_{\epsilon,\epsilon} g_{\epsilon+\Omega}). \tag{32}$$

The coefficients $A^{(\pm)}$ and B are defined as

$$A^{(\pm)}_{\epsilon,\epsilon'} = 1 \pm (g_\epsilon g_{\epsilon'} + f_\epsilon f_{\epsilon'}), \tag{33}$$

$$B_{\epsilon,\epsilon'} = g_\epsilon f_{\epsilon'} + f_\epsilon g_{\epsilon'}, \tag{34}$$

$$C_{\epsilon,\epsilon'} = \zeta_\epsilon + \zeta_{\epsilon'}, \tag{35}$$

with $\epsilon = \epsilon'$ in this particular case. The anomalous function $\delta\hat{g}^{\rm an}$ is given by Eq. (24) with $\hat{\varrho}^{\rm an}_\Delta = 0$ and $\delta f^{\rm an}_Q(\epsilon, \epsilon')$ equal to

$$\delta f^{\rm an}_Q(\epsilon, \epsilon') = -iDQ^2(t_{\epsilon+\Omega} - t_\epsilon) \times \frac{(f^R_{\epsilon+\Omega} - f^A_{\epsilon+\Omega})A^{\rm an(+)}_{\epsilon,\epsilon} + (g^R_{\epsilon+\Omega} - g^A_{\epsilon+\Omega})B^{\rm an}_{\epsilon,\epsilon}}{C^{\rm an}_{\epsilon,\epsilon}}, \tag{36}$$

with $A^{\rm an(\pm)}_{\epsilon,\epsilon'} = 1 \pm (g^R_\epsilon g^A_{\epsilon'} + f^R_\epsilon f^A_{\epsilon'})$, $B^{\rm an}_{\epsilon,\epsilon'} = g^R_\epsilon f^A_{\epsilon'} + f^R_\epsilon g^A_{\epsilon'}$, $C^{\rm an}_{\epsilon,\epsilon'} = \zeta^R_\epsilon + \zeta^A_{\epsilon'}$, and $\epsilon' = \epsilon$.

Let us now find the variation of Δ. In the considered case, the self-consistency equation given by Eq. (19) is

$$\delta\Delta(t) = -i\lambda\int\frac{d\bar{\epsilon}}{2\pi}\int\frac{d\epsilon_-}{2\pi}{\rm Tr}\hat{\tau}_2\{\delta\hat{g}^{\rm reg} + \delta\hat{g}^{\rm an}\}\exp(i\epsilon_- t). \tag{37}$$

For the $\delta\Delta(t)$ averaged over time, we obtain

$$\delta\Delta = -i\lambda\int\frac{d\epsilon}{2\pi}{\rm Tr}\hat{\tau}_2\{\delta\hat{g}^{\rm reg} + \delta\hat{g}^{\rm an}\}|_{\epsilon_-=0}. \tag{38}$$

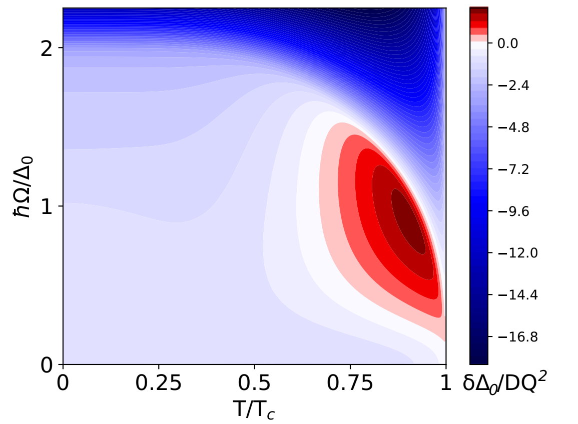

FIG. 2. Calculated time-averaged gap correction $\delta\Delta_0$ using Eq. (39) for $\gamma = 0.02\Delta_0$. Note, we set the polarization operator given by the factor of the left-hand side to 1, as it is solely positive and real. The area in red represents the area of stimulated (enhanced) superconductivity. Furthermore, the crossover boundary from light blue to dark blue in the figure mimics the superconducting gap temperature dependence. In particular, if one applies a pump-field with frequency Ω above 2Δ, direct pair-breaking processes are promoted resulting in a strong suppression of the gap. Thus, the positive region shows a time-independent increase in the order parameter, while the light and dark blue areas show a decrease/suppression of the order parameter.

This equation can be written in the form (see the Appendix)

$$4T\delta\Delta_0\Delta^2\sum_n\frac{1}{\zeta_n^3} = -DQ^2\left[T\Delta{\rm Re}\sum_n\frac{\omega(2\omega + i\Omega)}{\zeta^3_\omega\zeta_{\omega+i\Omega}} + \int\frac{d\epsilon}{2\pi}\delta f^{\rm an}_Q\right]. \tag{39}$$

In Fig. 2 we plot the dependence of $\delta\Delta_0$ (in the normalized form $\delta\Delta_0/DQ^2$) as a function of Ω and T for $\gamma = 0.02\Delta_0$, and Δ_0 is the value of the equilibrium gap at $T = 0$. Note $\delta\Delta_0/DQ^2$ does not depend on the probe-field frequency as the probe and the pump fields are assumed to be orthogonal to each other. Further, we set the factor in front of $\delta\Delta_0$ on the left-hand side of Eq. (39) to 1 as it is always positive and therefore has no influence on the area of enhancement. We also show the region in the (T, Ω) plane, where $\delta\Delta_0$ is positive, i.e., one finds a stimulation of superconductivity, especially in the vicinity of the superconducting transition temperature. Furthermore, smaller γ enlarges the region of stimulated superconductivity. One finds also a strong suppression of superconducting gap for $\Omega > 2\Delta(T)$. The results are consistent with those obtained previously in Refs. [102,103].

B. Second harmonic of $\delta\Delta$

Let us now consider the case $\mathcal{B}$ and set $\epsilon' = \epsilon + 2\Omega$. The variation of Δ can be represented as follows:

$$\delta\Delta_{2\Omega} = \frac{\delta\Delta^{\rm reg}_Q(2\Omega) + \delta\Delta^{\rm an}_Q(2\Omega)}{P^{\rm reg}_\Delta(2\Omega) + P^{\rm an}_\Delta(2\Omega)}. \tag{40}$$

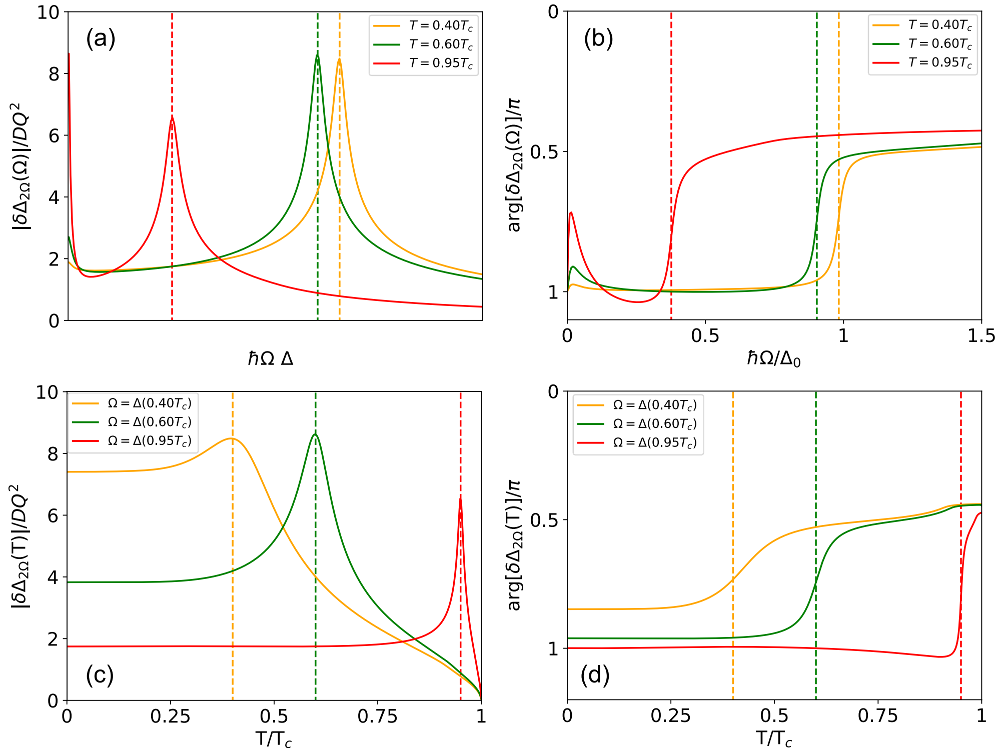

FIG. 3. Calculated frequency dependence of the amplitude (a) and the phase (b) of the gap oscillation $\delta\Delta_{2\Omega}$. Panels (c) and (d) show a corresponding temperature dependence of the gap oscillation $\delta\Delta_{2\Omega}$. We observe a peak accompanied by a phase shift at the resonance condition $\Omega = \Delta(T)$, which is highlighted by the dashed lines. The inset in (d) shows the equilibrium gap $\Delta(T)$ as a function of the temperature.

Here,

$$P_\Delta^{\rm an}(2\Omega) = -2\sinh(2\Omega\beta)\int \frac{d\epsilon}{2\pi} r_\epsilon \frac{A^{\rm an(+)}_{\epsilon,\epsilon+2\Omega}}{C^{\rm an}_{\epsilon,\epsilon+2\Omega}}, \tag{41}$$

$$P_\Delta^{\rm reg}(2\Omega) = 4T\,{\rm Re}\sum_n \left(\frac{A^{(+)}_{n,n+i2\Omega}}{C_{n,n+i2\Omega}} - \frac{1}{\zeta_n}\right), \tag{42}$$

where $\zeta_n = \sqrt{\epsilon_n^2 + \Delta^2}$ and the subindex n in $A_n^{(\pm)}$, B_n, and C_n denotes that the functions are expressed in terms of the Matsubara frequencies $\epsilon = i\epsilon_n = i\pi T(2n+1)$. The terms in the numerator are

$$\delta\Delta_Q^{\rm reg} = 2TDQ^2{\rm Re}\sum_{n\geqslant 0}\frac{f_{n+i\Omega}A^{(+)}_{n,n+i2\Omega} + g_{n+i\Omega}B_{n,n+i2\Omega}}{\zeta_n + \zeta_{n+i2\Omega}} \tag{43}$$

and

$$\begin{aligned}\delta\Delta_Q^{\rm an} = &- iDQ^2\sinh(2\Omega\beta)\int\frac{d\epsilon}{2\pi}\frac{r_\epsilon}{C^{an}_{\epsilon_-,\epsilon_+}}\\ &\times \big(A^{\rm an(+)}_{\epsilon_-,\epsilon_+}[F^{(+)} - F^{(-)}\tanh(\epsilon\beta)\tanh(\Omega\beta)]\\ &+ B^{\rm an}_{\epsilon_-,\epsilon_+}[G^{(+)} - G^{(-)}\tanh(\epsilon\beta)\tanh(\Omega\beta)]\big).\end{aligned} \tag{44}$$

The temperature factor $r(\epsilon,\Omega)$ is defined as

$$r_\epsilon = [\cosh(\epsilon_+\beta)\cosh(\epsilon_-\beta)]^{-1}, \tag{45}$$

where $\epsilon_\pm = \epsilon \pm \Omega$. The functions $F^{(\pm)}$, $G^{(\pm)}$ are given by

$$F^{(\pm)} = f_\epsilon^R \pm f_\epsilon^A,\; G^{(\pm)} = g_\epsilon^R \pm g_\epsilon^A. \tag{46}$$

At low frequencies, $\Omega \Rightarrow 0$, we obtain $\delta\Delta_Q^{\rm an} \Rightarrow 0$, and $\delta\Delta_Q^{\rm reg}$ has the same form as in a static case,

$$\frac{\delta\Delta_Q^{\rm reg}}{P_\Delta^{\rm reg}} = -DQ^2\frac{\sum_{n\geqslant 0}\frac{\epsilon_n^2}{\zeta_n^4}}{\sum_{n\geqslant 0}\frac{\Delta}{\zeta_n^3}}. \tag{47}$$

In Fig. 3 we plot the frequency dependence of the normalized variation of the second harmonic of Δ, $\delta\tilde{\Delta}_{2\Omega} \equiv \delta\Delta_{2\Omega}/DQ^2$ for $\gamma = 0.02\Delta_0$. As expected, the amplitude mode gives a resonant contribution around the corresponding value of $\Delta(T)$ with a characteristic phase shift of $\pi/2$. For the frequency-resolved second harmonic of $\delta\Delta_{2\Omega}$, we find a second peak at $\Omega \to 0$ for higher temperatures. This peak stems from the vanishing anomalous part of the polarization operator $P_\Delta^{\rm an}$ at these temperatures in the presence of disorder and a pump as shown in the Appendix. These peaks are never divergent as the regular part of both $\delta\Delta$ and the polarization operator are finite below T_c. Similar behavior was found previously in Ref. [64].

V. THE CURRENT INDUCED BY AN ac FIELD

To investigate the induced current, we consider an ac electric field incident on a superconducting film of thickness $2d$. The field consists of probe E_ω and pump E_Ω field components: $E(t) = E_\omega\cos(\omega t) + E_\Omega\cos(\Omega t)$. This field excites alternating currents $j(t)$ of various harmonics. In the third order in the amplitude E, the induced current consists of harmonics with frequencies ω, and $2\Omega \pm \omega$.

The current $j(t)$ is determined by

$$j(t) = i\frac{\sigma_0}{4e}\mathrm{Tr}\bigg[\hat{\tau}_3 \int d\tau\{\hat{g}([\hat{\mathbf{Q}},\hat{g}]\exp(i\Omega\tau) + [\hat{\mathbf{q}},\hat{g}]\exp(i\omega\tau))\}_3^K\bigg], \tag{48}$$

where $\hat{\mathbf{Q}} = \mathbf{Q}\hat{\tau}_3$, the momentum $\mathbf{Q}$ is the gauge-invariant quantity related to the phase of the superconductor χ, and the vector potential $\mathbf{A}$: $\mathbf{Q} = \nabla\chi - (e/c)\mathbf{A}$. In the case of $\chi = 0$, the vector potential $\mathbf{A}$ relates to the electric field as

$$\mathbf{E}_Q(t) = \frac{i\Omega}{e}\mathbf{Q}, \quad \mathbf{E}_q(t) = \frac{i\omega}{e}\mathbf{q}, \tag{49}$$

where ω and Ω is a probe (signal) and a pump frequency. These frequencies may in general coincide: $\omega = \Omega$.

We decompose all functions to the power of $\mathbf{Q}$ considering the quantity $DQ^2/\Delta(T)$ as a small parameter. In the lowest order, we obtain a linear response to the action of two signals with Ω and ω. It equals

$$j_1(t) = i\frac{\sigma_0}{4e}\int\frac{d\epsilon}{2\pi}\big[\mathbf{Q}\,\exp(i\Omega t)\{\hat{g}_{\epsilon-\Omega}\langle\hat{g}_\epsilon\rangle\}_0^K + \mathbf{q}\,\exp(i\omega t)\{\hat{g}_{\epsilon-\omega}\langle\hat{g}_\epsilon\rangle\}_0^K\big]. \tag{50}$$

Here $\langle\hat{g}\rangle = \hat{\tau}_3\hat{g}\hat{\tau}_3$, $\{\cdots\}_0 = (1/2)\mathrm{Tr}\{\cdots\}$, $\hat{g}^R_{\epsilon-\omega} \equiv \hat{g}^R_0(\epsilon-\omega)$, etc. The current $j_{1q}(t) \sim \mathbf{q}\,\exp(i\omega t)$ can be represented as

$$j_{1q}(t) = i\frac{\sigma_0}{4e}\,\mathbf{q}\,\exp(i\omega t)\big[J_1^{\mathrm{reg}} + J_1^{\mathrm{an}}\big], \tag{51}$$

with

$$J_1^{\mathrm{reg}} = \int\frac{d\epsilon}{2\pi}\big\{\hat{g}^R_\epsilon\big\langle\hat{g}^R_{\epsilon+\omega}\big\rangle t_{\epsilon+\omega} - \hat{g}^A_\epsilon\big\langle\hat{g}^A_{\epsilon+\omega}\big\rangle t_\epsilon\big\}_0, \tag{52}$$

$$J_1^{\mathrm{an}} = -\int\frac{d\epsilon}{2\pi}\big\{\hat{g}^R_\epsilon\big\langle\hat{g}^A_{\epsilon+\omega}\big\rangle\big\}_0(t_{\epsilon+\omega} - t_\epsilon). \tag{53}$$

The next correction to the first ω harmonic in the presence of a pump is

$$\delta j(t) = i\frac{\sigma_0}{4e}\mathbf{q}\int d\tau\,\exp(i\omega\tau)\{\delta\hat{g}(t,\tau)\langle\hat{g}_0(\tau-t)\rangle + \langle\hat{g}_0(t-\tau)\rangle\delta\hat{g}(\tau,t)\}_0^K. \tag{54}$$

In the Fourier representation, the current acquires the form

$$\delta j_\omega(t) = i\frac{\sigma_0}{4e}\mathbf{q}_\omega\sum_{\omega,\nu,\mu}\exp\left((i\omega + \Omega_{\nu+\mu})t\right) \times \int\frac{d\epsilon}{2\pi}\{\delta\hat{g}_{\epsilon,\epsilon+\Omega_{\nu+\mu}}\langle\hat{g}_{\epsilon+\omega+\Omega_{\nu+\mu}}\rangle + \langle\hat{g}_{\epsilon-\omega}\rangle\delta\hat{g}_{\epsilon+\Omega_{\nu+\mu}}\}_0^K. \tag{55}$$

Here $\delta\hat{g}_\epsilon$ contains the terms of the order of Q^2, and the summation is performed for $\pm\omega$, $\Omega_\nu = \pm\Omega_\mu = \pm\Omega$. Therefore, the current $\delta\boldsymbol{j}_\omega(t)$ contains harmonics $\delta\boldsymbol{j}_{\omega,\Omega}\infty\delta\boldsymbol{j}_{\omega,0}(Q^2)\exp(i\omega t)$, $\delta\boldsymbol{j}_{\omega,3}(Q^2)\exp\left(i(2\Omega\pm\omega)t\right)$. The first harmonic corresponds to $\Omega_\nu = -\Omega_\mu = \Omega$, whereas the third harmonic corresponds to $\Omega_\nu = \Omega_\mu = \Omega$. In obtaining this equation, we used the relation $\delta\hat{g}_{\epsilon,\epsilon'} = \delta\hat{g}_\epsilon 2\pi\delta(\epsilon' - \epsilon \pm \Omega_{\nu+\mu})$.

The correction $\delta\hat{g}_{\epsilon,\epsilon'}$ consists of all second-order combinations of $\mathbf{Q}$ and $\mathbf{q}$ and we took into account that $\mathbf{Q}\perp\mathbf{q}$ and $|\mathbf{q}| \ll |\mathbf{Q}|$, which leaves only the term $\propto Q^2$. First, we consider the correction to the conductance caused by the Eliashberg effect (EE).

A. Correction to the current due to EE

In this case, the frequency $\Omega_{\nu+\mu} = \Omega_\nu + \Omega_\mu = 0$ and the correction $\delta\hat{g}_{\epsilon,\epsilon'}$ can be written as follows:

$$\delta\hat{g}_{\epsilon,\epsilon'} = (DQ^2)\delta\hat{g}_\epsilon 2\pi\delta(\epsilon - \epsilon') + (-\Omega). \tag{56}$$

Then, the integral Eq. (54) acquires the form

$$\boldsymbol{j}^{\mathrm{EE}}(t) = i\frac{\sigma_0}{4e}\mathbf{q}_\omega\exp(i\omega t)J^{\mathrm{EE}}, \tag{57}$$

with

$$J^{\mathrm{EE}} = \int\frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^R_\epsilon\big\langle\hat{g}^K_{\epsilon+\omega}\big\rangle + \delta\hat{g}^K_\epsilon\big\langle\hat{g}^A_{\epsilon+\omega}\big\rangle + \big\langle\hat{g}^R_{\epsilon-\omega}\big\rangle\delta\hat{g}^K_\epsilon + \big\langle\hat{g}^K_{\epsilon-\omega}\big\rangle\delta\hat{g}^A_\epsilon\big\}_0. \tag{58}$$

The Keldysh component $\delta\hat{g}^K_\epsilon$ is defined as

$$\delta\hat{g}^K_\epsilon = \delta\hat{g}^{\mathrm{reg}}_\epsilon + \delta\hat{g}^{\mathrm{an}}_\epsilon, \tag{59}$$

$$\delta\hat{g}^{\mathrm{reg}}_\epsilon = t_\epsilon\big(\delta\hat{g}^R_\epsilon - \delta\hat{g}^A_\epsilon\big). \tag{60}$$

The integral J^{EE} consists now of three terms,

$$J^{\mathrm{EE}} = J^{\mathrm{reg}} + J^{\mathrm{AN}} + J^{\mathrm{an}}. \tag{61}$$

The regular part is

$$J^{\mathrm{reg}} = \int\frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^R_\epsilon[\langle\hat{g}_{\epsilon+\omega}\rangle t_{\epsilon+\omega} + \langle\hat{g}_{\epsilon-\omega}\rangle t_\epsilon]^R - \delta\hat{g}^A_\epsilon[\langle\hat{g}_{\epsilon+\omega}\rangle t_\epsilon + \langle\hat{g}_{\epsilon-\omega}\rangle t_{\epsilon-\omega}]^A\big\}_0. \tag{62}$$

The anomalous terms J^{AN} and J^{an} are equal to

$$J^{\mathrm{AN}} = -\int\frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^R_\epsilon\big\langle\hat{g}^A_{\epsilon+\omega}\big\rangle(t_{\epsilon+\omega} - t_\epsilon) + \delta\hat{g}^A_\epsilon\big\langle\hat{g}^R_{\epsilon-\omega}\big\rangle(t_\epsilon - t_{\epsilon-\omega})\big\}_0, \tag{63}$$

$$J^{\mathrm{an}} = \int\frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^{\mathrm{an}}_\epsilon\big\langle\hat{g}^R_{\epsilon-\omega} + \hat{g}^A_{\epsilon+\omega}\big\rangle\big\}_0. \tag{64}$$

Here, the $\delta\hat{g}^{R(A)}_\epsilon$ matrix is

$$\delta\hat{g}^{R(A)}_\epsilon = \frac{1}{2\zeta^{R(A)}_\epsilon}\{\hat{\tau}_3(\delta g_\Delta + \delta g_Q) + i\hat{\tau}_2(\delta f_\Delta + \delta f_Q)\}^{R(A)}. \tag{65}$$

The anomalous function $\delta\hat{g}^{\mathrm{an}}_\epsilon$ is

$$\delta\hat{g}^{\mathrm{an}}_\epsilon = iDQ^2\frac{t_{\epsilon+\Omega} - t_\epsilon}{\big(\zeta^R_\epsilon + \zeta^A_\epsilon\big)}\big\{N_3^{\mathrm{an}}\hat{\tau}_3 - i\hat{\tau}_2N_2^{\mathrm{an}}\big\}, \tag{66}$$

where $N^{\mathrm{an}}_{2,3}$ are

$$N_3^{\mathrm{an}} = G^{(-)}_{\epsilon+\Omega}A^{\mathrm{an}(-)}_{\epsilon,\epsilon} - F^{(-)}_{\epsilon+\Omega}B^{\mathrm{an}}_{\epsilon,\epsilon}, \tag{67}$$

$$N_2^{\mathrm{an}} = F^{(-)}_{\epsilon+\Omega}A^{\mathrm{an}(+)}_{\epsilon,\epsilon} + G^{(-)}_{\epsilon+\Omega}B^{\mathrm{an}}_{\epsilon,\epsilon}. \tag{68}$$

From the current, we can derive the correction to the complex conductivity

$$\delta\sigma(\omega,\Omega) = \frac{\sigma_0}{4\omega}J^{\mathrm{EE}}(\omega,\Omega), \tag{69}$$

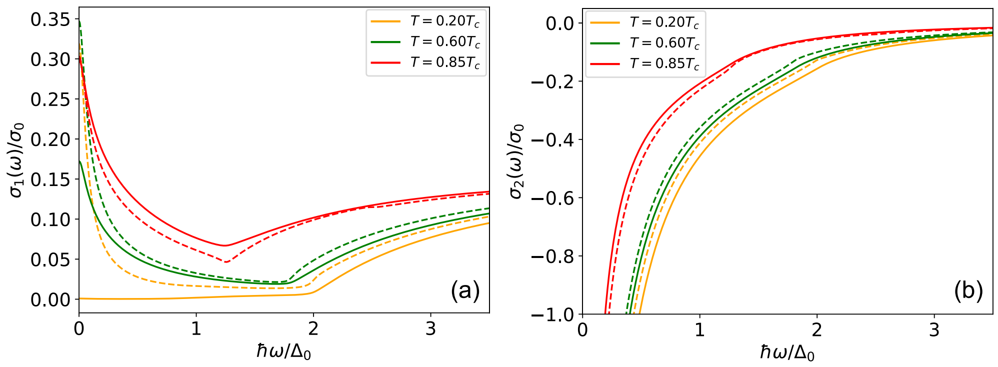

FIG. 4. Calculated photoexcited conductivity $\sigma(\omega, \Omega)$. The real part (a) of the conductivity $\sigma_1(\omega)$ and the imaginary part (b) $\sigma_2(\omega)$ are shown as a function of the probe frequency ω. The dashed (solid) curves represent the photoexcited (equilibrium) conductivity. The calculations were done for $DQ^2 = 0.005\Delta(T)$. The pump frequency is fixed at $\Omega = 2\Delta(T)$.

while the equilibrium conductivity is given by

$$\sigma^{(1)}(\omega) = \frac{\sigma_0}{4\omega}\big[J_1^{\mathrm{reg}} + J_1^{\mathrm{an}}\big]. \tag{70}$$

In Fig. 4 we show the total conductivity $\sigma(\omega, \Omega) = \sigma^{(1)}(\omega) + \delta\sigma(\omega, \Omega)$ (real and imaginary part) as a function of the probe frequency, ω, in comparison to its equilibrium behavior, $\sigma^{(1)}(\omega)$, for several temperatures and a pump frequency of $\Omega = 2\Delta(T)$. The used parameters are $\gamma = 0.02\Delta_0$ and $DQ^2 = 0.005\Delta(T)$. DQ^2 is chosen to be temperature-dependent as we need to satisfy $DQ^2 \ll \Delta(T)$, i.e., that we are far from the critical current for all temperatures. Observe that within the linear response and low temperatures $T = 0.2T_c$ the optical conductivity is gapped approximately to $2\Delta(T)$ and its evolution with temperature follows the standard behavior, dictated by the thermal excitation of quasiparticles in equilibrium superconductor in the presence of disorder; see, for example, Ref. [105]. The effect of the pump at low temperature ($T = 0.2T_c$) can be viewed as the effect of the "effective temperature" as $\sigma_1(\omega)$ become gapless and increases at lower ω due to the pair-breaking effect of the pump pulse and the resulting formation of the quasiparticles in a similar fashion as the increasing temperature would do. The situation, however, changes for higher temperatures and especially for $T = 0.85T_c$ where one observes the Eliashberg effect in which $\sigma_1(\omega)$ shows much stronger gap features than in the equilibrium. We observe a decrease of the spectral weight of the $\sigma_1(\omega)$ in the sub $2\Delta(T)$ region, i.e., a decrease of the in-gap states for $T = 0.85T_c$. Further, we also observe a slight shift of kink of the conductivity, which is located at $\omega = 2\Delta(T)$, to a higher frequency. As we assumed, $DQ^2 \ll \Delta(T)$ and therefore the correction $\delta\Delta_0(T)$ is also small compared to $\Delta(T)$; see Eq. (40), which corresponds to the shift of the gap edge. We observe the same trend to a lower effective temperature in the imaginary part of the conductivity $\sigma_2(\omega)$ as well, which is most evident in the region $\omega \leqslant 2\Delta(T)$. Further, the enhancement of the conductivity at low ω in the presence of a pump is related to that obtained earlier in the case of a Josephson junction [19] and of a uniform superconductor [106].

In Fig. 5 we show the behavior of $\sigma(\omega, \Omega)$ at a low temperature of $T = 0.1T_c$ as a function of the probe (a) and the pump frequency (b) for $\gamma = 0.02\Delta_0$ and $DQ^2 = 0.1\Delta(T)$. One could clearly see that the effect of the pump arises for $\Omega \geqslant \Delta(T)$ and is the most prominent one for $\Omega \sim 2\Delta(T)$. Note that we plot the curves in Figs. 4 and 5 for particular magnitudes of ratio $p = DQ^2/\Delta$, which is assumed to be small. But all the functions (corrections of the Green's functions and variation of Δ) are proportional to this parameter. Therefore, an increase of p means stretching the graphs along the y-axis.

B. Amplitude of the third harmonic

Here, we consider an amplitude of the third harmonic of the form $j^{\mathrm{TH}}(t) \propto j^{\mathrm{TH}} \exp{(i(2\Omega \pm \omega))}$. In this case, the frequency $\Omega_{\nu+\mu} = \Omega_\nu + \Omega_\mu = 2\Omega$ and the correction $\delta\hat{g}_{\epsilon,\epsilon'}$ can be written as follows:

$$\delta\hat{g}_{\epsilon,\epsilon'} = (DQ^2)\delta\hat{g}_\epsilon 2\pi\delta(\epsilon_- + 2\Omega). \tag{71}$$

The current in Eq. (54) is given by

$$\boldsymbol{j}^{\mathrm{TH}}(t) = i\frac{\sigma}{4e}\mathbf{q}_\omega \exp{(i(2\Omega \pm \omega)t)}J^{\mathrm{TH}}, \tag{72}$$

with J^{TH} equal to

$$J^{\mathrm{TH}} = \tilde{J}^{\mathrm{reg}} + \tilde{J}^{\mathrm{AN}} + \tilde{J}^{\mathrm{an}}. \tag{73}$$

The currents $\tilde{J}^{\mathrm{reg}}$, $\tilde{J}^{\mathrm{AN}}$, and $\tilde{J}^{\mathrm{an}}$ are defined as follows:

$$\tilde{J}^{\mathrm{reg}} = \int \frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^R_\epsilon[\langle\hat{g}_{\tilde{\epsilon}}\rangle t_{\tilde{\epsilon}} + \langle\hat{g}_{\epsilon-\omega}\rangle t_{\epsilon+2\Omega}]^R - \delta\hat{g}^A_\epsilon[\langle\hat{g}_{\tilde{\epsilon}}\rangle t_\epsilon + \langle\hat{g}_{\epsilon-\omega}\rangle t_{\epsilon-\omega}]^A\big\}_0, \tag{74}$$

$$\tilde{J}^{\mathrm{AN}} = -\int \frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^R_\epsilon\big\langle\hat{g}^A_{\tilde{\epsilon}}\big\rangle(t_{\tilde{\epsilon}} - t_{\epsilon+2\Omega}) + \delta\hat{g}^A_\epsilon\big\langle\hat{g}^R_{\epsilon-\omega}\big\rangle(t_\epsilon - t_{\epsilon-\omega})\big\}_0, \tag{75}$$

$$\tilde{J}^{\mathrm{an}} = \int \frac{d\epsilon}{2\pi}\big\{\delta\hat{g}^{\mathrm{an}}_\epsilon\big\langle\hat{g}^R_{\epsilon-\omega} + \hat{g}^A_{\tilde{\epsilon}}\big\rangle\big\}_0, \tag{76}$$

where $\tilde{\epsilon} = 2\Omega \pm \omega$ and the matrix $\delta\hat{g}^{R(A)}_\epsilon$ is given by

$$\delta\hat{g}^{R(A)}_\epsilon = \frac{1}{\zeta^{R(A)}_\epsilon + \zeta^{R(A)}_{\epsilon+2\Omega}}\big\{\delta\hat{\Delta}_{2\Omega} - \hat{g}_\epsilon\delta\hat{\Delta}_{2\Omega}\hat{g}_{\epsilon+2\Omega} + iDQ^2[\langle\hat{g}_{\epsilon+\Omega}\rangle - \hat{g}_\epsilon\langle\hat{g}_{\epsilon+\Omega}\rangle\hat{g}_{\epsilon+2\Omega}]\big\}^{R(A)}. \tag{77}$$

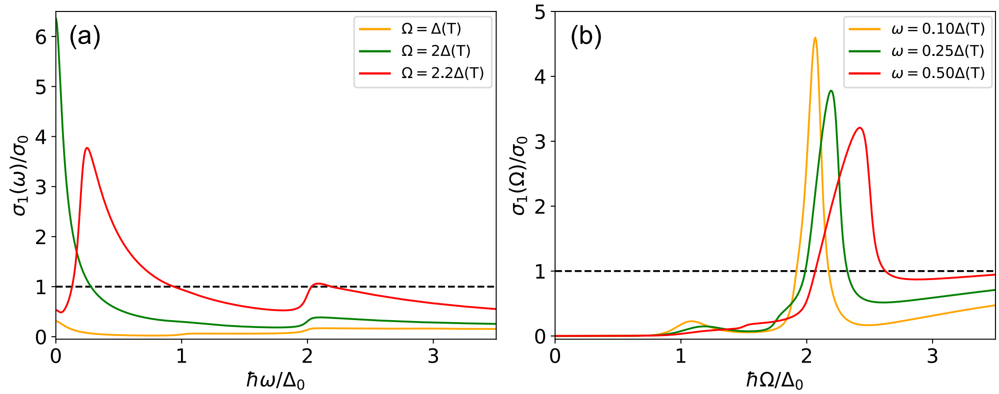

FIG. 5. Calculated real part of the photoexcited complex conductivity, $\sigma(\omega, \Omega)$. In (a) $\sigma_1(\omega, \Omega)$ is shown as a function of the probe frequency ω for various pump frequencies Ω. In (b) the real part of the conductivity $\sigma_1(\omega, \Omega)$ is shown as a function of the pump frequency Ω for various fixed probe frequencies ω. The calculation were done for $DQ^2 = 0.1\Delta(T)$ at $T = 0.1T_c$.

The anomalous function $\delta\hat{g}^{\mathrm{an}}_{\epsilon}$ is

$$\begin{aligned}\delta\hat{g}^{\mathrm{an}}_{\epsilon} = \frac{1}{\zeta^R_{\epsilon} + \zeta^A_{\epsilon+2\Omega}}\big\{\big[\delta\hat{\Delta}_{2\Omega} - \hat{g}^R_{\epsilon}\delta\hat{\Delta}_{2\Omega}\hat{g}^A_{\epsilon+2\Omega}\big](t_{\epsilon} - t_{\epsilon+2\Omega})\\ + iDQ^2\big[\big\langle\hat{g}^R_{\epsilon+\Omega}\big\rangle - \hat{g}^R_{\epsilon}\big\langle\hat{g}^R_{\epsilon+\Omega}\big\rangle\hat{g}^A_{\epsilon+2\Omega}\big](t_{\epsilon+\Omega} - t_{\epsilon+2\Omega})\\ + iDQ^2\big[\big\langle\hat{g}^A_{\epsilon+\Omega}\big\rangle - \hat{g}^R_{\epsilon}\big\langle\hat{g}^A_{\epsilon+\Omega}\big\rangle\hat{g}^A_{\epsilon+2\Omega}\big](t_{\epsilon} - t_{\epsilon+\Omega})\big\}. \quad (78)\end{aligned}$$

In Fig. 6 we plot the temperature dependence of the third-harmonic current contribution normalized to $j_0 = \sigma DQ^3 e^{-1}$ and $\gamma = 0.02\Delta_0$ and $\omega = \Omega$. For this, we also separate the amplitude (Higgs) mode contribution j_{H} and the contribution of the direct action of the electric field j_{AAA} by separating $\delta\hat{g} = \delta\hat{g}_Q + \delta\hat{g}_{\Delta}$ in Eqs. (74)–(76). $\delta\hat{g}_{\Delta} \propto \delta\hat{\hat{\Delta}}$ describes the correction to the current arising from the correction to Δ, while $\delta\hat{g}_Q$ describes the direct coupling of the light to the condensate. This separation was already introduced earlier, when we defined $\delta\hat{g}$ in Eqs. (21) and (24).

As discussed in the Introduction, one of the most interesting questions is whether the amplitude (Higgs) mode contribution dominates over the contribution due to the direct action of an ac electric field in the diffusive superconductors. Indeed, we find that the amplitude mode contribution

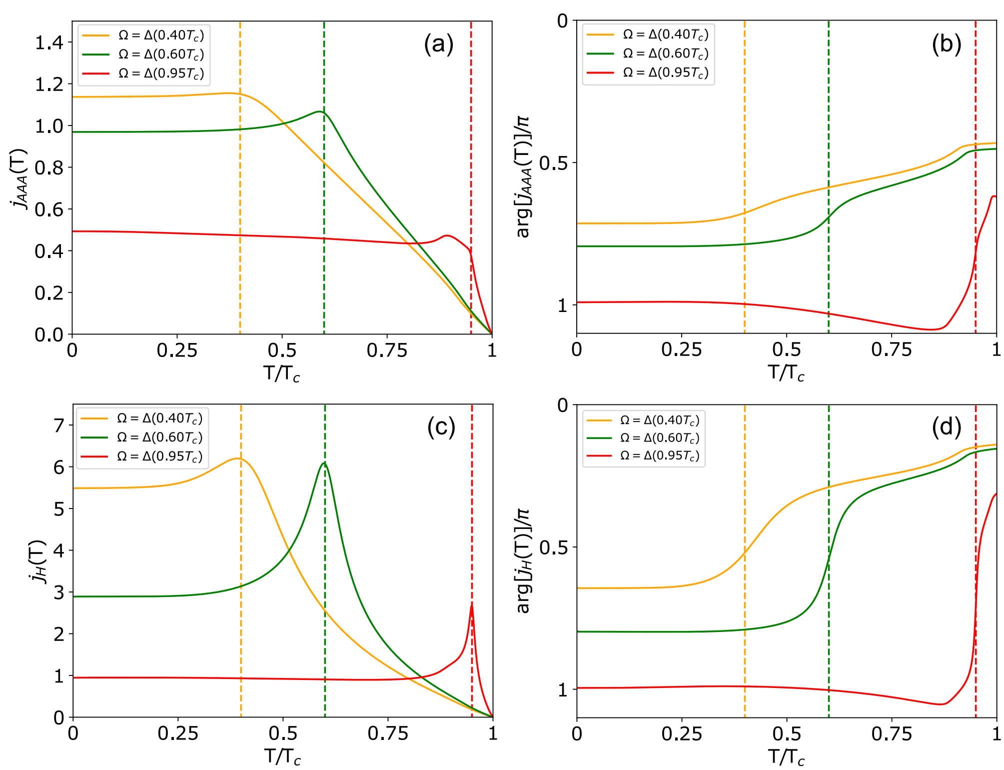

FIG. 6. Calculated temperature dependence of the THG currents. Parts (a) and (b) show the amplitude and phase of j_{AAA}. Parts (c) and (d) show the amplitude and phase of the Higgs contribution j_H. We observe that the Higgs contribution dominates the THG current and that both contributions possess a peak and a phase shift at the resonance condition $\Omega = \Delta(T)$ highlighted by the dashed lines.

dominates over the direct action of the electric field, and this is especially prominent if one looks at the phase shift of the argument of the THG current, consistent with previous calculations by various groups.

VI. REFLECTION AND TRANSMISSION COEFFICIENTS OF AN ac FIELD

We consider an electromagnetic wave incident on a superconducting film with thickness $2d$. The reflection (transmission) coefficient is determined from Maxwell's equations

$$\nabla \times \mathbf{H} = \frac{4\pi}{c}\mathbf{j} + \frac{\varepsilon}{c}\partial_t \mathbf{E}, \tag{79}$$

$$\nabla \times \mathbf{E} = -\frac{1}{c}\partial_t \mathbf{H}. \tag{80}$$

For a wave of a form $E \infty \exp(i\omega t - ikz)$, these equations can be written as

$$\mathbf{k} \times \mathbf{H} = \frac{1}{c}(4\pi i\sigma_\omega - \varepsilon\omega)\mathbf{E}, \tag{81}$$

$$\mathbf{k} \times \mathbf{E} = \frac{\omega}{c}\mathbf{H}, \tag{82}$$

or

$$\mathbf{k} \times (\mathbf{k} \times \mathbf{E}) = \frac{1}{c^2}(4\pi i\sigma_\omega\omega - \varepsilon\omega^2)\mathbf{E}. \tag{83}$$

The dispersion relation $\omega(k)$ follows directly from Eq. (83),

$$(ck_S)^2 = -4\pi i\sigma(\omega)\omega + \varepsilon\omega^2, S \text{ film}, \tag{84}$$

$$(ck_V)^2 = \varepsilon\omega^2, \text{ vacuum}. \tag{85}$$

Writing down the Maxwell equations for components of E and H, we find

$$\partial_z H = \frac{1}{c}[4\pi\sigma(\omega) + i\omega]E \equiv \epsilon_\omega E, \tag{86}$$

$$\partial_z E = \frac{i\omega}{c}H. \tag{87}$$

Thus, we obtain for the solution $E(z,t) \equiv E(z)\exp(i\omega t)$ and $H(z,t) \equiv H(z)\exp(i\omega t)$ with

$$E(z) = \begin{cases} E_{\text{in}}\exp(-ik_V(z+d)) + E_r\exp(ik_V(z+d)), & z < -d, \\ E_{\text{tr}}\exp(-ik_V(z-d)), & z > d, \end{cases} \tag{88}$$

and

$$H(z) = \sqrt{\epsilon_0}\begin{cases} -E_{\text{in}}\exp(-ik_V(z+d)) + E_r\exp(ik_V(z+d)), & z < -d, \\ -E_{\text{tr}}\exp(-ik_V(z-d)), & z > d. \end{cases} \tag{89}$$

Inside the S film, we have

$$E(z) = C\cosh(ik_S z) + S\sinh(ik_S z) \quad |z| < d, \tag{90}$$

$$H(t,z) = \sqrt{\epsilon_\Omega}[C\sinh(ik_S z) + S\cosh(ik_S z)], \quad |z| < d. \tag{91}$$

The matching conditions $[E]_{\pm d} = 0$ and $[H]_{\pm d} = 0$ yield

$$E_r + E_{\text{tr}} = E_{\text{in}}\frac{\sqrt{\epsilon_0} - \sqrt{\epsilon_\omega}\tanh(i\theta_S)}{\sqrt{\epsilon_0} + \sqrt{\epsilon_\omega}\tanh(i\theta_S)}, \tag{92}$$

$$E_r - E_{\text{tr}} = -E_{\text{in}}\frac{\sqrt{\epsilon_\omega} - \sqrt{\epsilon_0}\tanh(i\theta_S)}{\sqrt{\epsilon_\omega} + \sqrt{\epsilon_0}\tanh(i\theta_S)}, \tag{93}$$

where $\theta_S = k_S d$, $k_S c = \omega\sqrt{\epsilon_\omega}$, and $\epsilon_\omega = 4\pi\sigma(\omega)/i\omega + \epsilon_{S0}$.

As a result, we obtain the reflected and transmitted waves

$$E_r = E_{\text{in}}\frac{\tanh(i\theta_S)(\epsilon_0 - \epsilon_\omega)}{\mathcal{D}}, \tag{94}$$

$$E_{\text{tr}} = E_{\text{in}}\sqrt{\epsilon_\omega\epsilon_0}\frac{1}{\cosh^2(i\theta_S)\mathcal{D}}, \tag{95}$$

where $\mathcal{D} = [\sqrt{\epsilon_\omega} + \sqrt{\epsilon_0}\tanh(i\theta_S)][\sqrt{\epsilon_0} + \sqrt{\epsilon_\omega}\tanh(i\theta_S)]$. The reflected wave phase change ϕ_r is given by

$$\tan(\phi_r) = \frac{\text{Im}\{\tanh(i\theta_S)(\epsilon_0 - \epsilon_\omega)/\mathcal{D}\}}{\text{Re}\{\tanh(i\theta_S)(\epsilon_0 - \epsilon_\omega)/\mathcal{D}\}}, \tag{96}$$

which can also be measured in experiments. In the limit of a thick S film ($|\theta_S| \gg 1$), we obtain $\mathcal{D} = [\sqrt{\epsilon_0} + \sqrt{\epsilon_\omega}]^2$ and

$$E_r = E_{\text{in}}\frac{\sqrt{\epsilon_0} - \sqrt{\epsilon_\omega}}{\sqrt{\epsilon_0} + \sqrt{\epsilon_\omega}}, \tag{97}$$

$$E_{\text{tr}} = E_{\text{in}}\frac{\sqrt{\epsilon_0\epsilon_\omega}}{\cosh^2(i\theta_S)[\sqrt{\epsilon_0} + \sqrt{\epsilon_\omega}]^2}. \tag{98}$$

As expected, Eq. (98) shows that the amplitude of the transmitted wave is exponentially small: $E_{\text{tr}} \infty E_{\text{in}}\exp(-2d/d_{\text{skin}})$, where $d_{\text{skin}} = c/\sqrt{4\pi i\sigma(\omega)\omega}$ is the skin depth.

For a thin S film ($|\theta_s| \ll 1$) we plot the reflectivity $R(\omega) = \frac{|E_r|^2}{|E_{\text{in}}|^2}$ as a function of normalized frequency for various temperatures in Fig. 7 for $\gamma = 0.02\Delta_0$, $DQ^2 = 0.005\Delta(T)$, $\sigma_0 = 2 \times 10^{16}\ \text{s}^{-1}$, and $d = 12$ nm. Observe clear signatures of the Eliashberg effect for temperatures close to the superconducting transition temperature for both $\Omega = \Delta(T)$ and $\Omega = 2\Delta(T)$.

Next we look into the effects of parametric amplification. For that we assumed that the EM-field inside the thin S-film should be either a uniform or a slowly varying function. Thus, we take the average of the electric field inside the film:

$$\langle E_\omega^S \rangle = \frac{E_{\text{in}}\tan(\theta_s)}{2\theta_s}\left(1 + \frac{\sqrt{\epsilon_V} - \sqrt{\epsilon_\omega}\tanh(i\theta_s)}{\sqrt{\epsilon_V} + \sqrt{\epsilon_\omega}\tanh(i\theta_s)}\right). \tag{99}$$

Most importantly, we can now link the incoming electric field $E_{\text{in}}(\omega)$ with the outgoing field $E_r(2\Omega - \omega) = E_{\text{tr}}(2\Omega - \omega) =$

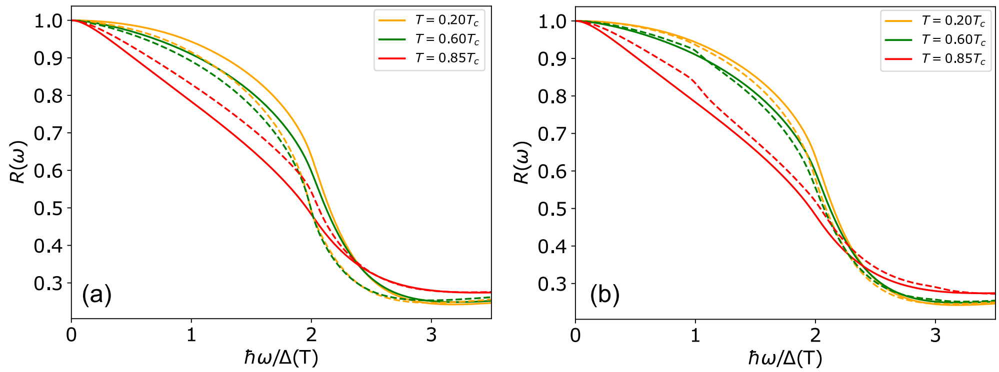

FIG. 7. The reflectivity $R(\omega) = \frac{|E_r|^2}{|E_{in}|^2}$ via Eq. (94). The solid lines represent the case of the equilibrium, while the dashed lines are obtained by taking the correction $\delta\sigma(\omega,\Omega)$ into account when determining ϵ_ω. The pump-frequency is fixed at $\Omega = 2\Delta(T)$ for (a) and $\Omega = \Delta(T)$ for (b). Further, $DQ^2 = 0.005\Delta(T)$, $\gamma = 0.02\Delta_0$, $\sigma_0 = 2\times 10^{16}\,\mathrm{s}^{-1}$, and a film thickness of $d = 12$ nm. For a pump of $\Omega = 2\Delta(T)$, we observe that the high-temperature reflectivity curve is getting enhanced under irradiation, while this is not the case for the lower-temperature curves. Lastly, we observe that for a lower pump-frequency $\Omega = \Delta(T)$ we see a small area of reflectivity enhancement for the lower-temperature curve of $T = 0.6T_c$, which is due to the redistribution of particles inside the gap and not to the Eliashberg effect.

$E_{2\Omega-\omega}$ via the THG current

$$j(2\Omega-\omega) = \frac{\Lambda_{\mathrm{ac}}(2\Omega-\omega)}{i\omega}\langle E^S_\omega\rangle = \sigma^{(1)}(2\Omega-\omega)E_{2\Omega-\omega}. \tag{100}$$

Using the definition from Eq. (99) for the field E_ω inside of the S film, we define the down-conversion intensity

$$R_{12}(2\Omega-\omega) = \left|\frac{\Lambda_{\mathrm{ac}}(2\Omega-\omega)}{i\omega\sigma^{(1)}(2\Omega-\omega)}\right|^2 \times\left|\frac{\tan(\theta_s)}{2\theta_s}\left(1+\frac{\sqrt{\epsilon_V}-\sqrt{\epsilon_\omega}\tanh(i\theta_s)}{\sqrt{\epsilon_V}+\sqrt{\epsilon_\omega}\tanh(i\theta_s)}\right)\right|^2, \tag{101}$$

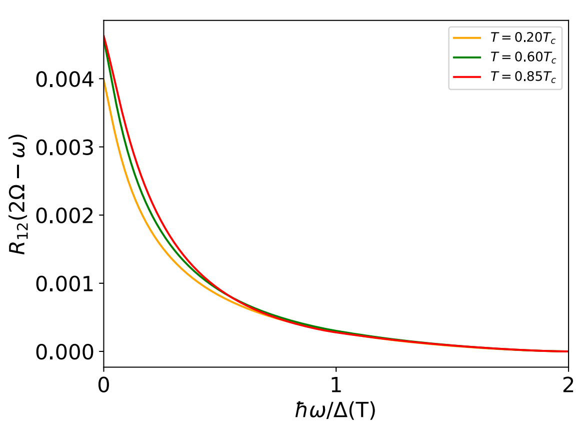

FIG. 8. The down-conversion intensity $R_{12}(2\Omega-\omega)$, i.e., $\frac{|\vec{E}_{2\Omega-\omega}|^2}{|\vec{E}_\omega|^2}$, as a function of the probe-frequency ω. The pump-frequency is fixed at $\Omega = \Delta(T)$ and $DQ^2 = 0.005\Delta(T)$. Further, $\gamma = 0.02\Delta_0$, $\sigma_0 = 2\times 10^{16}\,\mathrm{s}^{-1}$, and a film thickness of $d = 12$ nm. The highest intensity is achieved for $\omega\to 0$, while it vanishes at the points $\omega = 2\Omega$. For frequencies of $\Delta(T)\leqslant\omega\leqslant 2\Delta(T)$, the curves visually overlay.

which is similar to Ref. [70], with the important difference that all quantities entering its definition are now determined fully microscopically. In particular, the function Λ_{ac} is directly given by the THG current $j(2\Omega-\omega)$ and $\sigma^{(1)}$ is the linear response of the complex conductivity.

The down-conversion intensity $R_{12}(2\Omega-\omega)$ for $\Omega = \Delta(T)$ is shown in Fig. 8 as a function of the probe frequency ω for the same parameters as for the reflectivity. It is noted that in contrast to $R(\omega)$, we do not include the conductivity correction $\delta\sigma(\omega,\Omega)$ in the definition of ϵ_ω in Eq. (101). Similar to the phenomenological analysis presented in Ref. [70], we observe that the down-conversion intensity decreases rapidly in the region of $\omega < 2\Delta(T)$ and that it vanishes at $\omega = 2\Delta(T)$. Note that in Ref. [70] this quantity was related to the amplitude of the emitted idler mode, normalized by the amplitude of the incident signal beam and connected to the parametric amplification of superconductivity due to the Higgs mode.

In our case, we can indeed confirm that the behavior of the down-conversion intensity is expected due to the third-harmonic generation currents and excitation of the Higgs mode and the direct action of the electric field on the charge carriers in the superconducting state.

VII. CONCLUSIONS

To conclude, we have developed a theory of nonlinear effects arising in diffusive superconductors under the action of an ac electromagnetic fields $E(t)$. These effects are described in terms of quasiclassical matrix Green's functions $\hat{g}$, which consist of the retarded (advanced), $\hat{g}^{R(A)}$, and Keldysh Green's functions [93]. We use a method of the representation of the Keldysh function $\hat{g}^K$ as the sum of a regular, $\hat{g}^{\mathrm{reg}}$, and "anomalous," $\hat{g}^{\mathrm{an}}$, parts [4]. This trick allows one to avoid the method of analytical continuation [93]. Using this representation, we derive general expressions for $\hat{g}^{R(A)}$ and $\hat{g}^{\mathrm{an}}$. On the basis of this formalism, we obtain the variation of the order parameter $\delta\Delta$ caused by the ac field up to the second order in the magnitude of $E(t)$. In particular, we analyze the zero Fourier harmonic of $\delta\Delta_0$ (the Eliashberg effect [53]) and the second Fourier harmonic $\delta\Delta_2$. Furthermore, we calculated the ac currents $I(\Omega)$ induced by external electromagnetic fields. Analyzing the third-harmonic generating current, we indeed confirm that in the diffusive superconductors it is mostly dominated by the amplitude ("Higgs") mode and not by the direction action of the ac electric field. This is in contrast to the clean case.

Finally, we also analyze microscopically the down-conversion intensity, R_{12}, of the thin superconducting sample, the characteristic behavior of which was argued to be related to the parametric amplification of superconductivity. Although studying parametric amplification goes beyond the present theoretical analysis, we indeed see that a very similar behavior of R_{12} is expected due to the coupling to the amplitude mode and the direct action of the electric field in the third-harmonic generation currents. We obtained also a strong enhancement of a photoexcited ac conductance, which occurs at low frequencies and low temperatures. This issue deserves separate consideration.

Note that our formalism for calculation of the nonlinear currents can be used to analyze the data on transient transport in pumped conventional and unconventional superconductors. Furthermore, given the recent interest in the study of the dynamics of multiband high-T_c superconductors [107–109], it would be useful to generalize this approach to the case of these superconductors.

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APPENDIX

1. Anomalous Green's function

The Keldysh component of Eq. (14) can be written as

$$\hat{N}_\epsilon \delta\hat{g}^K - \delta\hat{g}^K \hat{N}_{\epsilon'} = -\hat{\mathcal{R}}^K(\epsilon, \epsilon'), \tag{A1}$$

where $\hat{N}_\epsilon = \epsilon\hat{\tau}_3 + \Delta i\hat{\tau}_2$, $\hat{\mathcal{R}}^K(\epsilon,\epsilon') = \{[\delta\hat{\Delta}, \hat{g}] + iD[Q\hat{\tau}_3\hat{g}\, Q\hat{\tau}_3, \hat{g}]\}^K$. The normalization condition for the Keldysh component is

$$\delta\{\hat{g}^R\hat{g}^K + \hat{g}^K\hat{g}^A\}_{\epsilon,\epsilon'} = 0 \tag{A2}$$

or

$$\delta\hat{g}^R\hat{g}^K_{\epsilon'} + \hat{g}^R_\epsilon\delta\hat{g}^K + \delta\hat{g}^K\hat{g}^A_{\epsilon'} + \hat{g}^K_\epsilon\delta\hat{g}^A = 0. \tag{A3}$$

Here $\hat{g}^K_\epsilon = (\hat{g}^R_\epsilon - \hat{g}^A_\epsilon)t_\epsilon$ is the Keldysh Green's function in equilibrium. The variation of the Keldysh function $\delta\hat{g}^K$ is represented as a sum of a regular part and the anomalous part $\delta\hat{g}^{\text{an}}$ [see Eq. (22)]

$$\delta\hat{g}^K = \delta\hat{g}^R t_{\epsilon'} - t_\epsilon\delta\hat{g}^A + \delta\hat{g}^{\text{an}}. \tag{A4}$$

Taking into account Eq. (A4), we can write Eq. (A3) as

$$\begin{aligned}0 = &\left(\delta\hat{g}^R\hat{g}^R_{\epsilon'} + \hat{g}^R_\epsilon\delta\hat{g}^R\right)t_{\epsilon'} - t_\epsilon\left(\delta\hat{g}^A\hat{g}^A_{\epsilon'} + \hat{g}^A_\epsilon\delta\hat{g}^A\right)\\ &+ \hat{g}^R_\epsilon\delta\hat{g}^{\text{an}} + \delta\hat{g}^{\text{an}}\hat{g}^A_{\epsilon'}.\end{aligned} \tag{A5}$$

The first two terms are equal to zero due to the variation of the normalization conditions for the matrices $\hat{g}^{R(A)}$,

$$(\delta\hat{g}\hat{g}_{\epsilon'} + \hat{g}_\epsilon\delta\hat{g})^{R(A)} = 0. \tag{A6}$$

Thus, we come to the equation for $\delta\hat{g}^{\text{an}}$,

$$\hat{g}^R_\epsilon\delta\hat{g}^{\text{an}} + \delta\hat{g}^{\text{an}}\hat{g}^A_{\epsilon'} = 0. \tag{A7}$$

Then, we multiply Eq. (14) for $\delta\hat{g}^{R(A)}$ by $t_{\epsilon'}$ (t_ϵ) and subtract (add) from (to) Eq. (A1). Taking into account the definition of the matrix $\hat{N}^{R(A)}_\epsilon = (\zeta_\epsilon\hat{g}_\epsilon)^{R(A)}$, we obtain

$$\begin{aligned}&(\zeta_\epsilon\hat{g})^R\delta\hat{g}^{\text{an}} - \delta\hat{g}^{\text{an}}(\zeta_{\epsilon'}\hat{g}_{\epsilon'})^A\\ &\quad = -\{\hat{\mathcal{R}}^K(\epsilon,\epsilon') - \hat{\mathcal{R}}^R(\epsilon,\epsilon')t_{\epsilon'} + t_\epsilon\hat{\mathcal{R}}^A(\epsilon,\epsilon')\}.\end{aligned} \tag{A8}$$

The matrix $\hat{\mathcal{R}}^K(\epsilon,\epsilon')$ contains only the Green's function in equilibrium when $\hat{g}^K_\epsilon = (\hat{g}^R_\epsilon - \hat{g}^A_\epsilon)t_\epsilon$. Using the normalization condition, Eq. (A7), we find for the anomalous function $\delta\hat{g}^{\text{an}}$

$$\delta\hat{g}^{\text{an}} = -\hat{g}^R_\epsilon\frac{\{\hat{\mathcal{R}}^K(\epsilon,\epsilon') - \hat{\mathcal{R}}^R(\epsilon,\epsilon')t_{\epsilon'} + t_\epsilon\hat{\mathcal{R}}^A(\epsilon,\epsilon')\}}{\zeta^R_\epsilon + \zeta^A_{\epsilon'}}. \tag{A9}$$

Substituting the known Green's functions into Eq. (A9), we come to Eqs. (24) and (25).

2. Useful functions

We list the definition of useful functions to avoid cluttering in the definition of $\delta\hat{g}_{\epsilon,\epsilon'}$,

$$N_3^{R(A)} = (A^{(-)}_\epsilon g_{\epsilon+\Omega} - B_\epsilon f_{\epsilon+\Omega})^{R(A)}, \tag{A10}$$

$$N_2^{R(A)} = (A^{(+)}_\epsilon f_{\epsilon+\Omega} + B_\epsilon g_{\epsilon+\Omega})^{R(A)}. \tag{A11}$$

The functions $N^{\text{an}}_{2,3}$, $G^{(-)}_{\tilde{\epsilon}}$, and $F^{(-)}_{\tilde{\epsilon}}$ are defined as follows:

$$N^{\text{an}}_3 = G^{(-)}_{\tilde{\epsilon}}A^{(-)}_\epsilon + F^{(-)}_{\tilde{\epsilon}}B, \tag{A12}$$

$$N^{\text{an}}_2 = F^{(-)}_{\tilde{\epsilon}}A^{(+)}_\epsilon + G^{(-)}_{\tilde{\epsilon}}B, \tag{A13}$$

$$G^{(-)}_{\epsilon+\Omega} = g^R_{\epsilon+\Omega} - g^A_{\epsilon+\Omega}, F^{(-)}_{\epsilon+\Omega} = f^R_{\epsilon+\Omega} - f^A_{\epsilon+\Omega}. \tag{A14}$$

The functions A, B, and C are

$$(A^{\pm}_\epsilon)^R = \left[1 \pm \left(g^2_\epsilon + f^2_\epsilon\right)\right]^R, \tag{A15}$$

$$B^R_\epsilon = 2(g_\epsilon f_\epsilon)^R, \tag{A16}$$

$$C^R_\epsilon = 2\zeta^R_\epsilon. \tag{A17}$$

Anomalous functions A^{an}_ϵ, B^{an}_ϵ, C^{an}_ϵ are

$$(A^{\pm}_\epsilon)^{\text{an}} = 1 \pm \left(g^R_\epsilon g^A_\epsilon + f^R_\epsilon f^A_\epsilon\right), \tag{A18}$$

$$B^{\text{an}}_\epsilon = g^R_\epsilon f^A_\epsilon + g^A_\epsilon f^R_\epsilon, \tag{A19}$$

$$C^{\text{an}}_\epsilon = \zeta^R_\epsilon + \zeta^A_\epsilon. \tag{A20}$$

Here we provide further expressions, which are useful in obtaining the final expressions for the Eliashberg effect and the third-harmonic generation currents in the main text.

For the $\delta\Delta(t)$ averaged in time, we have

$$\delta\Delta = -\lambda i\int\frac{d\epsilon}{2\pi}\text{Tr}\hat{\tau}_2\{\delta\hat{g}^{\text{reg}} + \hat{g}^{\text{an}}\}|_{\epsilon_-=0}. \tag{A21}$$

Taking into account the identity

$$\delta\Delta = \lambda\delta\Delta\int\frac{d\epsilon}{2\pi}\left[\frac{1}{\zeta^R_\epsilon}-\frac{1}{\zeta^A_\epsilon}\right]t_\epsilon \tag{A22}$$

and subtracting Eqs. (A21) and (A22), we obtain

$$0=\sum_{\nu,\mu}\int\frac{d\epsilon}{2\pi}\left[\mathrm{Tr}\{i\hat{\tau}_2(\delta\hat{g}^{\mathrm{reg}}+\hat{g}^{\mathrm{an}})\}+\left(\frac{1}{\zeta^R_\epsilon}-\frac{1}{\zeta^A_\epsilon}\right)t_\epsilon\delta\Delta\right], \tag{A23}$$

where $\zeta^{R(A)}_\epsilon=\sqrt{(\epsilon\pm i\gamma)^2+\Delta^2}$. This equation can be written in the form

$$4T\delta\Delta_0\Delta^2\sum_n\frac{1}{\zeta_n^3}=-i(DQ^2)\int\frac{d\epsilon}{2\pi}\frac{\delta f_Q^{\mathrm{reg}}+\delta f_Q^{\mathrm{an}}}{2\zeta_\epsilon}, \tag{A24}$$

where $\zeta_n=\sqrt{\epsilon_n^2+\Delta^2}$, $\epsilon_n=\pi T(2n+1)$. We used Eq. (30) with $f_\Delta=1+g_n^2+f_n^2$, $g_n^2=(\epsilon_n/\zeta_n)^2=1-f_n^2$.

The regular part at the right is

$$\begin{aligned}\int\frac{d\epsilon}{2\pi}\frac{\delta f_Q^{\mathrm{reg}}}{2\zeta_\epsilon}&=\int\frac{d\epsilon}{2\pi}\left[\left(\frac{A_+f_{\epsilon+\Omega}+Bg_{\epsilon+\Omega}}{2\zeta_\epsilon}\right)^R-(\cdots)^A\right]t_\epsilon\\&=\int\frac{d\epsilon}{2\pi}\left[\left(\frac{g_\epsilon(g_\epsilon f_{\epsilon+\Omega}+f_\epsilon g_{\epsilon+\Omega})}{\zeta_\epsilon}\right)^R-(\cdots)^A\right]t_\epsilon\\&=\Delta\int\frac{d\epsilon}{2\pi}\left[\left(\frac{\epsilon(2\epsilon+\Omega)}{\zeta^3_\epsilon\zeta_{\epsilon+\Omega}}\right)^R-(\cdots)^A\right]t_\epsilon.\end{aligned} \tag{A25}$$

We express the integral in terms of Matsubara frequencies

$$\int\frac{d\epsilon}{2\pi}\frac{\delta f_Q^{\mathrm{reg}}}{2\zeta_\epsilon}=\Delta\int\frac{d\epsilon}{2\pi}\left[\left(\frac{\epsilon(2\epsilon+\Omega)}{\zeta^3_\epsilon\zeta_{\epsilon+\Omega}}\right)^R-(\cdots)^A\right] \tag{A26}$$

$$=-4iT2\Delta\mathrm{Re}\sum_n\frac{\omega(2\omega+i\Omega)}{\zeta^3_\omega\zeta_{\omega+i\Omega}}. \tag{A27}$$

We took into account the term with $-\Omega$. Therefore, Eq. (34) yields

$$\begin{aligned}4T\delta\Delta_0\Delta^2\sum_n\frac{1}{\zeta_n^3}=&-8T\Delta DQ^2\mathrm{Re}\sum_n\frac{\omega(2\omega+i\Omega)}{\zeta^3_\omega\zeta_{\omega+i\Omega}}\\&-iDQ^2\int\frac{d\epsilon}{2\pi}\frac{\delta f_Q^{\mathrm{an}}}{2\zeta_\epsilon}.\end{aligned} \tag{A28}$$

3. Polarization operator

To understand the origin of the low-frequency peak in the $\delta\Delta_{2\Omega}$ we plot total polarization operator $P(2\Omega)=P^{\mathrm{reg}}_\Delta(2\Omega)+P^{\mathrm{an}}_\Delta(2\Omega)$ in Fig. 9 for $\gamma=0.02\Delta_0$ at various temperatures. We observe the emergence of a minimum for all three temperatures at the resonance frequency $\Omega=\Delta(T)$. Therefore, the low-energy peaks at $\Omega\to0$ in the second-harmonic correction $\delta\Delta_{2\Omega}$ are a consequence of the abrupt vanishing of $P^{\mathrm{an}}(2\Omega)$ at higher temperatures.

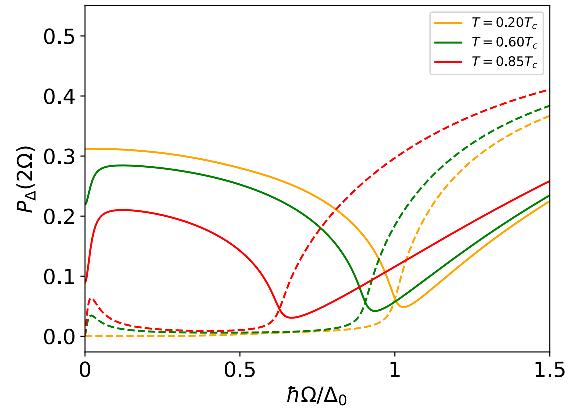

FIG. 9. The polarization operator $P(2\Omega)=P^{\mathrm{reg}}_\Delta(2\Omega)+P^{\mathrm{an}}_\Delta(2\Omega)$ as function of Ω. The solid (dashed) line is the real (imaginary) part of the polarization operator. We observe a minimum at the resonance frequency $\Omega=\Delta(T)$ for all temperatures, while for high temperatures we see a distinct second minimum for the limit $\Omega\to0$.

4. Third harmonic

The response to an external ac field with the frequency Ω_{in} is

$$I(t)\;=\sigma\;\exp\left(i(2\Omega+\Omega_{\mathrm{in}})t\right)Q_{\mathrm{in}}\int\frac{d\epsilon}{2\pi}J(\epsilon), \tag{A29}$$

where

$$\begin{aligned}J&=\int\frac{d\epsilon}{2\pi}\left[\delta\hat{g}\langle\hat{g}_{\epsilon+2\Omega+\omega}\rangle+\langle\hat{g}_{\epsilon-\omega}\rangle\delta\hat{g}^K_0\right]\\&=\int\frac{d\epsilon}{2\pi}\{\delta\hat{g}^R\left[\langle\hat{g}^R_{\epsilon+2\Omega+\omega}-\hat{g}^A_{\epsilon+2\Omega+\omega}\rangle t_{\epsilon+2\Omega+\omega}\right.\\&\quad+[\delta\hat{g}^Rt_{\epsilon'}-t_\epsilon\delta\hat{g}^A]\langle\hat{g}^A_{\epsilon+2\Omega+\omega}\rangle\langle\hat{g}^R_{\epsilon-\omega}\rangle[\delta\hat{g}^Rt_{\epsilon'}-t_\epsilon\delta\hat{g}^A]\\&\quad\left.+\langle\hat{g}^R_{\epsilon-\omega}-\hat{g}^A_{\epsilon-\omega}\rangle t_{\epsilon-\omega}\delta\hat{g}^A\right]+\delta\hat{g}^{\mathrm{an}}\left[\langle\hat{g}^R_{\epsilon+2\Omega+\omega}+\hat{g}^A_{\epsilon-\omega}\right\}_0.\end{aligned} \tag{A30}$$

5. Coefficients in the current (EE and third harmonic)

If $\Omega_{\mathrm{in}}=\Omega$, the total current is

$$I_{3\Omega}(t)\;=\sigma Q\exp(3i\Omega t)\int\frac{d\epsilon}{2\pi}\{\langle\hat{g}_{\epsilon-\Omega}\rangle\cdot\delta\hat{g}+\delta\hat{g}\cdot\langle\hat{g}_{\epsilon+3\Omega}\rangle\}^K_0, \tag{A31}$$

where $\delta\hat{g}=\delta\hat{g}(\epsilon,\epsilon+2\Omega)$. Here the matrix $\delta\hat{g}^{\mathrm{an}}_\mp\equiv\delta\hat{g}^{\mathrm{an}}(\tilde{\epsilon}_-,\tilde{\epsilon}_+)$ is defined as

$$\hat{g}^{\mathrm{an}}_\Delta=-r_\epsilon\frac{\sinh(2\Omega\beta)}{2}\frac{\delta\Delta_{2\Omega}}{C^{\mathrm{an}}_\mp}\left(g^{\mathrm{an}}_\Delta\hat{\tau}_3+f^{\mathrm{an}}_\Delta i\hat{\tau}_2\right), \tag{A32}$$

$$\hat{g}^{\mathrm{an}}_Q=-r_\epsilon\frac{\sinh(2\Omega\beta)}{2}\frac{iDQ^2}{C^{\mathrm{an}}_\mp}\left(g^{\mathrm{an}}_Q\hat{\tau}_3+f^{\mathrm{an}}_Q i\hat{\tau}_2\right). \tag{A33}$$

Thus, for J^{an} we find

$$\begin{aligned}J^{\mathrm{an}}=&\left\{\left(g^R_{\tilde{\epsilon}-2\Omega}+\hat{g}^A_{\tilde{\epsilon}+2\Omega}\right)\left(g^{\mathrm{an}}_\Delta+g^{\mathrm{an}}_Q\right)\right.\\&\left.+\left(f^R_{\tilde{\epsilon}-2\Omega}+f^A_{\tilde{\epsilon}+2\Omega}\right)\left(f^{\mathrm{an}}_\Delta+f^{\mathrm{an}}_Q\right)\right\}.\end{aligned} \tag{A34}$$

The coefficients $g^{\mathrm{an}}_{\Delta,Q}$ and $f^{\mathrm{an}}_{\Delta,Q}$ are defined as

$$g^{\mathrm{an}}_\Delta=B,\quad f^{\mathrm{an}}_\Delta=A_+, \tag{A35}$$

$$g_Q^{\mathrm{an}} = [G_+A_- - F_+B - t_\epsilon t_\Omega(G_-A_- - F_-B)], \tag{A36}$$

$$f_Q^{\mathrm{an}} = [F_+A_+ + G_+B - t_\epsilon t_\Omega(F_-A_+ + G_-B)]. \tag{A37}$$

The coefficients A, B, C are equal to

$$A_\pm = 1 \pm (g_-^R g_+^A + f_-^R f_+^A), \tag{A38}$$

$$B = g_-^R f_+^A + f_-^R g_+^A. \tag{A39}$$

The functions $G_\pm$, $F_\pm$ are defined as

$$G_\pm = g_\epsilon^R \pm g_\epsilon^A,\ F_\pm = f_\epsilon^R \pm f_\epsilon^A. \tag{A40}$$

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