

Nonlinear electromagnetic rectification of BCS-paired electrons at a superconductor surface

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A theory describing nonlinear electromagnetic rectification of BCS-paired electrons at a superconductor-vacuum interface by means of a monochromatic, plane electromagnetic wave incident at an oblique angle is presented. On the basis of a recently constructed nonlinear-response tensor, the forced nonlinear dc-current density is analyzed. A fundamental integro-differential equation incorporating the Meissner screening of the nonlinear response is established, and the prevailing dc-current density and the associated magnetostatic field are determined assuming the BCS-paired electrons to be specularly reflected from the surface. A self-consistency requirement for the forced nonlinear current density which has to be obeyed for any specular-reflection model is derived, and polarization selection rules for the incident electromagnetic field are established. For temperatures above the transition temperature, the present theory describes nonlinear electromagnetic rectification in a collisionless unpaired jellium. In the final part of the present work, the possibilities of achieving nonlinear electromagnetic rectification using incident waves of frequencies around the plasma edge, i.e., far above the superconducting gap frequency, are investigated. It is predicted that polariton-plasmon and plasmon-plasmon interactions can give rise to optical rectification.

I. INTRODUCTION

In the wake of the discovery^{1,2} of high- T_c superconductors, another domain of applications for solid-state optics is emerging. The primary reason that superconductor optics has become of interest stems from the fact that the high- T_c materials have much higher superconducting gap energies than the ones previously known. Hence, it is reasonable to expect optical studies in the infrared and far-infrared wavelength regime to contribute significantly to a better understanding of the electrodynamic properties of the paired many-body state of the electrons. Although the central theme in superconductor optics is concerned with the pairing interaction, it turns out³ that the new superconductors possess a number of other fascinating optical properties: e.g., plasma-edge resonances in the visible or near-infrared region, pronounced anisotropy effects, exotic interband phenomena arising from complicated band structures, and interesting Raman and excitonic interactions.

Recently^{4,5} the present author has suggested that the nonlinear optical properties of the superconducting state be investigated. In centrosymmetric superconductors, even-order nonlinear phenomena are of special importance for studies of the paired-electron state, since the pairing effect is basically of a nonlocal nature and since only nonlocality allows even-order nonlinear effects to occur in the bulk of a centrosymmetric medium.⁶⁻⁹ Thus, within the framework of the BCS-pairing approximation, a qualitative nonlocal microscopic theory of second-harmonic generation has been developed.⁵ Elements of this theory have also been analyzed numerically.^{10,11}

Second-harmonic-generation studies also open up the possibility of investigating the interaction of light with

the superconducting phase in the outermost atomic layers of the surface due to the fact that the inversion symmetry of the paired many-body state is broken in this region. Since nonlinear processes of odd order are allowed in the local (London) limit, these processes are expected *a priori* to be less sensitive to the pairing effect itself. On the basis of the phenomenological Ginzburg-Landau theory of superconductivity,¹² a local theory of third-harmonic generation has also been formulated.¹³ By extending this theory to include spatial changes in the order parameter, it is possible to give a simple phenomenological description of the second-harmonic-generation process.¹⁴ A reduction of the nonlinear, nonlocal theory to the Ginzburg-Landau limit and a comparison of the results obtained in the two formulations remain to be carried out.

Emission of nonlinear light from even-order processes gives rise to a net transfer of momentum to the paired many-body state. This electromagnetic rectification process, which also exists in the normal state, is especially important in the superconducting phase because the dc current generated can flow without resistance. A theory describing nonlinear electromagnetic rectification related to second-harmonic generation has been put forward by the present author.¹⁵ In this theory, special emphasis is devoted to a calculation and a subsequent discussion of the nonlinear, nonlocal response tensor of a homogeneous BCS-paired jellium.

In the present work, I shall undertake a theoretical study of the nonlinear rectification process in the case where a fundamental, plane-wave electromagnetic field is incident on a sharp superconductor-vacuum interface at an oblique angle. The rectification process is termed nonlinear since it exists only if nonlinear processes are taken into account in the field-matter interaction. By incor-

porating only lowest-order nonlinear effects, the nonlinear-response function (the coefficient of rectification) is independent of the intensity of the fundamental field. Three interesting articles,^{16–18} in which experimental observations of transient laser-induced currents in $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$, are reported, are of immediate relevance to the present work. The authors of Ref. 16 attempt to understand their observations with various well-known mechanisms and argue that another hitherto unidentified mechanism must give rise to these laser-induced signals. In Ref. 18, it is concluded that the laser-induced dc-current flow (i) can be induced over a large wavelength range of the incoming electromagnetic field, (ii) is nonlinear at each wavelength, (iii) scales as the sine of the angle of incidence of the laser beam for small angles, and (iv) is electric-dipole forbidden. Although it is not possible at the present stage of the art, both for theoretical (difficult and comprehensive numerical calculations are needed) and experimental (ultrahigh-vacuum experiments are necessary to avoid surface contamination) reasons, to make quantitative comparisons between the theory presented here and the experimental results of Refs. 16–18, it is worth emphasizing that the theory given below is in agreement, from a qualitative point of view, with the requirements of points (i)–(iv) stated above. It is noticed in Ref. 17 that room-temperature measurements suggest that the measured signals scale with the sample resistance as observed in photon-drag studies of tellurium single-crystals.

An alternative method of detecting the electromagnetic rectification phenomenon might be based on the principle that the magnetic flux—and hence the dc current—in a superconducting ring is quantized. Since it is possible to detect a single quantum of flux in a superconducting ring, the smallest possible amount of a rectified current, namely, that stemming from a single-nonlinear-photon process, should thus be detectable. In no other solid-state system does it seem feasible at present to investigate optical rectification processes at this single-event level. Furthermore, one would expect to be able to investigate nonlinear processes in the quantum-optical limit for the intensity of the fundamental beam. Electrodynamical rectification in a superconducting ring should also occur if the incident field has a frequency (far) below the gap frequency. This may imply that low-lying excited states having a net angular momentum and displaced from the ground state by an amount of energy much less than the gap energy can be studied via the nonlinear rectification phenomenon. Recently it has been predicted¹¹ that the nonlinear coefficients describing second-harmonic generation and electromagnetic rectification for frequencies close to the superconducting gap are enhanced by 2 orders of magnitude as the system is cooled below the transition temperature. It is possible to extend the present theory to include effects associated with the variation of the electron density at the surface. In turn, this may enable one to study, on the microscopic level, the profile of the dc-current density in the outermost atomic layers of the surface. Such an analysis might yield a more detailed picture of dc surface currents in conventional-superconductor experiments.

The present paper is organized as follows. In Sec. II, the field-induced dc-current density is studied and explicit expressions for the forced nonlinear current density and the free Meissner current density are given. In Sec. III, a fundamental integro-differential equation for the static vector potential is presented. Next, within the framework of the well-known specular-reflection model,^{19–21} an appropriate integro-differential equation for the vector potential in a semi-infinite superconductor exhibiting translational invariance parallel to the surface plane is established. This integro-differential equation is of major concern in the remaining part of the paper. In Sec. IV, the prevailing nonlinear dc-current density and the associated magnetostatic fields inside and outside the superconductor are calculated. To perform this calculation, it is assumed as an additional boundary condition that the first derivative of the nonlinear current density in the direction perpendicular to the surface vanishes at the surface. An important self-consistency requirement for the forced part of the nonlinear current density is obtained. Furthermore, the relation between the forced current density and the incident field is investigated, and polarization selection rules for the electromagnetic rectification process are established. In Sec. V, we specialize our results to a description of the rectification phenomenon that one would expect in a collisionless normal state. It is of interest to consider the possibilities for generation of nonlinear dc currents by means of optical fields, i.e., fields having frequencies somewhat above the superconducting gap. Thus, in Sec. VI, it is predicted that electromagnetic rectification can be established in a BCS-paired jellium with incident fields having frequencies in the vicinity of the plasma edge via polariton-plasmon and plasmon-plasmon interactions. This conclusion is drawn on the basis of a pole-structure analysis of the forced nonlinear current density. Finally, the electromagnetic rectification process is studied within the framework of a hydrodynamic model.^{21,22} In this model, which is especially adequate for numerical treatments, only the above-mentioned collective excitations are allowed. For temperatures not too close to the superconducting transition temperature, the hydrodynamic model predicts that the nonlinear response function increases proportionally to the square of the gap parameter. This means that one would expect an enhancement of 2 orders of magnitude in the nonlinear coefficient of the high- T_c superconductors relative to that of the lower- T_c ones previously known.

In Ref. 11, in the context of optical second-harmonic generation, some numerical results are presented for the so-called semilocal response tensor in the hydrodynamic approximation. Apart from a sign (see Ref. 5), this semilocal response tensor, which is responsible for a part of the second-harmonic response, is identical to the one used in the present work to describe the electromagnetic rectification process.

II. FIELD-INDUCED dc-CURRENT DENSITIES

A. Forced nonlinear current density

The lowest-order nonlinear interaction between a monochromatic, plane-wave electromagnetic field and an

infinitely extended spatially homogeneous superconductor with BCS pairing has been investigated by the present author.⁵ It was demonstrated that the second-harmonic generation stems from three basic processes: (i) simultaneous two-photon excitation, (ii) double nonlocal excitation, and (iii) semilocal excitation. In comparison, the nonlinear electromagnetic rectification phenomenon was shown to originate only in the semilocal processes.

In a generalized description, valid for a spatially inhomogeneous Cooper-paired superconductor, the forced nonlinear dc-current density, $\mathbf{J}_0^{\text{SL}}(\mathbf{r})$, induced at space point \mathbf{r} by means of semilocal (SL) interaction processes is given by

$$\mathbf{J}_0^{\text{SL}}(\mathbf{r}) = \frac{1}{2} [\mathbf{E}_1^*(\mathbf{r}; \omega) \int_{-\infty}^{\infty} \mathbf{R}_0(\mathbf{r}, \mathbf{r}'; \omega) \cdot \mathbf{E}_1(\mathbf{r}'; \omega) d^3 r' + \text{c. c.}] , \quad (1)$$

where $\mathbf{R}_0(\mathbf{r}, \mathbf{r}'; \omega)$ is a nonlocal vectorial response function relating the nonlinear dc-current density at point \mathbf{r} to the fundamental field, $\mathbf{E}_1(\mathbf{r}'; \omega)$, at neighboring points \mathbf{r}' . In Eq. (1), $\mathbf{E}_1(\mathbf{r}'; \omega)$ is the complex amplitude of an assumed monochromatic fundamental electric field, $\mathbf{E}_1(\mathbf{r}, t)$, viz.,

$$\mathbf{E}_1(\mathbf{r}, t) = \frac{1}{2} [\mathbf{E}_1(\mathbf{r}; \omega) e^{-i\omega t} + \text{c. c.}] , \quad (2)$$

with ω denoting the circular frequency. The semilocal interaction is so named since, in order to obtain the induced current density at \mathbf{r} , one requires the product of the *local* field (complex conjugated) $\mathbf{E}_1^*(\mathbf{r}; \omega)$ and the field $\mathbf{E}_1(\mathbf{r}'; \omega)$ prevailing in *neighboring* points \mathbf{r}' weighted by the response function $\mathbf{R}_0(\mathbf{r}, \mathbf{r}'; \omega)$. The presence of the nonlinear dc response of the superconductor is closely related to the well-known Meissner effect in the sense that the local interaction stems from certain off-diagonal elements of the diamagnetic current-density operator.

For a medium exhibiting translational invariance under infinitesimal displacements perpendicular to the z axis of a Cartesian (x, y, z) coordinate system, the response function has the form

$$\mathbf{R}_0 = \mathbf{R}_0(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z'; \omega) , \quad (3)$$

where $\mathbf{r}_{\parallel} = (x, y, 0)$ and $\mathbf{r}'_{\parallel} = (x', y', 0)$. When the response tensor has the form given in Eq. (3), it is convenient to Fourier transform the constitutive equation (1) in the coordinates perpendicular to the z axis. Hence, we write the quantities $\mathbf{R}_0(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z'; \omega)$ and $\mathbf{E}_1(\mathbf{r}; \omega)$ in the forms

$$\begin{aligned} \mathbf{R}_0(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z'; \omega) \\ = (2\pi)^{-2} \int_{-\infty}^{\infty} \mathbf{R}_0(z, z'; \mathbf{Q}_{\parallel}, \omega) e^{i\mathbf{Q}_{\parallel} \cdot (\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel})} d^2 \mathbf{Q}_{\parallel} \end{aligned} \quad (4)$$

and

$$\mathbf{E}_1(\mathbf{r}; \omega) = (2\pi)^{-2} \int_{-\infty}^{\infty} \mathbf{E}_1(z; \mathbf{Q}_{\parallel}, \omega) e^{i\mathbf{Q}_{\parallel} \cdot \mathbf{r}_{\parallel}} d^2 \mathbf{Q}_{\parallel} , \quad (5)$$

and the semilocal current density as

$$\mathbf{J}_0^{\text{SL}}(\mathbf{r}) = \frac{1}{2} [\mathbf{J}_0^{(+)}(\mathbf{r}) + \mathbf{J}_0^{(-)}(\mathbf{r})] , \quad (6)$$

where

$$\mathbf{J}_0^{(+)}(\mathbf{r}) = (2\pi)^{-2} \int_{-\infty}^{\infty} \mathbf{J}_0^{(+)}(z; \mathbf{Q}_{\parallel}) e^{i\mathbf{Q}_{\parallel} \cdot \mathbf{r}_{\parallel}} d^2 \mathbf{Q}_{\parallel} , \quad (7)$$

and

$$\mathbf{J}_0^{(-)}(\mathbf{r}) = [\mathbf{J}_0^{(+)}(\mathbf{r})]^* . \quad (8)$$

By inserting Eqs. (3)–(8) into Eq. (1), one obtains

$$\mathbf{J}_0^{(+)}(z; \mathbf{q}_{\parallel}) = (2\pi)^{-2} \int_{-\infty}^{\infty} \mathbf{E}_1^*(z; \mathbf{Q}_{\parallel} - \mathbf{q}_{\parallel}, \omega) \int_{-\infty}^{\infty} \mathbf{R}_0(z, z'; \mathbf{Q}_{\parallel}, \omega) \cdot \mathbf{E}_1(z'; \mathbf{Q}_{\parallel}, \omega) dz' d^2 \mathbf{Q}_{\parallel} , \quad (9)$$

and

$$\mathbf{J}_0^{(-)}(z; \mathbf{q}_{\parallel}) = [\mathbf{J}_0^{(+)}(z; -\mathbf{q}_{\parallel})]^* . \quad (10)$$

In the special case where the fundamental field consists of a single Fourier component perpendicular to the z axis, i.e.,

$$\mathbf{E}_1(z'; \mathbf{Q}_{\parallel}, \omega) = \mathbf{E}_1(z'; \mathbf{K}_{\parallel}, \omega) \delta(\mathbf{Q}_{\parallel} - \mathbf{K}_{\parallel}) ,$$

where $\delta(\mathbf{Q}_{\parallel} - \mathbf{K}_{\parallel})$ is the two-dimensional δ function, one has

$$\mathbf{J}_0^{(+)}(z; \mathbf{0}) = (2\pi)^{-2} \mathbf{E}_1^*(z; \mathbf{K}_{\parallel}, \omega) \int_{-\infty}^{\infty} \mathbf{R}_0(z, z'; \mathbf{K}_{\parallel}, \omega) \cdot \mathbf{E}_1(z'; \mathbf{K}_{\parallel}, \omega) dz' . \quad (11)$$

Since only the Fourier component $\mathbf{J}_0^{(+)}(z; \mathbf{0})$ is nonzero in the present case, the nonlinear current density can depend only on z , i.e., $\mathbf{J}_0^{\text{SL}} = \mathbf{J}_0^{\text{SL}}(z)$.

It is of importance for our subsequent analysis of the nonlinear electromagnetic rectification, carried out within the framework of the semiclassical infinite-barrier (SCIB) model,^{19–21} to consider the semilocal response of a homogeneous superconductor. Thus, for a medium exhibiting translational invariance also in the z direction, one has

$$\mathbf{R}_0 = \mathbf{R}_0(\mathbf{r} - \mathbf{r}'; \omega) . \quad (12)$$

Involving now the Fourier transformations

$$\mathbf{E}_1(z; \mathbf{Q}_{\parallel}, \omega) = (2\pi)^{-1} \int_{-\infty}^{\infty} \mathbf{E}_1(Q_{\perp}, \mathbf{Q}_{\parallel}, \omega) e^{iQ_{\perp} z} dQ_{\perp} , \quad (13)$$

$$\mathbf{J}_0^{(+)}(z; \mathbf{Q}_{\parallel}) = (2\pi)^{-1} \int_{-\infty}^{\infty} \mathbf{J}_0^{(+)}(Q_{\perp}, \mathbf{Q}_{\parallel}) e^{iQ_{\perp} z} dQ_{\perp} , \quad (14)$$

and

$$\begin{aligned} \mathbf{R}_0(z - z', \mathbf{Q}_{\parallel}, \omega) = (2\pi)^{-1} \int_{-\infty}^{\infty} \mathbf{R}_0(Q_{\perp}, \mathbf{Q}_{\parallel}, \omega) \\ \times e^{iQ_{\perp}(z - z')} dQ_{\perp} , \end{aligned} \quad (15)$$

one obtains, by combining Eqs. (9) and (13)–(15), the relation

$$\mathbf{J}_0^{(+)}(\mathbf{q}) = (2\pi)^{-3} \int_{-\infty}^{\infty} \mathbf{E}_1^*(\mathbf{Q}-\mathbf{q}, \omega) \times \mathbf{R}_0(\mathbf{Q}, \omega) \cdot \mathbf{E}_1(\mathbf{Q}, \omega) d^3Q \quad (16)$$

with $\mathbf{Q} = \mathbf{Q}_{\parallel} + Q_{\perp} \mathbf{e}_z$ and $\mathbf{q} = \mathbf{q}_{\parallel} + q_{\perp} \mathbf{e}_z$, \mathbf{e}_z being a unit vector in the z direction. If the fundamental field is just a monochromatic plane wave, i.e., $\mathbf{E}_1(\mathbf{Q}, \omega) = \mathbf{E}_1(\mathbf{K}, \omega) \delta(\mathbf{Q} - \mathbf{K})$, Eq. (16) is reduced to

$$\mathbf{M}_0(\mathbf{Q}, \omega) = \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{Q}) \left[(u_{\mathbf{k}}^2 u_{\mathbf{k}+\mathbf{Q}}^2 - v_{\mathbf{k}}^2 v_{\mathbf{k}+\mathbf{Q}}^2) \left[\frac{1}{\hbar\omega + E_{\mathbf{k}} - E_{\mathbf{k}+\mathbf{Q}}} + \frac{1}{\hbar\omega - E_{\mathbf{k}} + E_{\mathbf{k}+\mathbf{Q}}} \right] (f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{Q}}) \right. \\ \left. + (v_{\mathbf{k}}^2 u_{\mathbf{k}+\mathbf{Q}}^2 - u_{\mathbf{k}}^2 v_{\mathbf{k}+\mathbf{Q}}^2) \left[\frac{1}{\hbar\omega - E_{\mathbf{k}} - E_{\mathbf{k}+\mathbf{Q}}} + \frac{1}{\hbar\omega + E_{\mathbf{k}} + E_{\mathbf{k}+\mathbf{Q}}} \right] (1 - f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{Q}}) \right]. \quad (19)$$

In Eqs. (18) and (19), $-e$, m , and \mathbf{k} denote, respectively, the charge, the mass, and the wave vector of the electron. The normalization volume of the superconductor is V . In the derivation of Eq. (19), relaxation phenomena, e.g., impurity scattering, have been neglected. This implies that \mathbf{M}_0 and hence \mathbf{R}_0 are real quantities. The probability amplitudes u_{κ} and v_{κ} for the pair state ($\kappa \uparrow, -\kappa \downarrow$) being empty and full, respectively, and the quasiparticle distribution function f_{κ} can be expressed in terms of the gap-dependent quasiparticle excitation energy $E_{\kappa}(\kappa = \mathbf{k}$ or $\mathbf{k} + \mathbf{q})$ in the usual way.

B. Free Meissner current density

The forced dc current density, $\mathbf{J}_0^{\text{SL}}(\mathbf{r})$, generated by the fundamental electromagnetic field will give rise to a dc magnetic field inside the superconductor. In turn, this magnetic field will be partly screened by the induction of a linear dc-current density in the vicinity of the surface. This, in the present context, the so-called free dc-current density (or Meissner current density), is responsible for the famous Meissner effect²³ in the linear electrodynamics of superconductors. The Meissner current density is linearly and nonlocally related to the prevailing magnetic field inside the superconductor. To determine the prevailing magnetic field, a self-consistent solution to the electrodynamic problem must be determined (cf. Sec. III). By denoting the self-consistent solution for the dc vector potential (in a suitable gauge) by $\mathbf{A}_0(\mathbf{r})$, the Meissner (\mathbf{M}) current density at space point \mathbf{r} is given by

$$\mathbf{J}_0^{(+)}(\mathbf{0}) = (2\pi)^{-3} \mathbf{E}_1^*(\mathbf{K}, \omega) \mathbf{R}_0(\mathbf{K}, \omega) \cdot \mathbf{E}_1(\mathbf{K}, \omega). \quad (17)$$

The semilocal response tensor $\mathbf{R}_0(\mathbf{Q}, \omega)$ has been calculated¹⁵ within the framework of the pairing approximation. The result obtained for a Cooper-paired jellium is the following:

$$\mathbf{R}_0(\mathbf{Q}, \omega) = -\frac{\hbar e^3}{4m^2 \omega^2 V} \mathbf{M}_0(\mathbf{Q}, \omega), \quad (18)$$

where

$$\mathbf{J}_0^{\text{M}}(\mathbf{r}) = \int_{-\infty}^{\infty} \vec{\mathbf{S}}_0(\mathbf{r}, \mathbf{r}') \cdot \mathbf{A}_0(\mathbf{r}') d^3r'. \quad (20)$$

In the case where the superconductor exhibits infinitesimal translational invariance parallel to the plane $z=0$, the response tensor has the form $\vec{\mathbf{S}}_0(\mathbf{r}, \mathbf{r}') \equiv \vec{\mathbf{S}}_0(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z')$. By writing the components of $\mathbf{J}_0^{\text{M}}(\mathbf{r})$, $\vec{\mathbf{S}}_0(\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}, z, z')$, and $\mathbf{A}_0(\mathbf{r}')$ in the generic form

$$\mathbf{Z}[\mathbf{R}_{\parallel}, z, (z')] = (2\pi)^{-2} \int_{-\infty}^{\infty} \mathbf{Z}[z, (z'); \mathbf{Q}_{\parallel}] e^{i\mathbf{Q}_{\parallel} \cdot \mathbf{R}_{\parallel}} d^2Q_{\parallel}, \quad (21)$$

where $\mathbf{R}_{\parallel} = \mathbf{r}_{\parallel}, \mathbf{r}'_{\parallel}$, or $\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}$, one obtains in the mixed Fourier representation

$$\mathbf{J}_0^{\text{M}}(z; \mathbf{Q}_{\parallel}) = \int_{-\infty}^{\infty} \vec{\mathbf{S}}_0(z, z'; \mathbf{Q}_{\parallel}) \cdot \mathbf{A}_0(z'; \mathbf{Q}_{\parallel}) dz'. \quad (22)$$

In the case of complete translational invariance, i.e., for $\vec{\mathbf{S}}_0(\mathbf{r}, \mathbf{r}') = \vec{\mathbf{S}}_0(\mathbf{r} - \mathbf{r}')$, one has, by introducing

$$\mathbf{Z}(R_{\perp}, \mathbf{R}_{\parallel}) = (2\pi)^{-1} \int_{-\infty}^{\infty} \mathbf{Z}(Q_{\perp}, \mathbf{Q}_{\parallel}) e^{iQ_{\perp} R_{\perp}} dQ_{\perp}, \quad (23)$$

the algebraic relation

$$\mathbf{J}_0^{\text{M}}(\mathbf{Q}) = \vec{\mathbf{S}}_0(\mathbf{Q}) \cdot \mathbf{A}_0(\mathbf{Q}). \quad (24)$$

Within the pairing approximation, the linear response tensor $\vec{\mathbf{S}}_0(\mathbf{Q})$ is given by the well-known expression²⁴

$$\vec{\mathbf{S}}_0(\mathbf{Q}) = -\frac{ne^2}{m} \vec{\mathbf{1}} + \left[\frac{e\hbar}{2m} \right]^2 \frac{2}{V} \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{Q}) \otimes (2\mathbf{k} + \mathbf{Q}) \left[(u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{Q}} + v_{\mathbf{k}} v_{\mathbf{k}+\mathbf{Q}})^2 \frac{f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{Q}}}{E_{\mathbf{k}+\mathbf{Q}} - E_{\mathbf{k}}} \right. \\ \left. + (u_{\mathbf{k}+\mathbf{Q}} v_{\mathbf{k}} - v_{\mathbf{k}+\mathbf{Q}} u_{\mathbf{k}})^2 \frac{1 - f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{Q}}}{E_{\mathbf{k}+\mathbf{Q}} + E_{\mathbf{k}}} \right], \quad (25)$$

where n is the conduction-electron density, $\vec{\mathbb{1}}$ is the unit tensor, and \otimes stands for the tensor product.

III. FUNDAMENTAL INTEGRO-DIFFERENTIAL EQUATION FOR THE STATIC VECTOR POTENTIAL

A. General case

In order to determine the Meissner screening of the nonlinear dc-current density induced by the fundamental electromagnetic field a self-consistent solution for the vector potential $\mathbf{A}_0(\mathbf{r})$ is needed, cf. Eq. (20). Once $\mathbf{A}_0(\mathbf{r})$ is obtained, the prevailing dc-current density

$$\mathbf{J}_0(\mathbf{r}) = \mathbf{J}_0^{\text{SL}}(\mathbf{r}) + \mathbf{J}_0^{\text{M}}(\mathbf{r}) \quad (26)$$

inside the superconductor can be found. By combining the magnetostatic Maxwell equation $\nabla \times \mathbf{B}_0(\mathbf{r}) = \mu_0 \mathbf{J}_0(\mathbf{r})$, where $\mathbf{B}_0(\mathbf{r}) = \nabla \times \mathbf{A}_0(\mathbf{r})$, with the explicit constitutive equation for the Meissner current density [Eq. (20)], one obtains the following inhomogeneous integro-differential equation for the vector potential:

$$\nabla \times [\nabla \times \mathbf{A}_0(\mathbf{r})] - \mu_0 \int_{-\infty}^{\infty} \vec{\mathbb{S}}_0(\mathbf{r}, \mathbf{r}') \cdot \mathbf{A}_0(\mathbf{r}') d^3 r' = \mu_0 \mathbf{J}_0^{\text{SL}}(\mathbf{r}) . \quad (27)$$

With a knowledge of the fundamental electric field inside

$$(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \frac{\partial^2 \mathbf{A}_0(z; \mathbf{0})}{\partial z^2} + \mu_0 \int_0^{\infty} \vec{\mathbb{S}}_0(z, z'; \mathbf{0}) \cdot \mathbf{A}_0(z'; \mathbf{0}) dz' + \frac{\mu_0}{2} [\mathbf{J}_0^{(+)}(z; \mathbf{0}) + (\mathbf{J}_0^{(+)}(z; \mathbf{0}))^*] = 0 , \quad (29)$$

where, cf. Eq. (11),

$$\mathbf{J}_0^{(+)}(z; \mathbf{0}) = (2\pi)^{-2} \mathbf{E}_1^*(z; \mathbf{K}_{\parallel}, \omega) \int_0^{\infty} \mathbf{R}_0(z, z'; \mathbf{K}_{\parallel}, \omega) \cdot \mathbf{E}_1(z'; \mathbf{K}_{\parallel}, \omega) dz' . \quad (30)$$

We emphasize here that although not indicated explicitly in the notation, $\mathbf{J}_0^{(+)}(z, \mathbf{0})$ is a function of both \mathbf{K}_{\parallel} and ω . In turn, the nonlinear dc-vector potential $\mathbf{A}_0(z; \mathbf{0})$ will, of course, depend on the frequency and the angle of incidence of the fundamental field.

C. Specular-reflection model

In order to determine the vector potential $\mathbf{A}_0(z; \mathbf{0})$ in the vicinity of the superconductor-vacuum interface, the surface-sensitive response functions $\vec{\mathbb{S}}_0(z, z'; \mathbf{0})$ and $\mathbf{R}_0(z, z'; \mathbf{K}_{\parallel}, \omega)$ have to be calculated. In the present work, a widely used phenomenological microscopic surface model, namely, the so-called specular-reflection model,^{19–21} is the basis for the determination of the response functions. The specular-reflection model allows us to express the surface response in terms of the bulk response tensors which in Fourier-transformed form are given by Eqs. (18) [with (19)] and (25), using the appropriate values for \mathbf{Q} . Although it would be highly desirable to construct a theory that goes beyond the specular-reflection model, it is felt that substantial insight can be gained with use of

the superconductor, the driven nonlinear current density $\mathbf{J}_0^{\text{SL}}(\mathbf{r})$ is a prescribed function of \mathbf{r} .

B. Semi-infinite superconductor with translational invariance parallel to the surface

It is not possible in the general case to obtain a closed-form solution for the vector potential in Eq. (27). Hence, to make progress in our basic physical understanding of the nonlinear electromagnetic rectification phenomenon we shall consider the case where the superconductor occupies the half-space $z > 0$ [in a Cartesian (x, y, z) coordinate system], the rest of the space being vacuum. Furthermore, we shall assume that the superconductor exhibits translationally invariant properties parallel to the surface. Now, if the superconducting electrons interact with a plane, monochromatic electromagnetic wave of frequency ω incident from vacuum at an oblique angle $\theta = \arcsin(c_0 \mathbf{K}_{\parallel} / \omega)$, where \mathbf{K}_{\parallel} is the component of the wave vector parallel to the surface, the fundamental electric field inside the superconductor is given by

$$\mathbf{E}_1(\mathbf{r}, t) = \frac{1}{2} [(2\pi)^{-2} \mathbf{E}_1(z; \mathbf{K}_{\parallel}, \omega) e^{i(\mathbf{K}_{\parallel} \cdot \mathbf{r}_{\parallel} - \omega t)} + \text{c.c.}] . \quad (28)$$

Since the vector potential in the present case is a function of z only, one obtains in the mixed Fourier representation the following integro-differential equation for the self-consistent vector potential:

a phenomenological microscopic surface model for which many calculations can be done analytically.

In the specular-reflection model, quantum interference effects at the surface are neglected.^{19,25} Consequently, the response functions are given by

$$\vec{\mathbb{S}}_0(z, z'; \mathbf{0}) = \vec{\mathbb{S}}_0(z - z', \mathbf{0}) + \vec{\mathbb{S}}_0(z + z', \mathbf{0}) \cdot \vec{\alpha} \quad (31)$$

and

$$\mathbf{R}_0(z, z'; \mathbf{K}_{\parallel}, \omega) = \mathbf{R}_0(z - z'; \mathbf{K}_{\parallel}, \omega) + \vec{\alpha} \cdot \mathbf{R}_0(z + z'; \mathbf{K}_{\parallel}, \omega) , \quad (32)$$

where

$$\vec{\alpha} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix} . \quad (33)$$

To solve Eq. (29) within the framework of the semiclassical infinite-barrier (SCIB) model, i.e., the specular-reflection model with the neglect of quantum interference effects at the surface, we introduce the effective fields

$$\mathbf{A}_0^{\text{eff}}(z; \mathbf{0}) = \Theta(z) \mathbf{A}_0(z; \mathbf{0}) + \Theta(-z) \vec{\alpha} \cdot \mathbf{A}_0(-z; \mathbf{0}), \quad (34)$$

and

$$\begin{aligned} \mathbf{E}_1^{\text{eff}}(z; \mathbf{K}_{\parallel}, \omega) &= \Theta(z) \mathbf{E}_1(z; \mathbf{K}_{\parallel}, \omega) \\ &+ \Theta(-z) \vec{\alpha} \cdot \mathbf{E}_1(-z; \mathbf{K}_{\parallel}, \omega), \end{aligned} \quad (35)$$

where Θ is the Heaviside unit-step function.

$$\mathbf{J}_0^{(+), \text{eff}}(z; \mathbf{0}) = (2\pi)^{-2} [\mathbf{E}_1^{\text{eff}}(z; \mathbf{K}_{\parallel}, \omega)]^* \int_{-\infty}^{\infty} \mathbf{R}_0(z-z'; \mathbf{K}_{\parallel}, \omega) \cdot \mathbf{E}_1^{\text{eff}}(z'; \mathbf{K}_{\parallel}, \omega) dz' . \quad (37)$$

On the basis of the above considerations, it is realized that the integro-differential equation

$$\begin{aligned} (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \frac{\partial^2 \mathbf{A}_0^{\text{eff}}(z; \mathbf{0})}{\partial z^2} + \mu_0 \int_{-\infty}^{\infty} \vec{\mathbb{S}}_0(z-z'; \mathbf{0}) \cdot \mathbf{A}_0^{\text{eff}}(z'; \mathbf{0}) dz' \\ + \frac{\mu_0}{2} \left\{ (2\pi)^{-2} [\mathbf{E}_1^{\text{eff}}(z; \mathbf{K}_{\parallel}, \omega)]^* \int_{-\infty}^{\infty} \mathbf{R}_0(z-z'; \mathbf{K}_{\parallel}, \omega) \cdot \mathbf{E}_1^{\text{eff}}(z'; \mathbf{K}_{\parallel}, \omega) dz' + \text{c. c.} \right\} = 0, \end{aligned} \quad (38)$$

within the framework of the SCIB model, has the same solutions as Eq. (29) in the half-space $z > 0$.

IV. PREVAILING MAGNETOSTATIC FIELDS AND dc-CURRENT DENSITIES

In the following we shall, taking Eq. (38) as a starting point, determine and discuss the prevailing magnetostatic fields inside and outside the superconductor and the associated nonlinear dc-current density in the Cooper-paired jellium.

A. Nonlinear static vector potential

To solve Eq. (38), it is convenient to introduce a Fourier decomposition of $\mathbf{A}_0^{\text{eff}}(z; \mathbf{0})$, viz.,

$$\mathbf{A}_0^{\text{eff}}(z; \mathbf{0}) = (2\pi)^{-1} \int_{-\infty}^{\infty} \mathbf{A}_0^{\text{eff}}(q_{\perp}, \mathbf{0}) e^{iq_{\perp}z} dq_{\perp}. \quad (39)$$

By means of the inverse transformation of that in Eq. (39) and the corresponding inverse transformations of

$$\mathbf{J}_0^{(+)}(q_{\perp}, \mathbf{0}) = (2\pi)^{-3} \int_{-\infty}^{\infty} [\mathbf{E}_1^{\text{eff}}(K_{\perp} - q_{\perp}, \mathbf{K}_{\parallel}, \omega)]^* \mathbf{R}_0(K_{\perp}, \mathbf{K}_{\parallel}, \omega) \cdot \mathbf{E}_1^{\text{eff}}(K_{\perp}, \mathbf{K}_{\parallel}, \omega) dK_{\perp}. \quad (44)$$

For notational simplicity we have omitted to add a superscript “eff” to $\mathbf{J}_0^{(+)}(q_{\perp}, \mathbf{0})$ and $\mathbf{J}_0^{\text{SL}}(q_{\perp}, \mathbf{0})$. It appears from Eqs. (39)–(44) that the dc-vector potential inside the superconductor in the specular reflection model can be expressed in terms of the *bulk* response functions, the actual wave vectors being $q_{\perp} \mathbf{e}_z$ and $\mathbf{K} = K_{\perp} \mathbf{e}_z + \mathbf{K}_{\parallel}$ for $\vec{\mathbb{S}}_0$ and \mathbf{R}_0 , respectively. Hence, in the following we shall use the notation $\vec{\mathbb{S}}_0(q_{\perp}, \mathbf{0}) \equiv \vec{\mathbb{S}}_0(q_{\perp} \mathbf{e}_z)$, $\mathbf{R}_0(K_{\perp}, \mathbf{K}_{\parallel}, \omega) \equiv \mathbf{R}_0(\mathbf{K}, \omega)$, and also $\vec{\mathbb{G}}_0(q_{\perp}, \mathbf{0}) \equiv \vec{\mathbb{G}}_0(q_{\perp} \mathbf{e}_z)$, $\mathbf{J}_0^{\text{SL}}(q_{\perp}, \mathbf{0}) \equiv \mathbf{J}_0^{\text{SL}}(q_{\perp} \mathbf{e}_z)$, $\mathbf{J}_0^{(+)}(q_{\perp}, \mathbf{0}) \equiv \mathbf{J}_0^{(+)}(q_{\perp} \mathbf{e}_z)$, $\mathbf{A}_0^{\text{eff}}(q_{\perp}, \mathbf{0}) \equiv \mathbf{A}_0^{\text{eff}}(q_{\perp} \mathbf{e}_z)$, and $\mathbf{E}_1^{\text{eff}}(K_{\perp} \mp q_{\perp}, \mathbf{K}_{\parallel}, \omega) \equiv \mathbf{E}_1^{\text{eff}}(\mathbf{K} \mp q_{\perp} \mathbf{e}_z, \omega)$.

It is a straightforward matter to show from Eq. (25)

The SCIB model allows us to rewrite the Meissner and semilocal current densities in terms of the effective fields. Thus, one has

$$\mathbf{J}_0^{M, \text{eff}}(z; \mathbf{0}) = \int_{-\infty}^{\infty} \vec{\mathbb{S}}_0(z-z'; \mathbf{0}) \cdot \mathbf{A}_0^{\text{eff}}(z'; \mathbf{0}) dz', \quad (36)$$

and $\mathbf{J}_0^{\text{SL}, \text{eff}}(z; \mathbf{0}) = \frac{1}{2} [\mathbf{J}_0^{(+), \text{eff}}(z; \mathbf{0}) + \text{c. c.}]$, with

$\mathbf{E}_1^{\text{eff}}(z; \mathbf{K}_{\parallel}, \omega)$, $\mathbf{R}_0(z-z'; \mathbf{K}_{\parallel}, \omega)$, and $\vec{\mathbb{S}}_0(z-z'; \mathbf{0})$ one can derive an expression for the Fourier amplitude $\mathbf{A}_0^{\text{eff}}(q_{\perp}, \mathbf{0})$. Thus, by multiplying Eq. (38) with $\exp(-iq_{\perp}z)$ and integrating over z from $-\infty$ to ∞ , one obtains the following result for the Fourier amplitude of the dc-vector potential:

$$\mathbf{A}_0^{\text{eff}}(q_{\perp}, \mathbf{0}) = \vec{\mathbb{G}}_0(q_{\perp}, \mathbf{0}) \cdot [\mathbf{g}_0 - \mu_0 \mathbf{J}_0^{\text{SL}}(q_{\perp}, \mathbf{0})], \quad (40)$$

where

$$\vec{\mathbb{G}}_0(q_{\perp}, \mathbf{0}) = [(\mathbf{e}_z \otimes \mathbf{e}_z - \vec{\mathbb{1}}) q_{\perp}^2 + \mu_0 \vec{\mathbb{S}}_0(q_{\perp}, \mathbf{0})]^{-1}, \quad (41)$$

$$\mathbf{g}_0 = 2 \begin{bmatrix} \partial A_{0,x}(z \rightarrow 0^+; \mathbf{0}) / \partial z \\ \partial A_{0,y}(z \rightarrow 0^+; \mathbf{0}) / \partial z \\ 0 \end{bmatrix}, \quad (42)$$

and

$$\mathbf{J}_0^{\text{SL}}(q_{\perp}, \mathbf{0}) = \frac{1}{2} \{ \mathbf{J}_0^{(+)}(q_{\perp}, \mathbf{0}) + [\mathbf{J}_0^{(+)}(-q_{\perp}, \mathbf{0})]^* \} \quad (43)$$

with

that $\vec{\mathbb{S}}_0(q_{\perp} \mathbf{e}_z)$ is in diagonal form in our Cartesian (x, y, z) coordinate system. By means of the transverse (T) $S_0^{\text{T}}(q_{\perp})$ and longitudinal (L) $S_0^{\text{L}}(q_{\perp})$ response functions, which are functions of the scalar quantity q_{\perp} , only, it is realized that $\vec{\mathbb{G}}_0(q_{\perp} \mathbf{e}_z)$ can be written in the form

$$\vec{\mathbb{G}}_0(q_{\perp} \mathbf{e}_z) = \frac{\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z}{\mathcal{N}_0^{\text{T}}(q_{\perp})} + \frac{\mathbf{e}_z \otimes \mathbf{e}_z}{\mathcal{N}_0^{\text{L}}(q_{\perp})}, \quad (45)$$

where

$$\mathcal{N}_0^T(q_\perp) = \mu_0 \mathcal{S}_0^T(q_\perp) - q_\perp^2, \quad (46)$$

$$\mathcal{N}_0^L(q_\perp) = \mu_0 \mathcal{S}_0^L(q_\perp). \quad (47)$$

One should notice that $\vec{G}_0(q_\perp \mathbf{e}_z)$ plays the role as a Fourier-transformed magnetostatic Green's function, re-

lating the vector potential to the source current density in Fourier space.

The magnetostatic vector potential inside ($z > 0$) the superconductor now can be obtained inserting Eq. (40) [with Eq. (45)] into Eq. (39). Since $\mathbf{A}_0^{\text{eff}}(z > 0; \mathbf{0}) = \mathbf{A}_0(z; \mathbf{0})$, we obtain

$$\mathbf{A}_0(z; \mathbf{0}) = (2\pi)^{-1} \int_{-\infty}^{\infty} \left[\frac{\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z}{\mathcal{N}_0^T(q_\perp)} + \frac{\mathbf{e}_z \otimes \mathbf{e}_z}{\mathcal{N}_0^L(q_\perp)} \right] \cdot [\mathbf{g}_0 - \mu_0 \mathbf{J}_0^{\text{SL}}(q_\perp \mathbf{e}_z)] e^{iq_\perp z} dq_\perp, \quad z > 0, \quad (48)$$

or, equivalently,

$$\mathbf{A}_0(z; \mathbf{0}) = (2\pi)^{-1} \int_{-\infty}^{\infty} \left[\frac{\mathbf{g}_0 - \mu_0 \mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z)}{\mathcal{N}_0^T(q_\perp)} - \frac{\mu_0 \mathbf{J}_\perp^{\text{SL}}(q_\perp \mathbf{e}_z)}{\mathcal{N}_0^L(q_\perp)} \right] e^{iq_\perp z} dq_\perp, \quad (49)$$

where $\mathbf{J}_\parallel^{\text{SL}} = (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{\text{SL}}$ and $\mathbf{J}_\perp^{\text{SL}} = \mathbf{e}_z \cdot (\mathbf{e}_z \cdot \mathbf{J}_0^{\text{SL}})$ are the semilocal current densities parallel (\parallel) and perpendicular (\perp) to the surface, respectively.

B. Self-consistency requirement

If one calculates

$$\lim_{z \rightarrow 0^+} \left[\frac{\partial}{\partial z} [(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{A}_0(z; \mathbf{0})] \right]$$

from Eq. (49) and combines the result with Eq. (42), one realizes that in order for our theory to be self-consistent we must require

$$\mathbf{g}_0 \left[\frac{i}{\pi} \int_{-\infty}^{\infty} \frac{q_\perp e^{iq_\perp 0^+}}{\mathcal{N}_0^T(q_\perp)} dq_\perp - 1 \right] = \frac{i\mu_0}{\pi} \int_{-\infty}^{\infty} \frac{q_\perp \mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z)}{\mathcal{N}_0^T(q_\perp)} e^{iq_\perp 0^+} dq_\perp, \quad (50)$$

using the shorthand notation

$$\lim_{z \rightarrow 0^+} \int (\dots) e^{iq_\perp z} dq_\perp \equiv \int (\dots) e^{iq_\perp 0^+} dq_\perp.$$

The self-consistency requirement in Eq. (50) can be simplified considerably utilizing that

$$\int_{-\infty}^{\infty} F(q_\perp) \frac{e^{iq_\perp 0^+}}{q_\perp} dq_\perp = \pi i \lim_{q_\perp \rightarrow \infty} F(q_\perp), \quad (51)$$

if F is an even function of q_\perp . Now, since $q_\perp^2 / \mathcal{N}_0^T(q_\perp)$ is an even function of q_\perp with the limiting value -1 for $q_\perp \rightarrow \infty$ [$\mathcal{S}_0^T(q_\perp) \rightarrow -ne^2/m$ for $q_\perp \rightarrow \infty$], one has

$$\frac{i}{\pi} \int_{-\infty}^{\infty} \frac{q_\perp e^{iq_\perp 0^+}}{\mathcal{N}_0^T(q_\perp)} dq_\perp = 1. \quad (52)$$

By comparing Eqs. (50) and (52), it is realized that the self-consistency relation can be written as

$$\int_{-\infty}^{\infty} \frac{q_\perp \mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z)}{\mathcal{N}_0^T(q_\perp)} e^{iq_\perp 0^+} dq_\perp = \mathbf{0}. \quad (53)$$

A final simplification of Eq. (53) can be achieved by noting that

$$\mathbf{J}_\parallel^{\text{SL}}(-q_\perp \mathbf{e}_z) = \mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z).$$

The quantity $\mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z)$ is an even function of q_\perp , since the effective semilocal current density parallel to the surface is an even function of z , i.e.,

$$\mathbf{J}_0^{\text{SL,eff}}(z; \mathbf{0}) = \mathbf{J}_0^{\text{SL,eff}}(-z; \mathbf{0}).$$

Since

$$q_\perp^2 \mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z) / \mathcal{N}_0^T(q_\perp)$$

is an even function of q_\perp , Eq. (51) thus shows that the self-consistency relation in Eq. (53) can be reformulated in the form

$$\lim_{q_\perp \rightarrow \infty} \mathbf{J}_\parallel^{\text{SL}}(q_\perp \mathbf{e}_z) = \mathbf{0}. \quad (54)$$

The requirement in Eq. (54) is fulfilled for the collective-mode model discussed in Sec. VI.

C. Additional boundary conditions and nonlinear current density

A complete determination of the nonlinear vector potential $\mathbf{A}_0(z; \mathbf{0})$ [Eq. (49)] requires a knowledge of $\mathbf{g}_0 = (g_{0,x}, g_{0,y}, 0)$. Once \mathbf{g}_0 is known, the static vector potential outside the superconductor can be calculated. To obtain \mathbf{g}_0 , the so-called additional boundary conditions (ABC's) are needed. For the specular-reflection model, these are

$$\lim_{z \rightarrow 0^+} [\mathbf{e}_z \cdot \mathcal{J}_0(z; \mathbf{0})] = 0 \quad (55)$$

and

$$\lim_{z \rightarrow 0^+} \left[(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \frac{d\mathcal{J}_0(z; \mathbf{0})}{dz} \right] = 0, \quad (56)$$

where

$$\mathcal{J}_0(z; \mathbf{0}) = \mathbf{J}_0^M(z; \mathbf{0}) + \mathbf{J}_0^{\text{SL}}(z; \mathbf{0}) \quad (57)$$

is the total current density at a distance z from the surface.

Let us demonstrate that the condition [Eq. (55)] that the normal component $\mathcal{J}_0^\perp(z; \mathbf{0}) \equiv \mathbf{e}_z \cdot \mathcal{J}_0(z; \mathbf{0})$ of the total current density vanishes at the surface is already fulfilled, using the self-consistent vector potential in Eq. (49). Since

$$\mathcal{J}_0^\perp(z; \mathbf{0}) = \int_{-\infty}^{\infty} S_0^\perp(z-z'; \mathbf{0}) A_{0,z}(z'; \mathbf{0}) dz' + \mathbf{J}_1^{\text{SL}}(z; \mathbf{0}), \quad (58)$$

where $\mathbf{J}_1^{\text{SL}}(z; \mathbf{0}) = \mathbf{e}_z \cdot \mathbf{J}_0^{\text{SL}}(z; \mathbf{0})$, as one readily realizes via Eq. (36), one obtains by taking $A_{0,z}(z'; \mathbf{0})$ from Eq. (49)

$$\mathcal{J}_0^\perp(z; \mathbf{0}) = -\frac{\mu_0}{2\pi} \int_{-\infty}^{\infty} \frac{\mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)}{\mathcal{N}_0^\perp(q_1)} \left[\int_{-\infty}^{\infty} S_0^\perp(z-z'; \mathbf{0}) e^{-iq_1(z-z')} dz' \right] e^{iq_1 z} dq_1 + \mathbf{J}_1^{\text{SL}}(z; \mathbf{0}). \quad (59)$$

Because the integral over z' equals $S_0^\perp(q_1)$, it follows immediately from Eq. (47) and the Fourier expansion of $\mathbf{J}_1^{\text{SL}}(z; \mathbf{0})$ that one has

$$\mathcal{J}_0^\perp(z; \mathbf{0}) = 0. \quad (60)$$

We have demonstrated now that the total dc-current density perpendicular to the surface is zero everywhere and thus also at the surface [Eq. (55)].

It is expected that the current density is independent of z , since the equation of continuity $\nabla \cdot \mathcal{J}_0 = 0$ requires that $\partial \mathcal{J}_0(z; \mathbf{0}) / \partial z = 0$.

The additional boundary condition in Eq. (56) that requires that the derivative with respect to z of the tangen-

tial component,

$$\mathcal{J}_0^\parallel(z; \mathbf{0}) = (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathcal{J}_0(z; \mathbf{0}),$$

of the total current density vanishes at the surface enables us to determine \mathbf{g}_0 . To calculate \mathbf{g}_0 , we take as a starting point the following equation for the total current density parallel to the surface:

$$\mathcal{J}_0^\parallel(z; \mathbf{0}) = \int_{-\infty}^{\infty} S_0^\parallel(z-z'; \mathbf{0}) (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{A}_0(z'; \mathbf{0}) dz' + \mathbf{J}_1^{\text{SL}}(z; \mathbf{0}). \quad (61)$$

By inserting the expression in Eq. (49) for $\mathbf{A}_0(z'; \mathbf{0})$ into Eq. (61) and changing the order of integration one obtains

$$\mathcal{J}_0^\parallel(z; \mathbf{0}) = (2\pi)^{-1} \int_{-\infty}^{\infty} \frac{\mathbf{g}_0 - \mu_0 \mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)}{\mathcal{N}_0^\parallel(q_1)} \left[\int_{-\infty}^{\infty} S_0^\parallel(z-z'; \mathbf{0}) e^{-iq_1(z-z')} dz' \right] e^{iq_1 z} dq_1 + \mathbf{J}_1^{\text{SL}}(z; \mathbf{0}). \quad (62)$$

Utilizing Eq. (46) and that the integral over z' equals $S_0^\parallel(q_1)$, Eq. (62) can be written as

$$\mathcal{J}_0^\parallel(z; \mathbf{0}) = \frac{\mathbf{g}_0}{2\pi} \int_{-\infty}^{\infty} \frac{S_0^\parallel(q_1)}{\mathcal{N}_0^\parallel(q_1)} e^{iq_1 z} dq_1 - \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{q_1^2 \mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)}{\mathcal{N}_0^\parallel(q_1)} e^{iq_1 z} dq_1. \quad (63)$$

By calculating $d\mathcal{J}_0^\parallel(z; \mathbf{0})/dz$, by noting that the functions $q_1^2 S_0^\parallel(q_1)/\mathcal{N}_0^\parallel(q_1)$ and $q_1^4 \mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)/\mathcal{N}_0^\parallel(q_1)$ are even functions of q_1 , and by utilizing that $q_1^2 S_0^\parallel(q_1)/\mathcal{N}_0^\parallel(q_1) \rightarrow ne^2/m$ for $q_1 \rightarrow \infty$, it follows, via Eq. (51), that

$$\frac{d\mathcal{J}_0^\parallel(z; \mathbf{0})}{dz} \Big|_{z \rightarrow 0^+} = -\frac{ne^2}{2m} \mathbf{g}_0 + \frac{1}{2} \lim_{q_1 \rightarrow \infty} \left[\frac{q_1^4 \mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)}{\mathcal{N}_0^\parallel(q_1)} \right], \quad (64)$$

and thus via the ABC in Eq. (56)

$$\mathbf{g}_0 = -\frac{m}{ne^2} \lim_{q_1 \rightarrow \infty} [q_1^2 \mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)]. \quad (65)$$

By inserting Eq. (65) into Eq. (49), the nonlinear dc-vector potential $\mathbf{A}_0(z; \mathbf{0})$ inside the superconductor has been completely determined in terms of the prescribed Fourier components $\mathbf{J}_0^{\text{SL}}(q_1 \mathbf{e}_z)$ of the forced nonlinear current density.

D. Nonlinear magnetic field inside and outside the superconductor

The self-consistent nonlinear dc magnetic field inside the superconductor, $\mathbf{B}_0(z)$, is obtained by inserting Eq. (49) into the expression $\mathbf{B}_0(z) = \nabla \times \mathbf{A}_0(z; \mathbf{0})$. Thus,

$$\mathbf{B}_0(z) = \frac{i}{2\pi} \mathbf{e}_z \times \int_{-\infty}^{\infty} [\mathbf{g}_0 - \mu_0 \mathbf{J}_1^{\text{SL}}(q_1 \mathbf{e}_z)] \frac{q_1}{\mathcal{N}_0^\parallel(q_1)} e^{iq_1 z} dq_1. \quad (66)$$

One should notice that $\mathbf{B}_0(z)$ does only depend on the component of the forced nonlinear current density parallel to the surface. This is to be expected since the *total* current flows parallel to the surface. The magnetic field just inside the surface is given by

$$\mathbf{B}_0(z \rightarrow 0^+) = \frac{i}{2\pi} \mathbf{e}_z \times \int_{-\infty}^{\infty} [\mathbf{g}_0 - \mu_0 \mathbf{J}_{\parallel}^{\text{SL}}(q_{\perp} \mathbf{e}_z)] \frac{q_{\perp}^2}{\mathcal{N}_0^{\text{T}}(q_{\perp})} \frac{e^{iq_{\perp} 0^+}}{q_{\perp}} dq_{\perp}. \quad (67)$$

By means of Eq. (51) and the self-consistency requirement in Eq. (54), Eq. (67) can be reduced to

$$\begin{aligned} \mathbf{B}_0(z \rightarrow 0^+) &= \frac{1}{2} \mathbf{e}_z \times \mathbf{g}_0 \\ &= \begin{bmatrix} -\partial A_{0,y}(z \rightarrow 0^+; \mathbf{0}) / \partial z \\ \partial A_{0,x}(z \rightarrow 0^+; \mathbf{0}) / \partial z \\ 0 \end{bmatrix}, \end{aligned} \quad (68)$$

where the second equality is obtained from Eq. (42). The result in Eq. (68) can, of course, be obtained directly from the equation $\mathbf{B}_0(z) = \nabla \times \mathbf{A}_0(z; \mathbf{0})$ in the limit $z \rightarrow 0^+$.

The translational symmetry parallel to the surface dictates that the nonlinear magnetic field in the vacuum, \mathbf{B}_0^V , can be a function of z , only, i.e., $\mathbf{B}_0^V = \mathbf{B}_0^V(z)$. In turn, since the Maxwell equations imply $\nabla \cdot \mathbf{B}_0^V = 0$ and $\nabla \times \mathbf{B}_0^V = \mathbf{0}$, it follows that $\partial B_{0,x}^V / \partial z = \partial B_{0,y}^V / \partial z = \partial B_{0,z}^V / \partial z = 0$. Hence, \mathbf{B}_0^V is also independent of z . To calculate the constant value of \mathbf{B}_0^V , the boundary conditions for the magnetic field are employed. Since the normal component of \mathbf{B}_0^V is continuous at the surface and since the current density $\mathcal{J}_{\parallel}^{\text{NL}}(z; \mathbf{0})$ is nonsingular at the surface ($z \rightarrow 0^+$), the tangential component of the magnetic field must also be continuous at the boundary. This implies immediately [cf. Eq. (68)] that

$$\mathbf{B}_0^V = \frac{1}{2} \mathbf{e}_z \times \mathbf{g}_0. \quad (69)$$

In terms of the wave-vector spectrum, $\mathbf{J}_{\parallel}^{\text{SL}}(q_{\perp} \mathbf{e}_z)$, of the forced current density, the prevailing nonlinear current density [Eqs. (60) and (63)] and the associated magnetic fields inside [Eq. (66)] and outside [Eq. (69)] the superconductor have now been obtained. It is a remarkable feature that *only the transverse Meissner response kernel* $S_0^{\text{T}}(q_{\perp})$ [via $\mathcal{N}_0^{\text{T}}(q_{\perp})$] *contributes to the nonlinear electromagnetic rectification process.*

E. Fundamental field and forced nonlinear current density

The strength of the forced nonlinear current density depends on the Fourier spectrum of the fundamental field inside the superconductor, cf. Eqs. (43) and (44). Within the framework of the specular-reflection model, the Fourier amplitude of the fundamental field can be determined by the same procedure as used in the normal metallic state.²⁶ Thus, to obtain the result for $\mathbf{E}_1^{\text{eff}}(K_{\perp}, \mathbf{K}_{\parallel}, \omega)$, etc., one need only to replace the normal-state linear conductivity tensor by the corresponding superconducting one in the normal-state expression for the field. Doing this, we find

$$\mathbf{E}_1^{\text{eff}}(\mathbf{K}, \omega) = \vec{\mathbf{G}}_1(\mathbf{K}, \omega) \cdot \mathbf{g}_1, \quad (70)$$

where the Fourier-space propagator $\vec{\mathbf{G}}_1(\mathbf{K}, \omega)$ is given by

$$\vec{\mathbf{G}}_1(\mathbf{K}, \omega) = \frac{\vec{\mathbb{1}} - \mathbf{e}_{\mathbf{K}} \otimes \mathbf{e}_{\mathbf{K}}}{\mathcal{N}_1^{\text{T}}(K, \omega)} + \frac{\mathbf{e}_{\mathbf{K}} \otimes \mathbf{e}_{\mathbf{K}}}{\mathcal{N}_1^{\text{L}}(K, \omega)}. \quad (71)$$

The denominators

$$\mathcal{N}_1^{\text{T}}(K, \omega) = \left[\frac{\omega}{c_0} \right]^2 \left[1 + \frac{i\sigma^{\text{T}}(K, \omega)}{\epsilon_0 \omega} \right] - K^2, \quad (72)$$

and

$$\mathcal{N}_1^{\text{L}}(K, \omega) = \left[\frac{\omega}{c_0} \right]^2 \left[1 + \frac{i\sigma^{\text{L}}(K, \omega)}{\epsilon_0 \omega} \right], \quad (73)$$

contain the transverse [$\sigma^{\text{T}}(K, \omega)$] and longitudinal [$\sigma^{\text{L}}(K, \omega)$] parts of the linear conductivity tensor [$\vec{\sigma}(\mathbf{K}, \omega)$] of the superconductor at the fundamental frequency. On the basis of the general expression²⁴

$$\begin{aligned} \vec{\sigma}(\mathbf{K}, \omega) &= \frac{ine^2}{m\omega} \vec{\mathbb{1}} - \frac{i}{\omega} \left[\frac{e\hbar}{2m} \right]^2 \frac{1}{V} \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{K}) \otimes (2\mathbf{k} + \mathbf{K}) \\ &\quad \times \left[(u_{\mathbf{k}} u_{\mathbf{k}+\mathbf{K}} + v_{\mathbf{k}} v_{\mathbf{k}+\mathbf{K}})^2 (f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{K}}) \left(\frac{1}{E_{\mathbf{k}+\mathbf{K}} - E_{\mathbf{k}} + \hbar\omega} \right. \right. \\ &\quad \left. \left. + \frac{1}{E_{\mathbf{k}+\mathbf{K}} - E_{\mathbf{k}} - \hbar\omega} \right) \right. \\ &\quad \left. + (u_{\mathbf{k}+\mathbf{K}} v_{\mathbf{k}} - v_{\mathbf{k}+\mathbf{K}} u_{\mathbf{k}})^2 (1 - f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{K}}) \left(\frac{1}{E_{\mathbf{k}+\mathbf{K}} + E_{\mathbf{k}} - \hbar\omega} \right. \right. \\ &\quad \left. \left. + \frac{1}{E_{\mathbf{k}+\mathbf{K}} + E_{\mathbf{k}} + \hbar\omega} \right) \right], \end{aligned} \quad (74)$$

the transverse and longitudinal response tensors are obtained from the relation

$$\vec{\sigma}(\mathbf{K}, \omega) = (\vec{\mathbb{1}} - \mathbf{e}_{\mathbf{K}} \otimes \mathbf{e}_{\mathbf{K}}) \sigma^T(K, \omega) + \mathbf{e}_{\mathbf{K}} \otimes \mathbf{e}_{\mathbf{K}} \sigma^L(K, \omega). \quad (75)$$

In Eqs. (71) and (75), $\mathbf{e}_{\mathbf{K}}$ denotes a unit vector parallel to the \mathbf{K} direction. The quantity $\mathbf{g}_1 = (g_{1,x}, g_{1,y}, 0)$ in Eq. (70) is determined by the boundary conditions for the fundamental electromagnetic field at the surface, of which an explicit expression is given below.

With a knowledge of $\mathbf{E}_1^{\text{eff}}$, the forced nonlinear dc-current density can be obtained from Eqs. (43) and (44).

Since $\mathbf{R}_0(\mathbf{K}, \omega) = R_0(K, \omega) \mathbf{e}_{\mathbf{K}}$, one has choosing the plane of incidence to coincide with the x - z plane

$$\mathbf{R}_0(\mathbf{K}, \omega) \cdot \mathbf{E}_1^{\text{eff}}(\mathbf{K}, \omega) = \frac{R_0(K, \omega)}{\mathcal{N}_1^T(K, \omega)} \frac{K_{\parallel}}{K} g_{1,x}, \quad (76)$$

as can be seen with use of Eqs. (70) and (71). Hence, it is realized that in order to obtain electromagnetic rectification an irrotational (L) part is needed in the fundamental field inside the Cooper-paired jellium. Since only $\mathbf{J}_{\parallel}^{\text{SL}}(q_{\perp} \mathbf{e}_z)$ is needed to obtain the dc-magnetic field and the associated dc-current density we make use of the equation

$$\begin{aligned} (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot [\mathbf{E}_1^{\text{eff}}(K_{\perp} - q_{\perp}, \mathbf{K}_{\parallel}, \omega)]^* &= \left[\frac{(K_{\perp} - q_{\perp})^2}{\mathcal{N}_1^T([K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2]^{1/2}, \omega)} + \frac{K_{\parallel}^2}{\mathcal{N}_1^L([K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2]^{1/2}, \omega)} \right] \frac{g_{1,x}^* \mathbf{e}_x}{K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2} \\ &+ \frac{g_{1,y}^* \mathbf{e}_y}{\mathcal{N}_1^T([K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2]^{1/2}, \omega)} \end{aligned} \quad (77)$$

to get

$$\begin{aligned} (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{(+)}(q_{\perp} \mathbf{e}_z) &= (2\pi)^{-3} \int_{-\infty}^{\infty} \left[\frac{(K_{\perp} - q_{\perp})^2}{\{\mathcal{N}_1^T([K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2]^{1/2}, \omega)\}^*} + \frac{K_{\parallel}^2}{\{\mathcal{N}_1^L([K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2]^{1/2}, \omega)\}^*} \right] \\ &\times \frac{|g_{1,x}|^2 \mathbf{e}_x}{K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2} + \frac{g_{1,x} g_{1,y}^* \mathbf{e}_y}{\{\mathcal{N}_1^T([K_{\parallel}^2 + (K_{\perp} - q_{\perp})^2]^{1/2}, \omega)\}^*} \left] \frac{K_{\parallel}}{K} \frac{R_0(K, \omega)}{\mathcal{N}_1^L(K, \omega)} dK_{\perp}, \end{aligned} \quad (78)$$

where \mathbf{e}_x and \mathbf{e}_y are unit vectors along the x and y directions. Our final result for the Fourier amplitude of the forced nonlinear current density flowing along the surface is obtained by inserting the expression in Eq. (78), plus an analogous one obtained by the replacement $q_{\perp} \rightarrow -q_{\perp}$ and a complex conjugation, into

$$\mathbf{J}_{\parallel}^{\text{SL}}(q_{\perp} \mathbf{e}_z) = \frac{1}{2} (\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \{ \mathbf{J}_0^{(+)}(q_{\perp} \mathbf{e}_z) + [\mathbf{J}_0^{(+)}(-q_{\perp} \mathbf{e}_z)]^* \}. \quad (79)$$

F. Polarization selection rules

The possibilities for achieving nonlinear electromagnetic rectification depend on the state of polarization and on the angle of incidence of the fundamental field. To investigate the polarization selection rules, we use the continuity of the tangential components of the fundamental electric and magnetic fields at the surface to express the quantity \mathbf{g}_1 in terms of the amplitude $\mathbf{E}_1^i(\mathbf{K}_i, \omega)$ of the incident (i) fundamental field,

$$\mathbf{E}_1^i(\mathbf{r}, t) = \frac{1}{2} [(2\pi)^{-3} \mathbf{E}_1^i(\mathbf{K}_i, \omega) e^{i(\mathbf{K}_i \cdot \mathbf{r} - \omega t)} + \text{c. c.}] \quad (80)$$

outside the surface.²⁰ In Eq. (80), the incident wave vector is $\mathbf{K}_i = \mathbf{K}_{\parallel} + K_{\perp}^i \mathbf{e}_z$, where $\mathbf{K}_{\perp}^i = [(\omega/c_0)^2 - K_{\parallel}^2]^{1/2}$. As the outcome of this calculation one obtains

$$g_{1,x} = \frac{4\pi E_1^{i,p}(\mathbf{K}_i, \omega)}{\frac{\omega}{c_0} \int_{-\infty}^{\infty} \left[\frac{K_{\perp} \left[K_{\perp} + \left(\frac{c_0 K}{\omega} \right)^2 K_{\perp}^i \right]}{\mathcal{N}_1^T(K, \omega)} + \frac{K_{\parallel}^2}{\mathcal{N}_1^L(K, \omega)} \right] \frac{e^{iK_{\perp}^i 0^+}}{K_{\perp}^i K^2} dK_{\perp}}, \quad (81)$$

and

$$g_{1,y} = \frac{4\pi E_1^{i,s}(\mathbf{K}_i, \omega)}{\int_{-\infty}^{\infty} \left[1 + \frac{K_{\perp}}{K_{\perp}^i} \right] \frac{e^{iK_{\perp}^i 0^+}}{\mathcal{N}_1^T(K, \omega)} dK_{\perp}}, \quad (82)$$

where $E_1^{i,p}$ and $E_1^{i,s}$ are, respectively, the p - and s -polarized components of the incident field amplitude.

It appears from Eq. (78) that in order to obtain a forced nonlinear current density, $g_{1,x}$ must be nonzero. This means according to Eqs. (81) and (82) that an s -polarized incident field cannot give rise to electromagnet-

ic rectification. If the incident field is p polarized, $g_{1,y}=0$, so that the forced current density is parallel to the plane of incidence [cf. Eq. (78)]. In turn, it follows from Eqs. (63) and (66) that also the self-consistent current density is parallel to the plane of incidence and the magnetic field is perpendicular to this plane. If the incident light has both a p and an s component, $\mathbf{J}_{\parallel}(z; \mathbf{0})$ is not confined to the scattering plane, nor is $\mathbf{B}_0(z)$ perpendicular to this plane. The magnetic field, however, is always parallel to the surface plane.

V. COLLISIONLESS NORMAL STATE

In the last decade, the optical second-harmonic generation in centrosymmetric metals has been extensively investigated.⁹ The associated dc reaction on the conduction electrons of the metals has not been studied until now. To observe this optical rectification process in the normal metallic state, one would like to increase the electronic relaxation time significantly relative to that of conventional second-harmonic-generation experiments. Usu-

ally, impurity scattering and electron-phonon interactions are the main sources for the conduction-electron damping. Thus, it is suggested that nonlinear optical experiments be performed on especially pure samples at low temperatures.

The theory presented in the preceding sections applies to the collisionless normal jellium state when setting $T > T_c$. The analytical expressions for the response tensors $\mathbf{R}_0(\mathbf{K}, \omega)$, $\vec{\mathbf{S}}_0(q_1 \mathbf{e}_z)$, and $\vec{\sigma}(\mathbf{K}, \omega)$ are much simpler in the normal state than in the superconducting state. The transition to the normal state (NS) is obtained following the standard procedure. Thus, one obtains, for the nonlinear response function in Eq. (18), the formula¹⁵

$$\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega) = -\frac{\hbar e^3}{2m^2 \omega^2 V} \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{Q}) \frac{f_{\mathbf{k}}^{\text{NS}} - f_{\mathbf{k}+\mathbf{Q}}^{\text{NS}}}{\hbar\omega + \epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}+\mathbf{Q}}}. \quad (83)$$

The expression for the linear conductivity tensor is reduced to the well-known form²¹

$$\vec{\sigma}^{\text{NS}}(\mathbf{Q}, \omega) = \frac{ine^2}{m\omega} \vec{\mathbb{1}} + \frac{2i}{\omega} \left[\frac{e\hbar}{2m} \right]^2 \frac{1}{V} \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{Q}) \otimes (2\mathbf{k} + \mathbf{Q}) \frac{f_{\mathbf{k}}^{\text{NS}} - f_{\mathbf{k}+\mathbf{Q}}^{\text{NS}}}{\hbar\omega + \epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}+\mathbf{Q}}} \quad (84)$$

in the normal state. The expression for $\vec{\mathbf{S}}_0(\mathbf{Q})$ in the normal phase can be obtained easily from Eq. (84) utilizing the relation

$$\vec{\mathbf{S}}_0^{\text{NS}}(\mathbf{Q}) = \lim_{\omega \rightarrow 0} [i\omega \vec{\sigma}^{\text{NS}}(\mathbf{Q}, \omega)]. \quad (85)$$

Thus,

$$\vec{\mathbf{S}}_0^{\text{NS}}(\mathbf{Q}) = -\frac{ne^2}{m} \vec{\mathbb{1}} - \left[\frac{e\hbar}{2m} \right]^2 \frac{2}{V} \sum_{\mathbf{k}} (2\mathbf{k} + \mathbf{Q}) \otimes (2\mathbf{k} + \mathbf{Q}) \frac{f_{\mathbf{k}}^{\text{NS}} - f_{\mathbf{k}+\mathbf{Q}}^{\text{NS}}}{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}+\mathbf{Q}}}. \quad (86)$$

It is important from a quantitative point of view to be in the possession of closed-formed expressions for the response tensors in Eqs. (83), (84), and (86). By replacing the Fermi-Dirac distribution function by a step function ($T=0$ approximation), i.e., $f_{\mathbf{k}}^{\text{NS}} = \Theta(k_F - |\mathbf{k}|)$, where Θ is the Heaviside unit-step function, it is a straightforward, but tedious matter to carry out the summations over \mathbf{k} space (see the Appendix). By introducing the ‘‘classical’’ (u) and ‘‘quantum-mechanical’’ (z) nonlocality parameters $u = \omega/(Qv_F)$ and $z = Q/(2k_F)$, respectively, v_F being the Fermi velocity of the electron, one obtains the following expression for the nonlinear response function:

$$\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega) = \frac{e^3 k_F^2}{4\pi^2 \hbar m \omega^2} u \left[1 + \frac{1}{4z} \left[[1 - (u+z)^2] \ln \left| \frac{u+z+1}{u+z-1} \right| + [1 - (u-z)^2] \ln \left| \frac{u-z-1}{u-z+1} \right| \right] \right] \mathbf{e}_{\mathbf{Q}}, \quad (87)$$

where $\mathbf{e}_{\mathbf{Q}} = \mathbf{Q}/Q$. If the magnitudes of the relevant electromagnetic wave vectors are small in comparison to the Fermi wave vector ($z \ll 1$), Eq. (87) is reduced to the form

$$\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega; z \rightarrow 0) = \frac{e^3 k_F^2}{4\pi^2 \hbar m \omega^2} u \left[2 + u \ln \left| \frac{u-1}{u+1} \right| \right] \mathbf{e}_{\mathbf{Q}}. \quad (88)$$

To lowest order in u^{-1} , one obtains, from Eq. (88), the previously established¹⁵ hydrodynamic result

$$\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega; z \rightarrow 0)|_{\text{hydro}} = -\frac{ne^3}{2m^2 \omega^3} \mathbf{Q}. \quad (89)$$

The analytical expressions for the transverse ($\sigma_{\perp}^{\text{NS}}$) and longitudinal ($\sigma_{\parallel}^{\text{NS}}$) parts of the linear conductivity tensor are well known.^{27,20,21,28} Thus,

$$\sigma_{\text{T}}^{\text{NS}}(\mathbf{Q}, \omega) = \frac{3ine^2}{8m\omega} \left[z^2 + 3u^2 + 1 - \frac{1}{4z} \left[[1 - (z-u)^2] \ln \left| \frac{z-u+1}{z-u-1} \right| + [1 - (z+u)^2] \ln \left| \frac{z+u+1}{z+u-1} \right| \right] \right], \quad (90)$$

and

$$\sigma_{\text{L}}^{\text{NS}}(\mathbf{Q}, \omega) = \frac{3ne^2u^2}{2im\omega} \left[1 + \frac{1}{4z} \left[[1 - (z-u)^2] \ln \left| \frac{z-u+1}{z-u-1} \right| + [1 - (z+u)^2] \ln \left| \frac{z+u+1}{z+u-1} \right| \right] \right]. \quad (91)$$

Utilizing Eq. (85), it follows that the transverse ($S_{0,\text{T}}^{\text{NS}}$) and longitudinal ($S_{0,\text{L}}^{\text{NS}}$) parts of the Meissner kernel are

$$S_{0,\text{T}}^{\text{NS}}(Q) = -\frac{3ne^2}{8m} \left[z^2 + 1 - \frac{(1-z^2)^2}{2z} \ln \left| \frac{z+1}{z-1} \right| \right], \quad (92)$$

and

$$S_{0,\text{L}}^{\text{NS}}(Q) = 0, \quad (93)$$

respectively. One should emphasize, in relation to Eq. (93), that only the gauge-invariant,²⁴ transverse part of the Meissner kernel contributes to the dc-current density and the associated magnetic field [cf. Eqs. (46), (63), and (66)]. In the small-wave-vector limit ($Q \ll 2k_F$), one obtains, to lowest order in z , the hydrodynamic result

$$S_{0,\text{T}}^{\text{NS}}(Q)|_{\text{hydro}} = -\frac{ne^2}{m} \left[\frac{Q}{2k_F} \right]^2 \quad (94)$$

for the transverse part of the free dc response. The classical Boltzmann-equation expression and the hydrodynamic result for $\sigma_{\text{T}}^{\text{NS}}(Q, \omega)$ and $\sigma_{\text{L}}^{\text{NS}}(Q, \omega)$ are well known and need not be reproduced here. By comparison of Eqs. (87) and (91), it is realized that R_0^{NS} and $\sigma_{\text{L}}^{\text{NS}}$ are related via the simple equation

$$R_0^{\text{NS}}(Q, \omega) = \frac{ieQ}{2m\omega^2} \sigma_{\text{L}}^{\text{NS}}(Q, \omega). \quad (95)$$

Below T_c , the relation between these two quantities is more complicated mainly because of the different ways in which the coherence factors enter the expression for $R_0(Q, \omega)$ and $\sigma_{\text{L}}(Q, \omega)$ [cf. Eqs. (18), (19), and (74)].

VI. ELECTROMAGNETIC RECTIFICATION BY POLARITON-PLASMON AND PLASMON-PLASMON INTERACTIONS

A. Pole structure of the forced nonlinear current density

For frequencies in the vicinity of and above the plasma edge, the fundamental field inside the BCS-paired jellium is dominated by that associated with the excitation and propagation of polaritons and plasmons. In implicit form, the inverse dispersion relations for polaritons [$\kappa^{\text{T}} = \kappa^{\text{T}}(\omega)$] and plasmons [$\kappa^{\text{L}} = \kappa^{\text{L}}(\omega)$] are determined by the conditions $\mathcal{N}_1^{\text{T}}(\kappa^{\text{T}}, \omega) = 0$ and $\mathcal{N}_1^{\text{L}}(\kappa^{\text{L}}, \omega) = 0$, respectively. In the actual case, where the wave-vector component along the surface (i.e., K_{\parallel}) is fixed, the wave-vector components perpendicular to the surface of the polariton ($\kappa_{\perp}^{\text{T}}$) and plasmon ($\kappa_{\perp}^{\text{L}}$) are given implicitly by

$$\mathcal{N}_1^{\text{T}}(K_{\parallel}, \kappa_{\perp}^{\text{T}}, \omega) = 0 \quad (96)$$

and

$$\mathcal{N}_1^{\text{L}}(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega) = 0. \quad (97)$$

To investigate the nonlinear electromagnetic rectification process associated with the excitation of the above-mentioned collective modes, we consider the general expression for the forced nonlinear current density [Eqs. (78) and (79)]. In the complex K_{\perp} plane, the integrand of Eq. (78) has poles at

$$K_{\perp} = \pm \kappa_{\perp}^{\text{L}} = \pm [(\kappa^{\text{L}})^2 - K_{\parallel}^2]^{1/2}. \quad (98)$$

To ensure that these poles, and those given below, do not lie on the real K_{\perp} axis, an infinitesimal small but positive imaginary number is added to the frequency, i.e., $\omega \Rightarrow \omega + i0^+$. Hence, choosing $\text{Im}\kappa_{\perp}^{\text{L}} > 0$, the pole at $K_{\perp} = \kappa_{\perp}^{\text{L}}$ lies in the upper half-plane. Since \mathcal{N}_1^{T} and \mathcal{N}_1^{L} are even functions of K_{\perp} , the poles in Eqs. (96) and (97) are always symmetrically placed with respect to the origin. To determine the remaining poles of the integrand in Eq. (78), we note that the solutions to $[\mathcal{N}_1^{\text{T}}(K, \omega)]^* = 0$ and $[\mathcal{N}_1^{\text{L}}(K, \omega)]^* = 0$ are $K = \pm(\kappa^{\text{T}})^*$ and $K = \pm(\kappa^{\text{L}})^*$, respectively. This implies that the rest of the poles are located at

$$K_{\perp} = q_{\perp} \pm (\kappa_{\perp}^{\text{T}})^* = q_{\perp} \pm \{[(\kappa^{\text{T}})^*]^2 - K_{\parallel}^2\}^{1/2} \quad (99)$$

and

$$K_{\perp} = q_{\perp} \pm (\kappa_{\perp}^{\text{L}})^* = q_{\perp} \pm \{[(\kappa^{\text{L}})^*]^2 - K_{\parallel}^2\}^{1/2}. \quad (100)$$

In choosing $(\kappa_{\perp}^{\text{T}})^* = +\{[(\kappa^{\text{T}})^*]^2 - K_{\parallel}^2\}^{1/2}$ and $(\kappa_{\perp}^{\text{L}})^* = +\{[(\kappa^{\text{L}})^*]^2 - K_{\parallel}^2\}^{1/2}$, the principal interval for the arguments is $(-\pi, \pi)$. With the additional choice $\text{Im}\kappa_{\perp}^{\text{T}} > 0$ (for $\kappa_{\perp}^{\text{T}} = [(\kappa^{\text{T}})^2 - K_{\parallel}^2]^{1/2}$), the poles

$$-p_{\text{T}}^*(q_{\perp}) \equiv q_{\perp} - (\kappa_{\perp}^{\text{T}})^* \quad (101)$$

and

$$-p_{\text{L}}^*(q_{\perp}) \equiv q_{\perp} - (\kappa_{\perp}^{\text{L}})^* \quad (102)$$

are located in the upper half-plane of the complex K_{\perp} plane. Introducing the quantities

$$\mathcal{R}_I(K_{\parallel}, \kappa_{\perp}^I, \omega) \equiv \lim_{K_{\perp} \rightarrow \kappa_{\perp}^I} \left[\frac{K_{\perp} - \kappa_{\perp}^I}{\mathcal{N}_1^I(K_{\parallel}, K_{\perp}, \omega)} \right], \quad I = \text{T or L} \quad (103)$$

it is possible to show that the residues associated with the (assumed) first-order poles $\kappa_{\perp}^{\text{L}}$, $-p_{\text{T}}^*$, and $-p_{\text{L}}^*$ are given by \mathcal{R}_{L} , $-\mathcal{R}_{\text{T}}^*$, and $-\mathcal{R}_{\text{L}}^*$, respectively. By (i) neglecting possible branch-cut contributions, and (ii) noticing that

$$\{(\kappa_{\perp} - q_{\perp})^2 / [\mathcal{N}_{\perp}^T(K_{\parallel}, K_{\perp} - q_{\perp}, \omega)]^* + K_{\parallel}^2 / [\mathcal{N}_{\perp}^L(K_{\parallel}, K_{\perp} - q_{\perp}, \omega)]^*\} / [K_{\parallel}^2 + (\kappa_{\perp} - q_{\perp})^2] \rightarrow 0$$

for $K_{\perp} \rightarrow q_{\perp} \pm iK_{\parallel}$ so that the integrand in Eq. (78) has no poles at $K_{\perp} = q_{\perp} \pm iK_{\parallel}$, residue calculation in the upper half-plane gives

$$\begin{aligned} \frac{1}{2}(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{(+)}(q_{\perp} \mathbf{e}_z) = & \frac{i}{4\pi^2} \left\{ \left[\left[\frac{(\kappa_{\perp}^L - q_{\perp})^2}{[\mathcal{N}_{\perp}^T(K_{\parallel}, \kappa_{\perp}^L - q_{\perp}, \omega)]^*} + \frac{K_{\parallel}^2}{[\mathcal{N}_{\perp}^L(K_{\parallel}, \kappa_{\perp}^L - q_{\perp}, \omega)]^*} \right] \frac{|g_{1,x}|^2 \mathbf{e}_x}{K_{\parallel}^2 + (\kappa_{\perp}^L - q_{\perp})^2} \right. \right. \\ & + \left. \frac{g_{1,x} g_{1,y}^* \mathbf{e}_y}{[\mathcal{N}_{\perp}^T(K_{\parallel}, \kappa_{\perp}^L - q_{\perp}, \omega)]^*} \right] \frac{K_{\parallel}}{\kappa_L} R_0(K_{\parallel}, \kappa_{\perp}^L, \omega) \mathcal{R}_L(K_{\parallel}, \kappa_{\perp}^L, \omega) \\ & - \left[\left[\frac{(\kappa_{\perp}^T)^*}{\kappa_{\perp}^*} \right]^2 |g_{1,x}|^2 \mathbf{e}_x + g_{1,x} g_{1,y}^* \mathbf{e}_y \right] \frac{K_{\parallel} R_0(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^T)^*, \omega) \mathcal{R}_T^*(K_{\parallel}, \kappa_{\perp}^T, \omega)}{\{K_{\parallel}^2 + [q_{\perp} - (\kappa_{\perp}^T)^*]^2\}^{1/2} \mathcal{N}_{\perp}^L(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^T)^*, \omega)} \\ & \left. - \frac{K_{\parallel}^3 |g_{1,x}|^2 \mathbf{e}_x R_0(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^L)^*, \omega) \mathcal{R}_L^*(K_{\parallel}, \kappa_{\perp}^L, \omega)}{(\kappa_{\perp}^*)^2 \{K_{\parallel}^2 + [q_{\perp} - (\kappa_{\perp}^L)^*]^2\}^{1/2} \mathcal{N}_{\perp}^L(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^L)^*, \omega)} \right\}. \end{aligned} \quad (104)$$

The poles associated with the integrand occurring in the expression for $(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{(+)}(-q_{\perp} \mathbf{e}_z)^*$ are determined from the condition $[\mathcal{N}_{\perp}^T(K, \omega)]^* = 0$, $\mathcal{N}_{\perp}^T(K_{\parallel}, K_{\perp} + q_{\perp}, \omega) = 0$, and $\mathcal{N}_{\perp}^L(K_{\parallel}, K_{\perp} + q_{\perp}, \omega) = 0$. This means that the corresponding poles are located at the following positions:

$$K_{\perp} = \pm (\kappa_{\perp}^L)^*, \quad (105)$$

$$K_{\perp} = -q_{\perp} \pm \kappa_{\perp}^T, \quad (106)$$

$$K_{\perp} = -q_{\perp} \pm \kappa_{\perp}^L. \quad (107)$$

The three poles in the upper half-plane are located at

$-(\kappa_{\perp}^L)^*$, $p_T = -q_{\perp} + \kappa_{\perp}^T$, and $p_L = -q_{\perp} + \kappa_{\perp}^L$, and the corresponding residues are $-\mathcal{R}_L^*$, \mathcal{R}_T , and \mathcal{R}_L , respectively. The distribution of all the upper half-plane poles in the integrand of $\mathbf{J}_0^{SL}(q_{\perp} \mathbf{e}_z)$ is tabulated in Table I. We notice from this table that the poles associated with $(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{(+)}(q_{\perp} \mathbf{e}_z)$ can be obtained from those appearing in $(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{(+)}(-q_{\perp} \mathbf{e}_z)^*$ by a mirroring in the imaginary axis. For completeness, the values of the residues connected to the various poles are also indicated in Table I. On the basis of the pole structure obtained above, we obtain, by contour integration in the upper half-plane, the result

$$\begin{aligned} \frac{1}{2}(\vec{\mathbb{1}} - \mathbf{e}_z \otimes \mathbf{e}_z) \cdot \mathbf{J}_0^{(+)}(-q_{\perp} \mathbf{e}_z)^* = & \frac{i}{4\pi^2} \left\{ - \left[\left[\frac{[q_{\perp} - (\kappa_{\perp}^L)^*]^2}{\mathcal{N}_{\perp}^T(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^L)^*, \omega)} + \frac{K_{\parallel}^2}{\mathcal{N}_{\perp}^L(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^L)^*, \omega)} \right] \frac{|g_{1,x}|^2 \mathbf{e}_x}{K_{\parallel}^2 + [q_{\perp} - (\kappa_{\perp}^L)^*]^2} \right. \right. \\ & + \left. \frac{g_{1,x}^* g_{1,y} \mathbf{e}_y}{\mathcal{N}_{\perp}^T(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^L)^*, \omega)} \right] \frac{K_{\parallel}}{\kappa_L^*} R_0^*(K_{\parallel}, -(\kappa_{\perp}^L)^*, \omega) \mathcal{R}_L^*(K_{\parallel}, \kappa_{\perp}^L, \omega) \\ & + \left[\left[\frac{\kappa_{\perp}^T}{\kappa_{\perp}} \right]^2 |g_{1,x}|^2 \mathbf{e}_x + g_{1,x}^* g_{1,y} \mathbf{e}_y \right] \frac{K_{\parallel} R_0^*(K_{\parallel}, \kappa_{\perp}^T - q_{\perp}, \omega) \mathcal{R}_T(K_{\parallel}, \kappa_{\perp}^T, \omega)}{[K_{\parallel}^2 + (\kappa_{\perp}^T - q_{\perp})^2]^{1/2} [\mathcal{N}_{\perp}^L(K_{\parallel}, \kappa_{\perp}^T - q_{\perp}, \omega)]^*} \\ & \left. + \frac{K_{\parallel}^3 |g_{1,x}|^2 \mathbf{e}_x R_0^*(K_{\parallel}, \kappa_{\perp}^L - q_{\perp}, \omega) \mathcal{R}_L(K_{\parallel}, \kappa_{\perp}^L, \omega)}{\kappa_{\perp}^2 [K_{\parallel}^2 + (\kappa_{\perp}^L - q_{\perp})^2]^{1/2} [\mathcal{N}_{\perp}^L(K_{\parallel}, \kappa_{\perp}^L - q_{\perp}, \omega)]^*} \right\}. \end{aligned} \quad (108)$$

In order to determine in the collective mode approximation, the forced contribution to the current density $\mathbf{J}_0^{\parallel}(z; \mathbf{0})$, and the magnetic field $\mathbf{B}_0(z)$, one has to calculate the pole contributions to the integrals

$$\Gamma_{(n)} \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{q_{\perp}^n \mathbf{J}_0^{SL}(q_{\perp} \mathbf{e}_z)}{\mathcal{N}_0^T(q_{\perp})} e^{iq_{\perp} z} dq_{\perp}, \quad n = 1 \text{ and } 2 \quad (109)$$

[cf. Eqs. (63) and (67); magnetic field, $n=1$; current density, $n=2$]. By inserting Eq. (104) into (109), it is realized that poles are located in the complex q_{\perp} plane at the positions

TABLE I. Pole structure of the forced nonlinear current density $\mathbf{J}_0^{SL}(q_{\perp} \mathbf{e}_z)$ in the complex K_{\perp} plane. The pole locations in the upper half-plane and the associated residues are tabulated. Notice that the poles are placed symmetrically with respect to the imaginary K_{\perp} axis.

Pole locations	Associated residues
κ_{\perp}^L	\mathcal{R}_L
$p_T = \kappa_{\perp}^T - q_{\perp}$	\mathcal{R}_T
$p_L = \kappa_{\perp}^L - q_{\perp}$	\mathcal{R}_L
$-(\kappa_{\perp}^L)^*$	$-\mathcal{R}_L^*$
$-p_T^* = q_{\perp} - (\kappa_{\perp}^T)^*$	$-\mathcal{R}_T^*$
$-p_L^* = q_{\perp} - (\kappa_{\perp}^L)^*$	$-\mathcal{R}_L^*$

$$q_{\perp} = \kappa_{\perp}^L \mp (\kappa_{\perp}^T)^* , \quad (110)$$

$$q_{\perp} = \kappa_{\perp}^L \mp (\kappa_{\perp}^L)^* , \quad (111)$$

$$q_{\perp} = (\kappa_{\perp}^T)^* \pm \kappa_{\perp}^L , \quad (112)$$

$$q_{\perp} = (\kappa_{\perp}^L)^* \pm \kappa_{\perp}^L . \quad (113)$$

The poles located in the upper half-plane are $q_{\perp} = \kappa_{\perp}^L - (\kappa_{\perp}^T)^*$, $q_{\perp} = \kappa_{\perp}^L - (\kappa_{\perp}^L)^*$, and, provided that $\text{Im}\kappa_{\perp}^L > \text{Im}\kappa_{\perp}^T$, $q_{\perp} = (\kappa_{\perp}^T)^* + \kappa_{\perp}^L$. For the first two poles, the associated residues are $-\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^T, \omega)$, and $-\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega)$, respectively. The pole at $q_{\perp} = (\kappa_{\perp}^T)^* + \kappa_{\perp}^L$ is twofold degenerated and the two related residues are (in case) $\mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^L, \omega)$ and $\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^T, \omega)$. One pole is located at the real axis, namely, at $q_{\perp} = 2 \text{Re}\kappa_{\perp}^L$. This pole also exhibits twofold degeneracy with residues $\frac{1}{2}\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega)$ and $\frac{1}{2}\mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^L, \omega)$. The remaining poles in Eq. (109) are obtained via the contribution from Eq. (108) to the integrand. Thus, by direct inspection, one locates poles at

$$q_{\perp} = (\kappa_{\perp}^L)^* \pm \kappa_{\perp}^T , \quad (114)$$

$$q_{\perp} = (\kappa_{\perp}^L)^* \pm \kappa_{\perp}^L , \quad (115)$$

$$q_{\perp} = (\kappa_{\perp}^T) \mp (\kappa_{\perp}^L)^* , \quad (116)$$

and at the positions given in Eq. (111). The poles in the upper half-plane or on the real axis stemming from Eq. (108) are at the following positions: $q_{\perp} = (\kappa_{\perp}^L)^* + \kappa_{\perp}^T$ (provided that $\text{Im}\kappa_{\perp}^T > \text{Im}\kappa_{\perp}^L$); [twofold-degenerate residues: $\mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^T, \omega)$ and $\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega)$], $q_{\perp} = (\kappa_{\perp}^L)^* + \kappa_{\perp}^L$ [on the

TABLE II. Pole structure of $\Gamma_{(n)}$ in the complex q_{\perp} plane. The pole locations in the upper half-plane and on the real axis and the associated residues are tabulated together with the degeneracy of each pole position. Only the poles in the three upper rows of the table give a net [polariton-plasmon (TL) and plasmon-plasmon (LL)] contribution to $\Gamma_{(n)}$.

Pole locations	Degeneracies	Associated residues
$\kappa_{\perp}^L - (\kappa_{\perp}^T)^*$	1	$-\mathcal{R}_{\perp}^*$
$\kappa_{\perp}^L - (\kappa_{\perp}^L)^*$	1	$-\mathcal{R}_{\perp}^*$
$2i \text{Im}\kappa_{\perp}^L$	1+1	$-\mathcal{R}_{\perp}^*$ and $-\mathcal{R}_{\perp}^*$ $\frac{1}{2}\mathcal{R}_{\perp}, \frac{1}{2}\mathcal{R}_{\perp}^*$ and
$2 \text{Re}\kappa_{\perp}^L$	2+2	$\frac{1}{2}\mathcal{R}_{\perp}, \frac{1}{2}\mathcal{R}_{\perp}^*$ $\mathcal{R}_{\perp}, \mathcal{R}_{\perp}^*$
$\kappa_{\perp}^L + (\kappa_{\perp}^T)^*$ if $\text{Im}(\kappa_{\perp}^L - \kappa_{\perp}^T) > 0$	2	$\mathcal{R}_{\perp}, \mathcal{R}_{\perp}^*$
$(\kappa_{\perp}^L)^* + \kappa_{\perp}^T$ if $\text{Im}(\kappa_{\perp}^T - \kappa_{\perp}^L) > 0$	2	$\mathcal{R}_{\perp}, \mathcal{R}_{\perp}^*$

real axis, and with twofold-degenerate residues: $\frac{1}{2}\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega)$, $\frac{1}{2}\mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^L, \omega)$, $q_{\perp} = \kappa_{\perp}^T - (\kappa_{\perp}^L)^*$ [residue, $-\mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega)$]. A tabulation of the final pole structure and the associated residues is presented in Table II.

Now, a straightforward, but tedious contour integration in the upper half of the complex q_{\perp} plane allows us to determine $\Gamma_{(n)}$. It turns out from such a calculation that the poles on the real axis ($q_{\perp} = 2 \text{Re}\kappa_{\perp}^L$) and the poles $q_{\perp} = \kappa_{\perp}^L + (\kappa_{\perp}^T)^*$ and $q_{\perp} = \kappa_{\perp}^T + (\kappa_{\perp}^L)^*$ give no net contribution to $\Gamma_{(n)}$. In explicit form, one thus obtains the following result:

$$\Gamma_{(n)} = \Gamma_{(n)}^{(+)} + \Gamma_{(n)}^{(-)} , \quad (117)$$

where

$$\begin{aligned} \Gamma_{(n)}^{(+)} = & \frac{1}{4\pi^2} \frac{K_{\parallel}}{\kappa_{\perp}^L} \mathcal{R}_0(K_{\parallel}, \kappa_{\perp}^L, \omega) \mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^L, \omega) \left\{ \left[\left[\frac{(\kappa_{\perp}^T)^*}{\kappa_{\perp}^T} \right]^2 |g_{1,x}|^2 \mathbf{e}_x + g_{1,x} g_{1,y}^* \mathbf{e}_y \right] \mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^T, \omega) \right. \\ & \times \frac{[\kappa_{\perp}^L - (\kappa_{\perp}^T)^*]^n}{\mathcal{N}_0^T(\kappa_{\perp}^L - (\kappa_{\perp}^T)^*)} \exp\{i[\kappa_{\perp}^L - (\kappa_{\perp}^T)^*]z\} \\ & \left. + \left[\frac{K_{\parallel}}{\kappa_{\perp}^L} \right]^2 |g_{1,x}|^2 \mathbf{e}_x \mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega) \frac{(2i \text{Im}\kappa_{\perp}^L)^n}{\mathcal{N}_0^T(2i \text{Im}\kappa_{\perp}^L)} \exp[-2(\text{Im}\kappa_{\perp}^L)z] \right\} , \quad (118) \end{aligned}$$

and

$$\begin{aligned} \Gamma_{(n)}^{(-)} = & \frac{1}{4\pi^2} \frac{K_{\parallel}}{\kappa_{\perp}^L} \mathcal{R}_0^*(K_{\parallel}, -(\kappa_{\perp}^L)^*, \omega) \mathcal{R}_{\perp}^*(K_{\parallel}, \kappa_{\perp}^L, \omega) \\ & \times \left\{ \left[\left[\frac{\kappa_{\perp}^T}{\kappa_{\perp}^T} \right]^2 |g_{1,x}|^2 \mathbf{e}_x + g_{1,x}^* g_{1,y} \mathbf{e}_y \right] \mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^T, \omega) \frac{[\kappa_{\perp}^T - (\kappa_{\perp}^L)^*]^n}{\mathcal{N}_0^T(\kappa_{\perp}^T - (\kappa_{\perp}^L)^*)} \exp\{i[\kappa_{\perp}^T - (\kappa_{\perp}^L)^*]z\} \right. \\ & \left. + \left[\frac{K_{\parallel}}{\kappa_{\perp}^L} \right]^2 |g_{1,x}|^2 \mathbf{e}_x \mathcal{R}_{\perp}(K_{\parallel}, \kappa_{\perp}^L, \omega) \frac{(2i \text{Im}\kappa_{\perp}^L)^n}{\mathcal{N}_0^T(2i \text{Im}\kappa_{\perp}^L)} \exp[-2(\text{Im}\kappa_{\perp}^L)z] \right\} . \quad (119) \end{aligned}$$

The polariton-plasmon contribution to the electromagnetic rectification process is given by the terms containing the factor

$$\exp[i(\text{Re}\kappa_{\perp}^{\text{L}} - \text{Re}\kappa_{\perp}^{\text{T}})z] \exp[-(\text{Im}\kappa_{\perp}^{\text{L}} + \text{Im}\kappa_{\perp}^{\text{T}})z]$$

and its complex conjugate. The plasmon-plasmon contribution, which only can give rise to a nonlinear current density in the scattering plane, is given by the terms containing the spatially nonoscillating factor $\exp[-2(\text{Im}\kappa_{\perp}^{\text{L}})z]$.

B. The quantities \mathbf{g}_0 and \mathbf{g}_1 in the pole approximation

To determine the prevailing nonlinear magnetostatic field and the associated dc-current density in the collective-mode approximation, one has to calculate \mathbf{g}_0 via the asymptotic expression in Eq. (65). Since

$$\begin{aligned} \lim_{q_{\perp} \rightarrow \infty} \sigma^{\text{T}}(K_{\parallel}, \kappa \pm q_{\perp}, \omega) &= \lim_{q_{\perp} \rightarrow \infty} \sigma^{\text{L}}(K_{\parallel}, \kappa \pm q_{\perp}, \omega) \\ &= \frac{ine^2}{m\omega}, \end{aligned} \quad (120)$$

with $\kappa = \kappa_{\perp}^{\text{L}}, \kappa_{\perp}^{\text{T}}, -(\kappa_{\perp}^{\text{L}})^*,$ or $-(\kappa_{\perp}^{\text{T}})^*$, one obtains the fol-

lowing asymptotic behaviors:

$$\lim_{q_{\perp} \rightarrow \infty} \mathcal{N}_{\perp}^{\text{T}}(K_{\parallel}, \kappa \pm q_{\perp}, \omega) = \lim_{q_{\perp} \rightarrow \infty} (-q_{\perp}^2) \quad (121)$$

and

$$\lim_{q_{\perp} \rightarrow \infty} \mathcal{N}_{\perp}^{\text{L}}(K_{\parallel}, \kappa \pm q_{\perp}, \omega) = \left[\frac{\omega}{c_0} \right]^2 \left[1 - \left[\frac{\omega_p}{\omega} \right]^2 \right], \quad (122)$$

where $\omega_p = [ne^2/(m\epsilon_0)]^{1/2}$ is the plasma (circular) frequency. By combining the results in Eqs. (121) and (122) with the fact that

$$\lim_{q_{\perp} \rightarrow \infty} R_0(K_{\parallel}, \kappa \pm q_{\perp}, \omega) \propto \lim_{q_{\perp} \rightarrow \infty} q_{\perp}^{-3}, \quad (123)$$

it is a straightforward matter to demonstrate that the terms in Eqs. (104) and (108) which contain $R_0(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^{\text{T}})^*, \omega)$, $R_0(K_{\parallel}, q_{\perp} - (\kappa_{\perp}^{\text{L}})^*, \omega)$, $R_0^*(K_{\parallel}, \kappa_{\perp}^{\text{T}} - q_{\perp}, \omega)$, and $R_0^*(K_{\parallel}, \kappa_{\perp}^{\text{L}} - q_{\perp}, \omega)$ do not contribute to \mathbf{g}_0 . Hence, by means of the relation in Eqs. (121) and (122), one obtains, by inserting Eqs. (104) and (108) into Eq. (65), after a few steps of algebraic manipulations,

$$\begin{aligned} \mathbf{g}_0 &= \frac{mK_{\parallel}}{4\pi^2 ine^2} \left[\left[\frac{R_0(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega) \mathcal{R}_{\text{L}}(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega)}{\kappa_{\text{L}}} - \frac{R_0^*(K_{\parallel}, -(\kappa_{\perp}^{\text{L}})^*, \omega) \mathcal{R}_{\text{L}}^*(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega)}{\kappa_{\text{L}}^*} \right] \left[\frac{K_{\parallel}^2 c_0^2}{\omega^2 - \omega_p^2} - 1 \right] |g_{1,x}|^2 \mathbf{e}_x \right. \\ &\quad \left. - \left[\frac{g_{1,x} g_{1,y}^*}{\kappa_{\text{L}}} R_0(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega) \mathcal{R}_{\text{L}}(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega) - \frac{g_{1,x}^* g_{1,y}}{\kappa_{\text{L}}^*} R_0^*(K_{\parallel}, -(\kappa_{\perp}^{\text{L}})^*, \omega) \mathcal{R}_{\text{L}}^*(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega) \right] \mathbf{e}_y \right]. \end{aligned} \quad (124)$$

It is a noteworthy feature that \mathbf{g}_0 depends on the polariton field only via the quantities $g_{1,x}$ and $g_{1,y}$. In the pole approximation, these are given by

$$g_{1,x} = \frac{2c_0 K_{\perp}^i}{i\omega} \frac{E_{\perp}^{i,p}(\mathbf{K}_i, \omega)}{\left[\frac{c_0 \kappa_{\text{T}}}{\omega} \right]^2 \frac{K_{\perp}^i + \kappa_{\perp}^{\text{T}}}{\kappa_{\text{T}}^i} \mathcal{R}_{\text{T}}(K_{\parallel}, \kappa_{\perp}^{\text{T}}, \omega) + \left[\frac{K_{\parallel}}{\kappa_{\text{L}}} \right]^2 \mathcal{R}_{\text{L}}(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega)} \quad (125)$$

and

$$g_{1,y} = \frac{2E_{\perp}^{i,s}(\mathbf{K}_i, \omega)}{i \left[1 + \frac{\kappa_{\perp}^{\text{T}}}{K_{\perp}^i} \right] \mathcal{R}_{\text{T}}(K_{\parallel}, \kappa_{\perp}^{\text{T}}, \omega)}. \quad (126)$$

C. Hydrodynamic model

In the preceding part of this paper, the electromagnetic rectification stemming from polariton-plasmon and plasmon-plasmon interactions has been discussed without reference to a specific model for the linear and nonlinear bulk responses. By using excitation frequencies in the vicinity of the plasma edge, it is reasonable to describe the collective excitations on the basis of the well-known hydrodynamic model.²² For this model, nonlocal effects can be neglected in the transverse part of the linear-response tensor so that the polariton and plasmon dispersion relations take the forms

$$\kappa_{\perp}^I = \left[\frac{\omega^2 - \omega_p^2}{a_I} - K_{\parallel}^2 \right]^{1/2}, \quad I = \text{T or L}, \quad (127)$$

where $a_{\text{T}} = c_0^2$ and $a_{\text{L}} = 3\alpha(\omega, T)$, possibly with ω replaced by $\omega + i/\tau$ to account, via a damping constant τ^{-1} , phenomenologically for irreversible relaxation processes. The quantity $3\alpha(\omega, T)$ is the diffusion coefficient for the superconducting phase. Its explicit expression can be found in Ref. 29. For frequencies somewhat above the gap frequency, 3α equals the diffusion coefficient of the normal state, i.e., $3\alpha \approx 3v_F^2/5$. The residues \mathcal{R}_{T} and \mathcal{R}_{L} to be used are given also in Ref. 29 (use only the local value for \mathcal{R}_{T}). In the near-local (hydrodynamic) approximation one obtains for the nonlinear response function \mathbf{R}_0 the following explicit result:

$$\mathbf{R}_0(K_{\parallel}, \kappa_{\perp}^{\text{L}}, \omega) = \beta(\omega, T) \kappa_{\text{L}}, \quad (128)$$

where

$$\beta(\omega, T) = -\frac{\hbar^2 e^3}{12\pi^2 m^3 \omega} \int_0^\infty k^4 \left[\frac{2f_{\mathbf{k}}(1-f_{\mathbf{k}})}{k_B T \omega^2} \left[1 - \frac{\Delta_{\mathbf{k}}^2(T)}{E_{\mathbf{k}}^2} \right] + \frac{\hbar^2 \Delta_{\mathbf{k}}^2(T)}{E_{\mathbf{k}}^3} \frac{1-2f_{\mathbf{k}}}{(\hbar\omega)^2 - (2E_{\mathbf{k}})^2} \right] dk. \quad (129)$$

In the normal state, β is reduced to $\beta(\omega, T > T_c) = -ne^3/(2m^2\omega^3)$, cf. Eq. (89). The normal-state value of β is also the value obtained at frequencies far above the gap frequency. From Eq. (128), one obtains

$$\mathbf{R}_0^*(K_{\parallel}, -(\kappa_{\perp}^I)^*, \omega) = \beta^*(\omega, T)(\kappa_{\perp}^M)^*,$$

where $\kappa_{\perp}^M \equiv \mathbf{K}_{\parallel} - \kappa_{\perp}^I \mathbf{e}_z$ is obtained by mirroring (M) κ_{\perp}^I in the surface plane. In the collisionless limit ($\tau \rightarrow \infty$), $\beta^* = \beta$.

On the basis of Eqs. (127)–(129), together with the relevant ones from Ref. 29, explicit expressions for the prevailing dc-current density and the associated magnetic field can be obtained in a way which is adequate for numerical studies.

For the lower- T_c , conventional superconductors, specific superconducting features can probably be neglected in Eqs. (127)–(129) since the plasma frequency is far above the gap frequency. For the high- T_c superconductors, the situation is more complicated since the plasma frequency is lower and the gap frequency higher.³

Even when lower- T_c , conventional superconductors are considered, specific superconducting effects are still to be expected in the electromagnetic rectification process due to the presence of the Meissner screening in Eq. (63) [$S_0^T(q_{\perp})$ and $\mathcal{N}_0^T(q_{\perp})$ being involved], and the function $\mathcal{N}_0^T(q_{\perp})$ in the forced parts of Eqs. (63) and (66). Finally, let us point out that while it is impossible to describe the generation of the forced nonlinear current density within the framework of a local model, the free Meissner current density can in some case be treated on the basis of a local (London³⁰) approach. It is so because nonlocal effects occur only in even orders of q_{\perp} in $S_0^T(q_{\perp})$. In the London limit, S_0^T is given by

$$S_0^T = -\frac{ne^2}{m} + \frac{2e^2 \hbar^2}{3m^2 k_B T} \int_{-\infty}^{\infty} k^2 f_{\mathbf{k}}(1-f_{\mathbf{k}}) \frac{d^3 k}{8\pi^3}, \quad (130)$$

as is well known.²⁴

APPENDIX: CALCULATION OF ANALYTICAL EXPRESSION FOR $\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega)$ IN $T=0$ APPROXIMATION

To obtain an analytical expression for $\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega)$ let us rewrite Eq. (83) as follows:

$$\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega) = -\frac{\hbar e^3}{2m^2 \omega^3 V} \sum_{\mathbf{k}} f_{\mathbf{k}}^{\text{NS}} \left[\frac{2\mathbf{k} + \mathbf{Q}}{\hbar\omega + \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k} + \mathbf{Q}}} - \frac{2\mathbf{k} - \mathbf{Q}}{\hbar\omega + \varepsilon_{\mathbf{k} - \mathbf{Q}} - \varepsilon_{\mathbf{k}}} \right]. \quad (A1)$$

The form in Eq. (A1) is readily obtained making the substitution $\mathbf{k} \rightarrow \mathbf{k} - \mathbf{Q}$ in the summation containing the quantity $(2\mathbf{k} + \mathbf{Q})f_{\mathbf{k} + \mathbf{Q}}^{\text{NS}}/(\hbar\omega + \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k} + \mathbf{Q}})$. As is usual, we then replace the summation over the closely spaced \mathbf{k} points by an integral over \mathbf{k} space, i.e., $V^{-1} \sum_{\mathbf{k}}(\dots) \rightarrow \int(\dots) d^3 k / (8\pi^3)$. Since \mathbf{R}_0^{NS} is an uneven function of k_x and k_y in a Cartesian (k_x, k_y, k_z) coordinate system having the k_z axis along the direction of \mathbf{Q} , one must have $\mathbf{R}_0^{\text{NS}} \parallel \mathbf{e}_Q$. Thus, it is realized that the integrals which have to be calculated are of the types

$$I_{\pm} = \int_{-\infty}^{\infty} \frac{(2k_z \pm Q) f_{\mathbf{k}}^{\text{NS}}}{\hbar\omega - \frac{\hbar^2}{m} k_z Q \mp \frac{\hbar^2 Q^2}{2m}} \frac{d^3 k}{8\pi^3}. \quad (A2)$$

By introducing spherical coordinates (k, θ, φ) , performing a trivial φ integration, and making use of the $T=0$ approximation $f_{\mathbf{k}}^{\text{NS}} = \Theta(k_F - k)$, the two integrals of Eq. (A2) takes the form

$$I_{\pm} = \int_0^{k_F} \int_0^{\pi} \frac{(2k \cos\theta \pm Q) k^2 \sin\theta}{\hbar\omega - \frac{\hbar^2 Q}{m} k \cos\theta \mp \frac{\hbar^2 Q^2}{2m}} \frac{d\theta dk}{4\pi^2}. \quad (A3)$$

Performing the θ integration, one obtains

$$I_{\pm} = \frac{1}{4\pi^2 \beta} \left[-\frac{4}{3} k_F^3 - \left[\frac{2\alpha_{\pm} \pm Q}{\beta} \right] \int_0^{k_F} k \ln \left| \frac{\alpha_{\pm} - \beta k}{\alpha_{\pm} + \beta k} \right| dk \right], \quad (A4)$$

where $\alpha_{\pm} = \hbar\omega \mp \hbar^2 Q^2 / (2m)$ and $\beta = \hbar^2 Q / m$, and then after some integration by parts,

$$I_{\pm} = \frac{1}{4\pi^2 \beta} \left\{ -\frac{4}{3} k_F^3 - \left[\frac{2\alpha_{\pm} \pm Q}{\beta} \right] \left[\left[\frac{k_F^2}{2} - \frac{\alpha_{\pm}^2}{2\beta^2} \right] \ln \left| \frac{\alpha_{\pm} - \beta k_F}{\alpha_{\pm} + \beta k_F} \right| - \frac{\alpha_{\pm} k_F}{\beta} \right] \right\}. \quad (A5)$$

Finally, by utilizing the relation

$$\frac{\alpha_{\pm} - \beta k_F}{\alpha_{\pm} + \beta k_F} = \frac{u \mp z - 1}{u \mp z + 1}, \quad (A6)$$

writing down the expression for $I_+ - I_-$, and carrying out a number of trivial algebraic manipulations, one obtains the expression for $\mathbf{R}_0^{\text{NS}}(\mathbf{Q}, \omega)$ given in Eq. (87).

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