High-frequency electrodynamic response of strongly anisotropic clean normal and superconducting metals

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We consider the influence of quasiparticle relaxation and nonlocality on the complex conductivity and microwave surface impedance of isotropic and quasi-two-dimensional metals in the normal and superconducting states for arbitrary electronic parameters intermediate between the classical skin effect and extreme anomalous limits. We describe the superconducting state by a two-fluid model with a nonlocal, retarded quasiparticle response and derive an expression for the surface impedance at low temperatures in the extreme anomalous limit. We show how microwave measurements can be used to probe the *k* dependence of the superconducting order parameter in layered materials.

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I. INTRODUCTION

The high-frequency electromagnetic response of normal and superconducting metals, measured by the complex surface impedance $Z_s = R_s + iX_s$, provides a valuable probe of their electronic properties. Early measurements by Pippard[1] and Chambers[2] demonstrated the power of such measurements in the determination of the Fermi surface (FS) of copper.[1,2] More recently, microwave measurements on $YBa_2Cu_3O_{7-\delta}$ helped establish the *d*-wave nature of the anisotropic high-temperature cuprate superconductors[3,4] and have provided important information on the temperature-dependent density and lifetime of the thermally excited quasiparticles.[5]

We have recently reported first Z_s measurements of superconducting Sr_2RuO_4 with a transition temperature $T_c \sim 1.4$ K,[6] which is believed to be a spin-triplet superconductor with *p*- or possibly *f*-wave symmetry of the order parameter.[7–9] Crystals of Sr_2RuO_4 can be grown with extremely high purity (a few ppm) having very long quasiparticle lifetimes, $\tau \sim 10^{-11}$ s, and mean-free paths, $l = v_F \tau \sim 1$ μm.[10] The quasiparticle lifetime at low temperatures can, therefore, become comparable with the microwave period $\omega\tau \sim 1$ and the mean-free path can exceed both the classical skin depth $\delta = (2/\mu_0 \omega \sigma_0)^{1/2}$ and London penetration depth $\lambda_L = (\mu_0 n e^2/m)^{-1/2}$: μ_0 is the vacuum permeability, v_F the Fermi velocity, *n* the effective quasiparticle density, *m* their effective mass, and $\sigma_0 = ne^2\tau/m = \tau/\mu_0\lambda_L^2$ the dc conductivity. Long relaxation times and nonlocal effects strongly influence the surface impedance of normal metals and superconductors.[6,11,12]

In Sec. II A and II B, we compute the surface impedance of a simple isotropic metal with an electromagnetic response intermediate between the classical skin effect and extreme anomalous limits. The results are then extended to strongly anisotropic, quasi-two-dimensional (2D), normal metals for microwave geometries of experimental interest (Sec. II C). In Sec. III A and III B, we introduce and analyze a simple two-fluid model to describe the influence of quasiparticle relaxation and nonlocality in the superconducting state. The extension of such a model to superconductors with a nodal order parameter, such as the cuprates or Sr_2RuO_4, is discussed in Sec. III C.

II. ELECTRODYNAMIC RESPONSE OF CLEAN NORMAL METALS

A. Normal skin effect and extreme anomalous limits

For a normal metal when $l \ll \delta$, the electromagnetic response at high frequencies remains local, with a frequency-dependent, complex-valued, conductivity $\sigma_\omega = \sigma_0/(1+i\omega\tau)$ and a surface impedance, $Z_s = (1+i\omega\tau)^{1/2} Z_0$, where $Z_0 = (i\mu_0\omega/\sigma_0)^{1/2} = \mu_0\omega\lambda_L \times (1+i) \times (2\omega\tau)^{-1/2}$ is the classical skin-effect result.[12] In the extreme anomalous limit, $\omega\tau$ and $\beta \gg 1$, Z_s approaches $\gamma\mu_0\omega\lambda_L \times (1+i\sqrt{3}) \times \beta^{1/3}$, where $\gamma = 4/9(\sqrt{3}/2\pi)^{1/3} = 0.289$, and $\beta \equiv v_F/\omega\lambda_L = [\omega\tau(\lambda_L/l)]^{-1}$ is a nonlocality parameter. In this limit, Z_s is determined by the extremal area of the FS parallel to the direction of current flow, which enabled Pippard to map out the FS of copper.[1] Such a regime can easily be realized for conventional metals at microwave frequencies ($f = 10$–60 GHz). However, the smaller v_F and larger λ_L values of the cuprates and ruthenates lead to smaller β values (β is only ~ 5 for Sr_2RuO_4 at $f = 20$ GHz, where we have taken $\lambda_L \sim 170$ nm and $v_F \sim 10^5$ m/s [Ref. 13]). Microwave measurements on such materials therefore probe the intermediate region between the classical and extreme anomalous limits.

B. Intermediate case for isotropic metals

Reuter and Sondheimer (RS) were the first to develop a comprehensive nonlocal theory for the surface impedance Z_s of *normal metals* based on the Boltzmann equation using a path-integral approach.[11] An alternative derivation, based on the Fourier components of the penetrating microwave field, was introduced by Pippard[1] and discussed by Kittel,[12] which for specular reflection gives results that can be shown to be identical to RS for all values of $\omega\tau$ and β. We adopt here the latter approach to derive the surface impedance Z_s of 3D and quasi-2D metals in *both* the normal *and* superconducting states as a function of $\omega\tau$ and β, for arbitrary relaxation times and nonlocal corrections.

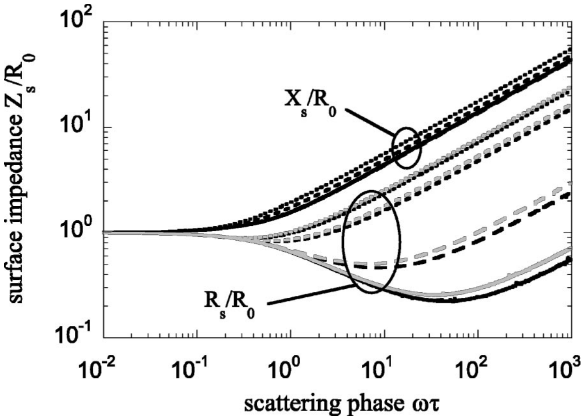

FIG. 1. $Z_s(\omega\tau)$ computed from Eq. (1), normalized to R_0, for $\beta=0.5$, 1, 5, 10 (solid, coarse dashed, fine dashed, and dotted curves) for a 3D (black curves) and a quasi-2D metal (gray curves).

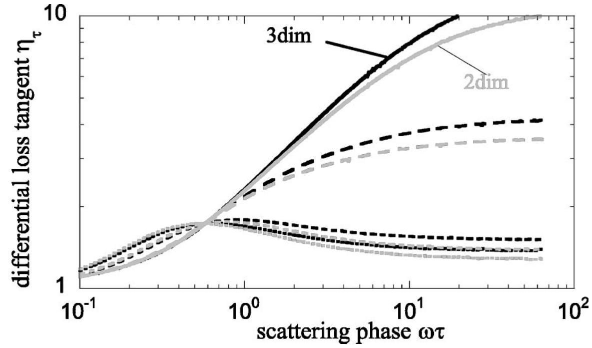

FIG. 2. Differential loss tangent $\eta_\tau(\omega\tau)$ corresponding to the data in Fig. 1.

We start by extending the RS and Pippard results for a spherical FS to arbitrary $\omega\tau$ and β values.[6,14] We assume for simplicity a single quasi-free-electron band with an isotropic scattering time and specular reflection at the surface. The results can easily be extended for diffuse scattering at the surface and for metals like the ruthenates with more than one contributing conduction band. The surface impedance is determined by the ratio of the tangential electric-to-magnetic field at the surface of a metal, which can be expressed in terms of the wave-vector-dependent transverse conductivity $\tilde{\sigma}_{3d}(q)$,[12]

$$Z_s=\frac{2i\mu_0\omega}{\pi}\int_0^\infty\frac{dq}{q^2+i\mu_0\omega\tilde{\sigma}_{3d}(q)}. \quad (1)$$

For Fourier components of the penetrating field varying as $e^{i(qx+\omega t)}$, the three-dimensional, nonlocal, and relaxation-time-dependent, transverse conductivity is given by

$$\tilde{\sigma}_{3d}(q)=\frac{3}{4}\sigma_0\int_{-1}^{+1}d(\cos\theta)\frac{\sin^2\theta}{1+i(\omega\tau+ql\cos\theta)}, \quad (2)$$

where θ is the angle at which the quasiparticles travel from the normal to the surface. We write $\tilde{\sigma}_{3d}(q)=\sigma_\omega\times\kappa_{3d}(q')$, where σ_ω accounts for relaxation while the function

$$\kappa_{3d}(q')=\frac{3}{2}\frac{(1+q'^2)\tan^{-1}(q')-q'}{q'^3} \quad (3)$$

accounts for nonlocality, setting $q'\equiv ql/(1+i\omega\tau)$. $\kappa(q')$ decreases from unity for small q' values to $3\pi/4q'$ at large values. Inserting these asymptotic values into Eq. (1), we recover the usual results for Z_s in the classical skin effect and extreme anomalous limits.

The black curves in Fig. 1 show Z_s computed from Eqs. (1) and (2) for various values of the nonlocality parameter β as a function of $\omega\tau$. The surface resistance and reactance have been normalized to their classical skin-effect values R_0, i.e., evaluated in the absence of relaxation and nonlocality. For small $\omega\tau$, only relaxation is important with R_s decreasing and X_s increasing to first order as $R_0(1\pm 1/2\omega\tau)$. For large $\omega\tau$ in the absence of nonlocal corrections, X_s approaches the limiting value $\mu_0\omega\lambda_L$ (near perfect diamagnetic screening), while R_s decreases towards the frequency-independent limiting value $\mu_0\lambda_L/2\tau$. Nonlocality leads to an increase of both R_s and X_s for large $\omega\tau$ values, with a limiting $\omega^{2/3}$ frequency dependence and a β-dependent ratio that approaches $1/\sqrt{3}$ for sufficiently large β.[1,11]

In practice, it is often easier to extract changes ΔX_s rather than absolute X_s values from microwave measurements. The differential loss factor, $\eta_\tau\equiv(\partial R_s/\partial\omega\tau)/(\partial X_s/\partial\omega\tau)$, can be determined accurately without the need for absolute calibration.[6] Figure 2 displays typical results for the $\omega\tau$ dependence of η_τ for different β values. While η_τ increases continuously in the local limit, nonlocal effects lead to a β-dependent limiting value at large $\omega\tau$. For $\beta>1$, a peak in η_τ develops, the height of which depends on β and $\omega\tau$. Measuring Z_s over a range of frequencies and fitting to such curves allows, in principle, β and $\omega\tau$ to be determined at any temperature and to relate these parameters to FS properties.

C. Intermediate case for strongly anisotropic metals

We now study the electromagnetic response of strongly anisotropic, quasi-2D, metals, like the cuprates and ruthenates. For such materials, measurements are often restricted to platelet crystals or epitaxial films with their major flat faces parallel to the ab planes and edges parallel to the c direction. We indicate the alignment of the electromagnetic field H_{rf} relative to the crystal axes for the two most-frequently used measurement configurations (1 and 2) in Fig. 3, together with the corresponding orientation of the quasicylindrical Fermi surface in k space. The electromagnetic field induces currents as indicated by the arrows, which correspond to a displacement of the FS along the same direction

We first consider configuration 1 [Fig. 3(a)], with the c direction of the platelet crystal parallel to H_{rf}. The induced microwave currents flow parallel to the ab planes on both the major flat faces and the edge surfaces of the sample.[15] Because of the large demagnetizing effect, the induced microwave currents are strongly peaked at the outer edges of the crystal.[15,16] Conformal mapping and numerical methods enable us to determine the relative contributions to the microwave response from the inhomogeneous field distribution

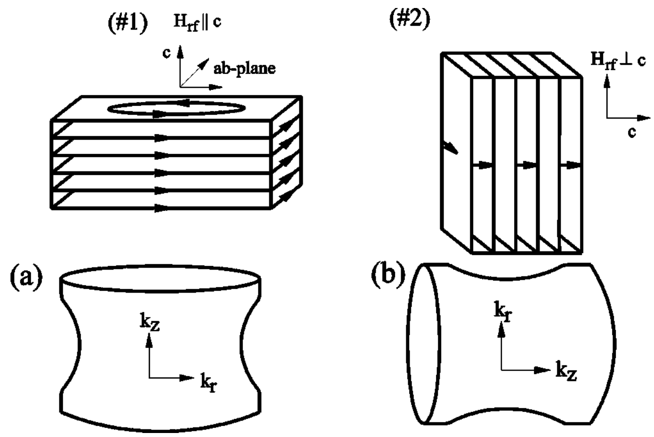

FIG. 3. Real- and k-space representations of a layered sample with a quasicylindrical FS in the two conventional configurations 1 (a) and 2 (b), where the currents decay within or perpendicular to the planes.

at the flat and edge faces of a thin rectangular strip accurately or a disk-shaped sample approximately. For an aspect ratio of 10, both faces contribute almost equally. We can ignore nonlocal corrections to the surface impedance from spatial variation of the field parallel to the faces, as these are only significant on a length scale of the order of the sample thickness, $d \gg X_s/\mu_0\omega$. For a 2D metal, nonlocality is only important on the edge surfaces, where the mean-free path in the ab planes can exceed the microwave penetration depth. Measurements with platelet crystals with different aspect ratios could be used to distinguish between the contributions from the edge and flat faces.

We first evaluate the surface impedance for the edge faces. In this configuration, the transverse conductivity for a 2D metal is given by

$$\tilde{\sigma}_{2d}(q) = \frac{1}{\pi}\sigma_{ab}\int_0^{2\pi} d\varphi \frac{\sin^2\varphi}{1+i(\omega\tau + ql\cos\varphi)}, \qquad (4)$$

where φ is the angle in the ab plane relative to the induced current flow and σ_{ab} is the in-plane dc conductivity. The corresponding nonlocality function $\kappa_{2d}(q') \equiv \tilde{\sigma}_{2d}(q)/\sigma_\omega$ is given by

$$\kappa_{2d} = \frac{2}{q'^2}(\sqrt{1+q'^2} - 1), \qquad (5)$$

which has a similar q' dependence as κ_{3d}, varying from unity for small q' to $2/q'$ for large arguments. The dependence of Z_s on $\omega\tau$ and β is therefore almost identical to that derived for a spherical FS, as illustrated by the gray curves in Figs. 1 and 2.

We now turn to the surface impedance for currents induced on the flat surfaces in both configurations 1 and 2 (Fig. 3). For an ideal 2D metal, quasiparticle motion is restricted to the ab planes. The microwave response, therefore, remains local at all frequencies with a surface impedance $Z_s = (1+i\omega\tau)^{1/2}(i\mu_0\omega/\sigma_{ab})^{1/2}$.

The cylindrical FS of real quasi-2D metals will be slightly warped, with radial Fourier coefficients of the FS of a simple tetragonal metal varying as $\Sigma_{m,n} a_{mn}\cos(4m\varphi)\cos(2n\pi k_z/k_c)$, where k_c is the Brillouin zone dimension in the c direction. Quasiparticles can therefore travel at a small angle ψ out of the planes and hence contribute to nonlocal effects, where

$$\psi = (2\pi k_F/k_c)\sum_{m,n} n a_{mn}\cos(4m\varphi)\sin(2n\pi k_z/k_c) \qquad (6)$$

and k_F is the in-plane Fermi wave vector. This effect can be treated as a small perturbation, resulting in an increase of the surface impedance by

$$Z_s = (1+i\omega\tau)^{1/2}\left[\frac{i\mu_0\omega}{\sigma_{ab}}\right]^{1/2}\{1 + i/2(\pi\beta A)^2\vartheta^3\}, \qquad (7)$$

where $A^2 = 2\Sigma_n n^2 a_{0n}^2 + \Sigma_{m\neq 0,n} n^2 a_{mn}^2$ and $\vartheta \equiv \omega\tau/(1+i\omega\tau)$. The corrections are only second order in the already small perturbation of the FS and can therefore be neglected in most cases of experimental interest, such as the very slightly warped Fermi surface of body-centered-cubic Sr_2RuO_4.[17]

In configuration 2, the microwaves also penetrate the edge surfaces perpendicular to the ab planes, where the electrodynamic response is governed by the dc c axis conductivity σ_c. For strongly anisotropic metals, $\sigma_c \ll \sigma_{ab}$, so that quasiparticle relaxation and nonlocality can generally be neglected. The c-axis surface impedance will then be given by the classical skin-effect result $Z_{s,c} = (i\mu_0\omega/\sigma_c)^{1/2}$.

The effective surface impedance in this configuration will be $\alpha_{ab}Z_{s,ab} + \alpha_c Z_{s,c}$, where α_{ab} and α_c are the fractional areas of the surfaces parallel and perpendicular to the planes (we assume that all crystal dimensions are much larger than microwave penetration lengths). Although measurements in this configuration are uncomplicated by demagnetization and nonlocal effects, measurements on samples of different thickness, cut from the same parent crystal, have to be performed to extract Z_{ab} and Z_c.[3]

III. ELECTRODYNAMIC RESPONSE OF CLEAN SUPERCONDUCTORS

A. Two-fluid model

We now consider the influence of relaxation and nonlocality on the electromagnetic response in the *superconducting* state. We extend the standard two-fluid model by incorporating an effective transverse conductivity

$$\tilde{\sigma}_s(q) = \frac{1}{i\mu_0\omega\lambda_L^2}\left[(1-f_n) + i f_n \frac{\omega\tau}{1+i\omega\tau}\kappa(q')\right], \qquad (8)$$

where f_n is the normal state fraction of quasiparticles and $\kappa(q')$ accounts for the nonlocal effects as before. We assume that all microscopic details related to the density of states and coherence effects can be included in the temperature and frequency dependences of f_n and τ.[18,19] While we retain the nonlocal response of the thermally excited quasiparticles, we assume a local response for the superconducting pairs, which is appropriate in the extreme type-II limit

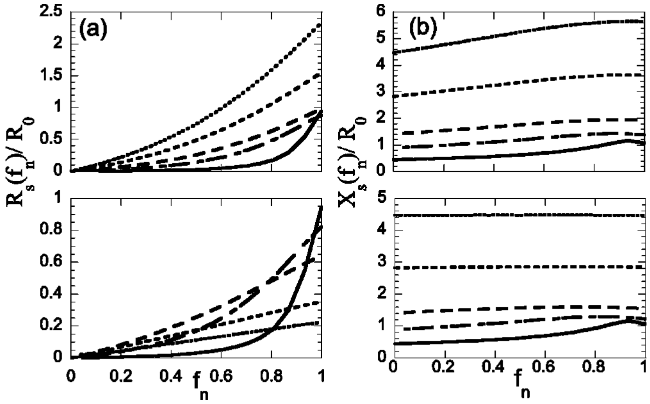

FIG. 4. (a) $R_s(f_n)/R_0$ derived from the two-fluid model for $\omega\tau=0.1$, 0.4, 1, 4, and 10 (solid, short-long dashed, long dashed, short-dashed and dotted curves), for $\beta=0.1$ (bottom), and 10 (top). (b) Corresponding $X_s(f_n)/R_0$ for $\beta=0.1$ (bottom) and 10 (top).

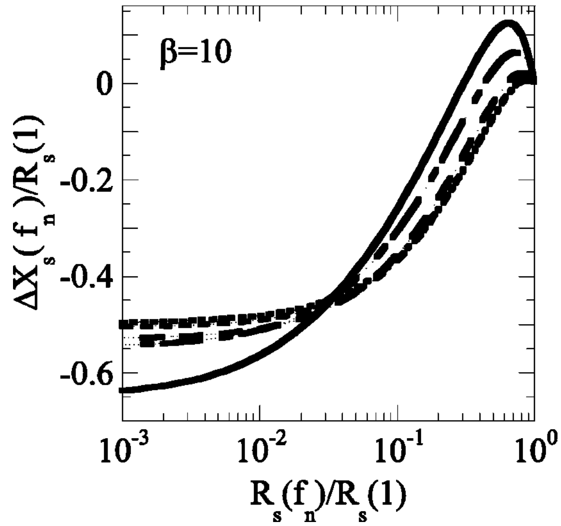

FIG. 5. Parametric plot of $\Delta X_s(f_n)$ versus $R_s(f_n)$, both normalized to $R_s(1)$, for $\omega\tau=0.1$, 0.4, 1, 4, 10 (solid, short long dashed, long dashed, short dashed, dotted curves) and $\beta=10$, calculated for a 3D metal using $v_F=8\times10^4$ m/s and $f=10$ GHz.

and if complications from non s-wave symmetry are ignored (see Sec. III C). This assumption enables us to use a phenomenological approach instead of fully nonlocal microscopic theories.[18,20]

In the absence of nonlocal corrections, we recover the usual two-fluid result $Z_s=i\mu_0\omega\lambda_L[1-f_n/(1+i\omega\tau)]^{1/2}$, which has been used to extract $f_n(T)$ and $\tau(T)$ for the cuprates.[3–5,19,21] A similar approach can be used to extract these parameters for Sr_2RuO_4, from Z_s measurements with the ab planes parallel to the microwave fields, in both configurations 1 and 2. However, for superconductors intermediate between the classical and extreme anomalous limits, and for microwave fields parallel to the edge faces of ruthenate crystals in configuration 1, nonlocal corrections have to be considered. In both cases, we initially ignore any k dependence of the quasiparticle properties. We can then compute Z_s from Eq. (1) using the generalized conductivity defined in Eq. (8) including the q-dependence of $\kappa(q')$ in Eqs. (3) and (5).

The surface impedance now depends on three parameters f_n, $\omega\tau$, and β. Figures 4(a) and 4(b) illustrate the f_n dependences of R_s and X_s, normalized to R_0, for various $\omega\tau$ values and small ($\beta=0.1$) and significant nonlocal corrections ($\beta=10$). The results are shown for a spherical FS but differ by only a few percent from those of a quasi-2D metal.

For all β and $\omega\tau$ values $R_s(f_n)$ decreases monotonically below T_c, though the rate of decrease is related to that of the penetration depth, which is strongly $\omega\tau$ dependent. For small $\omega\tau$ values there is a pronounced peak in $X_s(f_n)$ below T_c, which moves to lower temperatures (smaller f_n) with increasing purity and frequency, as observed in recent measurements on Sr_2RuO_4.[6] The peak in the reactance is a generic feature of the two-fluid model[22] and has the same origin as the peak observed in $X_s(H_{dc})$ in measurements of cuprate bicrystals.[23]

In Sr_2RuO_4 single crystals, this peak is a dominant feature of the X_s measurements because of the very long quasiparticle relaxation time. For less-pure superconductors the peak occurs rather close to T_c and tends to be obscured by fluctuations and impurity broadening of T_c as well as by coherence effects. The initial increase in reactance below T_c corresponds to a *decrease* in shielding. This arises because, for small $\omega\tau$, the normal and superconducting shielding currents are in phase quadrature. On entering the superconducting state, the contribution to the shielding currents from quasiparticles initially decreases as Δf_n, while the contribution from Cooper pairs only increases as Δf_n^2. When $\omega\tau\gg1$, quasiparticles in the normal state are almost as effective as superconducting pairs in shielding the microwave fields, so there is little change in X_s below T_c, as illustrated in Fig. 4(b) and confirmed in our measurements. Nonlocality only becomes important for $\omega\tau\geq0.1$, where it has an increasingly large influence on the shape of the curves below T_c.

To extract reliable information on f_n, $\omega\tau$, and β when nonlocality is important, requires accurate measurements over a range of frequencies in order to fit data to computed results. A useful alternative is to measure changes in the surface reactance against the surface resistance. In typical cavity perturbation measurements, such data can be extracted with high precision.[6] Figure 5 shows parametric plots of ΔX_s versus R_s, normalized to the surface resistance at T_c, resulting from Fig. 4 for selected $\omega\tau$ values and $\beta=10$. Comparing measured data with such computed results, $\omega\tau$ and β values and hence accurate f_n values can be extracted at any temperature within the superconducting state.

B. Extreme anomalous limit of the generalized two-fluid model

It is instructive to consider the limiting form of Z_s at low temperatures as $f_n\to0$. In the absence of nonlocality and relaxation, the surface resistance $R_s=(1/2)\mu_0^2\omega^2\lambda^3f_n\sigma_0$. However, for a clean superconductor with $l\gg\lambda_L$, the quasiparticles only spend a small fraction of their lifetime within the penetration length. One might anticipate, that the losses would therefore be reduced by a factor $\sim\lambda_L/l$. To evaluate the superconducting surface impedance in the *extreme anomalous limit*, we write Eq. (1) as

$$Z_{s,\infty}=\frac{2i\mu_0\omega}{\pi}\int_0^\infty\frac{dq}{q^2+\lambda_L^{-2}[1-f_n+2if_n\omega\tau(ql)^{-1}]},$$
$$=\frac{2i}{\pi}\mu_0\omega\lambda(T)\int_0^\infty\frac{x\,dx}{x(x^2+1)-\mathrm{i}a}, \quad (9)$$

where $x\equiv q\lambda(T)$, $a\equiv 2f_n/[(1-f_n)\beta(T)]$, $\beta(T)=v_F/\omega\lambda(T)$, and $(1-f_n)\lambda_L^{-2}=\lambda^{-2}(T)$. For comparison with the cuprates and ruthenates, we have used the limiting 2D nonlocal expression for the transverse conductivity, according to the discussion of Eq. (5).

In this limit, the integral is dominated by small x values in the denominator, $x\ll|ia|$, reflecting the long mean-free paths of the quasiparticles. To first order in a Eq. (9) yields

$$Z_s\approx i\mu_0\omega\lambda(T)[(1-a)-i(2/\pi)a(\ln 1/a+1/2)]. \quad (10)$$

The surface resistance is given by $R_{s,\infty}\approx(8/\pi)R_s(\lambda_L/l)\ln(\beta/2f_n)$, which contains a logarithmic correction to our anticipated result and leads to a weaker frequency dependence than in the local limit. We note also that nonlocality leads to a linear frequency dependence of the penetration depth through the parameter a.

C. Implications of a nodal order parameter

It is important to consider how the previous results would be affected by a non-s-wave symmetry of the superconducting order parameter. Of particular interest are the layered superconductors with possible nodes along specific directions, such as the $(k_x^2-k_y^2)$ dependence for the d-wave cuprates and the $(k_x\pm ik_y)$ or $(k_x-ik_y)(k_x^2-k_y^2)$ dependences recently suggested for Sr_2RuO_4, consistent with a p- or f-wave symmetry of the order parameter.[7–9]

For platelet crystals in a perpendicular field, only quasiparticles traveling parallel to the edge directions contribute to the electromagnetic response of the edge faces. Microwave measurements therefore provide a powerful tool to investigate the k dependence of the quasiparticle properties. In particular, one would expect very different values and temperature dependences of the surface impedance for [100] edges, where the quasiparticles would experience the full energy gap, and for [110] edges, which would probe the nodal regions. For non-s-wave superconductors, the existence of surface states[24] and the unavoidable depression of the order parameter by nonspecular reflection at the surface could also lead to significant sample geometry effects, further complicating the application and interpretation of our simple two-fluid model. Finally, it might be necessary to take into account the enhanced coherence length along any nodal directions, which may result in a nonlocal response of the superconducting pairs. All such considerations require microscopic modeling, beyond the scope of the present paper.

IV. SUMMARY

We have calculated the electromagnetic response of 3D and 2D metals in both the normal and superconducting states for arbitrary relaxation times and mean-free paths. The influence of nonlocality on the measured surface impedance of quasi-2D metals depends strongly on the microwave configuration used. For measurements with microwave field and induced currents parallel to the ab planes, nonlocal corrections are unimportant. However, for configurations in which the microwave field is parallel to the c direction with currents induced in the ab planes, nonlocal corrections are similar to those of a 3D metal. The surface impedance in the *superconducting state* has been derived using a generalized two-fluid model where the response of the Cooper pairs remains local but the nonlocal response of the quasiparticles is retained. A simple way to compare the computed results with measured data and to extract the key physical parameters from microwave measurements is based on the parametric correlation between surface resistance and reactance. Finally, we consider nonlocal effects in non-s-wave superconductors and show that microwave measurements can, in principle, provide detailed information on any k dependence of the order parameter, though such measurements may be complicated by other geometrical and surface-dependent effects.

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