

fect the lifetimes of the sublevels, and will give them an additional width, which must be added to the gamma-ray and internal-conversion contributions in determining the overall width of the observed line. Nuclear-relaxation effects differ from the other contributions to the widths in that they can be different for different sublevels. Thus, if the nuclear relaxation is due to magnetic-dipole effects, a nucleus in the  $+\frac{3}{2}$  state can make transitions only to the  $+\frac{1}{2}$  state. One in the  $+\frac{1}{2}$  state, on the other hand, can make transitions to the  $+\frac{3}{2}$  state and also to the  $-\frac{1}{2}$  state, which is degenerate with the  $+\frac{1}{2}$  state. The width of a level  $n$  is given by the sum of the transition probabilities out of that level. Hence, for magnetic-dipole relaxation,

$$1/\tau_{3/2} = P_{3/2 \rightarrow 1/2},$$

and

$$1/\tau_{1/2} = P_{1/2 \rightarrow 3/2} + P_{1/2 \rightarrow -1/2}.$$

If the temperature is sufficiently high so that  $P_{3/2 \rightarrow 1/2} = P_{1/2 \rightarrow 3/2}$ , we expect that the  $\pm\frac{1}{2}$  levels in the excited state in Fig. 1 will be broader than the  $\pm\frac{3}{2}$  levels. This too will give rise to an asymmetry in the quadrupole pattern. The temperature dependence of this effect depends, however, on the temperature dependence of the nuclear spin-lattice relaxation time. If this time decreases as the temperature increases, than the asymmetry will become more pronounced with increasing temperature, as is the case for Goldanskii's effect. In order to observe effects of nuclear relaxation, the relaxation times must be of the order of, or shorter than, the radiative lifetime of the excited nuclear state, i.e., about  $10^{-8}$  sec. Such times are quite short for nuclear-relaxation

processes, but they are not unreasonable for nuclei in magnetic materials.

Experimentally, it is possible to distinguish relaxation-induced asymmetries from the Goldanskii mechanism, apart from the temperature dependence, by observing the quadrupole pattern from a single-crystal source. The relaxation effects will give a broadening which affects the same line regardless of the direction of observation. The anisotropic Debye-Waller factor, on the other hand, will broaden only those gamma rays which are emitted in a specific direction, namely, along the direction in which the ion can most easily recoil.

The phenomena described here are a part of a large number of interesting and unusual line shapes which should be observable whenever fluctuating fields are present. A theoretical treatment which can be applied to ferromagnetic and antiferromagnetic substances, as well as to paramagnets, and which includes the effects of fluctuating electric field gradients, has been worked out, and details of the theory and the experimental results to which it is applicable will be given separately.

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<sup>3</sup>A. M. Afanasev and Yu. Kagan [*Zh. Eksperim. i Teor. Fiz.* **45**, 1660 (1963) [translation: *Soviet Phys.-JETP* **18**, 1139 (1964)]] have given a detailed treatment of this aspect of the problem for one-phonon electronic relaxation processes.

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## MAGNETIC FIELD DEPENDENCE OF THE MICROWAVE SURFACE IMPEDANCE IN PURE SUPERCONDUCTORS\*

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There have been a number of experimental works<sup>1-6</sup> on the microwave surface impedance of superconductors in the presence of static magnetic fields. They have found, especially, that the surface impedance decreases as the static field increases for temperatures close to the critical temperature. The difficulty in accounting for this phenomenon in terms of the

two-fluid model has been stressed by Pippard.<sup>7</sup>

In the present note we will show that many of the features of the experimentally observed field dependence of the surface impedance follow directly from the current microscopic theory. We calculate the surface impedance in pure superconductors in the presence of magnetic fields by using the techniques developed

by Abrikosov, Gor'kov, and Khalatnikov<sup>8</sup> (AGK).

The calculation is much simplified through the observation that the effect of the static magnetic field is equivalent to the shift of the excitation energy of the quasiparticles from  $\epsilon_p$  to  $\epsilon_p - \vec{p} \cdot \vec{v}_S$ , where  $\vec{p}$  is the momentum of the quasiparticle and  $\vec{v}_S$  the flow velocity of the condensed pairs given as  $\vec{v}_S = (e/c)\vec{A}$ , where  $\vec{A}$  is the vector potential. There is, of course, another effect coming from the change of the equilibrium value of the ordering parameter  $\Delta$  in the presence of fields. The latter effect, which might be important in the case of alloys where the electronic mean free path is short, turns out to be extremely small in the case of pure superconductors. We discard it in the following consideration.

We assume that both the static magnetic field  $H$  and the high-frequency field  $H_{rf}$  are parallel to the surface of the sample and their mutual angle is  $\gamma$ . As we are interested in pure superconductors, we consider only the anomalous skin limit ( $kv > \Delta$ , where  $v$  is the Fermi velocity,  $\Delta$  the temperature-dependent ordering parameter, and  $k$  the momentum of the high-frequency field which is directed perpendicular to the surface). In the following we adopt the

units  $\hbar = k_B = 1$ .

Two Green's functions describing the current-carrying state has been derived previously.<sup>9</sup> Following AGK, we have for the Fourier transform of the current density<sup>8</sup>

$$j(k, \omega) = -\frac{3e^2 N \Delta}{4m v k} Q(\omega, \alpha, \gamma) A(k, \omega), \quad (1)$$

where  $N$  is the density of electrons and  $\omega$  the angular frequency of the incident electromagnetic wave. The parameter  $\alpha$  is given as

$$\alpha = (ev/c)A = (ev/c)\delta H, \quad (2)$$

where

$$\delta = \frac{\sqrt{3}}{2\pi} \left[ \frac{c^2 m v}{3\pi^3 e^2 N \Delta \tanh(\Delta/2T)} \right]^{1/3}$$

is the Pippard penetration depth.

$Q(\omega, \alpha, \gamma)$  is given as

$$Q(\omega, \alpha, \gamma) = \frac{1}{\pi} \int_0^{2\pi} \sin^2 \varphi d\varphi F(\omega, \alpha \sin(\varphi - \gamma)), \quad (3)$$

where

$$\text{Re}F(\omega, x) = \pi^2 \tanh[(\Delta + x)/2T], \quad (4)$$

and<sup>10</sup>

$$\text{Im}F(\omega, x) = \pi \int_1^\infty d\epsilon \left[ \epsilon \left( \frac{\omega}{\Delta} + \epsilon \right) + 1 \right] \left[ \tanh\left( \frac{\omega + \epsilon \Delta + x}{2T} \right) - \tanh\left( \frac{\epsilon \Delta + x}{2T} \right) \right] \left\{ (\epsilon^2 - 1) \left[ \left( \epsilon + \frac{\omega}{\Delta} \right)^2 - 1 \right] \right\}^{-1/2}. \quad (5)$$

In deriving the above expressions we assume  $\omega/\Delta \ll 1$ , which is the situation encountered in many experiments. Combining Eq. (1) with the Maxwell equation, we obtain the following expression for the surface impedance<sup>10</sup>:

$$Z(\omega, \alpha, \gamma)/R_n = 2i[\pi\omega/\Delta Q(\omega, \alpha, \gamma)]^{1/3}, \quad (6)$$

where  $R_n = \sqrt{3}(m v c^2 \pi / 3 N e^2)^{1/3} \omega^{2/3} / c^2$  is the surface resistance in the normal state.

We shall now consider some limiting cases.

(1) Low-temperature region where  $\Delta \gg T$ .—In this region we have

$$\text{Re}Q(\omega, \alpha, \gamma) = \pi^2 \left\{ 1 - 2 \sum_{n=1}^{\infty} (-1)^{n+1} e^{-n\Delta/T} \left[ I_0\left(\frac{n\alpha}{T}\right) + \cos 2\gamma I_2\left(\frac{n\alpha}{T}\right) \right] \right\}, \quad (7)$$

$$\text{Im}Q(\omega, \alpha, \gamma) = 4\pi \sum_{n=1}^{\infty} (-1)^{n+1} \sinh \frac{n\omega}{2T} K_0\left(\frac{n\omega}{2T}\right) e^{-n\Delta/T} \left[ I_0\left(\frac{n\alpha}{T}\right) + \cos 2\gamma I_2\left(\frac{n\alpha}{T}\right) \right], \quad (8)$$

and

$$\frac{Z(\omega, \alpha, \gamma) - Z(\omega, 0, \gamma)}{R_n} = \frac{8}{3\pi} \left( \frac{\omega}{\pi\Delta} \right)^{1/3} \left\{ \sum_{n=1}^{\infty} (-1)^{n+1} e^{-n\Delta/T} \left[ \sinh \frac{n\omega}{2T} K_0 \left( \frac{n\omega}{2T} \right) + \frac{\pi}{2} i \right] \left[ I_0 \left( \frac{n\alpha}{T} \right) + \cos 2\gamma I_2 \left( \frac{n\alpha}{T} \right) \right] \right\}, \quad (9)$$

where  $K_0$  is a Hankel function and  $I_0$  and  $I_2$  are modified Bessel functions. In this limit  $\alpha$  reduces to

$$\begin{aligned} \alpha &= \Delta_0 \left\{ \frac{\sqrt{2}}{2\pi\sqrt{\pi}} \left[ 3\pi \left( \frac{2\pi}{7\xi(3)} \right)^{1/2} \frac{\gamma}{\pi} \right]^{1/3} \frac{H}{H_c(0)} \right\}, \\ &= 0.368\kappa^{-1/3} \Delta_0 H/H_c(0), \end{aligned} \quad (10)$$

where  $H_c(T)$  is the temperature-dependent critical field,  $\Delta_0$  is the ordering parameter at  $T = 0^\circ\text{K}$ , and  $\kappa = 3\pi T_{c0}(mc/e)[2\pi m/7\xi(3)P_0^5]^{1/2}$  is the Ginzburg-Landau parameter.<sup>11</sup>

(2) Higher temperature region where  $(\omega T)^{1/2} \ll \Delta \simeq T$ . - In this region we have

$$\text{Re}Q(\omega, \alpha, \gamma) = \pi^2 \left\langle \tanh \left( \frac{\Delta+x}{2T} \right) \right\rangle; \quad (11)$$

$$\text{Im}Q(\omega, \alpha, \gamma) = \pi \left\{ \frac{\omega}{T} \left\langle \cosh^{-2} \left( \frac{\Delta+x}{2T} \right) \right\rangle \ln \left[ 2 \left( \frac{2\Delta}{\omega} \right)^{1/2} \right] + \frac{\omega}{\Delta} \left[ 1 - \left\langle \tanh \left( \frac{\Delta+x}{2T} \right) \right\rangle \right] - 2 \frac{\omega}{T} \left\langle P \left( \frac{\Delta}{T}, \frac{x}{T} \right) \right\rangle \right\}; \quad (12)$$

and

$$\begin{aligned} \frac{Z(\omega, \alpha, \gamma) - Z(\omega, 0, \gamma)}{R_n} &= \frac{2}{3\pi \tanh(\Delta/2T)} \left( \frac{\omega}{\pi\Delta \tanh(\Delta/2T)} \right)^{1/3} \left\{ \frac{\omega}{T} \left\langle \cosh^{-2} \left( \frac{\Delta+x}{2T} \right) - \cosh^{-2} \left( \frac{\Delta}{2T} \right) \right\rangle \ln \left[ 2 \left( \frac{2\Delta}{\omega} \right)^{1/2} \right] \right. \\ &\quad - \frac{\omega}{\Delta} \left\langle \tanh \left( \frac{\Delta+x}{2T} \right) - \tanh \left( \frac{\Delta}{2T} \right) \right\rangle - 2 \frac{\omega}{T} \left\langle P \left( \frac{\Delta}{T}, \frac{x}{T} \right) - P \left( \frac{\Delta}{T}, 0 \right) \right\rangle \\ &\quad \left. - i\pi \left\langle \tanh \left( \frac{\Delta+x}{2T} \right) - \tanh \left( \frac{\Delta}{2T} \right) \right\rangle \right\}, \end{aligned} \quad (13)$$

where

$$\langle f(x) \rangle = \frac{1}{\pi} \int_0^{2\pi} d\varphi \sin^2 \varphi f(\alpha \sin(\varphi - \gamma)), \quad (14)$$

and

$$P(\xi, \eta) = \int_0^{\infty} \frac{d\epsilon}{\epsilon^2 - 1} \frac{\cosh(\epsilon\xi + \eta) - \cosh(\xi + \eta)}{[\cosh(\epsilon\xi + \eta) + 1][\cosh(\xi + \eta) + 1]}. \quad (15)$$

(3) In the vicinity of the critical temperature where  $(\omega T)^{1/2} \gg \Delta$ . - Expanding Eqs. (11) and (12) in powers of  $\Delta/T$ , we have

$$\text{Re}Q(\omega, \alpha, \gamma) = \frac{\pi^2 \Delta}{2T} \left\{ 1 - \frac{1}{3} \left( \frac{\Delta}{2T} \right)^2 - \left( \frac{\alpha}{2T} \right)^2 \frac{2 + \cos 2\gamma}{4} \right\}; \quad (16)$$

$$\begin{aligned} \text{Im}Q(\omega, \alpha, \gamma) &= \pi \left\{ \frac{\omega}{T} \left\{ 1 - \left( \frac{\Delta}{2T} \right)^2 - \left( \frac{\alpha}{2T} \right)^2 \frac{2 + \cos 2\gamma}{4} \right\} \ln \left[ 2 \left( \frac{2\Delta}{\omega} \right)^{1/2} \right] \right. \\ &\quad \left. + \frac{\omega}{\Delta} \left\{ 1 - \left( \frac{\Delta}{2T} \right) \left[ 1 - \frac{1}{3} \left( \frac{\alpha}{2T} \right)^2 - \left( \frac{\alpha}{2T} \right)^2 \frac{2 + \cos 2\gamma}{4} \right] \right\} \right\}; \end{aligned} \quad (17)$$

and

$$\frac{Z(\omega, \alpha, \gamma) - Z(\omega, 0, \gamma)}{R_n} = -\frac{\pi}{6} \left( \frac{\pi\Delta^2}{2\omega T} + i \right)^{-4/3} \left\{ \frac{\omega}{T} \left[ \ln 2 \left( \frac{2\Delta}{\omega} \right)^{1/2} - \frac{1}{2} \right] + i \frac{\pi\Delta}{2T} \right\} (2 + \cos 2\gamma) \left( \frac{\alpha}{2T} \right)^2. \quad (18)$$

Equation (2) is rewritten as

$$\alpha = \Delta \frac{\sqrt{3}}{(2\pi)^2} \left[ \frac{\gamma(2\pi)^{1/2} \eta \zeta(3) \Delta}{\kappa T} \right]^{1/3} \frac{H}{H_c(T)},$$

$$= 0.32 \Delta \kappa^{-1/3} (1 - T/T_{c0})^{1/6} H/H_c(T). \quad (19)$$

We would like to make the following remarks:

(I) The shift of the surface reactance with magnetic field is positive at all temperatures, which follows from the fact that  $\text{Re}Q$  decreases as the magnetic field increases independently of temperature.

In the field close to the critical field, the  $\gamma$  dependence of the shift of the reactance is  $(1 + \cos 2\gamma)$  at low temperatures ( $\Delta \gg T$ ) and  $(2 + \cos 2\gamma)$  at higher temperatures ( $\Delta \ll T$ ).

(II) The shift of the surface resistance with magnetic field is positive at lower temperatures ( $\Delta \gg T$ ), becomes negative at higher temperatures [ $(\omega T)^{1/2} < \Delta \lesssim T$ ], and becomes positive again in the immediate vicinity of the critical

field [ $\Delta \approx (\omega T)^{1/2}$ ]. In the first two temperature regions [ $\Delta > (\omega T)^{1/2}$ ], where the inequality  $\text{Re}Q > \text{Im}Q$  holds, the above field dependence of the resistance comes from the fact that  $\text{Im}Q$  increases at low temperature ( $\Delta \gg T$ ), while it decreases at higher temperature ( $\Delta \lesssim T$ ), as the magnetic field increases.

In the vicinity of the transition temperature the direction of the shift of the resistance with magnetic field changes due to the change of the sign of the real part of  $[(\pi\Delta/2T) + i]^{-4/3}$  in Eq. (18).

The  $\gamma$  dependence of the shift of the resistance in a field close to the critical field is similar to that of the reactance stated above.

(III) From both Eqs. (10) and (19) we expect the largest change in the impedance for superconductors with the smallest Ginzburg-Landau parameter  $\kappa$ .

For superconductors having  $\kappa$  smaller than 0.05, and in a field close to the critical field where  $\Delta < \alpha$ , the gap in the excitation spectrum vanishes in the surface region of the specimen. In this case Eq. (9) no longer holds and we obtain

$$\frac{Z(\omega, \alpha, \gamma) - Z(\omega, 0, \gamma)}{R_n} = \frac{8}{3\pi} \left( \frac{\omega}{\pi\Delta} \right)^{1/3} (1 + \cos 2\gamma) \left[ \frac{|\alpha - \Delta|}{2(2\alpha\omega)^{1/2}} + \frac{i}{2} \arccos \frac{\Delta}{\alpha} \right], \quad \text{for } \Delta \gg T. \quad (20)$$

(IV) In the above treatment we assume that the vector potential  $A$  is uniform close to the surface. Actually  $A$  decreases as  $A(z) = A(0)e^{-z/\delta}$  where  $z$  is the distance from the surface of the specimen. This effect can be included in the present theory as follows: Assuming that  $Q(\omega, \alpha, \gamma)$  may be expanded in powers of  $\alpha$  as

$$Q(\omega, \alpha, \gamma) = Q(\omega) + \sum_{n=1}^{\infty} \alpha^{2n} Q_n(\omega, \gamma), \quad (21)$$

we find that the correct expansion including the effect of spatial variation of  $A$  is given as

$$Q'(\omega, \alpha, \gamma) = Q(\omega) + \sum_{n=1}^{\infty} \frac{2n+1}{(2n+1)^3 - 1} \alpha^{2n} Q_n(\omega, \gamma). \quad (22)$$

Qualitatively we expect that  $Q'$  does not differ much from  $Q$ .

(V) Concerning the temperature dependence of the shift of the resistance with magnetic field, the above results are in qualitatively good agreement with the recent experiments by Dresselhaus, Douglass, and Kyhl<sup>4</sup> and by Lewis.<sup>5</sup> On

the other hand, the agreement appears poor concerning the behavior of the reactance with field. According to the present theory, the shift of the reactance should be positive at all temperatures, while experimentally<sup>4</sup> it seems to change sign in the vicinity of the critical temperature. A possible origin of this discrepancy is that the approximation  $\omega/\Delta \ll 1$  fails and the above expressions [i.e., Eq. (18)] are not applicable in this temperature region.

In order to make a quantitative comparison with the present theory, a more detailed calculation is required for various values of  $\alpha$  and  $T$ , which will be carried out in a future communication.

In conclusion, the author would like to thank Professor Douglass for suggesting this problem to him and for stimulating discussions. He is also grateful to Professor Y. Nambu and Professor J. C. Phillips for their valuable comments on the results obtained.

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## STRONG-COUPLING SUPERCONDUCTOR AT NONZERO TEMPERATURE

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The strong-coupling superconductors Pb and Hg have long been considered anomalous in the deviation of their superconducting properties from the law of corresponding states and from the predictions of the simplest form of the BCS theory.<sup>1</sup> Recent calculations of the effective tunneling density of states for Pb are in good agreement with the experimentally measured tunneling anomaly near zero temperature.<sup>2-4</sup> In these calculations, the effective electron-electron interaction was represented by a retarded dressed-phonon exchange and a static Coulomb pseudopotential.

In this Letter we extend these calculations to nonzero temperatures and (1) evaluate the effective electron-phonon coupling constant for Pb using an approximate form of the matrix element suggested by Harrison<sup>5</sup>; (2) determine the frequency dependence of the gap function at nonzero temperature; and (3) discuss an anomaly in the effective tunneling den-

sity of states arising from recombination processes.

The electron-electron interaction for energy transfer  $q_0$  and momentum transfer  $q$  can be written in the form

$$V(q, q_0) = \frac{4\pi e^2}{q^2 \epsilon(q, q_0)} + \sum_{\lambda} |v_{q\lambda}|^2 \frac{1}{q_0^2 - \omega_{q\lambda}^2 + i\delta}, \quad (1)$$

where  $\epsilon(q, q_0)$  is the electron dielectric constant,  $\omega_{q\lambda}$  is the dressed-phonon frequency of momentum  $q$  and polarization  $\lambda$ , and  $v_{q\lambda}$  is the effective electron-phonon coupling matrix element. When  $q$  is outside the first Brillouin zone,  $\omega_{q\lambda}$  is the frequency corresponding to the appropriate reduced wave vector  $q - K$ , where  $K$  is a reciprocal lattice vector. Using (1) with  $q_0 = 0$  in  $\epsilon$ , a spherical band approximation, and keeping only the lowest order vertex parts, the integral equation for the energy-gap function  $\Delta(\omega)$  at nonzero temperature  $T = 1/k\beta$  is<sup>6</sup>

$$\Delta(\omega) = \frac{1}{Z(\omega)} \int_0^{\omega_c} d\omega' \text{Re} \left\{ \frac{\Delta(\omega')}{[(\omega'^2 - \Delta^2(\omega'))^{1/2}]^{1/2}} \left[ f(-\omega')K_+(\omega, \omega') - f(\omega')K_+(\omega, -\omega') - U_c \tanh\left(\frac{1}{2}\beta\omega'\right) \right] \right. \\ \left. + \frac{i\pi}{Z(\omega)} \int_0^{\omega_c} d\omega' \left\{ \frac{\Delta(\omega + \omega')}{[(\omega + \omega')^2 - \Delta^2(\omega + \omega')]^{1/2}} + \frac{\Delta(\omega - \omega')}{[(\omega - \omega')^2 - \Delta^2(\omega - \omega')]^{1/2}} \right\} \frac{\sum_{\lambda} a_{\lambda}^2(\omega') F_{\lambda}(\omega')}{e^{\beta\omega'} - 1} \right\}; \quad (2)$$