Transverse electromagnetic response in superconductors by acoustic techniques. I. Anomalies in the electromagnetic response in niobium

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The work reported here had as its purpose the study of the transverse *electromagnetic* response as a function of $\vec{q}$ in high-purity niobium by observing the attenuation of transverse acoustic waves near T_c in the high-ql regime. The associated contribution to the acoustic attenuation, the electromagnetic attenuation α_E, is expected to suffer a rapid decrease below T_c when $ql > 1$, reflecting the rise of the conductivity in the superconducting state; the remaining electronic contributions to the attenuation, comprising the so-called residual attenuation α_R, are expected to satisfy the more gradual temperature dependence predicted by Bardeen, Cooper, and Schrieffer for so-called case I absorption. Despite the fact that the measurements were made in single-crystal niobium having resistivity ratio of 5000 at frequencies to 450 MHz, corresponding to $ql \approx 20$ at T_c, the "rapid-fall" region was not in evidence in Nb either for $\vec{q}\,||\,[100]$ or for $\vec{q}\,||\,[110]$ and $\epsilon\,||\,[1\bar{1}0]$. The absence for given $\vec{q}$ and $\vec{\epsilon}$ of the so-called rapid-fall regime in niobium, which we first reported in 1971, represents the first known example of such behavior. The present paper, addressed primarily to the inevitable skepticism which ensued, confines itself to a detailed documentation of the evidence for absence of rapid fall in niobium. It is demonstrated that on the basis of present theory the absence of rapid fall in Nb cannot be attributed to high-frequency breakdown of electromagnetic screening.

I. INTRODUCTION

When transverse ultrasonic waves having propagation vector $\vec{q}$ interact with the conduction electrons of a metal under the condition $ql > 1$, where l is the electron mean free path, one of the major sources of coupling between the electrons and transverse phonons is the transverse electromagnetic interaction, associated with absorption coefficient α_E, the electromagnetic attenuation, and characterized by an essentially single value of $\vec{q}$ throughout the bulk of the metal. Due primarily to Meissner screening and the associated rapid increase of the imaginary part of the conductivity with reduction of T below T_c, the superconducting transition temperature, this interaction becomes ineffective close to T_c. The effect is that the observed acoustic absorption versus T decreases rather abruptly, as temperature is reduced below T_c. This corresponds to the so-called "rapid fall" of attenuation. In addition, one may distinguish in the residual electronic attenuation α_R two essentially separable remaining interactions associated not with the aforementioned electromagnetic coupling of electrons and ultrasonic phonons but with shear deformation[1] of the Fermi surface and the so-called collision-drag[2] or relative-velocity effect,[1] interactions which observe the less rapidly varying temperature dependence predicted by Bardeen, Cooper, and Schrieffer[3] (BCS) for case-I absorption: $\alpha_{RS}/\alpha_{RN} = 2f(\Delta(T))$. Here α_{RS}/α_{RN} is the ratio of superconducting-to-normal-state residual attenuation, and $f(\Delta(T))$ is the Fermi function of the BCS energy gap $\Delta(T)$. It is emphasized that since $\hbar\omega \ll \Delta(T)$ for the acoustic phonons in the present work, except within $\sim(10^{-4} - 10^{-5})$ K of T_c, the absorption is attributed only to the excited quasiparticles.

The peculiar advantage over ordinary microwave methods in studying the electromagnetic interaction using transverse acoustic waves is that, owing in part to the relatively low acoustic wave velocities ($\sim 10^{-5}c$), propagating-wave conditions are readily met, so that the observed absorption is associable with a single Fourier component having propagation vector $\vec{q}$. This method has been used by Fossheim[4] for determining in indium the so-called London penetration depth λ_L, the local penetration depth for $q\xi < 1$; here ξ is the coherence length. In a following paper (hereafter, II) Page and Leibowitz[5] report a study of the electromagnetic response in superconducting tin in which the q dependence of the electromagnetic response was determined. Since values of $q\xi$ ranging from $q\xi < 1$ to $q\xi > 1$ were attained, it was possible to study the nonlocal as well as the local response; in this way, for example, not only the London penetration depth but also the nonlocal or Pippard penetration depth $\lambda_P(q)$ was determined directly over a range of $q\xi$.

It is clearly of great interest to attempt to extend those methods to the case of niobium, a type-II superconductor having the highest known transition temperature of any element. We examine in the present paper the unexpected observation of the absence of acoustic evidence for transverse electromagnetic response in niobium for $\vec{q}\parallel[100]$, even for ql values of approximately 20. Similar results were observed in a different sample for $\vec{q}\parallel[110]$, $\vec{\epsilon}\parallel[1\bar{1}0]$, and $ql \sim 4$ at T_c. No evidence is found for the characteristically abrupt decrease of attenuation at T_c. We first reported the absence, for the given wave orientations, of the rapid-fall regime in Nb in 1971[6]; further observations on this anomaly in niobium were reported in 1972.[7] It was suggested to the authors that this behavior in niobium may be attributed to breakdown of electromagnetic screening due to high frequency (cf. Ref. 1). We show that on the basis of present theory this mechanism is not responsible for the observations. The conclusion, based on the observations near T_c as well as on other evidence which we shall discuss, is that the transverse electromagnetic response is ineffective for $\vec{q}\parallel[100]$ in niobium, even for $ql \approx 20$. Present evidence suggests that the same conclusion also applies for the case $\vec{q}\parallel[110]$, $\vec{\epsilon}\parallel[1\bar{1}0]$. To our knowledge this observation of absence of rapid fall in the $ql \gg 1$ regime in niobium[6,7] represents the first such instance in the literature since the rapid-fall phenomenon was first observed in 1958 in indium.[8] The present paper is addressed primarily to the inevitable skepticism which ensued, particularly with regard to electromagnetic breakdown as a possible cause of the anomaly, and therefore confines itself to a detailed documentation of the evidence for absence of rapid fall in niobium. A similar result has now been asserted for given orientation in zinc and cadmium, and in molybdenum.[9] It remains elsewhere to consider whether the shear-wave anomalies in all of these cases have a common explanation.

II. THEORY

It has been demonstrated[10,11] that when the given metal is a superconductor the several transverse phonon-electron interactions cited above may readily be separated experimentally as well as analytically, the former by determining the transverse ultrasonic absorption near T_c as a function of ql. Thus, the normalized electromagnetic attenuation contribution $\alpha_E/\alpha_N(T_c)$ is determined by measuring the rapid-fall fraction of the total absorption, that fraction of the total (or normal-state) attenuation α_N at T_c which disappears very rapidly as temperature is reduced from T_c; $\alpha_R/\alpha_N(T_c)$ is the normalized residual attenuation at T_c.[12] In Fig. 1 these regions are shown schematically. The rapid-fall region is shown as a discontinuity since its magnitude is halved in a very restricted temperature range[12] $\Delta T \equiv T_c - T$: $\Delta T/T_c \sim 0.001$. The temperature dependence of the residual attenuation has been shown theoretically[13] to satisfy BCS case-I absorption when the electron mean free path l is *impurity limited*. That is, $\alpha_{RS}(T)/\alpha_{RN}(T) = 2f(\Delta(T))$, where $f(\Delta(T))$ is the Fermi function for the temperature-dependent BCS energy gap $\Delta(T)$. While l is phonon limited in Nb at T_c, leading to deviations from the BCS curve, we have shown[7] that, in the $ql \gtrsim 1$ regime, the deviations systematically disappear with increasing ql. This is readily understood: When $ql \gg 1$ both α_S and α_N become independent of l, so that the phonon limitation of l becomes irrelevant in the $ql \gg 1$ region.

The residual attenuation $\alpha_R(T_c)$ is the sum of the collision-drag contribution α_C and α_D, attributable to shear deformation of the Fermi surface. In Ref. 10 it was demonstrated that these may be separated in a superconductor by determining the high-ql limiting value of $\alpha_R/\alpha_N(T_c)$, which is shown to reduce asymptotically, with increase of ql, to a constant representing α_D/α_N in the high-ql limit. These effects are shown schematically in Fig. 2. While we are particularly concerned with α_E in the present paper, some of the evidence for apparent absence of an α_E contribution requires that attention be given as well to certain properties of α_C and α_D which have significance for the ql dependence of α_E/α_N.

Pippard finds that

$$\alpha_R = \frac{\hbar q}{4\pi^3 M v_t} \int \frac{D^2 ql\, dS}{1+(ql)^2\cos^2\varphi} , \qquad (1)$$

where

$$D = K_{xy} + k_y \cos\varphi .$$

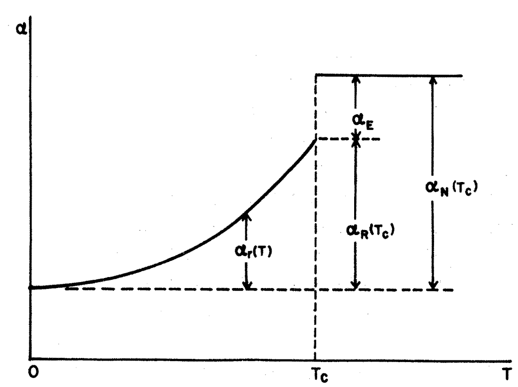

FIG. 1. Schematic representation of transverse acoustic-wave attenuation vs temperature for the case $ql > 1$.

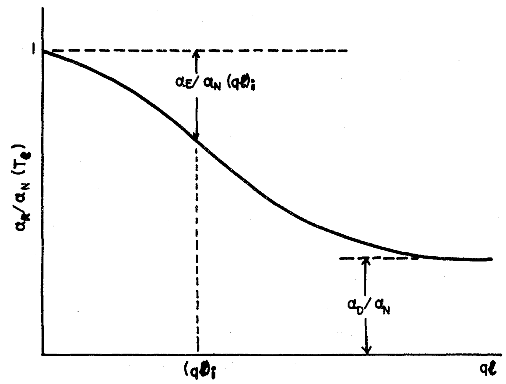

FIG. 2. Schematic representation of the residual attenuation (after Leibowitz, Ref. 10).

Here M is the density of the metal, v_t is the transverse acoustic wave velocity, and the z and x Cartesian axes represent the wave propagation and polarization directions, respectively. φ is the angle determined by electron propagation vector $\vec{k}$ and the x axis, and K_{xy} is a Fermi-surface deformation parameter defined by $dk_n = K_{xy}\omega_{xy}$; dk_n is the shift of the Fermi surface along a surface normal and ω_{xy} is the strain. In Ref. 10, $\alpha_D(T_c)$ and $\alpha_C(T_c)$ are defined as

$$\alpha_D(T_c) = \frac{\hbar q}{4\pi^3 M v_t} \int \frac{(ql) K_{xy}(K_{xy} + 2k_y \cos\varphi)\, dS}{1 + (ql)^2 \cos^2\varphi} \tag{2}$$

and

$$\alpha_C(T_c) = \frac{\hbar q}{4\pi^3 M v_t} \int \frac{(ql)\, k_y^2 \cos^2\varphi \, dS}{1 + (ql)^2 \cos^2\varphi} , \tag{3}$$

so that $\alpha_R = \alpha_D + \alpha_C$, the term α_D containing the deformation parameter as a factor. Thus we find $\alpha_R/\alpha_N \approx 1$ for $ql \lesssim 1$, and reducing with increase of frequency to a frequency-independent ratio given by α_D/α_N corresponding to the $ql \gg 1$ limit (see Fig. 2):

$$\frac{\alpha_D}{\alpha_N} \approx \oint RK_{xy}^2 \, d\psi \bigg/ \left(\oint RK_{xy}^2 \, d\psi + \frac{1}{\pi^2} \, \frac{\int D \tan\varphi \cos\psi \, dS}{\int R \cos^2\psi \, d\psi} \right) . \tag{4}$$

Here R is the product of the principal radii of curvature corresponding to angle ψ, which is the polar angle in a given effective zone. Correspondingly, the normalized electromagnetic contribution $\alpha_E/\alpha_N \approx 0$ for $ql \lesssim 1$ and increases monotonically with increasing ql, reflecting the decreasing importance of the interaction α_C/α_N. Note that the second term in denominator of Eq. (4) represents the α_E contribution for $ql \gg 1$.

Thus, as shown in the figure, the normalized electromagnetic contribution α_E/α_N becomes significant for $ql \gtrsim 1$. We have here, then, the basis for the $ql > 1$ requirement in order that a rapid-fall region be observable in the transverse-wave attenuation at T_c; with the onset of nonlocality with respect to electron trajectories the electromagnetic interaction becomes significant. For the free-electron case the functional dependence of ql may be stated explicitly. There is in that case no shear-deformation contribution[1] and therefore $\alpha_D = 0$. Further, it is demonstrated in Ref. 10 that α_R/α_N reduces from the relation determined by Eqs. (1) and (4) to the free-electron result[12] $(\alpha_C/\alpha_N)_{\rm FE}$:

$$\left(\frac{\alpha_C}{\alpha_N}\right)_{\rm FE} = g(ql) = \frac{3}{2(ql)^3} \left\{ [(ql)^2 + 1] \tan^{-1} ql - ql \right\} , \tag{5}$$

where $g(ql) \sim 1$ for $ql \lesssim 1$ and decreases monotonically with increase of ql. Accordingly, the electromagnetic contribution becomes significant with increase of ql, the free-electron ql dependence being given simply by $\alpha_E/\alpha_N = 1 - g(ql)$.

It will prove useful to summarize here some of the explicit properties of α_E/α_N. Assuming a wavelike dependence $e^{i(kx - \omega t)}$ for the transverse acoustic wave the electromagnetic attenuation takes the form[4,14]

$$\alpha_E = \frac{\mathrm{Re}(\sigma)}{M v_t} \left(\frac{4\pi n e \omega}{c^2 q^2} \right)^2 \left(\frac{1}{1 + [(4\pi\omega/q^2c^2)|\sigma|]^2 + 2[(4\pi\omega/q^2c^2)]\sigma_2} \right) , \tag{6}$$

where the complex conductivity $\sigma = \sigma_1 + i\sigma_2$ and n is the electron number density. There is obtained[4] for the superconducting-to-normal attenuation ratio for the electromagnetic interaction α_{ES}/α_{EN} the relation

$$\frac{\alpha_{ES}}{\alpha_{EN}} = \frac{(\sigma_{1S}/\sigma_{1N})(1 + \gamma^2)}{(\sigma_{2S}/\sigma_{1N} + \gamma)^2 + (\sigma_{1S}/\sigma_{1N})^2} . \tag{7}$$

Here the subscripts S and N refer to superconducting and normal state, respectively, and γ is defined by $\gamma = q^2c^2/(4\pi\omega\sigma_{1N})$. The conductivity ratio σ_{1S}/σ_{1N} has been calculated by Cullen and Ferrell[14]:

$$\frac{\sigma_{1S}}{\sigma_{1N}} = 1 + \frac{1}{2} \frac{\Delta(T)}{k_B T} \ln\left(\frac{8\Delta(T)}{e\hbar\omega} \right) - \frac{7\zeta(3)}{\pi^2} \left(\frac{\Delta(T)}{k_B T} \right)^2 , \tag{8}$$

where e is here the base of the natural logarithms and $\zeta(3)$, the Riemann ζ function of 3, has the value 1.202. For the ratio σ_{2S}/σ_{1N} in the free-electron case Fossheim[4] obtains

$$\frac{\sigma_{2S}}{\sigma_{1N}}=\frac{\hbar c^2}{v_t\, e^2\lambda_L(0)\,k_F^2}\,\frac{\xi}{\xi_0}\,\frac{F(q\xi)[1-(T/T_c)^4]}{H(ql)} . \quad (9)$$

Here the coherence lengths ξ and ξ_0 are related by $1/\xi=1/\xi_0+1/l$; $F(q\xi)$ is identical in form to $g(ql)$, defined by Eq. (5), the former expressing nonlocality with respect to coherence length ξ and the latter with respect to electron mean free path l. For convenience in the later data analysis we have introduced in Eq. (9) the function $H(ql) = (4/3\pi)(ql)g$. Thus $\sigma_{1N}=(\sigma_{1N})_{EA}H(ql)$, where the extreme anomalous conductivity $(\sigma_{1N})_{EA}=e^2k_F^2/(4\pi\hbar\omega)$.

The analysis summarized above has proved successful in interpreting the electromagnetic absorption in the local regime $q\xi<1$ in indium.[4] Further, as is shown in II,[5] it has been successfully applied, in pure tin, to the nonlocal regime $q\xi\gtrsim 1$, and in the normal state at frequencies in the electromagnetic breakdown region, where $\gamma=\delta/(\frac{1}{4}\lambda)\gtrsim 1$; here δ is the effective skin depth and λ the acoustic-phonon wavelength. Accordingly, it is of particular interest to determine whether the absence of rapid fall asserted in the present paper for niobium is in accord with the foregoing theory. Before we proceed to a review of the experimental data and analysis, we shall first briefly describe the experimental apparatus and procedures.

III. EXPERIMENT

The experimental procedures were similar to those used in our recent study[15] of magnetoacoustic oscillations in Nb. Certain additional constraints were imposed in the present instance, due primarily to the need for precise temperature measurements and control over a temperature range $\Delta T=T_c-T\sim 100$ mK and the associated requirement that ambient magnetic field be cancelled to ~0.001 G. For the latter, the cancellation of Earth's magnetic field in the $\alpha(\Delta T)$ experiments, three mutually orthogonal Helmholtz coils were used. The degree of field cancellation was determined component by component, at the actual sample site, by means of a flux-gate magnetometer.

The sample-holding block, supported by the experimental probe which extended into the cryostat, was machined from a solid piece of high-purity oxygen-free copper. A grid was scribed on the base of the block parallel to the top face of the probe to facilitate alignment of sample and transducer with the magnetic field [for the $\alpha(H)$ experiments]. The thermometry wires were inserted through a small metal tube running the length of the block. The rf leads to miniature coaxial cable (number 38 copper-wire center conductor with aluminized Mylar outer conductor) and the tuning shafts of thin stainless-steel tubing were left uncased. Plastic blocks, carefully machined from Delrin, held the capacitors for tuning circuits[15] and contained spring-loaded brass contacts for the transducer electrodes.

The tuning circuits in the sample holder (between coaxial cable and transducer) enhanced sensitivity and decreased crosstalk. Signal was observed to be 20 dB larger with the circuits than without at certain frequencies. Circuit components were changed in a systematic trial-and-error process until maximum signal and minimum crosstalk were observed for the center frequency of interest, approximately 315 MHz. When the circuits were properly tuned and impedance matched, dynamic impedance measurements indicated that the impedance at the center frequency was 50 Ω. The circuit configuration had certain thermal advantages to be discussed later.

Temperature was controlled by mounting a heater in the sample block. This block was surrounded by a vacuum can and the entire assembly then immersed in liquid helium. The helium bath and heater provided the heat sink and source. Temperature equilibrium for each current setting was reached within seconds. By changing the current in the heater a range of equilibrium temperatures could be achieved in the block. The heater was made from 100 ft of Evan Ohm wire, 49.9 Ω per ft, noninductively wound and thermally anchored to the block with G.E. lacquer. The tuning inductor served the additional purpose of providing a heat path from the sample to the block. The sample, the block, and germanium thermometer were thus closely coupled thermally.

Two mechanisms for thermally linking the copper block to the low-temperature reservoir, gaseous convection and thermal conduction, were carefully controlled to permit the required measurements to be taken. Thermal pinning through conduction was adjusted by trial and error. Once an optimum situation was found it was not changed through the course of the experiment. Gaseous pinning was standardized as follows. When the probe was assembled a rough-pump vacuum was drawn and a check made for leaks. The probe was then filled to 1 atm with helium gas and sealed. This configuration was used when taking total attenuation and magnetoacoustic measurements. For the very precise measurements near T_c, most of this gas was removed by a diffusion pump. This eliminated temperature fluctuations due to helium gas oscillations in the probe stem. Enough gas was left in the probe to provide pinning. With a little practice, very slow temperature sweeps could be made. Temperature-vs-attenuation measurements were made in both directions, heating up and cooling down to be sure temperature equilibrium of sample and thermometer was maintained. When more gas was pumped out of the probe, it was easier to go above 14 K, to 20 K. In this manner normal-state data above T_c were obtained. Helium gas was introduced into the probe when it was desired to return to helium temperature.

IV. DATA AND ANALYSIS

The canonical rapid-fall phenomenon, its absence for $ql<1$, and its increasing relative magnitude as ql increases from $ql\approx 1$ have been well documented. (For a review of the earlier experimental results, see Ref. 10.) In previous experimental studies of niobium (cf. Ref. 16) the absence of rapid fall was consistent with existing theory merely on the ground that $ql<1$ in the impure specimens then available. In the present work specimens having resistivity ratios of approximately 5000 were used which, for $\vec{q}\parallel[100]$ and transverse-wave frequencies to 435 MHz, permitted attainment of $ql\approx 20$. The estimated maximum ql value at T_c in the (110) specimen for $\vec{q}\parallel[110]$ and $\vec{\epsilon}\parallel[110]$ was approximately 4; the increased thickness of the (110) sample [0.78 mm as compared with 0.58 mm for the (100)], necessitated by the somewhat greater acoustic wave velocity and the associated increase in measured attenuation, resulted in a maximum useful transverse wave frequency of 375 MHz. In both instances, with $ql>1$, the theory reviewed in Sec. II would have predicted appearance of well-defined rapid-fall regions in the temperature dependence of attenuation near T_c, reflecting the turnoff of a measurable electromagnetic contribution to the total transverse phonon-electron coupling. Figures 3 and 4 show no evidence for the rapid-fall region; here the solid curves represent the BCS temperature dependence[3] for the case-I absorption, $\alpha_S/\alpha_N=2f[\Delta(T)]$. Note that the temperature range near T_c is examined in order to discriminate sensitively between the electromagnetic contribution and the BCS temperature dependence. Earth's magnetic field is cancelled to within ~0.001 G in order that distortion of $\alpha(T)$ near T_c be averted. The data in Fig. 3 range from $ql\approx 9$ (195 MHz) to $ql\approx 20$ (435 MHz), while those of Fig. 4 have an estimated ql range of approximately 2–3.5. Note the "approach from below" to the BCS curve as ql is increased. The effect, which we discuss below and have already reported,[7] has a ql dependence opposite to that associated with the electromagnetic interaction and is unrelated to it. Note in Fig. 3 the close agreement with BCS predictions when $ql\gg 1$.

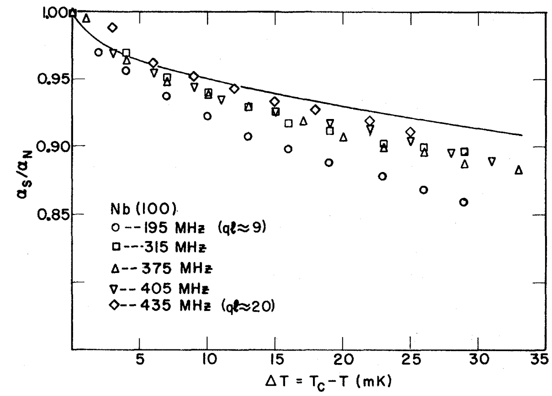

FIG. 3. Temperature dependence of the superconducting-to-normal attenuation ratio near T_c for $\vec{q}\parallel[100]$. Note the absence of the rapid-fall regime and the return to BCS temperature dependence (solid line) with increasing ql.

A. Determination of ql

Although the purity of the samples was high ($\rho/\rho_0\approx 5000$), direct determination of ql was essential since this value was intimately connected with the predicted observability of rapid fall. This was accomplished by two methods, computer fitting the Pippard[17] equation for $\alpha_N(ql)$ and observations on the transverse ($\vec{H}\perp\vec{q}$) magnetoacoustic effect. While Pippard gives explicit ql dependence of α_N over all ql only in the free-electron case, the sensitivity of the ql dependence to free-electron assumptions is significant only

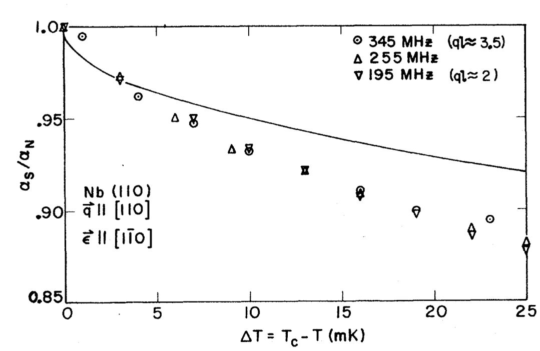

FIG. 4. Temperature dependence of the superconducting-to-normal attenuation ratio near T_c for $\vec{q}\parallel[110]$, $\vec{\epsilon}\parallel[1\bar{1}0]$. Note the absence of the rapid-fall regime. The ql range was not sufficient to test return to BCS temperature dependence (solid line).

in the region $ql \approx 1$; in the real Fermi-surface theory[1] α_N is shown to have the same ql dependence for $ql<1$ and for $ql>1$ as in the free-electron model. Of special interest in the present work is the determination of the $ql>1$ regime, and both the free-electron and real-metals theories predict $\alpha_N \sim q$ here. To supplement and check the ql determinations obtained using the Pippard criterion, the magnetoacoustic technique was used to provide a lower-limit ql estimate[18]: It should be noted that, for α_E and α_D at least, the electron-phonon coupling is confined essentially to the so-called "effective zones" of the Fermi surface, when $ql>1$; and under the assumption $v_t \ll v_F$ the effective zones are the loci satisfying $\vec{v}_F$ approximately normal to $\vec{q}$. Now the effective electrons for the case of zero applied field H are not, in general, the same as those for the magnetoacoustic measurements: In the latter case the observed attenuation is, of course, determined by the electrons lying on the cyclotron orbits, which in the transverse magnetoacoustic effect ($\vec{H} \perp \vec{q}$) are in a plane parallel to $\vec{q}$. In fact, the ql values determined by both methods turned out to be consistent with each other.[15]

For the first method, measurements were made of the total electronic attenuation at fixed temperature (T_c) as a function of frequency (see Fig. 5). The values of attenuation at T_c for these frequencies were then computer fitted to the Pippard equation for transverse waves $\alpha_N = K(1-g)/g$, where $g(ql)$ is given by Eq. (5). A refined grid-search technique[19] was used. This

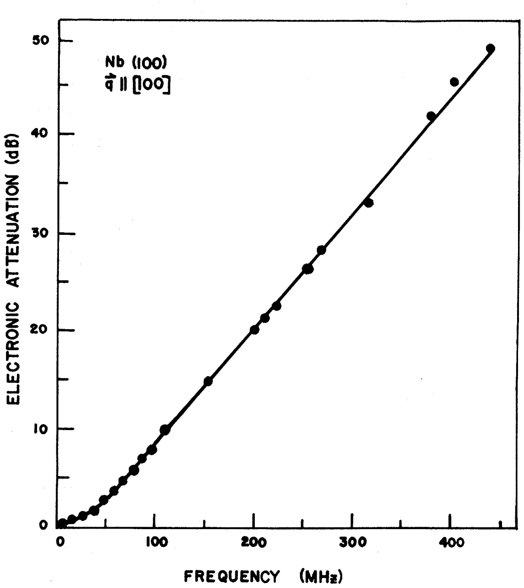

FIG. 5. Frequency dependence of electronic attenuation for $\vec{q} \parallel [100]$. Note the linear frequency dependence for $f>50$ MHz.

iterative process gave $ql \approx 4.4$ at T_c and a frequency of 100 MHz for the (100) sample. Thus at 435 MHz $ql \approx 20$ for T_c. For the (110) sample, ql was about 1 at 100 MHz and T_c. These values were consistent with those obtained from the transverse magnetoacoustic oscillations ($\vec{H} \perp \vec{q}$) which we have observed[15] in the same niobium specimens. It is concluded that measurements were made in the region $ql>1$.

B. Transition width

The possibility of broadening of the superconducting transition due to macroscopic inhomogeneities must be considered since the temperatures of special interest in the present work are in the range of tens of mdeg of T_c. The high resistivity ratio ρ/ρ_0 in the samples does not preclude the presence, for example, of inhomogeneous long-range strains which could conceivably modify $\alpha(\Delta T)$. At the same time, neither does the observation of the transition width in a different kind of measurement, such as that on resistivity $\rho(\Delta T)$, answer the concern satisfactorily, since the transition width observed may depend on the given transport mechanism. Thus, for example, a propagating acoustic wave may readily stimulate dislocation motion when resistivity is determined by an impurity-limited or phonon-limited electron mean free path. Accordingly, it is important to examine the width of the transition which occurs in the present measurements. Figure 6 shows x-y recorder data tracings of attenuation versus ΔT over the first few mdeg. As indicated by the arrows, data sweeps refer both to increasing and decreasing temperature, with sweep rate of about 1 mdeg per min. Note that the transition width lies within the noise and has an estimated upper limit of the order of 10^{-4} K.

Figure 3 and, less sensitively, Fig. 11 demon-

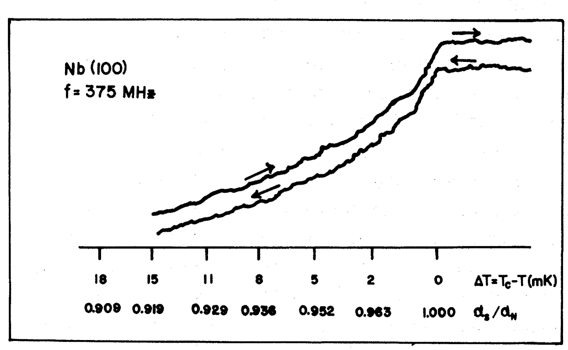

FIG. 6. x-y recorder data traces of attenuation ratio α_S/α_N vs ΔT (mdeg K), both for decreasing and increasing temperature. Ambient magnetic field is reduced to less than 3 mG.

strate that it is not reasonable to assume the attenuation data to be distorted over a temperature range much greater than the region of apparent broadening. Note from Figs. 3 and 11 that the $\alpha(\Delta T)$ curves change slope dramatically with change of ql, in the regime $ql > 1$. While such an effect, over the given restricted temperature range, is not readily attributable to macroscopic inhomogeneities,[20] it is accountable in terms of single-particle electron-phonon mechanisms, as discussed in Sec. IV E and Ref. 7. In point of fact, the data are seen to approach the BCS curve for $ql \gg 1$ as required. This is a result inconsistent with transition broadening on the temperature scale of interest in Fig. 3.

While significant broadening of the transition is not uncommon, especially in heavily cold-worked samples of refractory metals, the samples used in the present work were not only of uncommonly high purity for bulk Nb ($\rho/\rho_0 \approx 5000$), but also had been annealed near melting point ($T > 2000$ K) for 48 h in a vacuum of about 5×10^{-10} Torr. Hence it is no surprise that the observed broadening of the transition was as small as indicated in Fig. 6, a sharpness of transition comparable to that which we have observed in such metals as indium and tin.

C. Amplitude effects

A signal-amplitude-dependent attenuation attributable to dislocations, first reported[20] in Pb and subsequently studied in a number of other metals, can produce erroneous estimates of the electronic contribution to the acoustic attenuation. Also, the dislocation contribution can be temperature dependent even at liquid-helium temperatures, and different in the normal and superconducting states. These effects are attributable to the fact that the normal electrons exert a viscous drag on the dislocation motion excited by the acoustic wave. Such effects have been found to be significant only in the "soft" superconductors such as lead and indium. It has been anticipated that these contributions to the attenuation would prove unimportant in carefully prepared specimens of niobium. This, as we show below, proved to be the case.

An additional potential "amplitude-effect" error in the temperature dependence of the attenuation near T_c must be considered.[21,22] The induction field associated with the acoustic phonon-electron interaction can be big enough at very high acoustic amplitudes to generate local normal-state regions in the bulk of the material. Since these regions contribute attenuation larger than that corresponding to $\alpha_S(T)$, the effect is a potential source of nonlinear error in the estimation of electronic attenuation. While this effect involves the intermediate state and is not ordinarily of concern in a type-II superconductor, it also was sought in the data.

Another possible consequence of the use of high acoustic amplitudes must be considered. Since niobium is a type-II superconductor, it experiences only incomplete Meissner effect above H_{C1}. At sufficiently large wave amplitudes in the sample and at temperatures sufficiently close to T_c the local induction field associated with the acoustic wave could conceivably exceed H_{C1}, with the result that the $\alpha_E(T)$ data would be distorted. It must therefore be asked whether the apparent absence of rapid fall may be attributed to this cause. Again the data on $\alpha(T)$ near T_c taken at different power levels show that no such influence is in evidence. The power level of the input signal was decreased by 20 dB with no effect on $\alpha(T)$. It should be further noted that the signal suffered an additional attenuation of 50 dB in the sample at 430 MHz. The absence of observable α_E therefore would require that for no significant portion of the sample was the local induction field below H_{C1}, even when the signal level was attenuated by the amounts indicated. It should incidentally be noted that the signal levels from the transmitter never exceeded about 100 V measured at the sending transducer (with 20 dB additional attenuation, this is reduced to about 10 V). [Such levels are comparable with those corresponding to the linear region in the amplitude effect described in the last paragraph. Note that those measurements referred to indium, and that $H_{C1}(\text{Nb}) \gg H_C(\text{In})$ at a given reduced temperature t.]

The experimental demonstration that the aforementioned amplitude effects were not a factor in masking the effect of rapid fall is summarized in Fig. 7. Identical results were obtained for $\vec{q}\,\|\,[110]$, $\vec{\epsilon}\,\|\,[1\bar{1}0]$. The reduction in transmitter signal amplitude had no significant effect on the attenuation. We conclude that there were no signi-

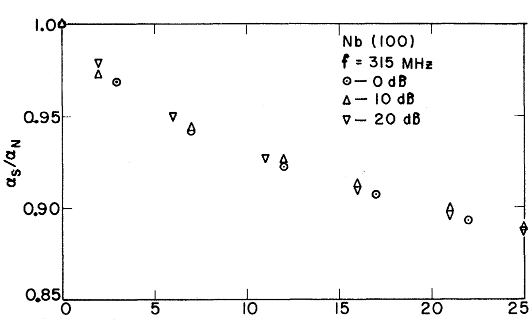

FIG. 7. Superconducting-to-normal-state attenuation ratio near T_c for different power-level inputs showing absence of amplitude dependence in the case Nb(100).

ficant amplitude effects in the niobium specimens at the power levels used and that amplitude effects were not masking rapid fall.

It has been noted that dislocations can produce temperature dependence of the attenuation even at liquid-helium temperatures. The absence of amplitude dependence does not preclude the possibility of dislocation as well as electronic contributions to *temperature dependence* of attenuation. This fact must be recognized in connection with our discussion of apparent absence of rapid fall. Fortunately, however, the rapid-fall phenomenon is largely confined to the order of tens of mdeg of T_c, a range over which the dislocation contribution would not be sensibly changed.[20]

D. Electromagnetic interaction: Frequency dependence

We consider next the possibility that the absence of rapid fall may be attributed to breakdown of electromagnetic screening[17] at the frequencies of these experiments ($f \leq 435$ MHz). When our preliminary results in niobium were first reported[6] and at the International Conference,[7] it was suggested to the present authors that, since the ql values were so high in the niobium experiments, the absence of rapid fall is attributable to breakdown of normal-state electromagnetic screening: The frequency is sufficiently high that the ionic-electronic current quasibalance condition is no longer applicable, and the electrons are no longer coupled to the transverse phonons via the time-varying local induction field. Accordingly, we shall now consider this mechanism, and show that on the basis of the present theory outlined in Sec. II, it is not responsible for the observations. The current imbalance becomes important when the effective skin depth δ becomes comparable with the acoustic wavelength λ, i.e., when $\gamma \approx 1$. From Eq. (6), the normal-state electromagnetic attenuation α_{EN} may be written[4]

$$\alpha = \frac{\sigma_{1N}}{Mv_t}\left(\frac{4\pi en\omega}{c^2q^2}\right)^2\left(\frac{1}{1+(1/\gamma)^2}\right) , \qquad (10)$$

where, it is recalled, $\gamma = \delta/(\frac{1}{4}\lambda) = q^2c^2/4\pi\omega\sigma_{1N}$. For $\gamma \ll 1$, α_{EN} increases with frequency, but for $\gamma \approx 1$ it "turns over." As the frequency increases further, the electromagnetic attenuation continues to decrease. In II Page and Leibowitz[5] demonstrate this screening breakdown for tin in the 1.0-GHz range. While the collapse of the current balance condition is expected commonly to occur for frequency $f \sim 1$ GHz, might it not develop at lower frequency in niobium, and thus account for the apparent absence of α_E in Nb? If electromagnetic breakdown were to occur at relatively low frequencies for the case of Nb, then α_E would be absent in the total electronic attenuation α_N at the frequencies of the present experiments; no rapid fall at T_c would be observed since α_E would no longer have been present in the normal-state attenuation.

The present theory does indeed suggest, as we shall show, that α_E breakdown should develop at relatively low frequency in Nb. However, the evidence in niobium is that such an effect does not occur. That is, the very absence of evidence for the onset and development of electromagnetic breakdown in the present work is further support for the assertion that, in fact, the α_E contribution is apparently absent in niobium even for $ql \approx 20$, when $\vec{q} \parallel [100]$: The observed complete absence of rapid fall in the experimental data would require that electromagnetic breakdown be already essentially complete even at the lowest observed frequencies, which ranged from 30 MHz (corresponding to $ql \approx 1$) to 435 MHz in the $\vec{q} \parallel [100]$ case. From Eq. (10) it is seen that the $\gamma = 1$ condition occurs at $f \approx 300$ MHz in the case of niobium. Recalling that $\gamma = q^2c^2/(4\pi\omega\sigma_{1N})$ and that σ_{1N} is proportional to k_F^2, it is recognized from Eq. (10) that the frequency scaling of the electromagnetic breakdown effect is determined for a given metal by the effective value of k_F. In paper II[5] the electromagnetic breakdown effect was demonstrated in tin. It is noted that the turnover effect in tin occurs at about 1 GHz, and that $\alpha_E(f)$ for tin conforms remarkably well with theory. Figure 8 compares $\alpha_E(f)$ for niobium and tin. For $\vec{q} \parallel [100]$ in tin the effective k_F value was determined to be $k_F = 2.0$ Å^{-1} (see paper II[5]). It was determined that the smallest value of effective k_F, $(k_F)_{\text{eff}}$, which could reasonably be chosen, in light of our magnetoacoustic results in niobium[15] and the augmented-plane-wave calculations of

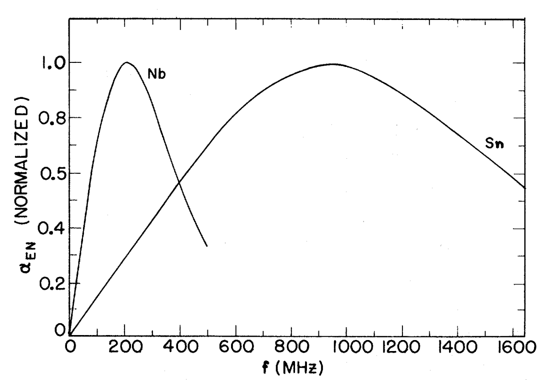

FIG. 8. Theoretical frequency dependence for the normal-state electromagnetic attenuation, showing effect of breakdown of electromagnetic screening for niobium and tin.

Mattheiss,[23] was $(k_F)_{\rm eff} \approx 0.5$ Å^{-1}; $(k_F)^2_{\rm eff} \equiv \int_0^{2\pi} |R|\, d\phi / \int_0^{2\pi} d\phi$, where R is the reciprocal of the Gaussian curvature and the integral is taken about the effective zone. In Fig. 8 it is seen that even for $k_F \approx 0.5$ Å^{-1} the breakdown effect is not significant in Nb below 200 MHz. And at this frequency, corresponding to $ql \approx 7$, rapid fall is expected to be very much in evidence: For $ql \approx 7$, $1 - g \approx 0.7$. Nevertheless, rapid fall is found to be absent for the entire ql range. Further, note that the experimentally determined normal-state attenuation $\alpha_N(f)$ given in Fig. 5 remains essentially linear to the highest frequency examined, 435 MHz, and thus gives no indication of a turnoff of the electromagnetic contribution to the observed attenuation. It is concluded that the absence of rapid fall is not attributable to electromagnetic breakdown and that, in point of fact, the absence of evidence for breakdown suggests absence of the electromagnetic coupling.

E. Electromagnetic interaction: Temperature dependence

Let us next consider the possibility that rapid fall in the electromagnetic interaction, corresponding to α_E, is in fact present in the total normal-state electronic attenuation α_N, but that its turnoff at T_c due to Meissner screening is not readily discernible experimentally. We saw (see Fig. 1) that the α_E and α_R regions are distinguished in practice from the rapid change of slope of $\alpha_S(T)$ near T_c; $\alpha_E(T)$ generally has a much stronger T dependence than does $\alpha_R(T)$, which follows roughly (see below) the temperature dependence given by the BCS relation, $\alpha_S/\alpha_N = 2f(\Delta(T))$. If, however, the temperature dependence of $\alpha_E(T)$ for niobium were to be anomalously slowly varying, it might not be readily discriminated experimentally from that of $\alpha_R(T)$. We therefore examine the theoretically predicted temperature dependence of α_E near T_c in niobium. In examining the temperature dependence of the electromagnetic interaction the behavior of niobium is compared with that of tin, a material which exhibits the rapid-fall behavior and which has been studied in detail (see paper II[5]).

The temperature dependence of Eq. (7) for α_{ES}/α_{EN} is determined by k_F and the conductivity ratios σ_{1S}/σ_{1N} and σ_{2S}/σ_{1N}; k_F is contained both in σ_{2S}/σ_{1N} and γ. Values for the conductivity ratios calculated as a function of temperature for both tin and niobium are shown in Fig. 9. The steeper slope of σ_{2S}/σ_{1N} for niobium relfects the choice of a smaller effective value of k_F for $\vec{q} \parallel [100]$ in niobium[15] than in tin. The effect on α_{ES}/α_{EN} is shown in Fig. 10. Thus, according to the present theory, reduction of temperature T would be expected to cause an even more rapid decrease of

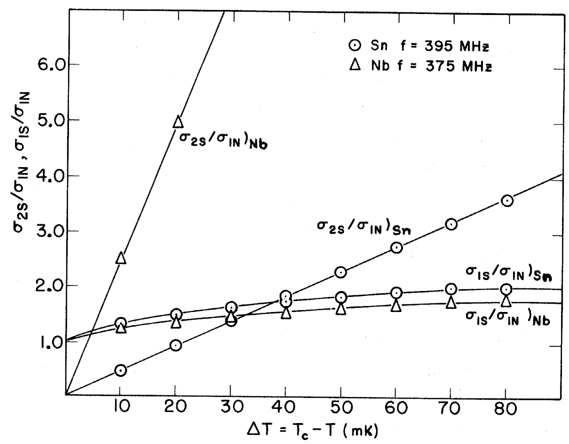

FIG. 9. Temperature dependence of the conductivity ratios σ_{1S}/σ_{1N} and σ_{2S}/σ_{1N} for tin and niobium near T_c. The indicated circles and triangles represent calculated points and not data.

$\alpha_E(T)$ in niobium than in tin. Yet, the canonical rapid-fall phenomenon has been well documented for tin (see paper II[5] and Ref. 10).

Since the theoretical results for Nb shown in Figs. 8 and 10 depend on the choice of k_F, the effect of larger k_F values must be considered. Clearly, an increase of k_F would shift the electromagnetic breakdown (see Fig. 8) to higher frequencies. Thus, our experimental observation of linearity in $\alpha_N(f)$ up to 435 MHz (the highest frequency attained) would not automatically contain the implication of absence of observable α_E. Correspondingly, a larger k_F value would result in a reduction of the sharpness of $\alpha_E(T)$ near T_c, represented in Fig. 10.

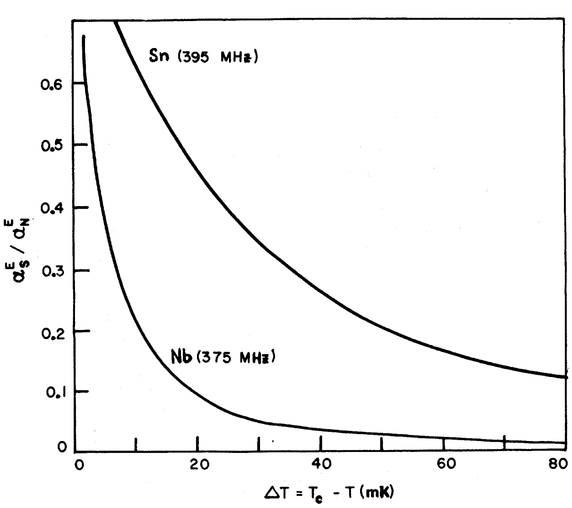

FIG. 10. Temperature dependence near T_c of the superconducting-to-normal-state ratio of the electromagnetic attenuation for Sn and Nb.

However, let us now consider the observed temperature dependence of α_S/α_N. Figure 11 shows that in the $ql \gg 1$ regime $\alpha_S/\alpha_N(T)$ agrees rather well with the BCS prediction. It is well established theoretically (cf. Ref. 13) that α_R/α_N should have the temperature dependence predicted by the BCS theory when $ql \gg 1$. If α_E were in fact contributing to the total electronic attenuation α_N, the observed temperature dependence would force the conclusion that α_E has the same temperature dependence as α_R in the superconducting state. Since α_E and α_R are expected to have qualitatively different temperature dependences,[3,4] a requirement that their temperature dependences be the *same*, in order to explain the above data, would certainly be most artificial.

Note, incidentally, the frequency dependence of $\alpha_S/\alpha_N(T)$ shown in Fig. 11. We have reported this effect elsewhere[7] and have attributed it to the modification of the BCS prediction for $\alpha_S/\alpha_N(T)$ (for longitudinal-wave case and the residual attenuation of shear waves) when the mean free path is phonon limited. That the mean free path in the temperature range examined in Fig. 11 is phonon limited is demonstrated experimentally by the observed temperature dependence (not shown) of α_N, reflecting[17] the temperature dependence of electron mean free path l (note that T_c/Θ_D for Nb is 0.033). The 30-MHz data correspond roughly to $ql \approx 1.4$, so that the data cover the approximate ql range 1.4–20; as ql becomes large relative to 1 both attenuation coefficients α_S and α_N become increasingly mean-free-path independent, with the effect that deviations from BCS predictions disappear. The effect is unrelated to our present concern, the electromagnetic absorption α_E. In point of fact, as we noted earlier, theory requires that α_E/α_N at T_c increase monotonically with ql; in the free-electron case $\alpha_E/\alpha_N(T_c) = 1 - g(ql)$. That is, the relative magnitude of the rapid-fall region should *increase* with frequency. That this is contrary to the behavior demonstrated in Figs. 3 and 11 further supports the assertion that the observed frequency dependence is unrelated to breakdown of Meissner screening, as manifested in the rapid-fall effect, and that α_E is in fact apparently absent in the total electronic attenuation α_N. The observation that $\alpha_S/\alpha_N(T)$ satisfies the BCS prediction when $ql \gg 1$, and that no rapid fall is observed, is consistent with the other evidence cited above for absence of α_E in niobium.

F. $\alpha_N(T)$ anomaly: Inadequacy of "developing" electromagnetic breakdown as an explanation

It has been pointed out[17] that as temperature is decreased, the normal-state attenuation is expected to increase and gradually approach a constant level. We have observed[7,24] a maximum in the attenuation-vs-temperature curves for Nb, not unlike that first reported by Robinson and Levy[25] for rhenium. We found[7,24] this maximum to occur at a fixed value of ql: As frequency was increased, the maximum shifted to higher temperatures in such a manner as to preserve ql = const (see Fig. 12). Fortunately, for the three highest frequencies used the maxima occurred above T_c, so that no magnetic field was needed to suppress the superconducting transition at those frequencies

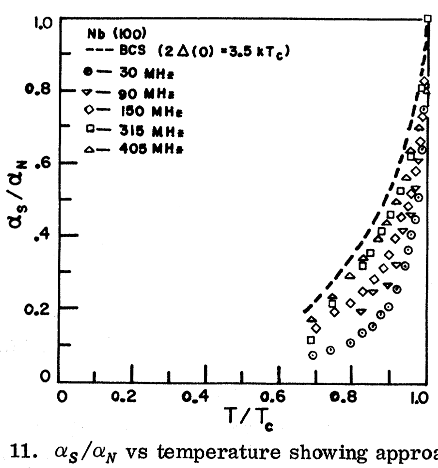

FIG. 11. α_S/α_N vs temperature showing approach to BCS prediction with increase of frequency.

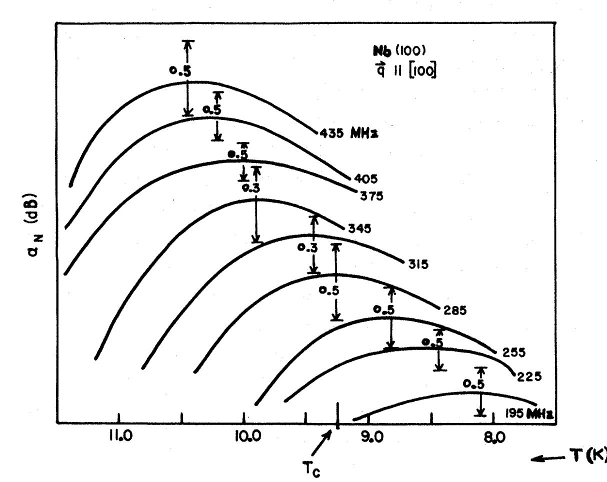

FIG. 12. Demonstration of maxima in the temperature dependence of the normal-state attenuation α_N at low temperature. Note that the temperature shift of the maximum with change of frequency is such as to maintain $ql \approx$ const. The α calibration (dB) is shown for each curve; there is an arbitrary off-set of ordinate between curves (after Blessing, Leibowitz, and Alexander, Ref. 22).

and thereby introduce the complicating influence of magnetoacoustic effects.[15] When these maxima in $\alpha_N(T)$ in niobium were first reported[7,24] the authors were urged to recognize that these maxima indeed gave evidence for the breakdown of electromagnetic interaction in niobium. But here again electromagnetic breakdown can be dismissed.

An explanation for the data in Fig. 12 based on electromagnetic breakdown would insist that as T decreases, and the phonon-limited mean free path $l(T)$ increases, the electromagnetic interaction would more closely approach the breakdown condition for given q. The result would then be a decrease of attenuation with further reduction of temperature. It has been found,[7,24] however, that the shift of the position of the maximum in $\alpha_N(T)$ with change of frequency is not consistent with the behavior expected if electromagnetic breakdown were responsible: The position of the maximum scales in such a manner that $ql \approx$ const[24] ($ql \approx 12$).

As was noted earlier, electromagnetic breakdown is determined by the factor γ, the effect becoming significant when $\gamma \approx 1$. The question to be examined is whether γ scales with ql, as does the effect summarized in Fig. 12. The parameter γ may be written as

$$\frac{\gamma}{\gamma_{EA}} = \frac{\pi}{2}\,\frac{(ql)^2}{[1+(ql)^2]\tan^{-1}ql - ql}\,, \qquad (11)$$

where $\gamma_{EA} = [2\pi\hbar c^2/(e^2 v_t^3)](f/k_F)^2$. We note that γ/γ_{EA} is proportional to $[(ql)g]^{-1}$. The near constancy of $(ql)_{\max}$, the ql value for which the maximum in $\alpha(T)$ occurred, was tested experimentally over more than a decade[24] of frequency range (150–435 MHz), a range corresponding approximately to ql values varying from 6 to 20. As is shown in Fig. 13, γ/γ_{EA} is constant to within 4% in this range. Consequently, we conclude that $\gamma \sim q^2$ and independent of l (and, therefore, T) to within 4%. That is, the behavior of this effect, in which $\alpha_N(T)$ maxima appear in the shear-wave attenuation, cannot be attributed to a "developing" electromagnetic breakdown.

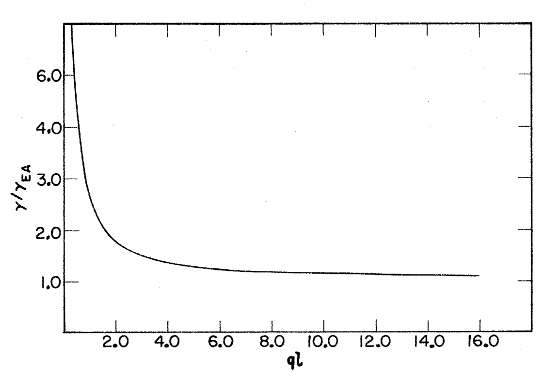

FIG. 13. γ/γ_{EA} as a function of ql, showing approximate constancy of the ratio at high ql.

G. Inadequacy of anomalously large a_D as explanation for absence of observable a_E

We recall (see Fig. 2) that, at high ql, $\alpha_N \approx \alpha_D + \alpha_E$ [see Eq. (4)]. We must ask whether the apparently negligible rapid fall found in the present work is attributable to $\alpha_D \gg \alpha_E$: Perhaps behavior of the field contribution α_E is, in fact, in accord with the theory summarized earlier, but it is the deformation contribution which is anomalous.[26] We find that the possibility can be readily dismissed.

From Fig. 3, which gives the temperature dependence of attenuation near T_c, an upper limit to α_E/α_N of about 1% can be estimated. In order for the α_D contribution to mask that of α_E, therefore, it would be required that $\alpha_D \gtrsim 100\alpha_E$. If we were to insist on an α_E contribution of "normal magnitude," the anomalously large α_D term would be reflected in a comparably inflated α_N, the total electronic attenuation. However, the experimentally determined value, albeit somewhat higher, is of the same order of magnitude as anticipated by the free-electron theory,[17] which gives $\alpha_N = NmV_F^{\omega}/Mv_t^2$; here N and m are the electron concentration and mass, respectively, V_F is the Fermi velocity, and M is the mass density of the metal. From this it is predicted that $\alpha_N \approx 696$ dB/cm at 400 MHz for Nb, which is to be compared with 780 dB/cm determined experimentally in the present work (in Fig. 5 it is seen that $\alpha_N \approx 44$ dB at 400 MHz for sample of thickness 0.58 mm, so that $\alpha_N \approx 780$ dB/cm for 400 MHz). If we use values of effective mass m^* and Fermi velocity V_F determined in independent studies[23,27,28] then the free-electron value of α_N is reduced by a factor in the range $\approx (0.24 - 0.42)$. From Eqs. (2)–(4) it is seen that

$$\frac{\alpha_D}{\alpha_{N\,\mathrm{fem}}} \approx \frac{3\hbar}{16\pi N m v_F} \oint R K_{xy}^2\, d\psi, \qquad (12)$$

which indicates explicitly the influence of the deformation parameter in modifying free-electron predictions.

While comparison with other metals is of limited significance, it nevertheless is to be noted that the attenuation level observed in the present study in Nb is of the same order of magnitude as we have observed in tin samples (of similar resistivity ratio) for which the shear-wave electromagnetic[5] and deformation[10] effects have been studied in detail, and for which α_E (400 MHz)≈ 490 dB/cm and α_D (400 MHz)≈ 800 dB/cm. (Since the data in the case of both metals were taken in the high-ql regime and since the v_t values are comparable in both cases, it is reasonable to compare the attenuations at the same frequency.) While α_N for tin is, in fact, even *larger* than for niobium, it is consistent with our thesis—that α_E is not in evidence in niobium—to compare the niobium attenuation only to the α_D term in tin. In summary, we find that α_N (Nb) is not anomalously large and that, accordingly, the data tend to dismiss the possibility of α_D values of magnitude sufficient to supress observable α_E in the manner hypothesized.

V. CONCLUSIONS

The experiments demonstrate the absence of observable electromagnetic interaction in the shear-wave electronic attenuation in niobium, and the inadequacy, from the point of view of the present theory, of high-frequency breakdown of electromagnetic screening as an explanation. Further support for this conclusion is derivable from the transverse magnetoacoustic study[15] which we have recently reported for niobium. The results point to an anomalous transverse electromagnetic response in niobium: For a transverse wave of propagation vector $\vec{q}$ the $ql \approx 1$ condition apparently does not mark the onset of nonlocality with respect to electron trajectories in niobium, at least for the cases $\vec{q} \parallel [100]$ and $\vec{q} \parallel [110]$, $\vec{\epsilon} \parallel [1\bar{1}0]$.

ACKNOWLEDGMENTS

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