Ultrasonic Attenuation of Longitudinal Waves in Pure Superconducting Niobium in High Magnetic Fields

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By adapting the approach of Kadanoff and Falko to the two-band model, the ultrasonic attenuation coefficient for longitudinal waves in two-band superconductors is obtained. The attenuation coefficient is expressed in terms of various correlation functions. Using the techniques developed by Maki for evaluating the various correlation functions for pure superconductors in high fields, the longitudinal attenuation for arbitrary ql is obtained for the case in which the propagation vector of the sound wave is parallel to the magnetic field. The $(H_{c2}-H)^{1/2}$ field dependence is still a feature of the two-band expression. This expression is used to explain the purity dependence observed in the ultrasonic attenuation in superconducting niobium placed in high magnetic fields.

I. INTRODUCTION

Recent calculation of the band structure of niobium[1] shows the Fermi surface to be comprised of three sheets. The electrons on the sheet centered around the symmetry point Γ are identified as s electrons[2] while those on the other two sheets are identified as d electrons. These sheets are seen to overlap in certain directions. A theory which describes superconductivity in niobium should reflect or take into account this band structure.[3] A model which contained an additional coupling term to account for possible pair formation of electrons from the different bands in the overlapping regions was introduced by Suhl, Matthias, and Walker.[4]

Niobium is also an intrinsic type-II superconductor. Therefore, niobium will enter into a mixed state[5] when placed in high magnetic fields. For type-II superconductors near the upper critical field, Maki[6-9] was able to obtain various transport coefficients for both dirty and pure superconductors. Both of his expressions for the ultrasonic attenuation and thermal conductivity coefficients in dirty superconductors showed a $(H_{c2}-H)$ field dependence, while those for clean superconductors showed a $(H_{c2}-H)^{1/2}$ field dependence. This latter field dependence has been seen in pure superconducting niobium in high fields.[10-15] Kagiwada *et al.*[10] obtained an exact fit of Maki's expression for the longitudinal ultrasonic attenuation for pure superconductors[6] by adjusting certain parameters. Forgan and Gough[11] discounted Kagiwada's fit of Maki's expression by pointing out that their measurements of ultrasonic attenuation in various purity niobium samples all showed the $(H_{c2}-H)^{1/2}$ field dependence, but that they also showed a purity dependence which was in disagreement with Maki's expression. A similar purity dependence in the longitudinal attenuation coefficients in various niobium samples in high fields has also been seen by Tsuda *et al.*[15] The quantitative agreement with Maki's expression grew worse as the purity of the sample became better.

The possible explanation of this dependence on a purity dependence in the density of states was discounted by Forgan and Gough since a false purity dependence in the magnetization in niobium would be implied. They thought that the purity dependence could be accounted for by properly treating the anisotropy of the Fermi surface in niobium.[3] We shall see that the purity dependence can easily be explained in terms of the two-band model. The better agreement with Maki's expression as the sample becomes dirtier is consistent with the two-band model. Garland[16] had shown that as a two-band superconductor becomes dirtier, its properties become more and more like those of a one-band superconductor.

The attenuation coefficient for the two-band superconductors in high fields will be expressed in terms of various correlation functions.[17] Generalizing the methods of Kadanoff and Falko,[18] we obtain the attenuation coefficient for longitudinal waves as

$$\alpha_h^S = \mathrm{Re}\,\frac{q^2}{i\omega\rho_{\mathrm{ion}}v_s}\Big[\langle[\tau_{szz},\,\tau_{szz}]\rangle_{q\omega} - \frac{2P_F^2}{3m_s}\langle[\tau_{szz},\,n_s]\rangle_{q\omega} + \Big(\frac{P_F^2}{3m_s}\Big)^2\langle[n_s,\,n_s]\rangle_{q\omega} + \langle[\tau_{dzz},\,\tau_{dzz}]\rangle_{q\omega} - \frac{2P_F^2}{3m_d}\langle[\tau_{dzz},\,n_d]\rangle_{q\omega} + \Big(\frac{P_F^2}{3m_d}\Big)^2\langle[n_d,\,n_d]\rangle_{q\omega}\Big],$$

where ρ_{ion} is the density of the ions, v_s is the ve-

locity of sound, $n_{s(d)}$ is the number operator for the $s(d)$ electrons, $\tau_{s(d)}$ is the stress tensor operator $s(d)$ band, $m_{s(d)}$ is the mass of the $s(d)$ electrons, and p_F is the Fermi momentum of the electrons. For the propagation vector of the sound wave being parallel to the magnetic field, the various correlation functions can be evaluated using a technique developed by Maki[6] to obtain the longitudinal attenuation coefficient for arbitrary ql.

The purity dependence will enter into the two-band expression for the attenuation coefficients through the ratios of densities of states $N_s/(N_s^\dagger N_d)$ and $N_d/(N_s^\dagger N_d)$. In the two-band model, it is possible for a small increase in the concentration of impurities to cause a rapid change in the density of state in the s band while causing a slight change in the d-band density of states. The rapid change in the magnetic properties will not occur since these properties are dominated by the d-band electrons.[19]

II. FORMULATION

The formulation of the ultrasonic attenuation coefficient in the two-band superconductor will follow somewhat the derivation in Kadanoff and Falko.[18] However, special care must be taken because of the different masses of the electrons in the two bands. Instead of a single equation of motion for the electrons, two equations of motion will be needed in the two-band model. The motion of the ions is still governed by a single equation. The three equations are combined into one by the use of Newton's third law. Following Tsuneto,[17] the motion of the electrons are transformed to a coordinate system which moves with the ions. The longitudinal attenuation coefficient is obtained directly from the single equation expressed in the new coordinate system. Using the method developed by Kadanoff and Falko for treating the effects of the long-range Coulomb interaction, the longitudinal attenuation coefficient is obtained in its final working form.

To begin, we define $\phi(r, t)$ as the displacement of an ion in the neighborhood of (r, t). Using Newton's second law, we have the equation of motion for the ions:

$$M\left(\frac{\partial^2}{\partial t^2}\right)\phi(r,t) = Ze\,\vec{\mathrm{E}}(r,t) + F_{i,e}\,, \tag{2.1}$$

where $\vec{\mathrm{E}}$ is the total electric field, Ze is the ionic charge, and $F_{i,e}$ is the force exerted on the ions by all of the electrons in both bands. The two equations of motion for the electrons are also obtained from Newton's second law and are

$$\left\langle m_s \frac{d}{dt} j_s(r,t) + \nabla\cdot\tau_s(r,t)]\right\rangle = -N_s e\vec{\mathrm{E}}(r,t) + f_{se,i} + f_{se,de}\,, \tag{2.2}$$

$$\left\langle m_d \frac{d}{dt} j_d(r,t) + \nabla\cdot\tau_d(r,t)]\right\rangle = -N_d e\vec{\mathrm{E}}(r,t) + f_{de,i} + f_{de,se}\,, \tag{2.3}$$

where (2.2) is the equation of motion for the s electrons and (2.3) is the equation of motion for the d electrons. The subscripts refer to the type of electrons being described. The current and stress tensor are defined as

$$j(r,t) = \sum_\sigma\left[\frac{(\nabla-\nabla')}{2im}\,\psi_\sigma^\dagger(r,t)\psi_\sigma(r,t)\right], \tag{2.4}$$

$$\tau_{ij} = \sum_\sigma\left[\frac{(\nabla-\nabla')_i}{2i}\,\frac{(\nabla-\nabla')_j}{2im}\,\psi_\sigma^\dagger(r',t)\psi_\sigma(r,t)\right]_{r=r'}. \tag{2.5}$$

The forces $f_{se,i}$, $f_{se,de}$, $f_{de,i}$, and $f_{de,se}$ are, respectively, the forces exerted on the s electrons by the ions, by the d electrons, and the forces exerted on the d electrons by the ions, and by the s electrons.

Using Newton's third law, we obtain

$$M\frac{d^2}{dt^2}\phi(r,t) = -\frac{Z}{N_s+N_d}\left\langle m_s\frac{d}{dt}\,j_s(r,t) + \nabla\cdot\tau_s(r,t)\right\rangle - \frac{Z}{N_s+N_d}\left\langle m_d\frac{d}{dt}j_d(r,t) + \nabla\cdot\tau_d(r,t)\right\rangle \tag{2.6}$$

This equation can be further reduced by assuming that $\phi(r, t)$ varies as

$$\phi(r,t) = \phi(q,\omega)\,e^{iq\cdot r - i\omega t}\,. \tag{2.7}$$

Equation (2.6) becomes

$$-M\omega^2\phi(r,t) = [Z/(N_s+N_d)]\langle im_s\omega\, j_s(r,t) - \nabla\cdot\tau_s\rangle + [Z/(N_s+N_d)]\langle im_d\omega j_d(r,t) - \nabla\cdot\tau_d\rangle\,. \tag{2.8}$$

Transforming to a coordinate system moving with the ions, the above equation becomes

$$\rho_{\mathrm{ion}}\omega^2\phi_i(q,\omega) - \tfrac{1}{5}\,v_{sF}^2 m_s N_s + v_{dF}^2 m_d N_d \times \{2q_i[\vec{\mathbf{q}}\cdot\phi(q,\omega)] + q^2\phi_i(q,\omega)\} + \{\langle[h_{si},\ h_{sj}]\rangle_{q\omega} + \langle[h_{di},\ h_{dj}]\rangle_{q\omega}\}\omega^2\phi_j(q,\omega) = 0\,, \tag{2.9}$$

where the ith component of $h_{s(d)}$ is

$$h_{s(d)i}(r,t) = q_j\tau_{s(d)ij}(r,t)/\omega - m_{s(d)}j_{s(d)i}(r,t)\,. \tag{2.10}$$

In the longitudinal case, where ϕ_i is parallel to the

wave vector $\vec{q}$, the sound-wave dispersion relation becomes

$$\rho_{\text{ion}}\omega^2 = \tfrac{3}{5}\,(N_s m_s v_{sF}^2 + N_d m_d v_{dF}^2)\,q^2 + (\langle[h_{sI}^L, h_{sI}^L]\rangle_{q\omega} + \langle[h_{dI}^L, h_{dI}^L]\rangle_{q\omega})\omega^2\,, \tag{2.11}$$

where

$$h_{s(d)I}^L(r,t) = \frac{q}{\omega}\,\tau_{s(d)zz}(r,t) - \frac{m_{s(d)}\omega}{q}\,n_{s(d)}(r,t)\,. \tag{2.12}$$

The longitudinal attenuation coefficient is given by

$$\alpha_L^S = -\,\text{Re}(i\omega/\rho_{\text{ion}}V_S)\{\langle[h_{sI}^L, h_{sI}^L]\rangle_{q,\omega} + \langle[h_{dI}^L, h_{dI}^L]\rangle_{q,\omega}\}\,. \tag{2.13}$$

The correlation functions in (2.13) are averaged in a system which includes the effects of the long-range electron-electron interaction between electrons in the same band. The interaction between electrons belonging to different bands were taken into account in (2.2) and (2.3) by the inclusion of $f_{se,de}$ and $f_{de,se}$ in the equations. The individual correlation functions in (2.13) can be treated in exactly the same way as in the Kadanoff-Falko paper. The resulting attenuation coefficient for the longitudinal sound wave is now given by

$$\alpha_L^S = \text{Re}\,\frac{q^2}{i\omega\rho_{\text{ion}}V_S}\Big[\langle[\tau_{szz},\tau_{szz}]\rangle_{q\omega} - \frac{2P_F^2}{3m_s}\langle[\tau_{szz},n_s]\rangle_{q\omega} + \Big(\frac{P_F^2}{3m_s}\Big)^2\langle[n_s,n_s]\rangle_{q\omega} + \langle[\tau_{dzz},\tau_{dzz}]\rangle_{q\omega} - \frac{2P_F^2}{3m_d}\langle[\tau_{dzz},n_d]\rangle_{q\omega} + \Big(\frac{P_F^2}{3m_d}\Big)^2\langle[n_d,n_d]\rangle_{q\omega}\Big]\,, \tag{2.14}$$

where the correlation functions are now averaged in the fictitious system with out any current-current interactions.

III. ATTENUATION COEFFICIENT FOR LONGITUDINAL WAVES

The various correlation functions for pure superconductors in high fields are evaluated using the techniques of the thermal Green's function. However, special care must be taken since it has been shown[20] that the perturbation expansion in terms of the order parameter will lead to unphysical results for the pure superconductor. Maki has used the similarity between the density of states for the pure superconductor in high fields and that of a current-carrying superconductor as a means of circumventing this difficulty.[6] To be able to apply Maki's techniques to the evaluations of the various correlation functions in the two-band case, we shall assume that the density of states for both bands are of the form

$$\frac{N(\omega)}{N(0)} = \int_{-\infty}^{\infty} d\alpha \int \frac{d\Omega}{4\pi}\,\rho(\alpha,\Omega)\,\text{Re}\Big(\frac{\omega-\alpha}{[(\omega-\alpha)^2-\Delta^2]^{1/2}}\Big)\,, \tag{3.1}$$

where

$$\rho(\alpha,\Omega) = [(\sqrt{\pi})\epsilon\,\sin\theta]^{-1}\exp\Big[-\Big(\frac{\alpha}{\epsilon\,\sin\theta}\Big)^2\Big]\,, \tag{3.2}$$

with ϵ given by

$$\epsilon = V_F[\tfrac{1}{2}eH_{c_2}]^{1/2} \tag{3.3}$$

and θ the angle between $\vec{p}$ (the momentum of the quasiparticle) and the magnetic field.

In the case where the propagation vector $\vec{q}$ of the sound wave is parallel to the magnetic field, the angular integrations involved in the various correlation functions are much simplified. The evaluations of these functions will lead to expressions exactly like Maki's[6] except for the band subscripts. Combining the resulting expressions for the various correlation functions, we obtain the longitudinal attenuation coefficient for arbitrary ql:

$$\frac{\alpha_L^S}{\alpha_L^n} = 1 - \frac{N_d}{N_s+N_d}\,\frac{\Delta_s}{2T}\int_{-\infty}^{\infty} d\alpha\,\phi_s(\alpha,ql)\cosh^{-2}\Big(\frac{\alpha}{2T}\Big) - \frac{N_s}{N_s+N_d}\,\frac{\Delta_d}{2T}\int_{-\infty}^{\infty} d\alpha\,\phi_d(\alpha,ql)\cosh^{-2}\Big(\frac{\alpha}{2T}\Big)\,, \tag{3.4}$$

where

$$\phi_{s(d)}(\alpha,ql) = \frac{1}{3X(y)}\int\frac{d\Omega}{4\pi}\,\rho_{s(d)}(\alpha,\Omega)(1-3z^2)\Big(\frac{1}{1+y^2z^2}\cdot\frac{1}{1-y^{-1}\tan^{-1}y} - \frac{3}{y^2}\Big)$$

$$= \frac{1}{3X(y)}\,\frac{1}{\sqrt{\pi}}\int_0^1 \frac{dz}{\epsilon_{s(d)}}\,\frac{(1-3z^2)}{(1-z^2)^{1/2}}\exp\Big[-\Big(\frac{\alpha}{\epsilon_{s(d)}}\Big)^2(1-z^2)^{-1}\Big]\Big(\frac{1}{1+y^2z^2}\cdot\frac{1}{1-y^{-1}\tan^{-1}y} - \frac{3}{y^2}\Big)\,, \tag{3.5}$$

with

$$X(y)=\frac{\tan^{-1}y'}{3(y-\tan^{-1}y)}-\frac{1}{y^2}\,,\qquad y=ql\,.$$

The asymptotic forms are given by

$$\frac{\alpha_L^S}{\alpha_L^n}=1-\frac{N_d}{N_s+N_d}\frac{\Delta_s}{2T}\left\{1-\frac{1}{2}\left(\frac{\epsilon_s}{2T}\right)^2\left[1+\frac{1}{y^2}\left(1-\frac{4}{15X(y)}\right)\right]+\frac{1}{2}\left(\frac{\epsilon_s}{2T}\right)^4\left[1+\frac{1}{y^2}\left(\frac{26}{15}-\frac{124}{315X(y)}\right)+\frac{1}{y^4}\left(1-\frac{4}{15X(y)}\right)\right]\right\}$$

$$-\frac{N_s}{N_s+N_d}\frac{\Delta_d}{2T}\left\{1-\frac{1}{2}\left(\frac{\epsilon_d}{2T}\right)^2\left[1+\frac{1}{y^2}\left(1-\frac{4}{15X(y)}\right)\right]\right.$$

$$\left.+\frac{1}{2}\left(\frac{\epsilon_d}{2T}\right)^4\left[1+\frac{1}{y^2}\left(\frac{26}{15}-\frac{124}{315X(y)}\right)+\frac{1}{y^2}\left(1-\frac{4}{15X(y)}\right)\right]\right\}\qquad \text{for } T\lesssim T_{cd}\,, \tag{3.6}$$

$$\frac{\alpha_L^S}{\alpha_L^n}=1-\frac{N_d}{N_s+N_d}\frac{\sqrt{\pi}\Delta_s}{3X(y)\epsilon_s}\left(\frac{1}{1-y^{-1}\tan^{-1}y}\left[\frac{1}{(1+y^2)^{1/2}}\left(1+\frac{3}{y^2}\right)-\frac{3}{y^2}\right]+\frac{3}{2y^2}-\frac{4T}{\sqrt{\pi}}\frac{\ln 2}{\epsilon_s}\left\{\frac{1}{1-y^{-1}\tan^{-1}y}\right.\right.$$

$$\times\left[\frac{1}{1+y^2}\left(1+\frac{3}{y^2}\right)-\frac{3}{y^2}\right]+\frac{6}{y^2}\left.\right\}-\frac{\pi^2}{3}\left(\frac{T}{\epsilon_s}\right)^2\left[\frac{1}{1-y^{-1}\tan^{-1}y}\left(1+\frac{3}{y^2}\right)\frac{y^2}{(1+y^2)^{3/2}}-\frac{9}{y^2}\right]\Bigg)-\frac{N_s}{N_s+N_d}\frac{\sqrt{\pi}}{3X(y)}\frac{\Delta_d}{\epsilon_d}$$

$$\times\left(\frac{1}{1-y^{-1}\tan^{-1}y}\left[\frac{1}{(1+y^2)^{1/2}}\left(1+\frac{3}{y^2}\right)-\frac{3}{y^2}\right]+\frac{3}{2y^2}-\frac{4T}{\sqrt{\pi}}\frac{\ln 2}{\epsilon_d}\left\{\frac{1}{1-y^{-1}\tan^{-1}y}\left[\frac{1}{1+y^2}\left(1+\frac{3}{y^2}\right)-\frac{3}{y^2}\right]+\frac{6}{y^2}\right\}\right.$$

$$\left.-\frac{\pi^2}{3}\left(\frac{T}{\epsilon_d}\right)^2\left[\frac{1}{1-y^{-1}\tan^{-1}y}\left(1+\frac{3}{y^2}\right)\frac{y^2}{(1+y^2)^{3/2}}-\frac{9}{y^2}\right]\right)\qquad \text{for } T\ll T_{cd}\,. \tag{3.7}$$

The field dependence enters in the behavior of the energy gaps in high magnetic fields. For the discussion in this section, we will assume that d-band energy gap is given by[19]

$$\Delta_d^2=\frac{1}{2\pi N_d}\,\frac{H_{c2}(T)-H}{1.16[2\kappa_2^2(t)-1]}\left(H_{c2}(t)-\tfrac{1}{2}t\,\frac{d}{dt}\,H_{c2}(t)\right)\,. \tag{3.8}$$

The field dependence of the lower energy gap shall be assumed to have a similar form in the temperature region above the lower transition temperature T_{cs}. Since the observed field dependence is definitely $(H_{c2}-H)^{1/2}$ in niobium, the assumption made above seems to be correct. In addition, we will assume that the energy gaps are proportional to one and other. This assumption is analogous to the observed behavior of the two gaps as the temperature varies, i.e., the recent discovery of Hafstom *et al.*,[21] that the relative strength of the two gaps remains constant over a wide temperature range.

IV. COMPARISON WITH EXPERIMENTAL RESULTS

Kagiwada *et al*[10] were able to fit their data on a niobium sample with a residual resistivity ratio (RRR) 300 with Maki's expression by using the value 1.5×10^{34} states/cm^3 erg for the density of states. To achieve a fit of the experimental data on niobium samples with RRR between 100 and 600, Tsuda *et al.*[15] used values ranging from 0.2 to 5.8 $\times10^{34}$ states/cm^3 erg (the lower values being required to fit the data on the purer samples). This variation in the density of states with the purity of the sample reflects the fact that the density of states as a parameter would have a purity dependence.[11,13,15] A strong purity dependence in the density of states would imply a similar dependence in the magnetic properties. However, this contradicts both the BCS theory, which predicts a weak purity dependence, and the observed magnetic properties in niobium,[15,22] which show a dependence stronger than the BCS prediction but not as much as would be implied by the purity dependence in the ultrasonic attenuation in pure niobium near the

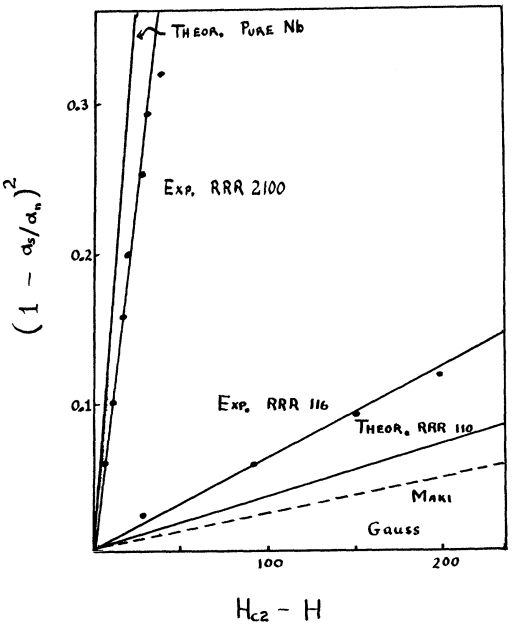

FIG. 1. The normalized attenuation in the mixed states of superconducting niobium as a function of the external field measured from H_{c2}. The theoretically predicted slopes of a pure sample and a RRR 110 sample are shown. (•) are the experimental values for RRR 2100 and RRR 116 samples as measured by Forgan and Gough. The bottom slope is the one resulting from substituting the best experimentally available values into Maki's one-band expression.

upper critical field. Another difficulty with the one-band analysis is the difficulty in seeing how a small change in the impurity concentration (a few ppm is all that is needed) can cause the rapid change in the density of states.[13]

The two-band expression for the ultrasonic attenuation in high fields allows an explanation without any of the drawbacks associated with the purity dependence in the one-band expression. It is possible for a small change in the impurity concentration to cause a rapid change in the density of states in the s band since the small impurity concentration may be quite large in comparison to the s-band density of states. A strong purity dependence in the magnetic properties would not necessarily be implied since the small changes in the impurity concentrations do not affect the density of states of the d electrons, which are the ones which dominate the magnetic properties in the two-band superconductors. It should be noted however, that the s electrons do play some role in determining these properties.[19]

In formulating the two-band expression for the attenuation coefficient, we have assumed that the Fermi surface can be considered to be a sphere. Hohenberg and Werthamer[23] have shown that the Fermi surface anisotropy can enhance the results obtained when using the spherical model by an enhancement factor. Ohtsuka and Takano[24] showed that the enhancement factor for niobium is 1.13. Therefore, it may be expected that the two-band expression (3.4) would predict numerical values lower than those observed.

To demonstrate the advantages of the two-band expression for the ultrasonic attenuation, we will show that some of slopes observed by Forgan *et al.*[11] of $(1-\alpha_L^S/\alpha_L^n)^2$ versus $(H_{c2}-H)$ can be obtained by substituting the best experimentally available numerical values into (3.7). The reason for not being able to duplicate all of the observed slopes is that the density-of-states ratio $N_s(N_s+N_d)$ is known for only two samples, one with a RRR of 110 and the other with a RRR over 2100. The numerical values for the extremely pure sample (RRR $\gtrsim$ 2100) are $N_s/(N_s+N_d)=0.12$, $\kappa_2=1.87$ at 4.2 °K, $H_{c2}=2700$ Oe at 4.2 °K and 4050 Oe at 0 °K, $v_{Fd}=3.0\times10^7$ cm/sec., $\Delta_s/\Delta_d=0.1$, $m_d=12.5m_s$, and $h(t)=H_{c2}(t)/dh/dt_{c2}=0.9$. The values for the RRR 110 sample are $N_s/(N_s+N_d)=0.015$, $\kappa_2=2.4$ at 4.2 °K, $H_{c2}=3808$ Oe at 4.2 °K, $v_{Fd}=3.0\times10^7$ cm/sec, $\Delta_s/\Delta_d=0.1$, $m_d=35m_s$, and $h(t)=0.7$.

Substituting the above values into (3.7), we obtain the two theoretical curves shown in Fig. 1. The experimental values for the RRR 2100 sample and RRR 116 sample measured by Forgan *et al.*[11] are shown for comparison. As we see, the theoretically predicted slope for pure niobium is greater than the observed slope for the RRR 2100 sample, while the predicted slope for the RRR 110 sample is less than the observed slope for the RRR 116 sample. The relative values of the two theoretical slopes to the two observed slopes are in keeping with the two-band model.

It is hoped that the density-of-states ratios for niobium of other purities can be obtained to further check the correctness of the two-band expression for the ultrasonic attenuation in the mixed states of transition-metal superconductors.

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