Comment on "Anomalous ultrasonic attenuation in pure superconducting Nb'' [†]

Marwan Nusair and I-Ming Tang

Department of Physics, American University of Beirut, Beirut, Lebanon

{Received 8 August 1973)

The recently reported measurements of the ultrasonic attenuation of both longitudinal and transverse waves in a pure niobium superconductor by Carsey and Levy are analyzed on the basis of a two-band description of the superconducting state. It is seen that some of the ambiguities resulting from a single-band analysis are not present in the two-band analysis.

I. INTRODUCTION

In a recent paper, ' Carsey and Levy have reported the measurements² of the attenuation of $110-$, 80-, 42. 5-, and 21-MHz longitudinal waves and 51 and 16.6-MHz transverse waves in a very pure niobium superconductor (a specimen with a residual resistivity ratio of over 7000). Comparing their longitudinal data with the model of ultrasonic attenuation of Maki, 3 which takes into account the increased absorption of phonons by electrons near the edge of the gap; with that of Fate et $al.$ ⁴ and Trivisonno et $al.$, 5 which assumes a difference of the electronic mean free paths in the normal and superconducting phases; and with that of Lacy and Daniel, ⁶ which assumes that the normalized longitudinal attenuation coefficient for a superconductor having a second energy gap⁷ is given by

$$
\frac{\alpha_S}{\alpha_N} = F_1(t) \frac{2}{e^{\Delta_1/kT} + 1} + [1 - F_1(t)] \frac{2}{e^{\Delta_2/kT} + 1},
$$

where

$$
F_1(t) = \frac{\Delta_1(t)}{\Delta_1(t) + \Delta_2(t)} + \text{const}
$$

and

$$
\Delta_i(t) = \Delta_i(0) \frac{\Delta_{\text{BCS}}(t)}{1.74 \ k_B T_c}
$$

Carsey and Levy concluded that while each of the above models was successful in explaining part of the data, none of them were complete in the sense that they were not able to explain all of the features seen in the attenuation data. The purpose of this paper is to present a two-band analysis of their data using expressions for the attenuation coefficients different from the phenomenological expression for the longitudinal expression proposed by Lacy and Daniel. Our expressions for the attenuation coefficients of longitudinal and transverse waves in a two-band superconductor is obtained from first principles in a manner similar to that used by one of the present authors to obtain the mixed-state attenuation coefficient in a two-band superconductor.⁸ Thus this paper serves also as continuation of that paper.

II. TWO-BAND ATTENUATION COEFFICIENTS

As was pointed out in Ref. 8, the electrons in the two bands should be governed by separate equations of motion since they are subjected to different constraints. The equations of motion are

$$
\left\langle m_s \frac{d}{dt} \overline{\mathbf{j}}_s(\mathbf{r}, t) + \nabla \cdot \overline{\mathbf{r}}_s(\mathbf{r}, t) \right\rangle
$$

= $- N_s e \overline{\mathbf{E}}(\mathbf{r}, t) + \overline{\mathbf{f}}_{se, i} + \overline{\mathbf{f}}_{se, de}$ (1a)

and

$$
\left\langle m_d \frac{d}{dt} \overrightarrow{j}_d(r, t) + \nabla \cdot \overrightarrow{\tau}_d(r, t) \right\rangle
$$

= $- N_d e \overrightarrow{\mathbf{E}}(r, t) + \overrightarrow{\mathbf{I}}_{de, i} + \overrightarrow{\mathbf{I}}_{de, se}$, (1b)

 λ

where $\overline{\mathbf{j}}_{s(d)}(r, t)$ and $\overline{\mathbf{T}}_{s(d)}(r, t)$ are the current and stress-tensor operators for electrons in the $s(d)$ band, respectively, and whose forms are the same as those defined for a one-band system. 9 The f's are the various forces acting on the electrons and which are defined in Ref. 8 along with all the other quantities. $\langle \cdots \rangle$ denotes an ensemble average over the ground state. These equations, (la) and (lb), along with the equation of motion of the ions

$$
M\frac{d^2}{dt^2}\,\vec{\phi}(r,\,t)=Ze\vec{\mathbf{E}}(r,\,t)+\vec{\mathbf{f}}_{i,\,e}\;, \tag{2}
$$

where $\phi(r, t)$ is the displacement of the ion in the neighborhood of (r, t) , can be combined into one via Newton's second law:

on for the longitudinal expression proposed by
by
by a significant deviation of
$$
(r, t)
$$
, can be combined into one
via Newton's second law:

$$
M \frac{d^2}{dt^2} \vec{\phi}(r, t) = -\frac{Z}{N_s + N_d} \left\langle m_s \frac{d}{dt} \vec{j}_s(r, t) + \nabla \cdot \vec{\tau}_s(r, t) \right\rangle - \frac{Z}{N_s + N_d} \left\langle m_d \frac{d}{dt} \vec{j}_d(r, t) + \nabla \cdot \vec{\tau}_d(r, t) \right\rangle.
$$
 (3)

Equation (3) can be further simplified by assuming that $\phi(r, t)$ varies as

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

(4)

This results in Eq. (3) becoming

$$
-M\omega^2 \overline{\phi}(r, t) = \frac{Z}{N_s + N_d} \langle im_s \omega \overline{j}_s(r, t) - \nabla \cdot \overline{\tau}_s \rangle + \frac{Z}{N_s + N_d} \langle im_d \omega \overline{j}_d(r, t) - \nabla \cdot \overline{\tau}_d \rangle . \tag{5}
$$

Transforming to a coordinate system moving with the ions, 10 the above equation becomes

$$
\rho_{\rm ion} \omega^2 \phi_i(q,\omega) - \frac{1}{5} (v_{sF}^2 m_s N_s + v_{dF}^2 m_d N_d) \left\{ 2 q_i [\bar{\mathbf{q}} \cdot \overline{\phi}(q,\omega)] + q^2 \phi_i(q,\omega) \right\}
$$

+
$$
\langle \langle [h_{si}, h_{sj}] \rangle_{\mathbf{q}\omega} + \langle [h_{di}, h_{dj}] \rangle_{\mathbf{q}\omega} \rangle \omega^2 \phi_j(q, \omega) = 0
$$
, (6)

where the *i*th component of $h_{s(d)}$ is

$$
h_{s} (a) i(r, t) = q_i \tau_{s} (a) i_j(r, t) / \omega
$$

- $m_{s} (a) j_{s} (a) i(r, t)$. (7)

For longitudinal waves, where ϕ_i is parallel to the wave vector $\dot{\tilde{q}}$, the sound-wave dispersion relation becomes

$$
\rho_{\text{ion}}\omega^{2} = \frac{3}{5} \left\{ N_{s}m_{s} v_{sF}^{2} + N_{d} m_{d} v_{dF}^{2} \right\} q^{2}
$$

$$
- \left\{ \left\langle \left[h_{s}^{\text{L}}, h_{s}^{\text{L}} \right] \right\rangle_{q\omega} + \left\langle \left[h_{d}^{\text{L}}, h_{d}^{\text{L}} \right] \right\rangle_{q\omega} \right\} ,\tag{8}
$$

where

$$
h_i^L = \frac{q}{\omega} \tau_{i, \text{ or }} (r, t) - \frac{m}{q} n_i(r, t) . \qquad (9)
$$

The sound-wave dispersion relation is different for transverse waves since in this case, the displacement vector $\phi_i(q, z)$ is perpendicular to the wave vector \overline{q} . Taking \overline{q} to be pointing in the z direction and the displacement vector to be in the x direction, Eq. (6) becomes

$$
\rho_{\text{ion}}\omega^{2} = \frac{1}{5} \left\{ N_{d}m_{d}v \frac{2}{dF} + N_{s}m_{s}v \frac{2}{sF} \right\} q^{2}
$$

$$
- \left\{ \left\{ \left[h_{s}^{T}, h_{s}^{T} \right] \right\}_{q\omega} + \left\{ \left[h_{d}^{T}, h_{d}^{T} \right] \right\}_{q\omega} \right\} \omega^{2} , \qquad (10)
$$

where

$$
h_i^{\mathrm{T}} = \frac{q}{\omega} \tau_{i, \, \mathbf{x}}(r, \, t) - m_i \, j_{i, \, \mathbf{x}}(r, \, t) \tag{11}
$$

The attenuation coefficients can be obtained directly from the sound-wave dispersion relations

(8) or (10). For longitudinal waves, the two-band expression for the attenuation coefficient is

$$
\alpha_{\rm L}^s = -\operatorname{Re}\frac{i\omega}{\rho_{\rm ion}V_s} \left\{ \left\{ \left[h_s^{\rm L}, h_s^{\rm L} \right] \right\}_{\rm q\omega} + \left\langle \left[h_d^{\rm L}, h_d^{\rm L} \right] \right\rangle_{\rm q\omega} \right\} \,, \quad (12)
$$

while for transverse waves it is

$$
\alpha_{\mathrm{T}}^{\mathrm{s}} = -\mathrm{Re}\,\frac{i\omega}{\rho_{\mathrm{ion}}V_t} \big\{ \big\{ \big[\, h_s^{\mathrm{T}}, \, h_s^{\mathrm{T}} \big] \big\}_{q\,\omega} + \big\langle \big[\, h_d^{\mathrm{T}}, \, h_d^{\mathrm{T}} \big] \big\rangle_{q\,\omega} \big\} \, . \tag{13}
$$

It must now be remembered that while the effects of the Coulomb interaction between electrons belonging to different bands were taken into account in Eqs. (la) and (lb), the effects of the long-range Coulomb interaction between electrons within the same band have not been taken into account. Therefore, the various correlation functions appearing in Eqs. (12) and (13) are averaged over a system containing long-range Coulomb interactions. While these interactions do not lead to large effects in the normal phase, they do lead to measureable effects in the superconducting phase.

It is, therefore, necessary to take into account these interactions when evaluating the correlation functions appearing in the definition of the attenuation coefficients. This is best done by going to the random-phase approximation (RPA) , ¹¹ The result of this treatment of the long-range Coulomb interactions is that the longitudinal attenuation coefficient becomes

$$
\alpha_{\rm L}^s = -\operatorname{Re}\frac{q^2}{i\omega\rho_{\rm ion}V_s} \sum_{i=s,\,d} \left\{ \langle \left[\tau_{izz},\,\tau_{izz}\right]\rangle'_{q\,\omega} - \frac{|\left\langle \left[\tau_{izz},\,n_i\right]\right\rangle'_{q\,\omega}|^2}{\left\langle [n_i,\,n_i]\right\rangle'_{q\,\omega}} \right\},\tag{14}
$$

while the transverse attenuation coefficient becomes

$$
\alpha_{\rm T}^s = -\operatorname{Re}\frac{q^2}{i\omega\rho_{\text{ion}}V_t} \sum_{i=s,d} \left\{ \langle \left[\tau_{ixz}, \tau_{ixz}\right] \rangle_{q\omega}^{\prime} - \frac{|\langle \left[\tau_{ixz}, j_{ix}\right] \rangle_{q\omega}^{\prime}|^2}{\langle \left[j_{ix}, j_{ix}\right] \rangle_{q\omega}^{\prime}|^2} \right\} \right. \tag{15}
$$

The primes denote the fact that the correlation functions are now being averaged over a fictitious system which does not contain the long-range Coulomb interactions.

The evaluations of the correlation function in Eq. (14) is straightforward and gives for the attenuation coefficient of longitudinal waves in a two-band superconductor

$$
\alpha_{\rm L}^{s} = \frac{N_{s} m_{s} V_{sF}}{\rho_{\rm ion} V_{s} l_{s}} \left(\frac{1}{3} \frac{(q l_{s})^{2} \tan^{-1} q l_{s}}{q l_{s} - \tan^{-1} q l_{s}} - 1 \right) 2 f(\Delta_{s}) + \frac{N_{d} m_{d} V_{dF}}{\rho_{\rm ion} V_{s} l_{d}} \left(\frac{1}{3} \frac{(q l_{d})^{2} \tan^{-1} q l_{d}}{q l_{d} - \tan^{-1} q l_{d}} - 1 \right) 2 f(\Delta_{d}) , \qquad (16)
$$

where $l_{s(d)}$ is the mean free path for the s (d) electrons and where $f(x)$ is the Fermi-Dirac function. By setting Δ_s and Δ_d , the two energy gaps, to zero in Eq. (16), we obtain the normal-state attenuation coefficient for longitudinal waves. Thus at T_c where both energy gaps vanish, we get $\alpha_L^s / \alpha_L^r = 1$ (α_L^r being the normal-state attenuation coefficient). The evaluation of the correlation functions in Eq. (15) gives the following two-band transverse attenuation coefficient:

$$
\frac{\alpha_T^s}{\alpha_T^m} = 2 \frac{\left\{ N_d m_d V_{dF} \left[1 - g(ql_d) \right] / l_d \right\} f(\Delta_d) + \left\{ N_s m_s v_{sF} \left[1 - g(ql_s) \right] / l_s \right\} f(\Delta_s)}{\left\{ N_d m_d V_{dF} \left[1 - g(ql_d) \right] / l_d g(ql_d) \right\} + \left\{ N_s m_s v_{sF} \left[1 - g(ql_s) \right] / l_s g(ql_s) \right\}} \right\} \tag{17}
$$

where $g(x)$ is the Pippard function

$$
g(x) = \frac{3}{2x^3} [(x^2 + 1) \tan^{-1} x - x].
$$
 (18)

Setting Δ_s and Δ_d to zero in Eq. (17) does not give unity since the presence of the Meissner effect in the superconductor precludes the existence of one of the normal-state damping mechanism. %hat is obtained by setting the energy gaps to zero is the initial drop of the attenuation just below the critical temperature T_c .

III. TWO-BAND ANALYSIS

The three features of the single-band analysis of the ultrasonic-attenuation data of Carsey and Levy, which are of particular interest to us, are the apparently different temperature behaviors of the energy gaps determined from the different sets of attenuation data, the different values of the energy

FIG. 1. Temperature behavior of the energy gap in niobium superconductor as determined from a singleband analysis of Carsey and Levy's attenuation data.

gap at $T = 0$ °K obtained from the single-band analysis of the different frequency-attenuation data, and the modification required of the Pippard function (18) so that the initial drop in the transverse attenuation conforms to the relation

$$
\alpha_{\rm T}^{\rm s}/\alpha_{\rm T}^{\rm n}=2g(ql)\tag{19}
$$

(this being the drop predicted in the single-band theory¹²). The apparently different temperature behaviors of the energy gap are clearly seen if we look at Figs. 5 and 6 of Ref. 1 and which are plotted together in Fig. 1 of this paper. The values of the energy gap at $T = 0$ °K predicted from a fit of the attenuation data to the single-band attenuation coefficients range from a value of $(1.72 \pm 0.02)k_B T_c$ to the value $(1.93\pm0.03)k_BT_c$ when only the lowtemperature data are considered, and from a value of $(3.52 \pm 0.1) k_B T_c$ to a value of $(5.20 \pm 0.15) k_B T_c$ when only the high-temperature data are considered.¹³ The greatest variation in the values of the energy gap comes from the transverse data: The high-temperature data for the 51-MHz wave indicates an energy gap $\Delta(0) = (3.79 \pm 0.10)k_B T_c$, while the high-temperature data for the 16.6-MHz wave implies an energy gap $\Delta(0) = (5.20 \pm 0.15)k_B T_c$.

The remaining feature is that the Pippard function $g(ql)$ had to be modified to¹⁴

$$
g'(ql) = g(ql) + [1 - g(ql)]/1.6 \tag{20}
$$

in order that the initial drop in the transverse attenuation be given by (19). Even with this modification, the fits of the transverse-attenuation data could not be made perfect at both high and low temperatures, simultaneously.

The two-band analysis of the data on the attenuation of the 110-, 80-, 42. 5-, and 21-MHz longitudinal waves and of the 51- and 16.6-MHz transverse waves in the niobium superconductor began by noting that the two two-band coefficients in Eq. (16) when normalized and in Eq. (17) depend on the ratio $(N_a m_d v_{dF})/(N_s m_s v_{sF})$, the two mean free paths l_s and l_d , and the two energy gaps Δ_s and Δ_d . By varying all the parameters, we noted that the two attenuation coefficients were most sensitive to variations in the values of the energy gaps and least sensitive to variations of the mean free paths. Because of the relative insensitivity to variations of the mean free paths, we did not take into ac-

FIG. 2. Temperature behavior of the primary energy gap in a niobium superconductor as determined from a two-band analysis of attenuation data.

count any possible temperature dependences in the mean free paths. Later, we found that the uncertainities in the values of the other parameters lead to errors greater than the possible errors resulting from our neglect of the possible temperature dependences in the mean free paths.

The actual two-band analysis of the data was in two steps. By attempting to account for the initial drop in the attenuation of the 51- and 16.6-NHz transverse waves with Eq. (17), we were able to determine the values of the ratio $(N_{\rm d}m_{\rm d}v_{\rm dF})/$ $(N_s m_s v_{sF})$ and of the d-band mean free path (the mean free path for the s electron was taken to be 40×10^{-4} cm) since the initial drop is independent of the energy gaps. We found that we were able to account for the drops in both the 51- and 16.6-MHz transverse attenuation by letting $(N_a m_a v_{dF})/$ $(N_sm_sv_{sF})$ equal 80 and l_d be equal to 20 $\times10^{-4}$ cm. The second phase of our analysis involved the fit of

~%'ork supported in parts by an Arts and Sciences Research Grant (American University of Beirut).

- 1 F. Carsey and M. Levy, Phys. Rev. B 7, 4123 (1973).
- 2 F. Carsey, Ph.D. dissertation (University of California, Los Angeles, 1971) (unpublished). 3 K. Maki, Phys. Kondens. Mater. 8, 305 (1969).
-
- $4W$. A. Fate, R. W. Shaw, and G. L. Salinger, Phys. Rev. 174, 413 (1968).
- 5J. Trivisonno, J. Washiek, M. J. Keck, and R. Reifenberger, J. Low Temp. Phys. 4, ⁹⁷ (1971).
- 6L . L. Lacy and A. C. Daniel, Phys. Rev. Lett. 27 ,

either the two-band longitudinal-attenuation coefficient [Eq. (16)] or the two-band transverse-attenuation coefficient $[Eq. (17)]$ to Carsey and Levy's data by varying the values of the two energy gaps. We found that a nearly perfect fit of all the data down to $0.3T_c$ could be achieved if we assumed the ratio between the energy gaps, Δ_d/Δ_s , was equal to 10. The temperature behaviors of the primary energy gap, Δ_d , obtained from the attenuation data for the different freouencies are shown in Fig. 2. As we see, most of the points lie on a single curve implying that all the attenuation data predict a similar temperature behavior of the energy gap.

All the attenuation data on the single niobium specimen of residual resistance ratio 7000 point to a primary energy gap $\Delta_d(0)$ equal to $(2.0 \pm 0.2)k_BT_c$. This value for the primary gap is in good agreement with the value $\Delta_{d}(0)=(1.95\pm0.02)k_{B}T_{c}$ obtained by MacVicar and Rose¹⁵ from some tunneling measurements done on niobium superconductors. The ratio between the values of the two energy gaps is the same as that obtained by other tunneling $measures¹⁶$ on niobium superconductors and by a two-band analysis of the specific-heat measure
ments on niobium superconductors.¹⁷ ments on niobium superconductors.¹⁷

It should be noted, however, that the temperature behavior implied in Fig. ² and the value of the primary energy gap do not agree with the BCS prediction even though the two-band model⁷ predicts that the d -band energy gap should be BCS-like. We believe that the non-BCS-like temperature behavior is most likely due¹⁸ to the anisotropy of the d -band Fermi surface and that the discrepancy between our value for the energy gap and the BCS value is the result of the strong-coupling nature of the niobium superconductor. However, we do not rule out the possibility that our neglect of the temperature dependences on the electronic mean free paths may be the cause of the observed deviation from the BCS predictions. This possibility can only be checked our when a theory for the temperature dependences of the electronic mean free paths in a two-band superconductor is formulated. (We expect that the temperature dependences will be more complicated than the temperature dependence proposed by Maki¹⁹ for the mean free path in an oneband superconductor.)

- 1128 (1971).
- 7 H. Suhl, B. T. Matthias, and L. R. Walker, Phys. Rev. Lett. 6, 677 (1961).
- 8 I. M. Tang, Phys. Rev. B 2, 2581 (1970).
- $9M.$ Nusair, M. S. thesis (American University of Beirut, 1973) (unpublished).
- 10 The reason for making this transformation is that as the sound waves travel through the lattice, the Bloeh periodicity of the lattice is destroyed. This means that the rest frame of the unperturbed lattice ean no longer support the Bloch states and we must, therefore, trans-

form to the frame in which Bloch's theorem holds, i. e. , the moving frame of the lattice. This amounts to a canonical transformation of the system in which the nodal

- surfaces of the sound waves are fixed. $¹¹I$. I. Falko and L. P. Kadanoff, Phys. Rev. 136 , A1170</sup> (1964).
- 12 T. Tsuneto, Phys. Rev. 121, 402 (1961); M. Levy,
- Phys. Rev. 131, 1497 (1963); L. T. Claiborne and R. W. Morse, Phys. Rev. 136, 893 (1964).
- 13 F. Carsey, R. Kagiwada, M. Levy, and K. Maki, Phys. Rev. B 4, 854 (1971).
- 14 This modification was made in Ref. 2.
- ¹⁵M. L. A. MacVicar and R. M. Rose, J. Appl. Phys. 39, 1721 (1968).
- '6J. W. Hafstrom and M. L. A. hiacVicar, Phys. Rev. B 2, 4511 (1970).
- $17C$. C. Sung and L. Y. L. Shen, Phys. Lett. 19, 101 (1965}.
- 18 If an anisotropy exists in the Fermi surface, the energy gap observed in the ultrasonic-attenuation measurements would be an averaged energy gap. At the lower temperatures, the smaller-energy-gap regions would contribute more heavily. Thus the apparent energy gap will be smaller when $T \ll T_c$ than would be expected from its value when $T \leq T_c$. See, R. D. Parks, Superconductivity (Marcel Dekker, New York, 1969), Chap. 4.
- ¹⁹K. Maki, Prog. Theor. Phys. 41, 586 (1969).