INVESTIGATION OF THE TUNNEL EFFECT IN LEAD AND THALLIUM UNDER PRESSURE

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A study was made of the usefulness of the tunnel effect in investigations of the superconductivity under pressure. Experimental results were obtained for lead and thallium films in superconductor-barrier-superconductor (S-I-S) tunnel systems at pressures up to 14 kbar. Data were obtained on the energy gap and characteristic frequencies of the phonon spectrum. Thick ($d > \xi_0$) films of lead, exhibiting a gap anisotropy, were investigated. It was found that this anisotropy was not greatly affected by the applied pressure.

1. INTRODUCTION

THE principal properties of the superconducting state can be described by the idealized Bardeen-Cooper-Schrieffer model[1] with an isotropic Fermi surface and an isotropic energy gap, which is in good agreement with numerous experimental results. However, there are discrepancies between the experimental data and this model. They include, for example, a structure in the tunnel density of states of some superconductors,[2] an energy gap anisotropy,[3] a departure from the law of corresponding states observed for many superconductors ($2\Delta/kT_c \neq 3.52$), etc. Recently started investigations of the tunneling of electrons through lead under pressure have already resulted in the discovery of a change in $2\Delta/kT_c$, caused by the compression of the lead, which is a superconductor with a strong electron-phonon interaction.[4-6]

The use of the tunnel effect in studies of the superconductivity under hydrostatic pressure opens up new possibilities because the tunneling of electrons can give detailed information on the microscopic properties of a superconductor. It is known that the differential conductivity of a tunnel contact is directly proportional to the density of states in a superconductor:

$$\sigma=\left(\frac{dI}{dU}\right)_S \Big/ \left(\frac{dI}{dU}\right)_N=\frac{N_S}{N_N}=\mathrm{Re}\frac{|\omega|}{[\omega^2-\Delta^2(\omega)]^{1/2}}, \qquad (1)$$

where N_S and N_N are the densities of states in the superconducting and normal states, $\Delta\omega$ is a complex energy-gap parameter,[7] and all the energies ω are measured from the Fermi level. Developments of the theory of tunneling have established[8-10] that the critical points in the phonon spectrum may be resolved easily as singularities in the second derivatives of the conductivity, i.e., in the dependence of $d\sigma/dU$ on U. The influence of the phonon spectrum on the superconducting density of states has been demonstrated convincingly in several experiments.[2 11-13] Rowell and McMillan[14] demonstrated that the tunnel-effect data can be used to find the function $\alpha^2(\omega)F(\omega)$, where $\alpha^2(\omega)$ is the intensity of the electron-phonon interaction and $F(\omega)$ is the energy distribution function of phonons. Finally, the experiments of Zavaritskiĭ[15] have shown that the tunnel effect approach is fruitful in investigations of the energy gap anisotropy.

The greatest sensitivity to changes in the energy spectrum is exhibited by tunnel systems of the superconductor-barrier-superconductor (S-I-S) type.[7] The present authors investigated such systems and the effect of pressure on the energy gap of superconducting films of lead, tin, indium, and thallium.[4,16,17]

The present paper reports new results obtained in an investigation of the tunnel density of states in lead and the first data resulting from a study of the gap anisotropy of lead and of the phonon spectrum of thallium under pressure.

2. EXPERIMENTAL TECHNIQUE

The technique used in the fabrication of thin-film Al-I-Pb or Al-I-Tl tunnel diodes was described in our earlier papers.[4,17] In contrast to the investigation reported in[4], the lead was evaporated onto a glass substrate kept at room temperature. This was particularly important in the observation of the anisotropy effects which were sensitive to the scattering by grain boundaries. An improvement in the technique of the preparation of the thallium diodes enabled us to store these diodes for a long time (5–10 days) at room temperature. This procedure ensured that the phonon singularities in the tunnel characteristics were clearly revealed; it also improved the quality of a barrier from the point of view of its suitability in investigations under pressure. The initial resistances of the junctions (at P = 0) were within the range 50–500 Ω.

The tunnel density of states and the phonon singularities were investigated using a modulation bridge method, similar to that described in[18]. The phonon effects were recorded at modulation levels amounting to 40–150 μV and the gap phenomena were recorded at levels of 1–10 μV. The modulation frequencies were 383 and 766 Hz. All the characteristics were plotted by an X-Y automatic recorder of the ÉPP-09 type. The static bias across a sample was measured by a high-resistance potentiometer to within ~1 μV.

The high pressures were produced in a bomb filled with a kerosene-oil mixture[19]; they were measured using the shift of the critical temperature of an indium

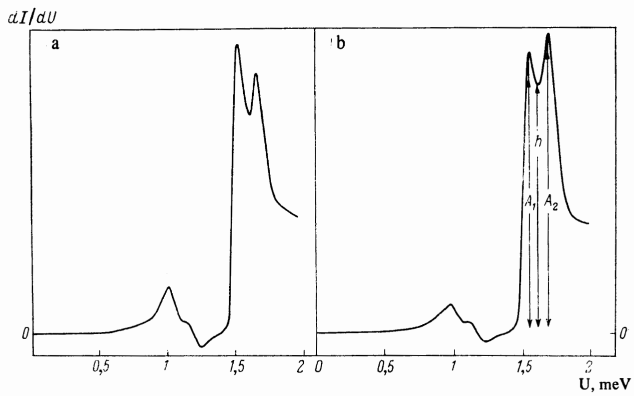

FIG. 1. Differential conductance of an Al-I-Pb* contact with a thick (~15000Å) film of lead (Pb*) at T = 1.17°K: a) $A_1/A_2 > 1$, $2\Delta_1$ = 2.55 meV, $2\Delta_2$ = 2.83 meV; b) $A_1/A_2 < 1$, $2\Delta_1$ = 2.49 meV, $2\Delta_2$ = 2.78 meV.

manometer.[17] The bomb containing the samples was cooled directly with liquid helium.

3. ENERGY GAP AND PHONON SPECTRUM OF LEAD

Although a direct proof of the anisotropy of the energy gap of superconductors was provided over a decade ago,[20] a complete understanding of this anisotropy has not yet been achieved[3], consequently detailed experimental and theoretical investigations of anisotropic superconductors are still desirable. Our aim was to obtain additional information on the gap anisotropy through experiments on anisotropic lead under pressure. The Fermi surface[21] and the phonon spectrum[22] of this metal have been investigated in detail. Bennett[23] suggested a theory of the anisotropy of the energy gap of superconducting lead, based on the assumption that the gap anisotropy was due to the anisotropy of the phonon spectrum. The fullest information on the gap anisotropy could be obtained by investigating single crystals. However, the anisotropy effects should be observed also in thick pure polycrystalline films if the mean free path of electrons is considerably longer than the coherence length, $l > \xi_0$.[23,24] This case was investigated experimentally by Campbell and Walmsley[25], and by Rochlin.[26]

A. Singularities in the Tunnel Conductance of Thick Films at Zero Pressure

An investigation was made on about 50 lead films, which were ~15 000 Å thick. The tunnel characteristics of the junctions exhibited clearly the singularities ($\Delta_{Pb} \pm \Delta_{Al}$) associated with the presence of two gaps Δ_1 and Δ_2 in the energy spectrum of lead (Fig. 1). The nature of the singularities in the characteristics $(dI/dU) = f(U)$ could be used to divide the samples investigated into two arbitrary groups: the first and larger group comprised those samples (~70% of all the samples) for which the ratio of the amplitudes at the singularities was $A_1/A_2 > 1$; the second group consisted of those samples for which this ratio was $A_1/A_2 < 1$. The structure of the characteristics at $eU = \Delta_{1,2}^{Pb} \pm \Delta_{Al}$ could be attributed to the presence of two groups of electrons with velocities normal to the barrier surface. Then, the ratio A_1/A_2 should be a measure of the contribution of a given group of electrons to the tunnel current. The value of A_1/A_2 was found to vary from sample to sample, ranging from a maximum of 1.35 to a minimum of 0.833. The best resolution of the singularities, i.e., high values of the ratio $A_{1,2}/h$ (h is the amplitude at the minimum between the two peaks), was achieved for lead films evaporated at a low rate (1–3 Å/sec) and annealed at 100°C for several hours after the fabrication of a junction. The maximum values of the ratio $A_{1/2}/h$ for these films were $(A_1/h)_{max} = 2.52$ and $(A_2/h)_{max} = 1.91$. The structure at $eU = \Delta_{1,2}^{Pb} - \Delta_{Al}$ was practically unresolved for samples with $A_{1,2}/h < 1.05$, which was explained satisfactorily by the theory due to Gorbonosov and Kulik,[24] according to which the amplitude of the singularities should shrink with decreasing temperature and should be

$$\left(1+\mathrm{th}\frac{\Delta_{Al}}{2T}\right)\Big/\,2\left(1-\mathrm{th}\frac{\Delta_{Al}}{2T}\right)$$

times smaller than the corresponding singularity at $eU = A_{1,2}^{Pb} + \Delta_{Al}$ (in our case, the two amplitudes differed by a factor of about 10).

The extremal values of the energy gaps, found from the dependences $(dI/dU) = f(U)$, were 2.49 ± 0.01 and 2.57 ± 0.01 meV for $2\Delta_1$, and 2.78 ± 0.01 and 2.85 ± 0.01 meV for $2\Delta_2$. As a rule, the samples belonging to the first group had larger values of Δ_1 and Δ_2. The results of the numerical calculations of Bennett[23] for polycrystalline samples (see Fig. 17a in[23]) were in satisfactory agreement with our data.

The theory[23,24] predicts singularities in the tunnel conductivity between polycrystalline films, due to the presence of critical points in the dependence $\Delta(n)$: there should be a local maximum, a minimum, and a saddle point. However, this theory is based on a model which presumes that tunnel transitions take place in films consisting of randomly oriented grains, so that the orientations of the momenta p and q with respect to the crystallographic axes are different. This case is difficult to realize experimentally because thick films deposited on various substrates exhibit a tendency to have a preferential orientation.[27] In particular, lead films deposited on glass are oriented preferentially along the [111] axis, perpendicular to the substrate surface.[28,29] We were able to prepare only two junctions, deposited simultaneously on the same substrate, which exhibited a complex structure (Fig. 2) similar to that predicted theoretically and which had the following critical points for 2Δ: 2.4, 2.49, 2.67, 2.78, and 2.92 meV.

B. Effect of Pressure

The samples with high values of the ratio $A_{1,2}/h$ were placed in the bomb and the measurements were carried out at high pressures. Figure 3 shows the $I = F(U)$ and $(dI/dU) = f(U)$ characteristics recorded at various pressures for the same sample. The results of a study of the influence of pressure on the energy gaps of lead are presented in Table I and in Fig. 4. At the maximum pressure employed, the difference $\Delta_2 - \Delta_1$ decreased slightly (see the inset in Fig. 3b):

$$eU = (\Delta_2-\Delta_1)_{P=0} - (\Delta_2-\Delta_1)_{p=14\,\mathrm{kbar}} = 16 \pm 4\ \mu\mathrm{V}.$$

The samples were prepared in such a way that the same substrate carried thick- and thin-film junctions (the configuration of the samples is shown in Fig. 3a). This enabled us to obtain, in the same experiment, additional information on the isotropic energy gap $2\Delta_{av}$. This method eliminated the error in the determination of the pressure when the results for the isotropic and anisotropic components of the gap were compared. At all pressures, we found that $\Delta_{av} = (\Delta_1 + \Delta_2)/2$. The pressure-induced change in the energy gap of thin films was in agreement with our earlier results.[4]

The features at $eU > \Delta_{Al} + \Delta_{Pb}$, associated with the phonon spectrum of the superconductor, were investigated using thin-film (~1000 Å) superconducting junctions. An investigation of the S-I-S system enabled us to resolve (and obtain information on) three peaks in the phonon spectrum obtained for the transverse polarization under pressure (Fig. 5). Table I gives the results of the present investigation, as well as the results obtained by others.[5,6,30] It is evident from Table I that, basically, the various tunnel-effect data are in good agreement.

The main results of the theory of anomalous superconductors due to Geĭlikman and Kresin[31]

$$\frac{2\Delta}{kT_c} = 3{,}52\left[1 + 5.3\frac{T_c^2}{\omega_0^2}\ln\frac{\omega_0}{T_c}\right], \tag{2}$$

and of the theory due to McMillan[32]

$$T_c = \frac{\omega_c}{1.73}\exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right] \tag{3}$$

can be applied to the experimental results.[6] For convenience, we can rewrite Eqs. (2) and (3) in the differential form:[33]

$$\frac{d\ln T_c}{dP} = \left[\frac{d\ln 2\Delta}{dP} + 5.3\frac{d\ln\omega_0}{dP}\cdot\left(2\ln\frac{\omega_0}{T_c} - 1\right)\frac{T_c^2}{\omega_0^2}\right]\left[1 + 5.3\frac{T_c^2}{\omega_0^2}\times\left(2\ln\frac{\omega_0}{T_c} - 1\right)\right]^{-1}, \tag{4}$$

$$\frac{d\ln T_c}{dP} = \frac{d\ln\omega_c}{dP} + \frac{1.23}{(\lambda - 0{,}11)^2}\cdot\sum_{\nu}^{3}\lambda_\nu\left(\frac{d\ln I_\nu}{dP} - 2\frac{d\ln\omega_\nu}{dP}\right). \tag{5}$$

Here, ω_0 is the characteristic frequency of phonons (ω_t or ω_l); ω_c is the cutoff frequency; λ is the elec-

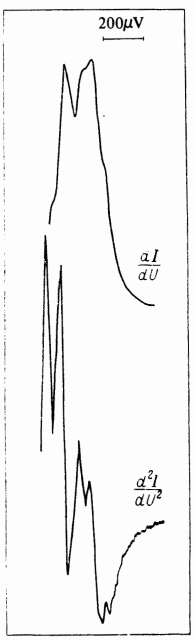

FIG. 2. Complex structure of the (dI/dU) = f(U) and (d^2I/dU^2) = F(U) characteristics of an Al-I-Pb* sample in the vicinity of the sum of the energy gaps $\Delta_{Pb} + \Delta_{Al}$, T = 1.16°K.

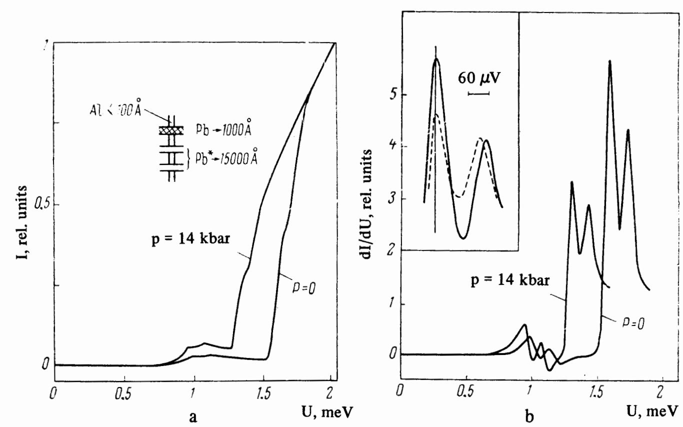

FIG. 3. Influence of high pressures on the characteristics of an Al-I-Pb* sample: a) I = f(U); b) (dI/dU) = F(U), T = 1.17°K. The insert shows the pressure-induced shift $\Delta_2^{Pb} - \Delta_1^{Pb}$ (the dashed curve corresponds to a pressure of 14 kbar).

Table I. Results of investigations of the tunnel effect in lead under pressure

	Investigated quantity X		
	$2\Delta_1$	$2\Delta_{cp}$	$2\Delta_2$
X, meV	2.56±0,01	2.72±0.01 2.71 [6]	2.85±0.01
dX/dP, 10^{-5} meV/bar	—2.22±0,15	—2,34±0.15 —2.16±0.2 [6]	—2.45±0.15
$d\ln X/dP$, 10^{-6} bar^{-1}	—8.7±0,6	—8.6±0.6 —10.1±0.8 [5] —8 [6]	—8.6±0.6

	Investigated quantity X			
	ω_t'	ω_t''	ω_t'''	ω_l
X, meV	3.88±0.02	4.52±0.02 4,45 [6] 4,6 [30]	4.9±0,02	8.55±0,02 8,5 [6]
dX/dP, 10^{-5} meV/bar	2.6±0.4	3.3±0.4 4.5±0.6 [6]	3.3±0,4	6,4±0.4 6±0.6 [6]
$d\ln X/dP$, 10^{-6} bar^{-1}	6,75±1	7,28±1 5.3±0,7 [30] 10.1 [6]	6.75±1	7.5±0.5 7±0.7[30] 7,05 [6]

tron-phonon interaction constant:

$$\lambda = \sum_{\nu}^{3} 2 \int_{0}^{\omega_c} \alpha_\nu^2(\omega) F_\nu(\omega) \frac{d\omega}{\omega},$$

and I_ν is a function which depends weakly on the phonon frequencies and pressure:[33]

$$d \ln I_t / dP = -0.1 \cdot 10^{-6}\ \text{bar}^{-1},$$
$$d \ln I_t / dP = 1.9 \cdot 10^{-6}\ \text{bar}^{-1}.$$

The results of the calculation of $d(\ln T_c)/dP$ made using Eqs. (4) and (5) as well as the data on the pressure-induced changes in the gap ($2\Delta_{av}$) and in the phonon frequencies (Table I), yield:

	$d \ln T_c/dP$, bar^{-1}
Eq. (4), $\omega_0 = \omega_t$:	$-4.95 \cdot 10^{-6}$
Eq. (4), $\omega_0 = \omega_l$:	$-6,9 \cdot 10^{-6}$
Eq. (5):	$-7,6 \cdot 10^{-6}$

Independent experiments give:

	[30]	[34]	[35]
$d\ln T_c/dP$, 10^{-6} bar^{-1}:	$-4,9$	$-5,37$	-6.75

Bearing in mind the approximate nature of the models used in[31,32], we find that the agreement between the calculated and experimental values is satisfactory. The experimentally determined values of $d(\ln 2\Delta_{1,av,2})/dP$ are equal, within the limits of the experimental error, which indicates that the phonon spectrum plays the dominant role in the change of $2\Delta/kT_c$ of lead under pressure.

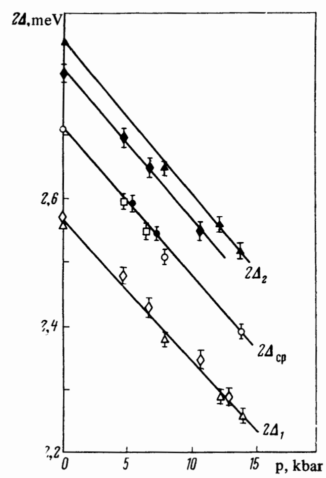

FIG. 4. Influence of high pressures on the energy gaps of superconducting lead. Different samples are represented by different symbols.

4. ENERGY GAP AND PHONON SPECTRUM OF THALLIUM

An investigation of the influence of pressure on the energy gap of thallium films was reported in[17]. The higher quality of our samples made it possible to extend somewhat the range of pressures and to carry out measurements of the shift of the characteristic frequencies in the phonon spectrum of this metal. The maximum value of R(300°K)/R(4.2°K) did not exceed 22 for any of the investigated films. The width of the superconducting transition, δT_c, ranged from 0.005 to 0.02°K. Prolonged annealing at room temperature improved considerably the superconducting characteristics of the films and the barrier properties of the tunnel diodes. A special feature of the films under pressure was the absence of a rise in T_c at low pressures, which should be typical of the bulk material.

The energy gap was determined from the $(dI/dU) = f(U)$ characteristics (Fig. 6). The maximum change in the gap at 13 kbar was $\Delta(P)/\Delta(0) = 3.60 \pm 1.7\%$ and the change in the critical temperature was $T_c(P)/T_c(0) = 3.5 \pm 0.5\%$. Thus, in the investigated range of pressures, the ratio $2\Delta/kT_c$ for thallium films remained constant within the limits of the experimental error.

Since the thallium junctions suitable for high-pressure measurements were prepared by a process which included the evaporation of thallium onto a cold substrate[17] and its subsequent annealing at room temperature, we determined the dependences $\sigma = f(U)$ and $(d^2U/dI^2) = F(U)$ for a large number of samples at zero (atmospheric) pressure. We found that the principal peaks in the phonon spectrum of thallium (Fig. 7) were located at $\omega_t = 3.99 \pm 0.03$ meV, $\omega_l = 9.5$

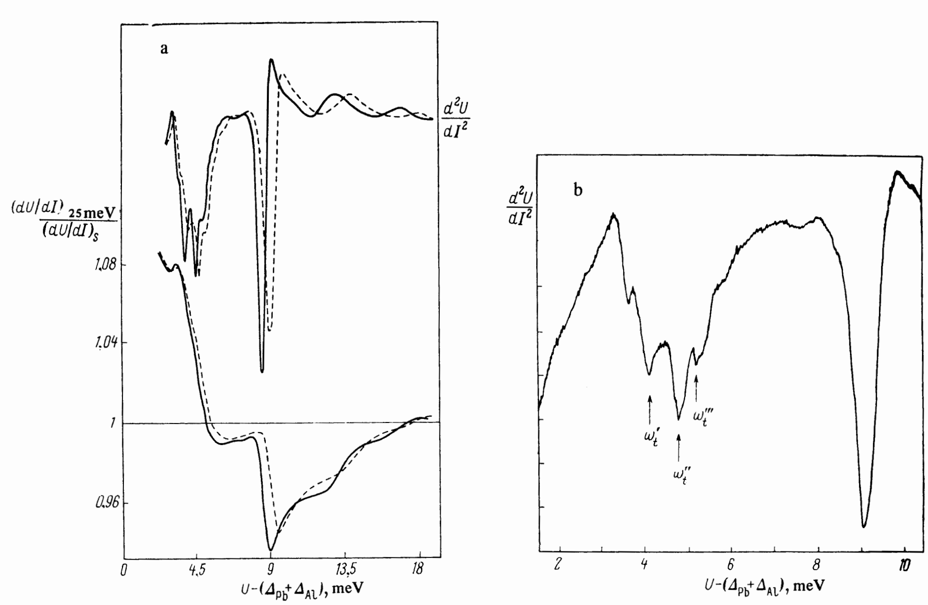

FIG. 5. a) Dependences of the characteristics $(dU/dI)_{25meV}/(dU/dI)_S$ and (d^2U/dI^2) on U for an Al-I-Pb sample at various pressures: the continuous curves correspond to P = 0 and the dashed curves to P = 8 kbar; the amplitude of (d^2U/dI^2) is normalized to the amplitude of ω_t. b) Actual trace of the second-harmonic voltage at P = 8 kbar for the same sample as in Fig. 5a; T = 1.17°K.

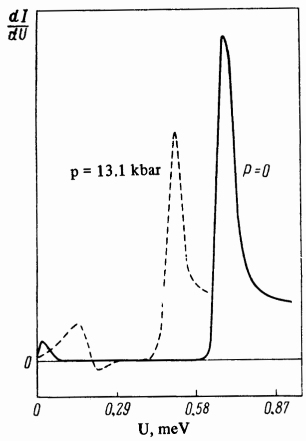

FIG. 6. Influence of high pressures on the differential conductance of an Al-I-Tl sample; T = 1.17°K.

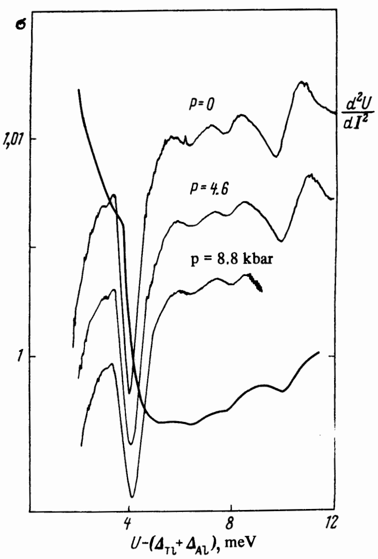

FIG. 7. Normalized conductance of an Al-I-Tl sample at P = 0 and the $(d^2U/dI^2) = f(U)$ characteristics at various pressures; T = 1.17°K.

± 0.03 meV, in good agreement with the results of Rowell and Kopf[2] and those of Clark.[36] The shift of the longitudinal vibrations ω_l was determined up to 5 kbar and the shift of the transverse vibrations ω_t was determined up to 9 kbar (Fig. 7). The values given in Table II are the averages of measurements of four samples. In contrast to lead, the shift was very weak for the transverse phonons ω_t. The values of $d(\ln \omega_\nu)/dP$ could be used to estimate the lattice Grüneisen constant γ_{ω_ν}. Using the tabulated compressibility of thallium, $\kappa = 2.77 \times 10^{-6}$, given in the review by Brandt and Ginzburg,[37] we found that $\gamma_{\omega_t} = 1.35$ and $\gamma_{\omega_l} = 2.1$. Obviously, these estimates were very rough since, to our knowledge, the experimental data on the compressibility of thallium (particularly thallium films) at low temperatures were not reliable.

5. CONCLUSIONS

By using the tunnel effect in investigations of superconductors under pressure, we were able to supplement the traditional measurements of $T_c(P)$ and $H_c(P)$ with the data on the shift of the energy gap and of the phonon spectrum under pressure. A theory of superconductors

Table II. Pressure-induced changes in the phonon frequencies and critical temperature of thallium films

	Investigated quantity X		
	ω_t	ω_l	T_c
X, meV	3.99	9.50	2.38° K
dX/dP, 10^{-5} meV/bar	1.48±0.7	5.45±1	$-(0.6\pm0.1)\cdot 10^{-5}$ deg/bar
$d\ln X/dP$, 10^{-6} bar^{-1}	3.7	5.75	$-2.52\cdot 10^{-6}$ deg/bar

with a strong electron-phonon coupling could explain satisfactorily the observed change in $2\Delta/kT_c$ for Pb as being due to a change in the phonon spectrum. It was found that the energy gap anisotropy of lead was basically unaffected in the investigated range of pressures. In principle, the tunnel effect could be used to determine directly the frequency dependence of the lattice Grüneisen constant:[38]

$$\gamma_{\omega_\nu} = \frac{1}{\varkappa}\frac{d\ln\omega_\nu}{dP}.$$

However, the experimental error in the determination of $d\omega_\nu/dP$ (Tables I and II) for lead and thallium was too large to allow us to separate the contributions of the long-wavelength (ω_t) and short-wavelength (ω_l) vibrations to the Grüneisen constant γ.

The results could be improved by the use of symmetrical superconducting tunnel junctions and very low temperatures. It would also be interesting to investigate the tunnel effect in bulk materials under pressure.

The use of high pressures meant that the tunnel contacts had to satisfy more than usually stringent requirements. Our investigation demonstrated the full "compatibility" of the electron tunneling method with the high-pressure technique. Difficulties were encountered, particularly in the changes and irreversibility of the contact conductance at pressures above 15 kbar; these effects were due to the deformation of the potential barrier and the participation of other (in addition to the tunnel effect) conduction mechanisms. However, these technical difficulties should not be insuperable. The search for new barrier materials and further improvements in the technology of the fabrication of aluminum oxide barrier diodes should stimulate investigations of the tunnel effect in superconductors under pressure.

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10