

Temperature dependence of the softening of pure lead during the superconducting transition

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The temperature dependence of the creep discontinuity $\delta\epsilon_{ns}$ and the ratio $\dot{\epsilon}_s/\dot{\epsilon}_n$ of the plastic deformation rates in the *S* and *N* states, in the superconducting transition of lead single crystals (99.999%) are investigated in the 1.6–7.2°K temperature range and in a wide deformation range. It is found experimentally that the nature of the temperature dependence of $\delta\epsilon_{ns}$ and the creep-rate ratio in the *S* and *N* states depend on the degree of deformation, i.e., on the structure imperfections of the crystal. Comparison with existing theories of softening yields satisfactory agreement with the fluctuation theory under the assumption of a one-band isotropic superconductor.

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1. INTRODUCTION

The discovery that metals become more plastic on going to the superconducting state^[1] has led to a large number of experimental and theoretical studies aimed at explaining the physical nature of this effect.^[20] These investigations have led to the observation of the dependence of the increased plasticity on various parameters such as the temperature, impurities, the deforming stress, the dislocation structure, etc.^[5–14] Particularly diligent investigations were made of the temperature dependence of the increased-plasticity effect. Their results turned out to be contradictory. Some workers obtained for the effect a temperature dependence close to that observed for the energy gap $\Delta(T)$,^[8] others obtained a dependence closer to that of the density of the superconducting electrons $\rho_S(T)$,^[13] and some of the experimental studies can be classified with equal degree of accuracy as leading to either of the aforementioned dependences. The cause of such a disparity between the experimental data should apparently be sought in the factor that different workers carried out their research under conditions that were not comparable. As a rule, the measurements were made in different temperature intervals, at different levels of the deforming stress, and on crystals with different impurity compositions, different orientations, and different defect structures. These factors, as follows from the theory, can significantly affect the magnitude and the behavior of the increased-plasticity effect.

The purpose of the present study was to investigate in detail the behavior of the flow-strain jump $\delta\epsilon_{ns}$ in the superconducting transition in a wide range of temperatures and deformations (up to fracture of the sample), to obtain the temperature dependence of the ratio $\dot{\epsilon}_s/\dot{\epsilon}_n$, of the rates of plastic deformations in the *S* and *N* states, and to compare the experimental results with those that follow from different theories of the increased-plasticity.

2. EXPERIMENTAL PROCEDURE

The measurements were made on pure lead (99.9999%) single crystals. To avoid effects of the orientation and to have similar dislocation structures, the single crystals were grown in batches of 10 each simultaneously in the same mold and with the same primer.^[21] The same primer was used to grow all the succeeding single crystals. As a result, all the investigated single crystals had the same orientation (the sample axis was between the

directions [100] and [110]), an identical impurity composition, and nearly the same defect structure. The reproducibility of the measurement data for different single crystals was therefore high enough and exceeded noticeably the reproducibility usually obtained in measurements of structure-sensitive properties with different samples. We tested 60 single crystals.

We measured the increment $\delta\epsilon_{ns}$ of the flow strain and the ratio of the flow rates at the instant of the transition of the crystal from the normal to the superconducting state, $\dot{\epsilon}_s/\dot{\epsilon}_n$, and the dependence of these quantities on the degree of crystal deformation at various temperatures in the interval 1.6 to 7.2°K. In the same temperature interval we obtained the strain hardening curves $\kappa(\epsilon)$, which are needed to find the hardening coefficients $\tau(\epsilon)$ of the investigated crystals, and we also measured the activation volumes $V(\epsilon)$ of the flow process. The sample was deformed by tension.

The transition from the normal to the superconducting state and back was effected by turning off and on a magnetic field produced by a superconducting solenoid in which the deformed sample was placed.

To obtain the relation $\delta\epsilon_{ns}(\epsilon)$ at a given degree of deformation, a stress increment $\Delta\tau = 20 \text{ g/mm}^2$ was used to obtain the next flow curve in the normal state. Then, a minute later, the sample was transformed from the normal to the superconducting state and the strain increment $\delta\epsilon_{ns}$ due to this transition was measured. The magnetic field was then turned on the sample made normal, the deformation of the crystal was increased 1–2%, the sample was made superconducting, and the flow-strain jump $\delta\epsilon_{ns}$ was measured. These measurements were repeated many times along the entire hardening curve from the yield point to the sample fracture, and the function $\delta\epsilon_{ns}(\epsilon)$ was plotted.

Measurement of the strain was automatic using an inductive pickup, appropriate amplification apparatus, and an electronic automatic potentiometer. The relative strain was measured with accuracy 2×10^{-5} .

When measuring the ratio $\dot{\epsilon}_s/\dot{\epsilon}_n$ of the flow-rates in the *S* and *N* states we used the same method of stepwise loading, except that the transition from the normal to the superconducting state was effected when the flow rate in the normal state reached a definite value, which was always the same. The rate was determined from the slope of the tangent to the flow curve.

The strain-hardening curves $\tau(\epsilon)$ were also plotted by the stepwise loading method, but the sample was always in the normal state during the course of deformation.

The dependence of the activation volume $V(\epsilon)$ on the degree of deformation was measured by a differential procedure described in [22]. To this end, as the crystal deformation was increased, every 2–3% we produced a flow whose rate was changed from $\dot{\epsilon}_1$ to $\dot{\epsilon}_2$ via a small increment of the deformation stress, by an amount $\Delta\tau = 2 - 3 \text{ g/mm}^2$. To calculate the activation volume we used the expression

$$V = kT \left[\frac{\ln(\dot{\epsilon}_2/\dot{\epsilon}_1)}{\Delta\tau} \right], \quad (1)$$

where $\dot{\epsilon}_1$, $\dot{\epsilon}_2$, and $\Delta\tau$ are the quantities indicated above, k is Boltzmann's constant, and T is the temperature of the crystal. The activation volume was measured with samples in the normal state.

Temperatures in the interval 1.6–4.2°K were obtained by pumping off liquid helium. To obtain temperatures from 4.2 to 7.2°K we used the following procedure. An additional thin-wall metallic dewar was placed inside a helium dewar on the immobile rod of the testing machine. The level of the liquid helium was somewhat lower than the upper edge of this dewar. The working volume with the sample, the superconducting solenoid, and the heater were thermally insulated. The sample temperature was raised by quasiadiabatic heating of the working volume to the required value, using a stabilizing feedback system. The temperature was measured with a semiconducting pickup with accuracy 0.01°K, and the temperature stability within this interval was not lower than 0.025°K.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows plots of $\delta\epsilon_{NS}$ against the degree of deformation of the crystal at various temperatures in the interval 1.6–7.2°K. Each curve is plotted from the measured strain jump in three single crystals. We see that the curves have a complicated stagewise-developed character, which reflects, as shown earlier in [23], the stagewise character of the hardening curve

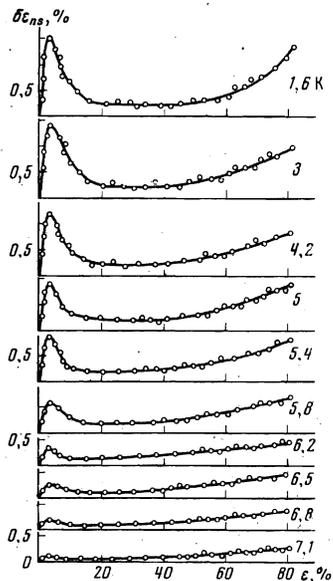


FIG. 1. Dependence of the flow-strain jump $\delta\epsilon_{NS}$ in the N-S transition on the degree of deformation in the temperature intervals 1.6 – 7.2°K.

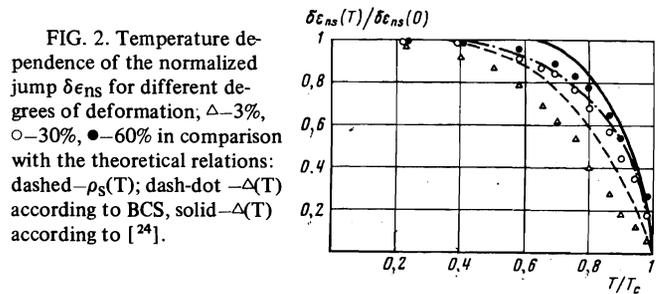


FIG. 2. Temperature dependence of the normalized jump $\delta\epsilon_{NS}$ for different degrees of deformation; Δ —3%, \circ —30%, \bullet —60% in comparison with the theoretical relations: dashed— $\rho_S(T)$; dash-dot— $-\Delta(T)$ according to BCS, solid— $-\Delta(T)$ according to [24].

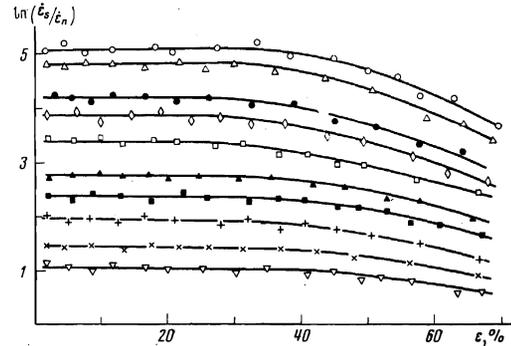


FIG. 3. Plot of $\ln(\dot{\epsilon}_S/\dot{\epsilon}_N)$ against the degree of crystal deformation at fixed temperatures: \circ —1.6 K; Δ —3 K; \bullet —4.2 K; \diamond —4.6 K; \square —5 K; \blacktriangle —5.4 K; \blacksquare —5.8 K; $+$ —6.2 K; \times —6.5 K; ∇ —6.8 K.

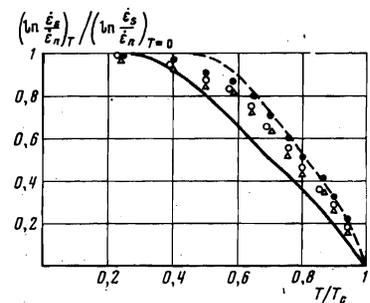


FIG. 4. Temperature dependence of $\ln(\dot{\epsilon}_S/\dot{\epsilon}_N)$ normalized to 0°K, for a deformation of 3% (Δ), 30% (\circ), and 60% (\bullet) as compared with the functions $\varphi'(T/T_C)$ of the theory [16] for $L = 100L_0$ (solid line) and $L = 10L_0$ (dashed).

of lead single crystals. It follows from the figure that raising the temperature leads to a decrease of the jump $\delta\epsilon_{NS}$ during all the deformation stages. From the curves of Fig. 1 for the deformations 3%, 30%, and 60%, corresponding to different stages of the hardening curve, we plotted the jump of the flow strain normalized to the extrapolated value of $\delta\epsilon_{NS}$ at 0°K, on the relative temperature T/T_C , which is shown in Fig. 2. We present here also the theoretical temperature dependence of the energy gap $\Delta(T)$ according to BCS, the experimental plot of $\Delta(T)$ for lead, [24], and the dependence of the density of the superconducting electrons $\rho_S(T)$.

Figure 3 shows plots of $\ln(\dot{\epsilon}_S/\dot{\epsilon}_N)$ on the degree of the deformation, obtained at various temperatures. It is seen that at low deformation the ratio $\dot{\epsilon}_S/\dot{\epsilon}_N$ of the flow rates in the S and N states depends little on the degree of deformation. An increase of the temperature and of the degree of deformation leads to a decrease of this ratio. From the curves of Fig. 3 for the same deformations we plotted $\ln(\dot{\epsilon}_S/\dot{\epsilon}_N)$ normalized to the value of $\ln(\dot{\epsilon}_S/\dot{\epsilon}_N)$ at 0°K, against the relative temperature T/T_C , as shown in Fig. 4. It is seen from Figs. 2 and 4 that the temperature dependence of the softening effect, remaining monotonic in all cases, is significantly different for different degrees of sample deformation, and that this difference exceeds the limits of experimental error. This indicates that the character of the temperature dependence of the

softening effect is strongly influenced by the defect structure of the crystal.

It was observed in^[10] that the temperature dependences of $\delta\epsilon_{NS}$ obtained at various stresses in polycrystalline indium and lead did not coincide. A similar result was obtained in^[25] in a measurement of $\delta\epsilon_{NS}$ of single crystals of very pure lead. In contrast to our present result, and also to the results by others,^[7-9, 12-14] the temperature dependences for lead in^[10] had a nonmonotonic character with a maximum at $T/T_c = 0.35$. The threshold stress, starting with which this nonmonotonicity was observed, amounted to $\sim 4 \text{ kg/mm}^2$. In our present investigations, however, even though the deforming stress exceeded 4 kg/mm^2 , no nonmonotonicity was observed in the temperature dependence of $\delta\epsilon_{NS}$. The discrepancy in the results of^[10] and other studies cannot be explained at present.

Let us compare the obtained experimental data with the existing theories of metal softening in the superconducting transition. In first papers on this phenomenon it was suggested that the effect is due to a decrease in the electron dragging of the dislocation as the metal goes into the superconducting state, and this leads to an increase of the dislocation velocity, i.e., the effect is purely dynamic. However a change of the strain rate of lead single crystals by a factor 4.4×10^3 did not lead to a noticeable change of $\delta\sigma_{NS}$,^[4, 7] in sharp contrast to one of the main conclusions of the dynamic theory, according to which the increased plasticity is proportional to the strain rate. Similar results were obtained in the deformation of indium,^[7, 12] niobium^[11], and the alloys Pb-Tl, Pb-Cd, and Pb-Sn.^[26] The weak dependence of the stress jump $\delta\sigma_{NS}$ on the strain rate of lead, observed in the superconducting transition by Fomenko^[14] at 1.65°K ($\delta\sigma_{NS}$ changed by a factor 1.6 when ϵ was changed by more than two orders of magnitude) does not prove the correctness of the dynamic theory, since the relation obtained in^[14] is weaker by many times than that predicted by the theory, and was observed only at very low temperatures. It must be borne in mind that a direct connection between the jump of the electron drag force and the magnitude of the softening exists in the dynamic theory only in those cases when the dislocations move with large velocities, at which their kinetic energy exceeds the potential barriers in the crystal. This purely dynamic situation is realized in plastic deformation apparently extremely rarely, the only exception being the active deformation at high velocities and large levels of the the deforming stress, and in experiments with pulsed loading of the crystals.

The flow for which the increased-plasticity effect in the N-S transition is clearly pronounced has patently a fluctuating behavior both in the normal and in the superconducting state, i.e., the velocity of the dislocations that ensure the flow strain is determined by the rate of their detachment from the dislocation-blocking barriers, and not by the quasiviscous velocity of the dislocations. It is therefore incorrect to compare the experimental relations obtained in the present study with the dynamic theory of increased plasticity.

Granato^[15] and Suenaga and Galligan^[13] have developed a theory of increased plasticity in the N-S transition; this theory is based on an inertial detachment of dislocations from local obstacles. Owing to the inertial forces, the dislocation can overcome the local barriers by purely mechanical means at stresses $1/2\sigma_c < \sigma < \sigma_c$,^[19]

where σ_c is the critical stress corresponding to the start of the mechanical detachment of the dislocation from the obstacle. The role of the inertial effects increases when the metal goes into the superconducting state because of the decrease of the electron drag coefficient.

The Granato theory, however, does not consider the case when the metal is deformed by a constant external stress, i.e., the creep, and therefore the results obtained in our investigation permit no comparison with a theory based on the model of inertial detachment of the dislocations. But even in the case of active deformation, for which the inertial theory is valid, a comparison of this theory with experiment is not quite legitimate because the inertial detachment mechanism was considered by Granato on the basis of an elementary act in which a single dislocation segment takes part, and no consistent transition was made from this act to the process of macroscopic plastic deformation described by the equation

$$\dot{\epsilon} = \rho b V(\sigma, T, \epsilon). \quad (2)$$

Therefore a comparison of the experimental results with the Granato theory, which is given in a number of papers, cannot be regarded as justified. Nonetheless, the inertial mechanism proposed by Granato seems important and it apparently can contribute to the increased plasticity in the N-S transition. To take it into account, however, further development of the theory is needed.

Natsik^[16] developed a theory of increased metal plasticity in the superconducting transition, based on fluctuating detachment of the dislocations from the local barriers. At temperatures above a certain characteristic value Θ introduced by the theory, the dislocation detachment is determined by the thermal fluctuations, and at $T < \Theta$ the principal role is played by quantum fluctuations. According to this theory, the probability of dislocation detachment is larger in the superconducting state than in the normal state. For the case of creep, the theory yields

$$\frac{\dot{\epsilon}_s}{\dot{\epsilon}_n} = \frac{\nu_s}{\nu_n}, \quad \delta\epsilon_{ns} = \frac{kT^*}{\kappa V} \ln \frac{\nu_s}{\nu_n}. \quad (3)$$

Here κ is the hardening coefficient, V is the activation volume, ν_s and ν_n are the frequencies of the attempts of the dislocation to overcome the barriers in the S and N states, and $T^*(T)$ is an effective temperature that depends on the temperature T and is equal to

$$T^*(T) = \begin{cases} T, & T > \Theta, \\ 1/2\Theta(1+T^2/\Theta^2); & T < \Theta. \end{cases} \quad (4)$$

Measurements of Θ for lead^[26] yielded a value 8°K . To compare the experimental data with Natsik's theory, a plot was constructed of $\kappa V \delta\epsilon_{NS}$ against the relative temperature T/T_c , using for this purpose the data of Fig. 2 and the experimentally obtained dependences of the hardening coefficient κ and of the activation volume V on the degree of deformation. Figure 5 shows this dependence for deformations of 3%, 30%, and 60%.

The ratio ν_s/ν_n in (3) depends, according to Natsik's theory, on the temperature and on the length L of the dislocation segment. At a length $L > L_0$ the ratio ν_s/ν_n takes the form

$$\frac{\nu_s}{\nu_n} = \begin{cases} 1/2(1+e^{kT^*/\Theta^2}); & T_0 < T < T_c, \\ L/L_0; & T < T_0. \end{cases} \quad (5)$$

Here L_0 is the critical length of the segment and deter-

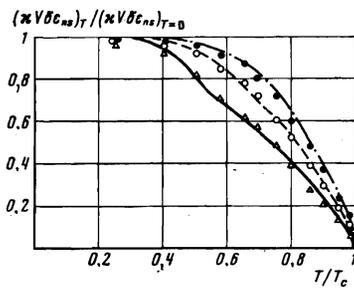


FIG. 5. Temperature dependence of the normalized quantity $\kappa V \delta \epsilon_{ns}$ for a deformation 3% (Δ), 30% (\circ), and 60% (\bullet), in comparison with the theoretical plots [16] of $\varphi(T/T_c)$ for $L = 100L_0$ (solid), $L = 50L_0$ (dashed), and $L = 10L_0$ (dash-dot).

mines the condition of strong damping; its value is $\sim 10^{-5}$ cm, [16] and

$$T_0 = \frac{\Delta(T)}{\ln(2L/L_0 - 1)}. \quad (6)$$

Figure 5 shows also the theoretical plots of

$$\varphi\left(\frac{T}{T_c}\right) = \frac{[T^* \ln(v_s/v_n)]_T}{[T^* \ln(v_s/v_n)]_{T=0}},$$

constructed with allowance for formulas (4)–(6) for dislocation-segment lengths $L = 100L_0$, $L = 50L_0$, and $L = 10L_0$. These lengths correspond to the inequality $L > L_0$. It follows from Fig. 5 that the theoretical plots are in satisfactory agreement with the experimental temperature dependences for the deformations $\epsilon = 3\%$, 30% , and 60% , respectively.

Thus, if the theory developed by Natsik is correct, this means that under these experimental conditions an increase of the degree of deformation of lead single crystals from 3% to 60%, decreases the length of the dislocation segments by a factor of ten. An estimate of the change of the dislocation-segment length can be obtained from measurements of the activation volume, which yield at 4.2°K the values $V = 7 \times 10^{-21}$ and 0.9×10^{-21} cm³ for the deformations 3% and 60%, respectively.

Thus, $L_3/L_{60} \sim V_3/V_{60} \sim 8$, which is in satisfactory agreement with the theory. Approximately the same value is obtained for the ratio of the activation volumes at other temperatures.

Figure 4 shows experimental plots of $\ln(\dot{\epsilon}_S/\dot{\epsilon}_N)$ normalized to 0°K for different degrees of deformation in comparison with the theoretical functions

$$\varphi'\left(\frac{T}{T_c}\right) = \frac{[\ln(v_s/v_n)]_T}{[\ln(v_s/v_n)]_{T=0}}$$

for different lengths of dislocation segments. For large degrees of deformation, the agreement between experiment and theory [16] is also satisfactory. However, although the experimental points corresponding to 3% deformation fall on a curve similar to the theoretical curve corresponding to $L = 100L_0$, they lie above the latter curve rather than on the curve itself. In this case the agreement between experiment and theory is less satisfactory.

It is possible that this difference is due to the fact that at low degrees of deformation the hardening of the sample is still not large and the instantaneous creep produced at the instant of the superconducting transition is appreciable, and this can lead to an overestimate of the flow rate measured in the superconducting state. The presented comparison of the experimental plots with Natsik's theory [16] based on the fluctuating surmounting of local barriers by the fluctuations shows that this theory describes satisfactorily the observed temperature

dependences of the increased-plasticity effect in the N-S transition.

It follows from the experiment and the theory that the form of the temperature dependence is determined essentially by the dislocation structure of the deformed metal, which is characterized in the experiment by the degree of deformation ϵ , and in theory by the dislocation-segment length L . The causes of the difference between the temperature curves of the increased-plasticity effect obtained by different workers [7-9, 12-14] now become clear. Depending on the degree of deformation, and consequently on the character of the defect structure, we can obtain temperature dependences close to those of the energy gap $\Delta(T)$ the density $\rho_S(T)$ of the superconducting electrons, and the critical magnetic field $H_{CR}(T)$ (Fig. 2).

Kuramoto et al. [25] measured the temperature dependence of the stress jump $\delta\sigma_{NS}$ in the region 0.5–6(5°K. The obtained curve was monotonic. In their opinion, the absence of a maximum on the curve means that the experimental results do not agree with Natsik's theory and cannot be explained by this theory. One cannot agree with this statement, however. Indeed, in Natsik's theory there can appear in some cases a maximum on the plot of the temperature dependence of $\delta\epsilon_{NS}$ or $\delta\sigma_{NS}$. For segment lengths $L > L_0$ this should be observed at temperatures $\Theta \ll T_c$. At $\Theta \gtrsim T_c$ (at the same segment lengths) the theory yields a monotonic temperature dependence. In the case of lead $\Theta \approx 8^\circ\text{K}$ and $T_c = 7.18^\circ\text{K}$, i.e., the case of a variation without a maximum is realized.

At $L \ll L_0$, for all values $\Theta \lesssim T_c$, the theory [16] predicts the appearance of a maximum of the temperature dependence of the increased-plasticity effect, but both in our study in [25] the object of the investigation was pure lead, for which this case is not realized. It might be assumed that a maximum of the temperature dependence of $\delta\epsilon_{NS}$ was observed in [10] because these measurements were made on polycrystalline lead, where the length of the dislocation segment could be less than L_0 in view of the presence of intergrain boundaries. However the maximum observed in [10] is not located in the temperature region that follows from the theory of [16].

Druinskiĭ and Fal'ko [17] generalized Natsik's theory to include the case of two-band superconductors, of which they consider lead to be an example. The flow strain jump $\delta\epsilon_{NS}$ is given in this theory by

$$\delta\epsilon_{ns} = \begin{cases} \frac{kT^*}{\kappa V} \ln \frac{1}{\Phi(T)}, & T_0 < T < T_c, \\ \frac{kT^*}{\kappa V} \ln \frac{B_n}{M\omega_0}, & T < T_0, \end{cases} \quad (7)$$

where the temperature T_0 determines the condition of weak and strong damping.¹⁾

The function $\Phi(T)$ is of the form

$$\Phi(T) = 2x_0(1 + e^{\Delta_1/T})^{-1} + 2(1 - x_0)(1 + e^{\Delta_2/T})^{-1}, \quad (8)$$

where $x_0 = (\lambda_1 m_1)^2 / [(\lambda_1 m_1)^2 + (\lambda_2 m_2)^2]$; λ_1 and λ_2 are constants of the electron-phonon interaction; m_1 and m_2 are the values of the effective masses of the electrons of bands I and II; $\Delta_1(T)$ and $\Delta_2(T)$ are the energy gaps of bands I and II, and for lead at 0°K their respective values are

$$\Delta_1(0) = 0.55T_c, \quad \Delta_2(0) = 2.05T_c.$$

For comparison of the experimental data with the theory [17, 18], we plotted in accordance with (7) and (8) the

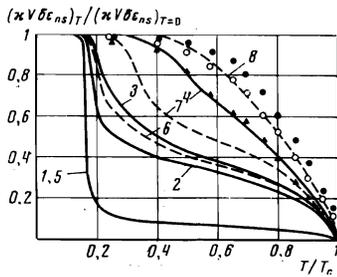


FIG. 6. Temperature dependence of the normalized quantity $\kappa V \delta \epsilon_{ns}$ for deformations 3% (Δ), 30% (\circ), and 60% (\bullet) as compared with the $\Psi(T/T_c)$ plots of the theory [17,18] for $L = 100L_0$, $x_0 = 0$ (curve 1); $x_0 = 0.5$ (curve 2), $x_0 = 0.9$ (curve 3); $x_0 = 1$ (curve 4)—solid lines—and for $L = 50L_0$ and $x_0 = 0$ (curve 5), $x_0 = 0.5$ (curve 6), $x_0 = 0.9$ (curve 7), and $x_0 = 1$ (curve 8)—dashed lines.

functions $\Psi(T/T_c) = T \ln[1/\Phi(T)]$, normalized to 0°K, for segments of length $L = 100L_0$ and $L = 50L_0$ at various values of the parameters x_0 of the theory in the interval $0 \leq x_0 \leq 1$. Figure 6 shows the functions $\Psi(T/T_c)$ for the indicated lengths of the segments and for the parameter x_0 equal to 0, 0.5, 0.9, and 1, as compared with the experimental ratios $(\kappa V \delta \epsilon_{ns})_T / (\kappa V \delta \epsilon_{ns})_{T=0}$ obtained from independent measurements. It follows from the figure that whereas the two energy gaps make comparable contributions to the increased-plasticity effect (for $L = 100L_0$ and $L = 50L_0$ and $x < 1$), the theoretical $\Psi(T/T_c)$ curves do not agree with the experimental ones. An agreement between the theoretical and experimental relations is observed in one of the limiting cases of the theory, when $x_0 = 1$. In this case the theoretical curves for the segments with lengths $L = 100L_0$ and $L = 50L_0$ coincide with the experimental plots for the deformations $\epsilon = 3\%$ and $\epsilon = 30\%$. In this case, according to (8), $\Psi(T/T_c)$ goes over into the function $\varphi(T/T_c)$, which describes the effect in Natsik's theory^[16], i.e., assuming a single-band isotropic superconductor.

The presented comparison of the experimental results with the theory of Druinskiĭ and Fal'ko allows us to conclude that lead is either a one-band superconductor, or for some reason the presence of a second energy gap in it exerts no influence on the increased plasticity effect.

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¹⁾In formula (7), the temperature T was replaced by the effective temperature $T^*(T)$, which made it possible to take into account the distinguishing features of the creep at very low temperatures.

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