

Effect of tin impurities on the temperature dependence of the increased plasticity of lead in superconducting transitions

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The effect of tin impurities and preliminary deformation on the temperature dependence of the increased plasticity effect $\delta\epsilon_{ns}$ during a superconducting transition in lead is investigated. The experiments show that an important feature of the temperature dependence of $\delta\epsilon_{ns}$ in doped and strongly-deformed lead alloys is its nonmonotonic variation at $T/T_c = 0.7$. The experimental data are compared with the fluctuation theory of increased plasticity of metals during an *N-S* transition and satisfactory qualitative agreement is observed. The temperature dependence of the energy gap $\Delta(T)$ in deformed and doped lead alloys is obtained on the basis of the experimental temperature dependence of $\delta\epsilon_{ns}$.

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1. INTRODUCTION

In the study of the temperature dependence of the enhanced plasticity of single crystals of pure lead (99.999%) during the superconducting transition, it was established in the creep process that the degree of deformation of the crystal, i.e., its defect structure, exerts a strong influence on the character of this dependence.^[1] A comparison of the results with the existing plasticity-enhancement theories has shown that they are best described by Natsik's fluctuation theory.^[2,3] The theory predicts that for crystals with an average dislocation-segment length $L \ll L_0$, where L_0 is a theoretical parameter on the order of 10^{-5} cm, a maximum should appear for certain superconductors on the plot of the temperature dependence of the plasticity enhancement. The condition for the appearance of this maximum is $T_c/\Theta \gtrsim 1$, where Θ is a characteristic parameter of the theory and corresponds to the temperature below which the creep process is determined by the quantum mechanisms,^[4] while T_c is the critical temperature of the superconductor. This condition is satisfied for lead, for which $T_c = 7.2^\circ\text{K}$ and $\Theta \approx 8^\circ\text{K}$.^[5]

Experiments on the influence of the degree of deformation on the character of the temperature dependence of $\delta\epsilon_{ns}(T)$ in pure lead have not succeeded in revealing this maximum.^[1] This is not surprising, however, since at the very largest deformations attained in these experiments ($\epsilon = 90\%$), the minimum dislocation-segment length was $L \approx 10L_0$, i.e., exceeded greatly the length at which theory calls for the appearance of the maximum. Suzuki and his co-workers^[6], in an investigation of the increased plasticity of pure lead, likewise failed to observe a maximum, and concluded therefore that the fluctuation theory is incorrect.

Thus, an experimental confirmation of the presence or absence of a maximum is an important test of the theory. We have therefore investigated the temperature dependence of the creep-deformation discontinuity $\delta\epsilon_{ns}$ in the *N-S* transition of deformed single crystals of lead doped with tin, an impurity that decreases the length of the dislocation segment.

2. EXPERIMENTAL PROCEDURE

The measurements were performed on single-crystal lead alloys with tin, Pb + 1 at.% Sn and Pb + 3 at.% Sn. The purity of the initial components amounted to

99.9998% for Pb and 99.999% for tin. The alloys were prepared in a vacuum of 10^{-4} mm Hg.

Lead and tin form a solid solution in which tin has a limited solubility, 3.4 at.% at room temperature. The tin impurity in the aforementioned alloys was therefore atomically-dispersed over the sample. The obtained alloy ingots served as the initial raw material for growing the single crystals by the procedure described in^[7]. All the investigated single crystals (75 pieces) were grown from one primer and had their dilatation axis oriented along $[120]$, i.e., the same orientation as the single crystals of the pure lead in^[1]. To obtain a uniform distribution of the impurities over the sample, and a similar initial dislocation structure, the investigated single crystals were subjected to homogenizing annealing for 15 hours at $T = 260^\circ\text{C}$. A spectral analysis has shown the impurities to be uniformly distributed over the sample length.

We measured the creep deformation increase $\delta\epsilon_{ns}$ at the instant of the superconducting transition as a function of the degree of deformation, in the temperature interval $1.6-7.2^\circ\text{K}$.

The transition from the normal to the superconducting state and back was effected by turning off or on a magnetic field produced by a superconducting solenoid, in which the deformed sample was located. The $\delta\epsilon_{ns}(\epsilon)$ dependence was obtained by the method of stepwise addition of load, as described in^[8]. The same method yielded, in the indicated temperature interval, plots of the strain hardening $\tau(\epsilon)$, which are needed to find the hardening coefficients $\kappa(\epsilon)$, and was also used to measure the activation volume $V(\epsilon)$ of the creep process.

The strain measurement accuracy was 0.2μ . The temperatures in the interval $1.6-4.2^\circ\text{K}$ were obtained by pumping off the vapor over liquid helium. To obtain stabilized temperatures in the interval $4.2-7.2^\circ\text{K}$ we used the procedure described earlier in^[1]. The temperature measurement accuracy was 0.01°K .

3. RESULTS AND DISCUSSION

Figures 1a and 1b show plots of $\delta\epsilon_{ns}$ against the degree of deformation for Pb + 1 at.% Sn and Pb + 3 at.% Sn single crystals in the interval $1.6-7.2^\circ\text{K}$. The experimental points on each curve are the results of measurements on three samples. As seen from the

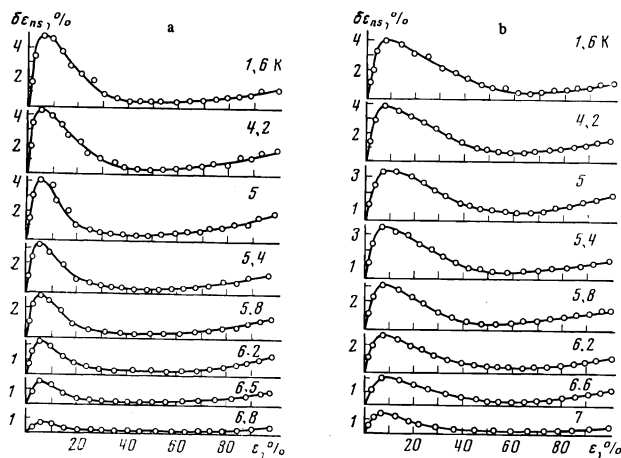


FIG. 1. Jump of the creep deformation $\delta\epsilon_{NS}$ in the N-S transition vs. the degree of deformation in the interval 1.6–7.2°K: a) in the alloy Pb + 1 at.% Sn; b) in the alloy Pb + 3 at.% Sn.

figures, the $\delta\epsilon_{NS}$ curves, just as in the case of pure lead,^[1] represent a number of stages. It was shown earlier in^[9] that this correlates well with the fact that the $\tau(\epsilon)$ curve goes through a number of stages.

Attention is called to the fact that in these alloys there is observed an extremely large discontinuity $\delta\epsilon_{NS}$ at the maxima of the curve, reaching 5% of the deformation in the Pb + 1 at.% Sn alloy and 4% in the Pb + 3 at.% alloy.

According to the data of Fig. 1a, for deformations 5, 60, and 90% of the Pb + 1 at.% Sn alloy and Fig. 1b for deformations 7, 60, and 90% of the Pb + 3 at.% Sn alloy, corresponding to different stages of the hardening curve $\tau(\epsilon)$, we plotted the dependence of the discontinuity of the creep deformation, normalized to the extrapolated value at 0°K, against the relative temperature T/T_c . These plots are shown in Figs. 2a and 2b, respectively.

Just as in the case of pure lead,^[1] the temperature dependences $C(T) = \delta\epsilon_{NS}(T)/\delta\epsilon_{NS}(0)$ are different for different degrees of deformation of the investigated samples.

An important feature of the temperature dependence of the enhanced plasticity in the superconducting transition of the investigated alloys is the presence of strongly pronounced maxima for the larger degrees of deformation. It follows from Figs. 2a and 2b that an increase of the impurity content leads to a change in the deformation at which the maximum is observed. Thus, for example, the curve for the alloy Pb + 3 at.% Sn at a deformation $\epsilon = 60\%$ has a maximum, whereas in the alloy Pb + 1 at.% Sn no maximum appears as yet at this deformation. In the latter alloy, however, the maximum is already clearly pronounced at $\epsilon = 90\%$. We recall that in pure lead no maximum was observed at this deformation. The temperature corresponding to the positions of all the observed maxima is approximately the same at $T \approx 5^\circ\text{K}$. For the alloy Pb + 3 at.% Sn at higher degrees of deformation of the crystal (60–90%), the plots of $C(T)$ are close to one another.

Let us compare the obtained experimental data with the fluctuation theory.^[2] The theory predicts that the defect structure of the superconductor, specified in terms of the length L of the dislocation segment, exerts an influence on the temperature dependence of the enhanced plasticity $\delta\epsilon_{NS}$ during the superconducting

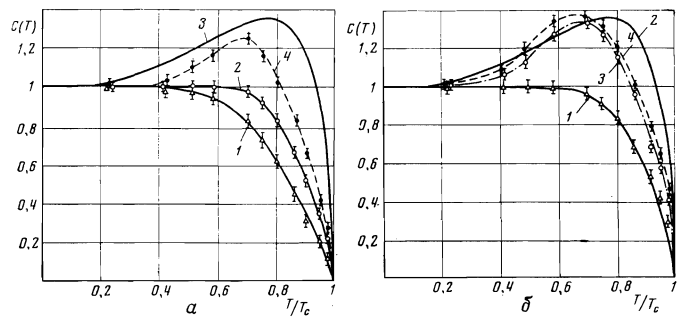


FIG. 2. a) Temperature dependence of the normalized jump $C(T) = \delta\epsilon_{NS}(T)/\delta\epsilon_{NS}(0)$ of the alloy Pb + 1 at.% Sn at the deformations: Δ —5%; \circ —60%; \bullet —90% (curve 4) in comparison with the curves of the theory^[3] for $L = 10L_0$ (curve 1), $L = 30L_0$ (curve 2), and $L \ll L_0$ (curve 3); b) Temperature dependence of the normalized jump $C(T)$ of the alloy Pb + 3 at.% Sn for the deformations: Δ —7%; \circ —60% (curve 3); \bullet —90% (curve 4) in comparison with the curves of the theory^[3] for $L = 4L_0$ (curve 1) and $L \ll L_0$ (curve 2).

transition. According to the theory, at $L > L_0$ we have

$$C(T) = \begin{cases} 1, & T < T_0 \\ \left(\ln \frac{2L}{L_0} \right)^{-1} \left(1 + \frac{T^2}{\Theta^2} \right) \ln \left[\frac{1}{2} (1 + e^{\Delta(T)/kT}) \right], & T_0 < T < T_c \end{cases}$$

and at $L \ll L_0$ we have

$$C(T) = (1 + T^2/\Theta^2) [1 - 4(1 + e^{\Delta(T)/kT})^{-2}], \quad (1b)$$

where T_0 determines the condition for weak and strong damping of the oscillating dislocation segment and is given by the expression

$$\Delta(T)/T_0 = \ln(2L/L_0 - 1).$$

These formulas were obtained under the assumption that the hardening coefficient κ and the activation volume V in the temperature region 0 to T_c do not depend on the temperature.

An analysis of the theoretical plots shows that at $L > L_0$, i.e., at low concentrations of the barriers that hinder the gliding of the dislocations, the plot of the function $C(T)$, remaining monotonic, changes with changing length L of the dislocation segment. At $L \ll L_0$, i.e., at large defect concentrations, $C(T)$ has a maximum, is independent of L , and is determined only by the parameters of the superconductor, namely by the temperature dependence of the energy gap $\Delta(T)$ and by the value of Θ .

As a check on the validity of the comparison of the obtained experimental data with the presented theoretical relations (1a) and (1b), measurements were made of the temperature dependence of the hardening coefficient κ and of the activation volume V of the investigated alloys. The measurements have shown that in the interval 1.6–7.2°K the indicated quantities are practically independent of the temperature.

Figure 2a shows the experimental temperature dependences of $C(T)$ for the alloy Pb + 1 at.% Sn for ϵ equal to 5, 60, and 90%. From the family of the theoretical curves calculated from formulas (1a) and (1b), the experimental relations agree well for $\epsilon = 5\%$ with the $L = 10L_0$ curve and for $\epsilon = 60\%$ for the curve for $L = 3L_0$. The experimental plot corresponding to $\epsilon = 90\%$ has a maximum; this means, according to the theory, that it corresponds to a dislocation segment $L < L_0$. However, the fact that it does not coincide with

the theoretical curve (1b) indicates that in this case L does not differ strongly from L_0 . Indeed, measurements of the dependence of the activation volume on the degree of deformation of this alloy have shown that as the deformation increases from 5 to 90% the value of V decreases by a factor 11, meaning that at $\epsilon = 90\%$ the length of the dislocation segment is $L \approx 0.8L_0$. Similar results were obtained for the alloy Pb + 3 at.% Sn (Fig. 2b). The curves corresponding to deformations 60 and 90% are close to each other. Measurements of $V(\epsilon)$ yielded for the curves corresponding to 7, 60, and 90% deformation dislocation-segment lengths 4, 0.4, and $0.25L_0$, respectively.

Thus, the experimental results are in qualitative agreement with Natsik's fluctuation theory.^[3] This agreement, however, allows us to draw more extensive conclusions. As seen from formula (1b), the temperature dependence of $C(T)$ is determined by the temperature dependence of two factors, $(1 + T^2/\theta^2)$ and $[1 - 4(1 + e^{\Delta(T)/kT})^{-2}]$. The first takes into account the fact that at low temperatures the creep has a quantum character—it is determined by the influence of the zero-point oscillations of the dislocation segments on the process whereby the dislocations surmount the barriers that delay them. With increasing temperature, this factor increases parabolically. The second factor takes into account the variation of the electronic energy spectrum of the superconductor and decreases monotonically with increasing temperature. The product of the factors leads to the appearance of a maximum on the $C(T)$ curve, and its experimental observation offers evidence in favor of the quantum nature of the creep at low temperatures.

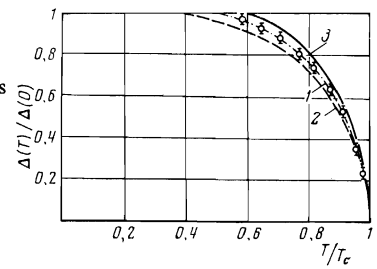
It should be noted that the maximum of the theoretical curve in Fig. 2b is shifted by a small amount relative to the maximum of the experimental curves towards higher temperatures. It can be assumed that the small shift of the theoretical and experimental $C(T)$ curves is due to the influence of the defect structure of the crystal (deformation and impurities) on the temperature dependence of the energy gap.^[10,11] If this assumption is correct, then Eq. (1b) enables us to determine from the experimental $C(T)$ dependence the temperature dependence of the gap $\Delta(T)$ in a crystal with defects. Indeed, it follows from (1b) that

$$\Delta(T) = kT \ln \left\{ \left[4 \left(1 - \frac{C}{(1 + T^2/\theta^2)} \right) \right]^{1/2} - 1 \right\}, \quad (2)$$

where $C = \delta\epsilon_{NS}(T)/\delta\epsilon_{NS}(0)$. The value of $\Delta(T)$ was determined for the alloy Pb + 3 at.% Sn deformed by 90%, i.e., for the alloy with maximum defect structure. The experimental values of $C(T)$ were taken from curve 3 of Fig. 2b.

Measurements of θ in this alloy yielded the same value as in pure lead, 8°K. The obtained temperature dependence of the normalized gap $\Delta(T)/\Delta(0)$ is shown in Fig. 3 (curve 1). The figure shows for comparison the temperature dependence of the gap $\Delta(T)$ in the BCS theory and the experimental $\Delta(T)$ dependence for pure lead.^[12] As seen from the figure, the dependence of the gap $\Delta(T)$ in the deformed lead alloy shifted from the gap dependence in pure lead towards $\Delta(T)$ in accord

FIG. 3. Temperature dependences of the energy gap: curve 1 was calculated from formula (2) from measurements of $\delta\epsilon_{NS}(T)$ in the Pb + 3 at.% Sn alloy for $\epsilon = 90\%$ in comparison with $\Delta(T)$ in accord with the BCS theory (curve 2) and the experimental plot of $\Delta(T)$ in pure lead^[12] (curve 3).



with the BCS theory. This behavior of the temperature dependence of the energy gap is in qualitative agreement with the theoretical calculations on the isotropization of the Fermi surface and with allowance for the change of the phonon spectrum in impurity and deformed superconductors.

The final judgement concerning the validity of the obtained relation can be made only after it is compared with the $\Delta(T)$ relation obtained from direct measurements for a superconductor with a large number of crystal-lattice defects. Such a comparison will serve as one more important verification of the fluctuation theory of increased plasticity in a superconducting transition. Unfortunately, no such measurements have been made so far.

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196