

THE SUPERCONDUCTING PROPERTIES OF RHENIUM

N. E. ALEKSEEVSKIĬ, M. N. MIKHEEVA and N. A. TULINA

Submitted to JETP editor November 25, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) 52, 875-881 (April, 1967)

The effect of plastic compression deformation on the superconducting characteristics of bulk samples of rhenium is investigated. A discontinuity is observed on the curve of the dependence of the critical temperature of the deformed samples on the value of $1/\alpha = r(4.2^\circ\text{K})/r(300^\circ\text{K})$. The discontinuity is attributed to the appearance of a new crystallographic modification of Re. In the range of not very large deformations the ratio H_{C2}/H_{Cm} depends linearly on $1/\alpha$; with increasing deformation the deviation from linearity becomes larger. The characteristic parameter κ grows with increasing deformation; this indicates that deformed rhenium changes from a type-I to a type-II superconductor. The maximum value of κ observed in the bulk samples is 2. The superconducting properties of rhenium films evaporated in vacuum are investigated; the behavior of the films is very similar to that of nontransition metals freshly precipitated at low temperatures. The values of κ and δ_{00} of the films are calculated on the basis of the experimental data.

WE have studied the superconducting characteristics of a series of bulk samples of rhenium, determined the effect of plastic compression deformation on them, and also carried out measurements of the superconducting characteristics of rhenium films.

In order to study the effect of plastic deformation on the superconducting properties of rhenium, we carried out several series of measurements with rhenium samples which had various values of α in their initial undeformed state. The samples underwent several deformations. After each deformation we determined the value of α and the critical temperature. The deformation led to a change in the initial value of α , the value of α attesting to the degree of deformation. The value of α varied in the investigated samples from 20 to 2700.

In addition, we determined the critical fields H_{C2} and H_{Cm} for most of the samples. The field H_{C2} was determined by extrapolating the curves of the dependence of the magnetic moment M on the field H . The value of H_{Cm} was found by integrating the area under these curves. The magnetic moment was determined ballistically. The investigated sample was transported from one measuring coil to the other with the aid of an eccentric rotated by a RD-09 motor. The latter was turned on periodically (with a period of 16–20 sec) for such a time as to allow the sample to move from the center of one coil to the center of the other. The motor was switched on by a time relay. The signals from the

measuring coils connected in opposition to each other were fed to a galvanometer whose deflection was recorded by a recorder modified according to the circuit proposed by Kosourov.^[1]

The critical temperature of the bulk samples was determined from resistivity measurements, as well as from data on the dependence of H_{Cm} on T near T_c .

The magnetic-moment curves of the initial undeformed rhenium samples were those characteristic of pure type-I superconductors. With increasing deformation the $M(H)$ curves became broader, similar to the magnetic-moment curves of alloys (Fig. 1).

Figure 2 shows the dependence of the critical temperature on $1/\alpha$. As is seen from these curves, in the region of small deformations, and consequently of small changes in the mean free path l of the electrons, the critical temperature remained unchanged. For values $1/\alpha = 0.01-0.015$ a jump is observed in the critical temperature, followed by linear increase.

The effect of a given compression deformation (of 10%) on the critical temperature was investigated on several polycrystalline rhenium samples in the form of short cylinders. It turned out that a 10-percent deformation increases T_c by 0.1°K . It should be noted that analogous behavior was observed in rhenium samples which underwent deformation in tension.^[2]

Figure 3 shows the dependence of H_{C2}/H_{Cm} on $1/\alpha$. Within the range of small values of $1/\alpha$ this

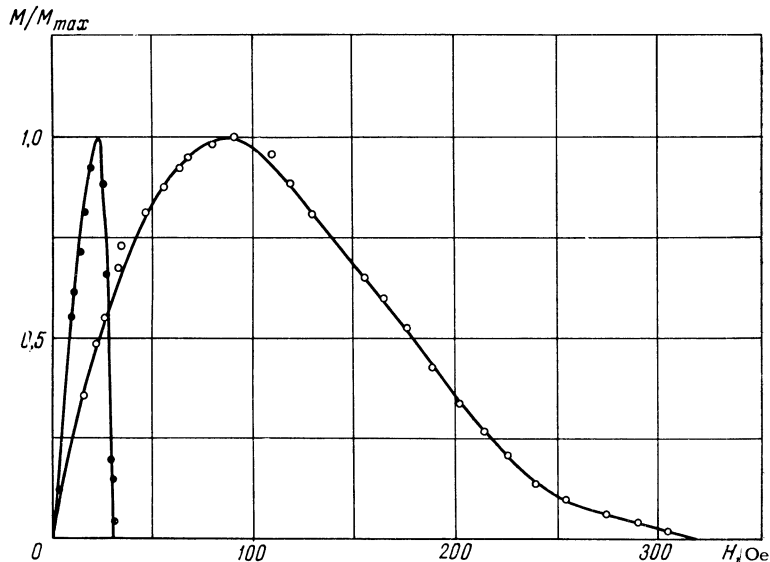


FIG. 1. Dependence of the relative magnetic moment on the field for a bulk single-crystal sample of rhenium: ●—initial sample, $\alpha = 2700$, $T = 1.63^\circ\text{K}$; ○—sample which underwent multiple deformation, $\alpha = 23$, $T = 1.615^\circ\text{K}$.

dependence is linear. Attention is drawn to the fact that for weakly deformed rhenium samples H_{C2} determined from extrapolation of the $M(H)$ dependence coincides with the first appearance of resistance. Full re-establishment of the resistance occurs in a field exceeding H_{2C} by a factor of about 1.7.

We investigated a large number of wire samples drawn to 0.2–0.3 mm through a draw plate. This wire had critical temperatures of about 3.5°K . In many wire samples the transition curve was step-like. This step-like nature disappeared after annealing, and the transition curve became smooth. Prolonged annealing in vacuum decreased, as a rule, the critical temperature down to the value characteristic for pure undeformed samples. Etching in hot acids also led to a decrease in the critical temperature. Radiochemical analysis of the

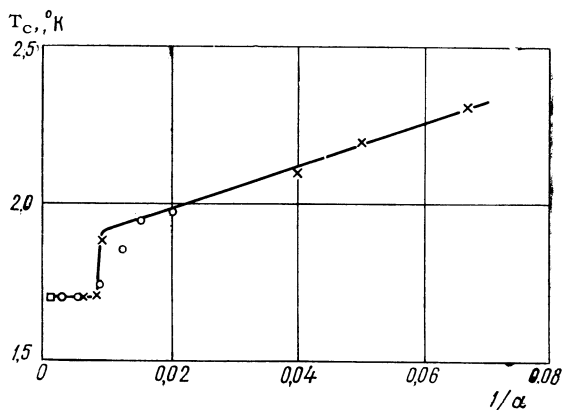


FIG. 2. Dependence of the critical temperature of bulk single-crystal samples of rhenium on $1/\alpha = r(4.2^\circ\text{K})/r(300^\circ\text{K})$: ○—samples of series 1, for the initial undeformed sample of this series $\alpha = 540$; ×—samples of series 2, for the initial undeformed sample of this series $\alpha = 178$; □—sample with the initial value $\alpha = 2700$.

rhenium wire for W, Ta, and Mo impurities indicated¹⁾ that the samples which we investigated contained no more than 10^{-4} per cent of these metals, which are the main impurities in rhenium, and that consequently they could not be responsible for the increase in T_c of the wires.

The above allows one to believe that the high critical temperature of the rhenium wire is due to the deformations and not to the chemical impurities. This is also indicated by the fact that the rhenium films obtained by evaporation of the metal of such a wire had low critical temperatures of about 2°K .

Such behavior of the rhenium wires is in agreement with the nature of the dependence of the critical temperature on the deformation which we obtained for deformed single-crystal samples of rhenium (Fig. 3). A critical temperature of the order of 3.0 – 3.5°K should be observed, according to this curve, in the case of samples with $1/\alpha \sim 0.14$ – 0.15 . For the wires which we investigated $1/\alpha \sim 0.1$. Since this value should refer to the most strongly deformed surface portion of the wire, the value of $1/\alpha \sim 0.1$ usually obtained for unannealed wire is in fairly good agreement with the results obtained for deformed samples.

We have also carried out measurements on rhenium films. The films were obtained by evaporation of the metal in a vacuum of 10^{-6} – 10^{-7} mm Hg onto a glass substrate in a device described in^[3]. As the evaporator we used a coil of rhenium wire heated up to 3000°C by a current. For films

¹⁾The authors express their sincere gratitude to Yu. B. Gerlit for carrying out the radiochemical analysis of the rhenium wires and for determining the thickness of the rhenium films by an activation method.

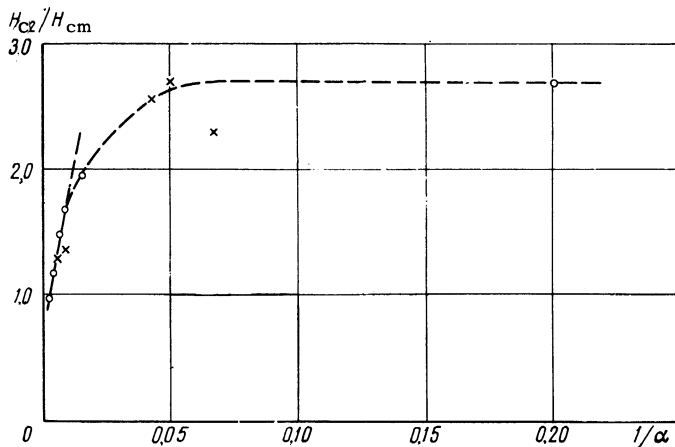


FIG. 3. Dependence of the value of the ratio H_{c2}/H_{cm} on $1/\alpha$: \circ —samples of series 1, for the initial undeformed sample of this series $\alpha = 540$; \times —samples of series 2, for the initial undeformed sample of this series $\alpha = 178$.

evaporated onto a liquid-nitrogen cooled substrate and subsequently annealed at room temperature, T_C was about 1.7–1.8°K. Films deposited onto a substrate at a temperature of 500–600°K during the evaporation went over into the superconducting state at 2.0–2.2°K. No correlation between the critical temperature and the film thickness was observed.

The transition of vacuum-evaporated rhenium films from the normal to the superconducting state was quite sharp (0.01°K). All the films which we investigated had a very high resistivity in the normal state (a film about 10^{-6} cm thick had a resistivity of about 1 ohm).

In their behavior in a magnetic field, the rhenium films are close to films of nontransition me-

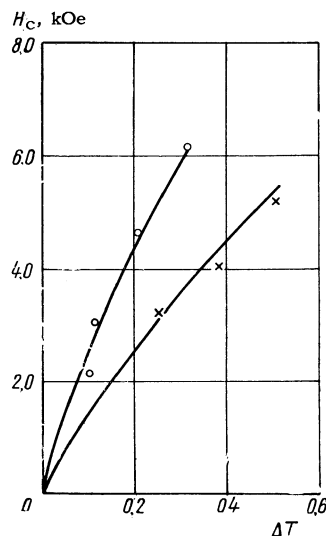


FIG. 4. Temperature dependence of the critical magnetic field of rhenium samples: \times — $d = 4.0 \times 10^{-6}$ cm, \circ — $d = 2.58 \times 10^{-6}$ cm.

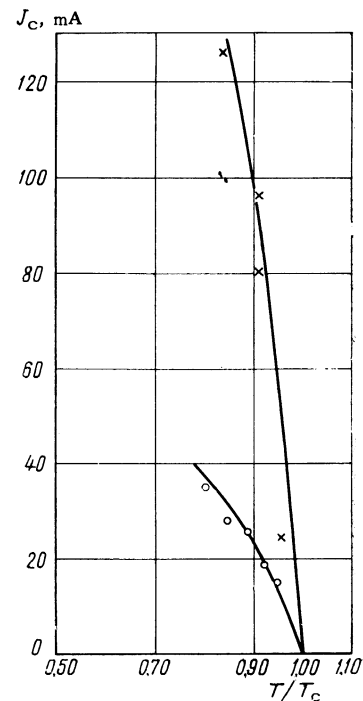


FIG. 5. Temperature dependence of the critical current of the rhenium films: \times — $d = 4.0 \times 10^{-6}$ cm, \circ — $d = 5.8 \times 10^{-7}$ cm.

tals freshly deposited at helium temperature: their critical currents are extremely small, and the critical magnetic fields are very high (Figs. 4 and 5). Thus, in the case of a film of thickness $d = 2.58 \times 10^{-6}$ cm, $(dH_c/dT)_{T_C} = 20$ kOe/°K.

The electron mean free path estimated for two rhenium films from their resistivity in the normal state was $l \sim 3 \times 10^{-7}$ cm. For comparison we indicate that aluminum films freshly deposited at 4.2°K have $l = 1.9 \times 10^{-7}$ cm and in the case of recrystallized aluminum films $l = 2.3 \times 10^{-6}$ cm.^[4]

It should be noted that rhenium films obtained by us by evaporating bulk rhenium with the aid of an electron beam differed in their properties from the films we described above: their transition to the superconducting state was very broad with respect to the temperature, and the critical temperatures exceeded 3°K. Such behavior is apparently explained by the less clean conditions under which the films were prepared and by their contact with air during their transfer from the evaporating chamber to the working Dewar.

If one considers the results obtained in bulk samples of rhenium, then one can conclude that pure rhenium with $\alpha = 2700$ is a type-I superconductor—its $M(H)$ curves exhibit a small hysteresis; the value of κ estimated from the formula $H_{c2} = \sqrt{2}\kappa H_{cm}$ is less than $1/\sqrt{2}$.

As the plastic deformation is increased, the rhenium begins to transform into a type-II super-

conductor; at the same time κ increases and approaches 2 for the most strongly deformed samples. In the region of not very large deformations H_{C2}/H_{Cm} depends linearly on $1/\alpha$; this could be considered to be in agreement with the GLAG theory.^[5] However, along with the coinciding functional dependence of H_{C2}/H_{Cm} on $1/\alpha$, a quantitative discrepancy between H_{C2}/H_{Cm} obtained from the theory and its value calculated on the basis of experimental data is observed.

In the region of large deformations the linear character of the dependence of H_{C2}/H_{Cm} on $1/\alpha$ is violated. It should be noted that the curve of the dependence $T_c(1/\alpha)$ experiences at the same time a jump. It is not excluded that such a jump can be related with a polymorphic transformation of rhenium. One could assume that as a result of plastic deformation there appears a grid of regions with maximum lattice distortions at whose locations there appears a new crystallographic modification of rhenium with a higher T_c . If it is assumed that the jump on the $T_c(1/\alpha)$ curve is related to the appearance of a new phase, then the critical temperature of this phase should be $\sim 2^\circ\text{K}$.

One can thus imagine one of the possible pictures of the change of the superconducting properties of rhenium with deformation: initially, for small deformations, T_c practically does not change and the rhenium changes from a type-I into a type-II superconductor; this is followed by a jump-like increase of T_c to 2.0°K owing to the appearance of a new phase. Further increase of deformation leads to a linear growth of T_c . Such a growth can be connected with an increase in the exponent in the expression

$$T_c = 1.14\Theta e^{-1/N(0)V}. \quad (1)$$

If one considers the data on rhenium films which we have obtained, one's attention is drawn to the fact that depending on the temperature of the substrate during the condensation the films had different critical temperatures: $1.7\text{--}1.8^\circ\text{K}$ at a substrate temperature of 90°K , and $2.0\text{--}2.2^\circ\text{K}$ at a substrate temperature of $500\text{--}600^\circ\text{K}$. These data can apparently be considered as a confirmation of the existence in rhenium of a new modification, as we already noted above.

Since the rhenium films which we investigated are close in their properties to those of nontransition metals freshly deposited at 4.2°K , one can apply to them the Abrikosov-Gor'kov theory of alloys.^[6] This enables one to determine the characteristic parameters κ and δ_{00} with the aid of the following expressions:

$$\kappa = 0.065ec\gamma^{1/2}/k\sigma, \quad (2)$$

$$\delta = \frac{c}{2\pi} \left(\hbar \left| \sigma \Delta \operatorname{th} \frac{\Delta}{2kT_c} \right| \right)^{1/2}, \quad (3)$$

$$\delta = \delta_{00}/\sqrt{1 - (T/T_c)^4}, \quad (4)$$

where σ is the conductivity of the films, Δ is the energy gap, δ is the penetration depth of the magnetic field, and γ is the coefficient of the linear term in the specific-heat expression²⁾.

For a film with a thickness $d = 2.58 \times 10^{-6}$ cm an estimate by means of these formulas yields $\kappa = 2.3$. An estimate of the value of κ of strongly deformed bulk rhenium according to Eq. (2) yields values closely coinciding with the values of κ determined from the expression $H_{C2} = \sqrt{2}\kappa H_{Cm}$.

The value of δ_{00} calculated from the conductivity of the films turns out to be $\delta_{00} = 32 \times 10^{-6}$ cm. If, on the other hand, one makes use of the data on the critical magnetic fields of the films and calculates δ_{00} from the relation

$$\frac{H_c}{2\Theta^2} = \sqrt[3]{\frac{2.57 \cdot 10^{-4}}{\Theta d}}, \quad \Theta = \sqrt{\frac{2e}{\hbar c}} H_{cm} \delta, \quad (5)$$

then δ_{00} turns out to be 33.5×10^{-6} cm, which is in good agreement with the first value. A somewhat larger value $\delta_{00} = 47.6 \times 10^{-6}$ cm is obtained from the expression $H_c/H_{Cm} = 2\sqrt{6}\delta/d$.

The values of δ_{00} which we have obtained for rhenium are apparently the highest known at present³⁾. The large value of the penetration depth of rhenium is in all probability a result of the fact that rhenium is a superconductor with weak coupling, i.e., it has a small electron-phonon interaction constant. If we make use of data on the specific heat of rhenium and take

$$\gamma = 2.55 \text{ mJ/mole-deg}^2, \quad \Theta = 405^\circ \text{K}, \quad T_c = 1.7^\circ \text{K},$$

then we can determine from expression (1) the electron-phonon interaction parameter V which turns out to be several times smaller than in the usual soft superconductors, and smaller by two orders of magnitude than that of lead.

In conclusion the authors consider it their pleasant duty to express their gratitude to B. D. Yurasov for help in the construction of instruments and Yu. A. Deniskin and A. A. Grigor'ev for help in carrying out the experiment.

²⁾It is assumed that the value of γ for a rhenium film does not differ from its value for the bulk metal.

³⁾It should be noted that the high values of δ_{00} for cadmium obtained in [7] are in all probability a consequence of sample contamination.

- ¹G. I. Kosourov, PTÉ No. 3, 90 (1956).
- ²J. Hauser and E. Buchler, Phys. Rev. 125, 142 (1962).
- ³N. E. Alekseevskiĭ and M. N. Mikheeva, JETP 31, 951 (1956), Soviet Phys. JETP 4, 810 (1957).
- ⁴I. S. Khukhoreva, Dissertation, Moscow State University, 1962.
- ⁵A. A. Abrikosov and L. P. Gor'kov, JETP 35, 1558 (1958) and 36, 319 (1959), Soviet Phys. JETP 8, 1090 (1959) and 9, 220 (1959).
- ⁶L. P. Gor'kov, JETP 36, 1918 (1959), and 37, 1407 (1959), Soviet Phys. JETP 9, 1364 (1959) and 10, 998 (1960).
- ⁷R. A. Hein and M. C. Steele, Phys. Rev. 105, 877 (1957).

Translated by Z. Barnea

110