

Study of Fermi-Surface Topology Changes in Rhenium and Dilute Re Solid Solutions from T_c Measurements at High Pressure

C. W. CHU*

Bell Telephone Laboratories, Murray Hill, New Jersey 07974

AND

T. F. SMITH* AND W. E. GARDNER*

Materials Physics Division, Atomic Energy Research Establishment, Harwell, Didcot, Berkshire, England

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The superconducting transition temperature has been measured as a function of hydrostatic pressure up to 18 kbar for single-crystal and polycrystal samples of pure rhenium and dilute solid solutions of Mo, W, and Os in rhenium. The pressure dependence for pure Re is anomalous and varies rapidly with solute additions. This anomalous behavior is explained in terms of an abrupt change in Fermi-surface topology under pressure.

INTRODUCTION

THE superconducting transition temperature T_c for the majority of the known superconducting elements has been studied as a function of pressure.¹ In general, T_c follows a monotonic function with pressure, increasing in a few cases, but usually decreasing. It has recently been demonstrated² for lead that, given an adequate knowledge of the pressure dependence of the phonon spectrum, the pressure dependence of T_c calculated from McMillan's³ expression which includes the effects of strong coupling, is in very good agreement with the experimental value. This agreement will very likely be achieved for all nontransition-metal superconductors, which, with the exception of thallium, exhibit a decrease of T_c with pressure. The situation for the transition-metal superconductors is more complex and a better understanding of the electron-electron interactions is required before such a calculation of the pressure dependence of T_c is possible.

The possibility of a nonmonotonic pressure dependence of T_c arising from pressure-induced abrupt changes in the topology of the Fermi surface has been discussed by Makarov and Bar'yakhtar.⁴ It has been suggested that such a transition is responsible for the anomalous pressure dependence of T_c for thallium. The extensive study of the pressure dependence of T_c for Tl and dilute Tl alloys by Lazarev and co-workers⁵ provides convincing support for this hypothesis.

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¹ For a general review see M. Levy and J. L. Olsen, *Physics of High Pressure and the Condensed Phase*, edited by A. Van Itterbeek (North-Holland Publishing Co., Amsterdam, 1965), p. 525.

² R. E. Hodder, *Phys. Rev. Letters* **22**, A8 (1969).

³ W. L. McMillan, *Phys. Rev.* **167**, 331 (1968).

⁴ V. I. Makarov and V. G. Bar'yakhtar, *Zh. Eksperim. i Teor. Fiz.* **48**, 1717 (1965) [English transl.: *Soviet Phys.—JETP* **21**, 1151 (1965)].

⁵ B. G. Lazarev, L. S. Lazareva, and V. I. Markarov, *Zh. Eksperim. i Teor. Fiz.* **44**, 481 (1963) [English transl.: *Soviet Phys.—JETP* **17**, 328 (1963)]; B. G. Lazarev, L. S. Lazareva, V. I. Makarov, and T. A. Ignat'eva, *Zh. Eksperim. i Teor. Fiz.* **46**,

The observation of a nonmonotonic pressure dependence of T_c for rhenium has been briefly reported.⁶ T_c for both single-crystal and polycrystal rhenium initially decreases with pressure, passes through a minimum at ~ 7 kbar, and then levels off between 13 and 18 kbar. The addition of Os rapidly displaces the minimum to lower pressures and the anomalous behavior disappeared entirely in alloys containing more than 0.6-at.% Os, for which T_c decreases almost linearly with pressure. This behavior suggested that an abrupt change in Fermi-surface topology also occurs in pure Re. Further measurements have now been made on dilute solid solutions of W and Mo in Re which support this proposal.

This paper includes full details of the preparation and results that have been obtained for polycrystal and single-crystal samples of pure Re and polycrystal samples of Re-Os, Re-Mo and Re-W solid solutions.

SAMPLE PREPARATION

Rhenium samples were cut from polycrystalline and single-crystal material obtained from Materials Research Corporation (MRC Grade 1, zone refined, quoted purity: 99.9 wt%). Spectroscopic analysis of a sample of the polycrystal material gave the major impurities in ppm atomic as W 1.4, Mo 12, Ni 9, Fe 4, Ca 2, K 20, and S 12 in reasonable agreement with a "typical" analysis supplied by MRC. Samples cut directly from the "as received" material had superconducting transition widths which extended over more than 1°K. A typical transition curve obtained for a polycrystalline sample (Fig. 1), shows a transition ranging from 3.1 to 1.7°K.

Such broad transitions have been observed in previous investigations and are attributed to the extreme sensitivity of the transition in Re to the amount of

829 (1964); **48**, 1065 (1965) [English transl.: *Soviet Phys.—JETP* **19**, 566 (1964); **21**, 711 (1965)]; N. B. Brandt, N. I. Ginzburg, T. A. Ignat'eva, B. G. Lazarev, L. S. Lazareva, and V. I. Makarov, *Zh. Eksperim. i Teor. Fiz.* **49**, 85 (1965) [English transl.: *Soviet Phys.—JETP* **21**, 1151 (1965)].

⁶ C. W. Chu, T. F. Smith, and W. E. Gardner, *Phys. Rev. Letters* **20**, 198 (1968).

TABLE I. Summary of metallurgical treatments and atmospheric pressure T_c for a number of Re samples.

Sample	T_c (°K)	Annealing conditions			Remarks
		Temperature (°C)	Time (h)	Vacuum (mm Hg)	
Re (polycrystalline)					
Re 1P	~3.1 -1.7				MRC, grade 1, 99.9 wt% purity
Re 2P	1.698-1.694				As received
Re 3P	1.695-1.692	1400	1	$2-3 \times 10^{-5}$	Arc melted, He atmosphere
Re 4P	1.696-1.694				
Re 5P	1.695-1.693	1350	1	$\sim 7.5 \times 10^{-5}$	Arc melted, Ar atmosphere
Re 6P	1.695-1.693				
Re (single crystal)					
Re 1S	~2.8 -2.0				Arc melted, He atmosphere
Re 2S	1.696-1.690	1600-1700	1	$3-5 \times 10^{-5}$	MRC, grade 1, 99.9 wt% purity
Re 3S	1.696-1.690	1500-1680	$1\frac{1}{2}$	$\sim 10^{-2}$	As received
Re 4S	1.695-1.694	1600-1750	1	$2-3 \times 10^{-5}$	Spark cut
Re 5S	1.695-1.694	1500	1	$6.7-7.5 \times 10^{-5}$	Cut on carborundum wheel
Re 6S	1.695-1.693	1500	$1\frac{1}{2}$	$4.5-5 \times 10^{-5}$	Cut on carborundum wheel
					Spark cut

plastic deformation and internal strain in the material. It was therefore necessary to sharpen the transition before the effect of pressure could be studied and it was found that inductive annealing *in vacuo* or, in the case of the polycrystal samples, arc melting in either a helium or an argon atmosphere produced satisfactory transition curves (Fig. 1). A full summary of the heat treatments is given in Table I.

Solid solutions of Os, W, and Mo in Re were prepared from MRC polycrystal Re and the appropriate amount of solute by arc melting in an argon atmosphere. In order to promote homogeneity, each sample was turned and remelted at least seven times. The more dilute samples (<0.5-at.% solute) were prepared by the addition of more Re to the alloy of next-highest concentration. As the weight losses which occurred during

melting were negligibly small, the quoted compositions are those calculated from the initial relative proportions of the constituents. The transition curves for the arc-melting alloy samples were sharp (between 1.5 and 5 mdeg wide) and this was taken to be indicative of good homogeneity and so no further treatment was given.

EXPERIMENTAL DETAILS

The sensitivity of the superconducting transition of Re to inhomogeneous strain⁷ placed stringent requirements on the achievement of hydrostatic pressures. A pressure transmission medium of micron-size Teflon particles, which has been used successfully in a number of previous investigations of the pressure dependence of T_c , was found to be unsuitable for the present measurements since it was observed that after the application of pressure there was a large irreversible shift in the zero-pressure value of T_c . This is illustrated in Fig. 1 which shows the effect on the transition curve of applying a pressure of 18.5 kbar with this medium. This led us to try a fluid medium of a 1:1 mixture of *n*-pentane and isoamyl alcohol contained in a self-sealing Teflon cell similar to that described by Jayaraman *et al.*⁸ The cell was pressurized at room temperature between two high-density alumina pistons in a $\frac{1}{4}$ -in. id hardened Cu-Be cylinder. The pressure was retained by a clamp arrangement for cooling to liquid-helium temperature. Cooling produced a pressure loss of 3-4 kbar. Pressures, as measured by a superconducting tin manometer,⁹ were achieved up to 19 kbar. Following the application of the maximum pressure with this arrangement the zero-pressure transition curve was found to be reproducible to within a millidegree. Some initial measurements were made using a hydraulic oil

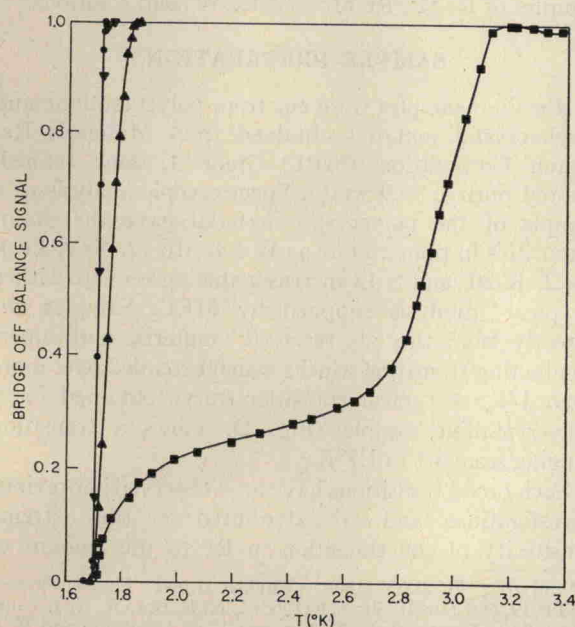


Fig. 1. Effect of strain on the superconducting transition curve for pure polycrystalline Re. ■, as received; ●, after arc melting; ▲, after application of 18.5 kbar (Teflon pressure medium); ▼, after application of 17.3 kbar (fluid pressure medium).

⁷ J. J. Hauser and E. Buehler, Phys. Rev. **125**, 142 (1962); N. E. Alekseyevsky, M. N. Mikheyeva, and N. A. Tulina, Zh. Eksperim. i Teor. Fiz. **52**, 875 (1967) [English transl.: Soviet Phys.—JETP **25**, 575 (1967)].

⁸ A. Jayaraman, A. R. Huston, J. H. McFee, A. S. Coriel, and R. G. Maines, Rev. Sci. Instr. **38**, 44 (1967).

⁹ T. F. Smith, C. W. Chu, and M. B. Maple, Cryogenics **9**, 53 (1969).