

# Magnetic and Resistive Transitions of Some Mo-Re Alloys

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Recent experimental work on the properties of pure metal<sup>1</sup> and alloy<sup>2-5</sup> superconductors with negative surface energies has confirmed the ideas of the GLAG theory.<sup>6-9</sup> Negative surface energy materials (superconductors of the second kind) are characterized by an order parameter  $K$  having values larger than 0.707.  $K$  is related to the bulk properties of the metal, and to the shape of the magnetization curves for this kind of materials. In contrast to superconductors of the first kind (with  $K$  less than 0.707) which return to the normal state at a single critical field, superconductors of the second kind possess two critical fields: the field at which flux initially penetrates,  $H_{c1}$ , and the field at which the sample is returned to the fully normal state,  $H_{c2}$ .

The present work on three metallurgically ideal Mo-Re alloy specimens gives further confirmation to the gross features of the GLAG theory. The samples were prepared by electron beam zone refining and were of compositions Mo<sub>0.98</sub>Re<sub>0.02</sub>, Mo<sub>0.85</sub>Re<sub>0.15</sub>, and Mo<sub>0.75</sub>Re<sub>0.25</sub>. The Mo<sub>0.98</sub>Re<sub>0.02</sub> and Mo<sub>0.75</sub>Re<sub>0.25</sub> were single crystals, while the Mo<sub>0.85</sub>Re<sub>0.15</sub> consisted of one large crystal and several smaller crystals located at one end. The samples were carefully cut with a diamond wheel and spark cutter and etched to remove any remaining surface strains. The final length was 5 in. and the diameter was 0.3 in. Magnetization measurements were made ballistically, the detector coil sampling a central portion of the specimen, which in the case of the Mo<sub>0.85</sub>Re<sub>0.15</sub> sample was the large single-crystal portion. Resistive transition measurements were made using the usual four-probe method.

The transitions from the superconducting to the normal state were studied over an extended temperature range. The Mo<sub>0.98</sub>Re<sub>0.02</sub> was found to be a superconductor of the first kind as was expected from the calculated value of  $K = 0.35$ . The other two specimens with  $K$  values greater than 0.707 (Mo<sub>0.85</sub>Re<sub>0.15</sub> with  $K = 2.62$ , Mo<sub>0.75</sub>Re<sub>0.25</sub> with  $K = 3.68$ ) showed the characteristic features of superconductors of the second kind. The relevant parameters for these materials are given in Table I.

TABLE I. Values of  $H_c$ ,  $\rho$ ,  $K$ , and  $T_c$  for Mo-Re alloys.

	Mo <sub>0.98</sub> Re <sub>0.02</sub>	Mo <sub>0.85</sub> Re <sub>0.15</sub>	Mo <sub>0.75</sub> Re <sub>0.25</sub>
$H_0^a$	132 G	1240 G	1600 G
$H_0^b$	...	1218 G	1515 G
$\rho$	0.98 $\mu\Omega$ -cm	5.44 $\mu\Omega$ -cm	7.48 $\mu\Omega$ -cm
$K^c$	0.35	2.49	3.70
$K^d$	...	2.62	3.45
$K^e$	...	2.76	3.90
$T_c$	1.13°K	7.99°K	10.25°K

<sup>a</sup> Calculated from the specific heat data of Morin and Maita (Ref. 11).

<sup>b</sup> From the areas under the experimental magnetization curves.

<sup>c</sup> Using the Gor'kov (Ref. 8) relation  $K = K_0 + 7.5 \times 10^3 \gamma^{1/2} \rho$ , where  $\gamma$  is the specific heat constant in erg/cm<sup>3</sup>-deg<sup>2</sup> and  $\rho$  is the residual resistivity in  $\Omega$ -cm. Note:  $K_0$ , the part of  $K$  independent of mean free path, is negligible for these alloys.

<sup>d</sup> Using the Abrikosov (Ref. 7) relation  $(H_{c1}/H_c)T_c = \sqrt{2} K$ .

<sup>e</sup> Using the Abrikosov (Ref. 7) relation  $4\pi \left( \frac{dM}{dH} \right)_{H_{c2}, T_c} = \frac{1}{1.18(2K^2 - 1)}$ .

Magnetization curves of the Mo<sub>0.75</sub>Re<sub>0.25</sub> and Mo<sub>0.85</sub>Re<sub>0.15</sub> are shown in Figs. 1 and 2. The Mo<sub>0.75</sub>Re<sub>0.25</sub> shows essentially reversible behavior and less than 2% trapped flux, which we believe to be the smallest yet reported for specimens with large values of  $K$ . Both samples show a steep slope in the magnetization curve near  $H_{c1}$ , and do not show the rounding associated with samples containing small chemical inhomogeneities or a defect structure.<sup>3</sup> The Mo<sub>0.85</sub>Re<sub>0.15</sub> sample shows more trapped flux, and thus would seem to be less ideal. An important difference between the samples as displayed in the curves is the oscillation in the magnetization between  $H_{c1}$  and  $H_{c2}$  for the Mo<sub>0.75</sub>Re<sub>0.25</sub>. Similar effects have been observed by DeSorbo<sup>10</sup> in other alloys and have

<sup>10</sup> W. DeSorbo, Phys. Rev. **130**, 2177 (1963).

<sup>1</sup> T. F. Stromberg and C. A. Swenson, Phys. Rev. Letters **9**, 370 (1962).

<sup>2</sup> J. D. Livingston, Phys. Rev. **129**, 1943 (1963).

<sup>3</sup> T. Kinsel, E. A. Lynton, and B. Serin, Phys. Letters **3**, 30 (1962).

<sup>4</sup> T. G. Berlincourt and R. R. Hake, Phys. Rev. **131**, 140 (1963).

<sup>5</sup> C. Chiou, R. A. Connell, and D. Seraphim, Phys. Rev. **129**, 1070 (1963).

<sup>6</sup> V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. **20**, 1064 (1950).

<sup>7</sup> A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].

<sup>8</sup> L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **37**, 1407 (1959) [English transl.: Soviet Phys.—JETP **10**, 998 (1960)].

<sup>9</sup> L. B. Gor'kov, Zh. Eksperim. i Teor. Fiz. **37**, 835 (1959) [English transl.: Soviet Phys.—JETP **16**, 593 (1960)].

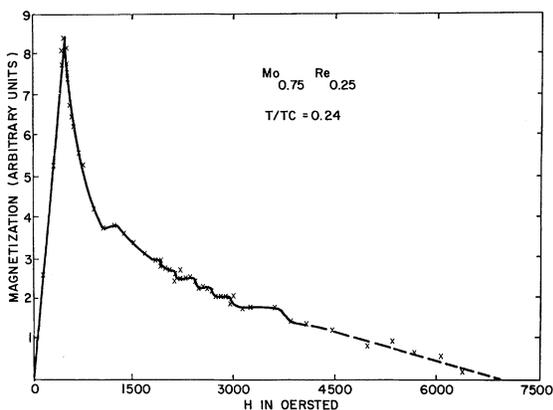


FIG. 1. Magnetization curve of  $\text{Mo}_{0.75}\text{Re}_{0.25}$  at a reduced temperature,  $T/T_c = 0.240$ .

been associated with flux jumping. In view of the otherwise ideal characteristics of this sample and the absence of such effects in the apparently less ideal  $\text{Mo}_{0.85}\text{Re}_{0.15}$  sample, it does not seem likely that a defect related flux jumping mechanism can explain

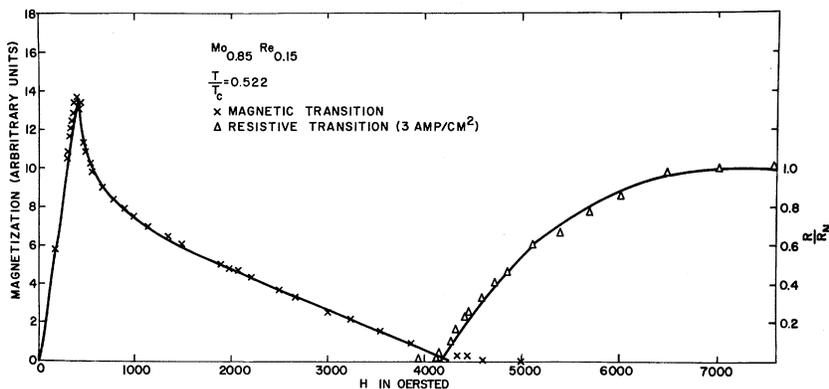


FIG. 2. Magnetization and resistive transition of  $\text{Mo}_{0.85}\text{Re}_{0.15}$  at a reduced temperature,  $T/T_c = 0.522$ .

this behavior. Further work is in progress to determine the origin of these oscillations.

A further characteristic observed in both samples is illustrated in Fig. 2 where it can be seen that zero magnetization does not correspond to the return of full normal state resistance as determined by low current density (1–3 A/cm<sup>2</sup>) measurements. The resistance transition is quite broad and extends to fields of the order of twice the field at which  $M = 0$ . The onset of resistance for the  $\text{Mo}_{0.85}\text{Re}_{0.15}$  sample corresponded at each temperature to the point at which the magnetization did become zero, while there was a considerable separation between these points for the  $\text{Mo}_{0.75}\text{Re}_{0.25}$  sample. Again the ideal nature of these specimens would seem to make questionable the presence of large scale chemical or physical inhomogeneities which would produce fila-

ments retaining superconductivity to these relatively much higher fields. Further measurements using higher current densities should give a better indication of the nature of the mechanism producing a reduced value of resistance at fields greater than the bulk upper critical field.

The temperature dependence of the lower critical field  $H_{c1}$  is shown in Fig. 3. It can be seen that in both samples the experimental data gives an excellent fit to a  $(T/T_c)^2$  plot.

In Fig. 4 the temperature dependence of the upper critical field  $H_{c2}$  is displayed. The data is normalized by plotting  $H_{c2}/KH_0$  as a function of reduced temperature,  $T/T_c$ .  $H_0$  is the thermodynamic critical field at  $T = 0$  and is calculated from the specific heat data of Morin and Maita.<sup>11</sup> The  $K$  used is the average of the  $K$  calculated by the several methods noted in Table I. The upper dashed curve shows the temperature dependence expected from Abrikosov's theory using the temperature dependence of  $K$  due to Ginzburg:

$$\frac{H_{c2}}{KH_0} = \frac{2\sqrt{2}[1 - (T/T_c)^2]}{[1 + (T/T_c)^2]}$$

while the lower solid curve is that predicted by Gor'kov for the supercooling field:

$$\frac{H_{c2}}{KH_0} = \left[ 1.77 - 0.43 \left( \frac{T}{T_c} \right)^2 + 0.07 \left( \frac{T}{T_c} \right)^4 \right] \times \left[ 1 - \left( \frac{T}{T_c} \right)^2 \right].$$

The experimental points for the  $\text{Mo}_{0.85}\text{Re}_{0.15}$  lie between these curves but closer to the Gor'kov dependence, as has been reported by Chandrasekhar, Hulm, and Jones<sup>12</sup> for niobium alloys in which the

<sup>11</sup> F. J. Morin and J. B. Maita, Phys. Rev. **129**, 1115 (1963).

<sup>12</sup> B. S. Chandrasekhar, J. K. Hulm, and C. K. Jones, Phys. Letters **5**, 18 (1963).

impurity scattering portion of  $K$  is dominant, and where  $H_{c2}$  was determined by the onset of resistance. In the case of  $\text{Mo}_{0.75}\text{Re}_{0.25}$  the temperature dependence of the field at which the onset of resistance occurs agrees with the data for the  $\text{Mo}_{0.85}\text{Re}_{0.15}$ , but

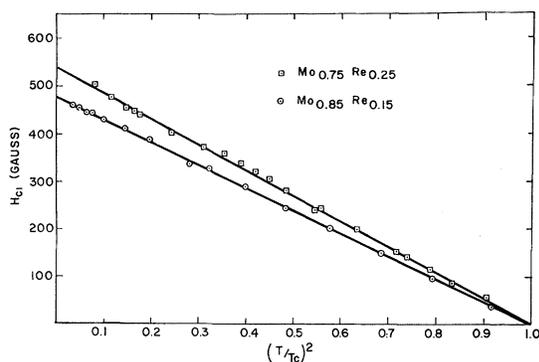


FIG. 3. Lower critical field,  $H_{c1}$ , for  $\text{Mo}_{0.85}\text{Re}_{0.15}$  and  $\text{Mo}_{0.75}\text{Re}_{0.25}$  vs reduced temperature squared,  $(T/T_c)^2$ .

$H_{c2}$  as determined by the magnetization measurements shows a considerable departure at lower temperatures. We do not know how to account for the discrepancy between the values of  $H_{c2}$  as determined by the magnetization and resistance measurements. However, since our  $K$  determined from  $(dM/dH)_{H_{c2}}$  shows good agreement with the values of  $K$  calculated in other independent ways, and since the values of  $H_0$  as determined from the areas under the magnetization curves are in reasonable agreement with those calculated from the specific heat, we believe that our magnetization curves do not contain any inherent experimental inaccuracies, and hence give the true bulk upper critical field.

In summary, therefore, measurements on carefully prepared, homogeneous, Mo-Re alloys give further support to the GLAG theory of superconductors of

the second kind. The lower critical field is found to have a  $T^2$  temperature dependence, while the upper critical field shows a temperature dependence not described adequately by present theory. Low current density resistive transitions show that full resistance is not restored until fields much greater than those required to produce zero magnetization are reached. Oscillations are observed in the magnetization of a sample showing a nearly reversible magnetization curve, and thus would seem to have their origin in a mechanism other than a large scale defect structure.

We wish to thank Robert Pautienus for his considerable assistance in assembling the apparatus and

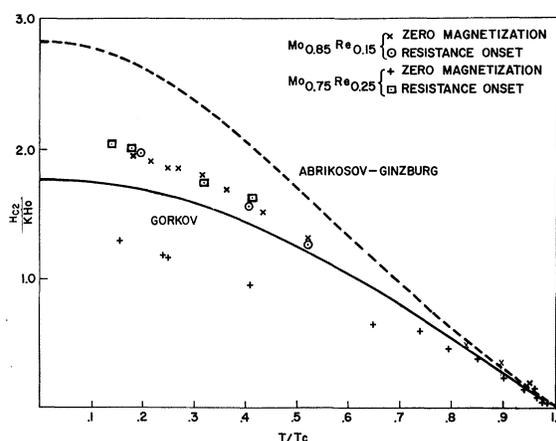


FIG. 4. Temperature dependence of upper critical field  $H_{c2}$  for  $\text{Mo}_{0.85}\text{Re}_{0.15}$  and  $\text{Mo}_{0.75}\text{Re}_{0.25}$ . Data is normalized by plotting  $H_{c2}/KH_0$  vs reduced temperature  $(T/T_c)$ . The data is compared to the predictions of Abrikosov-Ginzburg (upper dashed curve) and Gor'kov (lower solid curve).

in taking the measurements. Dr. Robert Gayley of the University of Maryland kindly made available a spark cutter. We appreciate receiving the data of Chandrasekhar, Hulm, and Jones in advance of its publication.