TABLE II. The tin energy gap from acoustic-attenuation data.[a]

Propagation direction θ	φ		Energy gap ($kT_c/2$)
0°	···	[001]	3.1±0.1
90°	0°	[100]	3.5±0.1
90°	12°		4.0±0.2
90°	18°		4.3±0.2
90°	30°		4.0±0.2
90°	45°	[110]	3.8±0.1

[a] Reference 6, p. 170.

above expression and should, in fact, be proportional to a weighted average of contributions corresponding to various Δ_m's from different pieces of the Fermi surface. It is reasonable to assume, however, that the lowest gap value listed in Table I, for a given propagation direction, is the dominant one due to the exponential character of (IV.4). Table II gives some results of acoustic attenuation experiments in which the experimental curves have been fitted into a single exponential term. It is interesting to note that experiment indicates that the gap observed for propagation in the basal plane takes a maximum value of 4.3 for the $\varphi=1.8°$ direction.[2] The trace of the third-zone surface in the (100) plane through Γ is largely determined by the lattice symmetry and the nearly-free-electron character of the surface. The normals to that piece are such that it only contributes for sound-propagation directions between $\varphi=18°$ and $\varphi=45°$. It is almost certain that the discontinuity at $\varphi=18°$ accounts for the observed extremum. It is quite likely that the addition of terms in (III.1) with a φ variation more rapid than $\cos 4\varphi$ would improve the numerical agreement, in particular, for propagation directions off the symmetry axes.

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Magnetic Properties of Superconducting Mo-Re Alloys

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This paper gives details of research carried out on the low-temperature properties of Mo–Re alloys in the superconducting state. Measurements were made on a 52–48% alloy of Mo–Re both unannealed and annealed, the alloy being in the form of fine wires. Micrograph studies were made to determine the percentage of the various phases present in each specimen. The low-temperature measurements covered observations of the magnetization as a function of applied magnetic field at various temperatures using two different techniques. The measurements yielded the critical magnetic fields H_{c1} and H_{c2} as a function of temperature and of the state of anneal, as well as of the transition temperature T_c. Estimates were made of $H_c(T)$ and the Ginzburg-Landau-Abrikosov-Gor'kov and Maki parameters κ, $\kappa_1(T)$, and $\kappa_3(T)$. Comparisons of the results are made with results obtained previously by us from resistivity measurements on the same alloy and by other authors on similar superconducting alloys; the comparisons show consistency in the data. Our evaluations of $\kappa_1(T)/\kappa_1(T_c)$ and of $\kappa_3(T)/\kappa_3(T_c)$ are consistent with the theory of Maki.

I. INTRODUCTION

THE work reported in this paper is an extension of an investigation of the general properties of Mo–Re alloys, which has been partly reported by two of us earlier.[1] This alloy, which has the general properties of a "high-field" superconductor, was chosen because its superconductivity can be quenched in readily available fields of less than about 40 kG, and because it was desired to study the properties of the normal state also. One of the primary motivations of the work was to make a wide variety of different measurements (resistivity, thermal conductivity, magnetic moment, etc.) on actually the *same* sample of each alloy in various states of anneal. Such a study was anticipated to yield a pattern of behavior showing some inner consistency.

This paper covers the measurements of the magnetic properties of Mo–Re alloys in the form of fine wires, both annealed and unannealed. The wire was from the same spool as that used previously in the electrical and

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[1] E. Lerner and J. G. Daunt, Phys. Rev. 142, 251 (1966).

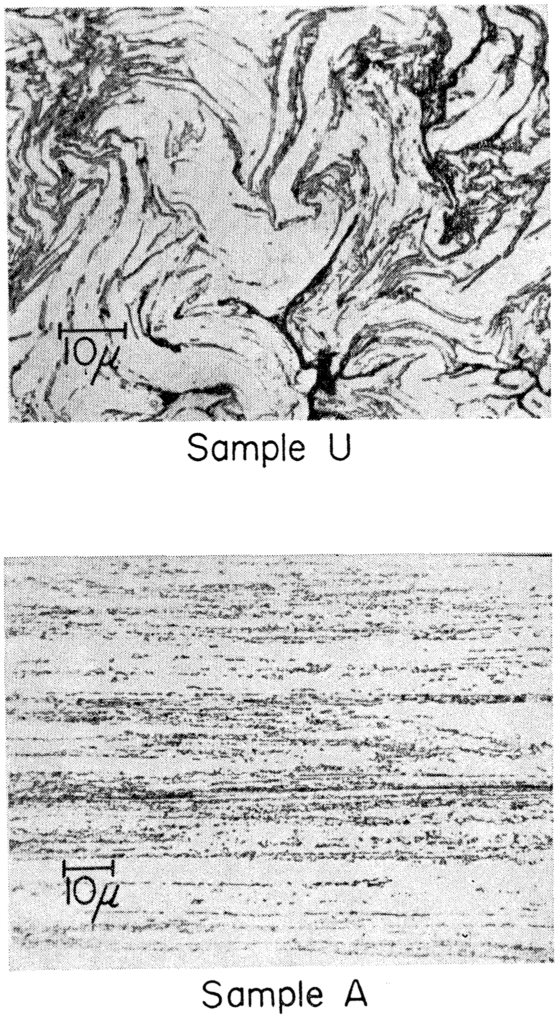

FIG. 1. Microphotographs of unannealed and annealed Mo–Re samples: sample U (1380× magnification); sample A (1000× magnification).

thermal measurements.[1] Data are given for T_c, H_{c1}, and H_{c2}, and estimates are made for H_c, $\kappa_1(T)$, $\kappa_1(T)/\kappa_1(T_c)$, and $\kappa_3(T)/\kappa_3(T_c)$; comparisons have been made with theory. In addition, some conclusions have been reached concerning the role played in the behavior of these alloys by annealing and by the presence of the σ phase.

II. SAMPLE SPECIFICATION

The sample consisted of a compact cylindrical bundle of superconducting Mo–Re (52–48 wt%) wires. Each wire was 0.024 cm in diameter, 1.21 cm long, and the bundle of diameter 0.98 cm weighed 6.68 g, giving a volume of metal equal to 0.48 cm^3. The wires in the bundle were held together by means of GE 7031 varnish. The sample was made from the same spool of wire, supplied by Chase Brass & Copper Company, that was used for the electrical and thermal resistivity measurements reported earlier by us.[1] Hereafter this earlier paper is referred to as I.

The chemical analysis of the wire is given in I. The 52–48% Mo–Re alloy contains two phases, a body-centered-cubic (bcc) β phase and a complex tetragonal σ phase, at temperatures below 1250°C.[2] Experiments were performed with the sample unannealed (sample U), i.e., 90% cold-worked as received from the manufacturer, and with the sample annealed (sample A). The annealing was in vacuum for 24 h at 1080°C with subsequent slow cool. From the microphotographs (see Fig. 1) the amount of σ phase in sample A was estimated by statistical sampling technique to be about 22%. Assessment of the amount of σ phase in sample U was not possible because of the masking effect of the cold work.

III. EXPERIMENTAL ARRANGEMENTS

The magnetic properties were measured by two different methods. In one, the magnetic moment was measured using a moving search coil, the so-called flip-coil method,[3] and in the other, referred to as the dynamic method, the search coil was fixed and the magnetic field swept.

A. The Flip-Coil Method

In our arrangement[4] an external homogeneous magnetic field, longitudinal to the sample, was supplied by a long solenoid submerged in a liquid-nitrogen bath. The sample was fitted tightly into a German-silver can which was mounted inside a vacuum jacket immersed in liquid helium. To assure good thermal contact between the sample and the can, silicone grease was used around the specimen. For measurements at temperatures above 4.2°K, a heater was wound around the can. The temperature of the sample was measured by means of a carbon resistor which made thermal contact with the can by means of GE varnish. The calibration of the thermometer was performed as described in I.

In order to calibrate this apparatus so that the magnetic moment of the sample could be directly related to

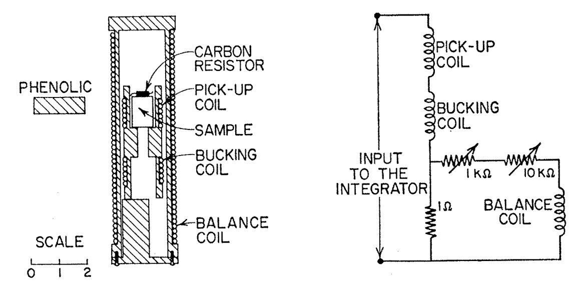

FIG. 2. (a) Diagram of sample and search coils used in the dynamic method. (b) Circuit showing search-coil connections.

[2] J. M. Dickinson and L. S. Richardson, Trans. Am. Soc. Metals **51**, 1055 (1959).
[3] K. Mendelssohn, Proc. Roy. Soc. (London) **A155**, 558 (1936).
[4] E. Lerner, M.S. thesis, Ohio State University, 1963 (unpublished).

the deflections of the ballistic galvanometer connected to the search coil; preliminary experiments were made using a pure-Pb sample of demagnetization factor $n=0.08$. The initial isothermal slope $(H/\theta)_i$ of external field versus galvanometer deflection in the completely diamagnetic region was observed and this was related to the magnetic moment of the sample m by the equation

$$k\theta=m=-HV/4\pi(1-n), \quad (1)$$

since in this initial region $\chi=-1/4\pi$. In this equation k is the calibration constant and V the volume of the specimen.

B. The Dynamic Method

In the dynamic method the external (longitudinal) magnetic field was changed continuously and the sample was fixed inside a pickup coil which was connected in series opposition with a similar bucking coil. The flux induced in the pickup coil by the superconducting sample was integrated by an Astrodata amplifier (Nanovolt Integrator, Model 122R) and the magnetization versus field curve recorded on an X-Y recorder. The pickup and bucking coils consisted of 1398 turns of No. 40 AWG copper wire wound in 10 layers with average turn diameter of 1.42 cm. Figure 2(a) shows the arrangement of these coils and the sample. To obtain a finer balance, a third coil [the balance coil of Fig. 2(a)] was wound as shown with 2160 turns of No. 34 AWG copper wire in five layers with average turn diameter 1.98 cm. The complete arrangement of coils and sample was immersed in a bath of liquid helium. The third coil was connected as shown in Fig. 2(b). All soldering was done with low-thermal-emf solder, and the other connections with copper screws and washers.

The temperature of the samples was measured by a carbon resistor attached to them with GE varnish and that of the bath by vapor-pressure thermometry. A heater was wound directly on the sample. It consisted of a few turns of Evanohm wire wound noninductively. This heater was used to warm the sample above its transition temperature and so to get rid of any frozen-in magnetic flux before starting each magnetization curve. The heater was also used to maintain the sample above 4.2°K, which was done by maintaining a small current through it when the level of the liquid-helium bath was below the sample.

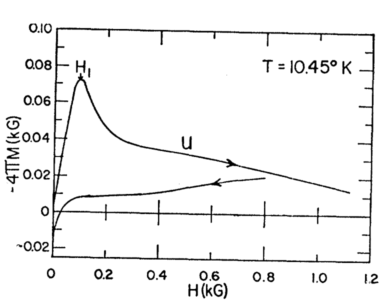

FIG. 3. Typical magnetization curve for sample U taken at $T=10.45$°K with the flip-coil method.

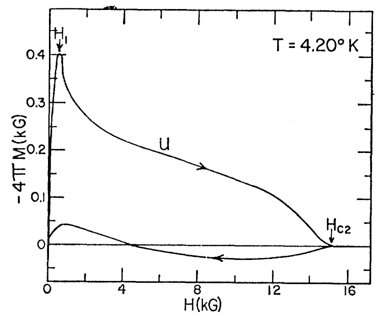

FIG. 4. Isothermal magnetization at $T=4.20$°K showing the magnetization of sample U as a function of external magnetic field H (kG) for both increasing and decreasing field using the dynamic method.

IV. EXPERIMENTAL RESULTS

A. The Flip-Coil Method

A typical isothermal measurement of the magnetization M of sample U versus applied field H taken at 10.45°K is given in Fig. 3. The critical magnetic field H_{c1} can be estimated from these isothermal magnetization curves after correction for the demagnetization factor n of the specimen. To get n from our experimental data, we assume that the observed initial slope $(H/\theta)_i$ of the external field versus galvanometer deflection corresponds to a completely diamagnetic susceptibility. Whence, using Eq. (1), the demagnetization factor n is given by

$$(1-n)=-(V/4\pi k)(H/\theta)_i, \quad (2)$$

provided that the magnetization is *uniform* throughout the specimen. This condition is apparently not fulfilled, since a literal application of Eq. (2), using our data, yields $n=0$. This would be the case if the individual wires of the bundle acted independently of one another, which is hardly reasonable. The other extreme case would be that of a solid cylinder with the same aspect ratio as the bundle. For this configuration the appropriate value of n is 0.16 (see Bozorth and Chapin[5]). The actual situation is presumably somewhere in between. Inasmuch as the filling factor of the bundle is about 0.5, we have assumed $n=0.08$, half the value for that of a solid cylinder.

For perfect diamagnetism and uniform sample magnetization $(-4\pi M)$ at the peak of the magnetization curve is equal to $H_{c1}/(1-n)$. For a nonuniform magnetization, due to nonellipsoidal geometry, we would expect the height of the observed peak to be reduced and blunted, as is evident in Figs. 3 and 4. For relatively narrow peaks, however, the resulting error in H_{c1}, selected by putting $H_1=H_{c1}(1-n)$, should be small. H_1 is the value of the external field at the peak in the magnetization curve, as marked, for example, in Figs. 3 and 4. No demagnetization correction is necessary for H_{c2}. The values of H_{c1} and H_{c2} obtained in this way for sample U are given in Table I and Figs. 5 and 6. In the

[5] R. M. Bozorth and D. M. Chapin, J. Appl. Phys. 13, 320 (1942).

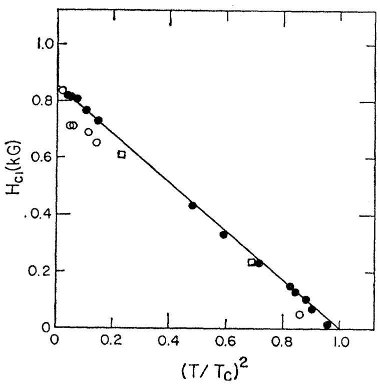

FIG. 5. Plot of H_{c1} (kG) versus $(T/T_c)^2$ for samples U and A using flip-coil and dynamic method as follows: solid circle—sample U (flip-coil method); open circle—sample U (dynamic method); open square—sample A (dynamic method).

flip-coil method the upper critical field H_{c2} was obtained at only two temperatures since sufficiently high fields were not obtainable with the liquid-nitrogen solenoid. The flip-coil method also permitted the transition temperature T_c of the samples to be observed. T_c was obtained from measurements of the remanent magnetization as a function of temperature. The temperature at which the frozen-in magnetic moment became zero then identified T_c. The value obtained was $T_c=11.1$°K for both samples U and A, in agreement with the resistivity measurements (see I).

B. The Dynamic Method

Typical magnetization curves for samples U and A taken at 4.20°K are shown in Figs. 4 and 7, respectively. H_{c1} and H_{c2} are obtained from these curves, as noted previously, taking $H_{c1}=H_1/(1-n)$. The results are listed in Table II and shown in Figs. 5 and 6.

The values of H_{c1} determined by the flip-coil method are more reliable than those obtained by the dynamic method. As was explicitly shown by the experiments of Goedemoed *et al.*,[6] heating effects associated with the transition occur in the dynamic method. As they pointed out, the magnetization in the case of a continuously varying field may show retardation effects and that their magnitude will be a strong function of the thermal time constants of the specimen. Although for uniform magnetization these effects should not be present at fields less than H_{c1}, in our case they may occur because of nonuniformity of the magnetization. With the flip-coil method the magnetization is measured point by point as the field is varied, and always sufficient time is allowed between successive readings for the attainment of equilibrium. It is concluded therefore that the most reliable data for H_{c1} are those given by the full curve of Fig. 5, which weights significantly the flip-coil data. It is of interest to note that H_{c1} appears in first approximation to be proportional to $(1-T^2/T_c^2)$. The retardation effects are less serious at the high-field end of the magnetization curves, and it is considered that the values of H_{c2} determined by the dynamic method are relatively free from the criticisms made above for similar determination of H_{c1}.

TABLE II. (a) Sample U (dynamic method). (b) Sample A (dynamic method).

(a) T (°K)	T/T_c	H_{c1} (G)	H_{c2} (kG)	(b) T (°K)	T/T_c	H_{c1} (G)	H_{c2} (kG)
1.24	0.112	...	19.5	1.16	0.104	...	16.2
1.61	0.145	836	20.1	1.94	0.175	...	15.0
2.48	0.224	...	18.3	2.27	0.204	...	14.3
2.62	0.236	713	17.9	2.98	0.268	...	13.8
2.70	0.243	713	17.0	3.48	0.314	...	13.3
3.20	0.288	...	16.5	3.72	0.335	...	13.1
3.80	0.342	682	15.9	4.20	0.378	...	12.5
4.20	0.378	651	15.0	4.50	0.410	...	11.5
4.20	0.378	651	15.1	5.25	0.473	...	10.9
4.35	0.392	...	14.8	5.40	0.486	613	9.8
4.70	0.424	...	12.8	6.40	0.576	...	8.0
5.80	0.522	...	11.0	7.40	0.666	...	6.1
6.10	0.549	...	10.6	7.62	0.685	...	6.2
				7.95	0.715	...	6.9
				9.20	0.829	238	...

V. DISCUSSION

It is of interest to compare the H_{c2} values given here with those determined by electrical-resistivity measure-

TABLE I. Sample U (flip-coil method).

T (°K)	T/T_c	H_{c1} (G)	H_{c2} (kG)
2.05	0.185	810	...
2.68	0.242	808	...
3.09	0.278	802	...
3.60	0.324	766	...
4.32	0.388	730	...
7.70	0.694	434	...
8.55	0.770	332	...
9.40	0.845	229	...
10.10	0.910	145	...
10.20	0.919	133	...
10.45	0.940	103	...
10.55	0.950	72	1.13
10.90	0.980	12	0.25

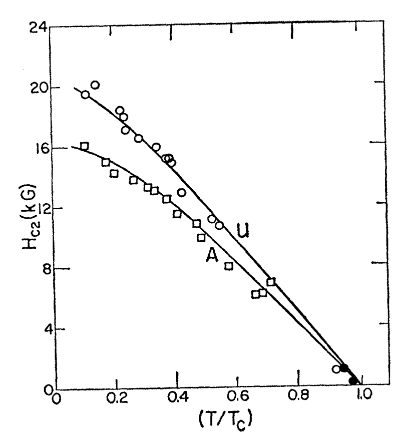

FIG. 6. Plot of observed values of H_{c2} (kG) versus (T/T_c) for samples U and A as follows: solid circle—sample U (flip-coil method); open circle—sample U (dynamic method); open square—sample A (dynamic method).

[6] S. H. Goedemoed, C. Van Kolmeschate, D. de Klerk, and C. J. Gorter, Physica 30, 1225 (1964).

ments $H_{c2}{}^r$ on Mo–Re wire taken from the same spool, as reported in I. For both samples and for all temperatures, $H_{c2}{}^r$ has higher values than H_{c2}, obtained from the magnetic measurements. It is possible that the values quoted for $H_{c2}{}^r$ are actually too high, because of the difficulty of determining precisely the exact magnetic field at which the first sign of resistivity appears as the field is increased. Even so, our results are qualitatively quite different from those of Joiner and Blaugher,[7] who found H_{c2} and $H_{c2}{}^r$ to be essentially the same for a very pure $Mo_{85}Re_{15}$ sample which consisted of a few large crystallites. On the other hand, as the work of Gygax, Olsen, and Kropschot[8] on indium-lead alloys clearly shows, $H_{c2}{}^r$ can take values either greater or less than H_{c2}, depending on the value of the measuring current density. As the measuring current tends to zero, they show that $H_{c2}{}^r$ tends to H_{c3}. We conclude therefore that for our Mo–Re samples only the values of H_{c2} taken by magnetic measurements are the valid ones. The value of $H_{c2}=15$ kG at $T=4.2°$K is in good agreement with Kunzler's[9] result on the first measurements on Mo–Re alloys.

Significant effects of annealing appear in the magnetization curves. On the other hand, the transition temperature appears to be independent of the state of anneal within the precision of measurement. Figure 6 shows that sample U showed higher H_{c2} values than sample A. The shape of the magnetization curve as the field is reduced from its maximum value to zero is completely different for samples U and A, as can be seen from Figs. 4 and 6. This kind of behavior was noticed by Livingston[10] in Pb–Cd and Pb–Na alloys and by Joiner[11] in In–Sn and In–Al–Sn alloys, who attributed it to precipitation of one component. Our results support this concept in that the microphotographs revealed precipitation of the σ phase in sample A.

For ideal type-II superconductors having reversible magnetization, as measured under conditions in which retardation effects are negligible, the critical field H_c is given by

$$\int_0^{H_{c2}} MdH=-\frac{H_c^2(T)}{8\pi}. \tag{3}$$

In our measurements made by the dynamic method the temperature of the sample was monitored and no temperature differences larger than about 0.1°K were observed. As a result the estimated errors in the evaluation of the integral in Eq. (3) introduced by retardation effects were 2% or less for temperatures at or below 4°K. Nevertheless, the observed irreversibilities in the magnetizations introduce possible errors in evaluation of $H_c(T)$ by Eq. (3) which are not readily assessible from the magnetic data alone. We therefore do not present tabulations of evaluations of $H_c(T)$ by Eq. (3) in our experimental results. However, it is possible to estimate these possible errors for temperatures near T_c in the following manner.

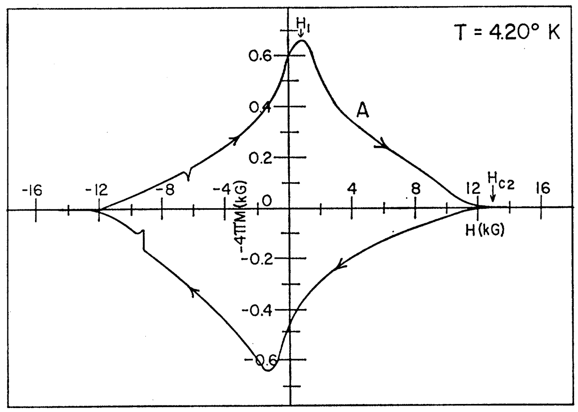

FIG. 7. Isothermal magnetization at $T=4.20°$K showing the magnetization of sample A as a function of external magnetic field H (kG) for both increasing and decreasing field using the dynamic method. The kinks in the curve are caused by flux jumps.

The value of $\kappa_1(T)$, formulated by Maki,[12] approaches that of κ defined by Abrikosov[13] as T tends to T_c and is given by

$$\kappa_1(T)=H_{c2}(T)/\sqrt{2}H_c(T). \tag{4}$$

Near T_c, therefore, we can make the approximation $H_c(T)\approx H_{c2}(T)/\sqrt{2}\kappa$ and employ Goodman's[14] expression

$$\kappa=\kappa_0+7.5\times10^3\gamma^{1/2}\rho \tag{5}$$

for estimating κ, using our previous determinations made in I of ρ and using Morin and Maita's[15] evaluations of γ. The $H_c(T)$ values determined in this way can then be compared with those estimated from Eq. (3).

Neglecting κ_0, the value of κ from Eq. (5) for sample U is 4.12. At, for example, $T/T_c=0.9$ the argument above yields a value of $H_c(T)$ equal to 0.41 kG. $H_c(T)$ as determined from the magnetization curve using Eq. (3) was 10% higher. A similar comparison at $T/T_c=0.8$ shows the same percentage difference. This comparison encourages us to present tentative estimates of $H_c(T)$ as deduced from our magnetization data using Eq. (3) and to evaluate therefrom $\kappa_1(T)$ from Eq. (4) and $\kappa_3(T)$, although we realize that these estimates of $H_c(T)$ may be about 10% too high and the corresponding values of $\kappa_1(T)$ the same percentage too low, because of the effects of the irreversibilities. The parameter $\kappa_3(T)$, introduced by Maki,[16] can be evaluated by use of

[7] W. C. H. Joiner and R. D. Blaugher, Rev. Mod. Phys. **36**, 67 (1964).
[8] S. Gygax, J. L. Olsen, and R. H. Kropschot, Phys. Letters **8**, 228 (1964).
[9] J. E. Kunzler, Rev. Mod. Phys. **33**, 501 (1961).
[10] J. D. Livingston, J. Appl. Phys. **34**, 3028 (1963).
[11] W. C. H. Joiner, Conference on Physics of Type II Superconductivity, Western Reserve University, Cleveland, 1964, p. IV-75 (unpublished).
[12] K. Maki, Physics **1**, 21 (1964).
[13] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].
[14] B. B. Goodman, IBM J. Res. Develop. **6**, 63 (1962).
[15] F. J. Morin and J. P. Maita, Phys. Rev. **129**, 1115 (1963).
[16] K. Maki, Physics **1**, 127 (1964).

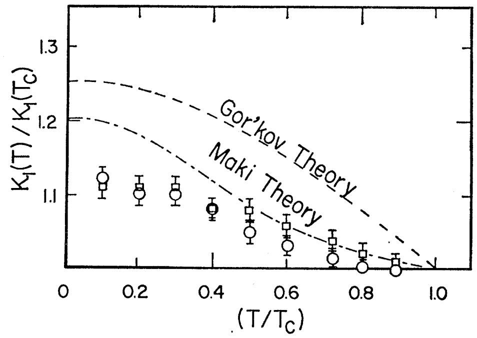

FIG. 8. Plot of our estimations of $\kappa_1(T)/\kappa_1(T_c)$ versus T/T_c. (Points: open circle—sample U; open square—sample A.) Dashed curve shows theoretical evaluation by Gor'kov (Ref. 18); dashed and dotted curve shows theoretical evaluation by Maki (Ref. 12).

Harden and Arp's[17] computations relating $\kappa_3(T)$ with H_{c1}/H_c. It should be pointed out also that the ratios $\kappa_1(T)/\kappa_1(T_c)$ and $\kappa_3(T)/\kappa_3(T_c)$, which can finally be deduced and which are the pertinent parameters for comparisons with theory, should suffer much less than the absolute values of $\kappa_1(T)$ and $\kappa_3(T)$ from the errors introduced by the irreversibilities in the magnetizations. In Table III we list these estimates of $H_c(T)$, $\kappa_1(T)$, and $\kappa_3(T)$. The values of $\kappa_1(T)$ and $\kappa_3(T)$ have been extrapolated to $T=T_c$ and these are also given in Table III in parentheses. It is evident, as can be seen in I also, that annealing has a marked effect on the κ values, lowering the magnitude of $\kappa_1(T)$ and raising that of $\kappa_3(T)$. Moreover, the value of $\kappa_1(T_c)$ for sample A given in Table III is in conformity with the findings in I of the κ values of annealed samples deduced from resistivity measurements.

Figure 8 shows the estimated values of $\kappa_1(T)/\kappa_1(T_c)$ versus T/T_c. This ratio increases with decreasing temperature by about 11% over the whole range of temperature from T_c to 0°K. It is of interest to note that it is relatively unaffected by the state of anneal of the samples. These results are in qualitative agreement with the theoretical predictions of both Gor'kov[18] and Maki,[12] which are shown by the broken lines in Fig. 8. Our results lie below both theories, but within our possible errors seem to support the theory of Maki. As is evident from Table III, $\kappa_3(T)/\kappa_3(T_c)$ also increases monotonically with decreasing temperature, as predicted by Maki.[16] Our estimations show that this ratio increases by about a factor of 2 in going from T_c to 0°K. There appears also to be, as an effect of the annealing, the lowering of $\kappa_3(T)/\kappa_3(T_c)$ values for the annealed sample by about 6%.

TABLE III. The columns under U and A refer, respectively, to the unannealed and annealed samples. The numbers in parentheses are extrapolations.

	$H_c(T)$ (kG)		$\kappa_1(T)$		$\kappa_3(T)$	
T/T_c	U	A	U	A	U	A
0.1	3.32	3.55	4.20	3.15	6.7	7.5
0.2	3.09	3.31	4.14	3.16	6.3	7.1
0.3	2.81	3.04	4.13	3.16	5.9	6.7
0.4	2.50	2.71	4.07	3.13	5.6	6.3
0.5	2.15	2.35	3.96	3.07	5.3	6.0
0.6	1.77	1.92	3.89	3.02	5.0	5.7
0.7	1.34	1.48	3.81	2.96	4.7	5.4
0.8	0.90	1.00	3.78	2.90	4.2	5.0
0.9	0.45	0.50	3.77	2.87	4.0	4.6
1.0	···	···	(3.76)	(2.84)	(3.6)	(4.3)

In principle it could be possible to calculate $\kappa_2(T)$ from magnetization curves using Maki's[12] extension of Abrikosov's theory,[13] using the equation

$$(dM/dH)_{H_{c2}}=[1.18(4\pi)(2\kappa_2^2(T)-1)]^{-1}. \qquad (6)$$

It was considered, however, that $(dM/dH)_{H_{c2}}$ is too sensitive to effects of irreversibilities in the magnetization to permit reliable evaluation of $\kappa_2(T)$ from Eq. (6). However, it appeared from our data for both samples A and U that $\kappa_2(T)$ diminished as T/T_c was reduced, in qualitative agreement with Maki's theory.

ACKNOWLEDGMENTS

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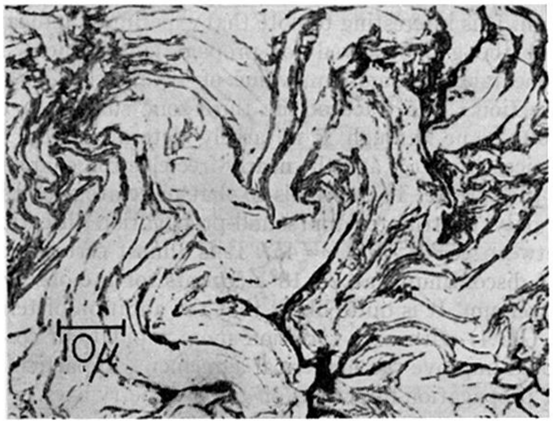
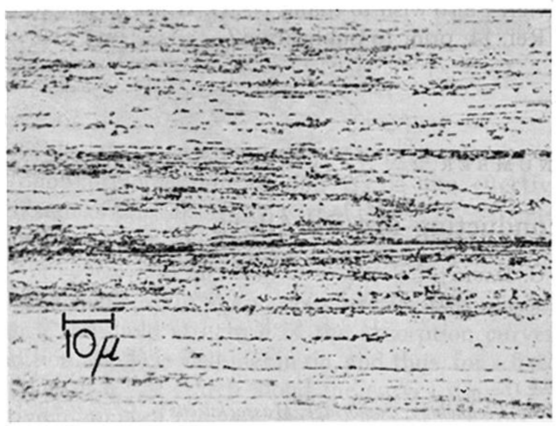


Sample U



Sample A

FIG. 1. Microphotographs of unannealed and annealed Mo–Re samples: sample U (1380× magnification); sample A (1000× magnification).