being fairly low, the shielding over the theoretical curve was fairly small. By decreasing the field, the trapped flux first decreases because diamagnetism is increasing, and then the trapped flux increases when diamagnetism disappears.

LIVINGSTON: This peak region occurs out in the high-field tail where the reversible curve has quite a measurable constant slope. We plot the difference and see quite a sizeable peak. It is a peak in hysteresis and is not associated with reversible curves in any way.

GOODMAN: I think one point which confirms Dr. Livingston's interpretation is that the increasing and decreasing curves are symmetrical with respect to what is predicted with reversible behavior. That should answer your question.

LIVINGSTON: It was in that curve, but I must remark in all fairness that not all curves come out so symmetric.

MENDELSSOHN: Just a mention to Professor Goodman, I am afraid a pure Pb ring does the same thing. It also yields a symmetrical magnetization curve.

Variation of the Critical Fields of Superconducting Lead with the Residual Resistivity

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INTRODUCTION

In this paper, experiments are reported on the influence of the residual resistivity ρ on the critical fields of superconducting lead. The residual resistivity has been increased by alloying, cold-working, or neutron-irradiation. For the description of the superconducting behavior, two groups of critical fields are defined.

I. Magnetization measurements as a function of a longitudinal field give: H_{c1}, the field at which the first macroscopic penetration occurs; H_{c2}, the field where the magnetic moment vanishes, or at least is smaller than the detection threshold; H_c, the thermodynamic critical field, as determined from the area under the magnetization curve.

In our experiments the magnetic moment M was measured with a null-coil magnetometer.[1] H_{c1} can be determined also from noise observations[2]; this effect is an analog of the Barkhausen noise in a ferromagnetic substance.

II. From resistance measurements as a function of the field, one can obtain the fields which restore a fraction 0.01, 0.50, or 0.99 of the normal resistance in longitudinal (l) or in transverse (tr) field: H_c (0.01, tr), H_c (0.99, tr), H_c (0.01, l), H_c (0.50, l), and H_c (0.99, l).

It was found that for a number of alloys these fields were much larger than H_{c2}. In order to study this more closely, the influence of the transport current on these fields was measured (J–H curves).

EXPERIMENTAL

Wires, diam 0.3 mm, made from lead–indium alloys were annealed for 14 days *in vacuo* at about 50°C below the melting point. The residual resistivity and the critical fields H_{c1}, H_{c2}, H_c, H_c (0.01, tr), H_c (0.99, tr), H_c (0.01, l), and H_c (0.99, l) were measured; the results are given in Table I. The J–H curves of the alloys were measured with J ranging from 0.15 $\mathrm{A \cdot cm^{-2}}$ to 4.6×10^3 $\mathrm{A \cdot cm^{-2}}$. The results on the lead–6.6 at. % indium alloy are given in Fig. 1 with a logarithmic J scale, and in Fig. 2 with a linear J scale.

For the deformation and irradiation, both at 78°K, 99.99% Johnson and Matthey lead was used. The deformation was done by cold-rolling to an increase in length of about 120%. The neutron-irradiation was carried out in the low flux reactor BR1 of the SCK/CEN at Mol (Belgium), the integrated flux being about 10^{18} fast neutrons per cm^2. The increase in residual resistivity ρ and H_c (0.50, l) were measured. The results are given in Table I; a more detailed description is given in Refs. 3 and 4.

DISCUSSION

The dependence of H_{c2} on the residual resistivity given by the GLAG theory has been shown by Livingston[5] to be in excellent agreement with experiment (sensitivity of $4\pi M = \pm 5$ Oe). Our results (sensitivity $4\pi M = \pm 1$ Oe) also agree with this

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[5] J. D. Livingston, Phys. Rev. 129, 1943 (1963).

TABLE I. Concentrations, residual resistivities ($\mu\Omega$-cm), and critical fields (oersted) at 4.2°K of lead–indium alloys and of cold-worked and neutron-irradiated lead.

at. % In	ρ	H_{c1}	H_{c1} (noise)	H_{c2}	H_c	$\frac{H_{c2}}{H_c}$	$H_c(0.01, \text{tr})$ $J = 10$ $(\text{A}\cdot\text{cm}^{-2})$	$H_c(0.99, \text{tr})$ $J = 10$ $(\text{A}\cdot\text{cm}^{-2})$	$H_c(0.01, \text{l})$ $J = 10$ $(\text{A}\cdot\text{cm}^{-2})$	$H_c(0.99, \text{l})$ $J = 10$ $(\text{A}\cdot\text{cm}^{-2})$
0	0.004	550	550	550	550	1				
0.03	0.07						490	610	675	865
0.06	0.14						530	920	710	1100
0.10	0.22						525	720	730	1200
0.16	0.35						535	810	730	1050
0.3	0.82	550		830	675	1.2	920	2350	1050	2750
0.5	1.1	600		960	700	1.4	740	1600	1050	2500
0.9	1.5	545		905	675	1.3	1000	2100	1150	2350
5.6	4.1	365		1450	645	2.2	2400	3900	2650	3900
6.6	5.0	335	250	1850	630	3.0	2400	3500	2900	3900
15.2	10.5	245		2650	650	4.1	2750	4350		
17.0	11.7	195	210	3300	575	5.8	2850	4650		
23.9	13.7	155	180	3550	545	6.5	4000	5850		
24.1	14.9	195	250	3450	620	5.5	3600	5750		

	ρ	$H_c(0.50, \text{l})$ $(J = 10\ \text{A}\cdot\text{cm}^{-2})$
Cold-rolled 99.99% Johnson–Matthey lead	0.17	1000 $(H_c + 450)$
Neutron-irradiated 99.99% Johnson–Matthey lead	0.015	600 $(H_c + 47)$

theory; see Fig. 3. The relation between H_{c1} and H_{c2} is found to be in accordance with Abrikosov's assumption for low κ values. With the resistance measurements, critical fields exceeding H_{c2} are observed; see Table I and Fig. 1. Superconductivity at fields above H_{c2} was found also by Berlincourt and Hake,[6] Autler *et al.*,[7] and Chiou *et al.*[8] However, from a linear J–H plot, as given in Fig. 2, it is seen that only at low current densities superconductivity above H_{c2} occurs. Extrapolation to $J = 0$ of the high current density part of the curve yields for H_c (0.01, l) a value equal to the magnetically determined H_{c2}.

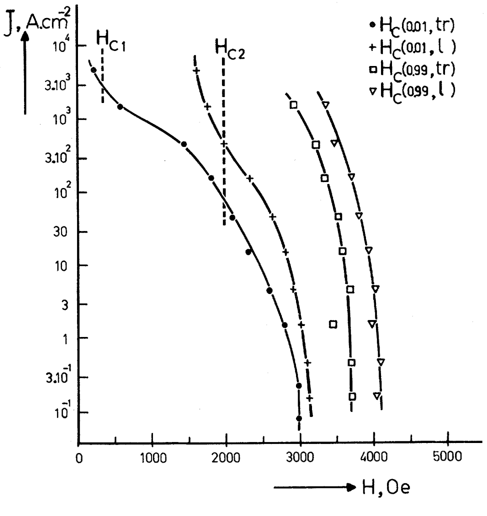

FIG. 1. J–H curves of a lead -6.6 at. % indium alloy, in transverse (tr) and longitudinal (l) fields. The critical fields are defined as the fields that restore 0.01 or 0.99 of the normal resistance. The values of H_{c1} and H_{c2} as found from magnetization measurements, are indicated.

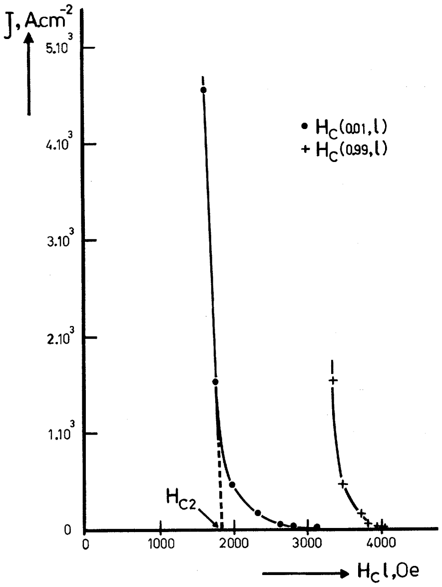

FIG. 2. Linear plot of J versus H_c (0.01, l) and H_c (0.01, tr) of a lead -6.6 at % indium alloy. The extrapolation of H_c (0.01, l) from high current densities to $J = 0$ is indicated.

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[6] T. G. Berlincourt and R. R. Hake, Phys. Rev. Letters **9**, 293 (1962); see also *Proceedings of the Eighth International Conference on Low Temperature Physics London, 1962* (Butterworths Scientific Publication Ltd., London, 1962). The critical field of a Ti–V alloy found from resistance measurements was higher than the calculated value of H_{c2} from the GLAG theory.

[7] S. H. Autler, E. S. Rosenblum, and K. H. Gooen, Phys. Rev. Letters **9**, 489 (1962). Superconductivity was found on niobium above H_{c2}, that was defined from resistance measurements.

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The field at which the last trace of superconductivity above H_{c2} disappears is considered as a new critical field and is called H_{c3}. It is obtained from the low current density part of the log $J - H_c$ (0.99, 1) curve, given in Fig. 1. H_{c3}/H_c is plotted vs ρ in Fig. 3; H_{c3} of the alloys having $\rho > 5$ $\mu\Omega$-cm was determined from the log J–H_c (0.99, tr) curves. The H_{c3}/H_c curve in Fig. 3 is nearly parallel to the H_{c2}/H_c curve if $\rho > 1$ $\mu\Omega$-cm, which is the critical value of ρ for the onset of type II superconductivity in lead. The slope of the $H_{c3}/H_c - \rho$ curve at $\rho = 0$ $\mu\Omega$-cm is about 5 $\mu\Omega^{-1}$ cm^{-1}.

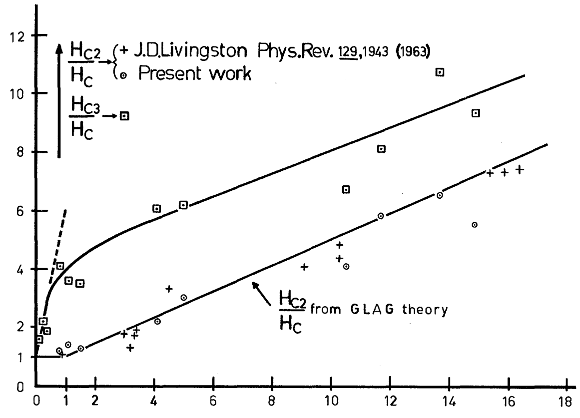

FIG. 3. H_{c2}/H_c calculated from the GLAG theory; H_{c2}/H_c from magnetization measurements; H_{c3}/H_c obtained from low current density resistance measurements vs the residual resistivity of lead alloys.

From the results of the above resistance measurements, one should expect a small magnetization to be found between H_{c2} and H_{c3}. This, however, was not shown by our magnetization measurements. The parts of the M–H curves near H_{c2}, which agree quantitatively with the Abrikosov theory, do not justify such an approach of M to the H axis, that would be needed to find a critical field $\gg H_{c2}$. From this it is concluded that the volume that remains superconducting above H_{c2} must be very small. This residual superconductivity might, therefore, be attributed to small inhomogeneities or structural defects.

Superconductivity above H_{c2} was found also on the cold-worked and the irradiated pure lead. The influence of deformation on the critical field, as inferred from resistance measurements, had already been established by Shaw and Mapother[9] for lead and by Hauser and Buehler[10] for niobium and rhenium. Magnetization measurements on our cold-worked and irradiated samples could not be carried out with the present magnetometer. However, ρ of these wires (see Table I) is far below the critical value of 1 $\mu\Omega$-cm, so according to the GLAG theory H_{c2} must be equal to H_c (=550 Oe for lead at 4.2°K). $[H_c\ (0.50, 1) - H_c]$ $(H_c\rho)^{-1}$ was found to be 5.5 $\mu\Omega^{-1}$ cm^{-1} for both cases and is equal to the initial slope of the H_{c3}/H_c vs ρ curve for the alloys. Apparently the critical field for superconductivity above H_{c2} is determined by the bulk resistivity ρ, at least below 1 $\mu\Omega$-cm, and not by the *type* of defect that causes ρ; and that is different for the three cases investigated: from recovery experiments it was found that, after cold-working, ρ stems from dislocations and point defects, and that ρ after irradiation mainly stems from point defects. In the alloys, ρ is caused by the solute atoms.

Inhomogeneities or structural defects may be thought to be responsible for superconductivity between H_{c2} and H_{c3}. The predominant effect of the bulk resistivity on the *value* of H_{c3} was shown in the preceding paragraph for $\rho < 1$ $\mu\Omega$-cm. At $\rho = 1$ $\mu\Omega$-cm, where type II superconductivity starts, the dependence of H_{c3} on ρ changes (see Fig. 3), which again might imply a relation between the bulk properties and superconductivity between H_{c2} and H_{c3}.

ACKNOWLEDGMENT

The authors are indebted to Professor Dr. J. Volger for helpful discussions.

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Discussion 5

N. R. WERTHAMER, *Bell Telephone Laboratories:* I would like to point out the relevance to the present discussion of the experiments by a number of workers on the superposition of a superconducting metal film on a normal metal substrate. In these experiments, the transition temperature of the film sandwich is lowered substantially when the thickness of the superconducting metal film becomes shorter than a coherence distance. Now, if a filament or sponge model is correct and we are to think of a wire or thread of superconducting metal embedded in an otherwise normal metal matrix, the superconducting filaments cannot be superconducting at all unless they are at least one coherence distance in thickness.

P. H. KEESOM, *Purdue University:* We have measured the specific heats of lead and indium alloys. We wanted to measure the normal state; first we used a 5-kG magnetic field to quench the superconductivity, and then one of 10 kG. In both cases, below one degree we find the specific heat for lead is about 5% larger than what you should expect for the extrapolation from higher temperatures. I wonder if H_{c3} has a large temperature dependence. Do you have any feeling about this temperature dependence?

W. F. DRUYVESTEYN, *Philips' Research Laboratories:* No, we haven't measured the temperature dependence of H_{c3}, but I think it may be the same temperature dependence as H_{c2}.

HAUSER: I also find some discrepancy in the measurements on niobium between the magnetic transition and the resistive transition and I wonder to what extent the difference between H_{c2} and H_{c3} is due to the difference in sensitivities in making a magnetic measurement and in making a resistive measurement.

DRUYVESTEYN: Yes, of course, it is possible. In that case we have to measure the magnetic moment with more sensitivity. But, of course, there is then no agreement with the GLAG theory, and H_{c3} must be H_{c2}.

Pseudoreversible Magnetization of Nb

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The application by Berlincourt and Hake[1] of the Ginzburg–Landau–Abrikosov–Gor'kov (GLAG) theory to the prediction of upper critical fields in type II, transition metal superconductors having varying degrees of defect concentration (cold-work) has been quantitatively satisfactory. Measurements of the resistance of Nb at liquid-helium temperatures by Autler, Rosenblum, and Gooen[2] and of the magnetization of high purity Nb by Stromberg and Swenson[3] lead to the conclusion that this metal is a type II superconductor with $\kappa \approx 1.1$. In fact, the latter group obtained an almost reversible magnetization curve for a relatively defect-free polycrystalline specimen in good qualitative agreement with the predictions of the Abrikosov[4] theory.

The phenomenological theory developed by Bean[5] and by Kim, Hempstead, and Strnad[6] on the assumption of a filamentary model of supercurrent transport has been successful in describing the size-dependent, irreversible magnetization of type II superconductors in the limit of defect concentration where the essentially diamagnetic nature of the material can be ignored. However, this nature is revealed in experiments by Hauser[7] and by Swartz[8] who found that thin films and finely divided samples of "hard" materials had a magnetization characteristic of the Abrikosov–Goodman[4,9] theories. In a recent paper, Silcox and Rollins[10] develop a magnetization theory which predicts hysteresis and size dependence with the incorporation of both Gorter–Anderson[11–13] flux pinning and the Abrikosov–Goodman[4,9] flux structure. The central result of this theory is the superposition of a symmetric (paramagnetic/diamagnetic) defect magnetization on the ideal, reversible curve.

We wish to describe a reversible measurement of the magnetization of a bulk sample of cold-worked Nb which supports the superposition concept and gives a result in good agreement with the Abrikosov[4] curve, though exhibiting a temperature dependence of the reduced upper critical field.

The sample used in this experiment was cylindrical, 0.635-cm diameter by 10.0-cm long and was fashioned from swaged and centerless ground, double-electron-beam-melted stock obtained from Wah Chang. It had good surface finish and prolate spheroidal ends with an eccentricity of 0.94; the estimated demagnetizing factor was 0.005. It was thermally isolated parallel to the bore axis of a NbZr solenoid with a field homogeniety of ±1.2% over the sample length. Flux penetration into the sample was measured from an *xy* plot of an integrated dB/dt signal obtained from a centered, 3-cm-long coil of known area-turns product (10^4 cm^2) surrounding it. A bucking coil, mounted axially parallel to the field probe and symmetrically with respect to its transverse centerline, canceled out the unwanted signal from the turns area of the probe. The effective noise level in this measurement corresponded to a change in the flux penetration of 800 quanta.

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