expansion coefficient of small single crystals. Errors due to changes, with temperature, of sample orientation and position are easily eliminated experimentally and temperature control by means of a gas stream is both convenient and especially good in a thermodynamic sense.

ACKNOWLEDGMENTS

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Magnetic Properties of Superconducting Lead-Base Alloys

J. D. LIVINGSTON
General Electric Research Laboratories, Schenectady, New York
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Magnetization measurements have been made at 4.2°K on a variety of binary lead-base alloys, with the purpose of determining the effects of solutes, precipitates, and dislocations on superconducting properties. Increasing solute concentration results in increasingly broad magnetic transitions, the results correlating with normal-state electron mean free path in quantitative agreement with "negative surface energy" theories of Abrikosov and others. Magnetic hysteresis and trapped flux are small for annealed single-phase specimens, but are greatly increased by plastic deformation and by cellular precipitation. The maximum magnetic field to which superconductivity persists in these alloys is apparently determined by the electron mean free path, whereas extended defects such as dislocations and precipitates are primarily responsible for magnetic hysteresis and trapped flux.

INTRODUCTION

MAGNETIC flux is excluded from a bulk sample of "soft" superconductor until the thermodynamic critical field H_C is reached, at which field the flux penetrates abruptly and superconductivity is destroyed.[1] This magnetic behavior is reversible, flux being expelled from the specimen when the field is lowered below H_C. Although many superconducting elements and some dilute alloys approximate this ideal behavior, some elements (e.g., Nb, Ta, V) and most alloys and compounds do not, and have been termed "hard" superconductors. In hard superconductors, superconductivity persists to a field (H_N) greater than both H_C, which is calculable on thermodynamic grounds from calorimetric data, and H_{FP}, the field of initial macroscopic flux penetration. The magnetic behavior is often also highly irreversible. The magnetic properties of soft and hard superconductors have been reviewed by Shoenberg.[2] Two seemingly alternative models to explain hard or high-field superconductivity are now prominent in the literature: Mendelssohn's filamentary model and the "negative surface energy" model.

Mendelssohn Model

In fields between H_{FP} and H_N a hard superconductor is apparently neither entirely in the superconducting state nor entirely in the normal state. Mendelssohn[3] suggested that hard superconductors are inhomogeneous in microstructure and that this allowed development of a "sponge" structure consisting of interconnecting filaments of superconducting material separated by regions of normal material. If the filament dimensions are comparable to the penetration depth, superconductivity can then persist to high fields, and if they form a multiply connected network, magnetic hysteresis will result. Bean[4,5] has recently extended this model to allow quantitative predictions of magnetic behavior, and has accounted for experimental results on bulk Nb_3Sn samples, including an observed dependence of magnetic hysteresis on bulk sample dimensions. A synthetic filamentary microstructure has been produced by pressing mercury into the pores of leached Vycor and the resultant properties were consistent with Bean's treatment.[6] For the natural high-field superconductors, however, it was not clear what features of the microstructure served as the superconducting filaments. Several authors[7–9] have suggested that dislocations play this role.

Several experimental results have raised difficulties about the filamentary picture of high-field super-

[1] It is assumed the specimen has zero demagnetizing coefficient and hence no "intermediate state" region.

[2] D. Shoenberg, *Superconductivity* (Cambridge University Press, New York, 1960).

[3] K. Mendelssohn, Proc. Roy. Soc. (London) **A152**, 34 (1935).

[4] C. P. Bean, Phys. Rev. Letters **8**, 250 (1962).

[5] C. P. Bean and M. V. Doyle, J. Appl. Phys. **33**, 3334 (1962).

[6] C. P. Bean, M. V. Doyle, and A. G. Pincus, Phys. Rev. Letters **9**, 93 (1962).

[7] R. W. Shaw and D. E. Mapother, Phys. Rev. **118**, 1474 (1960).

[8] J. J. Hauser and E. Buehler, Phys. Rev. **125**, 142 (1962).

[9] J. J. Hauser and E. Helfand, Phys. Rev. **127**, 386 (1962).

conductors. Specific heat measurements on V_3Ga under high magnetic fields[10] can be explained in the filamentary picture only by presuming the density of filaments so great that they make up most of the volume.[9] The magnetic properties of powder specimens[11] of Nb_3Sn, Nb_3Al, V_3Ga, and V_3Si have also been found to be inconsistent with a model of a small volume fraction of superconducting filaments. Another difficulty is that some alloys that retain superconductivity to high fields can show nearly reversible magnetic behavior.[12] This is inconsistent with a multiply connected filamentary structure, for which irreversibility is an inescapable feature.

Surface-Energy Model

Retention of superconductivity to high magnetic fields has also been explained by theories based not on inhomogeneities in the microstructure but on a "negative surface energy" of an interface between normal and superconducting phases. London[13,14] pointed out that the finite depth of flux penetration into a superconductor should make it energetically favorable for a superconductor in a magnetic field to split into a fine mixture of normal and superconducting regions, so distributed that the volume of the material remains mostly superconducting but appreciable flux penetration can nevertheless occur because the individual superconducting regions are very thin. Since the energy is thus appreciably lowered by the creation of many normal-superconducting interfaces, this flux penetration effect can be viewed as yielding an effective "negative surface energy." A soft superconductor does not behave in this way, so apparently must have positive contributions to the surface energy large enough to offset this negative term. (Ginzburg and Landau,[15] Pippard,[16,17] and Bardeen[18] have considered the total surface energy in further detail.) London pointed out, however, that the magnetic properties of *hard* superconductors *are* suggestive of such a splitting into a "mixed state."[19]

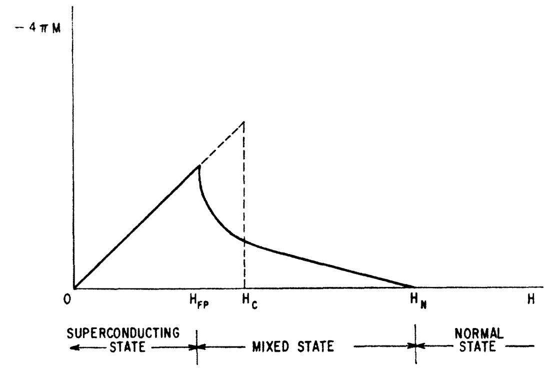

FIG. 1. Dashed curve: Magnetization curve for a soft superconductor. Solid curve: magnetization curve for a superconductor displaying "negative surface energy." M is the magnetization per unit volume, H is the applied magnetic field, H_C is the thermodynamic critical field.

The magnetic properties to be expected in such a case have been derived by Abrikosov[20] and by Goodman.[21,22] When a field H_{FP} ($<H_C$) is reached, the total surface energy becomes negative and a mixed state is formed, with the flux partially penetrating the specimen. In Abrikosov's picture the field initially penetrates in the form of separate filaments parallel to the field, these flux filaments (or "momentum vortices"[23] or "supercurrent vortices"[24]) each containing one quantum of flux (2.07×10^{-7} G-cm^2).[25] Further flux penetration continues over a large range of fields, complete penetration and complete transformation to the normal state not being achieved until the field H_N ($>H_C$). These treatments assume a defect-free homogeneous material, and the predicted magnetization behavior (compared in Fig. 1 with that of a soft superconductor) is reversible.[26]

These theories further indicate that a decreasing normal-state electron mean free path favors negative surface energy behavior.[21,22,27] A normal-superconducting "interface" in a soft superconductor is thought to be very broad—typically of the order of a micron—and in such a case positive contributions to the surface energy dominate. A decreased mean free path is expected to narrow this transition region at the bound-

[10] F. J. Morin, J. P. Maita, H. J. Williams, R. C. Sherwood, J. H. Wernick, and J. E. Kunzler, Phys. Rev. Letters **8**, 275 (1962).
[11] P. S. Schwartz, Phys. Rev. Letters **9**, 448 (1962).
[12] A. Calverley and A. C. Rose-Innes, Proc. Roy. Soc. (London) **A255**, 267 (1960).
[13] H. London, Proc. Roy. Soc. (London) **A152**, 650 (1935).
[14] F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950), Vol. I, pp. 125–130.
[15] V. L. Ginzburg and L. D. Landau, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 1064 (1950).
[16] A. B. Pippard, Proc. Cambridge Phil. Soc. **47**, 617 (1951).
[17] A. B. Pippard, Phil. Trans. Roy. Soc. (London) **248**, 97 (1955).
[18] J. Bardeen, Phys. Rev. **94**, 554 (1954).
[19] This "mixed state" should be distinguished from the "intermediate state" that exists in a narrow field range in soft superconducting specimens of nonzero demagnetizing coefficient due to external field inhomogeneities produced by the specimen shape. Both are pictured as mixtures of normal and superconducting regions, but the mixed state is on a scale of the order of the penetration depth, whereas the intermediate state is on a much larger scale, and has been directly observed by several techniques.

[20] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [translation: Soviet Phys.—JETP **5**, 1174 (1957)]; J. Phys. Chem. Solids **2**, 199 (1957).
[21] B. B. Goodman, Phys. Rev. Letters **6**, 597 (1961).
[22] B. B. Goodman, IBM J. Res. Develop. **6**, 63 (1962).
[23] G. B. Yntema, Proceedings of the Eighth International Low-Temperature Conference, London, 1962 (to be published).
[24] T. G. Berlincourt and R. R. Hake, Phys. Rev. Letters **9**, 293 (1962).
[25] The correspondence between Abrikosov's flux filaments and the flux quantum was pointed out to me by Professor M. Tinkham.
[26] The lower and upper transition fields characteristic of "negative surface energy" behavior were labeled H_{c1} and H_{c2} by Abrikosov[20] and by Goodman.[22] Because of conflict with earlier notation based on Ginzburg-Landau theory, several recent papers have reversed this notation, labeling the lower field H_{c2} and the upper field H_{c1}.
[27] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **37**, 1407 (1959) [translation: Soviet Phys.—JETP **10**, 998 (1960)].

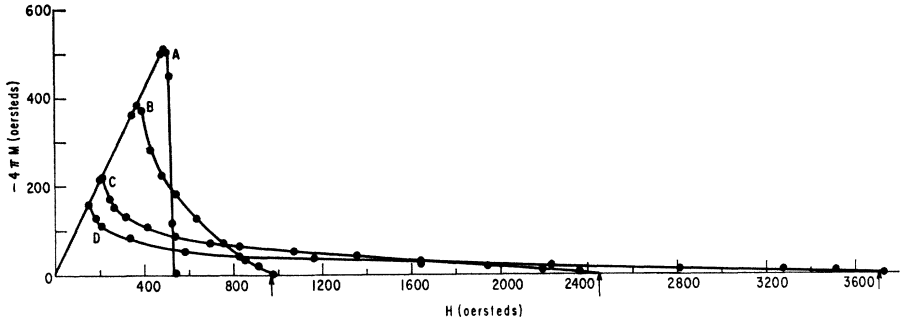

FIG. 2. Magnetization curves of annealed polycrystalline lead and lead-indium alloys taken in ascending field at 4.2°K. *A*—lead, *B*—lead-2.08 wt.% indium, *C*—lead-8.23 wt.% indium, *D*—lead-20.4 wt.% indium.

ary, the interface thus more nearly approaching the abrupt interface considered by London. The field can then penetrate well beyond the transition zone into fully superconducting material, more nearly achieving the full "negative surface energy" effect London calculated.

Rationale for Experiments

Neither of these seemingly alternative models alone can satisfactorily explain all the experimental observations on hard superconductors referred to above. It is clear that a major difficulty of interpretation has been lack of information on the structure sensitivity of various superconducting properties. In the present work a series of binary lead-base alloys has been studied with the purpose of determining the effects of solutes, precipitates, and dislocations on superconducting properties. Lead-base alloys were chosen both because lead is a well-studied soft superconductor with a convenient critical temperature and because magnetic properties of lead-thallium and lead-indium alloys had been used by Abrikosov as an example of "negative surface energy" behavior.

EXPERIMENTAL

A 500-g ingot of each binary alloy was prepared from high-purity materials by melting in a graphite crucible under purified argon and casting into a copper mold one inch in diameter. The metals used were Cominco Pb (99.999%), Vulcan Sn (99.999%), A. S. and R. In (99.999%), A. S. and R. Bi (99.999%), McKay Tl (99.98%), and Mallinckrodt (cp) Hg and Na. The ingot was swaged to 1/8-in. diam rod, from which magnetization specimens 3/4 in. long were prepared with ends rounded to more closely approximate an ellipsoid. Some of the rod was further swaged and drawn to produce 15 mil wire both for resistivity measurements and for magnetization measurements on bundles of 3/4-in. lengths (separated from each other by each being inserted in thin Nonex tubing).

Specimens of 1/8-in. diam and several inches long intended to be single crystals were produced of some of the more dilute alloys (2 wt.% In, 8 wt.% In, 5 wt.% Tl, 2 wt.% Bi) by the Bridgman technique. Specimens 3/4 in. long were spark cut; some were single crystals, others contained at most two or three crystals.

Specimens were annealed for one or more weeks under argon within about 20°C of the melting temperature unless otherwise specified. The two alloys that were quenched were first dropped into an iced brine bath, then rapidly transferred into liquid nitrogen. A satisfactory chemical polish for most of the alloys consisted of 8 parts glacial acetic acid to 2 parts of hydrogen peroxide (30%). Grain structures were revealed with an etchant consisting of 95 parts methanol to 5 parts

TABLE I. Concentrations, resistivities, and magnetic properties of the alloys studied. H_{FP} and H_N are as defined in Fig. 1, and H_{CA} is the thermodynamic critical field as estimated from the area under the ascending magnetization curve. Resistivities are in $\mu\Omega$-cm, magnetic fields in oersteds.

Solute	Wt. %	At. %	ρ(20.4°K)	$\Delta\rho$(20.4°K)	H_{FP}	H_{CA}	H_N
(Pb)	···	···	0.62	0	510	535	540
In	2.08	3.69	3.0	2.4	390	545	970
	8.23	13.9	10.9	10.3	220	510	2450
	20.4	31.6	17.0	16.4	155	495	3700
Tl	4.70	4.78	4.0	3.4	350	535	1020
	30.1	30.4	16.0	15.4	145	400	2900
Hg	4.91	5.05	9.7	9.1	235	580	2300
	9.79	10.1	16.5	15.9	220	590	4300
Bi	1.99	1.97	3.8	3.2	460	570	730
	10.0	9.9	10.9	10.3	290	640	2800
Sn	1.99	3.51	1.5	0.9	530	545	560
	7.02	12.9	···	(3.3)	450	650	1100
Na	0.20	1.55	5.1	4.5	280	610	2050
	1.0	8.32	···	(24.2)	190	545	6000

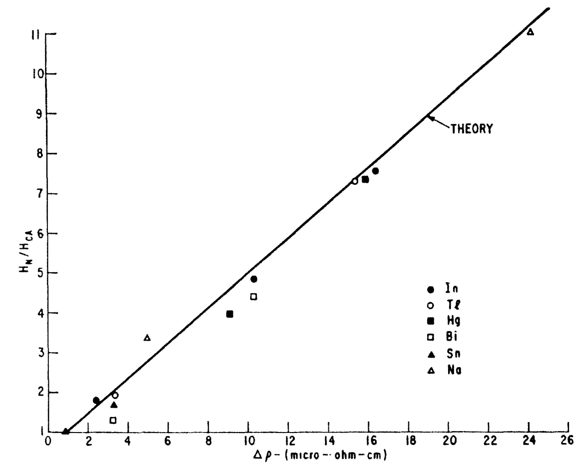

FIG. 3. Dependence of H_N/H_{CA} on $\Delta\rho$, the resistivity of the alloy at 20.4°K less the resistivity of lead at that temperature. The line labeled "theory" results directly from surface-energy theories, with no adjustable parameters.

nitric acid (70%). Most specimens were studied in the chemically polished condition.

Magnetization specimens were immersed in liquid helium in zero field. Measurements were then taken as a function of applied magnetic field by observing the deflection of a fluxmeter connected to a 2000-turn coil when the specimen was removed from or inserted into the coil. The magnetization M was taken as proportional to the fluxmeter deflection, which was calibrated for each specimen by assuming that the initial low-field readings represented complete flux exclusion from the specimen (perfect diamagnetism or $-4\pi M=H$). The magnetic field was measured to within ± 10 Oe, and $-4\pi M$ was measured to within ± 5 Oe. The rate of testing was such that the measurement at each field took about a minute, an entire magnetization curve about 40 min. Resistivity measurements on 6-in. lengths of wire were made with standard four-probe techniques.

RESULTS

Effect of Solutes

For polycrystalline lead and lead-indium alloys the magnetization curves taken in ascending field are shown in Fig. 2. Pure lead behaved as a soft superconductor,[28] but indium in solid solution alters the behavior to that predicted for a superconductor of negative surface energy. H_{FP} decreases and H_N increases as more and more indium is added.

Thallium, bismuth, mercury, tin, or sodium in solid solution similarly broaden the magnetic transitions. Table I shows the values of H_{FP}, H_N, and H_{CA} (the thermodynamic critical field as inferred from the area under the magnetization curve taken in ascending field) for the various alloys studied. The magnetic properties are seen to be much more sensitive to some solutes than to others. Sodium, which has a large effect on the normal-state resistivity, causes much more broadening of the transition per atom percent than does tin, which has a low resistivity per atom when added to lead. Comparing data from all the alloys, the experimental values of H_N/H_{CA} are found to correlate well with $\Delta\rho$ (Fig. 3), the difference between the resistivity of the alloy and that of pure Pb at 20.4°K.

In the 7.02 wt.% Sn and 1 wt.% Na alloys, quenching was necessary to retain a single-phase solid solution. For these alloys resistivity was not measured, but was estimated from the resistivity of more dilute alloys of these solutes by assuming $\Delta\rho$ proportional to the atom percent of solute. The general shape of the magnetization curves were like those in Fig. 2 except for the alloy containing 1.99 wt.% tin. In this alloy about 98% of the flux penetration occurred abruptly in the 530–560 Oe range, but a very small tail to the magnetization curve persisted to about 800 Oe. Further discussion of this observation will appear below. For this alloy the field by which the bulk of the transition had occurred (560 Oe) was listed as H_N in Table I.

The magnetization curves of the alloys were never completely reversible, a typical result being shown in Fig. 4. Well-annealed polycrystalline rod specimens (average grain diameter 0.03 to 0.05 in.), wire specimens (slightly smaller grains), and "single-crystal" specimens had very similar magnetization curves, the only consistent difference being that the wire specimens showed less remanent magnetization on return to zero field (i.e., less trapped flux). Remanent $4\pi M$ for rod specimens was between 5 and 40 Oe (except for the alloy containing 1.99 wt.% Bi, which for unknown reasons trapped between 120 and 180 Oe), whereas wire specimens never trapped more than 10 Oe. Further work is necessary to determine whether hysteresis and trapped flux can be further reduced by further increase in structural perfection of the specimens.

Whenever hysteresis is present, it is not rigorously correct to use the area under the ascending magnetization curve (the work done in driving the specimen normal) to calculate the thermodynamic critical field H_C (which is related to the difference in free energies between the normal and superconducting states). For these annealed single-phase specimens, however, the small hysteresis and the similarity of the theoretical curves (Fig. 1) to the experimental ones taken in ascending field (Fig. 4B) suggest that H_{CA} will probably be only a slight overestimate of H_C.

Effect of Cold Work

Some of the alloys were maintained near liquid-nitrogen temperature during and after a swaging operation from 3/4-in.-diam rod to 1/8-in.-diam rod. Samples were then prepared from this rod and the

[28] The transition is not completely abrupt because the demagnetizing coefficient of the specimen, although small, is not zero, and hence there is a small "intermediate state" region.

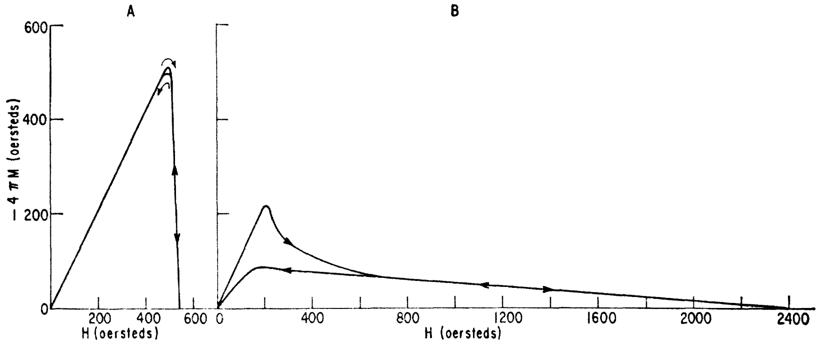

FIG. 4. Near reversibility of magnetic behavior of annealed *A*—lead, *B*—lead-8.23 wt.% indium.

magnetic properties measured before allowing the samples to warm much above liquid-nitrogen temperature. The resultant magnetization curve is much more hysteretic than for a well-annealed sample (Fig. 5). Markedly increased are the field for the first observable flux penetration (H_{FP}), the area under the ascending magnetization curve, and the remanent magnetic moment or trapped flux. The field H_N to which superconductivity apparently persists is little changed.

Subsequent annealing gradually removes the effects of deformation, as seen in Fig. 6. (This alloy, as most of the alloys studied, is capable of recrystallization at room temperature.) Reversible behavior first appears in the high-field region and moves to lower fields as annealing progresses, i.e., the high-field magnetic behavior is found to be less structure sensitive than the low-field magnetic behavior. For this particular sample the curves taken in ascending and descending field appear nearly symmetrically displaced from the reversible part of the curve for the annealed specimens. However, this was not a general rule, several other specimens having shown considerable asymmetry.

Effect of Precipitation

When an alloy containing 7.02 wt.% tin is quenched from a solution temperature of 250°C to low temperatures, the tin is largely retained in solution. However, the equilibrium solubility of tin in lead is less than 2 wt.% at room temperature, and about two hours at room temperature is sufficient for completion of a cellular precipitation that removes much of the excess tin from solid solution.[29,30] (Metallographic study confirmed that the cellular precipitation was completed within two hours.) The resultant two-phase structure consists of lamellae of tin-rich and lead-rich phases. The tin-rich phase is nonsuperconducting at 4.2°K.

The magnetization curve for lead-7.02 wt.% tin as-quenched (Fig. 7, curve A) is similar to that of the other solid solution alloys, although notably more hysteretic. The magnetic behavior after cellular precipitation is seen to be strikingly different (Fig. 7, curve B). The removal of much of the excess tin from solid solution has lowered H_N and raised H_{FP}, and the presence of a multiply connected two-phase structure has resulted in extreme hysteresis and flux trapping. From the value of H_N after cellular precipitation (about 800 Oe) and the correlation shown in Fig. 3, one would estimate that the cellular reaction has left about 4 wt.% Sn in solution in the lead-rich phase. It is also possible that the thinness of the lead-rich lamellae contributed to this H_N value, although the lamella thickness averaged about 5000 Å, considerably greater than the expected penetration depth.

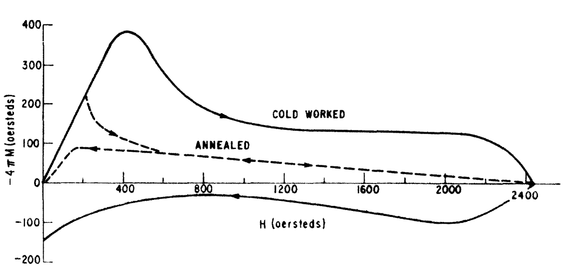

FIG. 5. Result of extreme cold working of the lead-8.23 wt.% indium alloy.

DISCUSSION

Although complete reversibility has not yet been achieved, the various lead-base solid solutions in the annealed condition closely approach the magnetic behavior predicted for superconductors with "negative

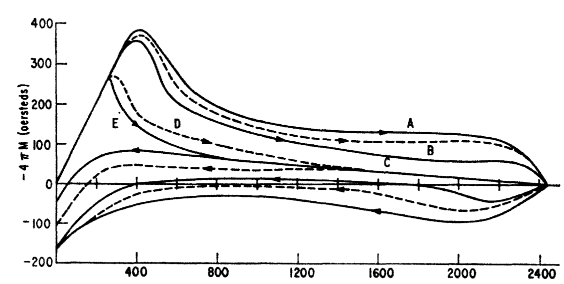

FIG. 6. Result of subsequent room-temperature annealing of the lead-8.23 wt.% indium alloy. *A*—as-cold swaged, *B*—annealed 30 min, *C*—1 day, *D*—18 days, *E*—46 days.

[29] D. Turnbull and H. N. Treaftis, Acta Met. 3, 43 (1955).
[30] D. Turnbull, Acta Met. 3, 55 (1955).

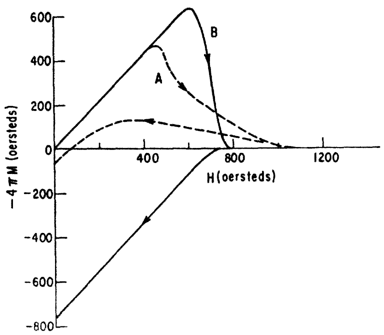

FIG. 7. Result of cellular precipitation in lead-7.02 wt.% tin alloy. A—as-quenched from 250°C, B—after 2 hours at room temperature, which allows completion of cellular precipitation.

surface energy." The H_N values obtained here for lead-indium and lead-thallium alloys agree well with those found earlier by Shubnikov *et al.*[31] Values of H_{FP}, H_{CA}, the hysteresis and the trapped flux differ from theirs, but this is not surprising in view of the structure sensitivity of these quantities (Fig. 5).

The results shown in Figs. 2 and 3 indicate that solutes in lead lower the surface energy and that the extent of the effect correlates with the solute's effect on normal-state resistivity, or electron mean free path, as predicted by surface energy theories.[21,22,27] According to Goodman[22] [his Eq. (6)] the Abrikosov theory leads to

$$H_N/H_C=\sqrt{2}\kappa_0+0.0106\gamma^{1/2}\rho, \qquad (1)$$

good for $H_N>H_C$, where γ is the coefficient of the electronic specific heat per unit volume in the normal state (in erg cm^{-3} deg^{-2}), ρ is the normal-state electrical resistivity in $\mu\Omega$-cm, and κ_0 is a constant that depends on the balance of surface energy terms for pure material with infinite mean free path. For lead, Goodman estimates κ_0 to be 0.4. Taking γ for lead as 1713 erg cm^{-3} deg^{-2},[32] and assuming it to be unchanged by alloying, the above equation for lead becomes

$$H_N/H_C=0.56+0.44\rho. \qquad (2)$$

We can now compare this prediction with the experimental results shown in Fig. 3, since by Matthiessen's rule ρ for the alloys at 4.2°K will be approximately the same as $\Delta\rho$ at 20.4°K. This theory predicts $H_N/H_C=1$ until $\Delta\rho=1$ $\mu\Omega$-cm, beyond which H_N/H_C should increase linearly with $\Delta\rho$ along the line marked "theory" in Fig. 3. The experimental data fit this prediction extremely well, especially considering the likelihood of change in γ by alloying, errors in H_{CA} (which is structure-sensitive) and uncertainty in the value of κ_0. Goodman[22] has already shown that the Pb-Tl data of Shubnikov *et al.*[31] fit this expression, but the present work has extended this to six different solutes with a wide range of resistivities per atom.

In a recent study of indium-base alloys, Seraphim[33] has shown that the appearance of the first traces of "hard" superconductivity as solutes are added to indium also correlates with the effect of solute on electron mean free path. The first evidence of "hard" behavior in his experiments was not apparent in the magnetization curves but in broadened transitions observed resistively. With still more solute he found that most of the flux still penetrated abruptly, but a small tail to the magnetization curve became observable, as for the lead-1.99 wt.% tin alloy tested here. Presumably with still higher solute contents he would have observed broadened flux transitions similar to those observed here on the remaining lead alloys. The more subtle traces of "hard" superconductivity Seraphim and others[34,35] have studied in dilute alloys may be due to a surface energy that is still positive throughout most of the specimen, but negative in the immediate vicinity of defects where the mean free path is locally decreased. Since such experiments have shown that resistive measurements are a more sensitive means of detecting small traces of remanent superconductivity than magnetization measurements, it is possible that in the present study small traces of superconductivity were present beyond the H_N determined magnetically. However, in view of the correlation between resistive and magnetic measurements on similar alloys observed by Shubnikov *et al.*[31] and the negligible increase in H_N resulting from deformation (at most perhaps 20 Oe—see Fig. 5) it is believed that the volume fraction remaining superconducting beyond the measured H_N must be extremely small.

More concentrated indium-base[36] and tin-base[37] alloys show magnetic behavior similar to that expected of negative surface energy superconductors, as do niobium-tantalum alloys[12] and a variety of transition metal alloys.[24,38] The magnetic properties of powdered specimens of Nb_3Sn, Nb_3Al, V_3Ga, and V_3Si[11] suggest that these intermetallic compounds also derive their high-field superconductivity from a negative surface energy, as does Goodman's[39] interpretation of specific heat measurements on V_3Ga. Hence, increasing evidence suggests that the fundamental origin of high H_N in most natural high-field superconductors is a negative surface energy of a superconducting-normal interface. Superconductors such as lead, indium, and tin show this behavior only when the mean free path is sufficiently decreased by the addition of alloying elements,

[31] L. V. Shubnikov, V. I. Khotkevich, U. D. Shepelev, and U. N. Riabinin, J. Exptl. Theoret. Phys. (U.S.S.R.) **7**, 221 (1937).
[32] D. L. Decker, D. E. Mapother, and R. W. Shaw, Phys. Rev. **112**, 1888 (1958).
[33] D. P. Seraphim, A.I.M.E. Annual Meeting, New York, February 1962 (to be published).
[34] P. R. Doidge, Phil. Trans. Roy. Soc. **248**, 553 (1956).
[35] M. D. Reeber, Phys. Rev. **117**, 1476 (1960).
[36] J. W. Stout and L. Guttman, Phys. Rev. **88**, 703 (1952).
[37] W. F. Love, Phys. Rev. **92**, 238 (1953).
[38] W. De Sorbo, Proceedings of the Eighth Annual Conference on Magnetism and Magnetic Materials, Pittsburgh, November 1962 [J. Appl. Phys. (to be published)].
[39] B. B. Goodman, Phys. Letters **1**, 215 (1962).

since for these materials κ_0 in Eq. (1) is less than $1/\sqrt{2}$. The electronic structure of some materials, however, may be such that κ_0 is greater than $1/\sqrt{2}$, and a decreased mean free path is then not necessary to produce $H_N > H_C$. Goodman[39] suggests that V_3Ga is such a material.

Plastic deformation (Fig. 5) and precipitation (Fig. 7) both markedly increase magnetic hysteresis and trapped flux. That extended defects such as dislocations and second-phase particles can have such effects has been reported earlier.[7,40–42] For these lead-base alloys where annealed specimens follow the magnetic behavior predicted by the surface energy theories of Abrikosov and others, the effects of deformation and precipitation presumably result from the interaction between extended defects and the flux filaments that are believed to be the elements of the mixed state. It is suggestive that the magnetization curves, when extended defects are present, closely resemble those expected from the Mendelssohn "sponge" model, and perhaps the two theories can be made into one by considering variations of the surface energy in the vicinity of defects.[34,43,44] However, more experimental and theoretical work is necessary before quantitative understanding of these effects can be achieved.

Since deformation is not expected to alter the mean free path of these alloys appreciably, and since H_N has been shown to depend on mean free path, it is not surprising that H_N is found to be insensitive to deformation (Fig. 5). In the case of precipitation, it is interesting that the two features usually associated with the magnetic properties of hard superconductors, high H_N and magnetic hysteresis, are oppositely affected by heat treatment (Fig. 7). This is a striking demonstration of the separate origin of the two effects, H_N being determined by the mean free path while the hysteresis depends on the presence of extended defects.

The measurements reported here have all been made at 4.2°K. Since alloying changes the critical temperature T_C, each alloy at 4.2°K was at a slightly different reduced temperature. Indium, thallium, and mercury decrease T_C, and bismuth increases it, but the changes for the alloys studied are not great.[45] Study of the magnetic properties of these alloys as a function of temperature would probably be interesting, but it is unlikely that any of the conclusions reached in this paper would require modification.

CONCLUSIONS

(1) The magnetization curves of annealed specimens of a variety of lead-base binary solid solution alloys approximate those predicted by "negative surface energy" theories of high-field superconductivity.

(2) The field to which superconductivity persists (H_N) increases with increasing solute concentration in a way that correlates with the normal-state electron mean free path in quantitative agreement with surface energy theories.

(3) Plastic deformation and precipitation from solid solution both increase magnetic hysteresis. H_N is decreased by precipitation but is almost unchanged by deformation. These results confirm that H_N is determined by the mean free path and indicate that extended defects such as dislocations and precipitates are primarily responsible for magnetic hysteresis and trapped flux.

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