ready trapped. The interaction between fluxoids with fields in opposite directions is attractive (this arises from the fact that the origin of the interaction between fluxoids is due mainly to the need to minimize the magnetic energy). If we assume that the attractive interaction occurs only between nearest neighbors we arrive at a picture something like Fig. 1(c) in which there is a "point of annihilation" or "no man's land" of width roughly the range of the interfluxoid forces lying between the two regions of flux. As the applied field is increased, the point moves further into the specimen until it reaches the center and all the reverse flux has been annihilated. The appropriate sequence EB of the magnetization curve is shown in Fig. 2. It seems likely that this section of the curve would be one of great instability particularly if ρF_P is not relatively uniform but shows nonuniformities at distances greater than the interfluxoid distance, and may correspond to the region of maximum probability of flux jumps.[6]

These curves shown in Fig. 2 show a considerable similarity to the curves observed in practice. Also, the curves shown in Fig. 1 show a similarity to equivalent curves shown by Kim and others.[6] A number of features, however, should be noted in relating these curves to practical curves. Two points have already been mentioned—the shape-dependence of the curve and the problem of the surface–fluxoid interaction between $+H_{c1}$ and $-H_{c1}$. A further point is that so far only nearest-neighbor contributions have been considered. It seems feasible to include further neighbors at the expense of greater complexity. Such an analysis would be possible with a digital computer and the effects may be important at fields close to H_{c2} where the fluxoids are closest together.

It is a pleasure to thank W. W. Webb for many interesting discussions and J. D. Livingston for discussions of his work.

Discussion 3

GOODMAN: These diagrams which we have just seen indicating the local flux density as a function of position during the course of magnetization cycles reminds one very much of the rather elegant films which Dr. DeSorbo showed at the Toronto meeting a few years ago in which one saw (I have in mind the film on niobium which we now know to be a London superconductor), as the external fields increased, a sharp front progressing into the superconductor. This rather sharp front is presumably connected with the vertical tangents in the magnetization curve which we have seen on the screen; the pinning down of the flux lines in certain places was indicated by the concavity of the front towards the outside. We also saw the flux jumps in that film and then finally, after the specimen had been through a number of hysteresis loops, Dr. DeSorbo pointed out to us that one could distinguish sometimes as many as two, or three, or perhaps more, different local values of the magnetic field corresponding to successive additions of flux lines of different signs. I think that perhaps some further experiments of this kind would be extremely valuable.

Defects and Magnetic Hysteresis in Type II Superconductors

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INTRODUCTION

Defects in type II superconductors produce magnetic hysteresis by interacting with, and thereby obstructing the motion of, the flux threads[1] of the mixed state. To move flux threads it then becomes necessary to build up a gradient in flux density, and a resultant magnetic driving force, strong enough to overcome the resisting force produced by the defects. A flux density gradient $\partial B/\partial x$ gives a driving force[2,3] per unit length on an individual flux thread of $(\alpha\phi_0\partial B/\partial x)/4\pi$, where $\alpha(B)$ is $dH(B)/dB$ for the ideal, reversible material, and ϕ_0 is the flux quantum.

Through this balance of forces the defect–flux thread interaction establishes a "critical" internal flux gradient or, equivalently, a "critical" internal

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current. Thus the "critical state" treatment of hysteresis in hard superconductors[4–6] becomes applicable, even though it was conceived from a different physical model. In particular, the internal flux gradients should lead to a specimen size dependence[4] of the magnetization curves.

Pb-base solid solutions have been shown to be type II superconductors,[1,7,8] with their upper critical fields determined by the solute content. Dislocations and precipitates introduced into these alloys produce hysteresis and trapped flux in magnetization measurements[8–10] and increased critical transport currents in resistive measurements.[11] These earlier results and others first reported here are discussed in terms of the interaction forces between defects and flux threads and the magnetic forces driving flux thread motion.

EXPERIMENTAL

Specimen preparation and measurement techniques have been reported earlier.[8,9] Magnetization M was measured at 4.2°K as a function of applied field H on specimens $\frac{1}{8}$ in. in diameter and $\frac{3}{4}$ in. long. If the critical internal current density J were independent of B, it could be easily determined directly from the magnetization curve.[4,5] Since J, in general, varies with B, more complex approaches are necessary to get an accurate $J(B)$ from the experimental $M(H)$. In this paper, however, we use mostly qualitative comparisons between magnetization curves to assess the effects on $J(B)$ of changes in various parameters.

RESULTS AND DISCUSSION

Type I vs Type II

Precipitated particles of Cd in a Pb–1.53 wt % Cd alloy produce much more hysteresis in a matrix that contains sufficient dissolved Cd to make it of type II than in a matrix of nearly pure Pb.[9] A type II superconductor is thus more structure-sensitive than one of type I, presumably because defects can interact more strongly with the mixed state than with the much coarser intermediate state of a type I superconductor.

Low-Field Hysteresis in Annealed Alloys

A well-annealed specimen of Pb–8.23 wt % In shows ideal reversible behavior at high fields [see Fig. 4(b) in Ref. 8]. In low fields flux expulsion is delayed and some hysteresis is observed, although flux continues to escape in decreasing field so that very little trapped flux remains at zero field. Pb–Tl alloys show similar behavior.[7]

Hysteresis caused by internal defects would be expected to increase at low flux densities, since the decreasing forces between widely spaced flux threads result in a decrease in $\alpha(B)$ and hence an increase in the $\partial B/\partial x$ necessary to produce flux motion. However, by chemically polishing an annealed specimen to about half its original diameter and remeasuring $M(H)$, it was found that this low-field hysteresis was *not* appreciably size-dependent. (Similar size-independent low-field hysteresis was observed by Swartz[12] in the limiting magnetization curve for small particles of Nb_3Sn and other compound superconductors.) This size independence indicates that appreciable flux gradients do not exist across the bulk of the specimen, and it is inferred that this low-field hysteresis results not from internal defects but from the specimen surface. The eventual escape of the flux when the Maxwell pressure of the external field is removed also suggests a surface origin of the hysteresis. Theoretical considerations of surface–flux thread interactions[13] indicate that even an ideal surface should provide a barrier to flux motion. Further experiments are underway to elucidate this surface hysteresis.

Hysteresis Produced by Dislocations

Magnetic hysteresis is markedly increased in single-phase Pb alloys by cold-swaging, and this hysteresis gradually decreases as the dislocation density is decreased by subsequent annealing (see Fig. 6 of Ref. 8). The magnetization curves of deformed specimens always show two maxima, the second indicating that $J(B)$ goes through a maximum shortly before decreasing to zero at the upper critical field. Similar peaks in critical transport currents have often been reported for cold-worked alloys.[14]

The $J(B)$ variations in these data cannot be caused by variations in the magnetic driving force, since $\alpha(B)$ from the reversible curve for this alloy is constant over most of this range of fields. Hence the variations in $J(B)$, including the maximum, must be caused by B dependence of the dislocation–flux

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thread interaction forces. Interpretation must await a satisfactory theoretical treatment of this interaction. More quantitative experiments are also clearly called for.

Specimen size dependence could not be satisfactorily investigated in these measurements.

Hysteresis Produced by Precipitates

Precipitation of second-phase particles has been shown to markedly increase magnetic hysteresis and flux-trapping.[8–10]

Precipitation in Pb–Sn and Pb–Cd alloys occurs discontinuously, cells of transformed two-phase material nucleating mostly at grain boundaries and growing into the grains. At fields high enough to drive the cells normal, the untransformed parts of the alloys, still superconducting, show reversible magnetic behavior [see Figs. 1(b), 3(b), and 4 of Ref. 9]. This demonstrates that *large* ($>100\ \mu$) normal inclusions (the cells) do *not* cause hysteresis.

The precipitates within the cells are normal inclusions on a much finer scale ($\approx 1\ \mu$) than the cells themselves, and cause considerable hysteresis when the matrix within the cells becomes superconducting (at lower fields). Critical currents here are large enough to produce 80–95% flux-trapping in this field range (upper critical fields of 900 Oe or less). Except very near the upper critical field, these currents need only flow to a small depth beneath the surface, and there is very little specimen size dependence.

Most of the data in these earlier studies document the changes resulting from isothermal precipitation, during which the volume fraction of precipitate and solute content of the matrix are continually changing, as are the size and shape of the precipitates. Preliminary results are now presented that are intended to separate the effects on hysteresis [hence on $J(B)$] of the degree of precipitate dispersion, volume fraction of precipitate, and matrix solute content (which determines the Ginzburg–Landau parameter κ).

Samples of two Pb–Sn alloys were solution treated, quenched, and held at room temperature for 3 h to allow completion of cellular precipitation. They were then heated to 95°C (in a water bath), a temperature at which volume diffusion of Sn in Pb is about 2000 times faster than at room temperature. The matrix Sn concentration is established at the equilibrium value of about 5 wt % Sn[9] very rapidly in the 11.1 wt % Sn alloy, but only after about 8 days in the 7.02 wt % alloys, since in the latter alloy considerable precipitated Sn must be redissolved. Beyond this time, further aging simply causes a coarsening of the dispersion of a constant volume fraction of precipitate in a matrix of constant composition and, hence, κ. Figure 1 shows the resulting changes in the magnetization curves. The changes A → B and C → D → E indicate a regular decrease in J with coarsening of the particle dispersion. Transmission electron microscopy reveals a large range of particle sizes at each aging time, but a gradual increase in average diameter from roughly 0.2 μ at 4 h to roughly 2 μ at 4 weeks.

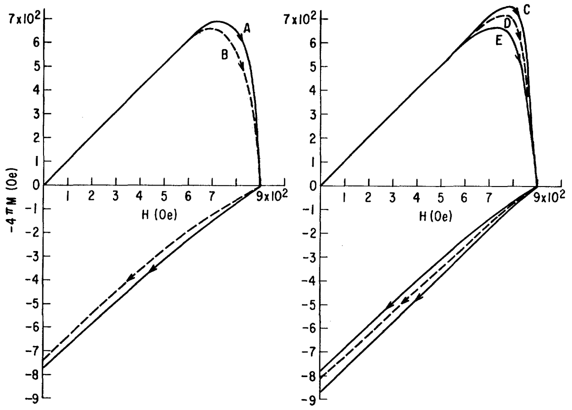

Fig. 1. Effect of coarsening of the precipitate dispersion on magnetization of binary Pb–Sn alloys. Left: Pb–7.02 wt % Sn aged 3 h at room temperature, and then A—8 days, B—23 days at 95°C. Right: Pb–11.1 wt % Sn aged 3 h at room temperature, and then C—4 h, D—2 days, E—23 days at 95°C.

Comparison of curves B and E in Fig. 1 indicates the effect of varying volume fraction with the same matrix and roughly the same particle size. Hysteresis is somewhat larger for the 11.1 wt % Sn alloy (containing about 9 vol. % precipitate) than for the 7.02 wt % Sn alloy (containing about 3 vol. % precipitate). A rough estimate of the average change in J with volume fraction can be obtained from the remanence and an assumption of constant J to the upper critical field.[5] This yields 2×10^4 A/cm^2 for curve B and 3×10^4 A/cm^2 for curve E, an increase in J of only 50% for a threefold increase in volume fraction f. This suggests $J \propto f^{\frac{1}{3}}$, but clearly more data are needed.

The addition of a third element, In, allows large variation of the matrix κ and also gives information on J over a larger field range. Magnetization curves for a Pb–In–Sn ternary alloy are shown in Fig. 2. From the decrease in upper critical field on precipitation, and from transmission electron microscopy, one estimates about 10 vol. % precipitate. Critical currents decrease at higher fields, and over much of the curve flux gradients exist throughout the specimen so that appreciable specimen size dependence is observed. Detailed comparison with the curves in Fig. 1 shows that at low fields (<700 Oe) the ternary alloy has lower critical currents than the binary alloys. Since the volume fraction of precipitate is comparable or higher, and since the particle size is not appreciably greater, it is concluded that this indicates a decrease in J with increasing κ.

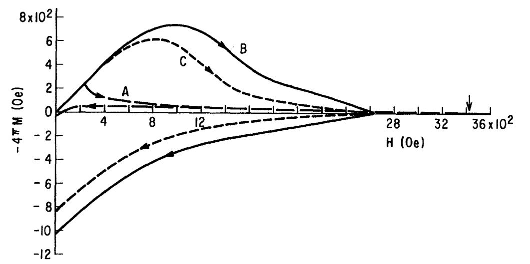

FIG. 2. Magnetization curves for a ternary alloy consisting of 78.4 wt % Pb, 10.0 wt % In, and 11.6 wt % Sn. A—as quenched from 260°C, B—after 1 day at room temperature, C—after 9 days at room temperature.

The decrease of hysteresis with aging time (Fig. 2 B → C) presumably results from coarsening of the precipitate, which is observed to occur very rapidly in this alloy, apparently by a second cellular process.

The Sn-rich precipitates in these various alloys presumably attract and pin flux threads by lowering the flux thread line energy, in particular the contributions of the "normal" core and the vortex supercurrents. The pinning forces caused by cavities have been calculated by Friedel *et al.*[2] They predict a decrease in J with: (a) increasing κ, (b) increasing interparticle spacing (hence, decreasing volume fraction), and (c) increasing particle size (once the particles are large enough to pin several flux threads simultaneously). Thus, the various experimental observations reported above are all qualitatively consistent with the expectations of Friedel *et al.* The observed decrease of J with coarsening of the particle dispersion is expected both from increasing interparticle spacing and from increasing particle size. With more extensive data, the effects of size and spacing can be more clearly separated, as has been done in the somewhat analogous problem of precipitation hardening.[15]

SUMMARY

1. Superconductors of type II are more structure-sensitive than those of type I.
2. Well-annealed specimens show a low-field hysteresis associated with a surface barrier to flux thread motion.
3. Dislocations produce a magnetic hysteresis with a maximum in $J(B)$ just below the upper critical field.
4. Precipitates produce considerable hysteresis, which is found to decrease with coarsening of the dispersion, increase with volume fraction of precipitate, and decrease with increasing matrix κ. These observations are qualitatively consistent with theoretical considerations of Friedel *et al.*

ACKNOWLEDGMENTS

This work has been assisted by transmission electron microscopy done by E. F. Koch and optical metallography done by W. A. Roman. C. P. Bean and H. W. Schadler contributed helpful discussions.

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Discussion 4

E. A. LYNTON, *Rutgers University:* Just a point of information: on the 1.5% bismuth as well as the 7% and 11% tin, you identify this as a type II superconductor; on what basis? Is the upper critical field, i.e., the field at which it becomes normal, much higher than the thermodynamic field?

J. D. LIVINGSTON, *General Electric Research Laboratory:* That was 1.5% cadmium. In an earlier paper I showed by an addition of a variety of solutes to lead, in single-phase alloys, that once you add enough solutes to produce a residual resistivity of about 1 μΩ-cm, the magnetization curves go from type I to type II. The upper critical field goes linearily with resistivity. In this case where one has hysteresis, I decided type I or type II on the basis of the upper critical field. The upper critical field goes above 540 Oe when you get enough solute in the matrix. Also from the phase diagram I can tell how much cadmium should be dissolved in this alloy, and I have shown that, in single-phase alloys, this should be a type II superconductor.

K. MENDELSSOHN, *Oxford:* I can't understand either why you call that a type II superconductor. You've got one critical field and not two. If you were to make a model such as Bean's artificial sponge, namely, one which is normally type I divided into fine, multiply connected regions you would get exactly your hysteresis figure.

LIVINGSTON: I have information in this case. I know what the concentration of the matrix is from my metallurgy. From the phase diagram and from a variety of other concentrations, which I've quenched from various temperatures, I know how much cadmium I have in solution. I know that that amount of cadmium in a single-phase material gives an ideal type II behavior. The cadmium in particles changes the magnetization curve. Now, you can call it type III if you want to, as the Bell people do, but if you add defects to a type II superconductor, you get this kind of magnetization curve.

J. J. HAUSER, *Bell Telephone Laboratories:* You showed a curve on a Pb alloy which showed a peak effect. Now unless you measured the current carrying capacity of this alloy, I wonder if this peak effect is not really due to the following —simply that the current carrying-capacity of this sample 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 $\text{A}\cdot\text{cm}^{-2}$ to 4.6×10^3 $\text{A}\cdot\text{cm}^{-2}$. 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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