PHYSICAL REVIEW VOLUME 159, NUMBER 2 10 JULY 1967

Hysteresis in Superconductors. I. Flux Trapping in Low- and High-κ Materials*

DONALD G. SCHWEITZER, M. GARBER, AND B. BERTMAN
Brookhaven National Laboratory, Upton, New York
(Received 19 January 1967)

Flux-trapping properties of type-II superconductors are described. A linear relationship between the remnant magnetization and the internal field is observed in the low-κ superconductors but not in Nb_3Sn and Nb_3Zr. It was found that the linear relationship and the associated slopes are to a large extent independent of much of the bulk and are related to surface currents. In addition, the linear relationships are observed only in low-κ superconductors that exhibit appreciable diamagnetic behavior.

I. INTRODUCTION

IN this and future papers we present new experiments on a variety of properties of hysteretic superconductors. The new information provides a critical test of models of hysteresis. In this paper we describe how the trapped flux or remnant magnetic moment varies with the average internal field present in the material during the application and removal of an external field. The materials studied had κ values in the range 0.8 to 3 for the most part. In addition, samples of commercial Nb_3Sn and Nb_3Zr were studied. Preliminary results were presented in previous papers.[1]

II. DESCRIPTION OF THE METHOD AND TERMINOLOGY

The measurements start with the sample cooled below its transition temperature in zero external field. One finds that the magnetization M in the low-κ materials is reversible, for some range of external field H, with a slope corresponding to complete diamagnetism (within the absolute accuracy of measurement, about 0.1%); that is, $-4\pi M=H$. In this paper only one sense of the applied field will be considered. Portions of the magnetization curves which have slopes equal to $-1/4\pi$, whether taken in the forward or reverse direction (i.e., with the field increasing or decreasing), are referred to as diamagnetics.

For a hysteretic or irreversible superconductor the internal field cannot, in general, be considered uniform even with zero demagnetization factor. In this work a quantity proportional to the total magnetic moment of the sample, and therefore to the *average* magnetization M, is measured. Assuming negligible end effects, i.e., that the internal fields do not vary in the z or field direction, we shall refer to the average value of the internal field over a plane normal to H as $\langle B\rangle$. These quantities are related by $-4\pi M=H-\langle B\rangle$. The magnetization curve is usually presented as a plot of $-4\pi M$ versus H. The vertical distance between an extension of the initial diamagnetic, $-4\pi M=H$, and the magnetization curve equals $\langle B\rangle$.

Once the external field exceeds the value H_{c1} and field penetrates the sample, the magnetization is no longer reversible. The reverse path falls below the initial magnetization curve and leads to a positive value of $4\pi M$ in *zero external field*. We shall call this quantity the trapped flux (TF). We shall present flux-trapping data in the form of graphs of TF versus H, the maximum field prior to removal of the field. (Since TF is zero until $H=H_{c1}$ and since relatively small values of TF can be measured in zero field, H_{c1} can be determined quite precisely by increasing the applied field in small steps until TF first appears. We did not make a study of H_{c1} in this work, however).[2]

A cycle of the sort just described, proceeding from zero field to a value of $H>H_{c1}$ and then back to zero, we call a minor hysteresis loop. As an illustration we show a moderately hysteretic sample of Pb (1.5 wt.% Bi) alloy in Fig. 1(a). The path OABC is a minor hysteresis loop with trapped flux equal to OC. The maximum value of the average internal field during this cycle $\langle B\rangle$ occurred at the point of reversal A and is shown as the vertical distance DA. In discussing the flux-trapping process we shall present the results in the form of curves of TF versus $\langle B\rangle$. For the above sample this is illustrated in Fig. 1(b). Curves of TF versus H and TF versus $\langle B\rangle$ were first presented by Schweitzer and Bertman.[1] These authors defined the degree of hysteresis of a given material as TF/H_{c2}, where in this expression TF is the value of the trapped flux after removing a field large enough to drive the sample completely normal. The limiting magnetization cycle obtained upon subsequent field cycling between the values 0 and H_{c2} (or H_{c3}) is termed the major hysteresis loop. In Fig. 1(a) this is shown as EFGHE. The properties of the minor and major hysteresis loops have been discussed previously.[1]

III. METHOD OF MEASUREMENT

Most of the magnetization measurements were made by a ballistic method. The voltage pulse which occurred when the sample was pulled from a pickup coil was

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] D. G. Schweitzer and B. Bertman, Phys. Letters **22**, 361 (1966); Phys. Rev. **152**, 293 (1966); Phys. Letters **20**, 339 (1966); **21**, 361 (1966).

[2] This technique has been used to determine H_{c1} of low-temperature superconductors by H. C. Hitchcock, Rev. Mod. Phys. **36**, 61 (1964); R. Hecht, RCA Rev. **24**, 453 (1964); and R. S. Kaeser, E. Ambler, and J. F. Schooley, Rev. Sci. Instr. **37**, 173 (1966).

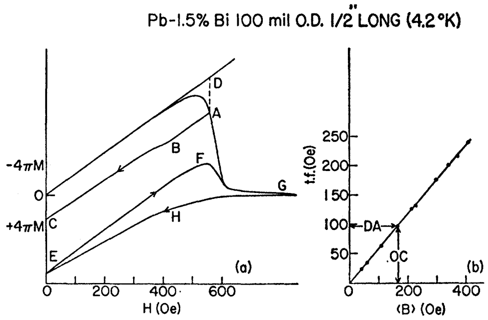

FIG. 1. (a) Magnetization curves of Pb (1.5 at.% Bi) at 4.2°K. The cycle OABC is an example of a minor loop. The cycle EFGHE is the major loop. The ordinate scale is given in arbitrary units. (b) Trapped flux (tf) versus $\langle B \rangle$ for this sample. An example of one of the points on this curve is [TF=OC, $\langle B \rangle$=DA], from (a).

integrated electronically. The sensitivity was approximately 10^{-8} V sec so that with a 5000-turn coil a flux change of 2×10^{-4} G cm^2 was detectable. For purposes of speed and convenience, the measurements were usually made with a reproducibility of 1% even though the available sensitivity made much greater precision possible.

The axes of the applied magnetic field, of the cylindrical samples and of the pickup coil, were collinear. Fields up to 2000 Oe were produced by a copper solenoid in the nitrogen bath. The samples were $\frac{1}{2}$-in. long and had various diameters between $\frac{1}{16}$-in. and $\frac{1}{8}$-in. It was verified by measurements on samples of the same material and same length but varying diameter that end effects could be neglected for the solid samples used. (The demagnetizing coefficient for a prolate spheroid of revolution of length/diameter=4 is 0.075 and it is unlikely that the effective demagnetizing coefficient of any of our samples exceeded twice this value.)

The magnetization data for a given sample were calibrated by setting $-4\pi M=H$ for the points on a diamagnetic. In some materials care is necessary because of flux penetration beginning at very low fields. For example, in a vanadium sample described below [see Fig. (11)], the initial magnetization curve departs from the diamagnetic at approximately 80 Oe. The calibration procedure was accurate to better than 0.1%.

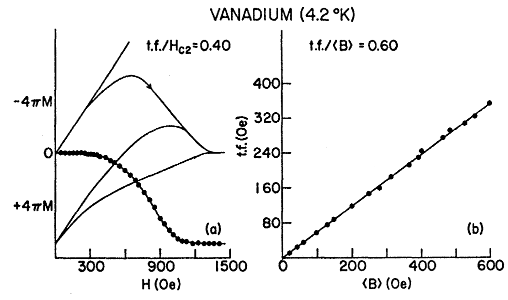

FIG. 3. Same as Fig. 2 for strongly hysteretic V at 4.2°K.

Data on Nb_3Zr wire and Nb_3Sn ribbon were taken with a P.A.R. vibrating-sample magnetometer[3] modified so that the axes of the field, sample, and pickup coils were collinear and parallel to the direction of vibration. A superconducting solenoid capable of producing 30 000 Oe was used with this apparatus.

Descriptions of the various samples will be given with the discussion of the results.

IV. RESULTS

In this section the results on a number of alloys and vanadium and niobium are summarized.

Low-κ alloys vary considerably in the degree of hysteresis they exhibit both from one alloy to another, as well as for a given alloy under different states of strain. It was possible, for example, to vary TF/H_{c2} for a Pb (1.5 wt.% Bi) alloy from 14 to 60% by simply bending and twisting the sample in liquid nitrogen. The same treatment applied to a Pb (13 wt.% Bi) alloy only produced an increase in TF/H_{c2} from 1.5 to 3%. For Sn (5 wt.% Sb) the value of TF/H_{c2} varied from 65% after an anneal to 95% after the mechanical working described. Niobium samples with TF/H_{c2} from 11 to 25% were studied. A high-purity vanadium sample had TF/$H_{c2}\cong$0.0%. After heating at 500°C at 10^{-3} Torr for 4 h so that the sample was quite tarnished, TF/H_{c2} was 25%. The surface of this sample was then "cleaned" by heating in vacuum, 10^{-7} Torr at 1000°C for different lengths of time. The resulting values for TF/H_{c2} varied up to 80%. Prolonged heating at 1000°C, 10^{-7} Torr, restored the sample to the nearly reversible condition.

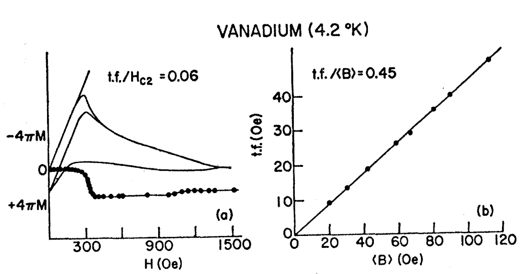

FIG. 2. Magnetization (recorder tracing) and TF (with data points) versus H at left; TF versus $\langle B \rangle$ at right. Slightly hysteretic vanadium at 4.2°K.

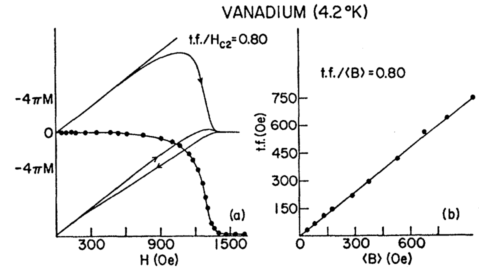

FIG. 4. Same as Fig. 2 for moderately hysteretic V at 4.2°K.

[3] Princeton Applied Research Corporation, Princeton, New Jersey.

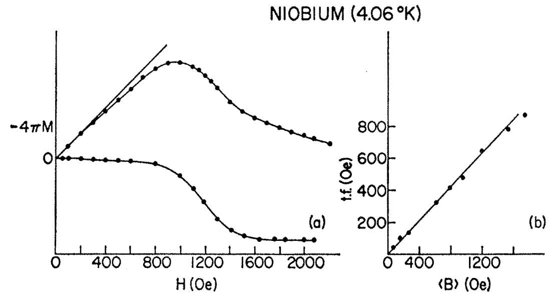

FIG. 5. Magnetization data for niobium, initial magnetization curve, TF versus H, and TF versus $\langle B\rangle$ shown (4.06°K).

An important characteristic of hysteresis is the way TF varies with $\langle B\rangle$. It was found earlier[1] that among the low-κ materials TF varied linearly with $\langle B\rangle$ nearly to the point at which the increase in the flux-trapping process stops. As was mentioned previously, even small values of TF can be measured with considerable accuracy in zero field. In this work, it was found that within $\sim$1 Oe the TF-versus-$\langle B\rangle$ curve goes through the origin, i.e., a portion of the flux is retained no matter how small $\langle B\rangle$ is.

Magnetization, TF-versus-H, and TF-versus-$\langle B\rangle$ curves for a number of the materials studied in this investigation are shown in Figs. 2–5. In Fig. 6 we have summarized the values of the ratio TF/$\langle B\rangle$ for all of the materials studied to date for which TF varies linearly with $\langle B\rangle$. Exceptions to this linear variation are the behaviors of the surface tarnished vanadium sample, and the commercial Nb_3Sn and Nb_3Zr. They are not included in Fig. 6. They are discussed below.

The value of TF/$\langle B\rangle$ is seen to lie in the range 0.40 to 0.58 for weakly hysteretic superconductors (TF/H_{c2}<50%). For superconductors with TF/$H_{c2}\gtrsim$ 50% the values of TF/$\langle B\rangle$ rise and approach unity.

The slope of the TF-versus-$\langle B\rangle$ curve for a given material remains the same when appreciable fractions

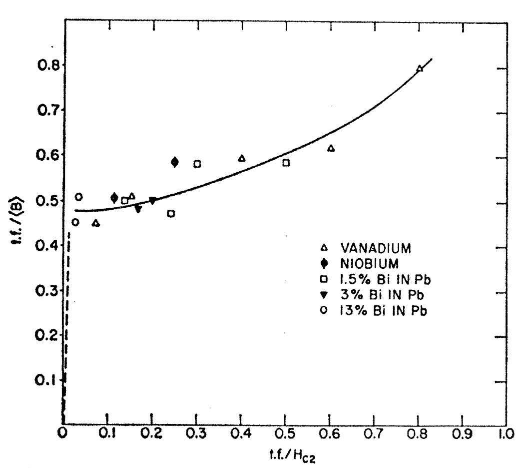

FIG. 6. Slope of TF versus $\langle B\rangle$ curves plotted against degree of hysteresis (TF_{max}/H_{c2}) for low-κ, type-II superconductors.

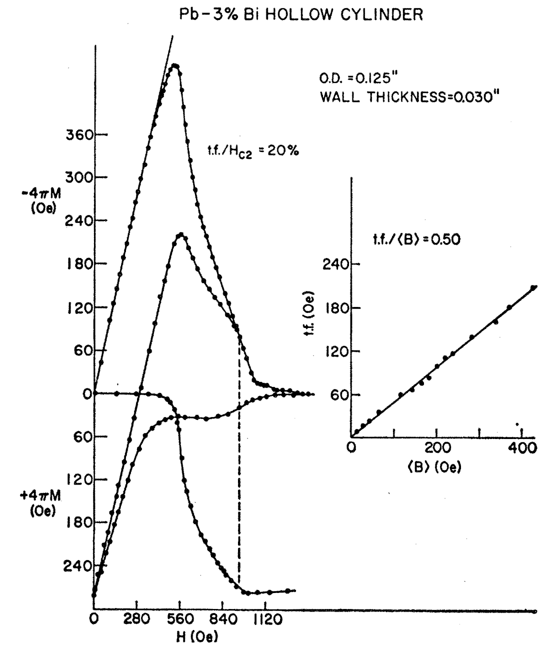

FIG. 7. Data for Pb (3 at% Bi) hollow cylinder, 0.125-in. o.d.×0.060-in. i.d.×$\frac{1}{2}$-in. long. These results are the same for all cylinders of this material with wall thickness $\geq$0.015 in. and for the solid.

of the interior are removed. Experiments on hollow cylinders give the same value of TF/$\langle B\rangle$ as is observed for the bulk for a wide range of wall thicknesses. In the limit of thin walls, however, TF/$\langle B\rangle$ approaches unity for all of the materials. Data for Pb (3 wt.% Bi) with a 0.010-in. wall (0.125-in. o.d.) in which TF/$\langle B\rangle\simeq$1 were given in Ref. 1. The corresponding data for a 0.030-in. wall (0.125-in. o.d.) cylinder are shown in Fig. 7. It was observed that for all cylinders with wall thickness$\gtrsim$0.015-in. TF/$\langle B\rangle$ is the same as for the solid, i.e., $\approx$0.50. In addition, the magnetization curves for the thicker hollow-cylinder samples are the same as for the solid (see following paper).

An interesting feature of some of the TF-versus-H curves which should be mentioned is the maximum.[4]

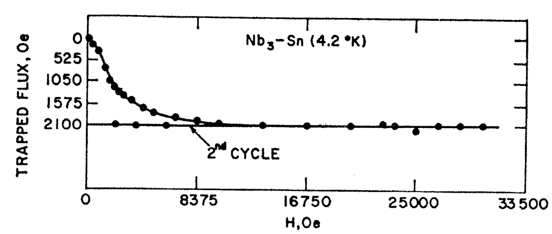

FIG. 8. Trapped flux in zero field after application and removal of progressively larger fields (H). Second-cycle data were obtained by repeating the procedure after the maximum field was applied and TF was present in the sample. For Nb_3Sn (RCA ribbon) at 4.2°K.

[4] S. H. Goedemoed, A. Van der Giessen, D. de Klerk, and C. J. Gorter, Phys. Letters **3**, 250 (1963); M. A. R. le Blanc and D. J. Griffiths, Appl. Phys. Letters **9**, 131 (1966).

This corresponds to a certain minor loop. As the sample is cycled to progressively larger fields, part of this flux is expelled. The effect, as we shall describe below, is quite large in the case of Nb_3Zr. After H_{c2} is reached there generally are no changes in the trapped flux; TF is constant with repeated cycling of the external field (Nb_3Zr is an exception).

The behavior of the magnetization curves upon field reversal is another important test for models of hysteresis. The reverse portion of any hysteresis loop for the materials represented in Fig. 6 always begins with a section which is a diamagnetic, such as AB in Fig. 1.

The diamagnetic indicates a region over which $\langle B \rangle$ is constant, and therefore the changes in magnetization are associated with a surface current.

We shall now discuss a number of materials which exhibit different behavior.

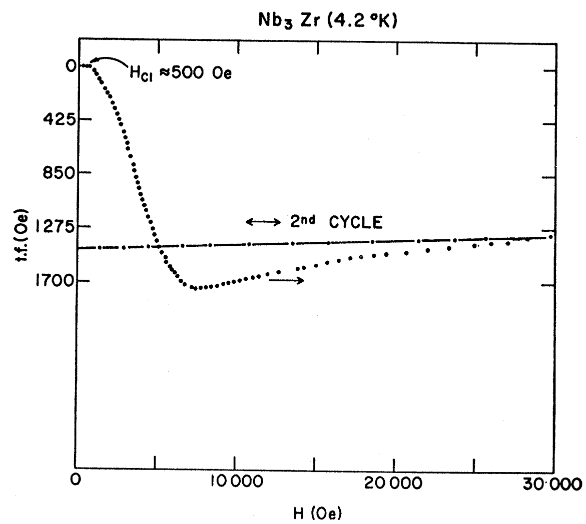

FIG. 9. Same as Fig. 8 for Nb_3Zr (0.010-in. Westinghouse wire) at 4.2°K.

V. MEASUREMENTS ON HIGH-FIELD SUPERCONDUCTORS

Flux trapping was studied in two samples of commercial high-field material: RCA Nb_3Sn ribbon and Westinghouse Nb_3Zr 0.010-in.-diam wire. The magnetization data were taken with the vibrating-sample magnetometer in fields provided by a superconducting magnet. The maximum field was 30 000 Oe. Therefore, the complete magnetization curves could not be obtained since the upper critical fields for these materials at 4.2°K are considerably in excess of this field. Flux trapping in Nb_3Sn appears to be complete, however, at external field values of 10 000 Oe as shown in Fig. 8. For Nb_3Zr, TF goes through a maximum with external field as was mentioned before. This occurs at the relatively low field of 8000 Oe as shown in Fig. 9.

As shown in Fig. 10 the curves of TF versus $\langle B \rangle$ are not linear for these materials. The curve bends over for

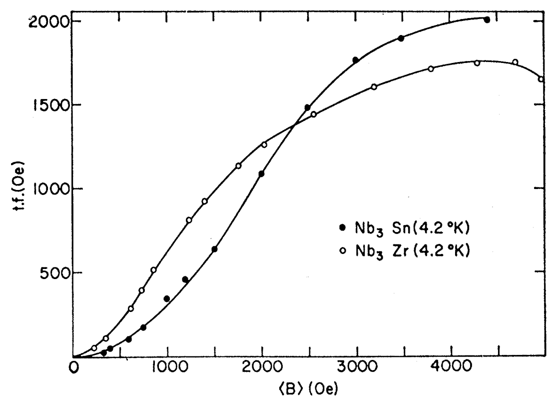

FIG. 10. TF versus $\langle B \rangle$ for the Nb_3Sn and Nb_3Zr shown in Figs. 8 and 9, respectively.

Nb_3Zr due to the maximum. For Nb_3Sn the curve is in the form of an S-shape; the initial slope is less than 0.1, which is considerably smaller than the values obtained for the low-κ materials previously discussed. The values of TF/H_{c2} are very small for these materials—roughly 2% for the Nb_3Zr and 0.7% for the Nb_3Sn. It was found that the maximum remnant flux in all the materials studied, independent of H_{c2}, is limited to a few kilogauss.

An interesting low-field superconductor which resembles the above and for which we can present more complete magnetization data is a vanadium sample with the surface oxidized. The magnetization data for this sample are shown in Fig. 11. In contrast to the vanadium samples described before both with lower and higher TF/H_{c2} values, this sample does not exhibit appreciable diamagnetism either on the forward magnetization

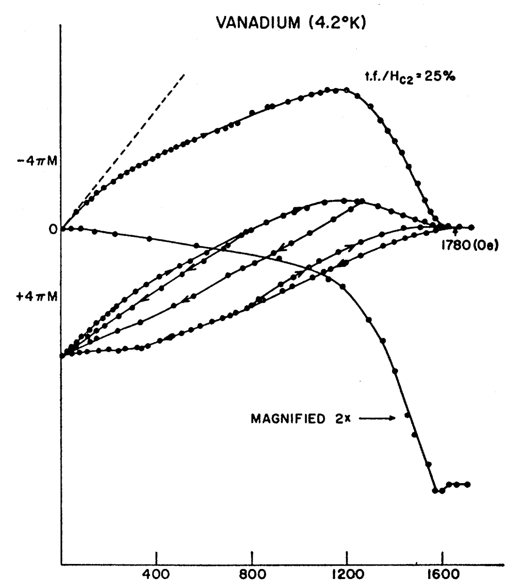

FIG. 11. Magnetization data for vanadium sample with tarnished surface.

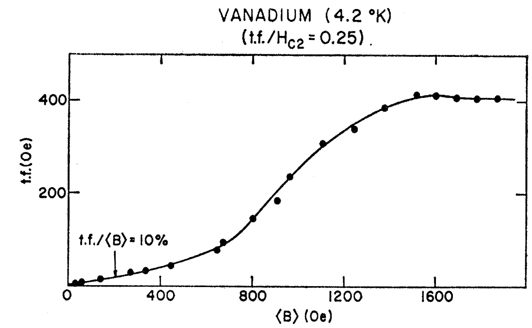

FIG. 12. TF versus ⟨B⟩ for the surface tarnished vanadium sample.

curve or on field reversals. In this respect the magnetization curves resemble those of Nb_3Sn and Nb_3Zr more than those of the low-κ materials. As can be seen from Fig. 12 the curve of TF versus ⟨B⟩ for this vanadium sample is similar to that for Nb_3Sn. These results show that a linear TF-versus-⟨B⟩ curve is associated only with materials for which there are large diamagnetics.

VI. DISCUSSION

We have described a method for analyzing hysteretic behavior by simultaneous measurements of magnetization curves and of the manner in which trapped flux varies with the internal field in the material. For low-κ materials with large diamagnetic behavior the relation between TF and ⟨B⟩ is linear. On the other hand, both high-κ and low-κ materials without appreciable diamagnetics show a nonlinear variation of TF with ⟨B⟩. In addition the hollow-cylinder experiments show that the linearity of the TF-versus-⟨B⟩ curves is not a property of the bulk and that the slope is independent of much of the bulk.

Models of hysteretic behavior are based on flux pinning at imperfections in the bulk of the material. The Bean model[5] in its simplest form assumes a uniform volume current density and leads to (a) a nonuniform internal field, (b) no diamagnetic behavior on external field reversal, and (c) a linear variation of TF with ⟨B⟩ with proportionality constant equal to $\approx$0.5 for an infinite slab. However, for a solid cylinder in an axial field, the variation of TF with ⟨B⟩ should be nonlinear. These predictions are not consistent with the observed pattern of behavior. For the materials for which TF$\simeq$0.5⟨B⟩ there is a large uniform component of the trapped flux. For the high-field materials, in which the evidence indicates little or no surface currents, the data are at variance with all the critical-state models.

The vanadium samples exhibit this inconsistency in a striking way. Only those samples with large diamagnetics have linear TF-versus-⟨B⟩ curves. The observed slopes for these samples varied from 0.45 to 0.80 with increasing hysteresis. These samples show a considerable component of the trapped flux to be associated with surface currents. On the other hand, the vanadium sample which shows no appreciable diamagnetic behavior exhibits a nonlinear variation of TF with ⟨B⟩.

The data presented here indicate that the surface currents do not originate from surface-pinning forces or steep field gradients in a thin outer shell. If steep field gradients existed in a thin outer shell only, the average value of the internal field would be very small for external fields less than H_{c2}. The forward part of the initial magnetization curve would resemble that of a type-I superconductor. At the same time field reversals, for $H<H_{c2}$, would lead to TF$\approx$0.5⟨B⟩. In fact, for samples which have this kind of magnetization curve, viz. the vanadium samples of Fig. 4, the slope approaches unity. Moreover, superconductors in which the hysteresis is due to surface pinning should continue to trap flux up to H_{c2}. Most of the materials discussed in this work show that the flux-trapping process is finished well below H_{c2}. In addition, magnetization changes upon field reversals in a surface-pinning model should involve changes in the internal fields that are initially small but keep increasing as H keeps changing. Such a model, therefore, does not allow for diamagnetic changes upon field reversals.

The Bean model is based on the postulate that the trapped flux is in a critical state, i.e., that the volume currents are everywhere critical. In the following paper we will discuss the results of extensive experimental tests of the critical-state assumption.

[5] C. P. Bean, Phys. Rev. Letters **8**, 250 (1962); C. P. Bean and M. V. Doyle, J. Appl. Phys. **33**, 3334 (1962); C. P. Bean, Rev. Mod. Phys. **36**, 31 (1964).