MAGNETIZATION STUDIES ON Nb_3Sn MULTIFILAMENTARY WIRE (*)

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ABSTRACT — The results and interpretation of magnetization measurements made on superconducting multifilamentary Nb_3Sn, obtained by the bronze route, are presented. Curves of first magnetization and quasi-static loops were obtained by electronic integration, the explanation of which is given in terms of the critical state model taking also into account the existence of a surface current. The dependence of the hysteretic loss on the surface magnetic field is yet considered and explained on the same basis. Finally a method of determining the variation of the critical current density with the magnetic induction by means of small loops is described.

In recent years great interest has been shown in many dc and ac applications such as superconducting magnets, superconducting electrical machines and superconducting power transmission. One of the materials which has received some attention in view of its high transition temperature is Nb_3Sn. In the beginning, because of the high hysteretic losses, it was not regarded as a potential ac conductor. However, recently, new techniques of fabrication were developed which allowed the losses to be reduced. The results we present in this communication were obtained on Nb_3Sn multifilamentary wire prepared in the UKAEA Establishment of Harwell by the bronze route.

The explanation ot the observed magnetic behaviour was based on Bean's critical model [1] applicable to hard superconductors. However, instead of considering, as Bean does, a constant critical current density, we suppose this density, J_c, dependent on the local magnetic induction, B, using Kim's relation $J_c = \alpha/(B + B_0)$.

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In this communication we begin by presenting briefly the related theory, after which the sample and experimental set-up are described. Then, we present and give explanation for the first magnetization curve and some of the symmetrical hysteretic loops we obtained. Finally we show a process by which one can get, easily, the relation $J_c = J_c(H)$ by means of small hysteretic loops recorded at different values of the applied magnetic field.

2 — THEORY

2.1 — *The Critical State*

The concept of the critical state was firstly introduced by Bean [1]. Its hypothesis are the following: *a*) there is a constant limit value for the critical current density, J_c, which is the maximum value that a supercondutor can carry without losses; *b*) this limit value J_c is locally induced by any e.m.f. no matter how small. Later it was admitted and proven experimentally that the critical current density depends on the local magnetic induction, $J_c = J_c(B)$. One of the functional relations which is frequently used is that proposed by Kim et al [2] $J_c = \alpha/(B+B_0)$, where J_c is the critical current density, B the magnetic induction and α and B_0 characteristic parameters of the material. The critical current density J_c has its origin in the existence of metallurgical defects which work as pinning centres of the fluxoids. Maxwell's equation $\nabla \times \vec{B} = k\vec{J}$ allows one to relate J_c with the fluxoid concentration, n, since we have $B = n\,\phi_0$, where $\phi_0 = 2.70 \times 10^{-15}$ Wb is the value of the magnetic flux associated with a fluxoid and $k = 4\pi/10$ with B in Gauss and J in A cm^{-2}.

In the case of a cylindrical sample Maxwell's law just referred can be written

$$\pm\int_{H_0}^{B(x)} \frac{dB}{J(B)} = kx \quad ,$$

x being measured in the relation to the surface and the sign $\pm$ appearing because the vector equation is written in a scalar form. The lower limit of the integral is the magnetic field seen by the bulk, which means that the surface magnetization is subtracted to the applied magnetic field. In the case of Nb_3Sn, the hysteretic contribution of the surface was not considered. However, we have taken into account the Meissner magnetization Me. According to Fietz and Webb [3] we consider $H_0 = H_a + 4\pi Me$, where H_a is the applied magnetic field.

2.2 — *Expressions for the case of full penetration*

When in every point of the sample there is magnetic field, we say that full penetration has ocurred. In this situation, critical current takes place all over the sample. We did use expressions (a), (b), (c) and (e) as given by Fietz et al [4]. As their expressions (d), (f), and (g) led us to incongruous results, we attempted to deduce new ones for the corresponding situations (see Appendix 1), which we refer in table 1.

2.3 — *Expressions for the case of partial penetration*

We have also deduced the corresponding expressions for the case in which the magnetic field does not penetrate the sample fully (see Appendix 2). The results are as follows, where H_{om} is the peak field seen by the bulk.

H_a increasing, $0 \leq H_0 \leq H_{om}$:

$$4\pi \overline{M} = \frac{2}{3\alpha k \omega} \left(\frac{(H_{om} + B_0)^2 + (H_0 + B_0)^2}{2} \right)^{3/2} -$$

$$- \frac{1}{3\alpha k \omega} ((H_0 + B_0)^3 + B_0^3) - \frac{B_0}{2\alpha k \omega} ((H_{om} + B_0)^2 - B_0^2) - H_a \,.$$

H_a decreasing, $H_{om} \geq H_0 \geq 0$:

$$4\pi \overline{M} = \frac{1}{3\alpha k \omega} ((H_0 + B_0)^3 + B_0^3) + \frac{B_0}{2\alpha k \omega} ((H_{om} + B_0)^2 -$$

$$- 2(H_0 + B_0)^2 + B_0^2) - \frac{2}{3\alpha k \omega} \left(\frac{(H_{om} + B_0)^2 - (H_0 + B_0)^2}{2} + B_0^2 \right)^{3/2} - H_a \,.$$

TABLE I

Validity Region	Average magnetization $4\pi\overline{M} = \int_0^{\omega} (B(x) - H_a)\,dx$
H_a increasing $0 < H_a < H_{c1}$, first magnetization	$4\pi\overline{M} = -H_a \quad (a)$
H_a increasing $H_{c1} < H_a < ((B_0 + H_{c1})^2 + 2\alpha k \omega)^{1/2} - B_0$ first magnetization	$4\pi\overline{M} = -H_a - \frac{(B_0 + H_a)^2}{\alpha k \omega}\frac{B_0}{6} - \frac{H_a}{3} + \frac{(B_0 + H_{c1})^2}{\alpha k \omega}\frac{B_0}{6} - \frac{H_{c1}}{3} \quad (b)$
H_a increasing $((B_0 + H_{c1})^2 + 2\alpha k \omega)^{1/2} - B_0 < H_a < H_p$	$4\pi\overline{M} = -(B_0 + H_a) - \frac{1}{3\alpha k \omega}\{[(H_0 + B_0)^2 - 2\alpha k \omega]^{3/2} - (H_0 + B_0)^3\} \quad (c)$
H_a decreasing $H_{c1} < H_a < ((H_p + B_0)^2 - 4\alpha k \omega)^{1/2} - B_0$	$4\pi\overline{M} = -(B_0 + H_a) + \frac{1}{3\alpha k \omega}\{[(H_0 + B_0)^2 + 2\alpha k \omega]^{3/2} - (H_0 + B_0)^3\} \quad (e)$
H_a increasing $-H_{c1} < H_a < H_{c1}$	$4\pi\overline{M} = B_0 + \frac{1}{3\alpha k \omega} B^3 - \frac{1}{3\alpha k \omega}(2\alpha k \omega + B_0^2)^{3/2} - H_a$
$H_{c1} < H_a < ((B_0 - H_{c1})^2 + 2\alpha k \omega)^{1/2} - B_0$	$4\pi\overline{M} = -\frac{1}{3\alpha k \omega}[(2\alpha k \omega + 2B_0^2 - (H_0 + B_0)^2)^{3/2} - (H_0 + B_0)^3] - \frac{B_0}{\alpha k \omega}((H_0 + B_0)^2 - B^2) + B_0 - H_a$

Obs.: H_p is the peak value of the applied field H_a.

The value of the average remanent magnetization, i.e. the value of the average magnetization when the applied field is zero, is obtained by putting $H_0=0$ in any of the above expressions:

$$4\pi\bar{M}_{rem}=\frac{2}{3\,\alpha\,k\,\omega}\left(\frac{(H_{om}+B_0)^2+B_0^2}{2}\right)^{3/2}-$$

$$-\frac{B_0}{2\,\alpha\,k\,\omega}(H_{om}+B_0)^2-\frac{1}{6\,\alpha\,k\,\omega}B_0^3.$$

3 — SAMPLES AND MEASUREMENT TECHNIQUE

The samples consisted of groups of 4 wires, approximately 4 cm long. These wires were cut from a stool produced by the bronze route, according to the technique initiated at A.E.R.E. Harwell in 1969 [5]. The basic building block of the composite is a 37 filament hexagon in a bronze matrix (Cu/7.5 *a* % Sn) which has been stacked in the configuration shown in the photomicrograph (Figure 1) with 37 of

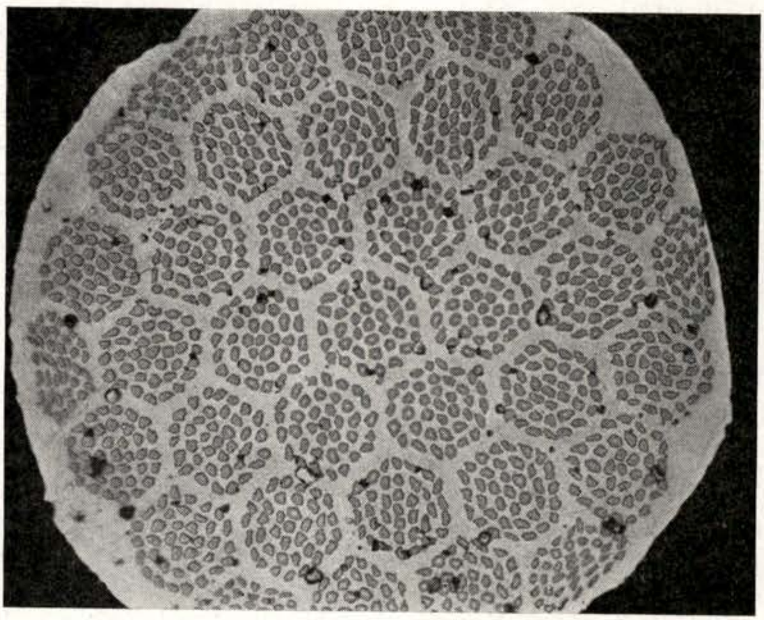

Fig. 1 — Wire of Nb_3Sn composite (37 × 37 filaments).

these blocks ($37 \times 37 = 1369$ filaments). The wire is drawn down to its final shape being simultaneously twisted: the diameter oi each filament is about 8 μm, the external diamater equal to 0.43 mm and the twist pitch equal to 6.44 mm. This wire is then introduced in a large quartz tube (~ 3 cm in diameter) which is placed in a tube furnace and kept under a dynamic vacuum better than 10^{-5} torr during 24 hours at

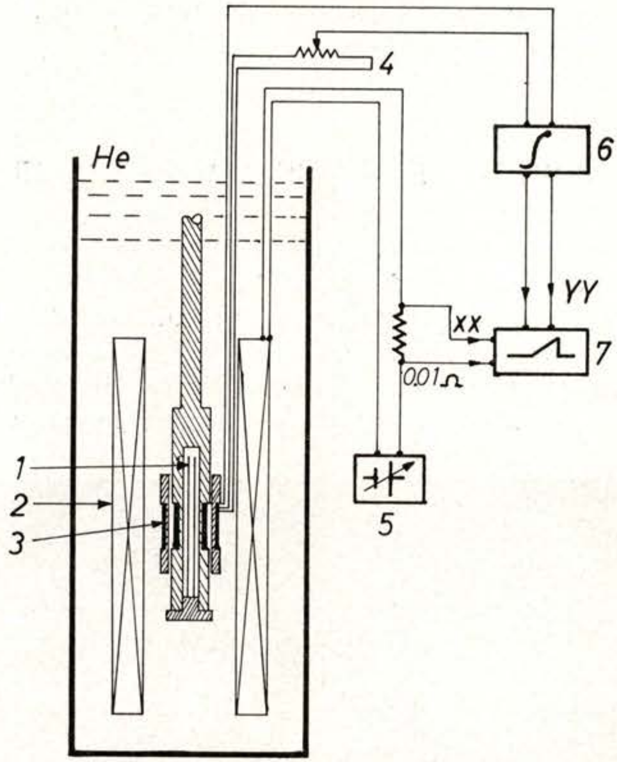

Fig. 2 — Circuit for electronic integration

1 — sample; 2 — superconducting magnet; 3 — pick-up coil; 4 — potential divider; 5 — sweeping d.c. power supply; 6 — integrator; 7 — x y recorder

750°C. The Sn diffuses from the matrix to the Nb filaments with the formation of a Nb_3Sn layer between the bronze and the Nb cores. After the heat treatment the sample cools down slowly under vacuum.

The experimental arrangement is shown in Figure 2. The magnetization is measured by electronic integration. The signal is obtained from a pick-up coil which consists of two coils connected in series opposition, the inner one with 10 000 turns and the outer with 527. The final compensation is obtained by means of a voltage divider connected between the ends of the last turn of the outer coil. The signal, after being integrated using an electronic integrator, was introduced in the Y-axis of a recorder. In the X-axis was introduced a signal taken from a resistor which was traversed by the current generating the magnetic field.

4 — RESULTS AND DISCUSSION

The curve of first magnetization and the loops for peak values of 1160, 1600, 2400 and 29680 Oe are presented in Figure 3 *a*, *b*, *c*, *d*, and *e*. They show the average magnetization, $-4\pi\bar{M}$, in terms of the applied field H_a. In full are the experimental curves and the dotted ones were evaluated with the above expressions deduced on the basis of the critical model using Kim's relation.

$\bar{M}$ varies linearly with H_a between -400 and 400 Oe, approximately, which can be interpreted as due to the Meissner current. Increasing H_a the magnetization reaches a maximum after which starts decreasing monotonically up to the peak field H_{am}. After this value has been reached and the external field starts to decrease it is not noticeable any hysteretic surface current: this behaviour is characteristic of high $\varkappa$ materials, as is the case of Nb_3Sn. In decreasing the magnetic field, the absolute value of the magnetization, after passing through zero, starts to increase, but with opposite sign, up to the remanent magnetization for $H_a = 0$.

Let us now look at the calculated curves. In their evaluation it was considered an average equilibrium magnetization, defined by the following pairs of values $\{H_a - 4\pi Me\} = \{(0,0); (400, 400); (500, 220); (800, 120); (>1200, 100)\}$. The best fitting for the loops whose peak values are 1200, 1600 and 2400 Oe was obtained by putting in Kim's relation $\alpha = 1{,}2 \times 10^{10}$ G-A/cm² and $B_0 = 1100$ Oe. The choice of these parameters gets further confirmation based on the variation of the remanent average magnetization, $4\pi\bar{M}_{rem}$, in terms of the applied magnetic field, as Figure 4 shows. The field of full penetration is, in this case, equal to 2200 Oe, approximately. For the curve of first magnetization and the loop of 29680 Oe peak field, the best fitting was with $\alpha = 5 \times 10^{10}$ G-A/cm² and $B_0 = 6300$ Oe.

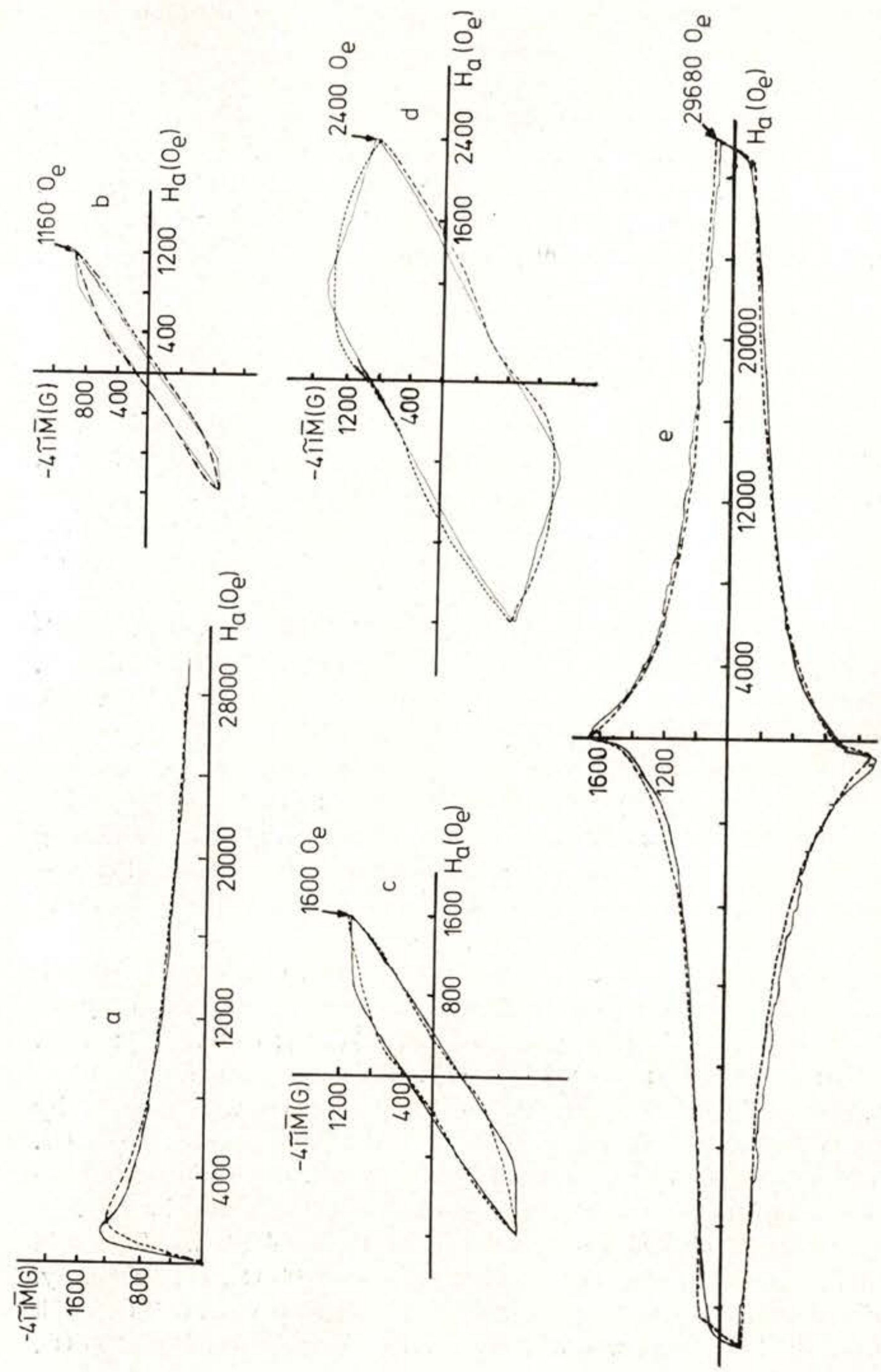

Fig. 3 — Curve of first magnetization and quasi-static loops.

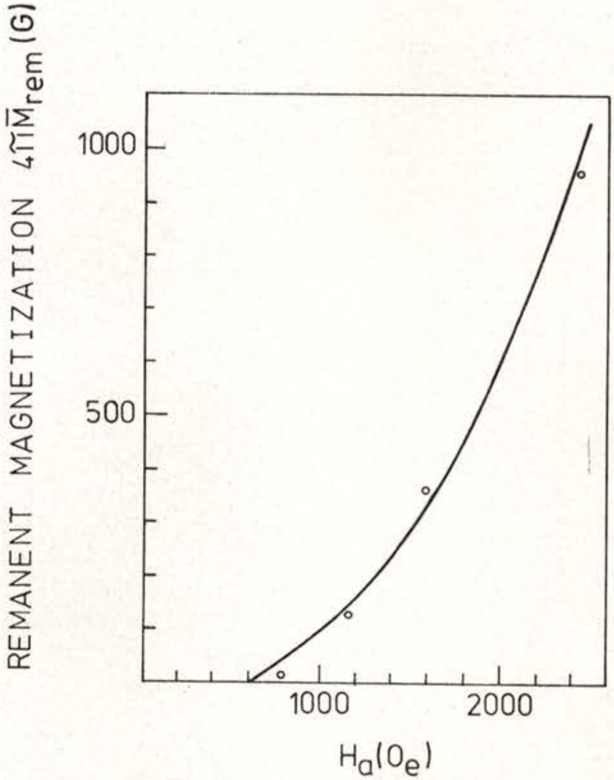

Fig. 4 — Dependence of the remanent magnetization on the applied magnetic field.

For these parameters the field of full penetration is equal to 2600 Oe approximately. Therefore, we are bound to consider the material well described magnetically by two Kim's relations: one for low fields (<3000 Oe) and the other for high fields. As Figure 5 shows these functions of critical current density vs. the applied magnetic field agree satisfactorily with the experimental one.

5 — DETERMINATION OF $J_c = J_c(H)$ BY MEANS OF SMALL LOOPS

The magnetization measurements brought the idea of using small loops traced at different values of the applied magnetic field to determine the relation $J_c = J_c(H)$. Let us imagine we reduce the value of the appied field from H_a to $H'_a < H_a$. If $\Delta H_a = H_a - H'_a$ is small it is justifiable to consider in this interval, the critical current density J_c as constant. The variation of the average induction $\Delta \overline{B}$ is then

$$\Delta \overline{B} = \frac{(\Delta H_a)^2}{4\, k\, \omega\, J_c} \quad .$$

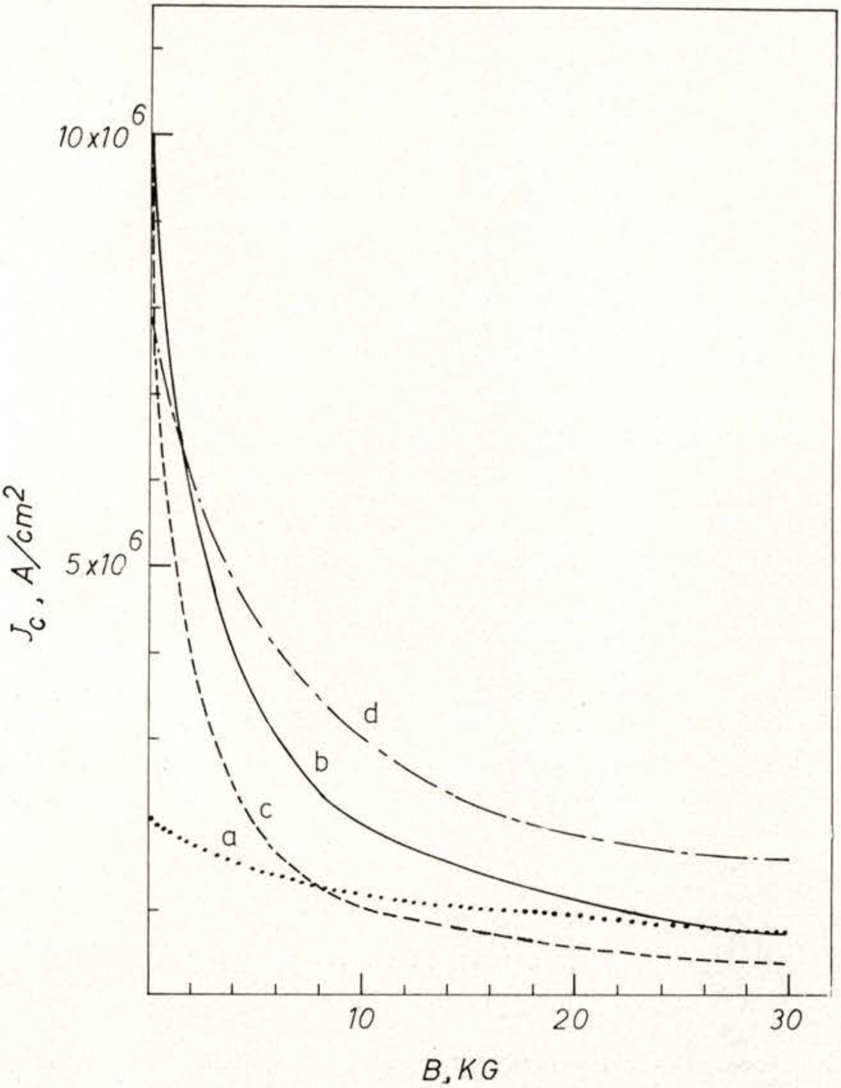

Fig. 5 — Dependence of the critical current density on the magnetic induction. *a* — experimental; *b* — Kim's relation $J_c = 2.5 \times 10^{10}/(B + 2300)$; *c* and *d* — obtained from quasi-static magnetization measurements.

Considering Kim's relation $J_c = \alpha/(B+B_0)$ and taking $B \simeq H_a$, which is reasonable, one gets

$$\frac{\Delta \bar{B}}{(\Delta H_a)^2} = \frac{H_a + B_0}{4\,\alpha k \omega} .$$

By the use of small loops, as those presented in Figure 6, we evaluate $\Delta \bar{B} = \Delta(4\pi \bar{M}) - \Delta H_a$. Then by choosing $\Delta \bar{B}/(\Delta H_a)^2$ as ordinate and H_a as abcissa we can easily obtain B_0 and $\alpha k \omega$. In our case, we get from Figure 7, $B_0 = 2\,300$ Oe and $\alpha k \omega = 1{,}0 \times 10^7$ which yields $J_c = 2.5 \times 10^{10}/(B+2300)$, which is represented in *b* of Figure 5. This curve is compared with the experimental one and those obtained from the quasi-static magnetization measurements *c* and *d* (Figure 5).

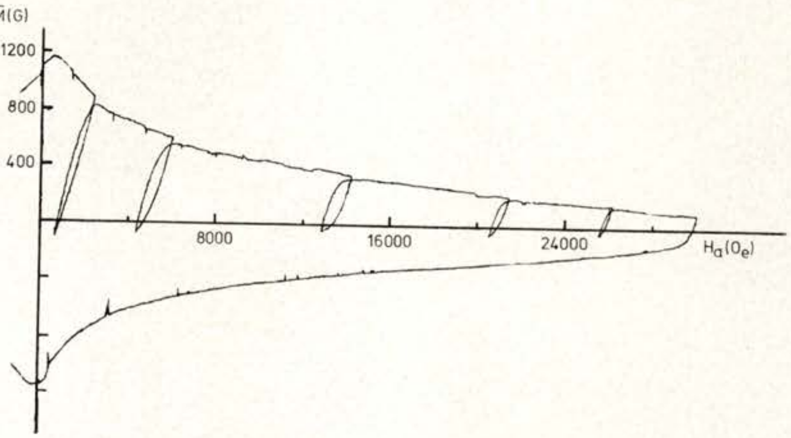

Fig. 6 — Example of experimental small loops.

This work was done as a member of the 'Projecto CF3' (INIC); the experiments took place during a stay in the UKAEA Establishme of Harwell, graciously financed by the Royal Society (London) and the Academia das Ciências de Lisboa to whom I express my gratitude. It is also an agreeable duty to thank Drs. J. A. Lee and J. P. Charlesworth, of Harwell, for the kindness of their hospitality.

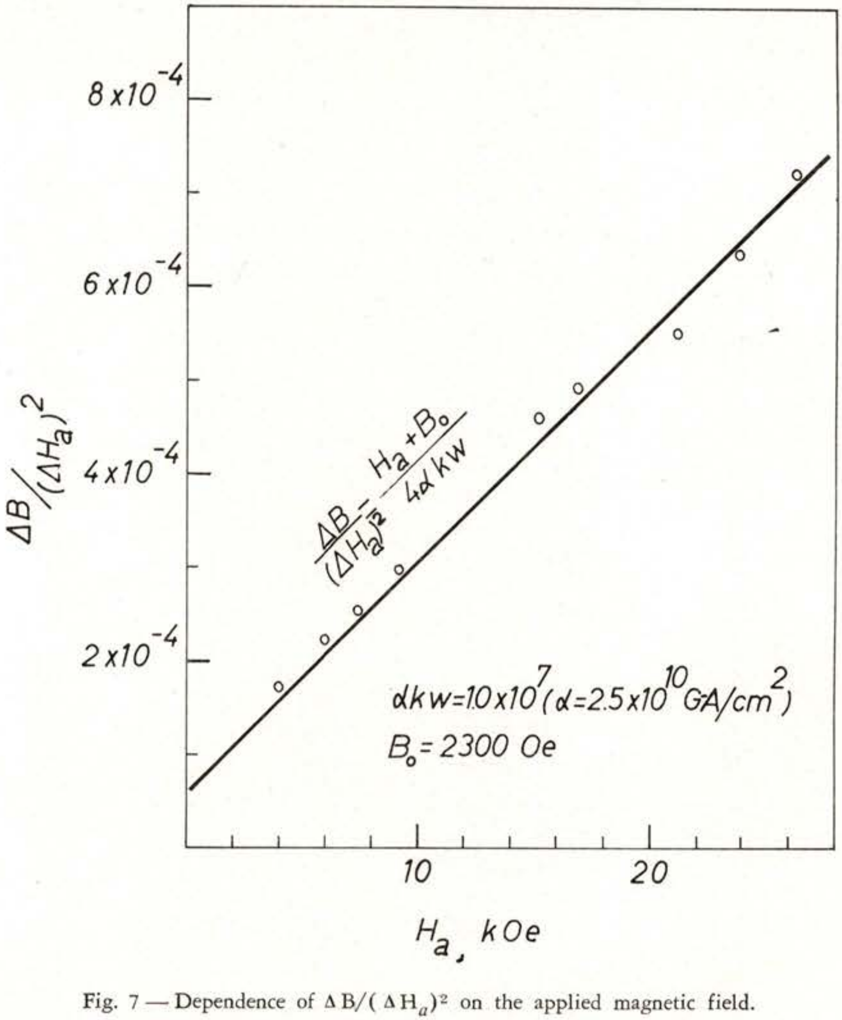

Fig. 7 — Dependence of $\Delta B/(\Delta H_a)^2$ on the applied magnetic field.

APPENDIX 1 — *Evaluation of the average magnetization for increasing H_a in case of full penetration and $0 < H_0 < (B_0^2 + 2\,\alpha\,k\,\omega)^{1/2} - B_0$.*

It is admitted in the following calculations that a superconducting slab, after being fully penetrated by the magnetic field in the negative direction, starts to be subjected to a positive applied magnetic field as Figure 8 shows.

From Maxwell's equation and supposing $J(B) = \alpha/(B + B_0)$ we have in $(0, d)$

$$-\int_{H_0}^{B(x)} (B + B_0)\, dB = \alpha k x$$

or

$$B(x) = ((H_0 + B_0)^2 - 2\alpha k x)^{1/2} - B_0 .$$

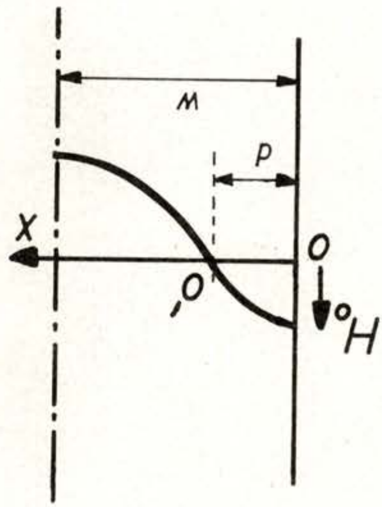

Fig. 8 — Profile of the magnetic induction for increasing H_a and $0 < H_0 < (B_0^2 + 2\alpha k \omega)^{1/2} - B_0$.

The value of d is determined by the condition $B(x) = 0$:

$$d = \frac{(H_0 + B_0)^2 - B_0^2}{2\alpha k}$$

Choosing the origin in $0'$ we have

$$-\int_{0}^{B(x)} (B + B_0)\, dB = \alpha k x$$

or

$$B(x) = B_0 - (2\alpha k x + B_0^2)^{1/2} .$$

The magnetic flux per unit length is then:

$$(0,\, d): \int_0^d \mathrm{B}\,(x)\; d\,x = \int_0^d \left\{((\mathrm{H}_0 + \mathrm{B}_0)^2 - 2\,\alpha\, k\, x)^{1/2} - \mathrm{B}_0\right\}\, d\,x\,;$$

$$(d,\, \omega): \int_0^{\omega - d} \mathrm{B}(x)\; d\,x = \int_0^{\omega - d} (\mathrm{B}_0 - (\mathrm{B}_0^2 + 2\;\alpha\, k\, x)^{1/2})\; d\,x =$$

$$= \mathrm{B}_0(\omega - d) + \frac{1}{3\,\alpha\, k}\;\mathrm{B}_0^3 - \frac{1}{3\,\alpha\, k}\,(2\,\alpha\, k\,(\omega - d) + \mathrm{B}_0^2)^{3/2}\;;$$

$$\text{Total } (0,\,\omega); -\frac{1}{3\,\alpha\, k}\left\{(2\,\alpha k \omega + 2\,\mathrm{B}_0^2 - (\mathrm{H}_0 + \mathrm{B}_0)^2)^{3/2} - (\mathrm{H}_0 + \mathrm{B}_0)^3\right\} -$$

$$-\frac{\mathrm{B}_0}{\alpha\, k}((\mathrm{H}_0 + \mathrm{B}_0)^2 - \mathrm{B}_0^2) + \mathrm{B}_0\,\omega \quad .$$

Then the average magnetization is

$$4\,\pi\,\overline{\mathrm{M}} = -\frac{1}{3\,\alpha\, k\,\omega}\left\{(2\,\alpha\, k\,\omega + 2\,\mathrm{B}_0^2 - (\mathrm{H}_0 + \mathrm{B}_0)^2)^{3/2} - (\mathrm{H}_0 + \mathrm{B}_0)^3\right\} -$$

$$-\frac{\mathrm{B}_0}{\alpha\, k\,\omega}\,((\mathrm{H}_0 + \mathrm{B}_0)^2 - \mathrm{B}_0^2) + \mathrm{B}_0 - \mathrm{H}_a \quad .$$

When $-\mathrm{H}_{c1} \leqslant \mathrm{H}_a \leqslant \mathrm{H}_{c1}$ the value of the magnetic field seen by the bulk, H_0, is zero and $d = 0$. Then the flux per unit length is

$$\mathrm{B}_0\,\omega + \frac{1}{3\,\alpha k}\;\mathrm{B}_0^3 - \frac{1}{3\,\alpha k}\,(2\;\alpha k\,\omega + \mathrm{B}_0^2)^{3/2}$$

and the average magnetization

$$4\,\pi\,\overline{\mathrm{M}} = \mathrm{B}_0 + \frac{1}{3\,\alpha\, k\,\omega}\,\mathrm{B}_0^3 - \frac{1}{3\,\alpha\, k\,\omega}\,(2\;\alpha k\,\omega + \mathrm{B}_0^2)^{3/2} - \mathrm{H}_a \;\; .$$

APPENDIX 2 — *Evaluation of the expressions for the average magnetization in the case of partial penetration*

We are going to deduce the expressions for the average magnetization in the case the maximum value of the magnetic field at the surface, H_{om}, is not enough for all the sample to be penetrated by the magnetic flux.

Decreasing Branch

We begin with the decreasing branch of the loop: the magnetic field at the surface, after passing by its maximum H_{om}, starts to decrease until it becomes zero. Figure 9 represents schematically the profile of the magnetic induction B (x) for a given value H_0 of the surface magnetic field.

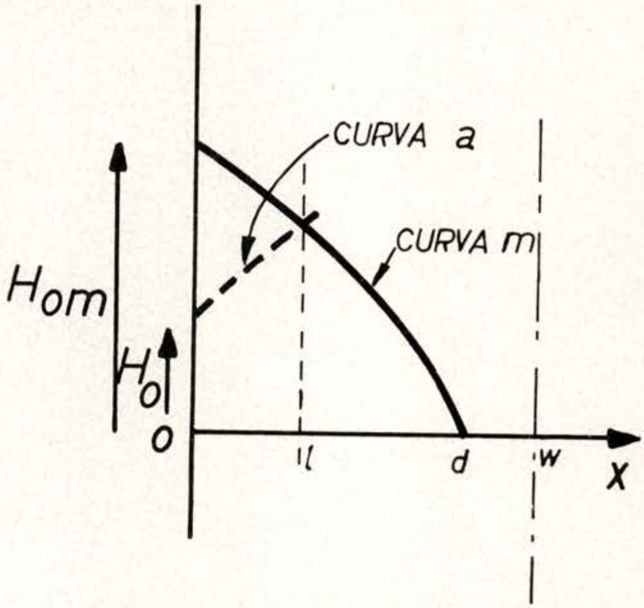

Fig. 9 — Profile of the magnetic induction for decreasing H_a in case of partial penetration.

By integrating

$$-\int_{H_{om}}^{B(x)} (B+B_0)\, dB = \alpha k x,$$

we get the expression of the curve m

$$B(x) = ((H_{om}+B_0)^2 - 2\alpha k x)^{1/2} - B_0.$$

Similarly, by integration of $\int_{H_0}^{B(x)} (B+B_0)\, dB = \alpha k x$, we obtain the expression of curve a

$$B(x) = ((H_0+B_0)^2 + 2\alpha k x)^{1/2} - B_0.$$

The value of the penetration depth of the magnetic field, d, is obtained considering $B(x)=0$ in the expression of the curve m

$$d=\frac{(H_{om}+B_0)^2-B_0^2}{2\alpha k}$$

Now we need to know the abcissae l of the point of intersection of the two curves. Its value is given by

$$l=\frac{(H_{om}+B_0)^2-(H_0+B_0)^2}{4\alpha k}$$

The flux can now be evaluated:

$$\int_0^l B(x)\,dx+\int_l^d B(x)\,dx=\int_0^l\left\{((2\alpha kx+(H_0+B_0)^2)^{1/2}-B_0\right\}dx+$$

$$+\int_l^d\left\{((H_{om}+B_0)^2-2\alpha kx)^{1/2}-B_0\right\}dx=\frac{2}{3\alpha k}\left(\frac{(H_{om}+B_0)^2+(H_0+B_0)^2}{2}\right)^{3/2}-$$

$$-\frac{B_0}{2\alpha k}\left((H_{om}+B_0)^2-B_0^2\right)-\frac{1}{3\alpha k}\left((H_0+B_0)^3+B_0^3\right).$$

Then the average magnetization is

$$4\pi\overline{M}=\frac{2}{3\alpha k\omega}\left(\frac{(H_{om}+B_0)^2+(H_0+B_0)^2}{2}\right)^{3/2}-\frac{B_0}{2\alpha k}\left((H_{om}+B_0)^2-\right.$$

$$\left.-B_0^2\right)-\frac{1}{3\alpha k}\left((H_0+B_0)^3+B_0^3\right)-H_a$$

Increasing Branch

Now we consider the situation where the magnetic field after having assumed its minimum value $-H_{om}$ increases from 0 to $+H_{om}$. Figure 10 shows the profile of the magnetic induction for a given value H_0 of the field at the surface. To calculate the magnetic flux per unit length we can add the flux between 0 and l' with twice the flux between $(d+l')/2$ and d:

$$\int_0^d B(x)\,dx=\int_0^{l'} B(x)\,dx+2\int_{\frac{d+l'}{2}}^d B(x)\,dx$$

In the interval $(0,\ l')$ we have $B(x)=((H_0+B_0)^2-2\alpha k x)^{1/2}-B_0$ and in $((d+l')/2,\ d)$, $B(x)=B_0-((H_{om}+B_0)^2-2\alpha k x)^{1/2}$ since the curve m' is symmetrical to the curve m (see Figure 10). The value of l' is determined putting $B(x)=0$ in the curve b:

$$l'=\frac{(H_0+B_0)^2-B_0^2}{2\alpha k}$$

Immediately we evaluate

$$(d+l')/2=\frac{(H_{om}+B_0)^2+(H_0+B_0)^2-2\,B_0^2}{4\,\alpha\,k}\ .$$

Now we can do the integration

$$\int_0^d B(x)\,dx=\int_0^{l'}\left[((H_0+B_0)^2-2\alpha k x)^{1/2}-B_0)\right]dx$$

$$+2\int_{(d+l')/2}^{d}\left[B_0-((H_{om}+B_0)^2-2\,\alpha k x)^{1/2}\right]dx\,,$$

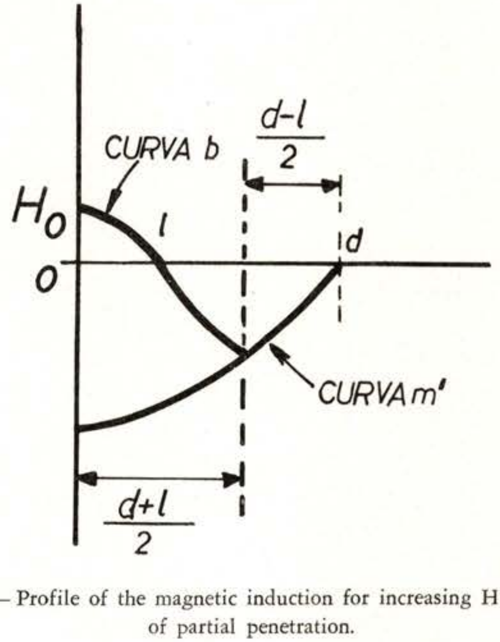

Fig. 10 — Profile of the magnetic induction for increasing H_a in case of partial penetration.

and finally obtain

$$4\,\pi\,\bar{M} = \frac{1}{3\,\alpha\,k\,\omega}\left[(H_0+B_0)^3+B_0^3\right] + \frac{B_0}{2\,\alpha\,k\,\omega}\left[(H_{om}+B_0)^2 - 2\,(H_0+B_0)^2 + B_0^2\right] - \frac{2}{3\,\alpha\,k\,\omega}\left[\frac{(H_{om}+B_0)^2-(H_0+B_0)^2}{2}+B_0^2\right]^{3/2} - H_a\,.$$

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