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IRREVERSIBLE MAGNETIZATION IN SUPERCONDUCTORS

by

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I. Introduction:

This study is an outgrowth of a previous work¹⁾ on superconducting wires carrying alternating currents. In that work it was shown that the critical current at which the wire entered the normal state was approximately that of Silsbee's hypothesis. However, an e.m.f. developed across the wire while it was in the superconducting state, and integration of the $V - I$ curve showed that there was a power loss associated with this e.m.f. The shape and magnitude of the $V - I$ curve suggested that magnetic hysteresis within the superconductor was an important factor. Dr. Houston recently suggested that flux flow may also be a factor; however, that has not been investigated in this study.

This present work was started to determine whether d.c. magnetization curves had sufficient hysteresis to account for the a.c. behavior. After it was shown that the d.c. hysteresis was large enough, the d.c. measurements were continued in order to describe better the sources of this irreversibility. D.c. measurements were made rather than a.c. because the former offered greater precision and sensitivity and better control of temperature, shape and magnetic field.

The three important sources of hysteresis investigated were: supercooling, geometric effects, and inhomogeneities. Supercooling refers to the existence of the normal state when the external field is below $H_c(T)$. By occurring only

for decreasing magnetic fields or temperatures, it leads to hysteresis. Multiply connected or irregularly shaped samples have hysteresis which arises as a geometric effect and is associated with nonuniform magnetization in the mixed or intermediate state. One example is a bump on an ellipsoid. The third source, inhomogeneities, refers to crystalline imperfections which are able to trap flux. These include impurities, strains, dislocations, grain boundaries, etc.

The object of this experiment was to make observations on each source with as little influence as possible from the other sources and to check for inter-relations between the sources. The behavior of these sources was then related to such important material characteristics as T_c , $H_c^{(0)}$, H_{c2} , etc. Most previous studies did not attempt to treat this wide range of variables, and in addition they did not have both the high precision and the good sensitivity for measuring the magnetization as were used in this study.

It is hoped that this experiment will help give better understanding of superconductivity by considering why there should be any irreversible behavior. Such an understanding may be quite useful for application to problems such as reducing a.c. losses or describing the irreversible behavior of superconducting solenoids.

II. Theory:

The magnetization of carefully prepared single crystal ellipsoidal superconductors is almost a reversible function of external field. Nevertheless, some hysteresis does occur for all samples and it is of interest to try to determine the source of this irreversibility. In order to differentiate between various sources it is necessary to understand the differing dependence of their hysteresis on external field, temperature, material and shape.

To provide a background for the discussion of irreversible behavior let us first review related reversible phenomena. As thermodynamics will be used, it is necessary to carefully define work, the extent of the system and other parameters. The work term dW implies work that is done on the external surroundings of the system. In general this work is represented by the product of an intensive parameter (generalized force such as pressure) multiplied by the change in an extensive parameter (generalized displacement such as volume). Thus the work done by expansion of a system is $dW = PdV$ or the total work for a large volume change with P a function of V is

$$W = \int_{V_i}^{V_f} P dV$$

For magnetic materials, the generalized force is the magnetic intensity H , and the generalized displacement may be either the magnetic induction B or the magnetization M .

Two possible definitions of the thermodynamic system are given by Callen²⁾ when he considers the work done on a battery by a thermodynamic system that is in the field of a solenoid powered by the battery. His first definition of the system divides the work into two parts. The first part is

$$dW_{\text{vac}} = - d \left[\frac{1}{8\pi} \int H_{\text{ext}}^2 dV \right] \quad (2-1)$$

and represents the work done by the vacuum. H_{ext} is the field intensity at the position of dV when there is no material in the solenoid. The second part is

$$dW_{\text{d}} = - \int H_{\text{ext}} dM dV$$

which represents the work done by magnetic changes of the elementary dipoles of the thermodynamic system. If the fields and magnetization are uniform as with an ellipsoid in an infinitely long solenoid

$$dW_{\text{tot}} = - \frac{V}{4\pi} H_{\text{ext}} dH_{\text{ext}} - V H_{\text{ext}} dM$$

By expanding the differentials and integrating over all space, Heine³⁾ shows that this is equivalent to

$$dW_{\text{tot}} = - \frac{V}{4\pi} H_i dB_i \quad (2-2)$$

Here H_i and B_i refer to the local fields within the thermodynamic system. London⁴⁾ preferred to use this system and

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these local values for his thermodynamic arguments.

Callen's second definition of the system excludes the vacuum by dropping the term given in Eqn. 2-1 and therefore has

$$dW_{TOT} = -V H_{EXT} dM \quad (2-3)$$

for an ellipsoid. Many recent authors such as Lynton⁵⁾ use this last definition of the system.

After selecting the system and variables, one can derive other useful thermodynamic relations for superconductors by using the laws of thermodynamics. Conservation of energy is required by the first law, which says

$$dU = dQ - dW \quad (2-4)$$

where dU is the increase of internal energy of the system, dQ is the energy entering the system as heat flow, and dW is the energy that left the system as work. As dQ is not an exact differential, one introduces the entropy S , which is a state function. The second law says

$$dS \geq \frac{dQ}{T} \quad (2-5)$$

It is often of interest to determine changes of internal energy that exclude that part associated with heat. For this TS is subtracted from the internal energy to give the Helmholtz free energy

$$F = U - TS \quad (2-6)$$

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This is a useful quantity, for it represents the mechanical part of the free energy that is available to do isothermal work. Equations 2-4, 2-5, and 2-6 give

$$dF \leq -dW - SdT,$$

which becomes

$$dF = -PdV + VH_{\text{ext}}dM - SdT \quad (2-7)$$

if the process is reversible.

Usually it is more convenient to measure changes of the intensive parameters P and H_{ext} rather than V and M . To obtain a function with P and H_{ext} as independent variables one uses the transformation

$$G = F + PV - VH_{\text{ext}}M,$$

which gives

$$dG \leq -SdT + VdP - VMdH_{\text{ext}} \quad (2-8)$$

where equality implies a reversible process. This Gibb's function is particularly important for phase changes that occur for constant values of the intensive parameters. Therefore a reversible phase transition must have $dG = 0$.

Although G is continuous, there may be discontinuities in the partial derivatives of G with respect to temperature at the phase transition. The order of a transition is defined as the lowest order derivative of G which is discontinuous.

If one uses the Gibb's free energy per unit volume g

instead of G , then $dG = 0$ also implies $dg = 0$. For the superconducting phase transition of a local region dV , the condition $dg = 0$ gives

$$g_n (H_{SN}, T, P) = g_s (H_{SN}, T, P).$$

Also, Equation 2-8 gives

$$\left[dg = -M dH_L \right]_{TP}$$

which shows that normal metal with negligible susceptibility has

$$g_n (H_{SN}, T, P) = g_n (0, T, P)$$

and that superconducting material has

$$g_s (H_{SN}, T, P) - g_s (0, T, P) = - \int_0^{H_{SN}} M dH_L$$

where H_L is the field intensity that would exist in the volume element dV if there were no material in dV , but the sample was otherwise unchanged. H_{SN} is the strength of H_L when the region dV completely enters the normal state. The thermodynamic bulk critical field is defined by the relation

$$\frac{H_{CS}^2}{8\pi} \equiv \int_0^{H_{SN}} M dH_L \quad (2-9)$$

Combining these relations gives

$$g_n (0, T, P) - g_s (0, T, P) = \frac{H_{CS}^2(T)}{8\pi} \quad (2-10)$$

Gorter and Casimir⁶⁾ estimated $g_s(O, T, P)$ by assuming a two fluid model with

x = fraction of normal electrons

$1 - x$ = fraction of superconducting electrons

and

$$g_s(O, T, P) = x^{\frac{1}{2}} g_n(T) + (1-x) B$$

where B is a constant and $g_n(T) = -1/2 \gamma T^2$ is the temperature dependent part of the Gibb's free energy per unit volume of free electrons. Minimizing $g_s(O, T, P)$ with respect to x , taking $x(T_c) = 1$ and using Equation 2-10 gives

$$x = \frac{T^4}{T_c^4} \quad \text{and} \quad H_{cs}(T) = H_{cs}(0) \left[1 - \frac{T^2}{T_c^2} \right] \quad (2-11)$$

Experimentally small deviations from this equation are observed. The B.C.S.⁷⁾ theory has been able to account for some of these deviations by using a temperature dependent energy gap. For low temperatures it predicts

$$H_{cs}(T) \approx H_{cs}(0) \left[1 - 1.07 \left(\frac{T^2}{T_c^2} \right) \right] \quad (2-12)$$

which is of proper sign to correct for V, In, and Sn, but is somewhat large. Lead and mercury need a correction of opposite sign, and it has been suggested that their low Debye temperatures necessitate corrections to the B.C.S. choice of a constant interaction potential.

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The two fluid model also predicts the temperature dependence of the penetration depth in London's⁴⁾ equations.

His equations are

$$\vec{E} = \frac{4\pi\lambda^2}{c^2} \dot{\vec{J}}$$

and

$$\frac{4\pi\lambda^2}{c} \nabla \times \vec{J} + \vec{H} = 0,$$

which give

$$\nabla^2 H = \frac{H}{\lambda^2}$$

λ is called the penetration depth and shows that magnetic fields are not excluded entirely from the sample but penetrate as

$$H(x) = H(0) e^{-\frac{x}{\lambda}}$$

In London's theory

$$\lambda^2 \equiv \frac{mc^2}{4\pi n_s e^2}$$

where n_s is the number of superconducting electrons. The two fluid model gives

$$n_s = n_{\text{TOT}} (1 - \alpha)$$

where $\alpha = \frac{T^4}{T_c^4}$ is the fraction of normal electrons and n_{TOT} is the total number of electrons. Combining shows that

$$\lambda(T) = \frac{\lambda(0)}{\sqrt{1 - \frac{T^4}{T_c^4}}}$$

This relation agrees rather well with experimental results and fairly closely approximates the B.C.S. prediction.

This penetration of magnetic fields reduces the magnetization from that expected for a body with $B_1 = 0$. The reduction means that Eqn. 2-9 does not have $H_{SN}(T) = H_{cB}(T)$, but has $H_{SN}(T) > H_{cB}(T)$ for samples with dimensions comparable to the penetration depth. Very thin films of thickness $a \ll \lambda$ have

$$H_{SN}(T) \propto H_{cB}(T) \frac{\lambda(T)}{a}$$

This penetration of magnetic fields seems to suggest that the Meissner expulsion of flux when the field is reduced from a large value is energetically unfavorable. The magnetic energy should be reduced by having the superconductor divide into superconducting threads which have dimensions smaller than λ and are separated from each other by extremely thin normal regions. London⁴⁾ solved this problem by arguing that there must be a surface energy associated with the normal to superconducting interface. He showed that the surface energy per unit area α_{NS} must satisfy the condition

$$\alpha_{NS} > \frac{\lambda}{2} \frac{H_{cB}^2}{8\pi}$$

However, his theory could not give a good estimate of the magnitude of α_{NS} , because it assumed that the superconductor was characterized by an ordered state that remained unchanged with position near the N-S interface. Likewise, the fraction of

normal electrons x in the two fluid model is a constant less than one in the superconductor and changes abruptly to one at the N-S interface.

In 1950 Pippard⁸⁾ suggested that x must be replaced by an order parameter that changes gradually near an interface. He approximated this variation by saying that it was equivalent to moving the interface a distance ξ , the coherence length, into the superconducting region. Because the free energy per unit volume of a superconductor is $\frac{H_{c0}^2}{8\pi}$ lower than that of the normal state, the free energy increase is approximately $\frac{A\xi H_{c0}^2}{8\pi}$ where A is the area of the interface. Subtracting the decrease in energy due to the magnetic field penetration gives the net surface energy

$$A \alpha_{NS} \approx \frac{A \xi H_{c0}^2}{8\pi} - \frac{A \lambda H_{c0}^2}{8\pi}$$

Often it is easier to use a quantity with a dimension of length rather than α_{NS} , and one uses Δ defined by

$$\Delta \equiv \frac{8\pi \alpha_{NS}}{H_{c0}^2} \approx \xi - \lambda \quad (2-14)$$

In the same year that Pippard introduced the idea of a coherence length, Ginzburg and Landau⁹⁾ developed an alternative method that was based on variation of the order parameter. Their calculation gave

$$\Delta \approx 1.89 \frac{\lambda_0}{K}$$

where κ was defined by

$$\kappa \equiv \left[\frac{2 e^*{}^2}{\hbar c^2} H_{c0}^2 \lambda_0^4 \right]^{\frac{1}{2}} \quad (2-15)$$

This relationship holds only for small κ and near the critical temperature. Gorkov's¹⁰⁾ derivation from the B.C.S. theory shows that the approaches of Pippard and Ginzburg are equivalent and κ can be derived in terms of ξ and vice versa. To do this it is necessary to use the relation shown by B.C.S.⁷⁾ that

$$\frac{1}{\xi_0} = \frac{\pi \epsilon_0(0)}{\hbar v_0}$$

where $2 \epsilon_0(0)$ is the energy gap at $T = 0$ and v_0 is the velocity of electrons at the Fermi surface.

Looking again at Equation 2-14, one sees that for $\xi < \lambda$ the surface energy becomes negative. Superconductors with negative surface energy are called type II to distinguish them from those with positive surface energy which are called type I. Type II superconductors can also be described as those with $\kappa \geq \frac{1}{\sqrt{2}}$. The magnetic properties of these two types of superconductors are different. Type II superconductors have a second order transition into the normal state, and although type I superconductors have a second order transition at T_c in zero field, they have a first order transition to the normal state in the presence of a magnetic field.

Abrikosov¹¹⁾ has shown that the Ginsburg-Landau equations indicate a nonzero order parameter above H_{cB} up to $H_{c2} = \sqrt{2} \kappa H_{cB}$ for $\kappa \geq \frac{1}{2}$. However, these type II superconductors have flux penetrating below H_{c2} . This flux is quantized and has associated current vortices. He also found that H_{c1} , the lowest field at which flux quanta exist in type II material, is lower than H_{cB} . The region between H_{c1} and H_{c2} is called the mixed state and exists even for a demagnetization factor D of zero. Goodman¹²⁾ gives an alternative derivation of H_{c1} , in which he essentially defines H_{c1} as the external field that gives zero net surface energy. The net surface energy is approximated by

$$[\alpha_{ns}]_{NET} \sim \frac{A \xi H_{cB}^2}{8\pi} - \frac{A \lambda H_{EXT}^2}{8\pi}$$

Thus he finds

$$H_{c1}^2 \sim H_{cB}^2 \frac{\xi}{\lambda}$$

However, de Gennes⁴⁰⁾ says that when one considers a flux quantum with diameter $\xi \ll \lambda$, minimizing the Gibb's free energy gives

$$\frac{H_{c1}}{H_{cB}} \sim \frac{\xi}{\lambda} \quad (2-16)$$

For ellipsoidal specimens with $D \neq 0$ there is flux penetration below H_{c1} for type II superconductors and below H_{cB} for type I. This follows from the condition

$$H_{INT} = H_{EXT} - 4\pi D M$$

which was derived by Stoner¹³⁾ for any ellipsoid with uniform magnetization. Before flux penetrates, a superconductor has

$H_{INT} = -4\pi M$, which gives

$$H_{INT} = \frac{H_{EXT}}{1-D}$$

However, when $H_{INT} = H_{cB}$ a type I superconductor enters the intermediate state. Abrikosov¹¹⁾ has suggested that an ellipsoidal type II superconductor may also have an intermediate state at $H_{int.} = H_{c1}$, but this has not been verified experimentally.

The intermediate state is characterized by rather large superconducting domains, as opposed to the uniform vortex structure of the mixed state. The large size of the domains is a result of the positive surface energy, which makes the vortex array energetically unfavorable. As the external field is increased above $H_{EXT} = H_{cB}(1-D)$ the domains shrink with an associated latent heat. London⁴⁾ averaged the fields over the superconducting domains and normal channels and used the free energy to derive the relation

$$-4\pi M = \frac{H_c - H_{EXT}}{D}, \quad (2-17)$$

which approximates the actual behavior in the intermediate state.

Another model of the intermediate state proposed by Kuper¹⁴⁾ represents the superconducting domains as ellipsoids. He minimizes the thermodynamic potential suggested by Landau for the intermediate state and finds the size and spacing of the ellipsoids. His model suggests a finite latent heat for the transition from the intermediate state to the normal state. For large ellipsoids it is quite small and the observed magnetization should appear linear as predicted by London.

Kuper says that smaller specimens may have an abrupt drop to zero magnetization at the transition to the normal state. In addition he predicts a horn-shaped rise in the magnetization curve near

$$H_{\text{EXT}} = H_c (1 - D)$$

The size of the horn is determined by the positive surface energy, the sample size, and the domain structure. The horn appears only for increasing fields and is therefore a source of hysteresis; however, this hysteresis is small for large samples.

Type I superconductors do have another form of hysteresis associated with their positive surface energy and latent heat. They can have either the normal phase existing below H_c or the superconducting phase existing above H_c . These phenomena are respectively called supercooling and superheating and mean that the magnetization curve is not reversible near H_c . Faber¹⁵⁾ has made a careful study of this and has

found that as in other first order phase transitions, a minimum sized nucleation center is necessary to initiate the phase change. He has also measured the velocity at which the phase boundary spreads from the nucleation center and has related it to eddy currents, the surface energy, the conductivity, and the external field.

Type II superconductors cannot have supercooling or superheating because their transition to the normal state is second order. This significant result stems from the difference between the domain structure and the uniform vortex structure of the mixed state and is based on Pippard's¹⁶⁾ statement that supercooling never occurs for second order phase transitions.

In addition to this hysteresis from supercooling, real ellipsoids can have other types of flux trapping. There are two important sources of this added irreversible behavior. First, there are geometric effects such as surface irregularities or voids within the sample, and secondly, there are inhomogeneities within the material of the sample.

The superconducting torus is a simple example that shows that flux trapping can arise from geometric effects. This shape was studied by Schoenberg¹⁷⁾ who used rings with $\frac{b}{a} \gg 1$ where b is the radius of the ring and a is the wire radius. His theoretical predictions shown in Figure 1 were in good agreement with his experimental results.

Initially at point O in Figure 1, no fields are present, then as $H_{\text{ext.}}$ is increased, currents are induced in the ring preventing flux change at the center of the ring. At point A, the sum of the external field plus the field of the induced currents equals $+H_c$ on the outer edge of the ring. For higher fields, flux penetrates to the center of the loop, thus maintaining $+H_c$ at the outer rim. The ring enters the intermediate state at B and cannot shield the interior from higher fields. For decreasing fields the ring changes from the intermediate state to the pure superconducting state at point B. Then flux at the center of the ring is trapped in an amount consistent with the condition that the field at the inner edge of the ring cannot exceed H_c . Below point D, currents are limited by the condition that the field on the outer edge of the ring cannot be lower than $-H_c$.

Dolecek¹⁸⁾ extended Schoenberg's analysis to a thick torus which had $\frac{b}{a} = 1.4$ and found the behavior shown in Figure 2. The residual moment with no external field is reduced from Schoenberg's value, but it will be shown that it is unlikely that trapping disappears even when $b = a$ and the body is not longer multiply-connected.

Another way to create a simply-connected body is to use the altered shape suggested by London.⁴⁾ This is shown in Figure 3 where the hole of the torus has been closed with the thin bridge of superconducting material to make the body

simply-connected. However, for decreasing fields the ring enters the pure superconducting state while the bridge is in the intermediate state thus preventing the bridge from completely expelling its flux. For a thin bridge the results will be similar to those of the hollow torus but thicker bridges will have excluded more flux before the loop becomes purely superconducting and therefore will give less flux trapping. Only ellipsoidal specimens with uniform magnetization would be expected to be free from this type of trapping.

Schoenberg's¹⁹⁾ tests on short tin cylinders illustrate this fact rather well. Fields parallel to the cylinder axis had much greater trapping than transverse fields on the same sample. Apparently, the large trapping for longitudinal fields stemmed from the sharp edges of the cylinder. This is the region of greatest nonuniformity in the intermediate state. Rounding the sharp edges with a file considerably reduced the trapping. The contrasting small trapping for transverse fields probably resulted from the unfavorable configuration for the formation of large trapping rings. The magnetization for transverse fields was approximately the same as that expected for an ellipsoidal specimen with a demagnetization factor of $1/2$. Schoenberg found that the trapping in longitudinal fields had the same temperature dependence as the critical field and that a magnetization curve at T_1 could be superimposed on that at T_2 if both $-4\pi M$ and $H_{\text{ext.}}$ were multiplied times the factor
$$\frac{1 - \frac{T_c^2}{T_2^2}}{1 - \frac{T_c^2}{T_1^2}}.$$

This shape dependent hysteresis can be responsible for trapping in ellipsoids that have surface irregularities. Consider an imperfection with a demagnetization factor greater than that of the ellipsoid as is shown in Figure 4. For purposes of illustration, the imperfection is chosen as half a thick torus and the temperature as $T = 0$. Before the half torus reaches point A with field $H_A(0)$ on Figure 2, there will be no trapping. If the imperfection is small and at the equator of the ellipsoid, then point A corresponds to an external field

$$H_{EXT} = H_A(0) \left[1 - D_{ELLIPSOID} \right].$$

Raising $H_{ext.}$ to some maximum value larger than this and returning it to zero results in a residual magnetization $-4\pi M_R$. This trapped magnetization increases linearly with H_{max} up to

$$H_{MAX} \approx H_B(0) \left[1 - D_e \right]$$

and is constant at $-4\pi M_{MAX}(0)$ for higher fields.

For a temperature $T > 0$, the half torus reaches point A at a lower field

$$H_A(T) = H_A(0) \left[1 - \frac{T^2}{T_c^2} \right].$$

Thus, the half torus starts trapping flux if the external field is raised to

$$H_{MAX}(T) = H_A(0) \left[1 - D_e \right] \left[1 - \frac{T^2}{T_c^2} \right]$$

If one applies

$$H_{MAX} \geq H_B(0) \left[1 - D_e \right] \left[1 - \frac{T^2}{T_c^2} \right]$$

and reduces $H_{ext.}$ to zero, then the residual magnetization will be

$$-4\pi M_{MAX}(T) = -4\pi M_{MAX}(0) \left[1 - \frac{T^2}{T_c^2} \right]$$

Of course it is not necessary for the imperfections to be shaped like the half torus, and there may be many types of irregularities. Probably those with sharp edges trap the most flux. This distribution of shapes will determine the way that $-4\pi M_{TRAP}$ varies with H_{max} , and therefore prediction of the functional relationship is quite difficult. For if H_{max} is raised to $H_{max} + \Delta H$, then $-4\pi M_T(H_{MAX})$ will be increased not only by regions which had no trapping at H_{max} but also by regions that had trapping at H_{max} and behave like Dolecek's torus between A and B.

Although calculation of $-4\pi M_T(H_{MAX}(T_1))$ at some temperature T_1 by considering the shape of surface imperfections would be quite difficult, it is not difficult to predict $-4\pi M_T(H_{MAX}(T_2))$ at some temperature T_2 after the trapping has been measured for all $H_{MAX} < H_c(1-D)$ at T_1 . As with Schoenberg's short cylinder, the magnetization curve at T_2 of each surface irregularity is found by multiplying both $-4\pi M(T_1)$ and $H_{ext.}(T_1)$ by the factor $\frac{1 - \frac{T_2^2}{T_c^2}}{1 - \frac{T_1^2}{T_c^2}}$.

Therefore, the net trapping at T_2 resulting from all contributing surface irregularities will be

$$-4\pi M_T (H_{MAX}[T_2]) = -4\pi M_T (H_{MAX}[T_1]) \left[\frac{1 - \frac{T_2^2}{T_c^2}}{1 - \frac{T_1^2}{T_c^2}} \right] \quad (2-18)$$

for the field

$$H_{MAX}[T_2] = H_{MAX}[T_1] \left[\frac{1 - \frac{T_2^2}{T_c^2}}{1 - \frac{T_1^2}{T_c^2}} \right] \quad (2-19)$$

If T_2 is taken as $0^\circ K$, this transformation gives $-4\pi M_T$ vs. H_{MAX} at $T = 0^\circ K$.

By measuring $-4\pi M_T$ vs H_{MAX} for two different temperatures it is possible to predict T_c . Dividing Equation 2-18 by 2-19, shows that

$$\frac{-4\pi M_T (H_{MAX}[T_2])}{H_{MAX}[T_2]} = \frac{-4\pi M_T (H_{MAX}[T_1])}{H_{MAX}[T_1]}$$

when

$$H_{MAX}[T_2] = H_{MAX}[T_1] \left[\frac{1 - \frac{T_2^2}{T_c^2}}{1 - \frac{T_1^2}{T_c^2}} \right] \quad (2-20)$$

Therefore, if one finds $H_{max}[T_2]$ and $H_{max}[T_1]$, which give the same value for $\frac{-4\pi M_T}{H_{MAX}}$, then the H_{max} values are related

by Equation 2-20, which can be solved to give

$$T_c = \sqrt{\frac{H_{MAX}[T_1] T_2^2 - H_{MAX}[T_2] T_1^2}{H_{MAX}[T_1] - H_{MAX}[T_2]}} \quad (2-21)$$

These predictions should also hold for type II superconductors as long as $H_{max} < H_{c1}(1-D)$. But they are generally not applicable after the sample enters the mixed or intermediate state, because then flux penetration is no longer limited to surface irregularities. This means that internal inhomogeneities can also cause trapping, and usually their hysteresis overshadows the small amount of flux trapped within surface irregularities.

The temperature and field dependence of trapping due to inhomogeneities is not easily predicted, but there are two models which seem fairly successful. Mendelssohn's²⁰⁾ sponge model is generally used for type I superconductors and two-phase alloys. Type II superconductors are usually treated with Anderson's pinning model^{21,22)}.

Mendelssohn described the sponge as "formed by annular regions of high threshold value impenetrable for magnetic flux that has once been caught in them." He said, "This had the effect of filling the material inside the meshes with flux which remained 'frozen-in' rendering it non-superconductive." However, he stated that a large fraction of the volume may be associated with the filaments of the sponge mesh. In addition he emphasized "that the whole of

the metal is superconductive, although part of it may be rendered normal by 'frozen-in' flux."²⁰⁾ Thus flux which is 'frozen-in' by a localized inhomogeneity may pass through regions with bulk critical field in order to escape at the sample surface.

In this article the sponge model will be interpreted in a somewhat modified picture. It is assumed that in addition to high field filaments, low critical field inclusions may permit a sponge structure. In this case material with the bulk critical field forms the sponge mesh. Also, instead of describing the mesh by a critical field, it is described by the critical current density. It will be shown that this treatment bears a great similarity to the pinning model, which has "frozen-in" flux because of forces acting on flux lines. But with type I superconductors the positive surface energy requires an interface between the "frozen-in" flux and the superconducting regions. The surface energy associated with this interface is more easily treated by a sponge model than a pinning model.

The critical current density is not as greatly influenced by filament size as the critical field. London⁴⁾ recognized the importance of the critical current density when he chose as a fundamental relation the equation

$$g_m - g_s = \frac{4\pi\lambda^2}{c^2} J_c^2 \quad (2-22)$$

Equation 2-10 relates this to H_{cB} . Bean²³⁾ used a sponge structure with constant critical current density in the filaments to predict the behavior of his sample, which was made by pressing lead into porous glass. Livingston²⁴⁾ has studied two-phase alloys with one phase superconducting and one phase normal. These normal inclusions did give a sponge-like behavior; however, often the phases were not completely segregated and the properties were influenced by having a solution of one component in the other.

A sponge structure is not possible in an ellipsoid of a pure metal without inhomogeneities. Stable sponges can form only at regions where the free energy or surface energy differ from the bulk properties. As the external field is reduced, inhomogeneities with high critical field or negative surface energy may form closed filament loops, which prevent the flux inside the loop from being expelled. Inhomogeneities with low critical field will serve as the normal channels between superconducting domains in the intermediate state, but they will not be able to expel their flux because the remainder of the ellipsoid will be in the pure superconducting state and will act similarly to Schoenberg's torus. The important range of external fields that determines whether an inhomogeneity will trap flux is that near the critical field and in the intermediate state. At lower fields flux may escape as with Schoenberg's torus, but a stability condition is mainly concerned with the manner in which the inhomogeneity

first forms a closed current loop at a field where homogeneous material would have expelled flux.

This suggests that an approximate stability condition is

$$\left| \Delta(g_n - g_s) \right| - \frac{A}{V} \alpha_{BF} \geq \frac{4\pi\lambda^2}{c^2} (J_T^2 + J_{MAG}^2) \quad (2-23)$$

where $\left| \Delta(g_n - g_s) \right|$ represents the difference in free energy per unit volume between the inhomogeneity and the bulk.

$\frac{A}{V}$ is the ratio of the surface area to the volume of the inhomogeneity, and α_{BF} is the surface energy required to create an interface between the bulk and the inhomogeneity. This interface is necessary to enable the two adjacent regions to carry different currents. The term on the right represents the energy per unit volume associated with electronic currents. J_T is the current density of the currents that shield the center of the loop from flux changes. J_{mag} is associated with the magnetization of the region carrying currents and is approximated by

$$\frac{4\pi\lambda^2}{c^2} J_{MAG}^2 \approx \int_0^{H_{FIL}} M_{FIL} dH_{FIL}$$

where H_{fil} is the magnetic intensity at that region. This term ascertains that the shielding current of a region goes to zero when that region reaches its critical field.

If α_{BF} is positive, then the size of a stable trapping center will be determined by $\frac{A}{V}$ which varies approximately as $\frac{1}{d}$ where d is the radius or smallest width of the inhomogeneity.

In this case, $|\Delta(g_n - g_s)|$ must be non-zero, which means that the critical temperature of the inhomogeneity must be different from that of the bulk. This is true when one can use Eqn. 2-10 and the observations by Pippard⁸⁾ and Muench²⁵⁾, which showed that impurities and strains change H_{cB} and T_c in such a way that $\frac{H_{cB}}{T_c}$ remains constant. As changes in T_c are usually small, $|\Delta(g_n - g_s)|$ will be small, and therefore stability for $\alpha_{BF} > 0$ requires $d \gg \lambda$.

Negative α_{BF} seems to favor very small d ; however, perhaps $[\alpha_{BF}]_{NET}$, as used to derive Eqn. 2-16, is more significant. In as much as $[\alpha_{BF}]_{NET}$ becomes positive for small H_{Fill} there may be small filaments with $d \lesssim \lambda$ that are stable at high fields near the intermediate state, but not near $H_{ext.} = 0$. Also since

$$\frac{A}{V} \alpha_{BF} \sim \frac{H_{cs}^2(\tau)}{8\pi} (\xi(\tau) - \lambda(\tau)) \sim \frac{H_{cs}^2(0)}{8\pi} (\xi(0) - \lambda(0)) \frac{\left(1 - \frac{T^2}{T_c^2}\right)^{\frac{3}{2}}}{\sqrt{1 + \frac{T^2}{T_c^2}}} \quad (2-24)$$

these small filaments will be more stable at low temperatures.

For both large and small filaments J_{mag} assures that J_T goes to zero at the critical field of the filament. A large filament would be influenced by J_{mag} in association with flux expulsion from the filament. From Eqns. 2-10 and 2-22 one finds that

$$J_c^2(\tau) \propto \frac{H_{cs}^2(\tau)}{\lambda^2(\tau)} = \frac{H_{cs}^2(0)}{\lambda^2(0)} \left[1 - \frac{T^2}{T_c^2}\right]^3 \left[1 + \frac{T^2}{T_c^2}\right]$$

Thus the temperature dependent critical current should vary as

$$J_c(T) = J_c(0) \left[1 - \frac{T^2}{T_c^2} \right]^{\frac{3}{2}} \left[1 + \frac{T^2}{T_c^2} \right]^{\frac{1}{2}}$$

Bardeen²⁶⁾ has made a study of how J_c is affected by the scattering of quasi-particles at impurities and thin film boundaries. He gives the slightly modified form for thin films

$$J_c(T) = J_c(0) \left[1 - \frac{T^2}{T_c^2} \right]^{\frac{3}{2}}$$

This form is simply related to the critical field by

$$J_c(T) = J_c(0) \left[\frac{H_{cB}(T)}{H_{cB}(0)} \right]^{\frac{3}{2}} \quad (2-25)$$

Assuming that $J_T(H_{ext}=0)$ has the same temperature dependence as J_c gives a prediction for the way that the trapping of a sponge structure will depend on temperature. Thus a fixed array of filaments should have

$$-4\pi M_T \propto \left[\frac{H_{cB}(T)}{H_{cB}(0)} \right]^{\frac{3}{2}} \quad (2-26)$$

This relation is not expected to hold if the stability conditions of Eqn. 2-23 allow changes in the number of stable filaments with temperature. Therefore, Eqn. 2-26 is not expected to describe the temperature region very near to T_c

or the very small high field filaments of Eqn. 2-24 which appear at low temperatures.

Glover²⁷⁾ has studied thin tin films and found that their critical currents are described rather well by Eqn. 2-25. However, Meiklejohn²⁸⁾ found that tantalum gave

$$J_c(T) = J_c(0) \left[1 - \frac{T^2}{T_c^2} \right]^N \left[1 + \frac{T^2}{T_c^2} \right]^{\frac{1}{2}}$$

where $N = 2.3$. He felt that his was due to gaseous impurities and showed that although pure tin films had $N = 1.5$, N increased as indium impurity was added and $N = 2.3$ for 3% indium. A sponge structure that is anchored on impurities probably will be affected in a similar way.

These predictions do not necessarily hold for type II superconductors whose flux penetration occurs with an ordered array of current vortices. In the homogeneous material the fluxoid interaction will prevent the formation of filaments that can trap flux. Motion of these current vortices is more easily described by the pinning model of Anderson²¹⁾ than the sponge model, but it is of interest to see what predictions may be expected from a sponge model.

As a start, one could consider a ring of type II material. As long as the field at the outer edge of the ring does not exceed H_{c1} , flux will be excluded from the center. Above H_{c1} flux penetrates because the ring enters the mixed state and can no longer support the transport currents necessary to shield the interior. Such shielding would require that the

array of current vortices have a nonuniform distribution in the ring.

Next, consider inserting a smaller ring inside the first. This ring would be shielded from flux changes and carry no current until the outer ring reached the mixed state. Then, however, if the outer ring had no transport currents, it could provide no shielding for the inner ring, which would also be required to enter the mixed state. To prevent this, the sponge model assumes the threads are quite small and limited by their current density rather than H_{c1} . This means the outer ring would shield the inner one completely until it reached its critical current density J_c . For higher fields it would still have a transport current determined by

$J_c^2 = J_T^2 + J_{MAG}^2$. It is easy to see that a torus composed of many filaments could shield the center better than a single large ring. The large ring could only have surface currents whereas the threads would have almost uniform current density across the cross section. This is similar to the mechanism proposed by Bean²³⁾ to describe his lead filled glass; however, he did not include J_{mag} in his calculations and therefore his J_T did not go to zero when the field was very high.

But, as was shown, large single phase materials complicate this picture with interface energies and vortex interactions. In order to justify the formation of independent filaments, there must be inhomogeneities. Following

a natural impulse, one says high field threads are formed. As there are three critical fields, this is not sufficient. If the bulk critical field is high, then the critical current density will also be high. But one must also consider H_{c1} and H_{c2} . Increasing H_{cB} but not κ increases both H_{c1} and H_{c2} . However, increasing κ but not H_{cB} increases H_{c2} and decreases H_{c1} .

The temperature dependence of H_{c1} and H_{c2} become quite complicated because they depend on both $H_{cB}(T)$ and $\kappa(T)$. Eqn. 2-15 from Ginzburg's⁹⁾ theory gives the temperature dependence of κ as

$$\kappa(T) = \kappa(0) \left[\frac{1}{1 + \frac{T^2}{T_c^2}} \right]$$

A modification proposed by Gorkov and calculated by Helfand²⁹⁾ gives a somewhat different temperature dependence. If Abrikosov's derivation of H_{c1} is used, this temperature dependent κ should also give H_{c1} . Harden and Arp³⁰⁾ have performed the numerical integrations necessary to find $\frac{H_{c1}}{H_{cB}}$ from κ or vice versa. Another interesting approach would be to calculate $\frac{H_{c1}}{H_{cB}}(T)$ from Eqn. 2-16 by using the B.C.S. theory to find $\frac{\epsilon}{\lambda}$. Perhaps this would agree with Hecht's³¹⁾ experimental results which found $\frac{H_{c1}(T)}{H_{cB}(T)} = \text{Constant}$.

Inasmuch as the derivation of H_{c1} from κ does not agree well with experiment, it is somewhat difficult to predict the temperature dependence of trapping. Threads that are large compared to λ give better shielding for large H_{c1} , and therefore their trapping will increase with $H_{cB}(T)$; however, their

dependence on κ (T) is uncertain. J_c should also increase with H_{cB} , so trapping for small filaments should also increase with H_{cB} . Whereas large threads may be rather ineffective for trapping in the mixed state, small threads may remain effective to high fields where their trapping will be influenced by the temperature dependence of H_{c2} .

Nevertheless, this sponge model is not well suited to describing the motion of current vortices, and a somewhat easier approach is to postulate force fields for inhomogeneities. These force fields arise from variation of the vortex's free energy with position due to inhomogeneities in the material and may alter the regular vortex pattern. As a result, several lines can collect at a minimum to form a bundle. It seems that this minimum would correspond to a minimum of $(g_n - g_s)$ rather than of g_s . Regions with low g_s would have high critical current density and act as barriers to the motion of vortices.

Pushing these flux lines into bundles creates flux gradients in the sample. The force per unit length on one flux line in a field gradient is

$$F = - \frac{\Phi_0}{4\pi} \frac{\partial H_{\text{LOCAL}}}{\partial x}$$

For equilibrium this force must be balanced by an equal and opposite force arising from the free energy variation associated with the barrier. This pinning force is therefore

$$F_p = \frac{\Phi_0}{4\pi} \frac{\partial H_{\text{LOCAL}}}{\partial x} .$$

By using the density and strength of pinning centers as variables, Campbell³²⁾ has developed an approximate method for deriving the magnetization of a specimen. The field gradient is expanded as

$$\frac{\partial H_{\text{LOCAL}}}{\partial x} = \frac{\partial H_{\text{LOCAL}}}{\partial B_{\text{LOCAL}}} \frac{\partial B_{\text{LOCAL}}}{\partial x}$$

and $\frac{\partial H_{\text{LOCAL}}}{\partial B_{\text{LOCAL}}}$ for the specimen with pinning centers is assumed to have the same value as $\frac{\partial H}{\partial B_{\text{REV}}}$ in the reversible material. After approximating the reversible curve with straight lines, he is able to predict the local induction in a long cylinder in a longitudinal external field. This induction is a function of radial position, external field, the pinning force per isolated line, and the details of the reversible curve.

With

$$4\pi M_{\text{AVE}} = \frac{1}{V} \int (B_{\text{LOCAL}} - H_{\text{LOCAL}}) dV$$

he finds that the magnetization curve is determined by $a\beta$ where

a = radius of cylinder

β = constant for material

$$\beta \propto (\text{pinning force on isolated line}) \left(\frac{1}{\text{separation of pinning centers}} \right)^2 \times \frac{\partial B}{\partial H_{\text{rev}}}$$

Figure 5 shows the magnetization curve for small $a\beta$, which implies widely separated, weak pinning centers. A larger amount of hysteresis is seen in Figure 6 for large $a\beta$. These

curves do not have the proper behavior for large H_{ext} , because Campbell assumed the force of a pinning center is inversely proportional to the number of lines present i.e. $F \propto \frac{1}{B_{\text{local}}}$. This form does not require the pinning force to go to zero at H_{c2} .

Cline³³⁾ is better able to treat the region near H_{c2} by using the critical current density determined from resistive measurements to calculate the magnetization. He uses Maxwell's equation

$$\frac{\partial H}{\partial x} = \frac{4\pi}{c} J(H)$$

to find the field gradient in the sample and gives the maximum gradient as

$$\left. \frac{\partial H}{\partial x} \right|_{\text{max}} = \frac{4\pi}{c} J_c(H)$$

It was seen from Campbell's discussion that the maximum pinning force is also given by the maximum field gradient.

$$F_p = \frac{\Phi_0}{4\pi} \frac{\partial H_{\text{local}}}{\partial x}$$

One would therefore expect the critical current density to be proportional to the pinning force for a given local induction. This relation $F_p \propto J_c$ is interesting in that it suggests that the pinning model and the sponge model are closely related. This should not be surprising, because both are dependent on variations in the free energy.

Although these predictions of Cline and Campbell give the approximate shape of the observed magnetization curves, there is need for some type of stability condition to determine the temperature dependence of trapping. Anderson and Kim³⁴⁾ have predicted a linear rise of J_c at low temperatures by treating the thermal activation of flux bundles; however, their model does not consider new pinning centers becoming stable at low temperatures. Also their model does not include the limitation imposed on $J_c(T)$ by $H_{c2}(T)$.

III. Experimental Design:

It was shown above that magnetic hysteresis is expected to depend on the material, temperature, applied field, shape and inhomogeneities. Therefore, the experimental arrangement was designed to allow variations in each of these parameters.

Of the three materials studied, two, tin and lead, were type I, and the other, niobium, was a type II superconductor. Although all samples were in ellipsoidal shape, they did not have the same demagnetization factor D . Some were spheres with radius .5 cm. and $D = 1/3$, while others had major axis 3 cm. and minor axis .635 cm. with $D = .06$.

The tin and lead samples were made here at Rice from ingots or granular material with a purity of 99.9% or better. The single crystal tin sphere, which had been grown in a mold, had a pockmark about 1 mm. deep where there had been a transition to grey tin. During the experiment, it was oriented so as to have the magnetic field perpendicular to the plane of this imperfection. Tin and lead ellipsoids with $D = .06$ were made by first melting the metal in a vacuum to form a cylinder. This cylinder was then cold-rolled to 50% diameter and cut on a lathe to the proper shape. The final dimensions after polishing with Crocus cloth were accurate to about .005". A lead sample with the same dimensions as the somewhat distorted niobium ellipsoid was made in this same manner. A third lead ellipsoid was made from a cylinder that

had not been cold-rolled.

The niobium ellipsoid with $D \sim .06$ was grown as a cylindrical single crystal by Semi-Elements Inc. with a purity of at least 99.9%. After its edges were rounded to approximately ellipsoidal dimensions, the sample was polished. For most points along the major axis, the radius of the circular cross section was accurate to a few %, but near the ends it was as much as 20 to 30% large. Although the sample was a single crystal, Laue back reflection x-ray photographs indicated strains giving lattice distortions as large as 1%. The niobium sphere, which was spark cut by Metals Research Ltd., had a purity of 99.9999% and no observable lattice distortion. It was also a single crystal and had been electro-polished.

In order to measure the hysteresis of these samples, it was important to measure the magnetization quite accurately. Careful consideration showed that both precision and sensitivity were most easily achieved by the direct approach using search coils and galvanometers. The search coils shown in Figure 7 were connected in series with opposite winding directions. Their output was fed into a galvanometer, and when the sample was moved from one coil to the other, the deflection was proportional to the magnetization. The stops shown in Figure 8 were adjusted to maximize this deflection for a given magnetization. At the start of each run, this adjustment was made and was found to remain so constant that

variations in the endpoints of the sample's travel contributed less than .05% error.

As both search coils were at liquid helium temperature, their combined resistance was only about 30 ohms, even though they had 10,000 turns. This is smaller than the critical damping resistance of the galvanometer, and therefore the deflection was closely approximated by the flux galvanometer expression

$$\theta_f - \theta_i \approx \frac{N_s [\phi_f - \phi_i]}{N_G \phi_M}$$

where

θ = deflection in radians

N_s = turns in both search coils

N_G = turns in galvanometer coil

$\phi_f - \phi_i$ = flux change in search coils

ϕ_M = constant determined by the magnetic field
in the galvanometer gap.

This expression holds only in the highly overdamped situation when the mechanical damping and the restoring torque of the suspension fiber can be neglected in the equation of motion for the galvanometer. But it has the somewhat surprising result that the largest deflections are obtained for galvanometers with small $N_G \phi_M$. Therefore, small magnetizations were measured with a voltage sensitive galvanometer having small $N_G \phi_M$ and a sensitivity of $10^{-8} \frac{\text{V.}}{\text{mm.}}$ on a scale at 4 meters. It had a deflection of 1 mm. if the average magnetization per unit volume of the ellipsoid was about .002 gauss.

However, this instrument had a short period, which caused a quick decay from the final deflection and made the integration of long pulses somewhat inaccurate. This, along with thermal emfs., limited the accuracy of measurements made with the voltage sensitive galvanometer to about 1%.

For more precise measurements, the output of the search coils was switched to a charge sensitive galvanometer that had large $N_G \Phi_M$ and a long period. This reduced the sensitivity to $.3 \frac{\text{gauss}}{\text{mm.}}$, but it gave readings that were reproducible to $\pm .1\%$ for magnetizations over 300 gauss. This galvanometer was located at the center of curvature of a scale with radius 2 meters and an arc-length of two meters. From measurements on the mutual inductance of an air core transformer, the system was determined to be linear to $\pm .1\%$ for deflections from 1 to 2 meters. Average magnetizations over 600 gauss were measured by using shunts that were calibrated during the run.

Although the search coils were wound by a self-feed lathe that rather accurately determined the number of turns per unit length, it was difficult to calculate the deflection for a given magnetization because of the ellipsoidal shape and the close proximity of the two search coils. Instead, calibrations were made by assuming that the sample was a perfect conductor in the superconducting state at low fields.

Some justification for this assumption arises from the fact that for a given ellipsoid, the ratio $\frac{\Delta \text{ deflection}}{\Delta H \text{ ext.}}$ was a constant within the limits of the galvanometer accuracy as

long as $H_{\text{ext.}}$ had not exceeded $H_c(1-D)$. It was also found that the ratio $K_{300^\circ\text{K}} = \frac{\Delta \text{ deflection}}{\text{Vol}_{300^\circ\text{K}} \Delta H_{\text{ext.}}}$ was constant to within $\pm 2\%$ for all samples with $D \sim .06$ regardless of which of the three metals was used. If the volumes were corrected to 4.2°K by using the available coefficients of thermal expansion, then $K_{4.2^\circ\text{K}}$ was constant to within $\pm .5\%$ for all materials. This spread may have stemmed from improper alignment of the major axis or irregularities in the shape.

With the experimental arrangement shown Figures 7 and 8, the magnetization was measured as a function of temperature and external field. The temperature was adjusted by regulating the pressure of the liquid helium. This pressure was measured with a mercury manometer and was held constant with a Cartesian diver manostat. Nevertheless, the temperature of the sample could not be determined and regulated to the expected accuracy. Moving the sample produced temperature fluctuations such as local heating of the sample holder by friction. In addition raising or lowering the sample stirred the bath and probably agitated the liquid surface, thereby bringing the liquid in contact with warmer regions. The pressure regulator response was not quick enough to compensate for these fluctuations, and for temperatures between 4.2°K and the λ point of helium, there were pressure variations as large as 5 mm.

There was also a tendency for temperature gradients to form within the bath. This was particularly observable when

the temperature increased after the system was sealed off from the pump. As cold helium is more dense than warm, it was possible to have the liquid surface warm to 4.2°K while the sample was below 3.0°K . The manometer indicated the temperature of the liquid surface and the sample temperature was given approximately by a carbon resistor. This effect was minimized by taking measurements only while decreasing the temperature, but pressure measurements were probably no more significant than ± 5 mm. above the λ point and ± 2 mm. below.

To permit all trapped flux to escape, it was necessary to raise the sample temperature above T_c . This was done by removing the adjustable stops and rotating the knob until the two foot long rack raised the sample above the helium bath. In this way the sample could be warmed while the bath was under vacuum, and if the sample was slowly returned into the liquid, the pressure of the bath was not greatly changed during this procedure.

After the temperature was stabilized, the magnetization was measured as a function of external field. This field was supplied by the end corrected solenoid shown in Figure 7. The end corrections were calculated to give $.1\%$ uniformity of the field for the 8 cm. along the axis where the sample traveled. A search coil with the same diameter as the sample showed that the predicted uniformity was achieved. Although greater uniformity would have been possible with a longer solenoid, the length $\ell = 25$ cm. was chosen to reduce joule heating. The

power generated in a solenoid with fixed inner and outer diameters may be shown to be

$$P \propto H^2 l \rho \quad (3-1)$$

where ρ is the resistivity of the wire. As the solenoid was in liquid air, it was necessary to keep P small. Number ten copper wire was used to construct the solenoid, which had a ten layer body with six layer end corrections. When calibrated with a proton N.M.R. probe, it produced $56.45 \pm .1\% \frac{\text{Oer.}}{\text{amp.}}$ in the uniform region.

Although Eqn. 3-1 is independent of wire size, large wire had two advantages. First, it saved labor by reducing the number of turns and second it reduced the resistance. The latter advantage permitted the use of a low voltage supply that had been designed for a superconducting solenoid. Originally, the system was designed to use a superconducting solenoid; however, it was found that the $N\ell \approx 25\%$ wire in the solenoid trapped flux and had remnant fields that would have obscured some measurements.

Figure 9 shows the low voltage source consisting of two 4 volt, 700 amp. hr. batteries, which could be used in series or parallel. The current was determined by measuring the voltage drop across the standard resistor R_s , which was calibrated to $\pm .05\%$. The Leeds and Northrup K3 Potentiometer gave the voltage drop if the galvanometer was nulled.

Generally it was necessary to take measurements for

either increasing or decreasing current, and it was important not to go beyond the test point. In this case the potentiometer was set to the desired voltage, then R_v was adjusted to null the galvanometer. This adjustment was made much easier by using the current regulator, which is shown in Figure 10 and was driven with the output voltage of the Fluke 840A Galvanometer. This output voltage, which was proportional to the inbalance of the galvanometer, modulated the regulator's normal current of about .2 amp. Thus, when the output voltage was applied to the regulator, the current through the solenoid was changed to better agree with the current implied by the potentiometer setting. The regulator circuit reduced by about 90% the difference between the actual current and that implied by the potentiometer setting. Although this was important to save time and to prevent going beyond the test point, the major purpose of the regulator was to compensate for current drift, and it provided stability better than .05% for several minutes. This source was satisfactory for currents up to 15 or 20 amperes and could therefore be used for tests on tin or lead.

However, niobium required as much as 75 amperes, which was delivered by a Nobatron voltage regulated supply. This source was powered by the 440 v., 3 phase, 60 cps. line and could deliver up to 500 amperes at any voltage between 18 and 36 volts. The output voltage, which was regulated to about $\pm 1\%$, was substituted for the batteries in Figure 9, and

nichrome resistors were substituted for the manganin R_v . When the current regulator was used, d.c. drift was held below .1% for currents below about 25 amperes, but this limit was exceeded by some line fluctuations that were quicker than the response of the regulator. For higher currents, the regulation decreased primarily from heating of the solenoid, which increased the resistance from its lowest value of .16 ohms. Near 75 amp., the current could change as much as 2-3% during a reading.

The procedure during an experiment was generally as follows with some modification for tin whose critical temperature was below 4.2°K. First, helium was transferred with the sample in the earth's field. Then at 4.2°K the sensitive galvanometer was calibrated by assuming that small external fields did not change the average induction of the sample.

Next a series of readings were made by first applying some $+H_{ext} = +H_{max} < H_c(1-D)$ and measuring the magnetization at this field with the accurate galvanometer. The field was reduced to zero and the sensitive galvanometer was used to detect the residual magnetization $-4\pi M_{R+}$. Then $-H_{max}$ was applied and the magnetization was recorded at that field. Once again the field was reduced to zero and the residual magnetization $-4\pi M_{R-}$ was measured. The trapped flux was taken to be

$$-4\pi M_T = \frac{-4\pi M_{R+} + 4\pi M_{R-}}{2}$$

(3-2)

The magnetizations at $H_{\max}$ were found to be linear with field and reversible within the accuracy of the galvanometer as long as the sample had not entered the mixed or intermediate state. Therefore, these readings were used to calibrate the charge sensitive galvanometer.

If the sample was in the mixed or intermediate state at $H_{\max}$, then the charge sensitive galvanometer could detect hysteresis, and the magnetization was measured at many fields between $+H_{\max}$ and $-H_{\max}$. When $H_{\max}$ was large enough to make the sample normal, the magnetization was approximately zero except for a small paramagnetic moment that was probably due to impurities. The hysteresis of the magnetization curves was independent of $H_{\max}$ for $H_{\max} > H_{SN}$.

After the magnetization curve was determined for a maximum field that transformed the sample entirely into the normal state, the sample was raised above the liquid helium surface and warmed above T_c . This removed all trapped flux, so that a set of readings similar to those described above could be made at a lower temperature. During a run, five or six different temperatures ranging from 1.5° K to 4.2° K were studied. For each ellipsoid, the experiment lasted from ten to fifteen hours and required about two liters of helium.

IV. Results:

The results are arranged according to material, and the different specimens will be specified by giving the sample number from the table. On graphs, the quantity $-4\pi M$ will be plotted rather than the magnetization M . This is done for convenience as gaussian units are used with the convention described by Jackson³⁵⁾ that $4\pi M$ has the dimensions of gauss. In this convention $\mu_0 = 1$ in the relation $B = \mu_0 H + 4\pi M$ with the understanding that B and $4\pi M$ are measured in gauss and H in oersteds.

1. Tin

Previous work by Pippard⁸⁾ and Carroll³⁶⁾ used the symbol α to represent the percent of trapped flux where

$$\alpha = \frac{-4\pi M_T \times 100}{H_c}$$

They found that α increased at low temperatures and also very near to T_c . A similar behavior is seen in Figure 11 for Samples 1 and 2. The low temperature end of this curve is expanded in Figure 12.

The large amount of spread in the data stems from different methods of reducing the field. Circled points were made by quickly reducing $H_{ext.}$ to zero from a field greater than H_c . This gave fairly repeatable results, perhaps $\pm 10\%$, but it did not give the smallest residual magnetization. A smaller value was observed when the field was reduced slowly to zero by measuring the magnetization at many fields between H_c and

O. This may be related to the time variation of the magnetization in the intermediate state. If $H_{\text{ext.}}$ were reduced slightly, $-4\pi M$ did not immediately attain its new equilibrium value, but approached somewhat as voltage grows in a capacitor. The increase towards the new equilibrium sometimes lasted several minutes, and although it was accelerated by mechanically moving the sample, it occurred even if the sample were stationary. Thus $-4\pi M_T$ in zero field was smaller for these slow measurements, because they probably allowed the domains to more closely approach an equilibrium configuration in the intermediate state than was possible if H_{ext} was quickly shut off.

The greatest amounts of trapped flux were recorded if $H_{\text{ext.}}$ was abruptly reduced to zero from a field in the intermediate region. This type of measurement also had the least reproducibility.

The rise of $\mathcal{C}$ near T_c was also observed by Pippard and Carroll and was found to be quite sensitive to inhomogeneities. As is seen by comparing Samples 1 and 2, annealing had a much greater effect on this region than on the low temperature region. The high temperature behavior may be related to the sponge model. Assume there are regions with low T_c that act as normal inclusions in a superconducting matrix near T_c . At lower temperatures, these become superconducting and therefore expel flux because of their positive surface energy. This is seen in Figure 13, which shows the

gradual expulsion of the earth's field as the temperature was reduced. Notice that the magnetization reaches its constant value at the same temperature as the minimum in α . A measurement of this type was not possible in the work of Pippard or Carroll because their systems were designed to measure only residual fields.

Another observation made possible by measurement of the magnetization in an external field is the broadening of the magnetic transition near T_c . The long tail at high fields in Figure 14 is accompanied by large trapped flux. Figure 15 shows that the tail and trapping were reduced by annealing. Such broadening is expected for a sponge model in which T_c is not a constant throughout the material. Unfortunately, the large value of $\frac{dH_c}{dT} \approx 80 \frac{\text{Oe}}{\text{K}}$ near T_c made temperature fluctuations quite important and gave the large scatter of points shown in Figures 14 and 15.

The rise in α at low temperatures also seems to be fairly well predicted by a sponge model. In this region the sponge is assumed to be comprised of a fixed number of small filaments whose trapping is proportional to their critical current densities. Figure 16 shows that $-4\pi M_T[H_c(T)]$ has the field dependence given by Equation 2-26. In order to reduce the scatter from that of Figure 12, $-4\pi M_T$ was measured in a consistent manner. First, the external field was raised above $+H_c(0)$ and below $-H_c(0)$. This was necessary to remove all influence from trapping at previous temperatures. Then

$-4\pi M_T$ became reproducible and was measured for $H_{\max} \gg H_c(T)$ as described in the previous section. Because these data were all taken while the bath was slowly warming, H_c is the average of readings taken before and after the magnetization measurement. During the measurements, H_c did not vary more than 2 or 3%, which is comparable to the spread of $-4\pi M_T$ at constant temperature. However, the major drawback of the warming was that the temperature could not be measured with the manometer.

In order to measure H_c vs. T as shown in Figure 17, measurements had to be made while the bath temperature was constant after being reduced from a higher temperature. The dashed line shows that the low temperature data agree fairly well with the B.C.S.⁷⁾ theory, and the region near T_c was in general agreement with other papers. It can be seen that $H_c(T)$ does not strictly have a $\left[1 - \frac{T^2}{T_c^2}\right]$ dependence and therefore a small correction should be made to $\left[\frac{H_c(T)}{H_c(0)}\right]^{\frac{2}{3}}$, but the data spread in Figure 16 did not seem to merit this calculation. A plot of $\text{Log}(-4\pi M_T)$ vs. $\text{Log}(H_c)$ had slope 1.48 at lower temperatures with a deviation near T_c where transition broadening and a tail appeared.

$-4\pi M$ vs. H_{ext} is shown for Sample 3 at several temperatures in Figure 19. According to Kuper's¹⁴⁾ prediction, the magnitude of the slope S_2 in the intermediate region should be about a percent greater than twice the initial slope S_1 .

Both S_1 and S_2 were determined by a least squares fit of the lowest temperature curve in Figure 19 and they had standard deviations of $\pm .2\%$ and $\pm .4\%$ respectively. S_1 was more accurate because it was not influenced by temperature fluctuations. It was found that $S_2 = -2.028 S_1$, which agrees with predictions as well as can be expected for the somewhat irregular sphere. The small rise of $-4\pi M$ above the straight line near $H_{\text{ext}} = H_c(1-D)$ in the intermediate state may be associated with the horn Kuper predicts for this region, or it may be a result of a sponge structure.

The two different supercooling transitions on this same curve show the necessity of having $H_{\text{max}} \gg H_c$ before reducing H_{ext} when measuring supercooling. Apparently, the large field was necessary to quench filaments that served as nucleation centers. It was also important to lower the field slowly for $H_{\text{ext}} < H_c$ as sharp jumps also lead to premature spontaneous transitions. For this specimen supercooling disappeared near T_c when the magnetic transition broadening and tail appeared.

Another measurement on supercooling is shown by the oscilloscope patterns in Figure 20. These were made by applying the output of the two search coils to the vertical input and setting the horizontal on a sweep rate of 50 milliseconds per cm. The external field was reduced from a large value to a value near H_c . Here a picture was taken of the voltage vs. time when the field was reduced by ΔH

with the sample remaining normal. Then the external field was lowered to a value just above the supercooling field with the sample still normal. When the same change ΔH was made, there was a supercooling transition. The picture of the voltage during this transition was superimposed on the previous picture.

The peak that appears when there is supercooling may be related to the formation of a sheath as discussed by Faber¹⁵). His sheath proceeds at a rate that is limited by eddy currents. The time to reach the peak for small ΔH corresponds to the time for the sheath to travel, at the velocity measured by Faber, from a nucleation center at one end of the specimen to the other end. The larger ΔH probably induced several nucleation centers and therefore reached the peak earlier. In addition, this earlier peak may partly stem from the lower external field because Faber has predicted that the sheath velocity is proportional to $(H_c - H_{ext})^{3/2}$. He says the tail at long times is caused by flux migrating through the sheath. The galvanometer was more sensitive for measuring this tail and showed flux escaping as much as 20 sec. later.

Except for supercooling, the intermediate region near H_c was reversible for the single crystal studied in Figure 19. Other hysteresis in decreasing fields first appeared near $H_c(1-D)$. That this is not always true can be seen in Figure 21. Here hysteresis appeared at all fields, perhaps due to

grain boundaries. Annealing this sample reduced the trapping by about 30%, but as can be seen from Figures 22 and 23, it did not make the intermediate region reversible. These two figures show the nature of trapping near $H_{\max}$ for $(1-D)H_c < H_{\max} < H_c$. Figure 18 shows $-4\pi M_T$ after $H_{\text{ext.}}$ is reduced to zero from $H_{\max}$ as above. The x-axis on this graph gives the average internal induction at $H_{\max}$,

$$\beta_{\text{INT. AVE.}} = H_{\text{INT. AVE.}} + 4\pi M_{\text{AVE}} = H_{\text{EXT}} + 4\pi M_{\text{AVE}} (1-D), \quad (4-1)$$

which is a measure of the flux penetration at $H_{\max}$.

It seems significant that $-4\pi M_T$ grows quite quickly with the initial flux penetration and then tapers off. This suggests that in the intermediate state flux migrates through the sample and anchors first in the inhomogeneous regions with small $G_N - G_S$. When the field is reduced, these regions are unable to expel the flux. The peak in Figure 18 probably results from a form of quenching due to shutting off the field quickly. Near the peak, the flux trapped at each imperfection had to escape by its own action. With greater flux penetration, the flux moving from regions that did not have small $G_N - G_S$ may have provided escape routes for some of the flux trapped on inhomogeneities.

2. Lead

Figure 24 shows H_c vs. T for lead in the accessible temperature range. The region near T_c could not be measured but a linear extrapolation gives $T_c \sim 7.35^\circ \text{K}$. This is higher

than the generally accepted value of 7.2°K as is expected because of lead's positive deviation from the line $H_c(T) = H(0) \left[1 - \frac{T^2}{T_c^2} \right]$. An extrapolation to $T = 0^\circ\text{K}$ gives $H_c(0) \sim 810_{\text{Oer.}}$ in good agreement with other experiments.

Also shown on Figure 24 is the temperature dependence of $-4\pi M_T (H_{\text{max}} > H_c)$. The best linear approximations give "critical temperatures" for trapping that are below 6°K . Of course, one does not expect trapping to become zero at 6°K , therefore a graph of $\text{Log } [-4\pi M_T]$ vs. $\text{Log } H_c$ was made. This gave a straight line with slope $N = 1.92$. Figure 25 shows $-4\pi M_T$ vs. $\left[\frac{H_c(T)}{H_c(0)} \right]^{1.9}$ and suggests that if a sponge is responsible for trapping in this sample, then the critical current density of the filaments is proportional to $\left(1 - \frac{T^2}{T_c^2} \right)^N$. However, N is not 1.5 as predicted by Bardeen, but is approximately 1.9.

Meiklejohn's²⁸⁾ experiment suggests that this increase of N is due to impurities. Another factor may be strains, for a Log-Log plot of the two points for Sample 7 gave a larger N than Sample 8. The only difference between these two samples was the annealing and although it may have reduced the impurity concentration in the filaments of the sponge, it also relieved strains. At the present there are no studies of the effect of strains on N .

Figure 37 shows the magnetization curve for the lead sample that had the least hysteresis. This sample had been annealed and electropolished. A similar curve was made for the specimen after annealing, but before electropolishing,

and it showed three to four times as much hysteresis.

Figure 28 was made before the sample was annealed and shows considerable hysteresis.

This last figure seems to support the predictions of the sponge model. The long tail that extended to fields higher than $1.6 H_c$ is expected for every small filaments. As the tail is almost symmetric about the x-axis for decreasing fields, the filaments themselves have small magnetization. These filaments also retard the penetration of flux in the intermediate state and thus the best linear approximation to the intermediate region gave a critical field that was about 5% higher than that of the annealed sample. As the external field is decreased below H_c , these filaments reach their critical currents and allow flux to escape. The re-traces on Figure 28 suggest that those filaments that are active in the tail region may not contribute much to $-4\pi M_T$. $-4\pi M_T$ reaches its maximum while H_{max} is still in the intermediate region.

Perhaps $-4\pi M_T$ is determined by regions with low H_{SN} , which are the first to be penetrated in the intermediate region, and the tail is due to regions with high H_{SN} . Figure 34 shows the separation between the magnetization curve for increasing $H_{ext.}$ and that for decreasing $H_{ext.}$ This separation is a measure of the maximum hysteresis at a particular external field and it shows that the curve has the least reversibility near $H_c(1-D)$. Here one expects the greatest hysteresis be-

cause both high H_{SN} and low H_{SN} inhomogeneities are effective. At lower fields, the critical current of high H_{SN} filaments is exceeded, and at higher fields, low H_{SN} regions serve as the normal channels for the intermediate state.

The shape of the curve in Figure 34 depends not only upon the sample number but also on the temperature. Figure 29 shows the increase of trapping near H_c that occurs at low temperatures. At 4.22°K the region near H_c was almost reversible with no tail and a slight amount of supercooling. As the temperature was reduced, a tail appeared, the intermediate region became less reversible, and supercooling disappeared. This may be related to the increased stability at low temperatures predicted by Equation 2-24 for small filaments.

Supercooling seems to disappear when the high field tail appears. Figure 30 shows the supercooling transition for a sample with no tail. Sample 6 even had a small amount of supercooling at 1.8°K, thereby showing that supercooling disappears at low temperatures because of inhomogeneities and not as an inherent property of lead.

In Figure 30 the external field was reduced from point S to point B, and then cycled to give the small magnetization loop. The highly reversible nature of the region can be seen from the graph; however, a small hysteresis was present. Nevertheless, the slope predicted the demagnetization factor rather accurately.

Figure 31 shows the results for the lead sample with the largest amount of trapping. This curve was unchanged

after the sample was annealed for a week at room temperature. The large difference between this sample and Samples 4 and 7 is believed to stem from cold-rolling because the samples were treated the same in all other ways. Such cold working probably reduced the size of casting voids and also formed small grains which are more favorable to self annealing at room temperature.

By showing the magnetization for both $+H_{\text{ext}}$ and $-H_{\text{ext}}$, Figure 31 illustrates the symmetry that is observed for all samples. If the external field is reduced from some field far above H_c to one far below $-H_c$, the magnetization curve will form half of an envelope. These envelopes are different shapes for different samples, and the one for Sample 9 was entirely below the x-axis in Figure 31. When the external field is increased from far below $-H_c$, a similar envelope is formed, which can be superimposed on the first by reversing the signs of $-4\pi M$ and H_{ext} .

The top half of Figure 31 shows how a point $(-4\pi M, H_{\text{ext}})$ that is inside the envelope may be reached. Retraces that start from a point on the envelope where

$$-H_c(1-D) < H_{\text{EXT}} < +H_c(1-D)$$

are linear as long as H_{ext} is limited to this region. The line from the envelope at $H_{\text{ext}} = 0$ to the first point marked A illustrates this fact. This line parallels the line that starts at the origin for the virgin specimen. Similarly, the

points marked R were linear and reversible. However, the points marked B, which started from a point on the envelope with $H_{\text{ext}} < -H_c(1-D)$, were not linear. The intersection of lines A and B shows that interior points may be reached by several paths.

The complicated behavior of retraces starting from the intermediate state as shown in Figure 28 reflects the interplay between the trapping mechanism and the Meissner expulsion of flux. As the retrace begins, sponge filaments shield the interior from flux changes. For lower fields, the filaments reach their critical current and flux escapes.

These retraces show that $-4\pi M_T$ grows quite rapidly with H_{max} in the intermediate state. Figure 26 shows $-4\pi M_T$ vs. H_{max} for this sample and others. A significant feature is the small $-4\pi M_T$ that appears for fields well below the intermediate state. This trapping seems to obey the predictions of the model for the effects of surface irregularities. By comparing Samples 4 and 5, one sees that annealing had little effect on the low field region. The significant difference occurs in the intermediate state where flux penetration in Sample 4 was delayed by sponge currents. This was expected as annealing is a process that changes inhomogeneities but not the shape of the surface. The specimen was then electropolished. Sample 6 shows that this reduced $-4\pi M_T$ at low fields but did not change the field of the sharp break that indicated the start of the intermediate state. Also shown on Figure 26 is the trapping

of Sample 9, which apparently had a different distribution of surface irregularities and therefore had its own characteristic shape for $-4\pi M_T(H_{\max})$.

In addition to these changes with surface condition, the geometrical model predicts the temperature dependence of the trapping. Figures 32 and 33 show the temperature dependence of $-4\pi M_T(H_{\max})$, and also the transformation of the curves to $T = 0^\circ \text{K}$ by using Equations 2-18 and 2-19. In spite of the large scatter in Figure 32, lines can be drawn for each temperature; however, these points reduced to $T = 0^\circ \text{K}$ seem to have a random distribution about a single line. The data in Figure 33 were good enough to use Equation 2-21 to predict $T_c = 7.3 \pm 5\%$.

The success of the geometrical model suggested that it may be possible to predict the temperature dependence of a.c. losses at low fields. One attempt was made, but the background noise was so large as to obscure low field 60 c.p.s. a.c. losses.

3. Niobium

As niobium is a type II superconductor, it had a different behavior from tin and lead. One salient difference seen in Figures 36 and 37 is the long reversible tail that extends out to H_{c2} on the magnetization curve for a niobium sphere. Although tin and lead's magnetizations became zero at H_{cB} as calculated from Eqn. 2-9, niobium's long tail extended above H_{cB} . This is true even though too large a value of H_{cB} is obtained from the areas under the curves for in-

creasing fields in Figures 36 and 37. These large values of H_{cB} probably arise from some pinning mechanism as predicted by Campbell in Figure 5. Stromberg's³⁷⁾ sample which had much smaller hysteresis also had a smaller H_{cB} .

Additional support for the presence of pinning forces in the sphere comes from the fact that the peak magnetization occurs for an external field greater than $H_{c1}(1-D)$ where H_{c1} is taken from Stromberg's results. Instead, as expected from Figure 5, $H_{c1}(1-D)$ agrees with the field where flux penetration can first be detected and trapping begins.

Figure 38 shows the temperature dependence of some important features in Figures 36 and 37. H_{c2} is in rather good agreement with Stromberg's results, and although H_{c2} vs. T^2 can be linearly extrapolated to give a low T_c , it must be remembered that the temperature dependence of χ makes the curve nonlinear. The curve for H_{max} at $\frac{-4\pi M_T}{H_{max}} = 5 \times 10^{-6}$ gives a fairly good estimate of the temperature behavior of $H_{c1}(1-D)$; however, it is probably influenced by pinning mechanisms. T_c obtained from a linear extrapolation of this curve is somewhat high, and it is even higher if a larger value of $\frac{-4\pi M_T}{H_{max}}$ is used. This behavior reflects the increased capability of pinning centers to delay the penetration of flux at lower temperatures.

In addition, this delay affects the temperature dependence of the external field at the peak magnetization. Linear extrapolation of this curve gives a low T_c because pinning centers are

more effective at low temperatures, and therefore flux penetration is delayed to larger external fields. The temperature dependence of this magnetization peak strongly influences the calculation of $H_{cB}(T)$ and gives the large spurious H_{cB} obtained by integrating the area under the magnetization curve.

Because of these pinning centers, it is rather difficult to determine $\mathcal{K}$. Figure 39 shows the wide range of values obtained by different methods of calculation. Generally, $\mathcal{K}$ is determined from the equation

$$H_{ca} = \sqrt{2} \mathcal{K} H_{cB} ,$$

but according to theory the same value should be obtained by using Harden and Arp's³⁰⁾ numerical integration of Abrikosov's equations. However, the disagreement in the experimental results for the two methods seems to exceed experimental error. Stromberg's results give a similar lack of agreement.

There is a similar spread in the predictions for the temperature dependence of $\mathcal{K}$ as is shown in Figure 40. All predictions were normalized to agree at 4.22° K; nevertheless, no two curves coincide. Helfand's²⁹⁾ calculations do seem to take a middle of the road stand between the experimental curves, but no conclusions seem warranted.

Figure 41 shows $\frac{-4\pi M_T}{H_{max}}$ vs. H_{max} , which was used to find the approximate temperature dependence of H_{c1} in Figure 38. The trapping in this curve is not necessarily similar to that appearing in Figure 33 for lead with $H_{max} < H_c(1-D)$. The trapping in Figure 41 appeared above $H_{c1}(1-D)$ and probably

arises from pinning centers within the material; whereas, that of lead arises from surface irregularities.

Accompanying this trapping is a deviation from the linear relation between $H_{\max}$ and $-4\pi M(H_{\max})$. This nonlinearity above $H_{c1}(1-D)$ comes from the penetration of flux as given by Equation 4-1. Figure 42 shows how $\frac{-4\pi M_T}{B_p}$ is related to the flux penetration B_p . An important feature of this figure is that annealing reduced $\frac{-4\pi M_T}{B_p}$ for small B_p . As shown in the table, the annealing was performed in a good vacuum and should not have introduced impurities. If annealing had introduced impurities, these would have their greatest concentration near the surface and therefore would have increased the trapping at low B_p . This occurs because initial flux penetration is limited to regions near the surface in materials with pinning centers. Even though these pinning centers are weak and do not give much trapped flux at $H_{\text{ext}} = 0$, they probably are strong enough to limit initial flux penetration to regions near the surface as predicted by Bean²³⁾ for materials governed by the critical current density. The retrace that starts near H_{c1} for Sample 11 in Figure 36 traps almost all flux near H_{c1} and thereby seems to justify the assumption that pinning centers are quite effective near H_{c1} .

The reduction of the magnetization peak in Figures 36 and 37 also seems to justify the assumption that annealing did not introduce impurities. It is somewhat surprising therefore that $-4\pi M_T(H_{\max} > H_{c2})$ was increased considerably

after annealing. Figure 35 gives the temperature dependence of this trapping. The annealed sample seems to have $-4\pi M_T \propto e^{-\frac{E}{kT}}$ suggesting that the pinning strength may have a Boltzman distribution with temperature. Nevertheless, Sample 11 had a more complicated behavior and indicates the need of a stability condition similar to Eqn. 2-23 giving the strength of trapping center necessary for trapping at a given temperature.

The behavior of Sample 10 differs considerably from that of Samples 11 and 12. Sample 10 acts similarly to Campbell's³²⁾ prediction for large $a\beta$ given in Figure 6. Figure 44 has a high magnetization peak, and the outer envelope in Figure 46 follows Campbell's predictions for decreasing fields. Although this sample was a single crystal, it apparently had more strains than the sphere. X-ray analysis showed lattice distortions as large as 1% for Sample 10, but could not resolve any for the sphere. Also the impurity level was probably higher in Sample 10; however, no analysis was made.

Figures 44 and 45 show an interesting behavior that seems to be a result of strains. The curves labeled "before quench" were made by cooling the sample slowly to liquid air temperature, then transferring liquid helium into the dewar. Those labeled "after quench" were measured after the sample had been plunged into liquid helium directly from room temperature. This had a profound effect on the magnetization curve, for in

addition to increasing the peak magnetization, it seems to have increased H_{c2} .

In contrast, Figure 45 shows that the surface losses were reduced by this quench. This seems reasonable because the rapid quench probably created strains in the surface irregularities which delayed flux penetration. Apparently, these strains of quenching could be relieved at room temperature, because this same sample was warmed to room temperature and tested several weeks later by slowly cooling it to 4.2° K. The results were similar to those labeled "before quench". At the same time another quenching experiment was made, and it gave results similar to those labeled "after quench".

For the slow cool the trapping shown in Figure 45 probably does stem from surface irregularities. Figures 47 and 48 show how $-4\pi M_T$ varies with H_{max} and T and how the curves can be reduced to $T = 0^\circ K$ by using Equations 2-18 and 2-19. In addition, by using Equation 2-21 with all possible temperature combinations and several values of $\frac{-4\pi M_T}{H_{max}}$, T_c was found to be $9.2^\circ K \pm 2\%$. The measurements in Figure 47 were made for $H_{max} < H_{c1}(1-D)$, and it seems that these results as well as those for lead with $H_{max} < H_c(1-D)$ were probably due to surface irregularities. This value of $T_c = 9.2^\circ K$ seems to support Hecht's³¹⁾ measurements in suggesting that H_{c1} has a simple parabolic dependence on temperature.

Figure 46 gives a series of envelopes that were made for Sample 10 for various H_{max} . These were made for the slowly cooled sample and show the significant hysteresis that appears

above H_{c1} . Considerable flux remains trapped in these curves when H_{ext} is reduced from H_{max} to 0. Using Eqn. 4-1 to calculate B_p at H_{max} gives the data necessary for Figure 43. This curve shows that initial flux penetration has $\frac{-4\pi M_T}{B_p} \sim .3$; but for larger B_p , $\frac{-4\pi M_T}{B_p}$ becomes constant about .7. It remains at .7 $\pm 2\%$ until $-4\pi M$ at H_{max} is almost zero. This is greater than the constant value of .5 predicted by Bean's²³⁾ model for a cylinder with constant J_c ; however, his predictions for $J_c(B_p)$ do give a value which may be larger than .5 depending on the properties of the material.

The outer envelope in Figure 46 for $-H_{max} < H_{c2}$ is of considerable interest. Figure 49 shows the behavior of this envelope for increasing H_{ext} at several different temperatures. Reducing all points to $T = 0^\circ K$ with the transformation

$$-4\pi M(0) = \frac{-4\pi M(T)}{1 - \frac{T^2}{T_c^2}}$$

and

$$H_{ext}(0) = \frac{H_{ext}(T)}{1 - \frac{T^2}{T_c^2}}$$

gives the single curve shown in Figure 50. In this case it was necessary to use $T_c = 7.9^\circ K$. This T_c was determined by finding H_{ext} which gave constant $\frac{-4\pi M}{H_{ext}}$ and using an equation similar to Equation 2-21. As this is approximately the T_c given by H_{c2} in Figure 38, it seems that large amounts of trapping are governed by H_{c2} .

Another interesting feature of Figure 50 is that certain points have considerably smaller magnetizations than expected. This seems to arise from flux jumps. These jumps could explain why several points are low at $H_{\text{ext}} = 0$. H_{ext} was generally changed quite abruptly from $-500_{\text{Oer.}}$ to 0, and such quick changes of field while tracing the outer envelope seem favorable to flux jumps. Other field changes were limited by the time constant of the circuit to about .1 sec.

However, quick changes on retraces from the envelope did not induce flux jumps. As with lead in Figure 31, retraces from the envelopes in Figure 46 were linear and reversible and had a slope within 2% of that for the virgin curve below H_{c1} . This was true as long as $-H_{c1}(1-D) < H_{\text{ext}} < H_c(1-D)$. Thus, there was almost no flux change within the sample while it followed these retraces. But when the sample followed an envelope, there was flux change and it apparently led to flux jumps. These jumps seem to depend on the ability of the sample to dissipate the thermal energy associated with flux changes inside the sample.

V. Discussion:

Since considerable data are presented in the results, it is important to decide what new information is contributed. Some of the data merely reaffirm previous experiments. For example, these supercooling studies have not contributed much new information, but they do help substantiate Faber's¹⁵⁾ description of the phenomenon. The time dependence shown in Figure 20 agrees rather well with his predictions for the way that eddy currents limit the rate of formation of a superconducting sheath from a nucleation center. He has also discovered the effect shown in Figure 19 that raising the external field far above H_c decreases the supercooling transition field.

However, it was important that supercooling be measured in this experiment to relate it to other hysteresis mechanisms. As there had been speculation²⁰⁾ that lead may be a type II superconductor at low temperatures, one important measurement was merely to prove that supercooling existed for lead. The experiment showed that lead is a type I superconductor, but inhomogeneities may produce a high field tail that prevents supercooling at low temperatures. For tin a tail appeared at high temperatures and prevented supercooling near T_c . Although inhomogeneities can prevent supercooling, it seems that supercooling has little influence on other sources of hysteresis. Thus after the supercooling transition in Figures 19 and 30, the magnetization curve is quite reversible showing that there is no residual hysteresis associated with supercooling.

Perhaps the most significant contribution of this experiment is the study of trapping by surface irregularities. There do not seem to be any other d.c. studies of this type of trapping that stems from geometric effects. That the trapping did originate at the surface seems to be substantiated by the results of annealing and electropolishing shown in Figure 26. Moreover, the model proposed for geometric effects seems to predict the temperature dependence of the trapping quite successfully.

The hysteresis that is attributed to inhomogeneities has been subject to many studies, and in each the wide variety of factors involved make quantitative predictions difficult. By not requiring a detailed description of the inhomogeneities, the sponge and pinning models are fairly well suited to describing this type of hysteresis.

This article differs from most treatments of the sponge model in emphasizing the temperature dependence of the critical current density rather than the critical field and in proposing stability conditions for the temperature at which certain types of inhomogeneities become effective. Assuming that the trapped flux depends on the critical currents of the filaments gives

$$-4\pi M_T(\tau) \propto J_c(\tau) \propto [H_c(\tau)]^N$$

Figures 16 and 25 support this prediction for low temperatures, but near T_c stability conditions must also be considered. The

increase near T_c in Figure 11 apparently stems from inhomogeneities with low critical temperatures. These act as normal inclusions at high temperatures and thereby give regions with $\alpha_{BF} = 0$ in Equation 2-23. At lower temperatures these inclusions become superconducting and the stability condition changes because α_{BF} is no longer zero. The expulsion of flux in Figure 13 results from inclusions becoming superconducting.

The high temperature sponge formed by these normal inclusions has filaments that are much larger than the penetration depth. This can be seen from the reversible high field tail of Figure 14. As with Schoenberg's torus in Figure 1, the tail arises from filaments that are in the intermediate state. In contrast, the irreversible tail in Figure 29 arises from filaments with thickness smaller than λ . Equation 2-23 predicts the increased stability of these very small, high field filaments at low temperatures. These small filaments themselves have negligible magnetization, but their trapping is governed by their critical current density.

Most of the results for niobium seem to substantiate the predictions for the pinning model made by other authors. Perhaps one innovation of this article is the transformation of the data from Figure 49 into that of Figure 50. Since $T_c = 7.9^\circ\text{K}$ was necessary for this transformation, it seems that the magnetization envelope for strong trapping centers may have the same temperature dependence as H_{c2} . Therefore

the trapped flux at $H_{\text{ext}} = 0$ will have the same temperature dependence as H_{c2} if there are no flux jumps. Figure 44 shows that strains made the pinning mechanism even more effective and suggests that an important source of strong pinning centers may be strains.

Weak pinning centers seem to describe the behavior of the niobium sphere. The reversible tails in Figures 36 and 37 show that these pinning centers are mainly effective for fields in the regions $-H_{c1} < H_{\text{ext}} < H_{c1}$. As predicted in Figure 5 for weak pinning material, the peak magnetization is too large and occurs for $H_{\text{ext}} > H_{c1}(1-D)$. Previous experiments detected no changes at $H_{c1}(1-D)$; ³⁸⁾ however, the high precision of this experiment showed that flux penetration begins there and the high sensitivity indicated an abrupt rise in $-4\pi M_T$ for $H_{\text{max}} > H_{c1}(1-D)$. Figure 35 shows that $-4\pi M_T$ for $H_{\text{max}} > H_{c2}$ does not have a simple temperature dependence and suggests that weak pinning centers may have quite complicated stability conditions.

The high precision and sensitivity of this experiment were quite helpful for analyzing the results of annealing the niobium sphere. In this analysis it was necessary to use Bean's assumption ²³⁾ that the induction in the sample decreases linearly with distance from the surface. At the surface $B_0 = H_{\text{sur}}$ and flux penetrates a distance δ , which is approximated by the condition

$$\pi r^2 B_r \approx 2\pi r \delta \frac{B_0}{2}$$

where $\pi r^2 B_p$ comes from Equation 4-2 and approximates the flux penetration averaged over the entire sample. From this

$$\delta \sim \sim \frac{B_p}{B_0}$$

Thus the x-axis in Figure 42 also gives δ . If one takes $B_0 \sim 1400$ gauss and $r = .5$ cm., then

$$\delta \sim \frac{B_p}{1400} \text{ c. m.}$$

This shows that the crossover in Figure 42 corresponds to $\delta \sim .01$ cm., and it was found that $-\frac{4\pi M_T}{B_p}$ continued to change for $B_p > 200$ gauss for the annealed sample. Apparently, the unannealed sphere had rather uniform internal properties, but the annealed sphere had properties varying over distances as large as $\delta \sim .1$ cm. Therefore an explanation of the annealing process must explain two features. It must allow a variation of the superconducting properties for distances as large as .1 cm., and it must also give decreased trapping for small B_p but increased trapping for large B_p .

Impurity diffusion in niobium is great enough to give concentration gradients over distances as large as .1 cm. This has been shown by Gibala and Wert³⁹⁾ who studied the diffusion of carbon, nitrogen and oxygen. They give the r.m.s. migration distance d for radial diffusion as $d = \sqrt{6Dt}$ where t is the time that diffusion is permitted and D is the diffusion constant. D may be described by the relation

$$D = D_0 e^{-\frac{\Delta H}{kT}}$$

where T is the temperature, k is Boltzman's constant and ΔH and D_0 are constants that depend upon the type of impurity. Using their values of ΔH and D_0 for our sphere, which was annealed 4 hours at 1100°C , gives the following values:

Oxygen	$d \sim .3 \text{ cm.}$
Nitrogen	$d \sim .05 \text{ cm.}$
Carbon	$d \sim .05 \text{ cm.}$

It does not seem strange that this diffusion of impurities increased the trapping at large B_p . This may be related to the stability conditions that are needed to describe the temperature dependence of the effectiveness of pinning centers. Perhaps more effective pinning centers were formed by impurity atoms diffusing to collect in aggregates or at dislocations. Figure 35 seems to support this argument because its curves are more widely separated at 4.2°K where stability requires stronger pinning centers than at 1.5°K . These results suggest that impurities may be an important type of weak pinning center.

As with many experiments, this one seems to have raised more questions than it answered. One experiment that it suggests is an investigation of the behavior of ellipsoids in a.c. fields. By comparing the a.c. and d.c. characteristics of these samples, it may be possible to see if low level a.c. losses are governed by the predictions for surface irregularities. In such an experiment it would also be possible to see how quenching, electropolishing and etching affect a.c. losses.

Perhaps the apparatus of this experiment could be used if a coil to monitor a.c. signals were wrapped on the sample. Several trials were made with the search coils as sensors, but the background noise was too large. Signals were observed only in the intermediate region where flux changes in the sample seemed to be considerably delayed by eddy currents.

A second experiment that could be made with this apparatus is an investigation of the region near H_{c1} for alloys of type II superconductors. The high precision and sensitivity may help eliminate the indetermination generally associated with the increased magnetization peak that arises from pinning. Perhaps measurements of this nature may suggest an approach that reconciles the difference between theoretical and experimental values of χ and H_{c1} . In addition, by measuring $\frac{-4\pi M_T}{B_p}$ vs. B_p in this region, it may be possible to study the uniformity of alloy concentrations and perhaps even to develop a tool for studying impurity diffusion in metals. In conjunction with this, known impurities could be diffused into ellipsoids to determine why pinning centers become more effective at low temperatures.

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TABLE

Sample Number	Material	Shape	Comments
1	Tin	D = .06	Cold rolled, lathe cut, sanded.
2	"	"	#1 annealed one week at 200°C in vacuum.
3	"	Sphere	Single crystal, pockmark.
4	Lead	D = .06	Cold rolled, lathe cut, sanded.
5	"	"	#4 annealed one week at 300°C in vacuum.
6	"	"	#5 electropolished.
7	"	D ~ .06	Cold rolled, lathe cut, sanded, shape of Sample 10.
8	"	"	#7 annealed 3 months at room temp., electropolished.
9	"	D ~ .06	Cast cylinder, lathe cut, sanded.
10	Niobium	D ~ .06	Single crystal, surface strains shown by x-ray.
11	"	Sphere	Single crystal, spark cut, electropolished.
12	"	"	#11 annealed 4 hours at 1100°C in 8×10^{-8} Torr.

Ellipsoids with D ~ .06 had a major axis of 3 cm. and a minor axis of .635 cm. Spheres had radius .5 cm.

FIG. 3

FIG. 4

FIG. 5

FIG. 6

FIG. 7

FIG. 8

CURRENT CIRCUIT

FIG. 9

CURRENT REGULATOR

FIG. 10

FIG. 13

FIG. 14

FIG. 15

FIG. 16

FIG. 17

FIG. 18

FIG. 19

FIG. 20

FIG. 21

FIG. 22

FIG. 23

FIG. 24

FIG. 25

FIG. 26

FIG. 27

FIG. 28

FIG. 29

FIG. 30

FIG. 31

FIG. 32

FIG. 33

FIG. 34

FIG. 35

FIG. 36

FIG. 37

FIG. 38

FIG. 39

FIG. 40

SAMPLE 12

FIG. 41

FIG. 42

FIG. 43

FIG. 44

FIG. 45

FIG. 46

FIG. 47

FIG. 48

FIG. 49

FIG. 50