

Studies of irreversible magnetization in superconductors—a review

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Abstract. Magnetic measurements in the superconducting state of the high temperature superconductors have been characterized by the feature of irreversibility. Similar effects have been known in the conventional type II superconductors for about three decades now, and have been studied in great detail during the last few years. Recent studies of magnetic irreversibilities, in both conventional and high temperature superconductors, will be reviewed here. Thermally-activated relaxation accompanies such irreversibilities, and studies on flux-creep will also be reviewed.

This review shall cover the measurement of isothermal magnetization curves, of ac susceptibility, of thermo-magnetic history effects in the magnetization at a particular field and temperature, and of flux creep. An understanding of these in terms of Bean's celebrated macroscopic model shall be discussed. We shall also cover measurements that confirm the existence of weak links in ceramic high-temperature, as well as in conventional multifilamentary, superconductors.

Keywords. Superconductivity; magnetization; history effects.

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1. Introduction

The macroscopic magnetic properties of high temperature superconductors (HTSC) have been very extensively studied. During the first two years after the discovery of the HTSC, these studies produced many interesting features associated with the fact that the magnetization is irreversible over most of the magnetic field (H) and temperature (T) region of the superconducting phase. Many of these hysteretic features were considered to be novel and unconventional in the context of conventional superconductors (see the review by Malozemoff 1989). The ceramic nature of the HTSC, and the propensity of the initially discovered HTSC to form twin boundaries, were some contrasts with conventional superconductors that, together with the magnetic properties, led to new ideas like the "superconducting-glass model" (Muller *et al* 1987; Morgenstern *et al* 1988). Various groups (Schiedt *et al* 1988; Ravi Kumar and Chaddah 1988; Fraser *et al* 1989; Grover *et al* 1989), however, strove to find similarities between the HTSC and a sub-class of conventional superconductors, viz. the hard type II superconductors. The last two years have seen increasing success, and growing acceptability of the latter approach. It must be noted that the extensive effort in HTSC has resulted in experiments that were not earlier attempted in conventional hard superconductors, while the short coherence length in HTSC has made many more phenomena easily observable. The attempt towards a common understanding of conventional and high temperature superconductors provide the basis—and its successes provide the reason—for this review.

As is well known (see Chandrasekhar 1969), the discovery of Meissner effect in type I superconductors had established that the magnetization at any particular field and temperature was independent of the thermomagnetic history by which the sample was brought to that (H , T) point. Abrikosov's (1957) theory of type II superconductors assumed spatial uniformity in the superconductor and does not cover hysteretic effects

in the magnetic behaviour. This theory provides a basis for calculating the equilibrium or reversible magnetization of type II superconductors (see, e.g. Hao *et al* 1991).

The experimental measurements of magnetization have, however, always been complicated by irreversibilities. In type I superconductors, the magnetization of a FC sample (i.e. a sample cooled from above T_c to T in a constant H) is almost always found to be smaller in magnitude than that of a ZFC sample (i.e. a sample cooled in zero field to T and then subjected to H). No discrepancy was found in high purity Hg, and very little in Sn (Mendelssohn 1963). The observed discrepancies have been ascribed to impurities, and also to intermediate state effects (see Grover *et al* 1990, and references therein). Amongst type II superconductors, isothermal magnetization curves without hysteresis could be obtained for Nb (Finnemore *et al* 1966), by using high purity samples that were further outgassed in high vacuum. Amongst the HTSC, however, it has so far not been able to produce samples where magnetic irreversibilities are not observed. The hysteresis in the isothermal magnetization of $\text{YBa}_2\text{Cu}_3\text{O}_7$, the most studied HTSC, disappears only at temperatures $> 0.9 T_c$. Also, the low temperature FC magnetization (M_{FC}) is smaller in magnitude than the ZFC magnetization (M_{ZFC})—for fields larger than 1 mT. The experimentalist is thus unable to compare isothermal magnetization measurements of HTSC with calculations of equilibrium magnetization. The increased extent of irreversibilities as one goes from type I to conventional type II to HTSC may be related to the decreasing coherence length. We shall comment on this briefly in the next section.

Attempts to understand the hysteresis in isothermal magnetization began with the macroscopic model of Bean (1962, 1964). This model was relevant to "hard" superconductors that show a large hysteresis, and it is such hard superconductors that are used in the manufacture of superconducting magnets. Many of the detailed studies on Bean's model thus took place in the realm of applied physics, and this review shall also cover some of the work done in that area during the last two decades. The advent of HTSC brought out many new physical phenomena related to magnetic irreversibilities. The generation of harmonics in the magnetic response to an ac field is one such phenomenon. Peaks in the temperature dependence of the imaginary susceptibility (χ'') is another. Large history effects in the magnetization, and its temporal decay because of thermal activation, are some more. Detailed experimental studies of these phenomena resulted in Bean's original model being developed by many groups in the last few years. These developments shall be covered here.

In this review we shall not attempt to present a historical development of the subject. In the next section we shall discuss why the absence of spatial uniformity, and the presence of preferred sites for vortex "pinning", is necessary for a superconductor to carry large currents in the mixed state. We shall argue that this criterion for a large critical current density J_c necessarily results in hysteresis in the magnetization (though the converse may not necessarily be true). In § 3 we shall review some of the interesting experimental observations on magnetic irreversibilities in the HTSC. In § 4 we shall present Bean's macroscopic model and all extensions developed using its basic premise. In § 5 we shall review measurements on conventional hard superconductors. We shall point out similarities (and contrasts) with the HTSC data presented in § 3, and shall also compare with the theoretical understanding of § 4. We shall conclude by discussing possible further work, and the author's personal views on some limitations of the macroscopic model.

2. Magnetic irreversibilities and J_c

As mentioned earlier, Abrikosov (1957) first predicted the existence of the mixed state of type II superconductors. The magnetic flux penetrates the specimens in the form of flux-vortex lines, where each line carries a single flux quantum ϕ_0 . Abrikosov's calculation showed that the free energy is minimized if the vortices are arranged in a triangular lattice.

While two neighbouring flux lines repel each other, it is obvious that for any uniform flux density the repulsive interactions between a flux line and all its neighbours cancel out. But for a regular flux-line lattice with such a uniform flux density, $\text{Curl } \mathbf{B} = 0$ and it follows from Maxwell's equation that $\mathbf{J} = (1/\mu_0) \text{Curl } \mathbf{B} = 0$ and so no macroscopic currents can flow within the superconductor (and only surface currents are possible). We shall now examine what happens when one tries to pass current through the superconductor in its mixed state.

Let us consider that the superconductor is a rectangular slab that is infinite along the Y- and Z-axis, that the field is applied along the Z-axis and the current J flows along the Y-axis. Maxwell's equation $\text{Curl } \mathbf{B} = \mu_0 \mathbf{J}$ then requires a gradient in the flux density along the X-axis. It is clear that the repulsive interaction can no longer cancel out. Each vortex experiences a net force in the direction in which B decreases, and this force would be proportional to both the density of vortices and the gradient, i.e. $|\mathbf{F}| \propto B(dB/dx)$. This driving force can be derived rigorously based on a thermodynamic approach (Campbell and Evetts 1972) and one obtains

$$\mathbf{F} = \mathbf{B} \times \text{Curl } \mathbf{H} = \mathbf{B} \times \mathbf{J} \quad (1)$$

where the relation is exact only in the limit that the equilibrium magnetization can be ignored (Anderson and Kim 1964; Brechna 1973). This force on each vortex line, which is equivalent to a Lorentz force, will cause the vortices to move. The vortex motion will produce an electric field across the specimen, thus developing a resistance (known as the flux flow resistance). We have thus argued that the existence of a macroscopic current in the mixed state of a uniform superconductor would result in a resistance.

To prevent the vortices from moving and to allow a finite current to flow without any resistance (i.e. $J_c \neq 0$), the superconductors must "pin" the vortices with a force that is larger than the corresponding Lorentz force. This immediately requires some preferred sites for pinning the vortices, and the superconductor must no longer be uniform.

A reversible isothermal magnetization requires, according to Abrikosov's theory, that the vortices be arranged in a triangular lattice whose side increases (decreases) continuously as the applied field decreases (increases). It is clear that if there are some preferred sites where vortices are pinned—a necessary requirement for a non-zero J_c —then the vortex lattice cannot respond continuously to external field changes. Hysteresis in the isothermal magnetization curve is thus a necessary consequence of a non-zero J_c .

The flux pinning thus determines the J_c , and also keeps the superconductor from reaching thermodynamic equilibrium in its magnetic properties. Its calculation has been a subject of great interest, and such calculations are reviewed by Ullmaier (1975) and Huebner (1979) and are beyond the scope of this review. There are two features

of such calculations which should, however, be mentioned here. First, the pinning arises because of the existence of an impurity site of dimension $\sim \xi$. It is clear that the incidence of such inherent impurities will increase as ξ decreases, a qualitative trend that matches the increasing incidence of irreversibilities observed as mentioned in the introduction. Secondly, the pinning force is not a constant but increases with field at low field, passes through a maximum, and then decreases (as the field rises) at high fields. This is a universal behaviour that has been explained by Kramer (1973) in the following way. At low fields the vortex density is small and a large distortion of the triangular lattice (with concomitant increase of energy), is required if the vortices are to coincide with pinning sites. As the field increases, the distortion required decreases and the pinning force increases. At large fields when the number of vortices exceeds the number of pinning centres, the pinning force starts decreasing with increasing fields. It is further argued by Kramer that the pinning force must scale with B/B_{c2} . While various forms of the field dependence are given it has recently been argued (Yeshurun and Malozemoff 1988; Tinkham 1988) that the pinning potential at large fields (i.e. vortex spacing $\ll \lambda$) should scale as $H_c^2 \xi/B$. It is argued that even though H_c is large, the very small ξ in the HTSC makes the pinning very weak. The HTSC thus combine the features of high incidence of irreversibility with weak pinning, and these underlie the various experimental features to be discussed in the next section.

3. Experimental results on HTSC

In this section we shall view the data as broadly divided into two classes. In the first we shall consider studies in which the sample is cooled in zero-field to $T < T_c$, and magnetic studies are made isothermally at T . In the second we shall consider samples which are subjected to temperature variations at constant field (like field-cooled samples), and are subjected to various permutations of isothermal field changes and iso-field temperature changes. These give rise to interesting history effects which were first highlighted in the HTSC (Muller *et al* 1987), but were later understood as common to all hard superconductors (Ravi Kumar and Chaddah 1988). Magnetization decay experiments, in which both field and temperature are held constant during the actual measurement, have been studied under both conditions after the discovery of the high T_c , and will be discussed in detail. For completeness, we shall also mention some other measurements where thermomagnetic history effects have been observed.

3.1 ZFC samples studied at fixed temperature

3.1.1 *Hysteresis curves*: Isothermal hysteresis curves are the most commonly reported magnetic measurement. It entails cooling a sample in zero field to a temperature T , and then measuring the magnetization as the field is increased to H_{\max} . This yields the virgin magnetization curve. The field is lowered to $-H_{\max}$ and then increased again to H_{\max} and one obtains the hysteresis curve. The magnetization is measured at each field by one of the various dc methods (Faraday Balance, vibrating sample magnetometer, or SQUID magnetometer). It has been found that the magnetization measured at each applied field does depend (for the HTSC) on how long one waits (after changing the field and before measuring M), but we shall consider such effects

only in § 3.1.5. While the virgin curve can only be measured by one of the dc methods, the hysteresis curve (between H_{\max} and $-H_{\max}$) can easily be measured by applying an ac field $H(t) = H_{\max} \cos \omega t$, and monitoring the voltage induced in a secondary coil surrounding the sample. The ac technique has the advantage of improved signal-to-noise ratio through the use of phase-sensitive detection. In the ac technique, the measured hysteresis curve at a given temperature T is independent of whether the ac field is switched on after cooling the sample, or whether the sample is cooled below T_c with the ac field (of amplitude H_{\max}) kept on. The experimental parameters in the measurement of hysteresis curves are the temperature T and the field H_{\max} . The measured hysteresis curve do not show a perceptible frequency dependence up to a few hundred Hertz, and we shall consider these effects in § 3.1.5.

We first consider the virgin magnetization curve. Maletta *et al* (1987) made such measurements on sintered La-Sr-Cu-O. They found that the M - H curve is linear for $H < 0.1$ mT, and shows a continuous deviation from linearity (i.e. M/H is continuously decreasing as H increases) at higher fields (see figure 1). Senoussi *et al* (1987) observed similar behaviour in sintered pellets of Y-Ba-Cu-O, while the data of Grover *et al* (1988) shows that the field to which linearity appears to exist increases by more than an order of magnitude if the pellet is powdered. This thus indicated flux penetration into intergranular links in the sintered pellets at very low fields. In single crystal samples of YBaCuO, the deviation from linearity first occurred at much larger fields (values up to 0.5 T were reported by Dinger *et al* 1987). While this field value (for reasons that shall be discussed in § 4, this field value is not associated with the lower critical field H_{c1}) appeared to increase as the sample size was increased from a grain ($\sim 10 \mu\text{m}$) to a single crystal (~ 1 mm), this was true only for measurements with "conventional" accuracy. As was noted by Malozemoff (1989), the deviation from linearity is gradual, and the field-value appears to decrease with increasing experimental accuracy. The deviation from linearity is seen at fields varying from 10 Oe to 100 Oe in single crystals of Bi-Sr-Ca-Cu-O (2212) (Lin *et al* 1988) and in fields tending to zero in Tl-2223 (Wolfus *et al* 1989). We must note that the region of linearity of the virgin magnetization curve is also orientation dependent, and measurements in both single crystals and oriented powders yield the general result that the linearity extends to much larger fields when the applied field is along the C-axis.

Hysteresis curves are reported more frequently and the initial measurements as a

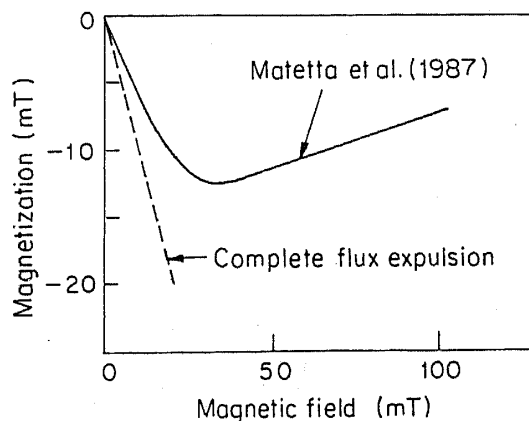


Figure 1. The measured magnetization curve is observed to deviate continuously from linearity.

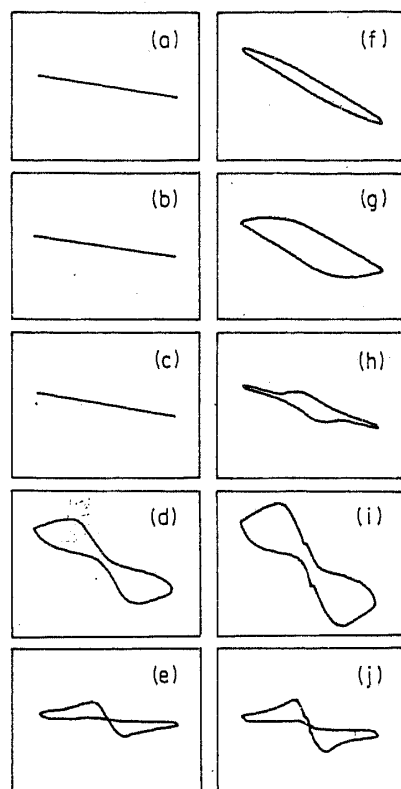


Figure 2. The magnetization hysteresis is shown at 77 K, as the field is cycled between H_{\max} and $-H_{\max}$, for various values of H_{\max} . Curves f to j are for a sintered pellet of $Y_{0.8}Dy_{0.2}Ba_2Cu_3O_7$, while curves a to e are obtained after powdering this pellet. The values of H_{\max} are 1 mT (a, f), 2 mT (b, g), 5.8 mT (c, h), 60 mT (d, i) and 150 mT (e, j). The vertical scale is arbitrary (Chaddah *et al* 1989a).

function of H_{\max} were reported by Senoussi *et al* (1987). For values of H_{\max} as low as 0.1 mT, a hysteresis appears in the sintered ceramic samples. This hysteresis increases as H_{\max} increases but, as depicted in figure 2, beyond a certain value of H_{\max} the hysteresis appears as a bubble which has closed and become reversible (within experimental accuracy) at high fields. At still higher H_{\max} , the hysteresis exists at all fields and the low field bubble evolves into a pair of kink-like anomalies. This is clearly depicted in figure 2. The low field bubble is attributed to hysteresis in the intergranular links and this is established (Grover *et al* 1988) by making measurements on the powdered pellet. As seen in figure 2, both the low-field bubble and the kinks disappear on powdering. The width of the low field bubble was found to depend on sample size and the intergranular J_c (Radhakrishnamurthy *et al* 1989), and the absence of hysteresis for H above a certain value was found to correlate with the decay of the intergranular J_c with increasing field (Mishra *et al* 1988).

The shape of the hysteresis loop depends on both H_{\max} and T , and we now show the change as a function of temperature. Detailed studies on single crystal of YBaCuO were made by Senoussi *et al* (1988). The behaviour of a powdered pellet is similar to that of an unaligned agglomeration of single crystals, and in figure 3 we show the results of Sarkissian *et al* (1989) on YBaCuO powder. The shape of the hysteresis curve undergoes a qualitative change above 10 K. It may be noted that the hysteresis curve at high temperature is confined to the second and fourth quadrants, unlike that

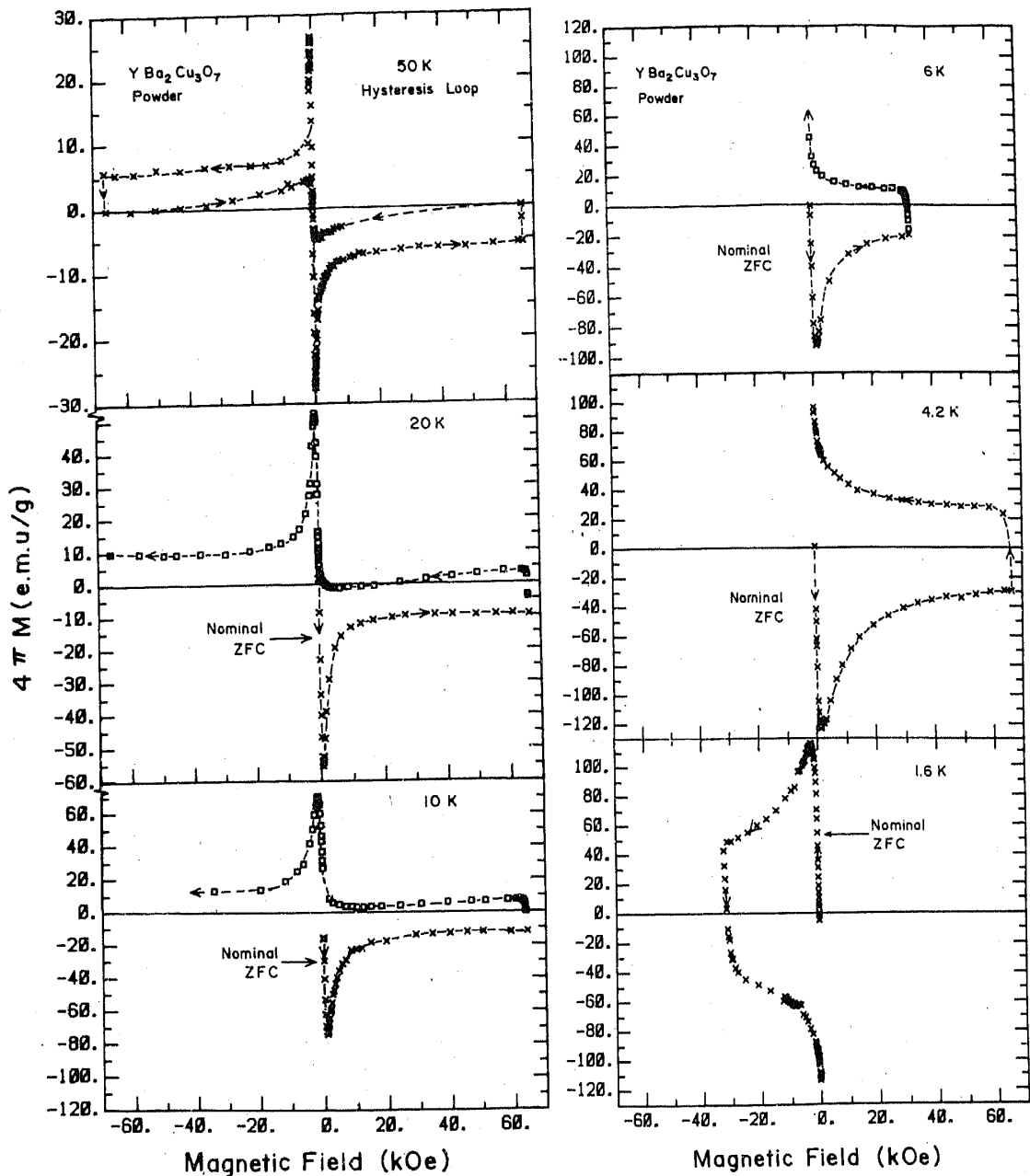


Figure 3. M vs H curves, as measured in YBaCuO powder by Sarkissian *et al* at different temperatures.

at low temperature. This qualitative change in the shape with temperature was first observed by Senoussi *et al* (1988) in single crystal YBaCuO . (A similar change in shape occurs, at 77 K, under particle irradiation or under special preparation conditions, and this will be discussed in §3.1.5). The small hysteresis seen at high temperatures is common to all HTSC, and is an indication of weak pinning in these materials.

We finally discuss the temperature evolution of hysteresis curves of sintered pellets. It was noted that the shape of the hysteresis curve evolves interestingly as one warms a pellet from 77 K to T_c . These changes in shape were attributed to the intergranular and intragranular responses, varying differently with temperature (Chaddah *et al*

1989a). Detailed studies of the temperature evaluation, for varying values of H_{\max} , have since been reported by Calzona *et al* (1989) and Shailendra Kumar *et al* (1990).

3.1.2. *AC loss and harmonic generation*: The existence of hysteresis in the magnetization curve implies an energy loss when the sample is placed in a cycling magnetic field. Similarly, the non-linear variation of M with applied field implies that the sample response to a field $H = H_{\text{ac}} \cos \omega t$ will contain higher harmonics and one can write in general that $M(t) = H_{\text{ac}} \Sigma [\chi'_n \cos n\omega t + \chi''_n \sin n\omega t]$. We shall review the data on power loss and on harmonic generation in this section.

The loss per cycle is simply the area contained within the hysteresis loop, and this loss manifests as an out-of-phase component in the ac response to a magnetic field. It is the latter feature that is used to measure the loss, and this is denoted by χ''_1 (or sometimes simply by χ''). The experimental technique for measuring the loss and for measuring the harmonics is thus identical. Similar to the case for measuring the ac hysteresis curve, the sample is exposed to an ac field $H(t) = H_{\max} \cos \omega t$. The voltage induced in the secondary coil is now fed to a lock-in, and the phase setting is done either in the paramagnetic state ($T > T_c$), or at low temperatures with H_{\max} kept so small that no hysteresis is detectable. The in-phase signal is then attributed to the real component of χ (denoted by χ') and the out-of-phase signal is related to χ'' , and thus to the ac loss. (We must mention here that it is sometimes assumed (Gotoh *et al* 1990) that $\chi'(H_{\text{ac}})$ gives the same result as a dc measurement of M/H with $H = H_{\text{ac}}$. This is not correct). In the same set-up, by having the reference signal at the appropriate harmonic $n\omega$, one can measure χ' and χ'' . We now discuss the highlights of such measurements.

χ'' has been measured as a function of temperature (for fixed H_{\max}) and this behaviour has been investigated for varying values of H_{\max} . In figure 4, we show a typical measurement of $\chi''(T)$ in YBaCuO pellet measured in $H_{\max} = 0.1 \text{ mT}$ (Grover and Chaddah 1991). The peak at higher temperature is due to losses in the intragrainar

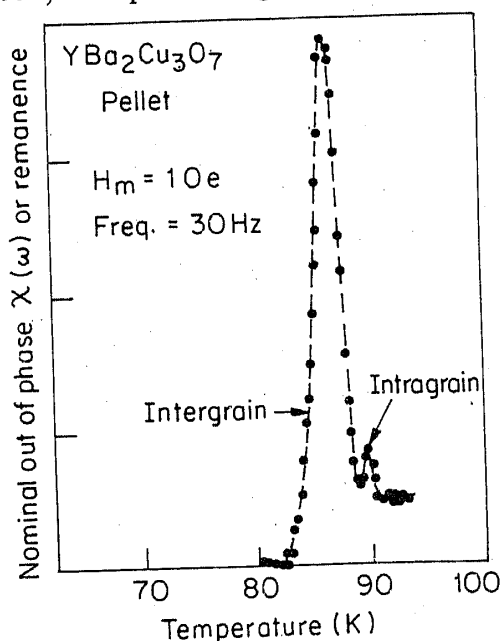


Figure 4. Temperature dependence of χ'' of YBaCuO pellet. The two peaks are identified with intergrain and with intragrain response. The intergrain peak disappears on powdering (Grover and Chaddah 1991).

region, while the lower temperature peak is due to losses in the intergranular region. This is easily verified by the disappearance of the low temperature peak on powdering the pellet, as was first pointed out by Goldfarb *et al* (1987). Single crystal samples also show only the higher temperature peak.

The variations with H_{\max} of the temperature at which the intergranular peak occurs has been studied by many authors (see, e.g. Calzona *et al* 1989; Muller and Pauza 1989; Ishida and Goldfarb 1990; Navarro *et al* 1990). Muller and Pauza have also studied $\chi''(T)$ when a variable dc field H_{dc} is superposed on $H_{\max} \cos wt$. One general feature of such studies is that as H_{\max} is increased, the intergranular peak shifts to lower temperature. A similar shift in the high-temperature peak is also seen (Malozemoff 1989), though such studies are limited because the fields required for observable temperature shifts are much larger.

The measurements of higher harmonics were reported by Muller *et al* (1989) who applied an ac field in the presence of a weak dc field. Ji *et al* (1989) presented detailed study of the generation of odd and even harmonics for varying H_{\max} and H_{dc} . They also emphasized that the existence of even harmonics implies a non-symmetric hysteresis loop and thus a field dependent J_c . We shall return to this point in §4. Navarro *et al* (1990), and Ishida and Goldfarb (1990) have studied χ' and χ'' as a function of temperature, for varying H_{\max} and H_{dc} , and these two are probably the most detailed studies of harmonic generation in YBaCuO pellets so far. All these studies are on sintered pellets since the generation of even harmonics from intergranular regions requires only small dc fields. Similar results should follow in powders and single crystal samples, though with much larger values of H_{\max} and H_{dc} .

3.1.3. RF shielding and microwave absorption: In this section we briefly discuss two experimental measurements that can monitor the flux penetration into a sample, in a way similar to the virgin magnetization curve. These two measurements have been frequently made on sintered samples, and thus correspond to flux penetration into the intergranular weak links.

In the first measurement one studies the magnetic shielding properties of a superconducting plate. An ac magnetic field, of variable amplitude, is applied through a primary coil positioned on one side of this plate, while a secondary coil positioned on the other side monitors the flux that is transmitted (Fiory *et al* 1988; Karthikeyan *et al* 1989). It has been observed that at low amplitude (~ 0.2 mT) of the applied field there is no measurable transmission. As the amplitude rises to about 0.5 mT, Karthikeyan *et al* (1989) observe a transmitted signal during that period of the cycle when the applied field is greater than 0.2 mT (see figure 5). This field value of 0.2 mT was observed to increase with the thickness as well as with the transport J_c of the superconducting plate. It thus appears (Karthikeyan *et al* 1990) that "perfect shielding" exists because of shielding currents set up in the intergranular region.

The frequency of the applied field in the shielding experiments can be varied from 100 Hz to 100 kHz (Karthikeyan *et al* 1989, 1990). While the transmitted waveform shows no change as the frequency is raised from 100 Hz to 10 kHz, a qualitative change occurs between 10 and 100 kHz, with the frequency at which the change occurs increasing with increasing transport J_c . This correlation indicates that at frequencies between 10 and 100 kHz the vortices get depinned and the magnetic response will now be qualitatively different from that in static fields.

At much higher frequencies (~ 1 GHz) microwave loss measurements look at the

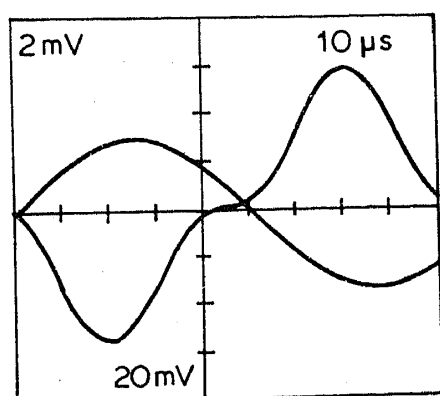


Figure 5. A typical waveform of the incident magnetic field (sinusoidal, 0.4 mT/div.) is shown, along with the distorted transmitted signal (Karthikeyan *et al*). Note that the transmitted signal is slightly phase-shifted.

magnetic response of a depinned vortex. The power in a commercial microwave cavity corresponds to the sample being exposed to magnetic fields of amplitude below 0.1 mT and no flux penetration is expected. When a dc field is superposed, a loss is observed because the dc field causes flux to penetrate in the intergranular region (see Ji *et al* 1990). It has been observed that this loss builds up when the dc field is raised beyond about 0.5 mT. It is at these field values that magnetization measurements show flux penetration in ceramic samples, and these microwave losses are also attributed to flux penetrating the intergranular weak links (see Sastry *et al* 1990a).

3.1.4 *Estimation of H_{c1} and J_c* : Most determinations of the lower critical field (H_{c1}) of HTSC have been made by measurements of the virgin magnetization curve of a ZFC sample. These measurements have assumed that the field at which the deviation from linearity is observed is H_{c1} . But this deviation is gradual and the onset is difficult to identify—and the reported value of H_{c1} appears to decrease with increasing experimental accuracy. As we shall argue in §4, there is reason to believe that the origin of the observed deviation from linearity may not be H_{c1} at all. We shall outline below some of the values quoted for H_{c1} , using this method.

The HTSC material most studied is YBaCuO. The deviation from linearity was observed, in single crystals of YBa₂Cu₃O₇ (with applied fields along the c-axis), at fields as varied as 0.8 T (Dinger *et al* 1987) and 0.1 T (Crabtree *et al* 1987). For a similar orientation, the deviation from linearity is seen at 10 mT in Bi₂Sr₂CaCu₂O₈ single crystals (Lin *et al* 1988) while deviation from linearity is observed in Tl₂Ba₂Ca₂Cu₃O₁₀ at fields below 0.5 mT (see Wolfus *et al*). While many researchers continue to quote H_{c1} values based on this deviation from linearity criterion, we have continuously emphasized the inapplicability of this criterion for HTSC (Chaddah 1987, 1988), and alternative methods of estimating H_{c1} are necessary (see Malozemoff 1989). While earlier attempts to estimate the deviation from linearity quoted $H_{c1}(0)$ values (for H parallel to ab plane) of about 20 mT in YBaCuO (Umezawa *et al* 1989), recent measurements (Umezawa *et al* 1990) have lowered this estimate to 7 mT. In crystals of the “248” compound of YBaCuO, Martinez *et al* (1990) have obtained an $H_{c1}(0)$ of 4 mT for the same field orientation. Martinez *et al* also vividly describe how the estimates of $H_{c1}(0)$ decrease as the measurement accuracy is improved.

The irreversibility observed in the hysteresis loop has been quoted most often to

estimate the critical current density J_c . The basis of this analysis is Bean's model (Bean 1964; Fietz and Webb 1969), which allows the determination of the field-dependent $J_c(B)$ by.

$$J_c(B) = k\Delta M/R$$

where ΔM is the hysteresis when the externally applied is increased or decreased about B/μ_0 . Here R is the dimension of the sample perpendicular to the field direction and k is a constant that depends on the geometry of the sample. This method of quoting J_c has become common because it is a contactless method that can be used on samples of arbitrarily small size. Amongst the initial successes of this method were the inferences (Finnemore *et al* 1987; Farrel *et al* 1987; Senoussi *et al* 1988) that $J_c(B)$ decreases sharply with increasing B . Secondly, it was observed that at large B , M does not scale linearly with sample size for sintered samples. This led to the conclusion (see § II B-3 of Malozemoff 1989) that these currents exist only within the grain and are blocked at grain boundaries. This view was strengthened by the observation of Oh *et al* (1987) that M varied linearly with the size of patterned thin films, as well as with thickness (for small size) in sintered samples (see Mizuno *et al* 1990).

Systematic attempts have been made to improve the J_c of the HTSC by varying the processing conditions or by introducing controlled number of defects into the bulk. In these cases the improvement in J_c is experimentally measured by an enhancement in the magnetization hysteresis. Jin *et al* (1989) have observed enhancements in ΔM (and thus in J_c) by a factor of ~ 400 at 1 T and 77 K by growing "melt-textured" samples instead of sintered ceramics. Murakami *et al* (1989) find that "Quench-melt growth" results in the bulk J_c (inferred from ΔM) rising to 1.5×10^4 A/cm² at 1 T and 77 K, a number slightly larger than that obtained by Jin *et al*. The same group (Murakami *et al* 1990) finds that single crystals grown with Y₂BaCuO₅ inclusion have much larger ΔM and yield $J_c > 3 \times 10^4$ A/cm² at 1 T and 77 K. Besides these reports, there are an enormous number of papers reporting magnetization hysteresis measurements as a step towards inferring J_c .

Neutron irradiation studies were found to increase the magnetization hysteresis for fast neutron fluences up to 10^{17} n/cm² (Umezawa *et al* 1987; Wisniewski *et al* 1988). Van Dover *et al* (1989) reported an inferred J_c of 6×10^5 A/cm² at 77 K and 0.9 T on irradiating YBaCuO single crystals with fast neutrons (7.9×10^{16} n/cm²). Van Dover *et al* (1990) and Civale *et al* (1990) have also reported a large hysteresis (and $J_c \sim 2 \times 10^5$ A/cm² at 77 K and 1 T) on irradiating YBaCuO crystal with 3 MeV protons to a fluence $> 10^{16}$ /cm². Amongst the more interesting inferences is drawn by Gyorgy *et al* (1990) who study the angular dependence of hysteresis to infer that, at least at low temperatures, twin boundaries do not play a significant role in pinning flux vortices.

3.1.5 Time-decay of magnetization: In an early paper after the discovery of HTSC, Muller *et al* (1987) reported a large decay in the magnitude of the ZFC magnetization of LaBaCuO ceramic. The decay was also noted to be non-exponential in time. These decays were extensively studied by Mota *et al* (1988 a, b, c) in sintered samples of HTSC, and they showed that the decay is logarithmic in time as

$$M(t) = M(t_0)[1 - b \log(t/t_0)]. \quad (2)$$

While similar logarithmic decays were observed (at fixed H and T) in samples prepared by various thermomagnetic histories, we shall concentrate first on ZFC samples. Yeshurun and Malozemoff (1988) first reported logarithmic time-decays in single crystals of YBaCuO, and large decays have since been established as intrinsic features common to all HTSC. We shall now discuss the salient features that have emerged from the extensive measurements on the time decay of ZFC magnetization.

As seen from (2), the decay rate $dM(t)/d \log t$ is proportional to $M(t_0)$, and this decay rate is denoted in literature by S . Since $M(t_0)$ differs from $M(t)$ only by a term proportional to b , one can also write $1/(M(t_0)) dM/d \log t$ as $[d \ln M/d \ln t]$. While the directly measured quantity is S , there has also been a lot of discussion regarding the derived quantity $(d \ln M/d \ln t)$.

Measurements of S during the virgin ZFC curves are made, at a fixed temperature, for various values of H . It is found that S increases with H as H^n for small H , where n is around 3 (Mota *et al* 1988a, b). S increases with H up to a certain field which we denote (for reasons to be discussed in §4) by H_1 , and then decreases with increasing H . The value of H_1 , at a given temperature, is different for different samples. In particular, it is much larger for single crystal samples (Yeshurun *et al* 1988) than for sintered pellets. The value of H_1 , for a given sample, increases as the temperature is decreased (Norling *et al* 1988; Shi *et al* 1990). It has been on experimental observation (see Grover and Chaddah 1991) that H_1 is larger than the field value at which M peaks in the virgin ZFC curve.

Mota *et al* (1988b, 1989) have also measured S for a ZFC sample which is subjected to an applied field H_m which is then reduced to zero. The decay rate is measured in the remanent state i.e. after the field has been lowered to zero. The remanent magnetization is positive but its decay again follows the form of (2). At low values of H_m , the value of S again varies as H_m^3 but is approximately one-fourth of that for the virgin ZFC sample exposed to H_m (Mota *et al* 1988b).

Rossel *et al* (1989) studied a ZFC sample of single crystal YBaCuO subjected to a field H at time $t = 0$, with the field being raised (suddenly) from H to $H + H'$ after t_w . The measurements of time-decay of magnetization were started at $t = t_w$, and S was found to show a discontinuity at approximately $t = 2t_w$. Kunchur *et al* (1990) made measurements on single crystals of BSCCO and found that the discontinuity occurred not at $2t_w$ but at $t_w + t'$. They showed that t' varies not just with t_w but also with H' and, further, with the temperature at which the ZFC sample was maintained. We consider the work of Kunchur *et al* as a complete work which unfolded the various relevant parameters and pointed to a proper understanding. We shall discuss their analysis in §4.

Measurements of the decay rate are being continuously used to estimate the flux-pinning potential U_0 (Hagen and Griessen 1989). As will be discussed in §4.8, the analyses used for such inferences is not fully established. The potential U_0 can also be obtained by measuring the resistivity (associated with flux creep) when a transport current $J \ll J_c$ is passed. Such attempts (Budhani *et al* 1990; Palstra *et al* 1990) yield values of U_0 which are much larger than those obtained by magnetization decay. A comparison of inferences from the two types of measurements is still the subject of discussion, and will not be attempted here.

Before concluding this discussion on flux creep, we must mention that the time scale on which flux creep manifests depends on the pinning strength. Thus intergranular

phenomena, as e.g. the shielding discussed in §3.1.3, show complications due to flux creep at frequencies of around 1 kHz. Nikolo and Goldfarb (1989) have observed that the peak in the ac loss (x_1'' of §3.1.2.) shifts slightly to higher temperature with increasing frequency. Similarly, Malozemoff (1989) argues that the peak in x_1'' in single crystals would also shift to higher temperatures, but the frequency range in which this would happen is ~ 1 MHz. It is the weaker pinning in HTSC that make complications related to creep so easily observable.

3.2 Thermomagnetic history effects

3.2.1. *Irreversibility temperature:* The existence of hysteresis, or any other form of irreversibility, in the measured magnetization implies that the value of M at a given field and temperature (H, T) will not be a function of H, T alone but will depend on the path of reaching this (H, T) point. Further, the measured value will generally not be the equilibrium value. In §3.1 we considered samples which were cooled in zero field to T_1 and then subject to various changes in H with T held fixed. In this section we consider experiments in which the sample is also subjected to temperature variations in a field. The most common example of the existence of such a history effect is the difference in the magnetization of FC and ZFC samples at the same H, T values. As depicted in figure 6, M_{FC} and M_{ZFC} are not equal in sintered HTSC over almost the entire range of temperature even in fields as low as 1 mT. Measurements in single crystal samples show similar disagreement between M_{FC} and M_{ZFC} except at temperatures close to T_c . The temperature above which M_{FC} and M_{ZFC} have no measurable difference has been referred to as the "irreversibility temperature" T_r . (We also note that the sample appears to be reversible for $T > T_r$.) One interesting feature first observed (Yeshurun and Malozemoff 1988) in YBaCuO crystals is that T_r decreases with increasing field and the following power law appears to hold for all

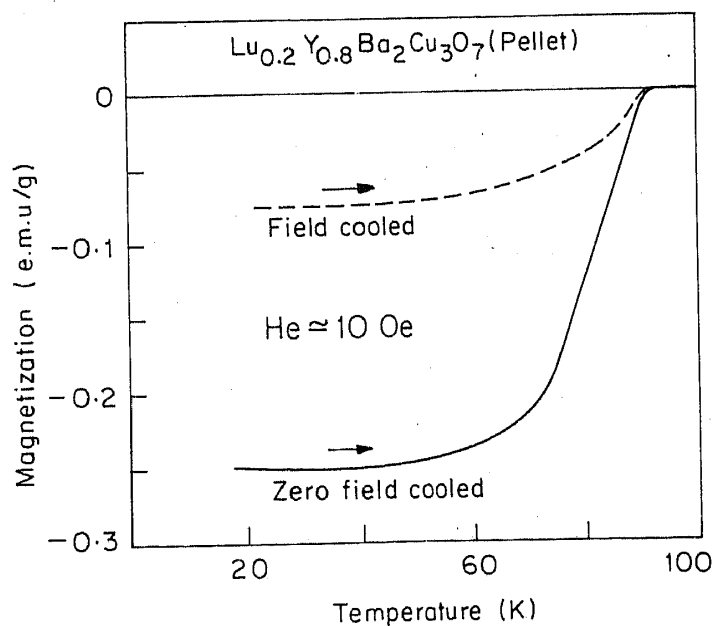


Figure 6. Temperature variation of FC and ZFC magnetization. The arrows indicate that both data were taken during warm-up (Grover *et al* 1988).

HTSC samples (Xu *et al* 1990).

$$(1 - T_r/T_c) = aH^q. \quad (3)$$

The above equation holds with T_c being the transition temperature in zero field. While the value of q in various reports varies in the range of 0.5 to 0.75 (Malozemoff 1990), Xu *et al* (1990) have noted that it is higher for thin film samples than for bulk samples. A second qualitative feature is that “ a ” is much larger for Bi- and Tl-based HTSC than for YBaCuO, indicating that irreversibilities set in at much higher temperature (~ 80 K in 2 T) in YBaCuO than in BSCCO (~ 30 K in 2 T). We shall now review some of the current discussions regarding T_r .

Xu *et al* (1990) have observed that the measured value of T_r is dependent on the size of the specimen as well as on the rate at which the magnetic field or the temperature is changed during the measurement process. T_r in YBaCuO is observed (Xu and Suenaga 1991) to increase by 1 to 2 K (which is 10 to 20% of $T_c - T_r$) when the rate of temperature change is increased. This latter feature appears to explain the discrepancy (Kritscha *et al* 1990) in T_r measured by ac and dc magnetization. The onset of irreversibility in transport and magnetomechanical effects have also been used to determine T_r (Malozemoff 1990), but the actual value of T_r determined by the various techniques differs by a few degrees in YBaCuO.

One also expects that hysteresis in the isothermal magnetization curve should also appear only at T_r . T_r is a function of H , however, and to determine $T_r(H)$ from isothermal hysteresis curve one has to make a measurement at T and observe the field H , above which ΔM vanishes. Such information can then be inverted to obtain $T_r(H)$. These measurements are tedious and it has been more common to measure the hysteresis curve at $T \ll T_r$ and then correlated $\Delta M(H)$ at T with $(T_c - T_r(H))$ determined from the temperature dependence of $M_{FC}(H)$ and $M_{ZFC}(H)$. As discussed in § 3.1.4, changes in the hysteresis curve have been observed under particle irradiation. Kritscha *et al* (1990) observed an inverse correlation between $\Delta M(H)$ and $(T_c - T_r(H))$ in neutron-irradiated crystals of BSCCO. Civale *et al* (1990) studied proton-irradiated crystals of YBaCuO and observed large changes in $\Delta M(H)$ as a function of proton fluence, but noted that $T_r(H)$ are “hardly changed”. Xu and Suenaga (1991) have noted, however, that if the data of Civale *et al* is plotted on log–log plots to check eq. (3), a consistent decrease of “ a ” with increasing ΔM is observed. This would be consistent, qualitatively, with the results of Kritscha *et al*, and with the idea that isothermal irreversibility (i.e. ΔM) and iso-field irreversibility have a common origin. Xu and Suenaga have further substituted Cu with $M = \text{Ni, Zn, Al and Fe}$ to study $\text{YBa}_2(\text{Cu}_{0.98}\text{M}_{0.02})_3\text{O}_7$. They observe changes in $\Delta M(H)$ and $(T_c - T_r(H))$, and find that the constant “ a ” decreases as ΔM increases, with eq. (3) being satisfied in all cases. To conclude this discussion, such studies clearly show the common origin of magnetic irreversibilities in isothermal and isofield cases, besides providing detailed data to test various theories of magnetic irreversibility.

3.2.2. *Thermal cycling in constant field:* Muller *et al* (1987) measured the magnetization of a ZFC sample of La(Ba)CuO subjected to a field of 30 mT at 4.2 K. The sample was then heated to $T_1 < T_c$, cooled back to 4.2 K, heated to $T_2 > T_1$, cooled back to 4.2 K and so on. All these thermal cyclings were made in a constant field, and the susceptibility (and magnetization) showed an interesting multi-valued behaviour

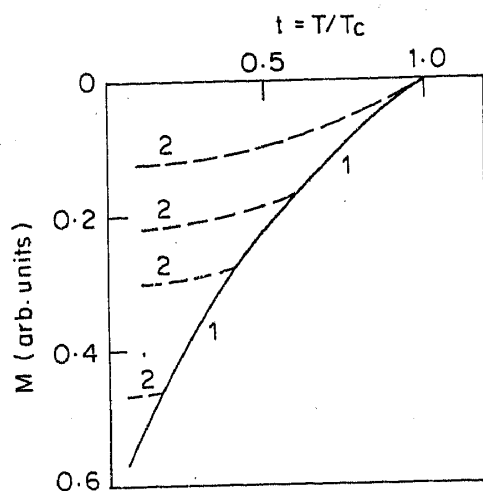


Figure 7. We show a schematic of M values obtained for a ZFC sample, subjected to a constant field H , as the temperature is cycled. Curve 1 is the ZFC envelope to the left of which lie all the M values that can be obtained. Curves 2 are obtained by cooling the sample during ZFC warm-up. The dashed lines indicate that these excursions from curve 1 are reversible.

schematically depicted in figure 7. The curve labelled by 1 is an enveloping curve to the left of which all accessible values of $M(T)$ lie for this particular H value. This is the curve which is traced if the sample is monotonically warmed from 4.2 K to T_c . If the sample is cooled at any intermediate temperature, then $M(T)$ does not retrace curve 1, but follows curve 2. Muller *et al* found that curve 2 was reversible under thermal cycling, while curve 1 was not. Various curves of the form of curve 2 are connected only via the curve 1. Schiedt *et al* (1988) observed similar behaviour in YBaCuO, and also in conventional superconductors. As we shall see in § 5, such a behaviour of M under iso-field temperature cyclings has now been established as a common feature of all hard superconductors.

3.2.3. Isothermal field changes on FC samples: The simplest example of this class of history effects is to cool a sample in a field H to a temperature T , and then isothermally reduce the field to zero. The sample develops a positive moment which is denoted by $M_{\text{rem}}(H, T)$. Malozemoff *et al* (1988) measured the T -dependence of M_{rem} for YBaCuO single crystal samples cooled in a field of 6 mT. They observed that at 4.2 K $M_{\text{rem}} = M_{\text{FC}} - M_{\text{ZFC}}$. Further, they noticed that this equality appears to persist for various values of field. This correlation between magnetization measurements in field-on and field-off cases provides a significant clue to the reason for history effects. We noted that M_{rem} is a special case of isothermal field changes after field-cooling, and these thermomagnetic histories should also be studied in detail (Grover *et al* 1989a, b). Our detailed measurements on conventional hard superconductors will be discussed in § 5—here we outline the results obtained by Sarkissian *et al* (1989) in YBaCuO powders. They cooled YBaCuO powder to 4.2 K in fields ranging from 0.15 T to 5.77 T. The sample showed some field-cooled magnetization which decreased with increasing field in this field range. After field-cooling, the field was decreased to zero in each case and the resulting M values are shown in figure 8 along with the ZFC hysteresis loop. The initial response of the sample in each case is to exclude the external field change, and the slope of $M-H$ for small field decrements is the same

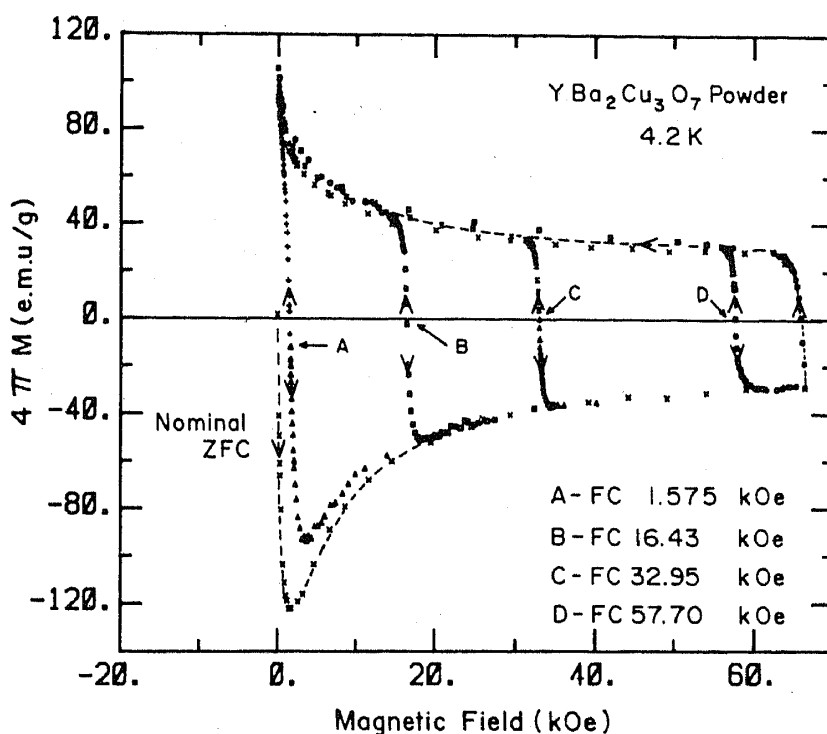


Figure 8. Forward and reverse isothermal $M(H)$ curves in YBaCuO powder (Sarkissian *et al*). For each of the runs the sample was cooled from 100 K to 4.2 K in the fields indicated, and the points marked A, B, C, D are the values of M_{FC} . The arrows indicate that both forward and reverse curves were separately measured for each field value.

as that of the virgin ZFC curve. This also explains the observation of Malozemoff *et al* (1988) since the fields in which they cooled the sample were comparatively small, and the "initial response" region continued till the field was reduced to zero. For large field decrements, the increase in M tapers off and the magnetization curve merges with the hysteresis curve of the virgin ZFC sample.

Sarkissian *et al* (1989) also subjected the field-cooled samples to isothermal field increase, and the initial $M-H$ curve again had the same slope as the virgin ZFC curve. The decrease in M again tapered off and the magnetization curve merged with the virgin ZFC curve—with the merger continuing into the hysteresis cycle. Sarkissian *et al* thus generalized the measurements of Malozemoff *et al* to cover various isothermal field variations after field cooling. They also reproduced many of the features observed in niobium by Grover *et al* (1989)—measurements that we will discuss in § 5.

3.2.4. History effects in related measurements: We discuss now some recent results of measurements of microwave surface resistance and of transport critical current density, in sintered polycrystalline samples. In both cases one is measuring the response of the intergranular region, and this response is determined by the local magnetic field in this region. Hysteresis in the microwave absorption (see Sastry *et al* 1990b and references therein) and in the transport J_c (see Majoros *et al* 1990 and references therein) have been noted as the applied magnetic field is cycled isothermally. The surface resistance has recently been found to be lower in a FC sample than that in a ZFC sample exposed to the same magnetic field (Ji *et al* 1990). The transport J_c in

a FC sample has also been found to be higher than in a ZFC sample in the same field (Mishra *et al* 1990). Both these groups, studying ceramic samples, have explained their results by arguing that the intergranular region sees a higher effective field in the ZFC case. We must stress that the history effect in this case does not have the same origin as the history effects discussed earlier. We believe that the sintered pellet is a two component (inter- and intra-grain) system, and these two components have different J_c and pinning strengths. It is this inhomogeneity of the sintered pellet that causes the history effects in microwave surface resistance and in transport J_c .

4. Macroscopic model of hard superconductors

In this section we shall review the various theoretical attempts at explaining, and at predicting, the experimental results in hard superconductors. While a large fraction of these theoretical studies were performed in the last few years, they are extensions and modifications of what was developed in the 1960's as the critical state model. Those early developments were reviewed by Campbell and Evetts (1972).

The idea of the critical state model was developed by Bean (1962, 1964) and, though some matters of detail were modified over the years, this macroscopic model is commonly referred to as Bean's model. The success of this model, as will be discussed in what follows, indicates that this macroscopic model appears to have captured the essential physics of hard superconductors. The present author's views regarding restrictions on its validity will be relegated to §6.

We must stress here that many theoretical developments have taken place in attempts to understand microscopically the pinning and depinning of vortices. The elastic constants of the vortex lattice, in particular the shear modulus and the tilt modulus, have been obtained (see Brandt 1990a, and references therein) in terms of microscopic parameters for both isotropic and anisotropic superconductors. Brandt (1990b) has calculated the critical shear stress, and the resulting J_c turns out to be much smaller than the measured J_c in YBaCuO. He thus argues that direct pinning of each vortex, by many pins along its length, is the dominant pinning mechanism in HTSC. Nelson (1988) has suggested that the vortex lattice melts when the thermal fluctuation of the vortex positions becomes comparable to the vortex spacing. Collective pinning of the vortices due to random pinning sites has been argued to give rise to a vortex glass phase (Fisher 1989). Recent scaling theories of collective pinning (Fisher *et al* 1991; Fiegel'man *et al* 1989) discuss thermally activated dissipation and yield a current dependence which predicts zero resistivity as the current density $J \rightarrow 0$. Detailed predictions of the J -dependent voltage, and of the magnetization decay rate, have been made by Fisher *et al* (1991). The various microscopic calculations discussed above offer differing explanations (Brandt 1990a, b) for the "irreversibility line", and their detailed predictions are still to be verified (or contradicted) convincingly by experiments. A detailed review of such calculations may thus be premature, and we now restrict to discussing the macroscopic model.

4.1. Bean's model

The version of Bean's model in common usage is at slight variance from the initial model of Bean (1962). It neglects the lower critical field H_{c1} as small. While this

approximation was made early (Bean 1964), it was then done in the spirit that the sample is exposed to a field $H \gg H_{c1}$ and the equilibrium magnetization is negligible. Attempts to incorporate H_{c1} (or the equilibrium magnetization) in such calculations have been made for conventional superconductors in the past (Bean 1962; Fietz *et al* 1964; Kes *et al* 1973; Clem 1979) and for the HTSC recently (Ravi Kumar and Chaddah 1988; Yeshurun *et al* 1988; Krusin-Elbaum *et al* 1990), but there is no agreement on how this should be incorporated. We shall now discuss the version of Bean's model in common usage (Ravi Kumar and Chaddah 1989; Chen *et al* 1990a, b; Yeshurun *et al* 1990; M Xu *et al* 1990) in which H_{c1} is simply ignored and put equal to zero. Questions about this version will be raised only in § 6.

Bean's model predicts the response of a hard superconductor, with a critical current J_c , to an isothermal variation of the external magnetic field as "any change in magnetic induction, *howsoever small*, felt by any region of the sample, will induce the full critical current density J_c to flow locally". The direction of J_c will depend, through Lenz's law, on the direction of the e.m.f. that accompanied the last local change in magnetic induction. Only three states of shielding current flow are possible with a given axis of the magnetic field: zero current in those regions that have never felt any isothermal change in B , and $\pm J_c \hat{u}$ where \hat{u} is perpendicular to the field axis. It is to be noted that this formulation includes the possibility that J_c is a function of B (Bean 1964).

4.2. Samples with zero demagnetization factor

Campbell and Evetts (1972) provided a method, for obtaining the virgin and hysteresis magnetization curve within Bean's model, for a cylindrical sample of arbitrary cross-section but of infinite length along the applied field direction. Such samples have a zero demagnetization factor. Bean's model gives the magnitude and direction of the shielding currents and the field profile is then obtained by substituting into Maxwell's equation to write

$$|\text{Curl } \mathbf{B}| = \mu_0 J_c$$

As argued by Campbell and Evetts, this equation is valid for an arbitrary cross-section, and can be written as

$$\left(\frac{dB}{dr}\right)_n = \mu_0 J_c(B) \quad (4)$$

where $(dB/dr)_n$ is the gradient in B along any normal to lines of constant B . These surfaces formed by the "lines of constant B " are uniquely specified by the cross-section and are independent of the form of $J_c(B)$. (The sample-surface is one such surface). The current flows along the lines of constant B , in a plane perpendicular to the infinite axis. When a ZFC sample is subjected to an increasing external field, the field penetrates only near the surface and the interior region, having $|\mathbf{B}| = 0$, is contained within a surface called the flux-front. This flux-front always coincides with one of the constant B surfaces, and moves inwards continuously as the external field is increased. The movement from one constant B surface to the next is along the normal to the surface at each point, and it follows that there will be discontinuities in the curvature of the surface once any centre of curvature is reached. It also follows from eq. (4) that the distance, from all points on a constant B surface, to the corresponding points

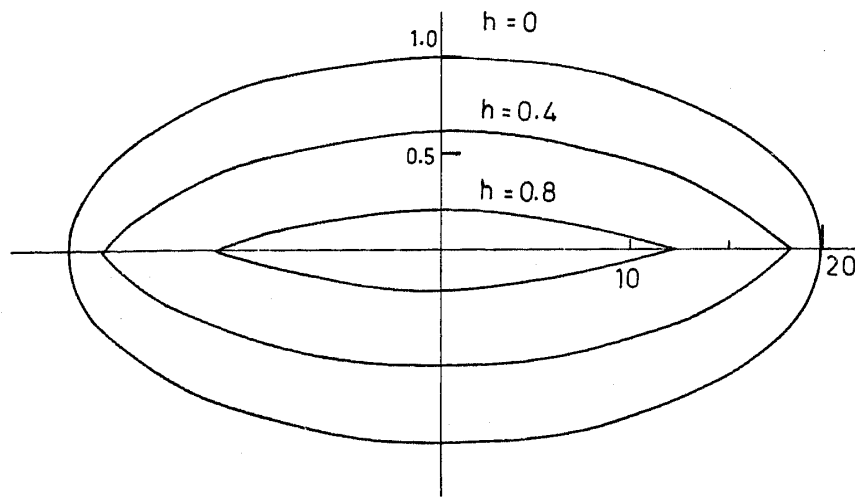


Figure 9. We show iso-field contours for an elliptical cylinder with major/minor axis ratio of $a/b = 20$. (Note the scale on the figure.) A centre of curvature is reached for $h > 0.05$, and the contours then have a discontinuous derivative on the major axis.

on the sample surface, is constant when measured along the normals specified above. The shape of these constant B surfaces, for a cylinder of elliptical cross-section, are schematically depicted in figure 9.

While eq. (4) is a first order partial differential equation, it is in principle integrable for arbitrary forms of $J_c(B)$. The shapes of the constant B surfaces are dictated only by the cross-section of the cylinder, and one can solve (4) for the virgin magnetization curves for various values of the applied field. The solution tells us which constant B surface the flux-front coincides with for a given applied field—and this constant B surface differs for different forms of $J_c(B)$. With this procedure it is also possible to obtain the hysteresis curves for arbitrary, but isothermal, variations of the applied field. We first review the calculations of hysteresis curves for various cross-sections with the assumption that J_c is independent of B . We then discuss its generalization for the case of rectangular cross-sections when the J_c along the two perpendicular directions (normal to the field direction) are not equal. We then go back to the case where J_c is independent of direction but depends on B . Finally, we comment on the possibility of incorporating the B -dependence of J_c and its angle dependence.

4.2.1 Hysteresis curves for constant J_c : Virgin and hysteresis curve calculations for this case were presented by Bean (1964) when the cylinder has a circular cross-section, and also when its cross-section is a rectangle, but with one dimension tending to infinity (this is, for obvious geometrical reasons, also referred to as the “slab” geometry). Campbell and Evetts considered the case of a finite rectangle, and, as discussed in §4.1, provided a method for arbitrary cross-sections. They provided results for an elliptic cross-section, but these are only for applied fields small enough that a centre-of-curvature is not reached. Bhagwat and Chaddah (1991) have presented solutions for the elliptical case for arbitrary values of applied field and have also provided solutions of the hysteresis curve. They solve instead of (4), the associated equation

$$\left(\frac{\partial B}{\partial x}\right)^2 + \left(\frac{\partial B}{\partial y}\right)^2 = (\mu_0 J_c)^2 \quad (5)$$

and choose the solution where (dB/dr) has the right sign. Chen *et al* (1990a, b) have considered regular polygons, and also other shapes which have parallel sides terminated by parts of a regular polygon or a circle. Based on all these calculations one can make some general observations about the virgin and hysteresis magnetization curve, schematically depicted in figure 10(a). The magnetization of the virgin curve saturates (at $-M_s$) at an applied field H_1 , which is the field at which the flux-front has collapsed (and encloses zero area). This field H_1 is dictated by J_c and the shortest dimension (D) of the cross-section as $H_1 = \mu_0 J_c D/2$. If the field is reversed from an arbitrary $H_m > H_1$, then the magnetization saturates at M_s for fields $H < (H_m - 2H_1)$. The value of M_s , and the shape of the magnetization curve in the region where magnetization varies with H , depend on the details of the cross-section (and not just on the shortest dimension). For some particular shapes we have listed below the values of M_s .

$$\text{Circle: } M_s = \mu_0 J_c D/6$$

$$\text{Slab: } M_s = \mu_0 J_c D/4$$

Finite rectangle [with sides being L and $D (L > D)$]: $M_s = \mu_0 J_c [D/4][1 - D/3L]$.
 [Ellipse major and minor axes being L and D , with $L/D \rightarrow \infty$]: $M_s = 2\mu_0 J_c D/3\pi$.

We wish to emphasize here that the magnetization of an infinite rectangle and an infinite ellipse approximating it are not equal, because the magnetization (per unit volume) has a non-vanishing contribution from the edges of the rectangle.

Since J_c does not depend on the field, it follows that the hysteresis curves for the case where the applied field is cycled about H_{dc} (i.e. between $H_{dc} + H_m$ and $H_{dc} - H_m$) are identical to the curves for $H_{dc} = 0$ except that they are suitably displaced. We shall make use of this feature in § 4.4.

4.2.2. *Anisotropic J_c in the rectangular case:* In the case of a rectangular cylinder the shielding currents, which flow along the lines of constant B , are always parallel to the two finite sides of the sample surface. (This situation is violated for samples of arbitrary cross-section, as is seen in the case of an ellipse). We denote the currents along the two direction by J_c^L and J_c^D . Then $(dB/dr)_n = \mu_0 J_c^L$ when we consider the movement of the flux-front parallel to the short side, and $= \mu_0 J_c^D$ for the perpendicular side. If we denote $(J_c^L/J_c^D) = K$, then (5) is replaced by

$$\left(\frac{\partial B}{\partial x}\right)^2 + \left(K \frac{\partial B}{\partial y}\right)^2 = (\mu_0 J_c^L)^2$$

which implies that the problem is equivalent (Grover and Chaddah 1991) to considering a rectangle of sides L and $(D' = KD)$ with $J_c = J_c^L$. The general results of § 4.2.1 hold with the constraint that the "shorter" dimension is no longer D , but the smaller of L and KD . The HTSC have very different values of J_c for currents in ab plane and along c -axis, and the "shorter" dimension is no longer determined from the physical dimensions alone. This feature was highlighted by Gyorgy *et al* (1989) while studying the magnetization curves of YBaCuO crystals with the field in the ab plane. It was pointed out by Peterson (1990) that the ratio K can be determined experimentally by studying samples of different physical dimensions.

4.2.3. *Field-dependent J_c* : As discussed in §4.2, (5) is in principle integrable for arbitrary $J_c(B)$. Bhagwat and Chaddah (1991) have recently argued that the field-dependent J_c problem is mathematically identical to the constant J_c problem provided one deals with a new field variable

$$h(B) = \int \frac{J_c(0)}{J_c(B)} dB.$$

We shall first review the calculations in literature for various forms of $J_c(B)$, and then discuss some general results provided by this idea of a new field variable.

Kim *et al* (1963) had considered a $J_c(B)$ that decays with field as

$$J_c(B) = \frac{J_c(0)}{1 + \frac{|B|}{B_0}}.$$

Kim's model has been used to calculate the virgin magnetization curves for cylinders whose cross-section is a circle, or an infinite rectangle (commonly referred to as a slab), or a finite rectangle (Kim *et al* 1963; Hulbert 1965; Watson 1968; Chaddah *et al* 1989b; Chen and Goldfarb 1989). In the limit $H_0 \rightarrow 0$, the form reduces to a power law and can be written in general as

$$J_c(B) = \frac{J_c(0)}{B^q}.$$

This model has been solved for the slab and cylinder geometries (Irie and Yamafuji 1967; Green and Hlawiczka 1967; Yeshurun *et al* 1990). An exponential model was introduced by Karasik *et al* (1971) and, independently for the HTSC, by Ravi Kumar and Chaddah (1989). Here

$$J_c(B) = J_c(0) \exp(-|B|/B_0)$$

and this has been solved for the slab, circular and rectangle geometries (Chaddah *et al* 1989b; Chen *et al* 1990a, b).

In figure 10(b) we show a schematic of the virgin magnetization curve for the case where J_c decreases with increasing B . We also show hysteresis curves, for field reversed from H_m , for a few values of H_m . This schematic brings out some general features of calculations of various models in which $J_c(B)$ decays with increasing B .

We note that the calculated virgin curve shows a minimum, and it never saturates. This minimum occurs at a field $H_v < H_1$, where H_1 is the field at which the flux-front collapses to enclose zero volume (Chaddah *et al* 1989b). The hysteresis curve for which $H_m = \infty$ provides an envelope that merges with the virgin curve at H_1 , and that has a minimum at a field $H_h < H_v < H_1$. A hysteresis curve for a cycle reversed at a finite H_m merges with the envelope hysteresis curve (for $H_m = \infty$) at $H_m - H'$, where H' is a function of H_m . For $H_m > H_1$, H' decreases as H_m increases and it follows from Bhagwat and Chaddah (1991) that H' can be obtained in terms of the generalized field variable h as

$$h(H_m - H') = h(H_m) - \mu_0 J_c D.$$

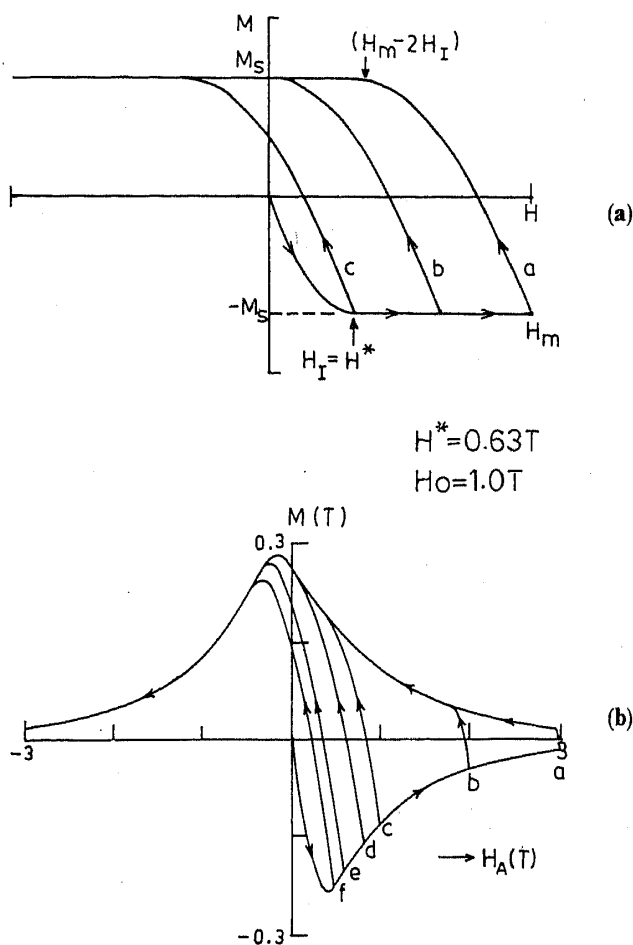


Figure 10. Schematic virgin and hysteresis magnetization curves are depicted, with the field reversal occurring from various values of H_m . In (a), we assume that J_c is a constant, while in (b), J_c is assumed to decrease with increasing field. Note that the magnetization curve on field reversal merges with the envelope curve at $H_m - h'$, with h' decreasing as H_m is lowered from 3 T to about 0.63 T (which is assumed to be H_{II} in this schematic).

We shall discuss the use of these general results in §4.3. We define the field H_{II} as that value of H_m for which $H' = 0$. We wish to emphasize that since J_c is a function of field, the hysteresis curve for the case where the field is cycled about H_{dc} is no longer simply related to the case where it is cycled about zero.

Calculations for a rectangular sample where J_c is a function of field and is anisotropic in that $J_c(0)$ is different for the two directions have not been reported so far. These correspond to scaling both the field variable and the sample dimensions, and should be straightforward. The problem where the field-dependence of J_c^L and J_c^D is different is also solvable, and appears to be relevant when there are different field dependences for J_c in the ab plane and along the c -axis.

4.3 Magnetization J_c

The calculations reviewed above gained importance because the hysteresis in the constant- J_c case saturates to a value which is linearly proportional to J_c . It was proposed (Bean *et al* 1962) that, even for J_c dependent on field, the hysteresis at any

field H should be linearly proportional to $J_c(H)$ as

$$J_c(H) = \Delta M(H)/KD \quad (6)$$

where K is a constant that depends on the geometry of the sample. As discussed in §3.1.4, the above relation has been widely used in the HTSC to estimate $J_c(H)$. Its usage was justified by Fietz and Webb (1969) who expanded the flux profile inside the sample in a Taylor series around the induction at the surface. Using $(dB/dr)_n = +(-)\mu_0 J_c$ for the field decreasing (increasing) case, they showed that (6) is valid to order dJ_c/dB . After the discovery of HTSC, we (Ravi Kumar and Chaddah 1989) calculated the hysteresis curve for a $J_c(B)$ decaying exponentially with $|B|$ and inferred back $J_c(B)$ from these calculated ΔM using (6). Since we found gross disagreements, we solved the hysteresis curves analytically in both our and Kim-Anderson forms of $J_c(B)$, for two different shapes of the cross-section. Following these calculations, the following general conditions were obtained (Chaddah *et al* 1989b) on the validity of (6). The first result is that (6) will underestimate $J_c(0)$ for all forms of $J_c(B)$ that decrease with increasing $|B|$. The extent by which $J_c(0)$ is underestimated will be more for larger samples. The $J_c(B)$ inferred using (6) will, in the low-field region, display a highly reduced field-dependence. Equation 6 will, however, provide a good estimate of the actual $J_c(B)$ for fields larger than H_1 . H_1 is the field for full penetration and its experimental determination from the merger of the virgin and hysteresis has been discussed in §4.2.3.

Finally, if the hysteresis curve is obtained by reversing the field from H_m , then (6) should not be used for $H > H_m - H'$, where H' is a function of H_m and has been again defined in §4.2.3. The use of (6) to infer $J_c(B)$ for Nb powder shows a smooth field dependence when the data is restricted to the field region defined above (Sarkissian *et al* 1991).

4.4 Response to ac fields

We now consider the response to applied fields of the form $H(t) = H_{dc} + H_{ac} \cos wt$. The analysis assumes the frequency w is small enough for the sample to respond quasi-statically. For $H_{dc} = 0$, the response is fully described by the hysteresis curve. The area enclosed within the hysteresis curve is equal to the power loss per cycle, and can be written as

$$\oint M dH = Q = \frac{H_{ac}^2}{2\mu_0} \chi_1'' \quad (7)$$

where χ_1'' is called the imaginary susceptibility. Calculations of χ_1'' (or power loss) have been explicitly performed in the case where J_c is independent of fields (see Wilson 1983), for both the slab and the circular cylinder geometries. Needless to say, calculations of χ_1'' are only one step away from those of these hysteresis curve, and are just a small exercise for all the cases discussed in §4.2. in which the solution of the hysteresis curve exists. One qualitative feature evident from the shape of the hysteresis curves is that at very low values of H_{ac} the area Q rises as H_{ac}^3 or faster, while at very large H_{ac} Q rises as H_{ac} or slower. This, alongwith (7), tells us that χ_1'' will show a peak as a function of H_{ac} . The position of this peak, as we have cautioned earlier (Bhagwat and Chaddah 1990), depends on the sample geometry considered and does not necessarily coincide with any particular field like H_1 .

The non-linear magnetic response implies that the magnetization in an ac field of frequency w will contain a response at higher harmonics nw , and this magnetization will not be in-phase with the applied field. One can thus write

$$M(t) = \sum_{n=1}^{\infty} [M'_n \cos nwt + M''_n \sin nwt]$$

or

$$\begin{aligned} \chi(t) &= M(t)/H_{ac} \\ &= \sum_{n=1}^{\infty} [\chi'_n \cos nwt + \chi''_n \sin nwt] \end{aligned}$$

where χ'_n and χ''_n are referred to as the real and imaginary parts of the complex harmonic susceptibility $x_n = x'_n - ix''_n$. We wish to stress here that while x'_1 is related to the area enclosed by the hysteresis loop through relation (7), no such simple identification can be made for any of the other components of x_n . In particular, x'_1 is not equal to $M(H_{ac})/H_{ac}$ in the general case.

Calculations of x'_n and x''_n are presented, for the field-independent J_c case, by Ji *et al* (1989) for the slab geometry. For $H_{dc} = 0$, the symmetry of the hysteresis loop [$M(t) = -M(t + \pi/w)$] results in $x'_n = x''_n = 0$ for even n . The presence of a finite H_{dc} translates the hysteresis loop without distortion in this case, and all even components continue to be zero. The vanishing of the even harmonics, for non-zero values of H_{dc} , is a feature inherent in Bean's assumption of field-independent J_c and holds for all sample shapes. Ji *et al* (1989) have stressed that all odd harmonics are proportional to H_{ac}/H^* for $H_{ac} < H^*$ in their results for the slab. This result breaks down in the geometry of the circular cylinder.

Calculations have also been done to obtain χ'_n and χ''_n for field-dependent J_c . Muller (1989) calculated χ'_1 and χ''_1 for $J_c(B)$ following Kim-Anderson model, both for $H_{dc} = 0$ and for finite H_{dc} . Ji *et al* (1989) have solved for the slab geometry for $J_c(B) = J_c(0)/|B|$, for $H_{dc} = 0$ and also approximately for non-zero H_{dc} . The symmetry of the hysteresis curve for $H_{dc} = 0$ again dictates that all even harmonics vanish in this case. The field-dependence of J_c shows up in that the even harmonics are non-zero for finite H_{dc} , and Ji *et al* predicted how various harmonics should vary as H_{dc}/H^* varies (for fixed H_{ac}).

In all the above calculations the magnetic fields (H_{dc} or H_{ac}) are scaled with respect to the parametric field of the sample ($H^* = \mu_0 J_c(0)D/2$). Ji *et al* discussed the temperature dependence of χ_n , for fixed H_{dc} and H_{ac} , by assuming a temperature dependence of $J_c(0)$. Muller (1989) calculated χ'_1 and χ''_1 as a function of temperature. Ishida and Goldfarb (1990) have calculated χ_n as functions of H^* (for various fixed H_{dc} and H_{ac}) and successfully compared the qualitative features of calculated $\chi_n(H^*)$ with measured $\chi_n(T)$ (as H^* decreases and T increases).

4.4.1. *Samples with two components*: As discussed in § 3, sintered pellets of HTSC are 2-component systems with different $J_c(0)$ values in the intra- and intergranular regions. Most experimental measurements predominantly look either at the intergranular response or at the intragranular regions. The measurements can then be compared with calculations for a single-component hard superconductor with the parameter H^* chosen appropriately. The 2-component superconducting pellet has to be treated carefully as a two component system only when the field lies in between the H^* values for the two regions (Radhakrishnamurthy *et al* 1989). This region of fields has not

really been treated quantitatively so far, and only qualitative explanations have been offered (Chaddah *et al* 1989a).

4.5 Thermomagnetic history effects

The change in magnetization of a ZFC sample that was subjected to a field H isothermally at T_1 , on heating to T_2 , is dictated by the change in $J_c(B)$ on warming from T_1 to T_2 (see, e.g. Wilson 1983). The magnetization at T_2 is then the same as if the sample was cooled in zero field only to T_2 and isothermally subjected to H . Ravi Kumar and Chaddah (1988) extended the ideas of the critical state model to calculate the change in magnetization when the sample is cooled back from T_2 to T_1 . Their work has been used to calculate the effect of various thermomagnetic histories (Grover *et al* 1989a, b), and the basic premises used are

1. All isothermal field changes cause shielding currents to be induced in accordance with Bean's model
2. Cooling a sample in a constant magnetic field causes a change in the magnetization that is driven only by the change in the equilibrium magnetization. The full equilibrium magnetization may not be seen because of flux trapping.
3. Warming a sample in a constant magnetic field causes any shielding currents that may be present to decay to the value $J_c(B)$ corresponding to the higher temperature. The equilibrium magnetization will decrease in magnitude, consistent with the condition that changes in equilibrium magnetization, as temperature is varied, are reversible.

Based on the above ideas, and assuming negligible flux-trapping on cooling, the magnetization of a ZFC sample subjected to a field H was calculated as the temperature is varied in constant H (Ravi Kumar and Chaddah 1988). The results qualitatively reproduce the experimental measurements of Muller *et al* (1987) discussed in §3. These calculations explain also the increase of the slope of the reversible regions with increasing temperature.

Assuming complete flux trapping (i.e. zero equilibrium magnetization), the same model was used to calculate the isothermal magnetization curves of samples cooled in various fields (Grover *et al* 1989a). Here only the qualitative features were predicted. The new feature brought out was that the sample will lose the memory of the field it was cooled in after it has been subjected, isothermally, to a certain minimum change in magnetic field. This minimum change required depends on the direction of field change, and correlates with the field (H_{FC}) the sample was cooled in. It is less for larger H_{FC} if the isothermal change has the same sign as H_{FC} .

4.6 A reinterpretation of the critical state model

Bean's model presents the response of a conductor, which has $\sigma = \infty$ for $J < J_c$ and $\sigma \rightarrow 0$ for $J > J_c$, to changes in the magnetic field. The conductor response involves setting up shielding currents in accordance with Lenz's law. As $J_c \rightarrow \infty$, the model reproduces the response of a perfect conductor. In that limit, for any change in the applied field, the change in the total flux contained in the sample is always zero. For a conductor with finite J_c , we postulated that Bean's model should correspond to the physical constraint that "for any change in the external field, the direction and

magnitude of the shielding currents set up is assumed to be such as to minimize the change of the total flux contained in the sample" (Chaddah *et al* 1989b). As noted subsequently (Bhagwat and Chaddah 1989), this statement appears as a logical outcome of Lenz's law and is also seen to be equivalent to the original statement of Bean's model for all the shapes having zero demagnetization factor (N) solved so far. The shielding currents have to satisfy $J \leq J_c(B)$, and Maxwell's equations must of course be satisfied.

Restating Bean's model as a condition of minimal flux entry (or expulsion) is necessary, as we shall see in §4.7, before the model can be solved for sample shapes with $N \neq 0$. In an attempt to approximately solve the case of a transverse cylinder ($N = 1/2$), Matsushita (1990) has also stated that the shielding current distributions "should be determined so as to minimize the variation in the inner magnetic flux." We shall now argue that this reinterpretation is analogous to the "minimally stable" criterion used to determine the shape of sand-piles (Bak *et al* 1987).

Analogies between sand-piles and the critical state of a hard superconductor have been drawn continuously in literature. De Gennes (1966) had compared the motion of vortices on depinning ($J > J_c$) to that of avalanches (local slope $\theta > \theta_c$) in sand-piles. Campbell and Evetts (1972) have noted that the shape of an evolving sand-pile is determined by (4) with B replaced by the height of the sand and J_c by the critical angle of repose θ_c . The field-increasing case corresponds to sand being filled into a hollow cylindrical container of appropriate cross-section with the inflow of sand occurring at the perimeter. The field-decreasing case corresponds to the container being lowered, with sand flowing out of the perimeter. In both cases, the flow of sand stops as soon as $\theta \leq \theta_c$ holds everywhere. The first such stable configuration, which Bak *et al* (1987) call as locally minimally stable, requires that the least amount of sand flows out of the container. Continuing the analogy between sand and B , the requirement of "minimum flux expulsion" suggests itself for the field decreasing case.

4.7 Samples with non-zero demagnetization factor

We have so far discussed only sample shapes that are infinitely long along the field axis and thus have a demagnetization factor $N = 0$. Experimental samples are of finite size, and $N = 0$ can be approximated by making D , the dimension perpendicular to the field, arbitrarily small. Taking this limiting case is not suitable for hard superconductors because the hysteresis ΔM also tends to zero as $D \rightarrow 0$, and any experimental manifestation of hysteresis requires a sample of finite D .

The properties of hard superconductors have been of great interest to those concerned with superconducting magnets (Brechna 1973; Wilson 1983). The magnet wire is usually experiencing a field perpendicular to its axis, and in this geometry $N = 1/2$. The solution for $N = 0$ samples discussed so far cannot be directly applied here.

Attempts to calculate magnetization curves for samples with $N \neq 0$ have been continuously made. All the efforts reviewed below have been made with Bean's simplifying assumption that J_c is independent of field. Campbell and Evetts obtained approximate solutions to the virgin magnetization curve in the limit of zero applied field, for a sphere ($N = 1/3$) and for an infinitely long circular cylinder perpendicular to the field ($N = 1/2$). In both these cases they obtained $M = -H/(1 - N)$ in the limit $H \rightarrow 0$. Clem and Kogan (1987) and Daumling and Larbalestier (1989) have calculated

the saturation magnetization for a sphere and a thin disc (with axis along the field) respectively. In both these cases the cylindrical symmetry dictates that shielding currents flow along $-\hat{e}_\phi$. Both the calculations assume that $-J_c\hat{e}_\phi$ is flowing everywhere and the saturation magnetization is obtained as the magnetic moment due to these persistent currents.

To obtain the magnetization curve one must obtain the shape of the flux-front and its evolution with increasing applied field. In the past, such an attempt has been made for the circular cylinder in a perpendicular field (Kato *et al* 1976; Wilson 1983; Matsushita 1990). In all these attempts a shape for the flux-front was assumed, with one free parameter being decided by the applied field h_a . For $H_a = 0$ the flux-front was constrained to coincide with the outer surface, while the field for full penetration H_1 was obtained when the flux front collapsed to a point. The direction of the shielding current is again determined by the symmetry of the problem, while these attempts assumed that its magnitude was J_c at all points enclosed within the sample surface and the flux-front. The parameter specifying the flux-front was obtained, for a given H_a , by requiring that $B = 0$ at the centre of the circular cross-section. These solutions were inherently approximate because, as recognized by Wilson (1983), the requirement that $\mathbf{B} = 0$ *everywhere within the flux-front* was never satisfied. These solutions were useful, however, in approximately calculating the hysteretic loss in ac fields. These showed that the loss in perpendicular geometry ($N = 1/2$) is more than in the field-parallel geometry ($N = 0$).

Bhagwat and Chaddah (1989) have recently presented solutions of Bean's model for sample shapes being an ellipsoid of revolution (field is along the axis of revolution) and for cylinders of elliptical cross-section (field is along one of the ellipse axis). Their work first brought out why a solution had not been obtained so far. When H_a is increased for a ZFC sample with $N = 0$, the local $B(r)$ at all points in the sample either increases or remains zero. For an $N \neq 0$ sample, on the other hand, the local $B(r)$ at some points in the sample must decrease even when H_a is increased. The region within the sample surface and the flux-front must carry shielding currents flowing along $-\hat{e}_\phi$ at some points and along \hat{e}_ϕ at others. In support of this argument, they could rigorously show that no solution for the flux-front exists if one assumes that shielding currents flow only along $-\hat{e}_\phi$.

If one averages the shielding current over a macroscopic region (of dimensions comparable to the inter-vortex spacing) around \mathbf{r} , and calls this averaged current density $J_{sh}(\mathbf{r})$, then one sees that $J_{sh}(\mathbf{r}) \leq J_c$. Bean's model is now invoked, following §4.6 by requiring that the change in the flux contained in the sample, for a given change in H_a , be minimized. Bhagwat and Chaddah (1989, 1990) could then obtain the virgin and hysteresis magnetization curves, and also the power loss per cycle (and thus χ''_1), for the sample shapes mentioned earlier. This work for shapes with $N \neq 0$ has not yet been extended to the case where J_c is field-dependent.

4.8 Calculation of time-decay of magnetization

At non-zero temperatures, the magnetization of a hard superconductor decays with time. The decay of magnetization is understood in terms of Anderson's (1962) idea of thermal excitation of flux bundles, over an effective pinning potential, in the direction of the flux gradient. If U_0 is the pinning potential in the absence of a current (or a flux gradient), then the effective pinning potential in the presence of J

and a local induction B is reduced to $U_0 - \alpha JB$ in the direction of the flux gradient, but is raised to $U_0 + \alpha JB$ in the opposite direction. Thermal activation of the flux bundle is possible in both directions, and gives rise to a resistivity (Anderson and Kim 1964; Campbell and Evetts 1972),

$$\rho \sim \frac{B}{J} \exp \left[-\frac{U_0}{KT} \right] \sinh \left[\frac{U_0 \cdot J}{KT \cdot J_c} \right].$$

If we consider a thin cylindrical shell, then the shielding current J in this decays as (Campbell and Evetts 1972)

$$J(t) = J(t_0) \left[1 - \frac{KT}{U_0} \log \frac{t}{t_0} \right]. \quad (8)$$

If one considers a thick sample where the induction is B and its gradient is ∇B (this assumes that B is much larger than ∇B times the sample thickness) one obtains (Beasley *et al* 1969; Xu *et al* 1989)

$$M(t) = M(t_0) \left[1 - \frac{KT}{U_0} \log \frac{t}{t_0} \right]. \quad (9)$$

We must mention here that (9) follows from (8) for a thin cylindrical shell. Hagen and Griessen (1989) have recently extended this formulation to replace a single U_0 by a distribution $f(U_0)$ due to possible disorder in the sample. The critical state model was first invoked, to explain the details of magnetization decay in HTSC, by Yeshurun *et al* (1988). They basically assumed constant- B shells in the sample, where the shielding current at each shell decays in accordance with (8). This then results, to first order in KT/U_0 , in the magnetization of the sample decaying as

$$M(t) = M(t_0) \left[1 - \frac{KT}{U_0} A(H, H^*(T), H_0(T)) \log \frac{t}{t_0} \right]. \quad (10)$$

In (10) we have incorporated the features that B varies inside the sample, and this variation is dictated by H^* and the applied field H . The field dependence of J_c is also incorporated through the parameters $H_0(T)$. The inclusion of the factor A is crucial to properly understand experimental temperature and field dependences of $[1/M(t_0)] dM/d \ln t$ (Chaddah and Bhagwat 1990, 1991) and its form depends on the thermomagnetic history. We now review recent efforts to calculate A for various experimental situations.

Most calculations have been performed for the case where T is held fixed and thus the only variable is H . Yeshurun *et al* (1988) assumed that J_c decays with field as a power law and calculated A for a ZFC sample subjected to a field H which is held constant. Chaddah and Ravikumar (1989a, b) performed the same calculation but for a J_c that decays exponentially with field. They also considered the case of a ZFC sample subjected to a field H which is then switched off, and the decay of the remnant magnetization is measured. The calculated $dM/d \ln t$ are depicted in figure 11. The general results that follows from these calculations are

- (i) For small H , the decay rate with field-on will be about four times that with field-off.
- (ii) The decay rate with field on will reach its maximum value at H_I ; and
- (iii) The decay rate for field off case will become independent of H for $H > H_{II}$.

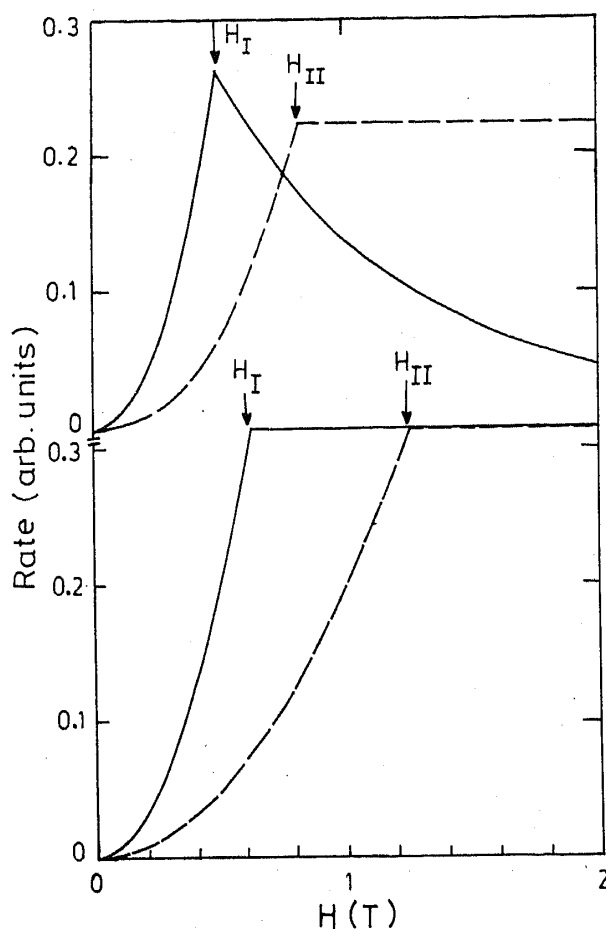


Figure 11. $[dM/d \ln t]$ is plotted for $H_0 = 1$ T (upper set of curves) and $H_0 = \infty$ (lower set). These calculations have used $H^* = 0.63$ T. Full line corresponds to field-on case and dashed line to field-off case (Chaddah and Ravi Kumar 1989b).

Kunchur *et al* (1990) have calculated A for the case of a ZFC sample (at fixed T) subjected to a field H (at time $t = 0$) which is increased by ΔH to H_1 (at time $t = \tau$). They calculated $M(t)$ for $t > \tau$, and found that A becomes independent of H (and depends only on H_1) at $t > \tau + \tau'$. They assumed that J_c is independent of field and calculated the dependence of τ' on τ , ΔH , and H_1 . Only recently (Chaddah and Bhagwat 1991) has an attempt been made to include the temperature dependence explicitly by incorporating the T -dependence of H^* and H_0 .

We are not discussing here the attempts to discuss time-decay of magnetization by going beyond the phenomenological critical state model. Some references to such attempts may be obtained from Malozemoff and Fisher (1990).

5. Experimental results on conventional hard superconductors—General features and their understanding

In this section we shall discuss measurements of irreversible magnetization in conventional hard superconductors. The irreversibilities in these materials could be substantially reduced by enhancing their purity etc., and were not extensively studied

as they were considered extrinsic. As argued in §2, magnetic irreversibilities are essential in superconductors for current carrying applications, and it was in 'applied superconductivity' literature that many studies appeared (see Brechna 1973; Clem 1979; Wilson 1983; Sampson 1986).

The extensive studies of magnetic irreversibilities in HTSC prompted similar measurements in conventional superconductors. While reviewing the work on conventional materials, we shall discuss the similarities and contrasts with HTSC. We shall discuss also the current understanding of each type of measurements within the model discussed in the last section.

5.1 ZFC samples studied at fixed temperature

5.1.1 *Hysteresis curves*: The first detailed measurements, on ZFC Nb₃Sn cylinders was reported by Bean (1962) in support of his model. By measuring virgin magnetization curves on a cylinder machined to two different radii, he could fit the data with a single value of J_c . In this analysis Bean also incorporated the lower critical field H_{c1} . This version of his model is not universally accepted and there is still no consensus on how H_{c1} is to be incorporated. We shall discuss this feature in §6. As discussed in the last section, the version that ignores H_{c1} as small (Bean 1964) is where there is universal agreement.

Bean's prediction of hysteresis that scales with sample size was qualitatively verified immediately, and detailed measurements were made in the 1960's on various hard superconductors such as Nb₃Sn (Kim *et al* 1963), Nb-Zr (Fietz *et al* 1964), Pb-Bi (Campbell *et al* 1968) etc. Most of these measurements are reviewed by Campbell and Evetts (1972). These measurements failed to show quantitative agreement with Bean's calculations assuming a constant J_c , and Kim's model assuming $J_c(B) \propto (1 + B/h_0)^{-1}$ was found to give good agreement in most cases. In some systems the assumption $h_0 \gg B$ could be made (Watson 1968), while Kim's model could not fit the NbZr data at fields above 1.5 T (Fietz *et al* 1964). The latter data led to the first proposal of an exponential decay of J_c with B , and the virgin magnetization curve was solved by Karasik *et al* (1971) to fit their NbTi data.

In the last few years conventional superconductors have been studied mainly for comparison with HTSC. The only new feature in the study of hysteresis curves has been attempts to measure the field for full penetration H_1 , and some numbers so obtained have been used to cross-check features in the time-decay measurements (Grover and Chaddah 1991).

The other samples of conventional hard superconductors in which detailed studies of hysteresis curves have been made in the last few years are the technologically relevant multifilamentary wires. As discussed in §4, the hysteresis would reduce with decreasing filament size, and so would the associated problems in the magnet for a high energy storage ring (Brianti 1986). Subject to the constraint that the filament dimension should remain constant along the entire length of the wire, technology for the fabrication of NbTi/Cu wire with filaments of 2 to 3 μm diameter has been developed (Sampson 1986; Larbalastier *et al* 1986). Such wires have shown very interesting hysteresis curves. Colling (1988) and Hlasnik *et al* (1985) have observed bubble shaped hysteresis curves similar to those seen in HTSC (§3.1.1). This bubble was also found to evolve into a pair of kink like anomalies when the cycling field

was increased from 8 mT to 0.3 T. The temperature evolution of the low field hysteresis curve was also studied, in a cycling field of 8 mT, as the temperature was raised from 4.5 K to 8.4 K (Collings 1988). This behaviour is in almost complete analogy to that seen in HTSC, and is attributed to weaklinks, formed by the copper matrix between the superconducting filaments. The proximity induced superconductivity in copper is believed to be destroyed in high fields. This understanding has been strengthened (and the technological hurdles in the use of this wire were overcome) when Ghosh *et al* (1988) showed that doping the copper matrix with 0.5 atomic percent manganese drastically reduced the low field hysteresis.

5.1.2 *AC loss and harmonic generation:* The out-of-phase susceptibility χ'' had been studied in conventional elemental superconductors like lead and tin (Maxwell and Strongin 1963; Ishida and Mazaki 1979) and Ishida and Mazaki concluded that a peak in χ'' is indicative of dislocations or inhomogeneity. Khoder (1983) studied a type II superconductor but for $H < H_{c1}$, while Hein (1986) studied χ'' in niobium but again at low fields. Hein also discussed the possible physics behind the loss mechanism, but did not attribute it to magnetic hysteresis alone. Ishida and Mazaki (1981, 1982) studied both the fundamental and higher harmonics of χ in a multiconnected superconductor, and explained the losses modelling the sample as a connected network of Josephson junctions. The ac losses in superconducting wires have, on the other hand, been explained as due to hysteresis alone, and good agreement with calculations based on Bean's model is obtained (see §8.2 of Chapter 8 of Wilson 1983). The agreement between measurements and calculations of the critical state model, observed in HTSC, thus appears to have settled an issue that remained controversial in conventional superconductors.

5.1.3 *Time decay of magnetization:* Time decay of magnetization was extensively studied in conventional hard superconductors in the 1960s (see Campbell and Evetts 1972; Kim and Stephen 1969 for reviews) and was understood in terms of Anderson's ideas (Anderson and Kim 1964). During the last few years measurements of magnetization decay in conventional superconductors have been performed at much smaller applied fields, and also under various thermomagnetic histories. These have been motivated, to a reasonable extent, by attempts to duplicate effects first observed in the HTSC. The manifestation of thermally activated creep, as has been noted by Campbell and Evetts (1972), was not apparent in conventional hard superconductors presumably because of large values of U_0/KT . Much higher accuracy now available in magnetization measurements (see e.g. the data of Mota *et al* 1989) may also be a possible cause for extensive measurements on conventional superconductors in recent years. Mota *et al* (1988c, d) have made detailed measurements in CeCu_2Si_2 in fields varying from 0.5 mT to 50 mT, and in an organic superconductor in fields from 0.2 mT to 2 mT.

Measurements were made, analogous to their studies in HTSC (Mota *et al* 1988a, b), on a ZFC sample with field on, and also after the field is switched off. Further measurements were also made in a bulk CeCu_2Si_2 sample, and also in a powder specimen from the same starting material. The field dependence of $dM/d \ln t$ was found to vary for small fields, as H^n with $n \approx 4$ in CeCu_2Si_2 and ≈ 3.5 in organic superconductors. $dM/d \ln t$ was found to flatten at higher fields, as expected (see §4.8) for a hard superconductor with J_c independent of field in the range studied. The

field-off decay was smaller, by a factor of 3 to 4, than the field-on decay rate. The decay rate at low fields was larger for powder sample. This would be explained (Chaddah and Ravikumar 1989a, b) by smaller H^* of the powder; and is also consistent with the measured decay rate saturating at low fields for the powder. The low-field data on conventional hard superconductors thus appears similar to that in HTSC, with the feature that J_c is independent of field at the low fields used.

Fraser *et al* (1989) studied the decay of magnetization in ZFC samples of V_3Si subjected to fields varying from 0.1 to 1 T. The decay rate at 4.2 K shows a peak at 0.4 T, and decays at higher fields. This is consistent with J_c decaying with increasing field, as for HTSC, and the field at which the minimum in magnetization is seen is only 0.25 T. This is again consistent with predictions of § 4 (see also Grover and Chaddah 1991).

Mota *et al* (1989) have also recently made measurements on Pb–In alloy and $PbMo_6S_8$ in the field-off case. In $PbMo_6S_8$, $dM/d \ln t$ increases with field as H^3 , while in Pb–In the decay rate saturates at the same field at which the remanent magnetization saturates. This field value thus corresponds to H_{II} defined in § 4, and the results are consistent with the calculations of § 4.8.

Rossel *et al* (1990) have observed memory effects in time-decay measurements on $PbMo_6S_8$ that are strikingly similar to what they observed earlier (Rossel *et al* 1989) in YBaCuO. While they continue to view this as due to a superconducting glass phase, this behaviour follows from the explanation of Kunchur *et al* (1990) as common to all hard superconductors.

5.2 Thermomagnetic history effects

History effects in conventional hard superconductors have been studied in recent years in niobium (Schiedt *et al* 1988; Grover *et al* 1989a, b; Sarkissian *et al* 1989) and in V_3Si (Fraser *et al* 1989). The study on V_3Si was a successful attempt to reproduce all the features observed by Muller *et al* (1987) in LaBaCuO. Since the need for a superconducting glass model was never felt for the extensively studied V_3Si , Fraser *et al* argue that their success places the superconducting glass model in doubt. In particular, they studied the effects of heating and cooling cycles, in constant applied field, on the magnetization of ZFC sample of V_3Si . They observed behaviour totally similar to that observed by Muller *et al* and this was consistent with the prediction of Ravikumar and Chaddah (1988) that the history effects were common to all hard superconductors. These history effects were explained, as discussed in § 4.5, by extending the ideas of Bean's model.

The studies of Grover *et al* (1989a, b) were motivated by a need to reproduce, and extend, the features observed by the IBM group (Malozemoff *et al* 1988) in YBaCuO. To ensure that their studies could capture the underlying physics, Grover *et al* studied two samples of high purity niobium that showed very different extents of flux trapping on field cooling. They first showed, as Fraser *et al* had done for V_3Si , that there exists a temperature $T^*(H)$ above which FC and ZFC magnetizations are the same. (Grover *et al* (1991) have recently studied $T^*(H)$ in lead discs. We shall not discuss that here since the phenomenon in lead appears to depend on sample shape, and is definitely not explained by the macroscopic model of § 4). They further reproduced the equality

$$M_{rem}(H, T) = M_{FC}(H, T) - M_{ZFC}(H, T) \quad (11)$$

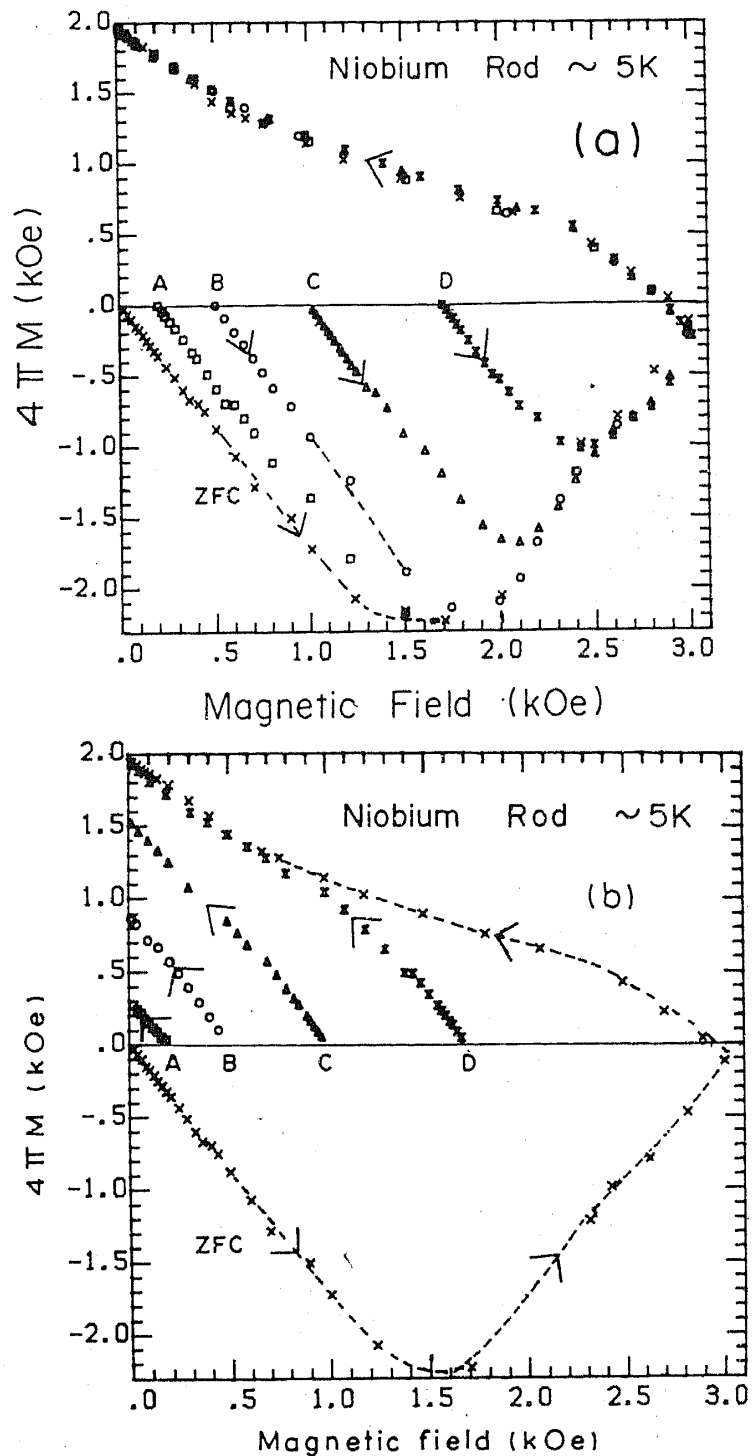


Figure 12. (a) Forward and hysteresis magnetization curves for a niobium rod sample cooled in various fields (Grover *et al* 1989a). The forward curves for FC samples finally merge with the ZFC curves as the field is increased. (b) Reverse magnetization curves for the same niobium rod after field-cooling. The reverse curves again finally merge with the ZFC hysteresis curve.

which was first reported by Malozemoff *et al* (1988) in YBaCuO. Grover *et al* showed that (11) is only satisfied at low T and small H . They also studied the isothermal magnetization curves of samples cooled in a field, and then subjected to a field

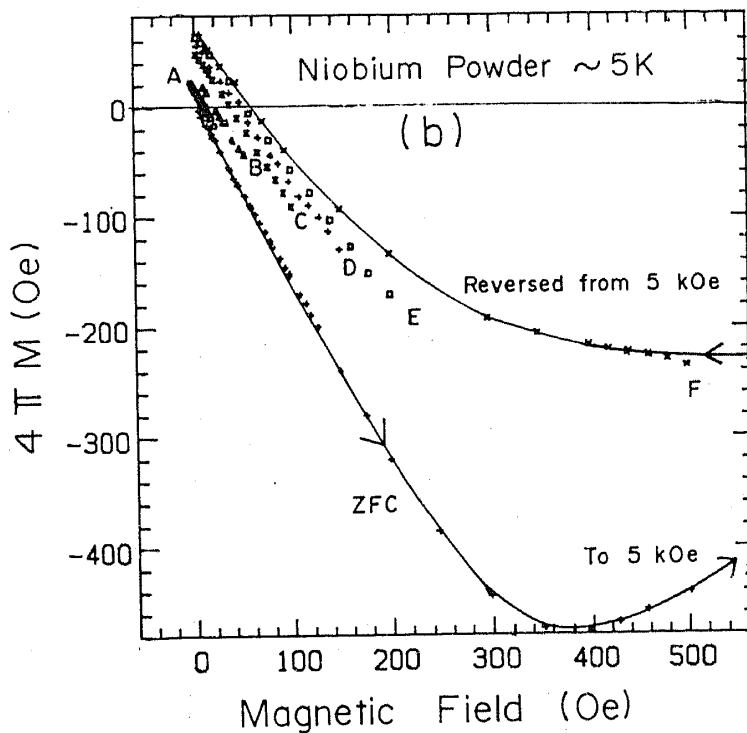
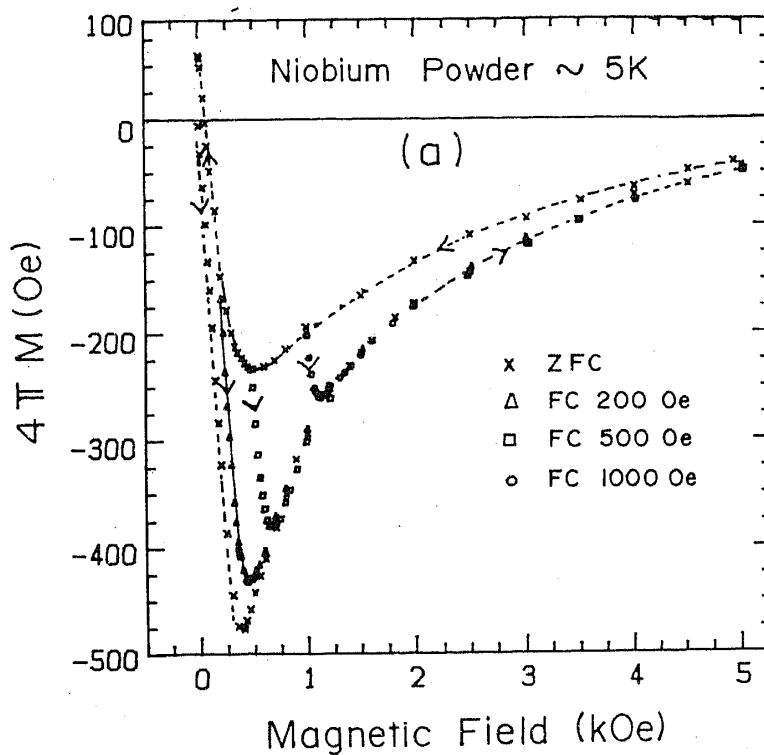


Figure 13. Data similar to that in figure 12, but for a niobium powder sample. This sample has a large M_{FC} , and also much smaller hysteresis, compared to the rod sample. (Grover *et al* 1989a).

variation. The general results of these measurements which are shown in figures 12 and 13, which were also later reproduced in YBaCuO by Sarkissian *et al*, were that:

- (i) The forward (and also the reverse) magnetization curves of a sample cooled in a

field H merges with the ZFC hysteresis curve after the field was increased (or decreased) by a certain H' . The increase H' required kept decreasing as H increased.

(ii) The equality in (11) is first replaced by $<$, and then by $>$, as either H rises towards $H_{c2}(T)$ or T rises towards $T_c(H)$. Both these features were qualitatively explained using the ideas of § 4.5. These studies on niobium thus stand out as a major test of the physical idea of Bean's model. They are also a rare example (in the last three years) of new features first observed in a conventional superconductor, and then reproduced in HTSC.

6. Discussion

As mentioned in the introduction, this review has been written as a success story of Bean's macroscopic model of a hard superconductor, and of its many extensions. The critical state model had earlier been accepted in the field of superconducting magnets, where conventional hard superconductors were extensively studied. The discovery of HTSC produced many new features in the measured magnetic irreversibilities. Over the last few years these features have been understood by extending Bean's model, but using the same underlying physical ideas.

The critical state model has some difficulties in including the equilibrium magnetization, and in handling the small field limit. The discussion in § 4 ignored H_{c1} as small. This was reasonable in conventional studies on hard superconductors where measurements were only made in larger fields (> 1 T). The studies in the last few years have been made, as discussed in this review, in fields as low as ~ 1 mT. In fact, once the applied field is less than H_1 , the model assumes that B at the flux-front goes smoothly to zero. The problems with the model are then the following. Firstly, there is a disagreement on whether the London screening currents (which contribute to the equilibrium magnetization M_{eq}) are set up at the flux-front (Bean 1962) or at the sample surface (de Gennes 1966). In recent years we have persisted with Bean's view (see Chaddah *et al* 1989a) that since one is talking of an average field (and a $J_c(B)$) at each point in the sample, the equilibrium response cannot be determined only at the surface. Krusin-Elbaum *et al* (1990) have imposed screening currents *both* at the surface and at the flux front, and have assumed that their magnitude adds up (irrespective of the applied field) to be that when the applied is only H_{c1} . Both Bean's model and de Gennes pictures dictate that the currents flow only at one surface. It suffices to state, at this stage, that the scheme of incorporating M_{eq} is not well established.

The second problem in the low field case is that the vortex separation is very large. It is not clear that the picture of replacing the vortex structure by macroscopic $J(r)$ and $B(r)$ is justified. This point was raised early (Anderson and Kim 1964), but did not warrant much discussion since the experiments were concerned with large applied fields. It probably needs to be addressed now.

A third problem concerns with the treatment of flux creep. In § 4.8 it was assumed that the critical state was established fast (i.e. $\sigma \rightarrow 0$ for $J > J_c$) and decays slowly (σ is large for $J < J_c$). Flux creep could then be treated as a perturbation of the critical state. In HTSC the change in σ , as J varies across J_c , may not be that discontinuous. This would specially be true at higher temperatures. The evolution of shielding currents (and thus of magnetization) with time probably requires more realistic calculations.

We have concentrated, amongst the HTSC, on reviewing the data on YBaCuO. This particular compound is the easiest to obtain as a single phase, and the easiest to measure. As single phase samples of other HTSC become common, their magnetization behaviour may not follow as well the model discussed in §4. The much larger flux creep seen in bismuth and thallium based cuprates would probably show the failings of the discussion in §4.8. HTSC with weak inter-layer coupling may also show up new features associated with two dimensionality. The future holds the promise of glorious uncertainties!

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