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Magnetic phase diagram of anisotropic layered superconductors via magnetization measurements for $H \| c$ in Bi2212

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Abstract. The $Bi_2Sr_2CaCu_2O_8$ system is viewed as an archetypal of superconductors modelled as Josephson coupled CuO_2 bilayers. The isothermal and temperature dependent DC and AC magnetization measurements for $H \| c$ in a single crystal of Bi2212 have been performed. Qualitative changes are observed to occur over a narrow range of temperature values before reaching the superconducting-normal transition. The observed behaviour can be ascribed to the rapid variation in the strength of the coupling between the superconducting CuO_2 planes (i.e., bilayers in the case of Bi2212). Strongly coupled planes behave like a 3D superconductor, whereas weakly coupled planes have a two component response attributable to 2D planes and interplanar couplings. We believe that this paper is a plethora of new findings. Our observations imply that resistivity across the planes becomes zero earlier than that within the planes. A new line (designated as $H_{2D}(T)$) above which the change in the electromagnetic response is dominated by quasi 2D-planes has been determined for the first time. This paper also contains the first observation of Differential Diamagnetic Effect (DDE) in the In-phase AC susceptibility data which signals the onset (at $T_{2D}(H)$) of dominance of response from 2D-planes. In addition to a host of interesting thermomagnetic history effects which are a consequence of interplay between the diamagnetic responses from the two components, a comparison of irreversibility lines (of the 3D state) determined by different methods on the same specimen of a HTSC is also being presented for the first time. We have come across Paramagnetic Meissner Effect (PME), first recognized in ceramic samples of Bi2212, in the temperature region of dimensional crossover in our single crystal sample, which *inter-alia* confirms our labelling of the two component behaviour. A schematic phase diagram summarizing the various transformations that can occur near T_c in the electromagnetic response of an anisotropic layered system has been drawn.

Keywords. Layered superconductors; dimensional crossover; magnetic phase diagram; Bi2212.

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1. Introduction

The difference in the electromagnetic response of high temperature superconductors (HTSC) vis-a-vis conventional low temperature superconducting alloys (e.g., the slower rate of decrease of electrical resistance with temperature at the superconducting transition and the absence of noticeable shift in the superconducting onset with moderate fields, etc.) have led to the pursuit of answers to the questions, like, whether

HTSC materials ever attain $R=0$ state, what the nature (topological character, size, etc.) of so called vortex lines in these materials is, and how the vortices arrange themselves, etc. (for a review, see [1]). A hallmark of cuprate HTSC is their layered structure and the accompanying large anisotropy. In recent years, a consensus appears to have emerged [1] that cuprate superconductors, with or without large anisotropy do really superconduct ($R=0$) in the vortex glass phase at low temperatures, and up to moderate field values. The glassy phase is believed to be a consequence of an inevitable presence of imperfections at the atomistic level in these materials. However, the details of the magnetic phase diagram in high field values at low temperatures or even in low fields at the elevated temperatures remain to be clearly revealed. In particular, in highly anisotropic compounds, like bismuth cuprates (BSCCO), where it is considered that vortices comprise of "wiggly flux lines" or "pancake" vortices confined to individual planes with correlations amongst them, the issue of the intrinsic strength of these correlations (and the influence of applied field and temperature on them) is in contemporary focus.

Kadowaki [2] and Kes *et al* [3] had been the first to point out a change in the character of diamagnetic response (i.e. the transformation in the sign of dM/dH from positive to negative value, before reaching the nominal zero field superconducting-onset temperature $T_c(0)$), in temperature dependent magnetization measurements for $H \| c$ in single crystal samples of $Bi_2Sr_2CaCu_2O_8$ (Bi2212). The Bi2212 system has been shown to comprise of Josephson coupled CuO_2 bilayers [4]. In this system, the diamagnetism increases as H increases at T close to $T_c(0)$ [2, 3]. A conventional analysis of such data implies that nominal $T_c(H)$ (or $H_{c2}(T)$) increases as H (or T) increases. However, such a response, since the initial works of Kadowaki [2] and Kes *et al* [3], has been considered to emanate from superconducting fluctuations confined to individual copper oxide planes (a two dimensional (2D) superconductivity *a la* 2D magnetism). The 2D superconductivity is full of exotic possibilities (e.g. see, [5, 6]), with novel features anticipated in the electromagnetic response of Josephson coupled planes (i.e., CuO_2 bilayers in the case of Bi2212). Of particular interest is the existence of a Kosterlitz-Thouless (KT) type of a transition, and the manifestations of possible spontaneous creation of vortex-antivortex pairs in individual planes [6]. From a theoretical treatment, suitable for bismuth and thallium cuprates, which incorporates the entropy contribution to the free energy due to thermal distortion, Bulaevskii, Ledvij and Kogan (to be referred as BLK) [6] showed that KT type transition can occur in these systems at a temperature $T_s < T_c(0)$. Their analysis also identifies a temperature at which the change in character of diamagnetic response occurs. It is argued that at $T \approx T^*$, the magnetization becomes field independent for $H \geqslant H_{cr}$. This, in turn, implies that temperature dependent magnetization ($M(T)$) curves for $H \geqslant H_{cr}$ would intersect at $T = T^*$. Above T^*, diamagnetic response is predicted to increase logarithmically with field, whereas somewhat below T^* the diamagnetic values decrease with field as in usual 3D superconducting materials. The temperature T^* is believed to lie below T_s and both of them lie in the region where coherence length ξ_c is much less than interplanar spacing s. The electrical resistivity and lower critical field H_{c1} are anticipated to fall to zero value at T_s instead of $T_c(0)$ [6].

Vacaru *et al* [7] found that H_{c1} data for $H \| c$ in a single crystal specimen of $Tl_2Ba_2CuO_6$ (Tl 2201) indeed linearly extrapolate to zero value at a temperature smaller than its $T_c(0)$. The temperature at which H_{c1} extrapolates to zero value is observed to lie in between $T_c(0)$ and the temperature at which change in character of diamagnetic response occurs in their sample. More recently, Brawner *et al* [8] have reported that, in a single crystal sample of Bi2212 (for $H \| c$), changes in slope values of $\mathrm{d}H_{c1}/\mathrm{d}T$ and $\mathrm{d}J_c/\mathrm{d}T$ occur at a temperature below $T_c(0)$. Their H_{c1} and $J_c(H=0)$ (as deduced from isothermal hysteresis value in zero field) values indeed collapse to zero at a temperature smaller than the nominal $T_c(0)$ in their sample. They attribute the sudden change in slope values $\mathrm{d}H_{c1}/\mathrm{d}T$ and $\mathrm{d}J_c/\mathrm{d}T$ to dimensional crossover effect. Brawner *et al* [8] further surmise that, after the collapse of H_{c1} and J_c, the diamagnetic susceptibility is *reversible*. Such an inference appears to be in contrast to the behaviour exhibited by a single crystal sample of Tl 2201 of Vacaru *et al* [7]. In Tl 2201, the irreversibility temperature $T_r(H)$ in $H = 1$ Oe lies above the temperature of collapse of H_{c1}. Another notable difference between the analysis made by Vacaru *et al* [7] and Brawner *et al* [8] is that the former group designates the temperature of $H_{c1} = 0$ as T_s, whereas, the latter group identifies T_s (the onset-temperature of K-T type of a transition) with the temperature at which slope value $\mathrm{d}H_{c1}/\mathrm{d}T$ exhibits a drastic change. In any case, the experimental data of both these groups and the scenario sketched by BLK [6] provide many interesting leads which must be followed to unravel the details of the magnetic phase diagram near $T_c(0)$. We have been concerned with the determination of irreversibility lines and lower critical fields by different magnetization techniques and methods in samples of Bi2212 [9–11]. We have reinitiated a series of new investigations by AC and DC magnetization techniques in high quality single crystal samples of Bi2212 for the said purpose. Our initial data show that as 3D to 2D crossover occurs, the magnetic response of a crystal transforms from that of a one-component system to two-component system corresponding to 2D planes and interplanar links. The two-component response is highly irreversible and the irreversibility persists up to $T_c(0)$. We identify the T^* and H_{cr} values of BLK [6] with (T_{2D}, H_{2D}) line (i.e., $\mathrm{d}M/\mathrm{d}H = 0$ line), above which the changes with field and temperature in the diamagnetic response are dominated by the response from 2D planes. Below this line, we see clear manifestations of interplay between diamagnetism of two components. Our new results lead us to schematically sketch the details of the magnetic phase diagram near $T_c(0)$. The key result and a summary of our new findings have been communicated earlier [12, 13]. The purpose of this paper is to report the details of new experiments which vividly elucidate the changes in magnetic response that occur during the dimensional crossover in highly anisotropic layered superconductors.

2. Experimental details

Our single crystal sample of $Bi_2Sr_2CaCu_2O_8$ ($1{\cdot}9 \times 1{\cdot}5 \times 0{\cdot}4\,\mathrm{mm}^3$) is cut from a boule grown by travelling solvent floating zone technique [14]. The DC and AC magnetization data for $H \| c$ have been recorded using a Quantum Design SQUID magnetometer ($MPMS_2$-system) and a home built mutual inductance bridge, respectively. The AC susceptibility system has a variable frequency 2-phase lock-in amplifier and a superposed DC field is produced using a superconducting solenoid and can be swept at a rate of 0·5 to 100 Oe/s at fixed temperatures. The bridge is usually operated at 119 Hz.

This paper is based on the following experimental runs.

2.1 Temperature dependent DC magnetization curves

We recorded magnetization data in $H = 0{\cdot}3$, 0·5, 1·3, 10, 50, 100 and 300 Oe, respectively along zero field cooled warm-up (M_{ZFC}, cycle 1), (very slow) field cool cool-down (M_{FCC}, cycle 2), field cool warm-up (M_{FCW1}, cycle 3) and warm-up after rapid (in 3 to 5 minutes) cool-down to 10 K in field (M_{FCW2}, cycle 4). No measurements can be made during rapid cool-down cycle in field. The T-dependent data have been recorded in small steps of 100 to 200 mK in the temperature regions of interest, and at each temperature a minimum wait time of 300 s was maintained. It was checked that a wait time longer than 600 s did not produce any further measurable change in magnetization values. The thermal equilibrium gets established rather slowly in Quantum Design SQUID magnetometer. Further, the overshoots and undershoots are not the same during warm-up and cool-down cycles. During cool-down, minimum undershoot is about 1 K in 60 to 90 K interval even when the set temperature is decreased by only 0·1 K. The overshoot/undershoot values during warm-up can be limited to less than 0·5 K provided the step size is not greater than 0·2 K.

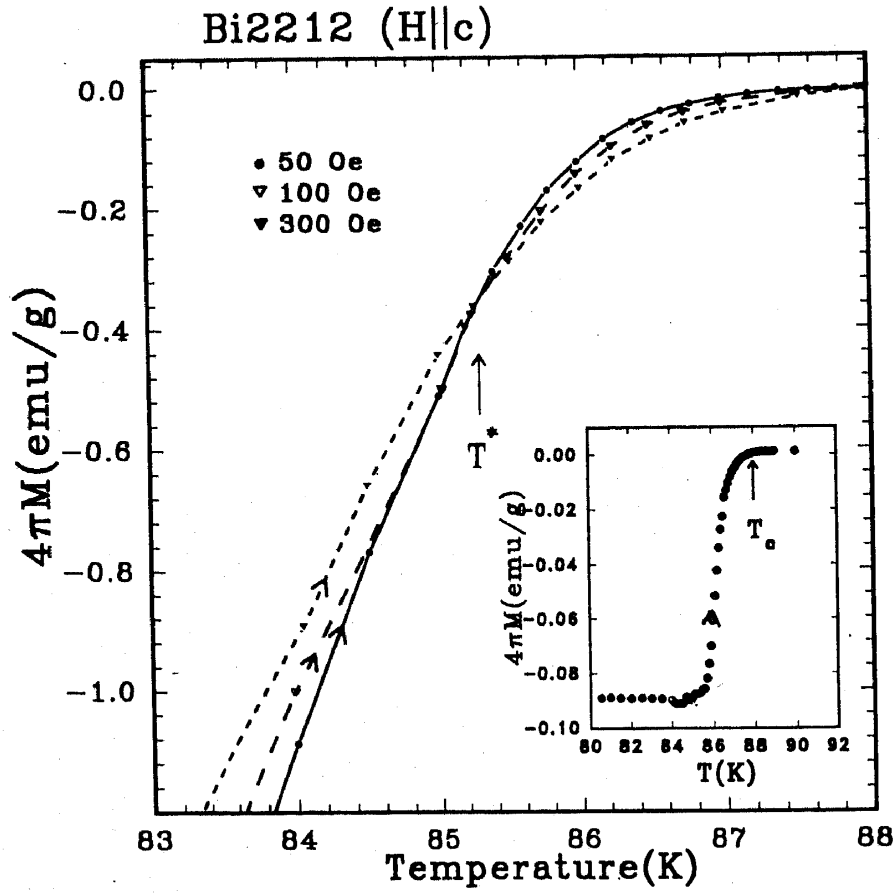

Figure 1. Zero field cooled magnetization curves, M_{ZFC} vs T, in a single crystal specimen of $Bi_2Sr_2CaCu_2O_8$ for $H(\parallel c) = 50$, 100 and 300 Oe, respectively. The temperature T^* identifies the midpoint of the temperature range encompassing the region of intersection of curves for $50\,\text{Oe} < H < 300\,\text{Oe}$. The inset shows the identification of so-called zero field transition temperature T_c via the onset of diamagnetic response in the temperature dependent magnetization curve recorded in the warm-up mode in the nominal residual field ($\approx 0{\cdot}3$ Oe) of the magnet. The sample was initially rapidly cooled down to 10 K in the same field.

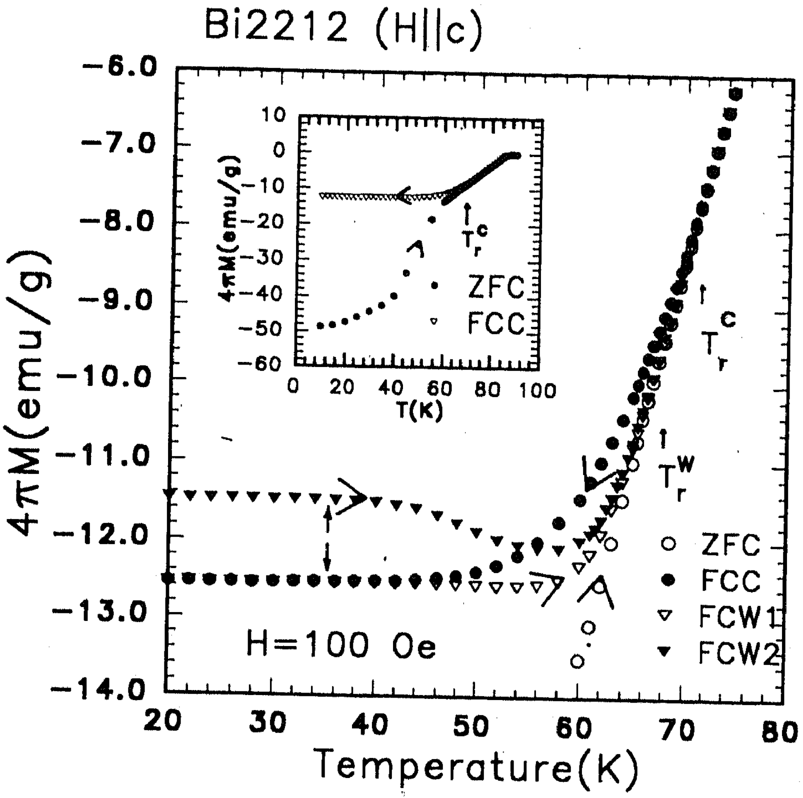

Figure 2. Temperature dependence of magnetization data in $H(\| c) = 100\,\mathrm{Oe}$ in Bi2212 sample. The acronyms ZFC, FCC, FCW1 and FCW2 identify the cycles zero field cooled warm-up, (slow) field cooled cool-down, (first) field cool warm-up and (second) field cool warm-up (after rapid field cool cool-down), respectively. The temperatures T_r^c and T_r^w identify the merger of M_{FCC} and M_{FCW1}/M_{FCW2} curves with M_{ZFC}. The temperature at which M_{FCW1} curve starts to bifurcate from M_{FCC} data can identify the limiting value of the lower critical field via the criterion suggested by Clem and Hao [19]. The inset shows $M_{ZFC}(T)$ curve over the entire temperature range.

The temperature dependent data can be used to determine the limiting values of irreversibility temperature and the lower critical field. These data also identify the temperature region across which the change in character of diamagnetic response occurs (i.e., change in dM/dH from positive to negative values, e.g., see figure 1). The merger temperature of M_{ZFC} with M_{FCC} and M_{FCW1}/M_{FCW2} identify the values of irreversibility temperatures $T_r^c(H)$ and $T_r^w(H)$ (e.g., see figure 2). In general, $T_r^c(H) > T_r^w(H)$ [11, 15–18] and Clem and Hao [19] have sought to provide an understanding of the difference between M_{FCC} and M_{FCW1} curves in the framework of critical state model. Clem and Hao [19] argue that M_{FCW1} values in an applied field H would start to differ from M_{FCC} values at a temperature T_{c1} such that the lower critical field at T_{c1} equals H. Following such a procedure, we may choose to pick out $H_{c1}(T)$ values also from the temperature dependent magnetization data.

2.2 *Isothermal DC magnetization hysteresis curves*

Magnetization hysteresis curves in fields up to $\pm 10\,\mathrm{kOe}$ have been recorded at close temperature intervals in the range 73 K to 86·7 K. For every run, the specimen was cooled down to a given temperature in nominal zero field, i.e., the residual field values

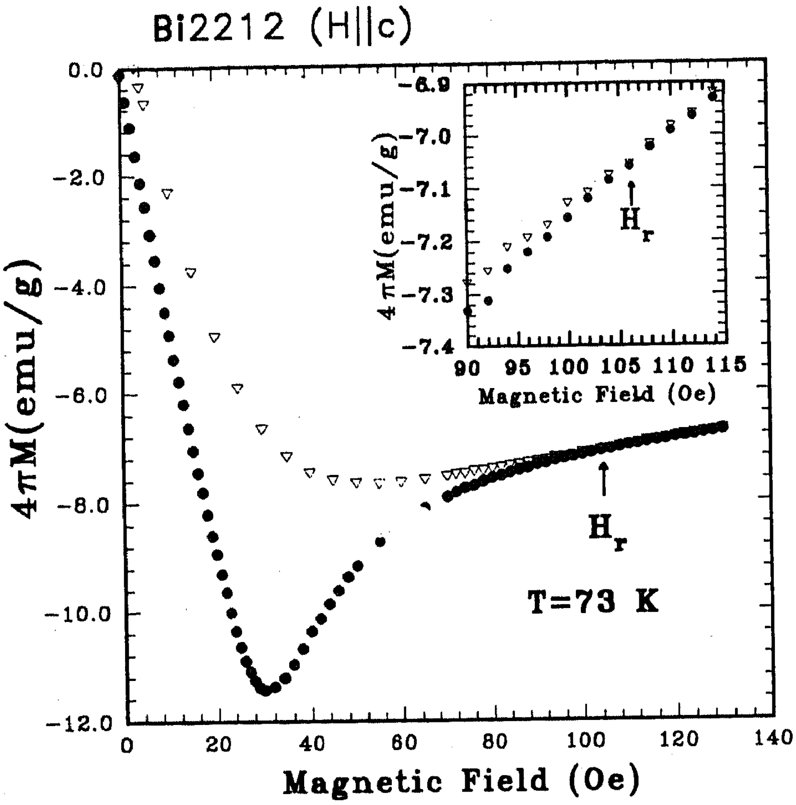

Figure 3. Isothermal magnetization curve in a Bi2212 sample ($H \parallel c$) at 73 K. The inset highlights the identification of irreversibility field $H_r(73\,\mathrm{K}) = 106$ Oe. It may be noted that T_r^c (100 Oe) = 72·5 K in figure 2.

of magnet (which were measured to lie between 0·3 to 0·4 Oe). For observing the details of magnetization curves in the low field region, the residual field of the magnet was specifically reduced to ≈ 1 mOe using Fluxgate option supplied by Quantum Design Inc. However, the smallest absolute field value which the power supply can reproducibly set is limited to 0·3 Oe and the smallest step size thereafter is limited to 0·1 Oe. The field values were incremented in non-overshoot mode, and without invoking the *hysteresis-option software*. The field increments using *hysteresis-option software* were completely avoided as it leads to various artifacts [20]. A minimum wait time of 60 s was observed at each field value.

The isothermal hysteresis curves are used to determine the values of irreversibility field $H_r(T)$ via the merger of $M - H$ curve recorded during the forward cycle with that recorded during the reverse cycle (e.g., see figure 3). The isothermal magnetization data also help to identify the field-temperature region across which dramatic changes occur in the shape and nature of hysteresis loops (e.g. see figures 4 and 5). During this changeover, the hysteresis curve appears to be a superposition of two components instead of the usual behaviour expected in an irreversible type II superconductor (cf. loops at 83 K and 86·7 K in figure 4).

2.3 *Temperature dependent in-phase (χ') and out-of-phase (χ'') AC susceptibility*

$\chi'(T)$ and $\chi''(T)$ data at a frequency of 119 Hz and with a peak amplitude in the range of 0·6 Oe to 1·3 Oe in superposed DC field values of nominal zero, 100 Oe and 300 Oe, respectively, have been recorded.

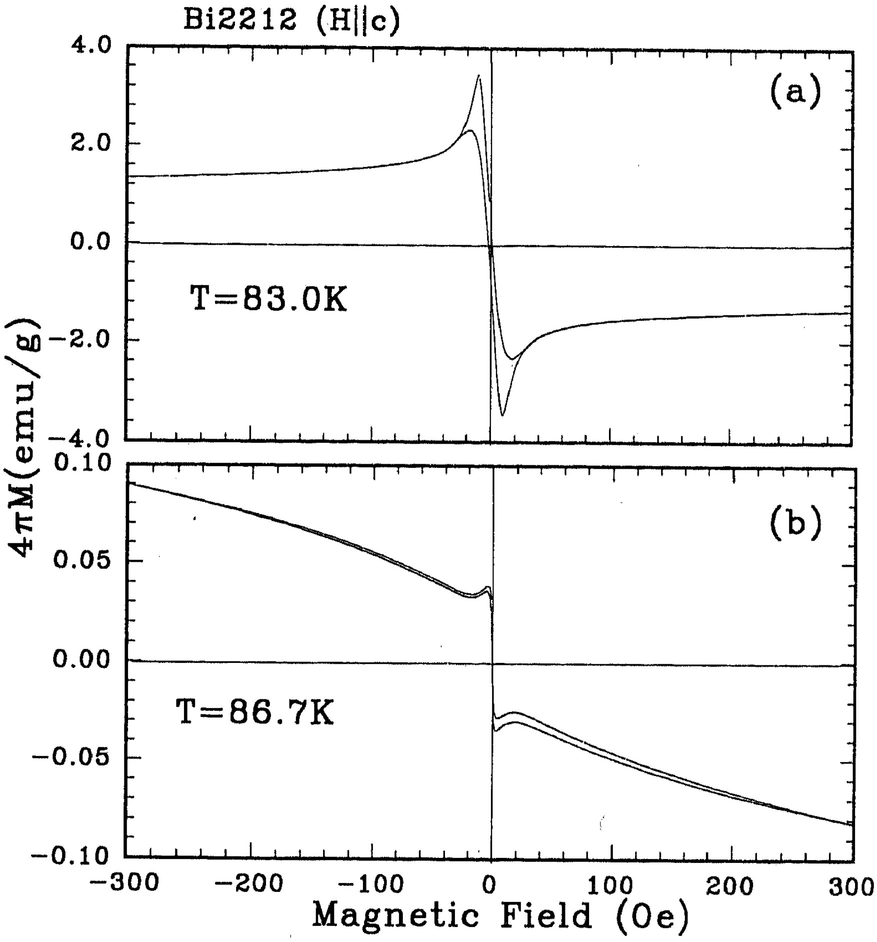

Figure 4. Comparison of magnetization hysteresis loops obtained between ± 300 Oe at 83 K ($< T^*$) and 86·7 ($> T^*$), respectively. Note the change in shape, increase in irreversibility field value and the loss of symmetry as the temperature crosses T^* value of about 85·3 K.

The $\chi'_H(T)$ data at fixed H identify the irreversibility temperature [21–24] as the onset-temperature (designated as $T_r^{\mathrm{DPE}}(H)$) of differential paramagnetic effect (DPE) (e.g., see figures 6 and 7).

From $\chi''_H(T)$ data in different H (e.g., see figure 7), one can pick out the temperature values of peak positions ($T^{\chi''}_{\mathrm{peak}}(H)$).

2.4 *Isothermal in-phase and out-of-phase AC susceptibility*

χ'_H and χ''_H data as a function of superposed DC field (up to 300 Oe) have been recorded at several temperature values lying between 60 K and T_c.

The χ'_H vs H plots at different T identify the irreversibility field as the field values of the onset of DPE (designated as $H_r^{\mathrm{DPE}}(T)$, e.g see figure 8), whereas χ''_H vs H data are used to pick the field values of peak positions in χ''_H (designated as $H^{\chi''}_{\mathrm{peak}}(T)$, e.g. see figure 9). In principle, the isothermal χ'_H and χ''_H data can also be used to confirm the 'temperature and field' values across which the qualitative changes in the shape of DC magnetization hysteresis loops occur. The detailed results and the analysis of AC susceptibility studies shall be reported elsewhere [24]. Only a part of AC susceptibility data necessary for the present objective have been included here.

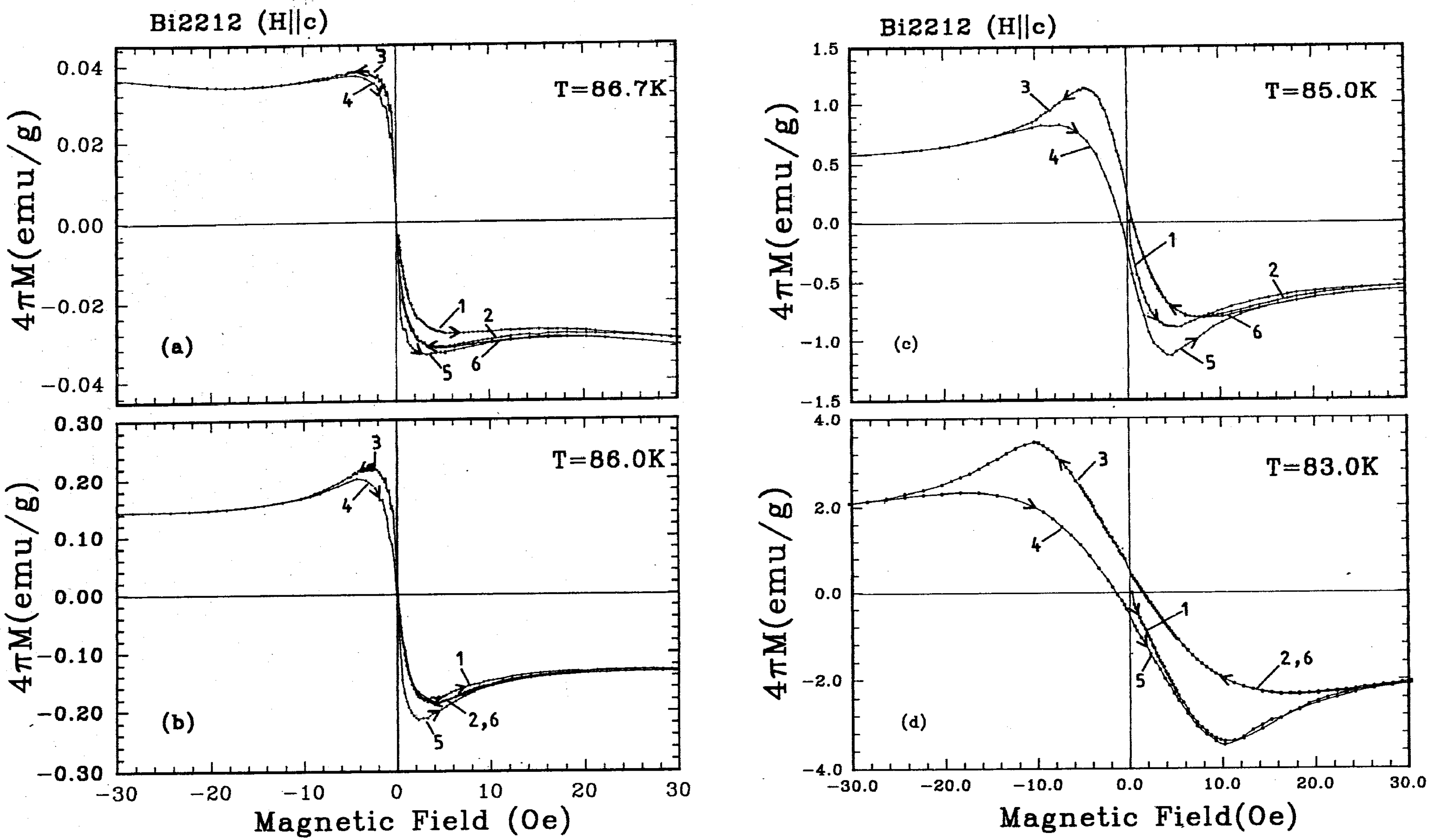

Figure 5. Comparison of the details of the magnetization hysteresis curves obtained at (a) 86·7 K, (b) 86 K, (c) 85 K and (d) 83 K, respectively. Each time the sample was cooled down to a given temperature in a residual field of about 1 mOe. Each of the complete cycles is as follows: 0 to + 30 Oe (1), + 30 Oe to 0 Oe (2), 0 to − 30 Oe (3), − 30 Oe to 0 (4), 0 to + 30 Oe (5) and 30 Oe to 0(6). Note the difference between curves 1 and 2 as the temperature changes from 86·7 K to 83 K. The irreversibility field as defined by merger of curves 3 and 4 would appear to decrease as T changes from 83 K to 86·7 K. However, the differences between curves 1 and 2 may imply otherwise.

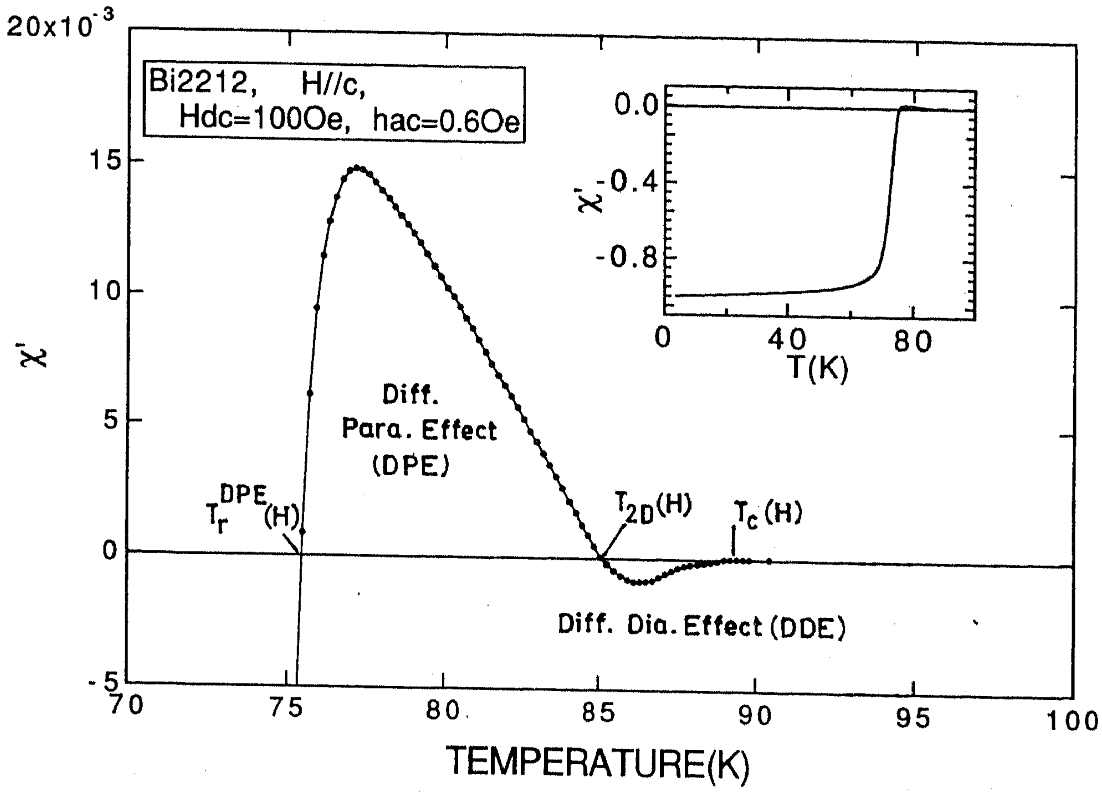

Figure 6. The in-phase AC susceptibility $\chi'(T)$ vs T near T_c in $H(\|c) = 100$ Oe. The onset and offset of positive response in $\chi'(T)$ identify the values of $T_r^{DPE}(H)$ and $T_{2D}(H)$, respectively. The amplitude of AC field was 0·6 Oe and frequency = 119 Hz.

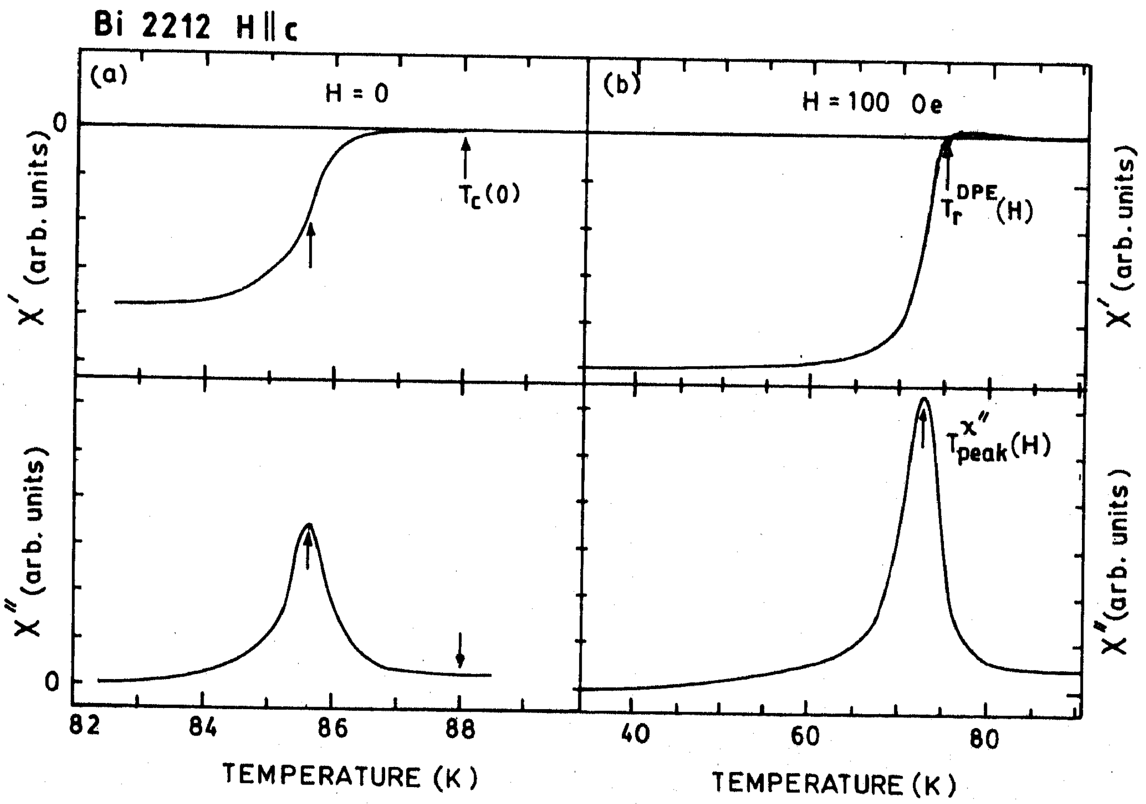

Figure 7. Comparison of in-phase and out-of-phase AC susceptibility in $H = 0$ and 100 Oe in Bi2212 sample.

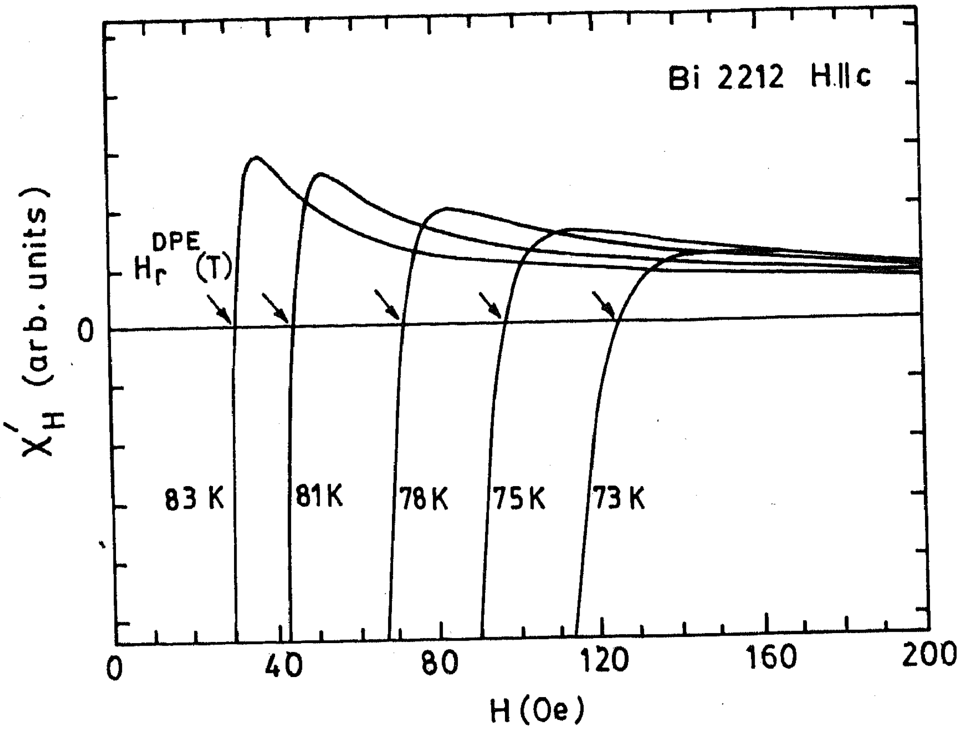

Figure 8. The in-phase AC susceptibility vs H data at different temperatures. The arrows identify the values of irreversibility field at different temperatures. These data have been obtained at a frequency of 119 Hz, the irreversibility fields are seen to increase as frequency increases (data not shown here).

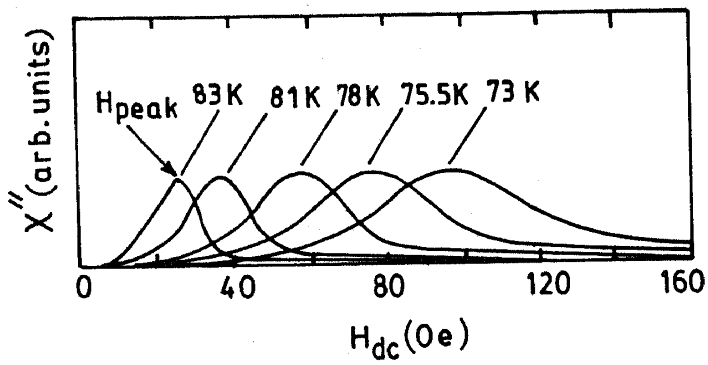

Figure 9. The out-of-phase AC susceptibility data vs H at different temperatures.

3. Results and discussion

3.1 *Dimensional crossover effect*

Figure 1 shows M vs T curves for $H = 50$, 100 and 300 Oe, respectively. It is apparent that these curves intersect at a temperature of $85{\cdot}3 \pm 0{\cdot}1$ K. At any temperature value below this, dM/dH values are positive whereas above this the dM/dH values cross over to the negative regime. The positive values imply a behaviour usually seen in the mixed state of any type-II superconductor. Figure 4 shows magnetization hysteresis loops in $\pm$ 300 Oe, at temperature values below and above the intersection temperature of 85·3 K. At 83 K, the hysteresis loop (figure 4(a)) has the shape and features normally ascribable to a hard type-II superconductor, whereas at 86·7 K, it is tempting to view the observed hysteresis curve (figure 4(b)) as the superposition of a minor hysteresis loop (limited to low field values) on the continuously increasing irreversible diamagnetic response extending to high field values. The isothermal magnetization data of figure 4 confirm that dM/dH values for $H \geq 30$ Oe are positive and negative at 83 K and 86·7 K, respectively. As remarked in the introduction, the response with negative dM/dH values is being perceived to arise from fluctuation effects confined to individual quasi-2D copper oxide planes. Taking a clue from well documented 2-component behaviour (ascribed to intragrain and intergranular regions) observed in magnetization hysteresis curves of ceramic samples of HTSC [25–28] or fine multi-filament wires of alloy superconductors (see, for instance, discussion in § 4 of [29] or [30]), we identify the hysteresis loop confined to lower field values with the response from interplanar linkages. At any temperature, as the applied field increases the interplanar links are expected to get severed resulting in response from only decoupled planes.

Two more noteworthy differences in the hysteresis loops displayed in figures 4(a) and (b) are, (i) the change (i.e., increase) in the irreversibility field value on increasing the temperature from 83 K to 86·7 K and (ii) the appearance of marked asymmetry (i.e., the absence of time reversal symmetry) in the hysteresis loop at 86·7 K. Both these changes are of considerable significance. The irreversibility in a hard type-II superconductor correlates with the macroscopic currents that flow in a hard type-II superconductor. Since the critical current density J_c decreases with increase of temperature or the field, the irreversibility field values are expected to decrease as T increases. The absence of time reversal symmetry has hitherto been noted in ceramic samples of $YBa_2Cu_3O_7$ by Roy *et al* [30] and is ascribed by them to the interplay between the diamagnetic responses from intragrain and intergranular regions. The explanation uses an idea due to Evetts and Glowacki [31], that in sintered samples, the intra-grain diamagnetic response M_g in an applied field H_o provides an additional field $H_g(M_g)$ in the intergrain region. The additional field value $H_g(M_g)$, thus, depends on the history of application of H_o via the history dependent magnetization value of the grain in field H_o.

The inset of figure 1 depicts warm-up M vs T curve obtained after rapidly cooling the sample in nominal residual field ($\approx 0·3$ Oe) of the magnet. The nominal zero field transition temperature $T_c(0)$, identified with the onset of curvature (w.r.t. base line) in the diamagnetic response, is reckoned as 88 K in our sample of Bi2212. The mid-point of sharp fall in diamagnetic response is located just above 86 K. The transition width of this specimen is very narrow and it correlates well with similar data reported in the literature in good quality samples of the highly anisotropic layered compounds (e.g., see figure 4 of [7]). The magnetization data contained in figures 1 and 4 motivates us to view the observed changes (e.g., at 85·3 K in figure 1) in magnetization behaviour before reaching the $T_c(0)$ value as the consequence of dimensional cross-over effect. So long as the inter-planar coupling is strong, the specimen has a response identifiable with a 3D type-II superconductor. The weakening of the coupling results in a quasi-two component behaviour with interesting interplay between the two components.

3.2 *Irreversibility line*

The fact that the flux pinning has to vanish at a temperature below $T_c(H)$ in a hard type-II superconductor inevitably leads to the existence of pinning-depinning transition (or an irreversibility line) above which the value of macroscopic critical current density J_c is zero (see schematic description in § 3 of [9]). Ever since the determination of an irreversibility line in a specimen of HTSC by Müller *et al* [32], a variety of techniques and procedures have been employed to determine quasi-irreversibility lines in HTSC as well as in conventional alloy superconductors. It is an experimental reality that irreversibility lines determined by different methods in the same sample do not overlap. However, all the irreversibility lines concur in conveying the message that values of irreversibility field/irreversibility temperature decrease as temperature/field increases. A systematic study in the samples of niobium (an elemental superconductor with $T_c = 9{\cdot}3$ K) have indicated a hierarchy amongst the irreversibility lines determined from different DC and AC magnetization measurements [9, 21, 22]. A complete understanding of this hierarchy is still to be attained. In any case, we find the existence of similar hierarchy in the quasi-irreversibility lines determined from different measurements in our single crystal sample of Bi2212.

Figures 2, 3, 6, 7, 8 and 9 exemplify the determination of values of temperature and field, in T-dependent and isothermal DC and AC magnetization measurements, which are considered to mark the transition from irreversible to reversible response. Figure 10 summarizes all the data obtained on irreversibility temperatures and fields for $T > 70$ K and $H < 300$ Oe. The hierarchy that $H_r^{\mathrm{DPE}}(T)$ line defines the upper boundary and $T_r^c(H)$ line is the lower limit of the irreversible to reversible transition region is clearly evident in figure 10. Clem and Hao [19] have argued that merger temperatures $T_r^w(H)$ at which M_{ZFC} data become indistinguishable from M_{FCW} data do not identify the real boundary of reversible to irreversible transition. $T_r^w(H)$ data points are observed to lie below the corresponding $T_r^c(H)$ data points in figure 10.

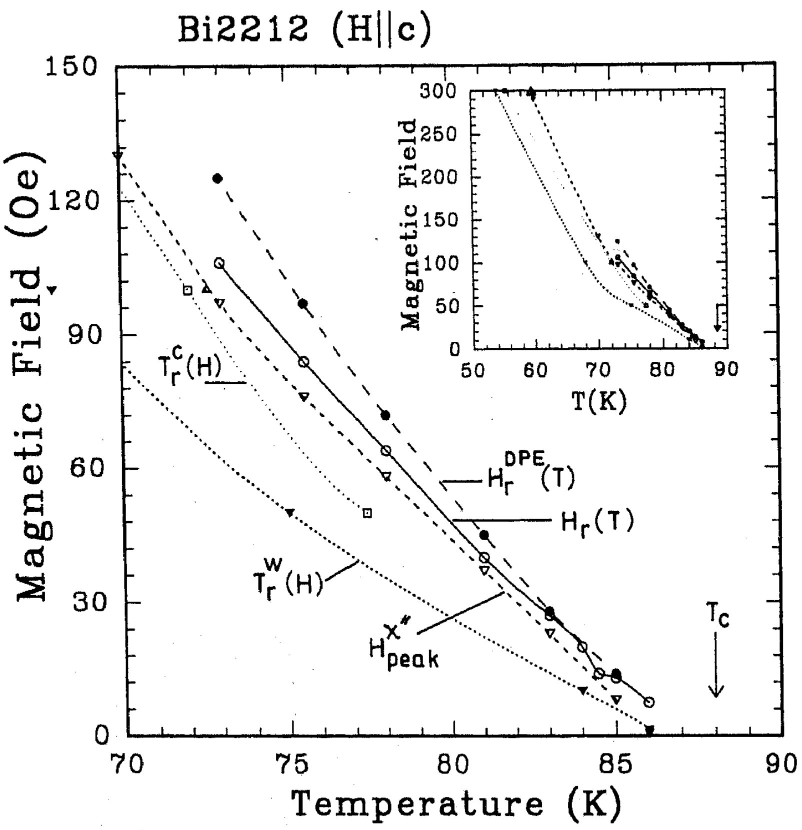

Figure 10. Comparison of quasi-irreversibility lines near T_c obtained by different procedures and techniques. The inset shows such data over an extended temperature range.

Also, the peaks in χ'' data are considered to identify the line that should lie below the true reversible to irreversible boundary [9]. However, $H^{\chi''}_{\rm peak}(T)$ line is observed to lie above the $T^c_r(H)$ line. This in turn implies that $T^c_r(H)$ line is located sufficiently below the actual irreversible to reversible boundary.

For our purpose it suffices to reckon here that $H_r(T)$ and $H_r^{\rm DPE}(T)$ lines would nearly linearly extrapolate to zero at a temperature value which is definitely smaller than the nominal $T_c(0)$ value of the sample. We have not included data points above 86 K in figure 10 because a little below this temperature the qualitative changes that occur in the irreversibility behaviour at low fields start to make the determined values of $H_r(T)$ depend on the maximum field to which the sample is exposed. This point is exemplified by a comparison of two hysteresis loops at 86·7 K displayed in figures 4(b) and 5(a), respectively. In figure 4(b), the irreversibility is seen to persist up to maximum field of 300 Oe, whereas, from figure 5(a), the irreversibility field value may be picked out to be equal to or less than 30 Oe. A similar comparison of hysteresis loops at 83 K in figures 4(a) and 5(d) shows that H_r values in the two figures are nearly the same.

3.3 *Two-component behaviour*

As stated earlier, the asymmetry in the hysteresis loop at 86·7 K (figure 4(b)) is a signature of interplay between the diamagnetism of two components. Several other interesting thermomagnetic history effects which are a consequence of interplay between the responses from two components in sintered samples of HTSC have been documented in the literature [33–36]. Two of these are the observed increase in transport $J_c(H)$ (i) on reversing the field, and (ii) on field cooling the sample in the given H. Both these are ascribable to additional field values arising from intragrain diamagnetism. The $J_c(H)$ values during the first ramp-up of applied field (after initially cooling the sample in zero field) are lower because the effective field (at a given applied H) experienced by intergrain regions during the first forward run is larger than that during the reversal of the applied field or after cooling the sample in a given applied field. We see somewhat analogous effects in $M-H$ data in the temperature region of dimensional crossover in our single crystal sample of Bi2212.

Figure 5(a) shows the details of the $M-H$ curves obtained at 86·7 K after ensuring the cool-down of the sample in a field of ≈ 1 mOe. The complete cycle is as follows: 0 to ± 30 Oe (curve 1), $+30$ to 0 Oe (curve 2), 0 to -30 Oe (curve 3), -30 to 0 Oe (curve 4), 0 to $+30$ Oe (curve 5) and $+30$ to 0 Oe (curve 6). It may be noted that curve 2 lies below curve 1, which was completely unexpected. This implies that diamagnetic values are larger on reversal of field from $+30$ Oe to 0 Oe as compared to those during the virgin ZFC run (0 to $+30$ Oe). On further cycling the field between ∓ 30 Oe, the hysteresis loop comprises of curves 3, 4, 5 and 6. It may also be noted that the difference between curves 1 and 2 is larger than that between curves 3 and 4 or curves 5 and 6. The irreversibility field value determinable from curves 3 and 4 or 5 and 6 is much smaller than that evident from curves 1 and 2. On subsequent repeated cycling the field between ± 30 Oe, the hysteresis loop would consist of curves equivalent to curves 3, 4, 5 and 6. We further find that, if the sample gets cooled down to 86·7 K in a nominal residual field of the magnet ($\approx 0{\cdot}3$ Oe), the hysteresis loop nearly comprises of curves 5, 6, 3 and 4, and no anomalous increase in diamagnetic response is evident on reversal of field after the first forward run. This point is well illustrated by a comparison between the two sets of data contained in figure 11. In

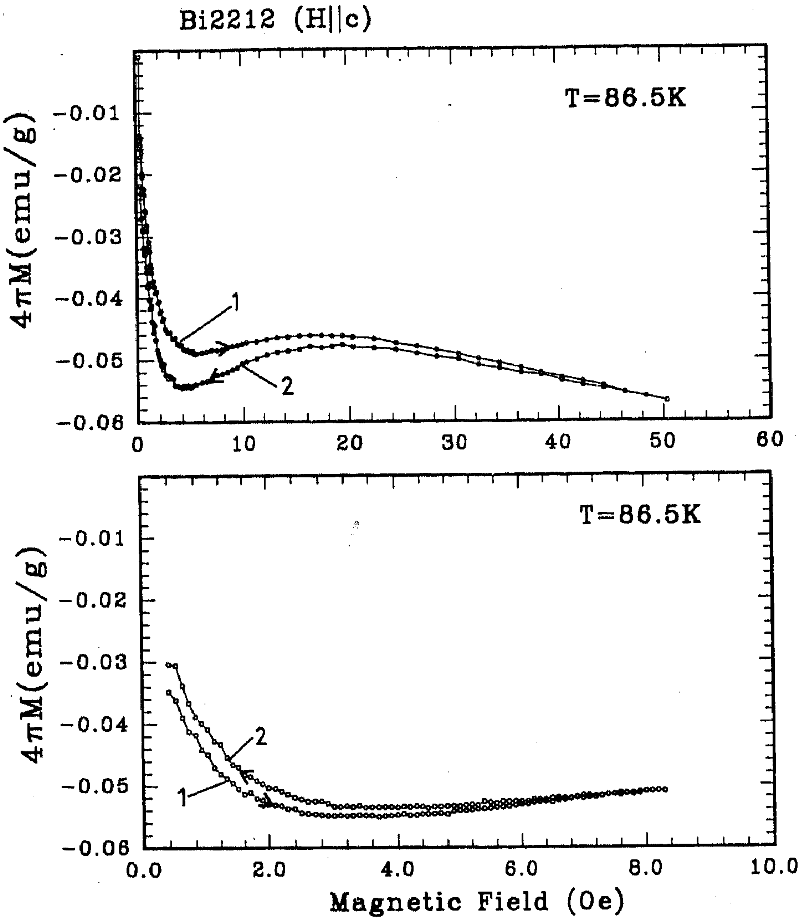

Figure 11. Comparison of magnetization curves obtained at 86·5 K in two different initial conditions. In figure 11(a), the sample has been cooled-down to 86·5 K in a residual field of 1 mOe, whereas the curves in figure 11(b) correspond to an initial cool-down to the same temperature in a nominal residual field of the magnet which was measured to be about 0·4 Oe.

figure 11(a), we have depicted $M-H$ curves up to 50 Oe after cool down of the sample in a field of 1 mOe, whereas figure 11(b) shows $M-H$ curves obtained after cool down of the sample to 86·5 K in nominal zero field ($\approx 0{\cdot}3$ Oe). In any case, the complete hysteresis loop obtained on repeated cycling the field between $\pm H_{max}$ is independent of the initial thermomagnetic history of the specimen. The complete loop comprises the asymmetry property reflecting the interplay between the diamagnetic responses from the two components.

A careful examination of the details of the magnetization curves obtained at 86 K, 85 K and 83 K (see, figures 5(b) to 5(d)) shows that vivid effects which reflect the interplay between the two components become less spectacular as the temperature is lowered from 86·7 K to 83 K. As remarked earlier, the irreversibility field value at 83 K is found to be nearly independent of the thermomagnetic history and the maximum field values between which the sample is cycled. It may be mentioned here that $H_r(T)$ values at 85 K and 86 K contained in figure 10 correspond to those obtained by ramping the field up to a maximum of 20 Oe.

3.4 *Dimensional crossover line*

The experimental data and their analysis presented so far implies that the imprints of two component response and the dimensional crossover effect are present before reaching $T_c(0)$ value. Further, if we accept the argument that response from 2D planes has the characteristic that dM/dH is always negative, we can look for this signature in the isothermal $M-H$ curves. For instance, it may be noted from $M-H$ curves at 86·7 K and 86·5 K (see, figure 4(b), 5(a) and 11(a)) that after the initial peak in magnetization located between 3 and 4 Oe, the magnetization curves turn over again at about 15 Oe and 20 Oe, respectively and above these field values the diamagnetic values increase as H increases (i.e., dM/dH is negative). We may take the second field value at which $dM/dH = 0$ to signify the onset of dominance of 2D like response and designate this field as H_{2D}. Figure 12 shows $M-H$ curves obtained at 83·5 K and 85·5 K, respectively. In this figure, the H_{2D} value at 85·5 K has been identified, whereas at 83·5 K, dM/dH values continue to remain positive up to 10 kOe (highest field up to which data have been recorded) and as such H_{2D} value cannot be ascertained at 83·5 K. We may remark here that some of H_{2D} values at $T < 85$ K evident in reference 13 are erroneous due to an inadvertent omission of subtraction of sample holder diamagnetism.

Figures 13(a) and (b) show the plots of H_{2D} values vs temperature on linear as well as semi-log scales. It is apparent that H_{2D} values show a slow increase as temperature is progressively lowered down from 86·7 K to 85·75 K. The fast rise in H_{2D} values sets in only below 85·5 K. The temperature interval between the drastic change in dH_{2D}/dT value (the dotted line in figures 13(a) and (b)) and the $T_c(0)$ value nominally

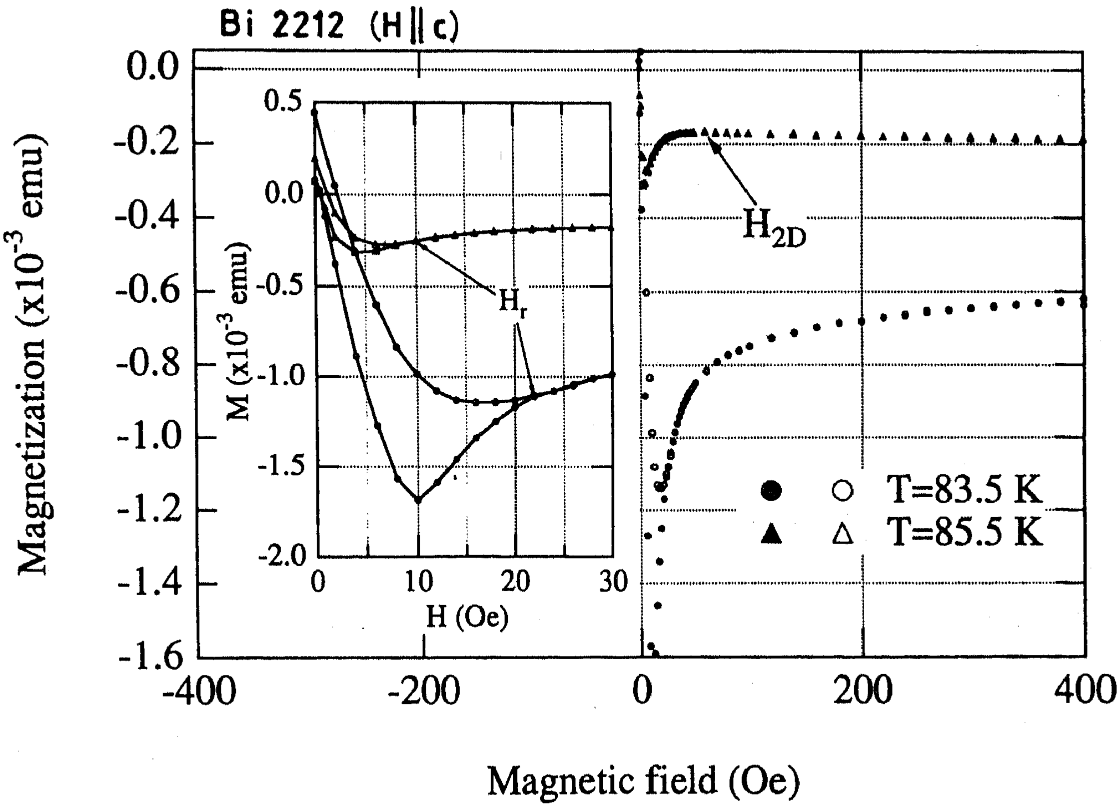

Figure 12. Isothermal M vs H curves at 83·5 K and 85·5 K, respectively.

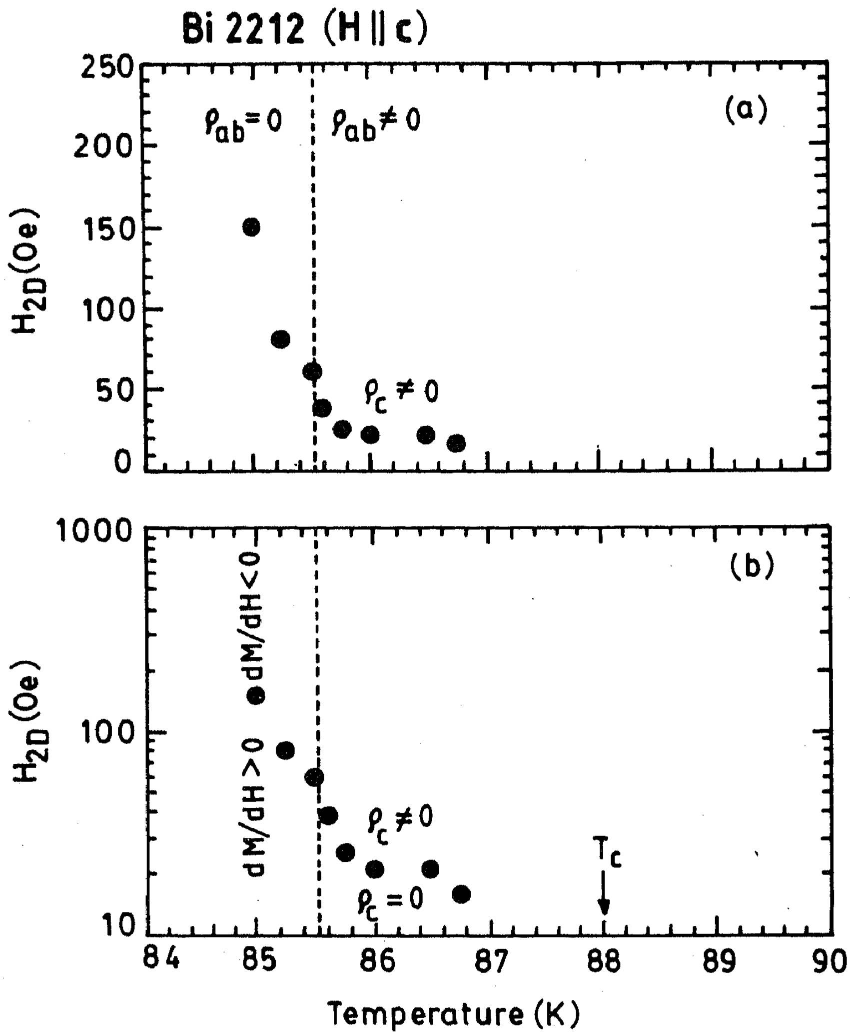

Figure 13. Plot of H_{2D} vs T values on (a) linear and (b) semilog scales. The temperature at which rapid rise in H_{2D} values sets in can be easily seen in these data.

marks the dimensional crossover region. From (H_{2D}, T_{2D}) line of figure 13(b), we note that H_{2D} value at the temperature of intersection ($= 85{\cdot}3 \pm 0{\cdot}1$ K) of $M - T$ curves (depicted in figure 1) is about 50 Oe. This immediately tempts us to identify T_{2D} values of figure 13 with T^* of BLK [6]. As remarked in the introduction section, in the framework of BLK treatment, T^* is the temperature at which $\mathrm{d}M/\mathrm{d}H = 0$ for $H > H_{cr}$ and above which $\mathrm{d}M/\mathrm{d}H$ values are negative. If our T_{2D} and T^* of BLK [6] are the same, it would imply that $M - T$ curves for all H would not intersect at the same T^*. As H values increase, the temperature-band of intersection of $M - T$ curves would expand towards the lower temperature. The overall shape of (H_{2D}, T_{2D}) line comprising of rapid build up of H_{2D} with T_{2D} in figure 13 indicates that the said temperature-band of intersection of $M - T$ curves is reasonably narrow for the range of laboratory field values across which $M - T$ curves are usually recorded by experimentalists (e.g., see figure 1 of BLK [6]). Recent high field ($1\,\mathrm{kOe} < H < 50\,\mathrm{kOe}$) $M - T$ curves (for $H \| c$) in our Bi2212 sample show that these curves intersect at $85{\cdot}5 \pm 0{\cdot}2$ K [37]).

BLK [6] have also talked of temperature T_s at which electrical resistance (and consequently J_c as well) and H_{c1} values vanish and above which electromagnetic response arises only from 2D planes. As found in § 3·2, the irreversibility lines of strongly coupled quasi-3D superconductor extrapolate to zero at a temperature larger than T^* but smaller than $T_c(0)$. We shall present our H_{c1} data in the next section. However, we wish to remark here that low field hysteritic behaviour attributed to interplanar links continues to persist up to $T_c(0)$, this implies that the imprints of two component behaviour probably survive up to $T_c(0)$. Lastly, it is not of place to recall here that BLK [6] have predicted that above T^*, the diamagnetic response increases logarithmically with field and Martinez *et al* [38] have sought to provide magnetization data from torque measurements in Bi2212 supporting this prediction. But, a cursory look at dM/dH values at $H > H_{2D}$ in our specimen of Bi2212 shows that diamagnetic values increase in a quasi-linear manner.

It is pertinent to also recall here an assertion recently made by Wan *et al* [39] on the basis of their electrical transport studies for $H \parallel c$ in Bi2212. They noted that the deduced values of resistivity (in zero applied field) within the planes (ρ_{ab}) go to zero at a temperature T_c^{ab} which is lower than the temperature T_c^c at which the resistivity across the planes ρ_c goes to zero. This led them to surmise that in the temperature interval $T_c^{ab} < T < T_c^c$ Josephson supercurrents flow across the resistive planes (which are presumably in the dissipative fluctuating superconducting state). It is tempting to correlate the temperature interval T_c^{ab} to T_c^c with the interval over which we observe spectacular manifestations of two component response in the $M-H$ data. Our identification of normal looking minor hysteresis loop confined to low field values with the component corresponding to interplanar links inherently implies the existence of supercurrents across the planes. We think that the temperature $T_c^{ab}(H=0)$ at which the resistivity within the planes goes to zero probably marks the transition from weakly coupled to strongly coupled response (from Josephson coupled quasi-2D planes) in the magnetization data. The (H_{2D}, T_{2D}) line of figure 13a appears to be made up of two parts corresponding to slower and faster increase in $H_{2D}(T)$ values. The temperature $T_c^{ab}(H=0)$ perhaps demarcates the two parts. In the nominal dimensional crossover region (temperatures above dotted line figures 13(a) and (b)), the (H_{2D}, T_{2D}) line is such that $\rho_c = 0$ below it but ρ_c values would become non-zero at somewhat above it. At temperatures lower than $T_c^{ab}(H=0)$, the (H_{2D}, T_{2D}) line is such that above this line the change is diamagnetic response (dM/dH values are negative) of the strongly coupled system is dominated by the response from quasi-2D planes. At $T < T_c^{ab}(0)$, the (H_{2D}, T_{2D}) line determined via $dM/dH = 0$ values does not bifurcate the regions of $\rho_c = 0$ and $\rho_c \neq 0$. The line demarcating the regions $\rho_{ab} = 0$ and $\rho_{ab} \neq 0$ probably lies below it whereas the line demarcating the region $\rho_c = 0$ and $\rho_c \neq 0$ would exist above it. These lebellings have been schematically depicted in figures 13(a) and (b).

3.5 *Paramagnetic Meissner effect*

Braunisch *et al* [40] reported observation of field independent positive magnetization value (designated as Paramagnetic Meissner Effect PME) in certain ceramic samples of Bi2212 on field cooling in very low field values (< 100 mOe). Their initial observations have since been confirmed by them [40] as well as others [41] in several other varieties of ceramic samples of Bismuth cuprates. The PME is believed [40, 41] to be a consequence of the occurrence of spontaneous orbital (superconducting) currents in certain loops containing weak links (the so called "π" junctions). For our purpose, it is enough to consider that PME is an attribute of the existence of weak links of certain strength in loops of certain geometrical shape and size. In view of the observation that the single crystal specimen of Bi2212 has two component response, like, the ceramic samples, in the temperature interval of dimensional crossover, we may expect to see the signature of PME in our single crystal sample as well.

One way to look for presence of PME is to measure FC magnetization in small fields at different temperatures. We measured FC magnetization in a field value of ≈ 1 mOe in the temperature range 83 K to 87·7 K. The positive magnetization signals with $4\pi M(\text{emu/cm}^3) \approx 50$ mOe are observed at $T > 85$ K. This magnitude compares

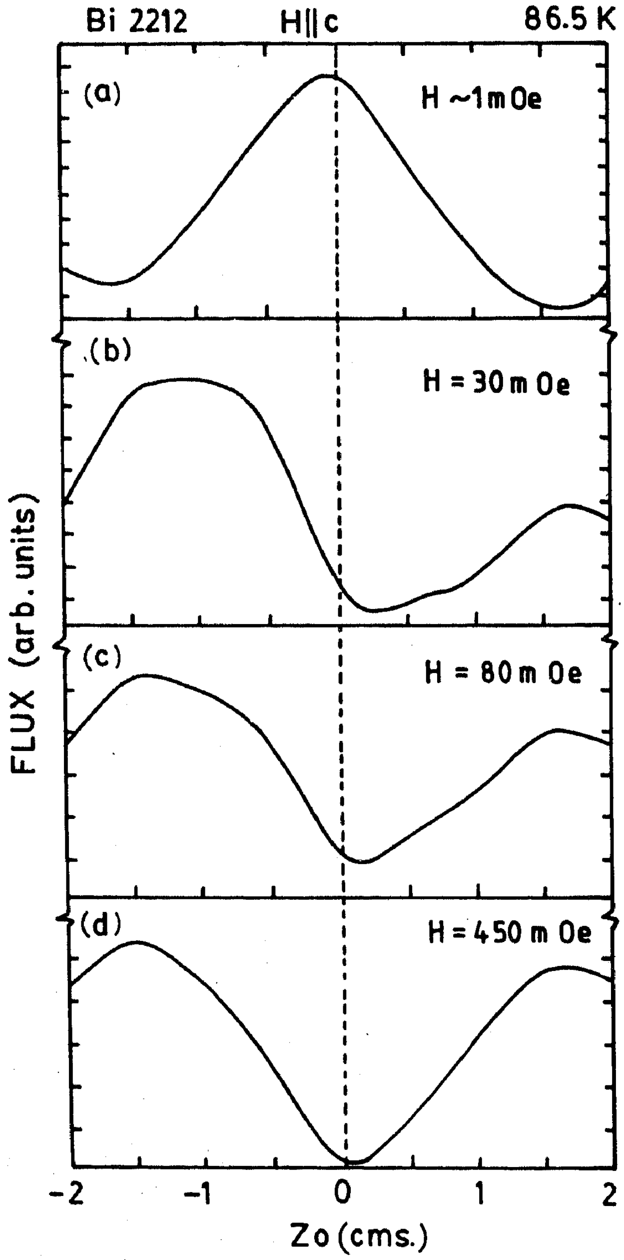

Figure 14. Flux detected as a function of distance from the centre of "second derivative" coil array of Quantum Design SQUID magnetometer at 86·5 K for $H \parallel c$ in Bi2212 at different values of field. The sample is initially cooled down to 86·5 K in a field of ≈ 1 mOe and flux profile is recorded over 4 cm scan (figure 14(a)). The field is then ramped up. Figures 14(b), (c) and (d) show the flux profiles in nominal field values of 30 mOe, 80 mOe and 450 mOe.

favourably with the values of such signals reported earlier [40–42]. After cooling the sample in $H \approx 1$ mOe, a further increase in applied field values up to a few hundered mOe pushes the sample magnetization towards negative values. At such fields, the diamagnetism due to surface screening current starts to dominate over positive magnetization from spontaneous orbital currents.

The magnet power supply of Quantum Design SQUID magnetometer ($MPMS_2$ system) is such that in its usual mode of operation, it does not reproducibly generate low filed values ($H < 300$ mOe). Hence, we are not confident of absolute values of field at $H < 0{\cdot}3$ Oe. In any case, we made a record of flux profiles as the applied field is slowly ramped up from nominal 10 mOe to 500 mOe at 86·5 K. Figure 14 shows flux profiles recorded in nominal field values of 1 mOe, 30 mOe, 80 mOe and 450 mOe. The profiles in 1 mOe (figure 14(a)) and in 450 mOe (figure 14(d)) are essentially dipolar like [9] with magnetization ($4\pi M$) values of $+8 \times 10^{-3}$ emu/g and $-1{\cdot}4 \times 10^{-2}$ emu/g, respectively. The profiles in figures 14(b) and 14(c) show large deviation from dipolar like response, thereby implying that the magnetization signal originating from states of superposition of spontaneous orbital currents and the surface screening current in low field values have significant contributions from quadrupolar moment [9, 43]. In view of our past experience with the analysis of deviations from dipolar like response in superconducting samples [43], we can confidently state that the profiles in figures 14(b) and (c) are neither an artifact of any inhomogeneity in the field along the axis of the magnet, nor the smallness of the absolute value of the magnetization signal.

3.6 *Critical fields*

It is now widely appreciated that the determination of precise values of lower critical field in HTSC is a daunting task for experimentalists (e.g., see remarks in [38] and description in § 8·2 of [9]). The simplest procedure of picking out field values at which isothermal M vs H curve deviates from linearity provides an upper limit on H_{c1} values, when the pinning is strong. We have stuck to this procedure only to ascertain values of $(1 - N_{\|})H_{c1}$ in our sample of Bi2212, where $N_{\|}$ is the demagnetization factor for $H \| c$. It suffices for our limited purpose here as we are interested in knowing the behaviour of H_{c1} vs T near T_c, where the pinning effects are not anticipated to be very strong.

Figure 15 shows a plot of values of $(1 - N_{\|})H_{c1}$ for $T > 70$ K, determined via nominal deviation from linearity criterion. It is apparent that $H_{c1}(H \| c)$ values linearly extrapolate to zero between 86·5 and 87 K. We are not confident of the occurrence of any drastic change (i.e., increase) in value of the slope dH_{c1}/dT just before the collapse of H_{c1} in our data. As stated in § 1, such a behaviour has been noted by Brawner *et al* [8] in Bi2212. In the usual mode of operation the Quantum Design SQUID magnetometer, even with the Fluxgate option facility, does not provide accurate values of deviation from linearity field lying in the range $\leqslant 1$ Oe. The value of $(1 - N_{\|})H_{c1}$ falls to $2{\cdot}0 \pm 0{\cdot}5$ Oe at 84 K in our data and above this temperature, our error bars are so large that they preclude any inference regarding the increase in values of the slope dH_{c1}/dT between 84 K and 87 K. Even in the absence of this effect in our data, it is satisfying to note that the $H_{c1}(T)$ values extrapolate to zero at a temperature value $\leqslant 87$ K. This temperature appears to be not very different from the temperature region of steep increase in diamagnetic response in the lowest field (see, inset of figure 1). This leads us to surmise that the latter identifies the

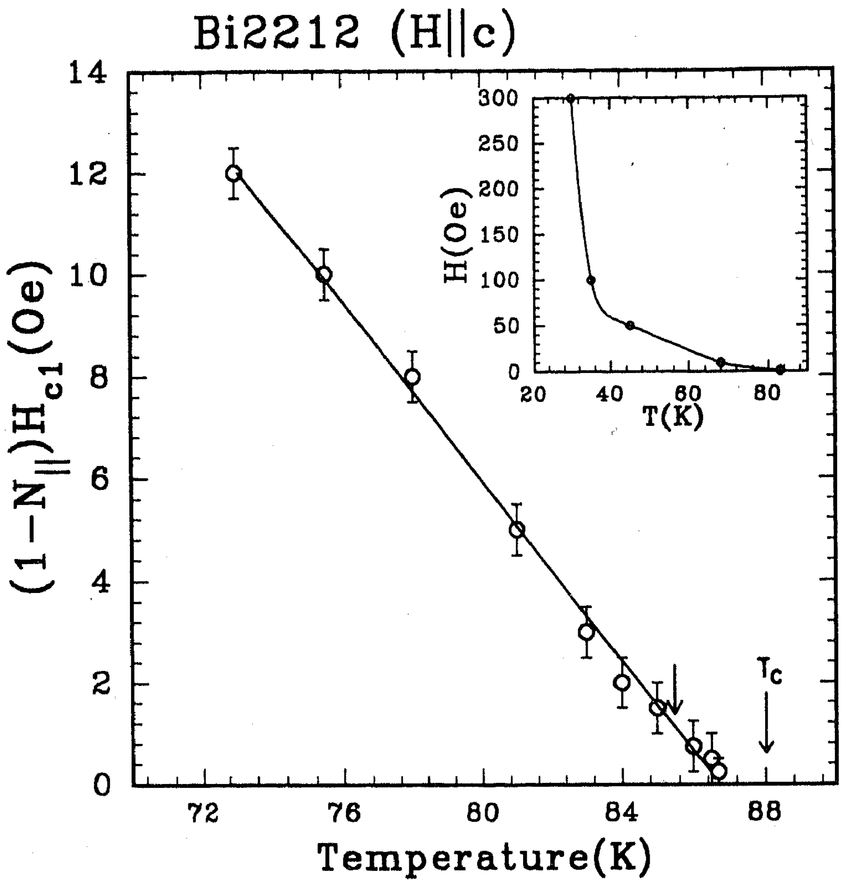

Figure 15. Plot of $(1-N_{\|})H_{c1}$ vs temperature for $H \| c$ in Bi2212. $N_{\|}$ is the demagnetization factor for $H \| c$. The values of $(1-N_{\|})H_{c1}$ have been determined via deviation from linearity criterion. The inset shows the field dependence of temperatures at which M_{FCC}/M_{FCW1} curves start to bifurcate or M_{FCW2} curve starts to show a downward trend.

temperature region of the occurrence of rapid change in the strength of the interplanar coupling (i.e., thermally induced dimensional crossover) in zero field. We feel that above 86 K, the strength of interplanar coupling stands considerably reduced, but, as stated earlier, some imprints of the two component response appear to survive up to $T_c(0)$.

The inset of figure 15 shows the field dependence of temperature values at which M_{FCW1} data start to differ from M_{FCC} data and the M_{FCW2} data start to show a downward trend (see figure 2 for data in $H = 100$ Oe). If we identify the $H - T$ curve in the inset of figure 15 with $H_{c1}(T)$ as per the suggestion of Clem and Hao [19], these limiting values of $H_{c1}(T)$ are somewhat smaller than the corresponding values determined from deviation from linearity criterion. Further, it may be noted that the temperature dependence of the curve in the inset of figure 15 shows a steep rise at $T < 40$ K. Such a behaviour has often been noted in temperature dependence of H_{c1} in HTSC. Burlachkov *et al* [44] have recently pointed out that the steep rise in H_{c1} at lower temperatures is caused by an additional surface barrier effect which becomes effective at high pinning strength. They argue that if a correct analysis of the data is made, the actual H_{c1} values would show the usual saturation effect in the lower temperature region.

The $H - T$ curve in the inset of figure 15 identifies the boundary at which M_{FCW1}

and M_{FCW2} data (as in figure 2) start to show the so-called negative peak effect, i.e., increase in diamagnetic response with increase in temperature. It may be mentioned that the negative peak effect stands enhanced in the warm-up data recorded after rapid cool down in field (cf. M_{FCW2} and M_{FCW1} data sets in figure 2). The negative peak feature has earlier been noted in ceramic samples of HTSC or specially prepared porous polycrystalline samples of an alloy superconductor Nb_3Sn [18]. Huyn [18] has sought to explain this effect in terms of interplay between responses from two components (i.e., grains and intergranular regions or weak links at grain boundary contacts in HTSC or alloy superconductor, respectively) in conjunction with thermal hysteresis of diamagnetism of first component, viz., grains. The central idea is that during cool-down, the flux escaping from the grain region gets trapped in intergranular regions. On warm-up, the flux movement inside and outside the grain regions is such that more net flux escapes from the entire sample resulting in enhanced diamagnetism. The two facts, that (i) the field cool magnetization value after rapid cool-down (the first data point of the data set marked FCW2 in figure 2) differs from the corresponding value after slow cool-down and (ii) the negative peak effect, can be qualitatively understood in terms of a scenario described by Huyn [18], which also implicitly confirms the existence of interplay between the responses from two components in a single crystal of Bi2212. When the interplanar coupling is strong, the usual electromagnetic response of the coupled system is indistinguishable from that of a 3D type-II superconductor. However, coupling manifests itself via the effects, like the two pointed out above. As stated in the introduction, Kleiner *et al* [4] have been the first to directly observe dc and ac Josephson effects at 4·2 K in Bi2212. More recently Nakamura *et al* [45] have described another manifestation of the coupling between planes via the drastic change in the initial magnetization curve at 10 K, when the applied field stood slightly misaligned w.r.t. a–b plane.

The assignment of upper critical field for the coupled 2D superconductor poses new issues. It is generally accepted that there is no well defined H_{c2} line for the highly anisotropic HTSC system, instead, there perhaps exists only a dimensional crossover region (e.g., see figure 3(b) of [1]). Kadowaki [2] has been amongst the firsts to point out that T-dependent magnetization data cannot be used to identify the said H_{c2} values as the onset of diamagnetism in these systems is gradual, rounded and somewhat obscure and the conventional analysis leads to an unusual inference that $T_c(H)$ increases with H [3].

When we consider the electromagnetic response of a layered HTSC in terms of a 2-component system, the above remarks regarding the absence of any signature and the difficulty in defining H_{c2} would apply to the component corresponding to 2D planes. The other component attributed to interplanar links and identified with the hysteresis loop confined to low field values does possess a feature which can be used to pick out a limiting value of the upper critical field of that component. In the temperature interval of dimensional crossover, H_{2D} is physically the field above which all interplanar links may be considered broken. Thus, H_{2D} at any T, as identified with the field value at which $dM/dH = 0$ in isothermal DC magnetization curve, defines a (lower) limit, which must correlate with the upper critical field H_{c2} of the interplanar component.

In principle, one can see a signature of H_{2D} via differential paramagnetic effect (DPE) in the in-phase AC susceptibility data as well. Figure 16 shows a schematic plot of isothermal DC magnetization and χ'_H vs H curves. The first field value at which χ'_H crosses from negative to positive values identifies $H_r(T)$, whereas the second

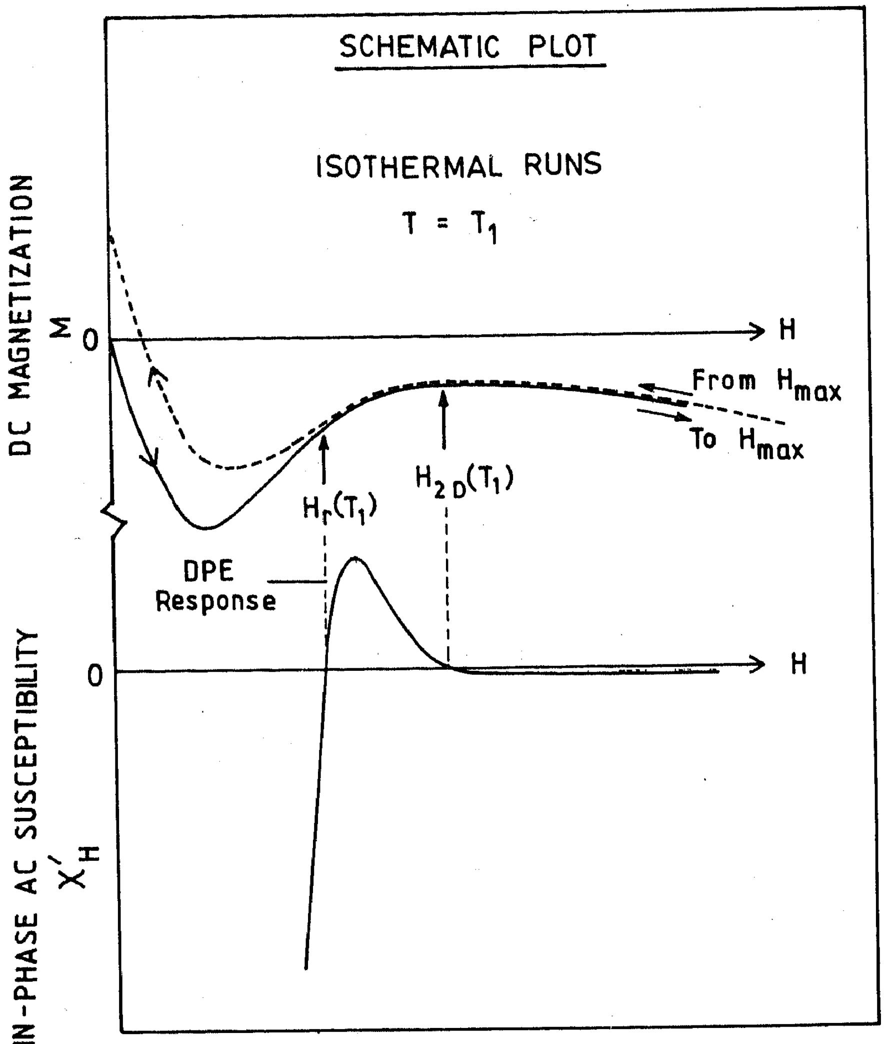

Figure 16. Schematic plot of isothermal M vs H and χ' vs H behaviour expected in a situation which encompasses dimensional crossover. The signatures of irreversibility field $H_r(T)$ and the dimensional crossover field $H_{2D}(T)$ are identified.

crossover, after which χ'_H saturates to a very small negative value, should identify the H_{2D} value. The second crossover is difficult to unambiguously pick out from the isothermal χ'_H data. However, it is possible to pick out the analog of second crossover corresponding to $T_{2D}(H)$ value in very carefully recorded temperature dependent $\chi'_H(H)$ runs [cf. figure 6]. Figure 6 shows DPE peak in χ' vs T data in $H = 100$ Oe. We feel that in T-dependent measurements, the temperature at which DPE signal crosses the baseline second time identifies $T_{2D}(H)$. Above $T_{2D}(H)$, the differential susceptibility (dM/dH) is negative and we choose to call the tiny negative peak as *Differential Diamagnetic Effect* (DDE). The DDE is a hallmark of dominance of diamagnetic response from decoupled 2D planes. The temperature at which DDE signal merges into base line identifies the so-called $T_c(H)$ value which correlates well with the similar value evident in DC magnetization data of figure 1. It may be appreciated that DPE and DDE responses are so small that DPE and DDE signals being reported in the present paper would rank amongst the first such observations in any single crystal specimen of HTSC. The presence of DPE and DDE type peaks in polycrystalline samples of Bi2212 can be noted in some of the data of Couach and Khoder [23]. Kadowaki *et al* [46] have also recently reported the field dependence of the temperature values (designated as $H_{c2}(T)$ by them) at which DPE signals merge into the base line in their single crystal sample of Bi2212 for $H \parallel c$. They find a steep fall in "$T_c(H)$" values (as H increases) across a narrow temperature region lying below $T_c(0)$. We feel that their "H_{c2}" vs T behaviour echoes the H_{2D} vs T dependence of our sample (figure 13) in the temperature interval of dimensional crossover. They have not succeeded in observing DDE peaks in their data.

If we examine the hysteresis loop at 86·7 K in figure 4(b) in the light of above discussion, we may note that irreversible behaviour continues up to field values much larger than the identified H_{2D} at this temperature. We find that the hysteritic behaviour asymptotically vanishes at $H \approx 1$ kOe (data not shown here). We speculate that the field value at which the irreversibility in diamagnetic response disappears may play the role of "H_{c2}" for the component representing the response from decoupled 2D planes.

3.7 *Irreversible behaviour near T_c at low fields*

The occurrence of dimensional crossover and the interplay between diamagnetism of two components bestows this system with anomalous looking features just below T_c in temperature dependent DC magnetization measurements. For instance, figure 17 displays M_{ZFC}, M_{FCC} and M_{FCW} curves obtained in 1·3 and 10 Oe in our sample of Bi2212. We note that whereas M_{FCW} curves merge into the respective M_{ZFC} curves, and the M_{FCC} curve do not do so. M_{FCC} curves cross the respective M_{ZFC} curves; above the crossover temperature, the M_{FCC} data points lie below the M_{ZFC} data. The anomaly that $M_{FCC}(T) > M_{ZFC}(T)$ is somewhat analogous to the observation that diamagnetic response during the reverse cycle is larger than that during the virgin ZFC run at 86·7 K (cf. curves 1 and 2 in figure 5(a)). Two interrelated inferences which further emerge from the observed anomalous behaviour are as follows: (i) Irreversible behaviour at low fields appears to survive up to T_c and (ii) $T_r^c(H)$ as defined by merger of M_{FCC} data into M_{ZFC} data is not relevant at small H.

We do not see anomalous behaviour in temperature dependent magnetization data at $H = 50$ Oe for which $T_r^c(H) = 77{\cdot}5$ K and $T_r^w = 75$ K. The (minimum) limiting value of the field in which we shall not observe anomalies in temperature dependent runs on our sample lies between 10 and 50 Oe. This leads us to surmise that the field value $H_{2D}(T^*)$ perhaps defines an upper limit below which anomalous effects would be visible. Figure 18 schematically sketches the anticipated behaviour in temperature dependent magnetization data across such a limiting field. The data of figure 17(a) and figure 2 can be identified with the schematic plots in figures 18(a) and (b), respectively.

Figure 19 summarizes further manifestations of the hysteritic response at low fields. The magnetization curves in field values smaller than $H_{2D}(T^*)$ are not expected to intersect, they asymptotically approach each other near the superconducting transition temperature (e.g., see $M_{ZFC}(T)$ data in 1·3 Oe and 10 Oe, respectively in figure 19(a)). Figure 19(b) shows a comparison of magnetization data recorded during warm-up (after initial rapid cool-down to 10 K in a field of about 0·3 Oe) with the data recorded during slow cool-down in the same field. These data exemplify the survival of irreversible behaviour in a small field of 0·3 Oe up to the nominal T_c value.

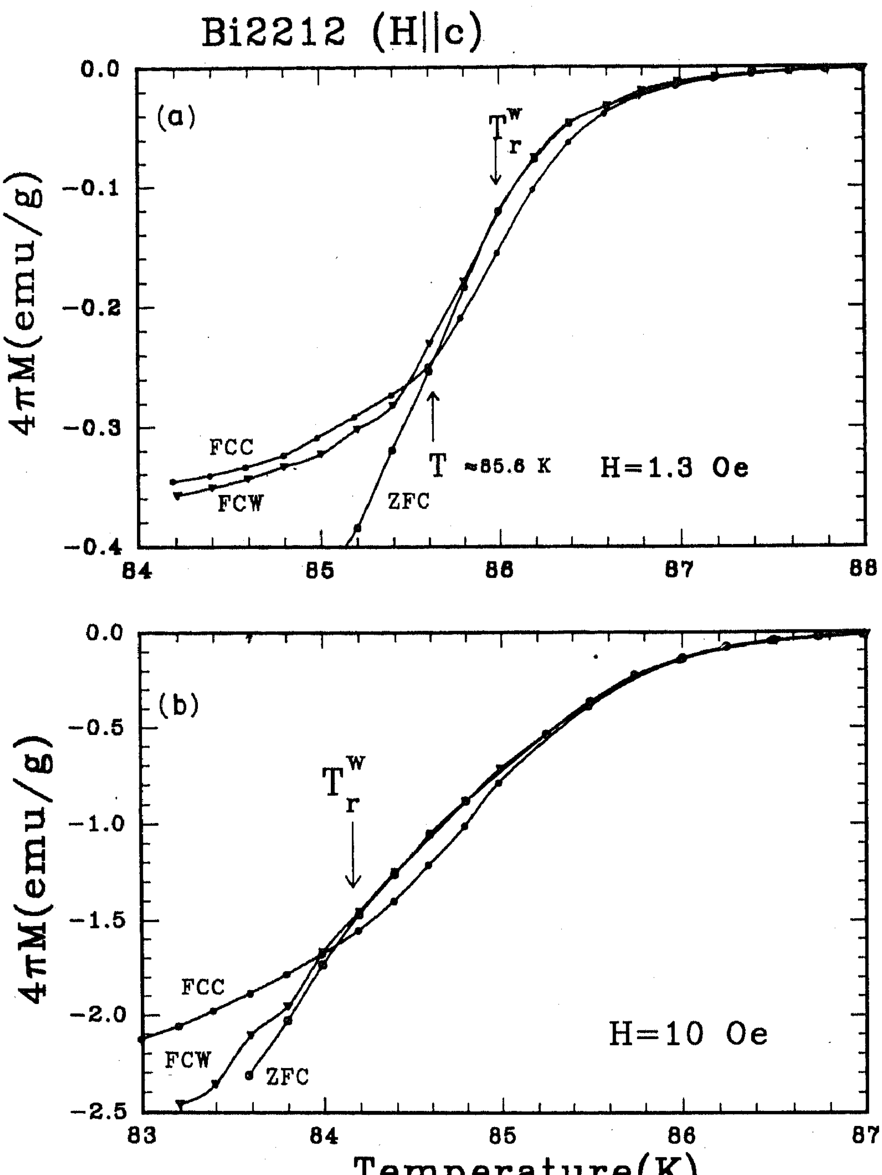

Figure 17. Temperature dependence of magnetization data in (a) $H \approx 1{\cdot}3$ Oe and (b) $= 10$ Oe, respectively.

3.8 *Magnetic phase diagram*

The magnetization data presented so far (for instance, see figures 10, 13 and 15) motivate us to propose revision and additions in the schematic phase diagrams drawn by Brawner *et al* [8] and Huse *et al* [1]. Our proposed schematic phase diagram near T_c is depicted in figure 20. We feel that on cooling down a specimen from higher temperature end, the superconducting fluctuations start to cause a deviation from the normal state resistivity $\rho(T)$ in the "H_{c2}" crossover region. Below this region there broadly exist three regions corresponding to response from strongly coupled 2D planes ($\equiv$ 3D superconductor), weakly coupled 2D planes (two-component response) and decoupled 2D planes. The usually determined irreversibility lines approximately demarcate the regions of reversible and irreversible response from the 3D superconductor. The H_{2D} line lies above the $H_r(T)$ line. The temperature at which

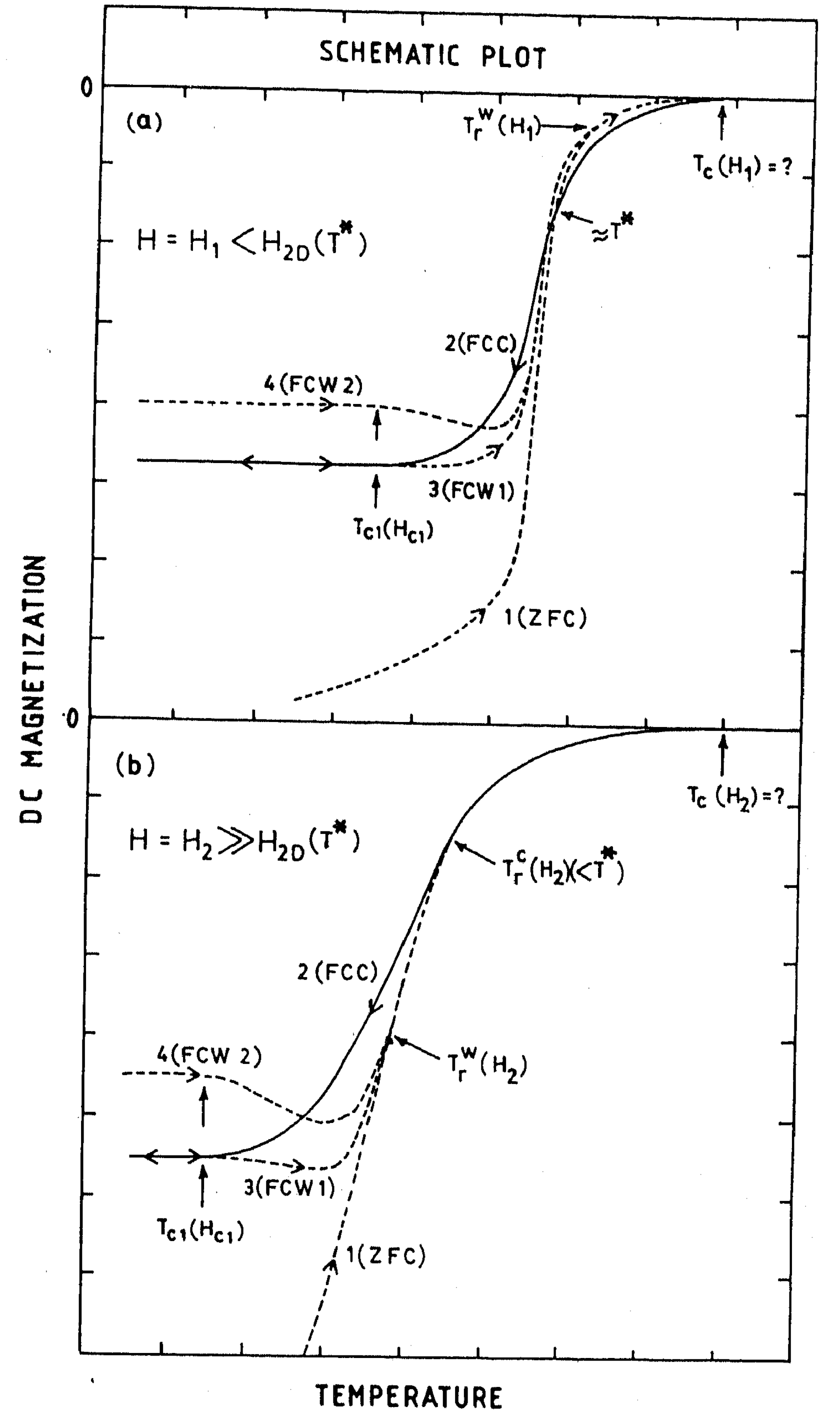

Figure 18. Schematic plot of temperature dependent magnetization data expected in (a) very small and (b) large applied fields. In (b), $T_r^c(H) > T_r^w(H)$ and no further anomalies are anticipated. In (a), $T_r^w(H) > T^*$, $T_r^c(H)$ cannot be defined and many anomalies are anticipated near T.

drastic change in the value of the slope dH_{2D}/dT (also, that of dH_{c1}/dT) occurs approximately identifies the onset of transformation from one component (3D superconductor) to two component (weakly coupled 2D planes) response. The $H_{2D}(T)$ line in the temperature interval of two component response approximately demarcates the field-temperature regions of $\rho_c = 0$ and $\rho_c \neq 0$.

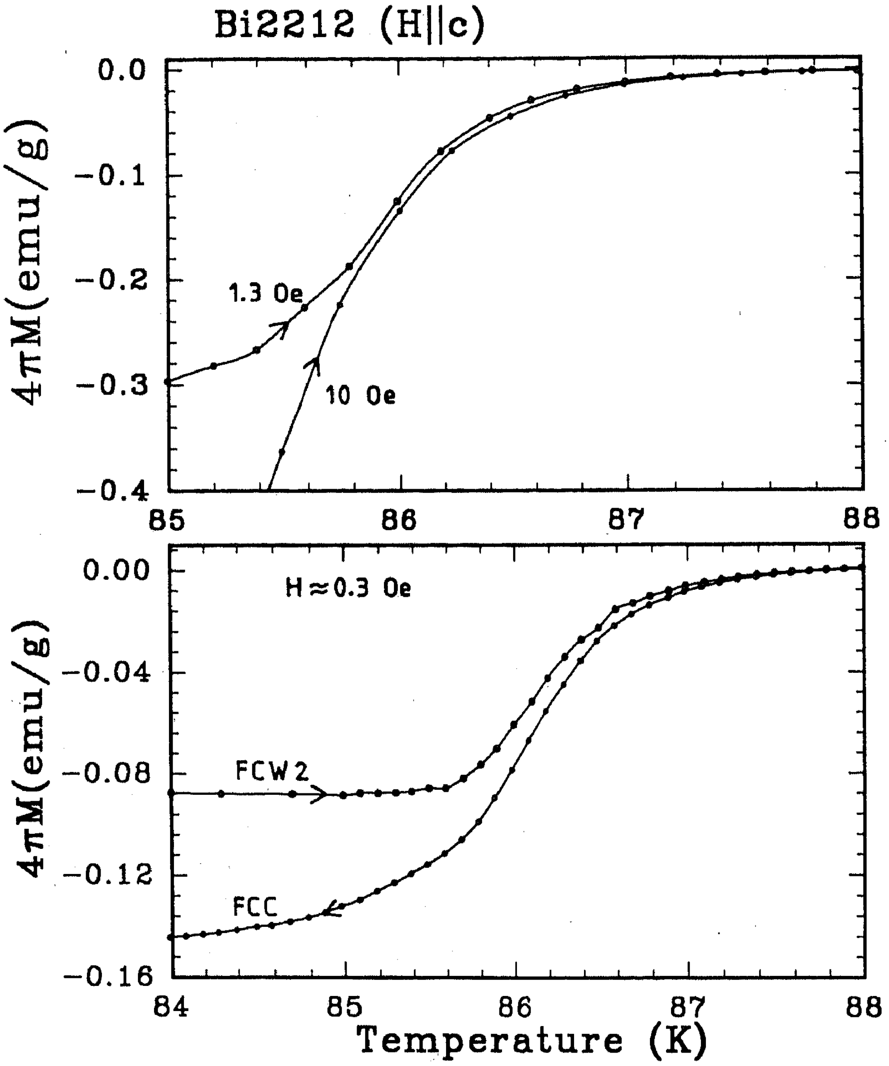

Figure 19. Temperature dependence of magnetization values near T_c in small field values. Figure 19(a) shows that warm-up magnetization curves obtained in $H = 1{\cdot}3$ Oe and 10 Oe, respectively, asymptotically meet at T_c. Figure 19(b) shows a comparison of field cool warm-up (after initial rapid cool down) and slow field cool cool-down magnetization data in the residual field of magnet ($\approx 0{\cdot}3$ Oe).

For the strongly coupled 2D planes, the $H_{2D}(T)$ line probably lies in between the $T_c^{ab}(H)$ and $T_c^{c}(H)$ lines. We have not extrapolated the $H_r(T)$ line across the transformation region of one component to two component response in view of the observed enhancement in the irreversibly field values due to interplay between the responses from two components. Further, the measured $H_{c1}(T)$ and $H_{2D}(T)$ in the region of 2-component response, may be taken as the lower and upper critical fields of the component describing the response from interplanar couplings.

It is also worthwhile to stress that our proposed phase diagram, in concept, is consistent with two other experimental facts about cuprate variety of HTSC, as enunciated by Geballe [47] and Chien *et al* [48], respectively. A unit cell of Bi2212 comprises of a pair of CuO_2 bilayers Josephson coupled to each other. The experimental data on BSCCO superlattice structures supports the view [47] that $\rho_{ab} = 0$ transition at about 85 K is a property of double pyramidal sequence (i.e., one CuO_2 bilayer). In $YBa_2Cu_3O_7$ system, one needs to consider at least two unit cells to

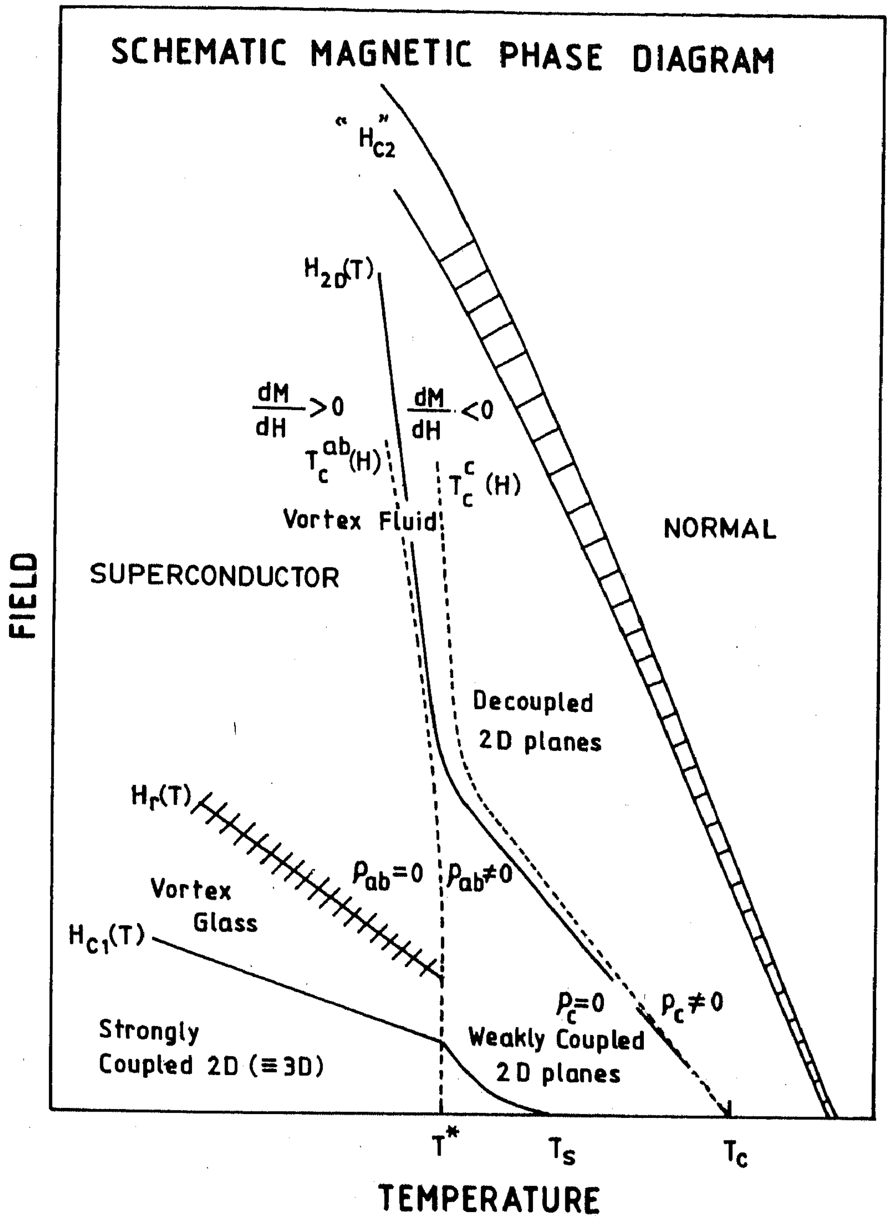

Figure 20. Schematic plot of proposed phase diagram in an anisotropic layered superconductor. For explanation of various lines, see text.

visualize the presence of a double pyramidal sequence (two CuO_2 layers separated by a Y layer). Chien *et al* [48] have reported that within the vortex liquid state of $YBa_2Cu_3O_7$ ($H \parallel c$), qualitative changes in dissipation behaviour occur across some identifiable field-temperature lines. The vortex motion correlated over a certain length scale gives way to a diffusive behaviour across a specific field-temperature line (see, H_k line in figure 4 of [48]). In our phase diagram, we expect the diffusive behaviour to occur above $T_c^c(H)$ line. The vortex motion may continue to remain correlated across $T_c^{ab}(H)$ line, somewhat analogous to the behaviour across H_0 line of Chien *et al* [48].

4. Summary

We have performed extensive isothermal and temperature dependent magnetization measurements for $H \parallel c$ in a high quality single crystal sample of $Bi_2Sr_2CaCu_2O_8$. The new results presented in this paper include the following:

1. Comparison of irreversibility lines (for $H \parallel c$) determined by different DC and AC magnetization methods and procedures in the same single crystal sample of a HTSC.
2. Identification and labelling of two component response across the dimensional crossover region and determination of $H_{2D}(T)$ line.
3. Identification of effects due to the interplay between the diamagnetic responses from two components.
4. Recognition of occurrence of qualitative changes in the irreversibility behaviour across the dimensional crossover.
5. Observation of paramagnetic Meissner effect in the temperature interval of dimensional crossover. This effect *inter alia* confirms our labelling of two component response.
6. Schematic representation of the observed anomalies near T_c in the temperature dependent magnetization data at low fields.

5. Conclusion

The modelling of highly anisotropic copper oxide superconductors as stacks of two dimensional superconducting planes Josephson coupled to each other is so appealing, that for long experimentalists have sought to provide evidences in favour of such a scenario in all sorts of magnetic and transport measurements. We feel that the results presented in this paper convincingly reaffirm the validity of Josephson coupled models. The isothermal and temperature dependent magnetization data near the superconducting to normal transition temperature comprehensively elucidate the changes that occur in the electromagnetic response over a narrow range of temperature values. The schematically sketched phase diagram in figure 20, in a sense, summarizes our present understanding of such systems. More experiments are in progress at the moment and the sketched diagram may undergo further revisions on analysis of newer results. We have refrained from making any quantitative contact with different theoretical models, including the one due to BLK [6], at this stage, as by and large no model predicts several of the key features of the scenario that appears to emerge from the results in our specimen of Bi2212.

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Post script

Isothermal magnetic hysteresis data for $H \parallel c$ in our specimen between 10 and 75 K (obtained since the submission of this manuscript) reveal the presence of additional two component responses at temperatures 25 K and 75 K. The magnetic response is one component like between 5 and 15 K, 35 K and 65 K, 78 K and 83 K. We now believe that in addition to intra-unit cell weak link there also exist inter-unit cell weak links having a certain heirarchy in any Bi2212 single crystal specimen. The Bi2212 single crystals are never perfectly stoichiometric and HREM pictures also reveal the presence of stacking faults as well as the intergrowth of unit cells of Bi2201 and Bi2223 in them. The inter-unit cell weak links are a material property whereas the intra-unit cell weak link is intrinsic (exists within a unit cell) and is the strongest. The breakage of intra-unit cell weak link gives rise to dimensional crossover phenomenon just below T_c, whereas the breakage of inter-unit cell weak links amount to division and further sub-division of a given crystal into aligned microcrystallites of progressively smaller dimension. The microcrystallites and the Josephson coupling across them can give rise to two component response, each of which is like a 3D type-II superconductor. In view of above scenario, it may be difficult to uniquely label the weak link responsible for negative peak effect in our figure 18 as well as for the effects described in references [4] and [45].