

Comparison of irreversibility temperatures determined via DC and AC magnetization techniques in conventional superconductors

A K GROVER*, S RAMAKRISHNAN†, RAVI KUMAR†,
P L PAULOSE†, S K MALIK† and P CHADDAH**

*Department of Physics, Panjab University, Chandigarh 160 014, India

†Tata Institute of Fundamental Research, Bombay 400 005, India

**Solid State Physics Division, Bhabha Atomic Research Centre, Bombay 400 085, India

MS received 1 July 1991

Abstract. A comparison has been made of irreversibility temperature determined by four different methods in few specimens of lead (type-I) and niobium (type-II). The merger of $M_{ZFC}(T)$ and $M_{FC}(T)$ curves give $T_r(H)$ values lower than those evident from vanishing the hysteresis in isothermal DC magnetization. The identification of peak temperature in $\chi_H''(T)$ data with $T_r(H)$ is appropriate only if the contribution from changes in the normal state electrodynamics can be isolated and the peak is narrow. The appearance of differential paramagnetic effect in $\chi_H'(T)$ data is adequate to imply reversibility, however, its efficacy to precisely locate irreversibility line remains to be established.

Keywords. Irreversibility line; AC and DC magnetization techniques; type-I and type-II superconductors.

PACS Nos 74·30; 74·55; 74·60

1. Introduction

Two basic properties which comprise the superconductivity phenomenon are the infinite conductivity and the complete expulsion of magnetic flux. The latter property ensures a thermodynamic response in a superconducting material. However, an incomplete expulsion of magnetic flux is a common occurrence in both type-I and type-II superconductors and such materials display a path dependent or hysteretic magnetic response. The advent of high temperature superconductors (HTSC) has given rise to the belief (Malozemoff 1990) that there exists a region of thermodynamic response just below the normal-superconducting phase boundary ($T_c(H)$ line) and the hysteretic behaviour sets in below an irreversibility (T_r, H_r) line. We have searched for such an irreversibility line by DC and AC magnetization techniques in few specimens of conventional low temperature type-I and type-II superconductors with an objective of comparing the relative efficacy of four different procedures usually employed to determine the irreversibility temperature in HTSC. To enumerate, these different procedures are (i) the merger of zero field cooled (ZFC) and field cooled (FC) DC magnetization curves measured in a constant field H , (ii) the vanishing of hysteresis in isothermal DC magnetization curves, (iii) the appearance of differential

* On leave of absence from Tata Institute of Fundamental Research, Bombay 400 005, India

paramagnetic effect (DPE) (Hein and Falge 1961) in the in-phase AC susceptibility $\chi'_H(T)$ measured in DC field H and (iv) the peak in out-of-phase AC susceptibility $\chi''_H(T)$. The results obtained illustrate the scope and limitations of each of these methods.

2. Experimental

2.1 Samples

The type-I and type-II materials chosen are lead ($T_c = 7.2$ K) and niobium ($T_c = 9.2$ K) respectively. The lead specimen (Grover *et al* 1990) is in the form of a disc (dia. = 4.15 mm, thickness = 2.15 mm) and niobium specimens (Grover *et al* 1989a, b) are in the form of a disc (dia. = 3.59 mm, thickness = 0.09 mm) and powder (sieved through 65 μ mesh). The Pb specimen is known to display more hysteretic behaviour when the field is applied perpendicular to the plane of the disc as compared to that in the parallel orientation (Grover *et al* 1990). The two specimens of niobium also differ in the degree of their irreversible behaviour (Grover *et al* 1989a, b).

2.2 Magnetic measurements

The DC magnetic measurements have been made by using a quantum design SQUID magnetometer. The AC magnetic measurements have been performed using a home-built mutual inductance system (Ramakrishnan *et al* 1985). For disc-shaped specimens, the DC field (coaxial to the AC field) was either parallel or perpendicular to the plane of the disc. The mutual inductance bridge was balanced with the sample in its superconducting state. The DC and AC magnetic measurements made include the following:

- (i) The isothermal $M - H$ curves at 4.7 K for H parallel and perpendicular to the plane of Pb and Nb discs. The $M - H$ curves in Nb powder at 4.2 K and 6.2 K.
- (ii) The ZFC and FC magnetizations in Pb disc at $H = 50, 100, 200, 300$ Oe for both orientations. Similar measurements at several DC fields from 20 to 3000 Oe in Nb powder specimens.
- (iii) The χ'_H and χ''_H at 21 Hz in Pb disc at 4.7 K as a function of DC field H in both orientations.
- (iv) The χ'_H and χ''_H at 21 Hz in Pb disc as a function of T at several H in both orientations. At $H = 100$ Oe in the perpendicular orientation, χ'_H and χ''_H data were also recorded by changing the amplitude (h_{ac}) as well as the frequency of AC field.
- (v) χ'_H and χ''_H at 21 Hz and 210 Hz respectively in Nb disc as a function of T at various H in the parallel orientation. χ'_H and χ''_H at few frequencies in Nb powder as a function of T at different chosen values of h_{ac} and in $H = 0$ and 100 Oe respectively.

Most of the results obtained are depicted in figures 1 to 11. The relevant parameters have been specified in the figures and wherever considered appropriate, and arrows have been put to indicate the way T and/or H were varied.

3. Results and analysis

3.1 Lead

Figure 1 shows $M - H$ curves in Pb disc specimen at 4.7 K for the parallel and perpendicular orientations. It appears that the field interval over which the forward and reverse magnetization curves overlap is larger in the parallel orientation of the given specimen. Figures 2a to 2d show χ'_H and χ''_H vs H at 21 Hz in the same specimen at 4.7 K. The data in figures 2a and 2c show the appearance of paramagnetic peak (designated as DPE) just before the material turns normal. The field interval over which the DPE is present is only marginally greater in the parallel orientation as compared to that in the perpendicular orientation. A comparison of the data in figures 2a and 2c with those in figures 1a and 1b respectively indicate that χ'_H data do not amount to tracing of the derivative of the DC magnetization hysteresis curve. It may be specifically noted that the slope values of the forward and reverse magnetization curves between 0 and 300 Oe are very different, however, the measured χ'_H values in the same interval along the forward and the reverse field directions are

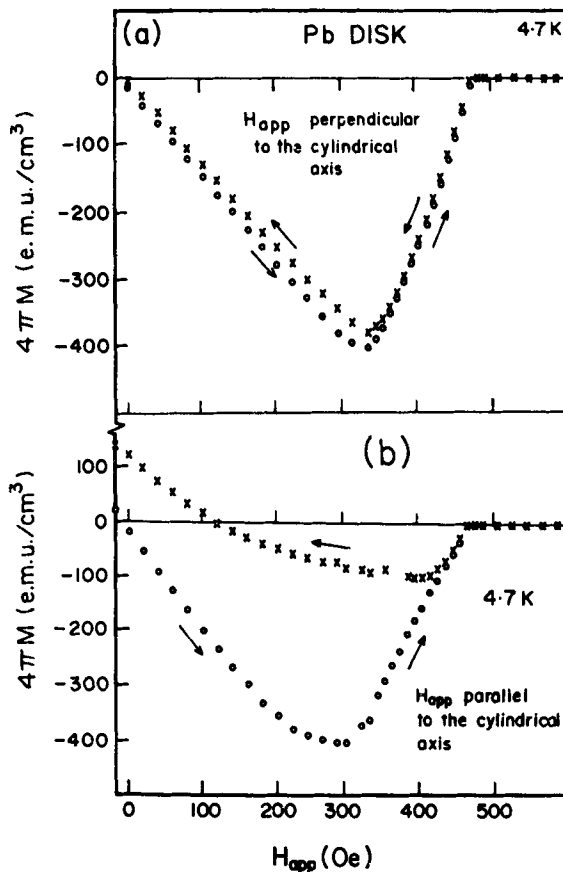


Figure 1. Magnetization hysteresis curve in Pb disc (dia = 4.15 mm, thickness = 2.15 mm) at 4.7 K for field applied nominally parallel and perpendicular to the disc plane.

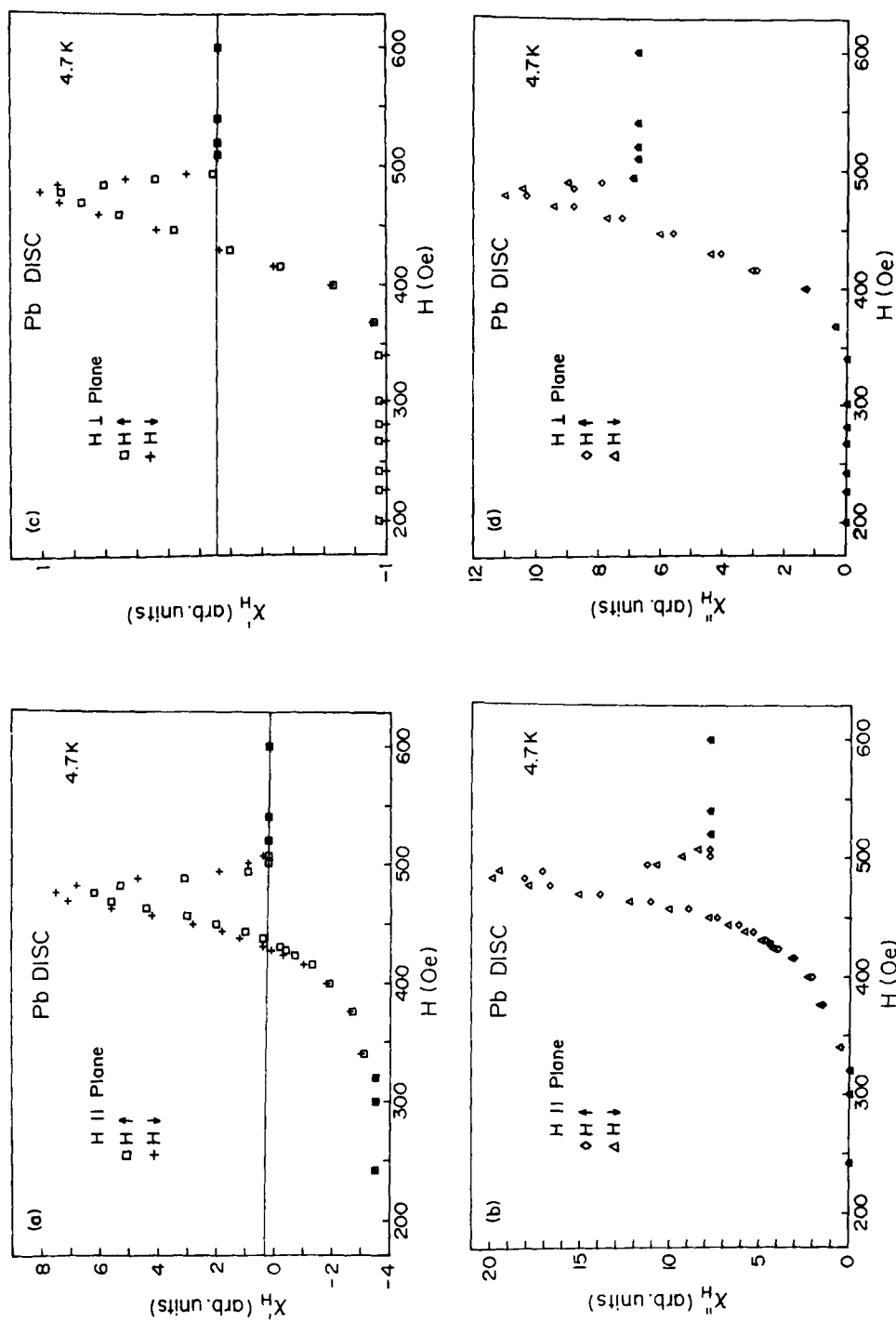


Figure 2. The isothermal χ_H'' and χ_H'' (measured at 21 Hz in h_{ac} of 1 Oe rms) vs H in Pb disc at 4.7 K for field applied in parallel ((a) and (b)) and perpendicular ((c) and (d)) orientations.

about the same. It had been pointed out by Hein and Falge (1961) that AC susceptibility measurements amount to tracing a minor hysteresis loop over the field interval $\Delta H(= \pm h_{ac})$ around a given $M(H)$ value of the hysteresis curve. The observation of DPE in a small value of h_{ac} necessitates the existence of nearly overlapping forward and reverse magnetization curves. Thus, the field interval over which DPE is present is a reliable indicator of quasi-reversible response. No magnetic hysteresis loss is anticipated in the field region of DPE in χ'_H data (cf. figures 2a and 2b and figures 2c and 2d respectively). However, χ''_H data of figures 2b and 2d show large increase in the said region. We believe that this is a consequence of changes in the normal state electrodynamic as H approaches the critical field value of the superconductor, somewhat akin to the occurrence of peak-like structure in $\chi'_0(T)$ data (Maxwell Strongin 1963; Hein 1986; Hein *et al* 1989; Geshkenbein *et al* 1991).

Figure 3 show the plots of $\Delta M(H)(\cong (M_{FC}(H) - M_{ZFC}(H)))$ vs T in Pb disc specimen at the H values indicated in both orientations. For a given H , $\Delta M(H)$ values in the perpendicular orientation are much larger, however, the $T_r(H)$ values that may be obtained from $\Delta M \rightarrow 0$ criterion appear to be nearly the same in the two orientations

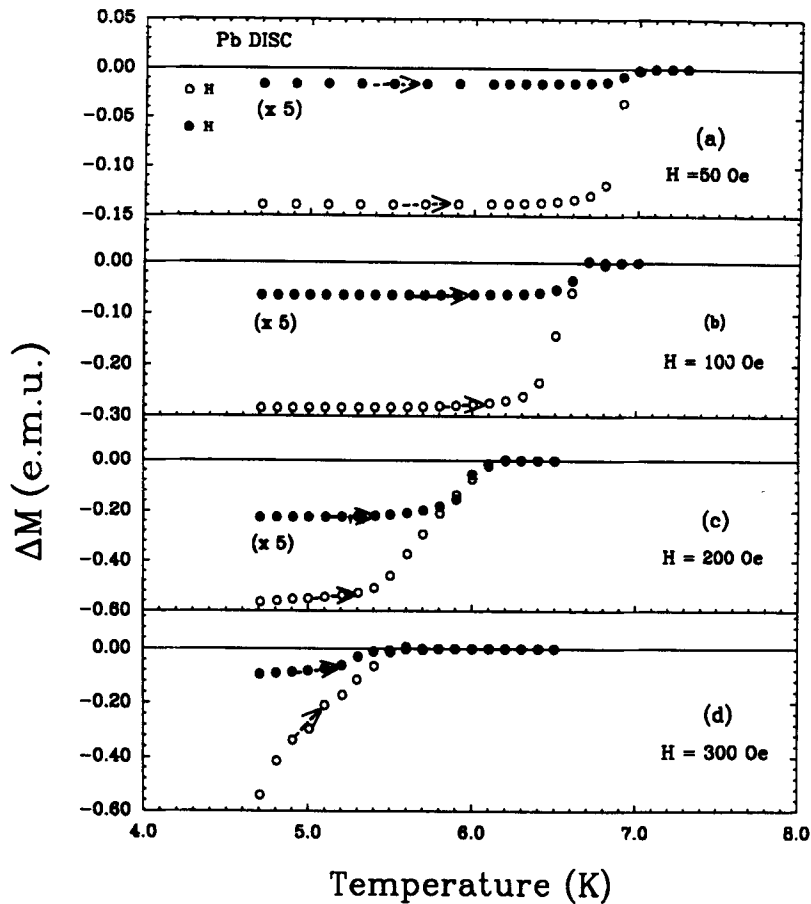
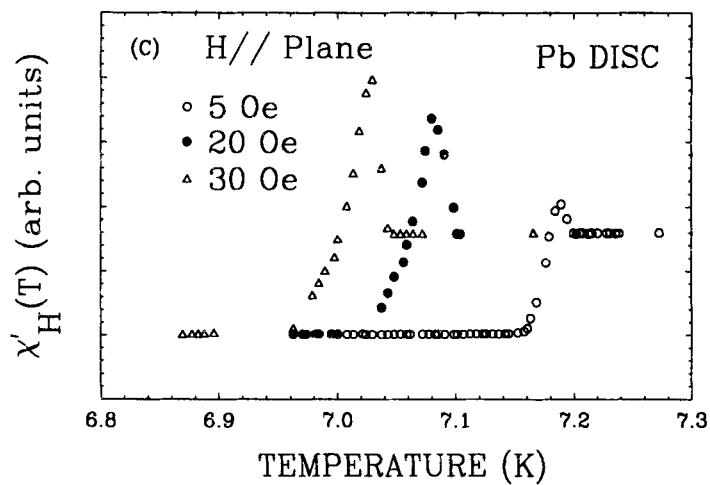
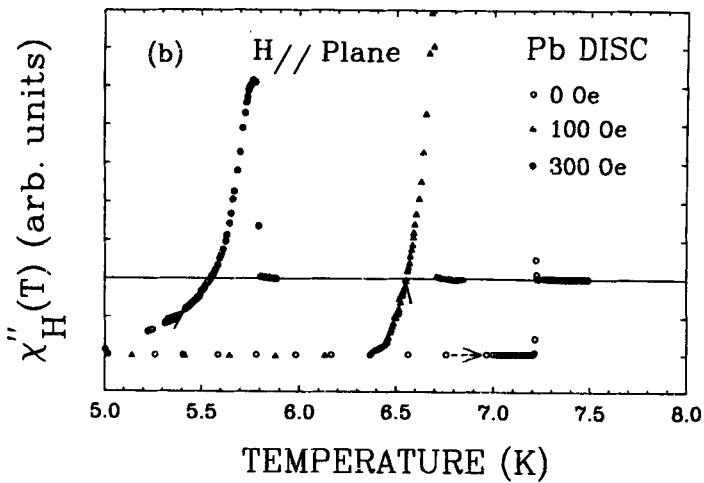
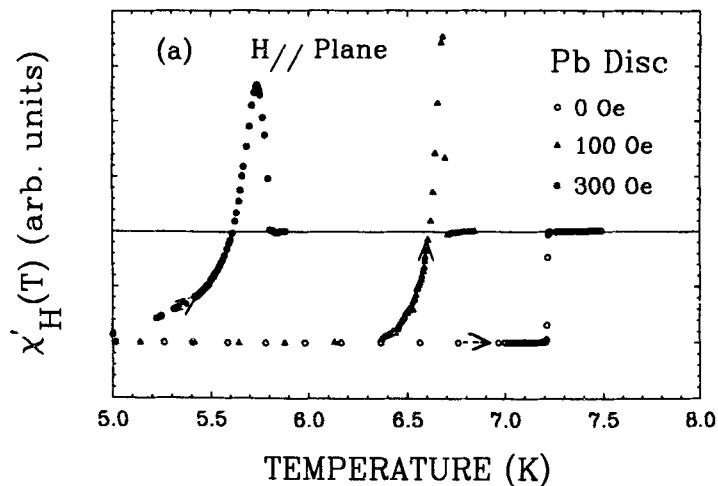


Figure 3. Plots of $\Delta M(H)(= (M_{FC}(H) - M_{ZFC}(H)))$ vs T in Pb disc for field applied parallel (open circle) and perpendicular (close circle) to the disc plane. The arrows mark the $T_r(H)$ values by $\Delta M(H) \rightarrow 0$ criterion.



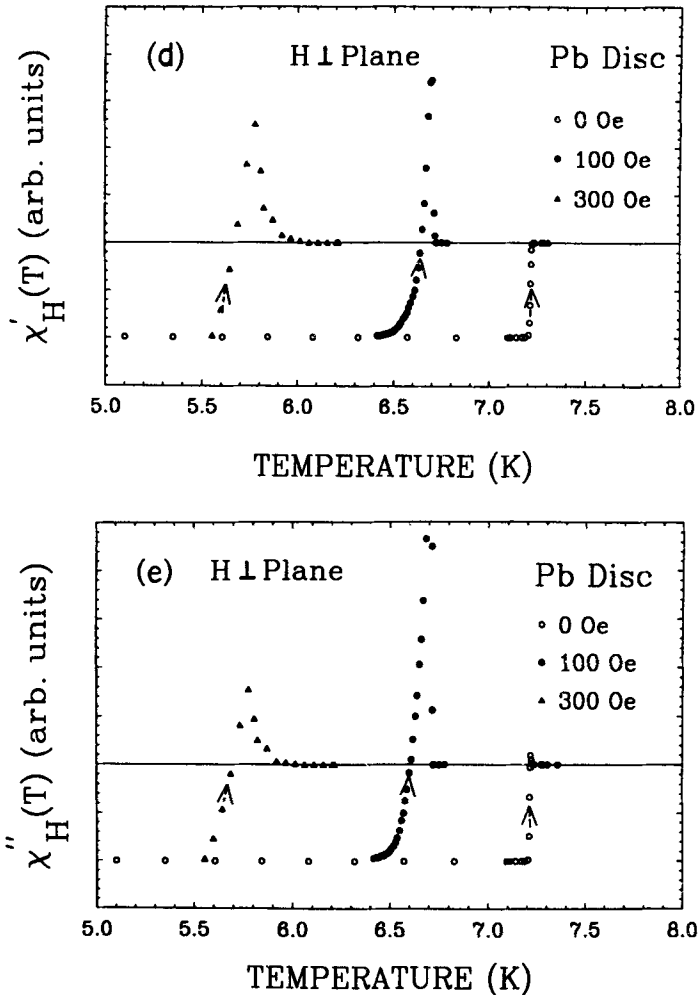


Figure 4. The temperature variation of χ'_H and χ''_H (measured at 21 Hz in h_{ac} of 1 Oe rms) in Pb disc specimen in the parallel ((a), (b) and (c)) and perpendicular ((d) and (e)) orientations. The data at different H values have not been scaled to one-another. Wherever considered appropriate, the horizontal lines have been drawn through susceptibility values in the normal state for reference purpose only.

Figures 4a to 4e show $\chi'_H(T)$ and $\chi''_H(T)$ data recorded at 21 Hz in an AC field of 1 Oe at few fixed DC fields in Pb disc specimen for parallel and perpendicular orientations. Figure 5 shows $\chi'_H(T)$ data at 21 Hz for $H = 100$ Oe in the perpendicular orientation for two values (0.3 and 2 Oe) of the AC energizing field. Figure 6 shows $\chi'_H(T)$ data in the same situation at 21 Hz and 210 Hz obtained with an h_{ac} of 1 Oe. In figures 4a and 4d, it may first be noted that no paramagnetic response is evident in $\chi'_0(T)$ data. However, a DC field of 5 Oe is adequate to elicit a DPE peak (see figure 4c). The $T_r(H)$ values that may be identified with temperature at which $\chi'_H(T)$ changes from positive to negative values are found to be nearly the same for the two orientations from the data recorded at 21 Hz (cf. figures 4a and 4d). The two sets of data in figure 5 show that the $T_r(H)$ values at 21 Hz is not dependent on the amplitude of h_{ac} . However, the two sets of data in figure 6 show that $T_r(H)$ value at given H

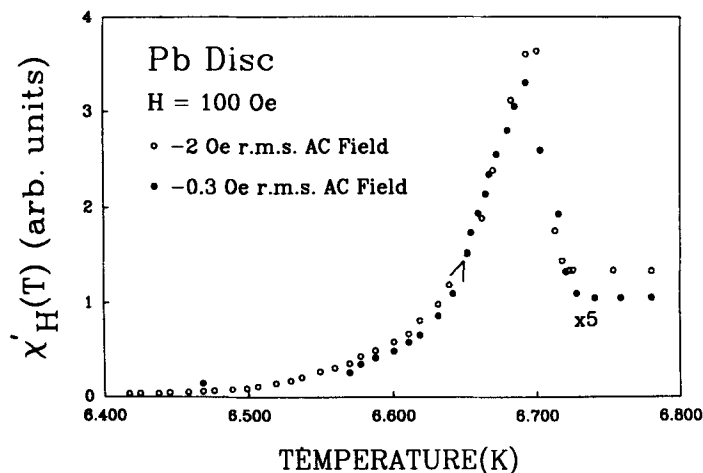


Figure 5. The temperature variation of χ_H'' measured at 21 Hz for two values of h_{ac} (0.3 and 2.0 Oe rms respectively) in Pb disc specimen for $H = 100$ Oe applied perpendicular to the plane of the disc.

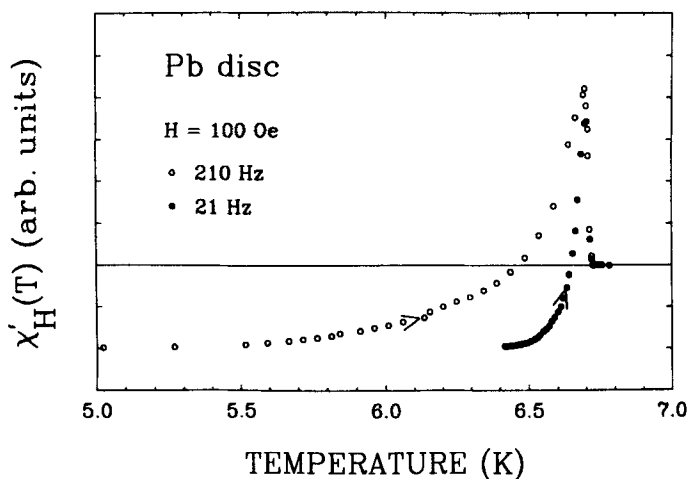


Figure 6. The temperature variation of χ_H'' measured with h_{ac} of 1 Oe rms at 21 Hz and 210 Hz respectively in Pb disc specimen with $H = 100$ Oe applied perpendicular to the disc plane.

estimated by the given criterion is a sensitive function of frequency of h_{ac} . $T_r(H)$ value appears to decrease as frequency increases. It is important to note that $T_r(H)$ values evident from DPE data at 21 Hz are consistent with the corresponding values estimated from DC magnetization data by $\Delta M(H) \rightarrow 0$ criterion. This fact appears to establish the efficacy of DPE data at low enough frequency to give reasonable estimate for $T_r(H)$ values.

The $\chi_H''(T)$ data in both the orientations show peak-like structures at $T_c(0)$ (see figures 4c and 4e). The peaks in $\chi_H''(T)$ shift to lower temperature and also broaden as H increases. The peak-like structure in $\chi_H''(T)$ is believed to have contributions from both the changes in normal state electrodynamic on approaching normal-

superconducting phase boundary as well as from the hysteresis loss phenomenon in the irreversible region of the superconducting state (Hein 1986; Hein *et al* 1989). The observation that the temperature intervals over which the DPE exists in $\chi'_H(T)$ data nearly coincide with the corresponding widths of the peaks in $\chi''_H(T)$ data, seem to imply that the latter structures mainly originate from the changes in the normal state electrodynamics. Thus, it may not be appropriate to identify the peak temperature in $\chi''_H(T)$ with $T_p(H)$ value.

The characteristic temperatures which can be noted from the experiments conducted on Pb disc specimen are $T_c(H)$, $T_r(H)$ (from DPE at 21 Hz) and $T_p(H)$ (peak temperature in $\chi''_H(T)$).

3.2 Niobium

Figure 7 shows DC magnetization hysteresis curve in the same specimen at 6.2 K, the inset of figure 7 shows the forward and reverse magnetization curves near H_{c2} value at 6.2 K on an expanded scale. Figure 8 shows temperature variation of ZFC and FC DC susceptibility values in Nb powder specimen at few H values. The data at several other H values have not been shown for brevity. The $H = 3$ kOe data of figure 8 show that χ_{ZFC} and χ_{FC} curves nearly merge into each other by 6.2 K, however, the inset of figure 7 indicates that the difference between forward and reverse $M - H$ curves persists far beyond 3.5 kOe at 6.2 K. Figure 9 shows the temperature dependence of the in-phase and the out-of-phase AC susceptibility values in the Nb powder at 21 Hz in h_{ac} of 1 Oe and in $H = 0$ and 100 Oe respectively. No DPE or any other peak-like structure can be seen in the χ'_H and χ''_H data of figure 9 as well as in other runs made by varying the frequency and the amplitude of AC field. We believe that the failure to observe DPE is consequence of absence of genuinely reversible region near the $T_c(H)$ line in the given Nb powder specimen. The situation in this case is that at a given H though M_{ZFC} approaches M_{FC} as T approaches a

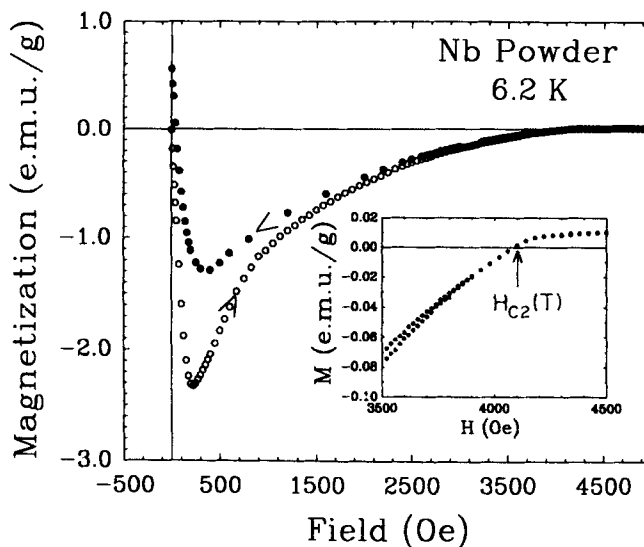


Figure 7. Magnetization hysteresis curve in Nb powder specimen at 6.2 K. The inset shows the forward and reverse $M - H$ curves near H_{c2} value.

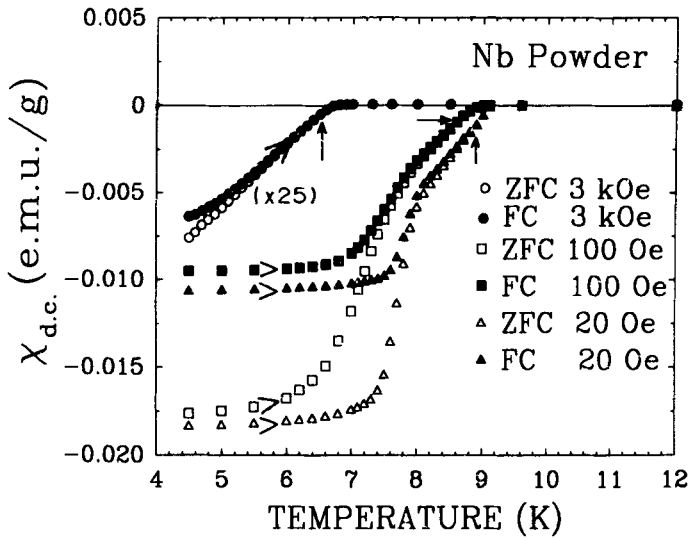


Figure 8. The temperature variation of ZFC and FC DC susceptibility values in Nb powder specimen at $H = 20, 100, 3000$ Oe respectively. The arrows identify the $T_c(H)$ values by $\Delta\chi(H) \rightarrow 0$ criterion.

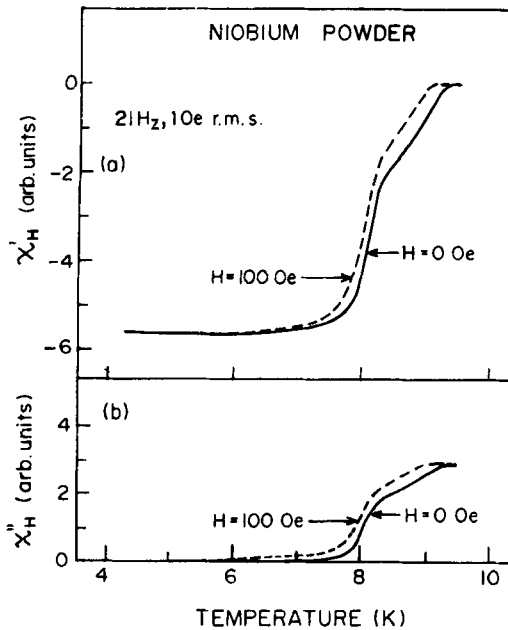


Figure 9. The temperature variation of χ'_H and χ''_H measured at 21 Hz in h_{ac} of 1 Oe in Nb powder specimen at $H = 0$ and 100 Oe respectively.

quasi-irreversibility temperature (determined via $\Delta\chi(H) \rightarrow 0$ criterion and marked by arrows in figure 8), the width of isothermal hysteresis remains significantly larger. The Nb disc is such that no magnetic flux escapes from it on field cooling and $M_{FC}(H)$ values are measured to be zero for the entire range of H values (Grover *et al* 1989a, b).

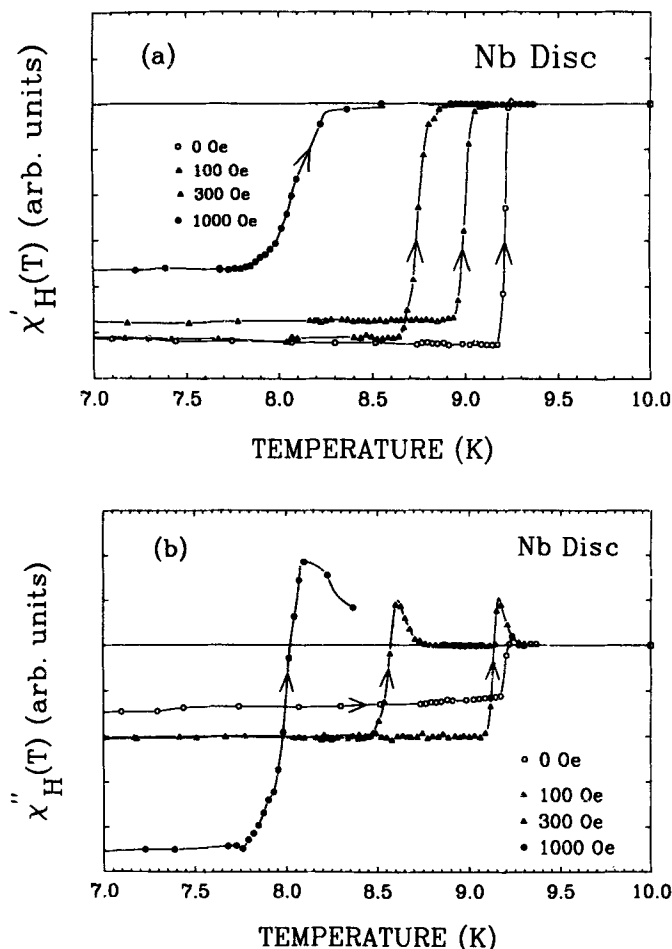


Figure 10. The temperature variation of χ'_H and χ''_H measured at 21 Hz in h_{ac} of 1 Oe in Nb disc at field values indicated in the parallel orientation. The continuous line joining the data points are just an aid to guide the eye. The data sets at different H are not normalized to one-another.

The M_{ZFC} values approach M_{FC} value only at $T = T_c(H)$. The isothermal forward and reverse $M - H$ curves are seen to meet (data not shown) at $H = H_{c2}(T)$. Figure 10 shows the $\chi'_H(T)$ and $\chi''_H(T)$ data in Nb disc specimen at some values of H . As anticipated, no DPE like structure is present in $\chi'_H(T)$ data. The peak-like structures are present in $\chi''_H(T)$ data. The peak is seen to broaden as H increases. The peak temperature in $\chi''_H(T)$ do not identify $T_r(H)$ values.

The characteristic temperatures that can be noted from the experiments conducted on Nb powder and disc specimens are $T_c(H)$ and $T_r(H)$ (from $\Delta\chi \rightarrow 0$ criterion) in the former and $T_c(H)$ and $T_p(H)$ (peak temperature in $\chi''_H(T)$) in the latter.

3.3 Power law relationship

Müller *et al* (1987) had introduced the fitting of $T_r(H)$ values determined from the merger of M_{ZFC} and M_{FC} curves to a power law relation, $(1 - T_r(H)/T_c(0)) \propto H^a$, and

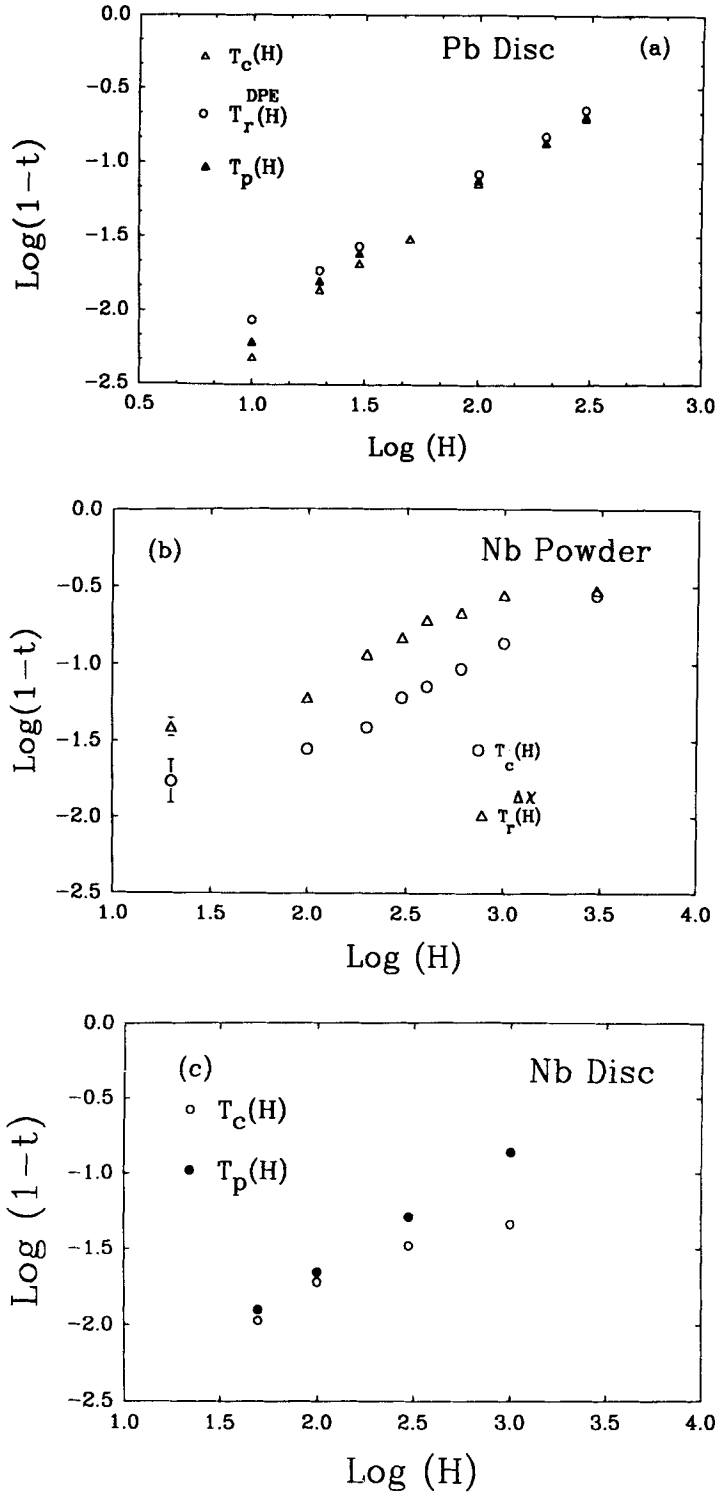


Figure 11. Log-log plots of $(1-t)$ vs H for different sets of t values in Pb disc (a), Nb powder (b) and Nb disc (c) specimens. The three sets of t values in Pb correspond to $T_c(H)/T_c(0)$, $T_r^{\text{DPE}}(H)/T_c(0)$ (DPE at 21 Hz) and $T_p(H)/T_c(0)$ respectively. The two sets each in Nb powder and disc specimens correspond to $T_c(H)/T_c(0)$ and $T_r^{\Delta x}(H)/T_c(0)$, and $T_c(H)/T_c(0)$ and $T_p(H)/T_c(0)$ respectively.

found a value of $2/3$ for the exponent q in a specimen of cuprate variety of HTSC. We display in figure 11 our attempt to fit different characteristic temperatures in Pb and Nb specimens to a power law behaviour. In Pb disc, $T_c(H)$, $T_r(H)$ and $T_p(H)$ appear to fit (figure 11a) to the power law with $q \cong 1$. In Nb powder, $T_c(H)$ and $T_r(H)$ values in the field range 0.1 to 1.0 kOe can be fitted (figure 11b) to power law with $q \cong 2/3$ and $11/20$ respectively. In Nb disc, $T_c(H)$ data do not appear to fit (figure 11c) to power law, whereas $T_p(H)$ values do with $q \cong 4/5$.

4. Summary and conclusions

To summarize, we have presented experimental data obtained by DC and AC techniques pertaining to the irreversibility phenomenon in superconducting specimens of Pb (type-I) and Nb (type-II) elements. The physical basis for irreversible behaviour in two types of superconductors are entirely different. Four different procedures have been looked into to ascertain the values of irreversibility temperatures $T_r(H)$. The $T_r(H)$ values determined from the merger of zero field cooled and field cooled DC magnetization data probably only specify a lower limit. The vanishing of hysteresis in isothermal magnetization curves appears to be a better criterion $T_r(H)$ from DC magnetic measurements—a feature also noted recently by Suenaga *et al* (1991). The identification of peak temperature in $\chi''_H(T)$ data with $T_r(H)$ value is appropriate only if the contribution from the normal state electrodynamics can be isolated from the hysteresis loss phenomenon and the peak in $\chi''_H(T)$ is narrow. The observation of differential paramagnetic effect (DPE) in $\chi'_H(T)$ is a qualitative feature whose presence is adequate to imply reversibility in the magnetization response, however, its efficacy to locate very precisely the T_r and H_r values remains to be established.

It may be argued that before measuring the irreversibility line in specimens of high temperature superconductors, one must first qualitatively establish the very existence of a reversible region by DPE in their $\chi'_H(T)$ data. The recent experiment of Khoder *et al* (1991) on Bi and Tl-based cuprates do show the presence of DPE. However, prior to them Hein *et al* (1989) did not succeed in their specific search for DPE in specimens of $\text{YBa}_2\text{Cu}_3\text{O}_7$ and had summarized that there is no genuine thermodynamic region in that HTSC system. The present data on conventional superconductors appears to strengthen the belief that may stem from the remarks of Hein *et al* (1989). Many interesting theoretical ideas (see Brandt 1991) such as flux lattice melting, flux depinning, vortex liquid to glass transition, etc., which pertain to the nature of irreversibility line, are under active consideration in connection with the physics of HTSC. Some of these may not be relevant for conventional superconductors. However, it is hoped that more experiments akin to the present work would eventually lead to find a reliable way to determine accurate values of $T_r(H)$.

References

- Brandt E H 1991 *Int. J. Mod. Phys. B* (to appear)
 Geshkenbein V B, Vinokur V M and Fehrenbacher R 1991 *Phys. Rev.* **B33** 3748
 Grover A K, Paulose P L, Chaddah P and Ravi Kumar G 1989a *Physica* **C162-164** 335
 Grover A K, Paulose P L, Chaddah P and Ravi Kumar G 1989b *Pramana - J. Phys.* **33** 297

- Grover A K, Ravi Kumar, Chaddah P, Subramanian C K and Sankarnarayanan V 1990 *Physica* **C170** 431
- Hein R A 1986 *Phys. Rev.* **B33** 7539 and references therein
- Hein R A and Falge Jr. R L 1961 *Phys. Rev.* **123** 407
- Hein R A, Hojaji H, Barkatt A, Shafii H, Michael K A, Thorpe A N, Ware M F and Alterescu S 1989 *J. Superconduct.* **2** 427
- Khoder A F, Couach M and Jorda J L 1990 *Phys. Rev.* **B42** 8714
- Malozemoff A P 1990 *MRS Bull.* **15** 50 and references therein
- Maxwell E and Strongin M 1963 *Phys. Rev. Lett.* **10** 212
- Müller K A, Takashige M and Bednorz J G 1987 *Phys. Rev. Lett.* **58** 1143
- Ramakrishnan S, Sundram S, Pandit R S and Chandra G 1985 *J. Phys.* **E18** 650
- Suenaga M, Ghosh A K, Youwen Xu and Welch D O 1991 *Phys. Rev. Lett.* **66** 1777