

SPIN-GLASS-LIKE FEATURES IN THE MAGNETIC BEHAVIOUR OF  
HIGH- $T_c$  SUPERCONDUCTORS

Y. Wolfus and Y. Yeshurun\*

Department of Physics, Bar-Ilan University, Ramat-Gan, Israel

and

I. Felner

Racah Institute of Physics, Hebrew University, Jerusalem, Israel

Abstract

Spin-glass-like models were invoked for explaining the irreversible magnetic susceptibility observed in high- $T_c$  superconductors. However, similar irreversible features are found in ordinary superconductors and are interpreted in terms of trapped flux. To demonstrate the difference in the magnetic response between ordinary superconductors and high- $T_c$  oxides we rotate the sample relative to the magnetic field and exhibit data for the angular dependence of the magnetization for Nb and for  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . The results for the latter resemble those obtained for classical spin-glasses but differ from those obtained for Nb.

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One of the intriguing features of the new high- $T_c$  superconductors<sup>(1,2)</sup>, found in magnetic measurements<sup>(3,4,5)</sup>, is the resemblance to the magnetic behaviour of spin glasses<sup>(6)</sup>. Most of the experimental claims for spin-glass-like features are based on irreversible properties found in the magnetic susceptibility. However, it is well known that in type II superconductors, extended lattice defects which act as pinning centers for the flux lines give rise to macroscopic irreversible phenomena<sup>(7)</sup>. The main purpose of this work is to experimentally compare irreversible properties in two systems: (i) Nb, an ordinary type II superconductor, and (ii)  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , an high- $T_c$  ceramic superconductor.<sup>(8)</sup> We describe two experiments. In the first one we measure the magnetic response of zero-field-cooled (ZFC) and of field-cooled (FC) samples. In the second we measure the response of the magnetization to rotation with respect to the external field. We

conclude that in some aspects  $\text{YBa}_2\text{Cu}_3\text{O}_7$  behaves differently than Nb and similarly to classical spin-glasses.

One of the characteristic fingerprints of spin-glasses is the "history dependence" of the magnetic response: The magnetic susceptibility of a sample which is cooled in zero field (ZFC) differs from that of a sample which is cooled in a field (FC). A very similar behaviour is found for the High- $T_c$  superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (Figure 1a). However, it is clear that on this basis the analogy to spin-glasses is some-

what superficial. The same history dependence is found for Nb (Figure 1b) where, using the superconductivity terminology, the ZFC and the FC curves represent the shielding and the Meissner effect respectively. The fact that we do not observe a full Meissner effect is attributed to trapped flux which is accumulated at higher temperatures during the cooling process, when the mixed-phase is crossed. Thus, due to the pinning forces, the number of flux vortices present at low temperature is higher than that in thermodynamic equilibrium and strongly depends on the magnetic history of the sample.

Another spin-glass characteristic is a field-induced macroscopic anisotropy. The remanent magnetization is coupled to the macroscopic anisotropy and can be rotated as a rigid body<sup>(9,10)</sup> but nonrigid rotations are observed for relatively high fields and temperatures.<sup>(11)</sup> We have recently reported<sup>(12)</sup> on similar features in a high- $T_c$  superconductor. We have studied the angular dependence of the field cooled magnetization of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  via measurements of the magnetization on a vibrating sample magnetometer (VSM) with a  $2\pi$ -rotating sample holder. In the FC measurements the sample was cooled in an applied field  $H$  from well above the transition temperature  $T_c$  to the measuring temperature, which is stabilized to better than 0.1 K. With the same field on, the sample was rotated by an angle  $\theta$  relative to the applied magnetic field. We then measured the magnetization  $M$  as a function of  $\theta$ .

Figure 2 exhibits typical  $M(\theta)$  data for FC runs in various fields for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (fig. 2.a) and for Nb (fig. 2.b) both taken at  $T = 4.2$  K. The most obvious feature in this figure is the strong angular dependence for the FC magnetization. To understand this feature we recall that in VSM measurements the measured magnetization  $M$  is the projection of the total sample magnetization  $\vec{m}$  on the direction of the field  $\vec{H}$ . The magnetization  $\vec{m}$  is composed of reversible and irreversible components:

$$\vec{m} = \chi\vec{H} + \vec{M}_{irr} \quad (1)$$

In the superconducting state the bulk susceptibility is of course isotropic and thus the reversible contribution  $\chi\vec{H}$  is always in the direction of  $\vec{H}$ . The irreversible part,  $\vec{M}_{irr}$ , on the other hand, might be coupled to the sample and rotate with it. It should also be taken into account that as a result of the external field,  $\vec{M}_{irr}$  might lag by an angle  $\Theta$  relative to the sample for a rotation  $\theta$  of the sample. Thus, the measured contribution of the irreversible magnetization is expected to be  $M_{irr}\cos(\theta-\Theta)$  where  $\theta \leq \theta$  and the measured magnetization is

$$M = \chi H + M_{irr}\cos(\theta-\Theta) \quad (2)$$

Least square analysis of the low-field data according to Eq.(2) yields  $\Theta = 0$ , implying a strong anisotropy. It is important to

note that in the low-field limit  $M(\theta)$  is reversible, i.e., the sample is rotated from 0 to  $2\pi$  and back to 0 and  $M(\theta)$  is only slightly altered.

Figure 2.a displays clearly the effect of the applied magnetic field on the angular dependence of the field-cooled magnetization for  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . For the convenience of representation we present  $M/H$  data. Generally speaking, all curves exhibit the same shape. However, note that  $M(2\pi) < M(0)$  and this difference is more pronounced for the higher fields. More important,  $M(\theta)$  is no longer reversible. This is demonstrated in the inset of Figure 2.a where we exhibit the angular dependence of the magnetization at 4.2 K for  $H = 1,500$  Oe. At an angle  $\theta'$  (which decreases with increasing field and temperature) there is a total breaking of the rigid moment and the FC curve coincides with the ZFC one. We note that very similar results, referred to as "broken rigidity", were observed in classical spin glasses.<sup>(10,11)</sup>

Rotating the sample in the presence of a field results in a torque on the trapped flux. Since  $M_{irr}$  (which is a result of the fluxons) is coupled to the sample due to pinning mechanism, the energy of the system increases by the rotation. If  $M_{irr}$  rotates as a rigid body, the increase in energy on rotating the sample by an angle  $\theta$  is given by:

$$\Delta \epsilon_{rot}(\theta) \equiv \epsilon(\theta) - \epsilon(0) \approx M_{irr} H (1 - \cos\theta). \quad (3)$$



When  $\Delta\epsilon_{rot}(\theta)$  becomes of the order of the pinning energy,  $\epsilon_p$ , we expect to find a breaking in the rigidity of  $M_{irr}$ .

We calculate  $\Delta\epsilon_{rot}$  according to equation (3) and find that the energy needed for breaking the pinning in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  at  $T = 4.2\text{K}$ ,  $H = 1500 \text{ Oe}$  is  $\epsilon_p = 7.8 \times 10^{-4} \text{ erg}$ . (We estimate the  $M_{irr}$  value by measuring the difference between the FC branch and the ZFC branch in zero rotating angle). Once the system gets enough energy to break the pinning, the amount of fluxons decreases in an irreversible process. The FC signal is not recovered when the sample is rotated back from  $2\pi$  to 0 (inset, fig. 2.a), demonstrating that the new configuration is energetically lower than the original FC configuration. This suggests that in high- $T_c$  superconductors, unlike spin glasses, the FC branch of the magnetization is an unstable branch. Measurements of the magnetization vs. time in those materials confirm this conclusion and show clear time dependence for both the ZFC and FC branches of the magnetization. (13)

To complete the experimental description, we compare the results with FC data for Nb (Figure 2.b). The figure exhibits  $M(\theta)$  dependencies for Nb which resemble those of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . There are, however, two important differences: (i)  $M(\theta)/H$  curves for Nb coincide for the various fields for most of the angular span. This result is surprising since  $M$  is expected to be linear with  $H$  only in the pure superconducting state. Here we see linearity of  $M$  and  $M_{irr}$  in the mixed phase. (ii) In

the high-field regime we observe a plateau above an angle  $\Theta_c(H)$ , implying that the magnetic moment is not capable of following the sample but the lag  $\Theta - \Theta_c$  is a constant (see inset, Figure 2.b). (Very similar results, though on a limited angular span, were obtained by Heise<sup>'14'</sup> in his torque-experiments). The plateau in high fields is limited to a small angular span and the overall shape of  $M(\Theta)$  is symmetric around  $\pi$  and is reversible, implying that  $M_{irr}$  is still a rigid body. We thus demonstrate that for Nb the macroscopic moment  $M_{irr}$  is able to recover after a full  $2\pi$  rotation. In other words, when  $\epsilon_{rot}$  is of the order of  $\epsilon_p$ ,  $M_{irr}$  is still a rigid body, but its orientation changes temporarily in order to decrease the system energy.

Recent theories<sup>'15-16'</sup> and simulations<sup>'15-17'</sup> predict field-induced frustration in granular superconductors, and as a result, glassy behaviour and irreversible response to magnetic field. The difference in the ZFC/FC branch, though consistent with the glassy picture, is found also in ordinary superconductors, and might be explained in terms of trapped flux. The rotation experiment, however, shows a clear distinction between the response of Nb and  $YBa_2Cu_3O_7$ . We note that the difference in the behaviour is really dramatic and is very unlikely that it is merely quantitative: For Nb we find rigid rotations at  $T/T_c \approx 0.5$  whereas for  $YBa_2Cu_3O_7$ , for comparable fields, the rigidity is broken for  $T/T_c < 0.05$ . Moreover, the behaviour found for  $YBa_2Cu_3O_7$  is very

similar to that found for classical spin-glasses. We therefore conclude that the rotation experiment gives support to the glassy picture suggested for the high- $T_c$  ceramics.

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Figure Captions

- (1) Zero-field-cooled (shielding) and field-cooled (Meissner) curves for (a)  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and (b) Nb.
  
- (2) Angular dependence of the FC magnetization for (a)  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and (b) Nb at 4.2K for various fields. The inset to (a) shows the irreversible angular dependence for  $H = 1.5\text{kOe}$ , at 4.2 K. The inset to (b) shows the lag  $\theta - \theta_0$  as a function of the rotation angle  $\theta$  for  $h = 1\text{kOe}$ .

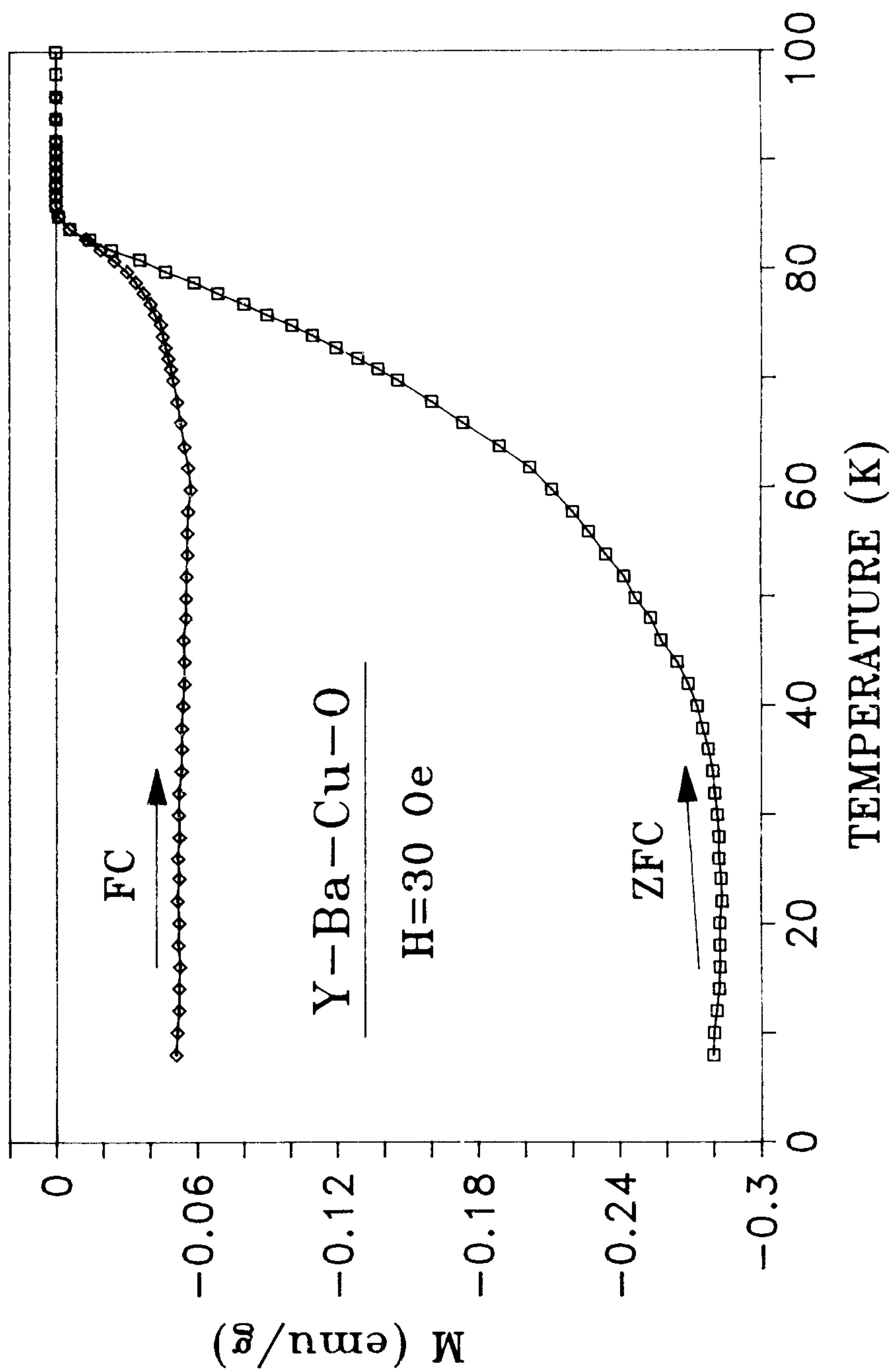


FIGURE 1a

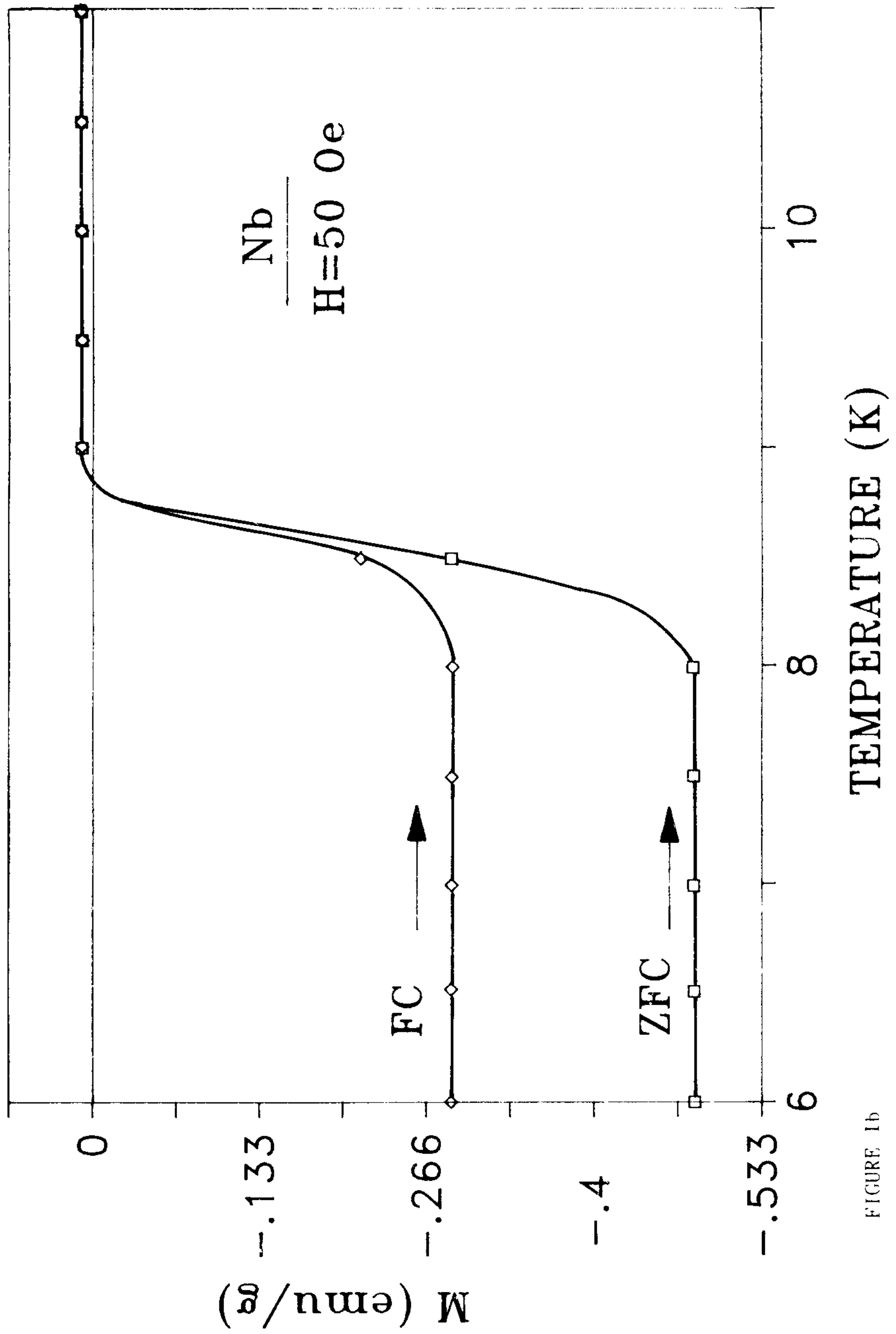


FIGURE 1b

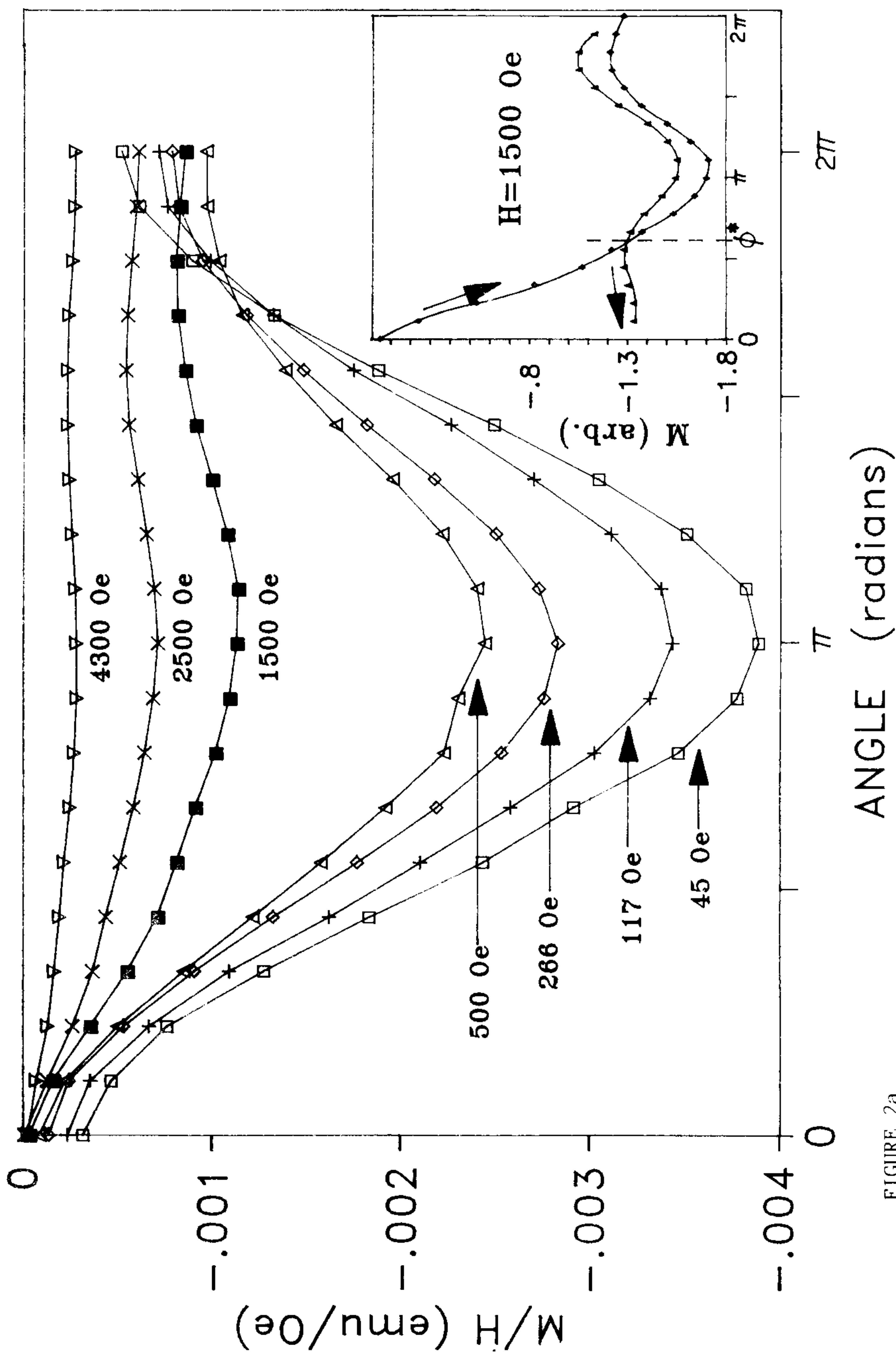
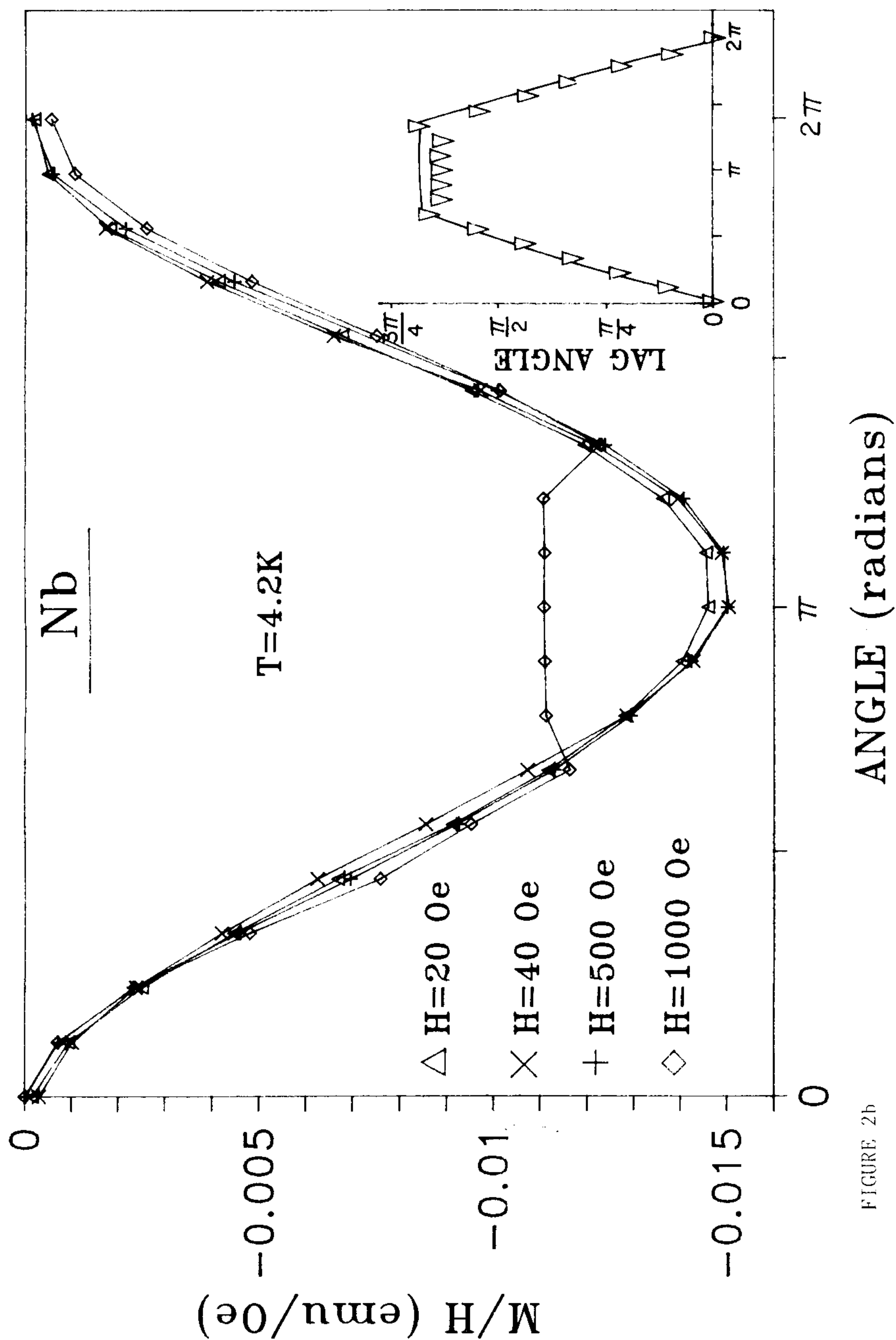


FIGURE 2a





ANGLE (radians)

FIGURE 2b