View metadata, citation and similar papers at core.ac.uk
brought to you by CORE
provided by NASA Technical Reports Server

N91-24082

# MELT-PROCESSED BULK SUPERCONDUCTORS: FABRICATION AND CHARACTERIZATION FOR POWER AND SPACE APPLICATIONS

**Hamid Hojaji, Aaron Barkatt and Shouxiang Hu**
**The Catholic University of American**
**Washington, D.C. 20059**

**Arthur N. Thorpe, Matthew F. Ware and David Davis**
**Howard University**
**Washington, D.C. 20059**

**Sidney Alterescu**
**NASA Goddard Space Flight Center**
**Greenbelt, MD 20910**

## ABSTRACT

Melt-process bulk superconducting materials based on variations on the base $YBA_2Cu_3O_x$ have been produced in a variety of shapes and forms. Very high values of both zero-field and high-field magnetization have been observed. These are useful for levitation and power applications. Magnetic measurements show that the effects of field direction and intensity, temperature and time are consistent with an aligned grain structure with multiple pinning sites and with models of thermally activated flux motion.

## HIGH-MAGNETIZATION SUPERCONDUCTING CUPRATES; FORMULATION, PROCESSING AND MICROSTRUCTURE

Recent studies conducted at the authors' laboratories have shown that the combination of melt processing of Y-Ba-Cu-O superconducting materials with the use of non-stoichiometric formulations makes it possible to produce materials with very high magnetization [1, 2]. Zero-field dc magnetization values of up to 18 emu/g and remanent magnetization ($M_+$ - M) values of up to 21 emu/g at a field intensity of 5 kOe have been observed at 77 K. The non-stoichiometric compositions mentioned above involve additions of excess yttrium, rare earths such as Gd, Tb, Ho and Yb, or transition metals such as Nb, to the base $YBa_2Cu_3O_x$ composition. The high magnetization of these materials has been concluded to be due to the controlled formation of a highly oriented grain structure, interspersed with second phase sites (in particular, $Y_2BaCuO_5$) which promote flux pinning. This approach has been shown suitable for the production of large pieces which have been fabricated to give a variety of shapes, including cylinders, bars, plates,a nd prototype bearing elements. Measurements of levitation forces and of magnetic stiffness gave high values for these materials, which were correlated with their high magnetization [3]. This makes these materials attractive for levitation applications such as levitation bearings. In addition, the approach described above has been shown to be suitable for the controlled production of materials with different types of hysteresis loops depending on the composition of the samples and on the procedure used to prepare them. In particular, it is possible to obtain, on one hand, materials with a high degree of flux trapping, i.e., a large remanent magnetization in zero field, and, on the other hand, materials which effectively pin the flux at higher fields. This increases the scope of applications of the materials to include, for instance, magnetic energy storage.

The melt-processed samples described above were recently subjected to extensive measurements of their magnetic properties, with an emphasis on flux trapping and flux creep, and on their magnetomechanical behavior as related to levitation and suspension.

## ANISOTROPIC MAGNETIC FEATURES AND FIELD DEPENDENCE

The measurement of dc magnetization and magnetic susceptibility was described in earlier paper [1, 4]. All the data in the present paper relate to a single melt-processed sample of $YBa_2Cu_3O_x$ without any additional dopants. The magnetization values obtained with this sample are somewhat lower than those

C-5

Y

obtained recently with the doped samples mentioned above; detailed magnetic studies of the latter samples are currently in progress.

It has been recently found that even though the melt-processed samples are polycrystalline, the effect of their grain orientation with respect to the applied magnetic field is very important. The effect of varying the orientation of a high-magnetization sample on the magnetization values at 77 K is shown in Figure !. It can be seen that rotation of the sample at an angle of $90^0$ with respect to the direction at which the maximum magnetization was observed caused the magnetization to drop by a factor of approximately 3.5, viz. from 15 emu/g to 4.3 emu/g. The combination of results obtained at the two extreme orientations with those obtained at other angles gives excellent agreement with a simple two-component model, according to which

$$M_r = M_r \parallel + (M_r \perp - M_r \parallel) * \cos^2 \theta \qquad (1)$$

where $M_r$ is the magnetization measured when the applied magnetic field, oriented at an angle $\theta$ with respect to the c axis of the $YBa_2Cu_3O_7$ grains, is removed. $M_r \parallel$ and $M_r \perp$ are the zero-field magnetization values obtained after applying fields parallel and perpendicular to the c axis or ab plane, respectively. The agreement between the model and the observed results is shown in Figure 2. The results show an excellent linear correlation between $M_r$ and $\cos^2 \theta$. The angular dependence of the magnetization observed in the present case is consistent with the behavior of the flux density in a layered superconductor with weak coupling between adjacent layers as analyzed by Kes et al [5]. However, in the present case better linear correlation was observed between $M_r$ and $\cos^2 \theta$ than between $M_r$ and $\cos \theta$.

The dependence of the remanent magnetization at 77 K on the maximum applied magnetic field is shown in Figure 3 for field intensities of up to 10 kOe. Both the values obtained in the direction of the c axis and those measured in the direction perpendicular to it are included. It can be seen that the magnetization observed in both directions levels off at fields of 2-3 kOe. A measurement of the initial slop of the M - H dependence was performed using very small applied fields. Under these conditions, the relationship between M and H is linear, as shown in Figure 4, and the values of the residual magnetization after removal of the field are very low, as shown in Figure 5.

## ANISOTROPY IN THE CRITICAL CURRENT DENSITIES

Bean's model [6] has been extensively used to obtain critical current densities, $J_c$, magnetization curves. Good agreement has been observed in several studies between the observed transport current density and the predictions of Bean's model [5a, 5b]. Recently, Hu et al [5c], developed an extended form of Bean's model for the specific case of a grain-aligned bulk superconductor. A simplified version of the equation in the case where the applied field H is parallel to the c-axis is given by:

$$\Delta M = \frac{1}{30} J_c^{ab} d \qquad (2)$$

where $J_c^{ab}$ is the critical current density parallel to the ab plane and d is the smaller dimension of the grains in the ab plane. With d = 0.2 cm, $\Delta$ M = 190 emu/cm$^3$ at H = 34 Oe, and $T_c$ = 77 K, $J_c^{ab}$ is calculated to be equal to 2.8 X $10^4$ A/cm$^2$. At H = 2 kOe, $J_c^{ab}$ is equal to 1.6 X 104 A.cm$^2$.

When the applied field is perpendicular to the c-axis, the calculated J values decrease to 7.3 X $10^3$ and 2.9 X $10^3$ A/cm$^2$ at 300 Oe and a 2 kOe, respectively.

The anisotropy in $J_c$, i.e., the ratio between the $J_c$ values corresponding to the two directions of the applied field, is about 4 at low fields, and it increases to about 5.5 at 2 kOe. Compared with the anisotropy of the single crystal [5d], which is about 175 at 40 K and H = 8 kOe, the much lower degree of anisotropy of the melt processed material characterized in this work is attributed to the much larger number of pinning sites uniformly distributed throughout the material.

## DEPENDENCE OF THE MAGNETIZATION ON TEMPERATURE AND TIME

Upon the dependence of the of the dc magnetic susceptibility and magnetization on temperature, it is observed that the values of these two magnetic properties decrease as the temperature is raised and approaches $T_c$. The data in Figure 6 represent magnetic susceptibility measurements in a very low field (110 Oe) at increasingly higher temperatures following the application and removal of a 2.5-kOe field at the lowest temperature of 77 K. Figure 7 likewise represents magnetization values obtained upon cooling to 66 K, applying a 10-kOe field and removing it, and then gradually raising the temperature of the sample. It is important to note that when the warm-up is interrupted and the temperature is lowered before being raised again, the magnetic susceptibility and the remanent magnetization remain constant until the temperature increases beyond the point at which the warm-up was interrupted. This shows that these experiments give a true measure of the dependence of the capability of the samples to trap magnetic flux, and that the results are not significantly affected by the interruptions in the heating schedule. The ensuing data can be interpreted as reflecting the role of thermal activation in flux trapping. (It should be noted that the apparent leveling off of the values of the magnetic properties at the lowest temperatures employed in the measurements shown in Figures 6 and 7 is an experimental artifact).

The temperature effects on the apparent activation energy of flux trapping are not strongly dependant on the orientation. Upon measuring the dc magnetization in the direction which yields the highest values, i.e., the direction perpendicular to the ab planes, the zero-field readings on the sample which was used to obtain the data in Figure 7 at temperatures of 77 K and 67 K were 15.3 and 37.6 emu/g, respectively. The corresponding readings in the direction parallel to the ab planes were 7.2 emu/g and 12.5 emu/g. The resulting ratio between $M_r \perp$ and $M_r \parallel$ was 2.99 at 77 K and 3.02 at 67 K. The observed temperature dependence in the direction perpendicular to the ab planes between 66 and 79 K can be described in terms of an activation energy of 3.86 kJ/mol or 0.0399 eV. A very similar activation energy was obtained when the temperature dependence was obtained when the residual magnetization at an applied field of 2.4 kOe was measured instead of the zero-field magnetization. It is important to note that the near-linear dependence of the magnetization on the temperature breaks down as the critical temperature $T_c$ is approached and is replaced by a tailing curve with progressively decreasing slope. Similar behavior has been observed upon examining the temperature dependence of the critical current density $J_c$ in thin films of $YBa_2Cu_{3O}x$ and flux creep [12] and which has been shown to be relatively insensitive to the presence of weak links [11].

Long-term measurements of the residual zero-field magnetization at a constant temperature of 77 K were carried out in order to characterize the rate of its decay as a result of flux creep. The results are shown in Figure 8. It can be seen that the decay of the magnetization in each of the two directions specified above follows a logarithmic rate law of the type:

$$M_r(t) = M_r(0)*[1-kT/E*\ln(1+t/t_0)] \qquad (3)$$

where $M_r(o)$ and $M_r(t)$ are the magnetization values observed initially and after time t, respectively, E is the activation energy obtained above, and $t_0$, is a characteristic decay time, amounting approximately $2*10^3$ seconds at 77 K. This model was originally developed by Hagen et al [13, 14] to describe thermally activated flux motion in high-$T_c$ superconductors consisting of many pinning regions.

In conclusion, the decay of $M_r$ with time in high-magnetization materials produced by melt processing of on-stoichiometric variants of $YBa_2Cu_3O_7$, together with the dependence of the magnetization on temperature and on grain orientation, are all consistent with the picture of a structure with multiple pinning regions which is subject to thermally activated flux motion. Furthermore, this picture is consistent with the oriented grain structure of these materials [1, 2], which contains micron- and submicron-sized second phase regions likely to provide sites for flux pinning.

## ACKNOWLEDGEMENTS

This study was supported by the National Aeronautics and Space Administration under Contract No. NAG 5-1017.

## REFERENCES

1. Hojaji, H., Barkatt, A., Michael, K. A., Hu. S., Thorpe, A. N., Ware, M. F., Talmy, I. G., Haught, D. A., and Alterescu, S.: Yttrium Enrichment and Improved Magnetic Properties in Partially Melted Ya-Ba-Cu-O Materials. J. Mater. Res. Vol. 5, No. 4, 1990, pp. 721-730.

2. Hojaji, H., Barkatt, A., Hu, S., Michael, K. A., Thorpe, A. N., Talmy, I. G., Haught, D. A., and Alterescu, S.: Superconducting Cuprates Prepared by the Melt Quench Process and Containing Excess Y or Additives. Mat. Res. Bull. Vol. 25, No. 6, 1990, pp. 765-777.

3. Moon, F. C., Chang, P. -Z., Hojaji, H., Barkatt, A., and Thorpe, A. N.: Levitation Forces, Relaxation and Magnetic Stiffness of Melt-Quenches $YBa_2Cu_3O_x$. Japan. J. Appl. Phys., in press.

4. Hojaji, H., Michael, K. A., Barkatt, A., Thorpe, A. N., Ware, M. F., Talmy, I. G., Haught, D. A., and Alterescu, S.: A Comparative Study of Sintered and Melt-Grown Recrystallized $YBa_2Cu_3O_x$. J. Mater. Res. Vol. 4, No. 1, 1990, pp. 28-32.

5. Kes, P. H., Aarts, J., Vinokur, V. M., and van der Beek, C. J.: Dissipation in Highly Anisotropic Superconductors. Phys. Rev. Lett. Vol. 64, No. 9, 1990, pp. 1063-1066.

6. Bean, C. P.: Magnetization of Hard Superconductors. Phys. Rev. Lett. Vol. 8, No. 6, 1962, pp. 250-253.

7. Murakami, M., Morita, and Koyama, N.: Magnetization of a $YBa_2Cu_3O_7$ Crystal Prepared by the Quench and Melt Growth Process. Japan. J. Appl. Phys. Vol. 28, No. 7, 1989, pp. L1125-L1127.

8. Salama, K, Selvamanickan, V., Gao, L., and Sun, K.: High Current Density in Bulk $YBa_2Cu_3O_x$ Superconductor. Appl. Phys. Lett. Vol. 54, No. 23, 1989, pp. 2352-2357.

9. Hu, S., Hojaji, H., Barkatt, A., Boroomand, M., Thorpe, A. N., and Alterescu, S.: Anisotropic Electromagnetic Features of a Grain-Aligned $YBa_2Cu_3O_x$ Bulk Superconductor. Phys. Rev. B, accepted for publication.

10. Swartzendruber, L. J., Roitburd, A., Kaiser, D. L., Gayle, F. W., and Bennett, L.: Direct Evidence for an Effect of Twin Boundaries on Flux Pinning in Single-Crystal $YBa_2Cu_3O_{6+x}$. Phys. Rev. Lett. Vol. 64, No. 4, 1990, pp. 483-486.

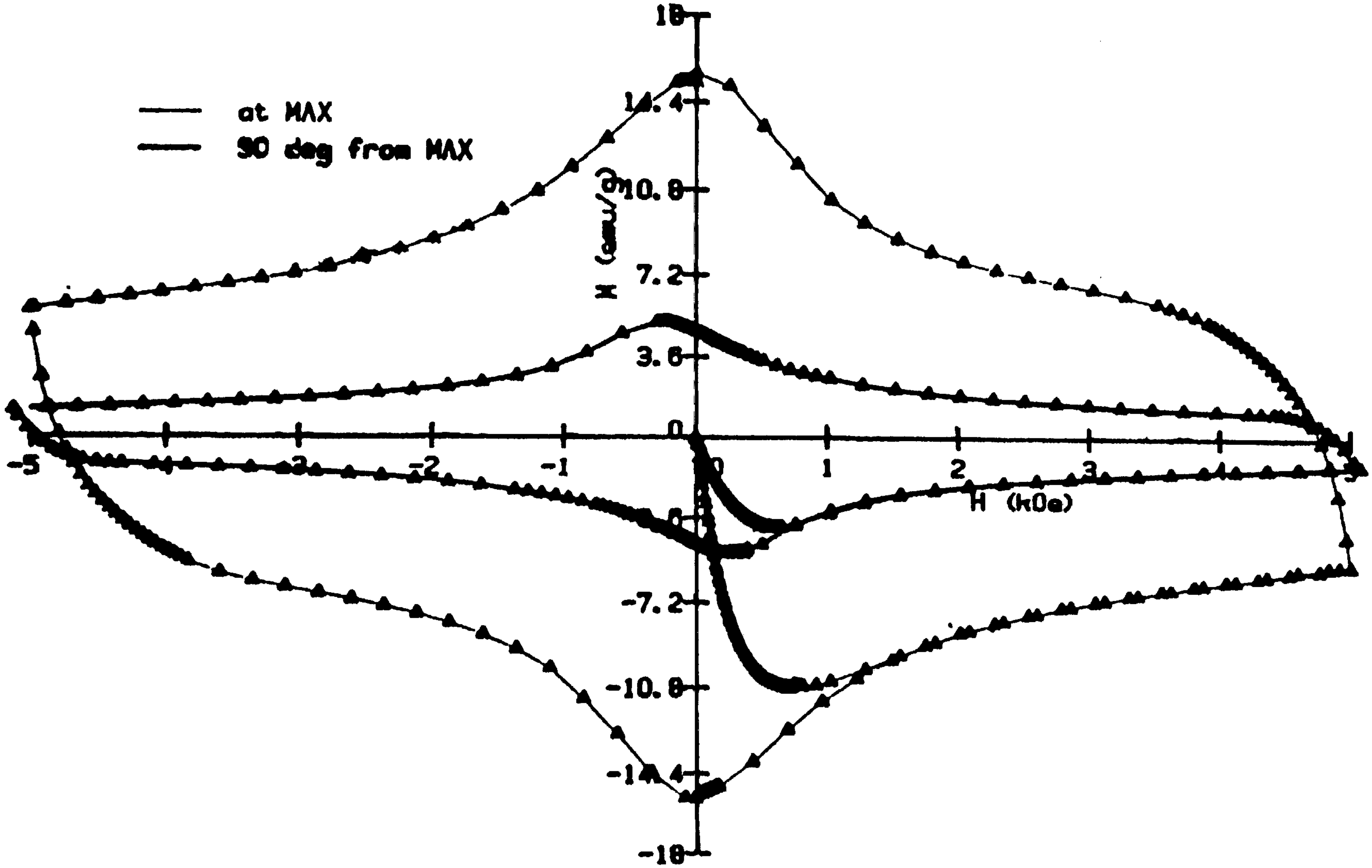
11. Savvides, N.: Flux Creep and Transport Critical Current Density in High-$T_c$ Superconductors. Physica. C Vol. 165, Nos. 5-6, 1990, pp. 371-376.

12. Anderson, P. W.: Theory of Flux Creep in Hard Superconductors. Phys. Rev. Lett. Vol. 9, No. 7, 1962, pp. 309-311.

13. Hagen, C. W., Griessen, R. P., and Salomons, E.: Thermally Activated Flux Motion in High-$T_c$ Superconductors: An Analytical Model. Physica. C Vol. 157, No. 2, 1989, pp. 199-208.

14. Mohamed, M. A.-K., Jung, J., and Franck, J. P.: Flux Creep and Penetration in Fe-doped $YBa_2Cu_3O_7$. Phys. Rev. B Vol. 41, No. 7, 1990, pp. 4286-4295.

Figure 1



T=LN2, mag moment=10; delay=100sec

Figure 2

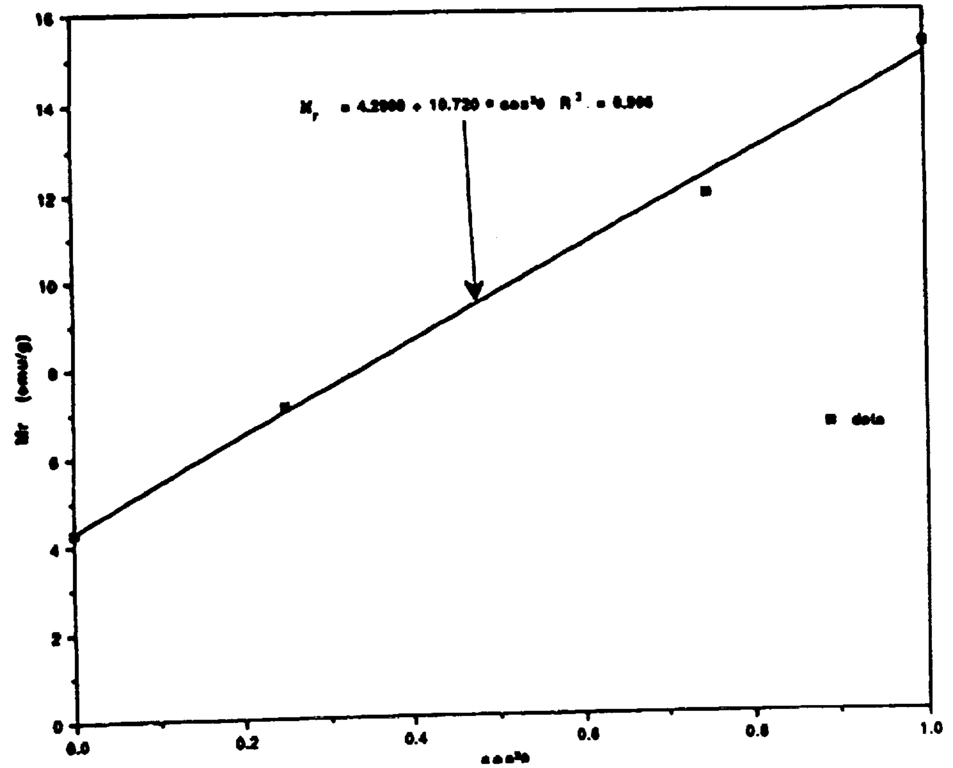


Figure 3

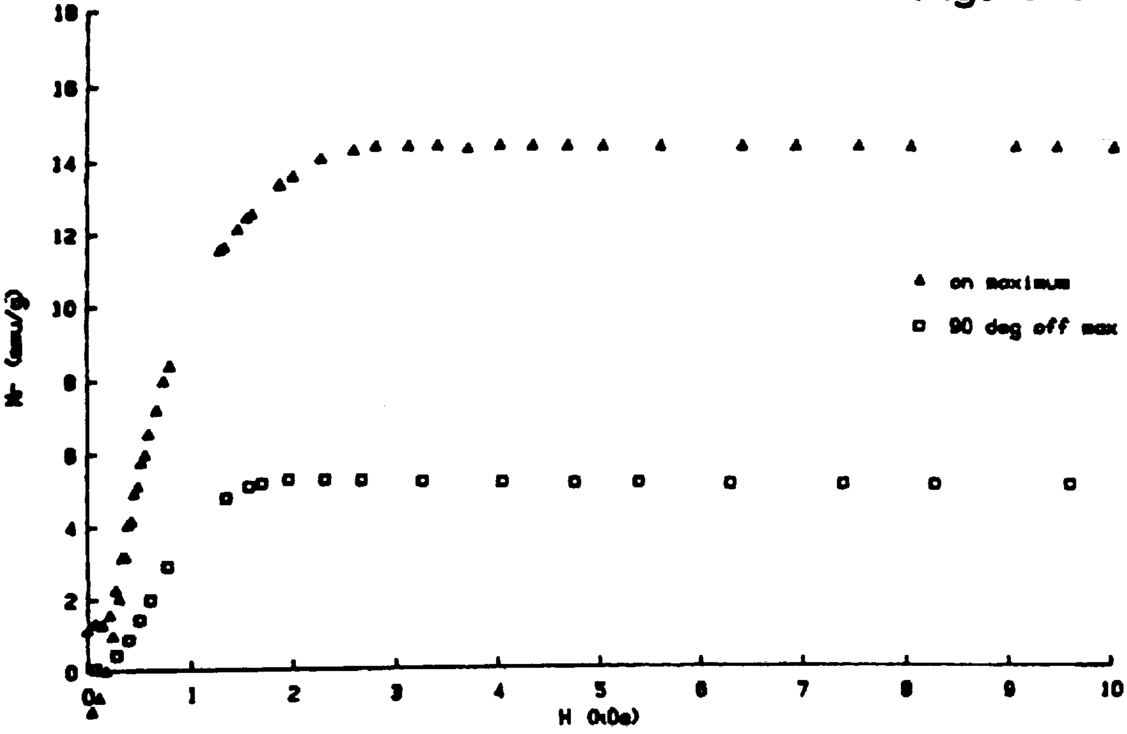


Figure 4

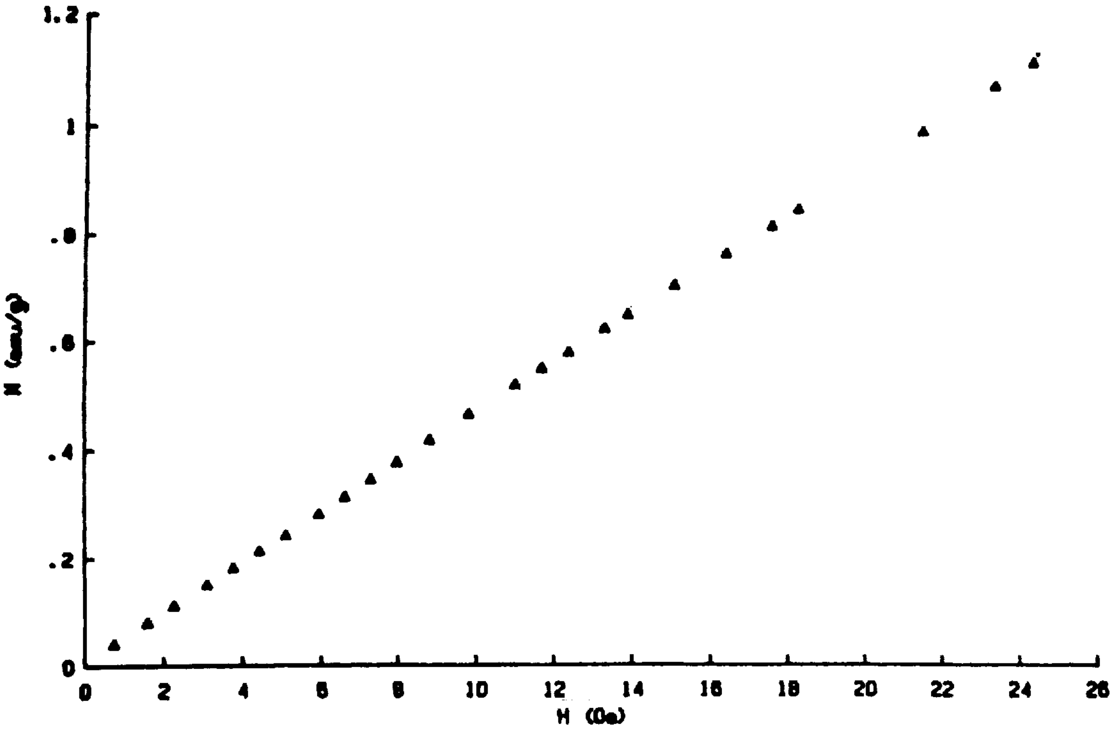


Figure 5

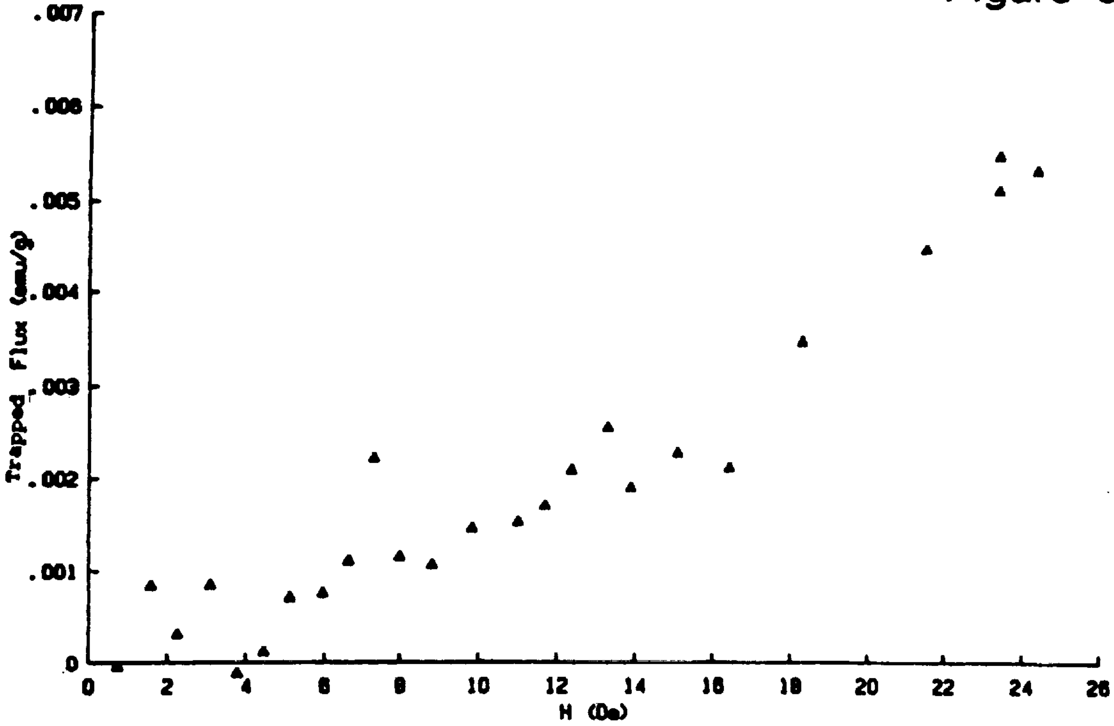


Figure 6

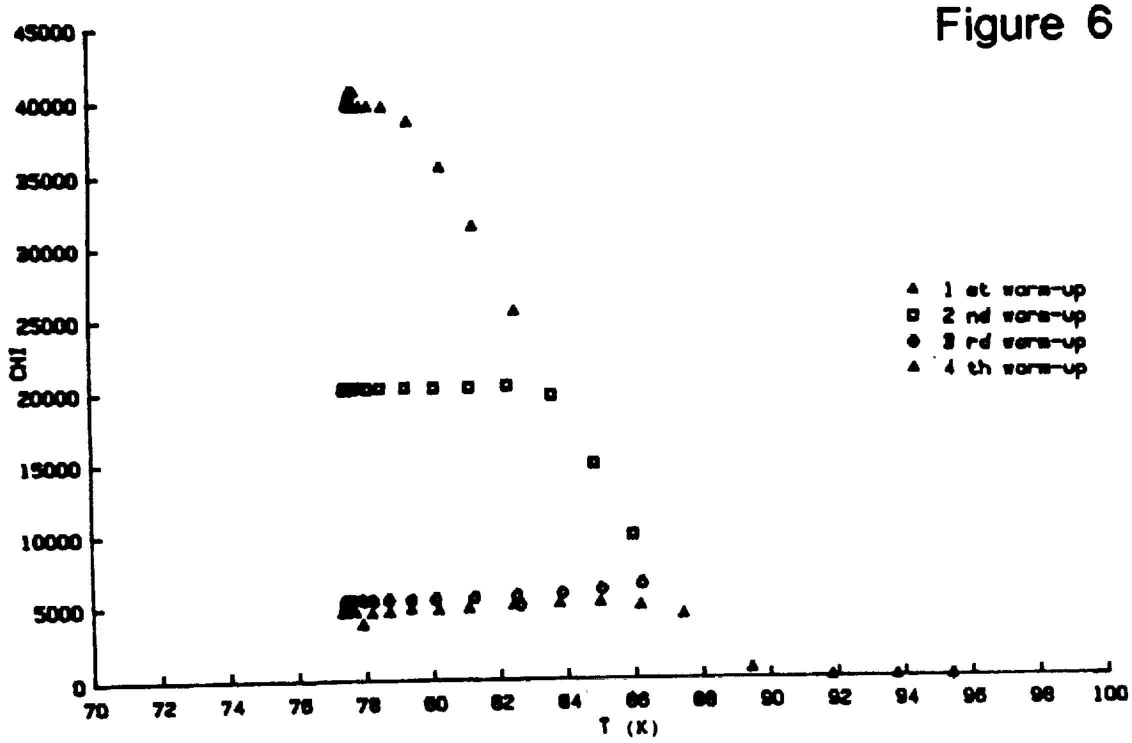


Figure 7

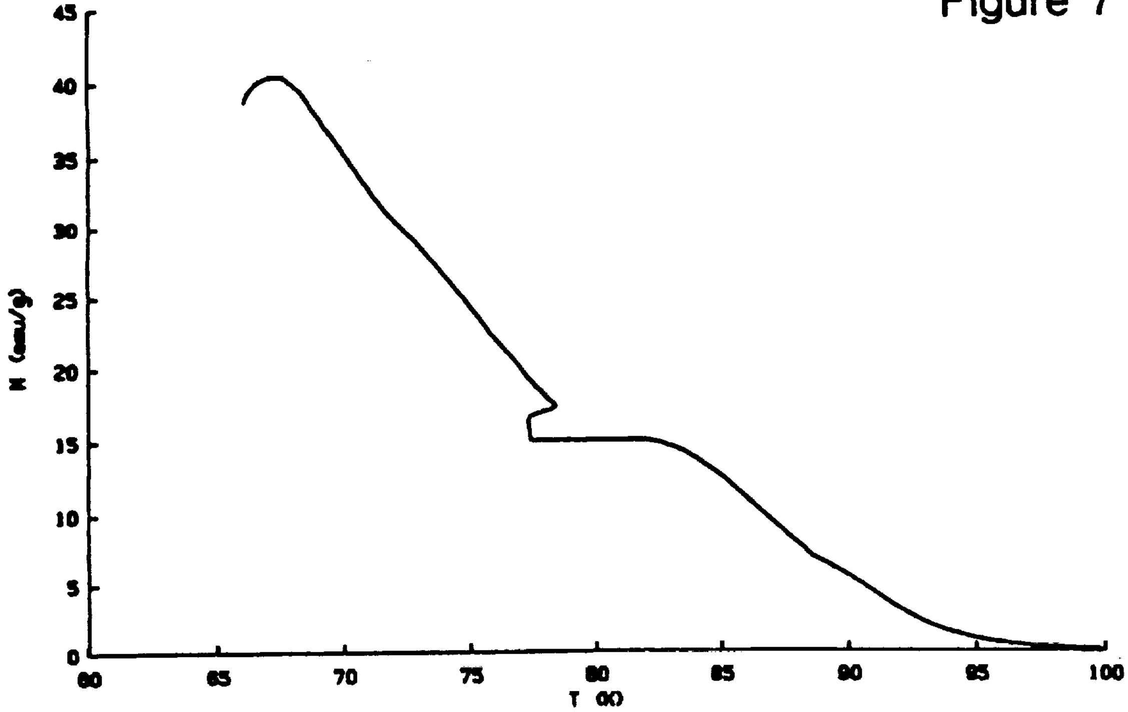
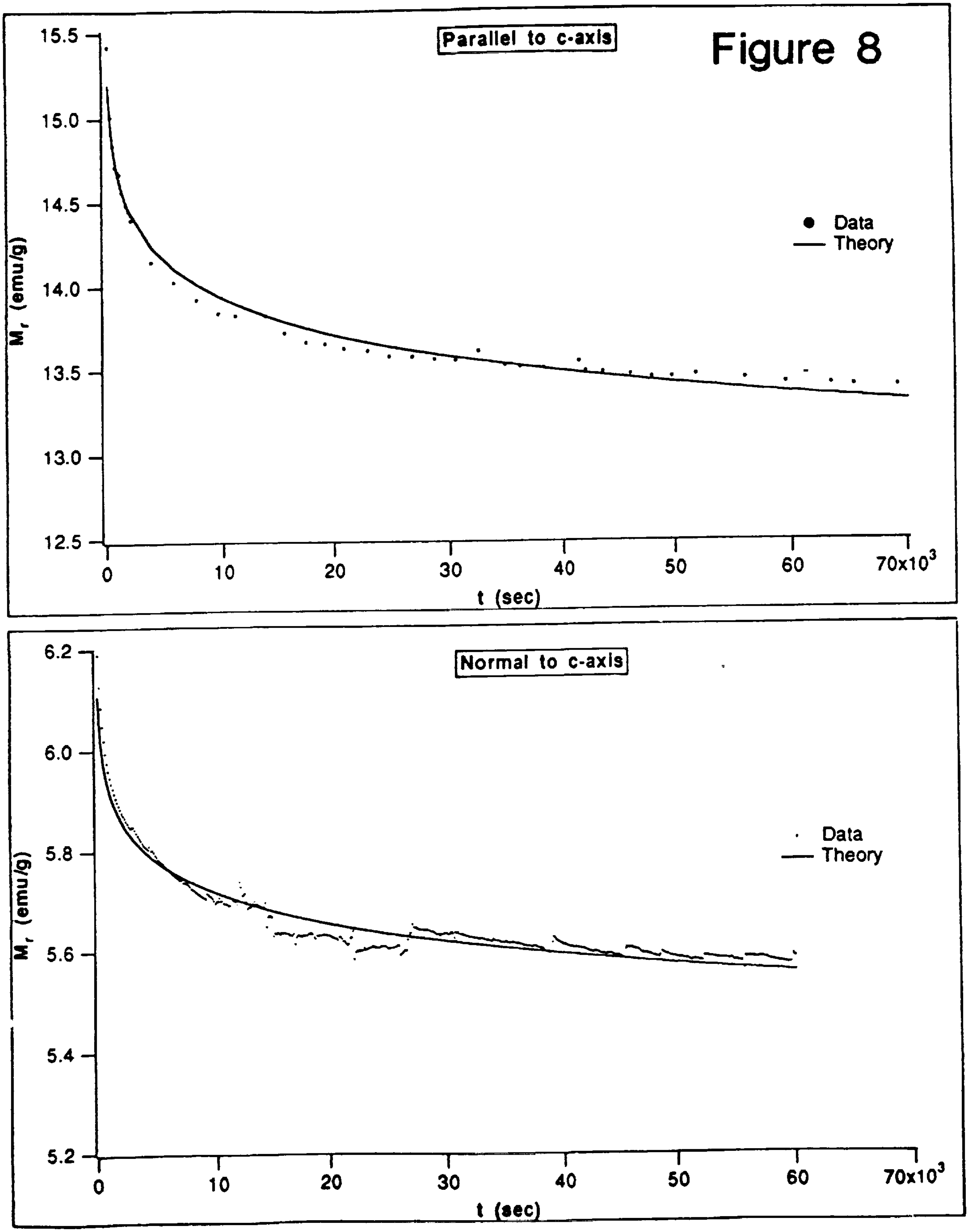


Figure 8


Parallel to c-axis
15.5
15.0
14.5
14.0
13.5
13.0
12.5
$M_r$ (emu/g)
● Data
— Theory
0
10
20
30
40
50
60
70x10$^3$
t (sec)
Normal to c-axis
6.2
6.0
5.8
5.6
5.4
5.2
$M_r$ (emu/g)
· Data
— Theory
0
10
20
30
40
50
60
70x10$^3$
t (sec)