ISSN 0029-3865

CBPF-NF-007/90

INTRINSIC DOMAIN WALL PINNING IN $Hf(Fe_{0.5}Al_{0.5})_2$

by

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Abstract

The magnetization vs. temperature curve for $Hf(Fe_{0.5}Al_{0.5})_2$ in low applied field presents a maximum and also an irreversibility up to near the Curie temperature. For temperatures lower than 7 K approximately, a critical field (propagation field) exist in the virgin magnetization curves. Even in high fields (70 kOe) the magnetization values are not saturated, suggesting high anisotropy energy and consequently narrow domain walls. An exponential temperature decrease of the coercive field is observed. These phenomena can be explained by intrinsic domain wall pinning. The time effect in the demagnetization region of the hysteresis loop was measured and it is related to the wall deformation (kink creation). A brief comparison with other systems is done.

Key-words: Intrinsic pinning; Laves phases; Intermetallic compounds; $Hf(Fe_{0.5}Al_{0.5})_2$; $A(Fl_{1-x}Al_x)_2$ A = (Y,Zr); Spin-glass.

Introduction

Some interesting magnetic properties arise on pseudo-binary intermetallic compounds $A(Fe_{1-x}Al_x)_2$ (A = Y, Zr) and $R(Fe_{1-x}Al_x)_2$(R = Dy, Er and Tb). According to Hilscher et al. [1] the Y system has the cluster glass regime in the concentration range from x = 0.10 to 0.35, while no freezing phenomena was observed up to the critical concentration x_c= 0.55 for $Zr(Fe_{1-x}Al_x)_2$. We have studied the compound $Hf(Fe_{0.5}Al_{0.5})_2$ to understand the role of Hf in the $A(Fe_{1-x}Al_x)_2$ system, in the intermediate concentration range. In this compound $Hf(Fe_{0.5}Al_{0.5})_2$ the magnetization vs. temperature curves shows a maximum and an irreversible behavior. The virgin magnetization vs. field curve at temperatures lower than 7 K approximately, presents a critical field (propagation field). For higher fields the magnetization approaches slowly to the saturation but even in high fields (70 kOe) the magnetization is not saturated. This behavior also arises in systems like $R(Fe_{1-x}Al_x)_2$(R = Dy, Er and Tb) [2-5], which have a high anisotropy energy and therefore, very narrow domain walls are present. For these narrow walls Zijlstra [6] showed that there is a difference of energy when the center of the wall is just at an atomic plane or just between two planes. This energy difference is an energy barrier and the wall energy along the crystal is

$$V(z) = V_0 + \Delta V\ \mathrm{sen}^2(\pi z/D) \qquad (1)$$

where V_0 is the wall energy in a minima potential, ΔV the energy barrier and D the interplanar distance. ΔV gives rise to an intrinsic coercive field

$$H_c = {}^1/_{2M_S} (\partial V/\partial Z)_{max} = {}^{\pi}/_{2M_S}\ {}^{\Delta V}/_D \ . \qquad (2)$$

Hilsinger and Kronmüller [7] have shown that this intrinsic coercivity is greater for narrow walls through a relationship $H_c \sim \Delta V \sim e^{-\pi(\delta/D)}$, where δ is the wall width. This intrinsic coervive force was observed in Dy single crystal [8] and more recently by Arnaudas et al. [9,10] in compounds $(Dy_xY_{1-x})Al_2$, $(Tb_xGd_{1-x})Al_2$ and $ErAl_2$. In these systems it was also observed a time effect in the demagnetization region of the hysteresis loop . This occurs because the walls move by thermal activation through a mechanism known as kink creation [11,12]. According to this model the activation energy is proportional to ${}^1/_{H_i}$, where H_i is the internal field. Such time dependence was also observed in $Hf(Fe_{0.5}Al_{0.5})_2$

In the present paper, we show the DC-magnetization vs. field, temperature and time, the temperature dependence of the coercive field and also the AC-susceptibility curve of the $Hf(Fe_{0.5}Al_{0.5})_2$, in order to elucidate the magnetization process at low temperature, and to compare with the systems mentioned above.

Experimental

The sample was prepared by arc-melting furnace under purified argon atmosphere in stoichiometric proportion. Elements were supplied by Johnson Matthey Chemicals LTD, Hf with 99.9% purity, Fe and Al with 99.999%. The single phase was ensured by X-ray diffractogram at room temperature. $Hf(Fe_{0.5}Al_{0.5})_2$ has a hexagonal C-14 Laves phase structure and the lattice parameters are a = 5.093 Å , c = 8.307 Å and c/a = 1.631 (They were extrapolated using Nelson-Riley function). The magnetization measurements were made using a PAR Vibrating Sample Magnetometer. The Curie temperature was obtained by extrapolation of the M^2 vs. T curve to $M^2 = 0$, for magnetization measurements in low field (60 Oe) and also by the maximum of χ_{AC}.

Results and Discussion

The magnetization vs. temperature measurements are shown in fig. 1. We note three important aspects in these curves:

1) The zero field cooling curve (ZFC) is almost constant up to 12 K approximately, above that there is a rapid increase. The applied field used was 60 Oe. There is a difference on the magnetization curves when the sample is cooled with and without applied field (Fig. 1a). The ZFC curve is irreversible, in contrast with field cooling (FC) curve that is reversible. This irreversibility happens from low temperatures up to near the Curie temperature.
2) For greater applied fields the maximum in ZFC curves is shifted to lower temperatures (Fig. 1b).
3) The χ_{AC} susceptibility has a sharp maximum at 32 K (Fig. 1c).

In the $Y(Fe_{1-x}Al_x)_2$ (0.1<x<0.35) there is also an irreversibility in the magnetization vs. temperature curves but related to spin-glass regime [1]. This irreversibility takes place from low temperatures up to the freezing temperature T_f, however, for the $Hf(Fe_{0.5}Al_{0.5})_2$ compound it takes place up to near the Curie temperature T_C. This is an important difference between these systems. In stoichiometric $Zr(Fe_{1-x}Al_x)_2$ no irreversibility was observed [1], although Hilscher [13] has used a higher applied field (500 Oe) in his measurements.

In the fig. 2 we show magnetization vs. applied field curves for some temperatures. The sample was always cooled in zero applied field. There is a more rapid increase after a critical field (propagation field).This critical field exist at temperatures below 7 K approximately. For higher fields the magnetization approaches slowly to the saturation but even in high fields (70 kOe) the magnetization is not saturated. It suggests a high anisotropy energy and consequently narrow domain walls, since the wall width is given by $\delta \sim \sqrt{A/K}$, where A and K are the exchange and anisotropy constants, respectively. $Zr(Fe_{1-x}Al_x)_2$ also shows high anisotropy energy and it is probable that this anisotropy supports a ferromagnetic alignment, avoiding the spin freezing effects [14].

We show in fig.3 the ZFC hysteresis loop at 2.0 K, where the maximum applied field is 13 kOe. No difference between ZFC and FC hysteresis loop was detected. We observe a time variation on the magnetization values in the range between 0 and H_c, where H_c is the coercive field. Although the observed effects of no saturation and of magnetization time dependence, the coercive field was defined as the field value corresponding to null magnetization. In fig.4 is presented a strong temperature dependence of the coercive field, which can be fitted by an exponential. This rapid decrease is closely related to the decrease of the wall pinning and it is responsible for the magnetization process at low temperatures.

To characterize the time effect, we stop the hysteresis loop on zero field and the time variation of the remanent magnetization is measured. We did it for some temperatures below Curie temperature. These curves can be fitted (fig.5) by,

$$M(t) = M_{To} - cte \ln t \qquad (3)$$

where M_{To} is a different constant for each temperature. The time effect is greater for fields near H_c. This time relaxation can be understood through the deformation of the wall surface (kink creation) [11,12] by a thermal activation process. The probability of the wall portion to jump one atomic spacing is

$$p = cte\ e^{-E_a/k_B T} \qquad (4)$$

where E_a is the activation energy and this probability is related to the time variation of the magnetization,

$$dM/_{dt} = cte\ e^{-E_a/k_B T} \qquad (5)$$

According to Egami [11], the energy variation with a kink creation is given by,

$$E = 2\pi r\sigma - \pi r^2 (2MH_i) \qquad (6)$$

for a cylindrical deformation, where σ is the kink energy per unit length, M is the magnetization per unit area of the atomic layer and H_i is the internal field. For the equilibrium condition, we obtain the critical radius and the activation energy,

$$r_c = {}^{\sigma}/_{2MH_i} \text{ and } E_a = {}^{\pi\sigma^2}/_{2MH_i} \qquad (7)$$

An interesting expression is obtained by substitution of the activation energy into Eq(5),

$$\ln ({}^{dM}/_{dt}) = \text{cte} - {}^{\pi\sigma^2}/_{2M}\ {}^{1}/_{k_B T}\ {}^{1}/_{H_i} \qquad (8)$$

This linear dependence $\ln({}^{dM}/_{dt}) \times {}^{1}/_{H_i}$ is observed for $Hf(Fe_{0.5}Al_{0.5})_2$ (fig.6).

According to Kütterer et al. [15] the coercive field has two contributions: the intrinsic coercive field (intrinsic pinning, characteristic of narrow domain walls), and the extrinsic coercive field (extrinsic pinning which is produced by all kinds of defects). At low temperatures, the first one dominates. Taking into account the rapid decrease of coercive field in the compound $Hf(Fe_{0.5}Al_{0.5})_2$, we believe that the coercivity is mainly produced by intrinsic pinning. This intrinsic pinning is present in several systems: $(Dy_xY_{1-x})Al_2$, $(Tb_xGd_{1-x})Al_2$, $ErAl_2$ [10,11], $R(Fe_{1-x}Al_x)_2$ (R = Dy, Tb and Er) [2-5]. All these systems have high anisotropy energy and narrow domain walls.

In the ZFC magnetization vs. temperature curve for 60 Oe (fig. 1a) the magnetization values below 12 K are almost constant, because the wall can not move, due to wall pinning. Only when the wall energy is high enough to deblock the wall (due to thermal energy) the magnetization values increase. For the magnetization vs. field curve a similar effect occurs, but in this case the energy is provided through the external applied field and moments interaction. When the sample is cooled in the presence of field, starting at any temperature , the walls can not move and we see the temperature dependence of spontaneous magnetization. These FC curves are reversible because there is not wall motion, existing only a spontaneous magnetization temperature dependence.

The values of the Curie temperature T_C determinated by extrapolation of the M^2 vs. T curve to $M^2 = 0$ and by the χ_{AC} are very close, say 33 and 32 K, respectively. Above T_C, $1/\chi$ vs. T obeys the Curie-Weiss law. The paramagnetic Curie temperature θ_P is 31 K and the effective magnetic moment μ_{eff} is 3.34 μ_B/Form. The magnetic moment obtained by extrapolation of the M vs. 1/H curve to 1/H = 0 on fields up to 70 kOe is 0.35 μ_B/Form. We don't know if the whole moment is attributed to iron. In recent papers, has been reported the existence of moment at A sites in AFe_2 (A = Y, Zr and Hf). From band structures calculations [16,17] it was shown to exist a small magnetic moment at the A site antiparallel to the Fe moment. More recently an experiment using polarized neutrons in YFe_2 [18], found $m_Y = -0.67$ μ_B and $m_{Fe} = 1.77$ μ_B. For the $Hf(Fe_{0.5}Al_{0.5})_2$ the iron moment is 0.35 μ_B or 0.78 μ_B if the Hf moment is nil or -0.43 μ_B [17] (calculated to Hf in $HfFe_2$), respectively. Our measurements are not able to clarify this problem and $Hf(Fe_{0.5}Al_{0.5})_2$ may be ferro or ferrimagneticaly ordered below T_C.

Conclusion

$Hf(Fe_{0.5}Al_{0.5})_2$ has a pure hexagonal structure, in contrast to the admixture of C-15 cubic and a small portion of C-14 hexagonal phases in $HfFe_2$. The maximum and the irreversibility behavior of the magnetization vs. temperature curve below T_C, the presence of the critical field below 7 K approximately, in the virgin magnetization vs. field curve, can be explained by intrinsic domain wall pinning and they are not cluster glass effects, like those observed in the $Y(Fe_{1-x}Al_x)_2$ system. These intrinsic pinning effects also are present in other systems like Dy single crystal [8], $(Dy_xY_{1-x})Al_2$, $(Tb_xGd_{1-x})Al_2$, $ErAl_2$ [10,11], $R(Fe_{1-x}Al_x)_2$ (R = Dy, Tb and Er) [2-5]. All these systems show high anisotropy energy and consequently have narrow domain walls. The time effect in the demagnetization region of the hysteresis loop is related to the wall deformation (kink creation) which is verified through the linear dependence $\ln({}^{dM}/_{dt}) \times {}^{1}/_{H_i}$, in agreement with the Egami model [11].

Acknowledgments

The authors acknowledg J. L. Tholence of the Centre de Recherche sur les Très Basses Températures/CNRS-Grenoble for the AC-susceptibility measurements and the Divisão de Informática of the Centro Brasileiro de Pesquisas Fisicas, in special Márcio P. de Albuquerque and Lauro Whately for the automatization of the magnetometer.

Figure Captions

Figure 1. (a) and (b) M/H vs. temperature for 60 and 700 Oe, respectively. (c) AC-susceptibility vs. temperature.

Figure 2. Magnetization vs. applied field at several temperatures.

Figure 3. ZFC Hysteresis loop at 2.0 K.

Figure 4. Temperature dependence of the coercive field. (●) experimental points, (——) exponential fit.

Figure 5. Time dependence of the remanent magnetization for some temperatures below T_C.

Figure 6. Linear dependence of the remanent magnetization rate vs. inverse of the internal field for some temperatures below T_C.

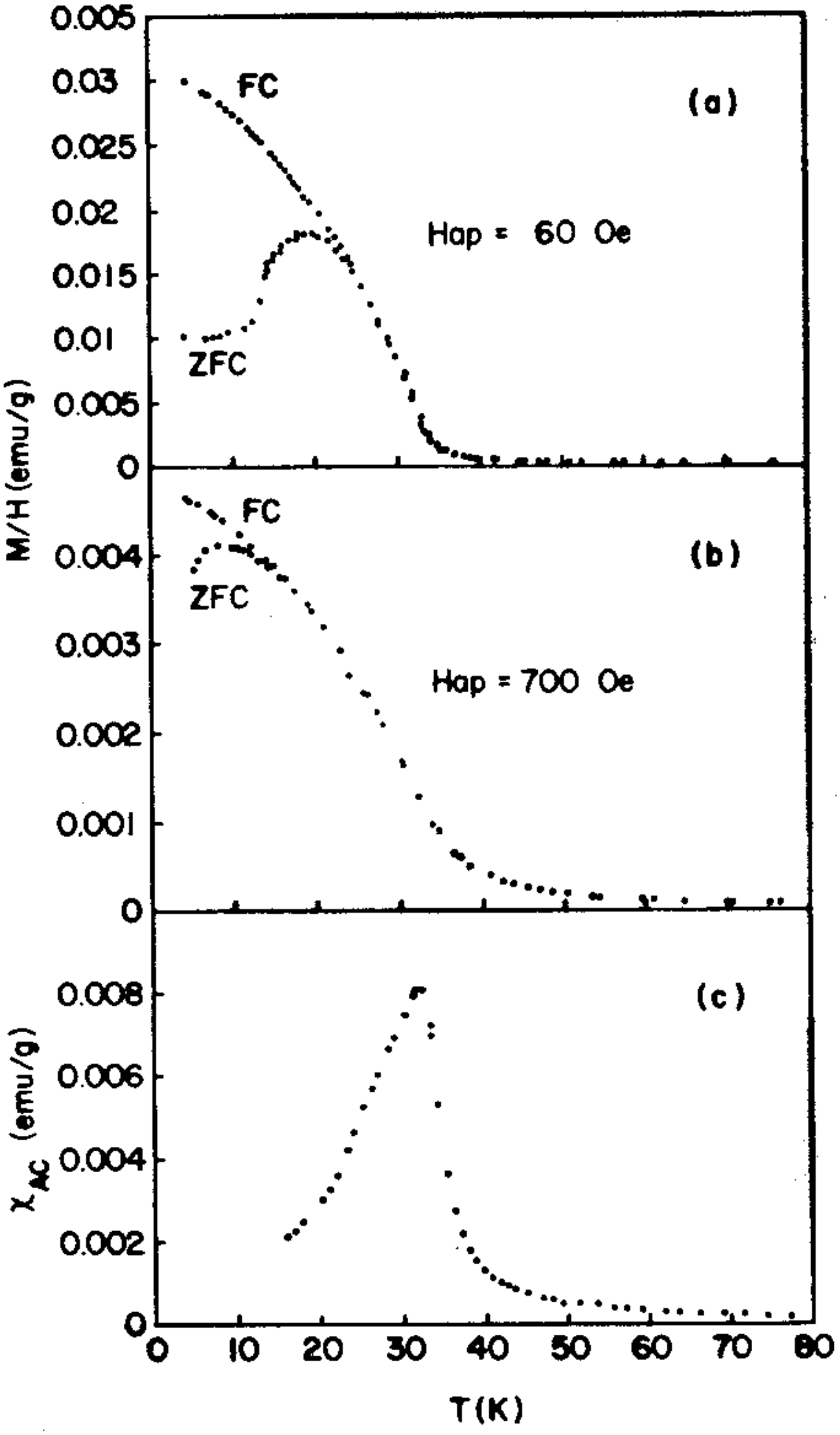
M/H (emu/g)
0.005
0.03
0.025
0.02
0.015
0.01
0.005
0
FC
ZFC
(a)
Hap = 60 Oe
0.004
0.003
0.002
0.001
0
FC
ZFC
(b)
Hap = 700 Oe
χ_{AC} (emu/g)
0.008
0.006
0.004
0.002
0
(c)
0
10
20
30
40
50
60
70
80
T(K)

Fig. 1

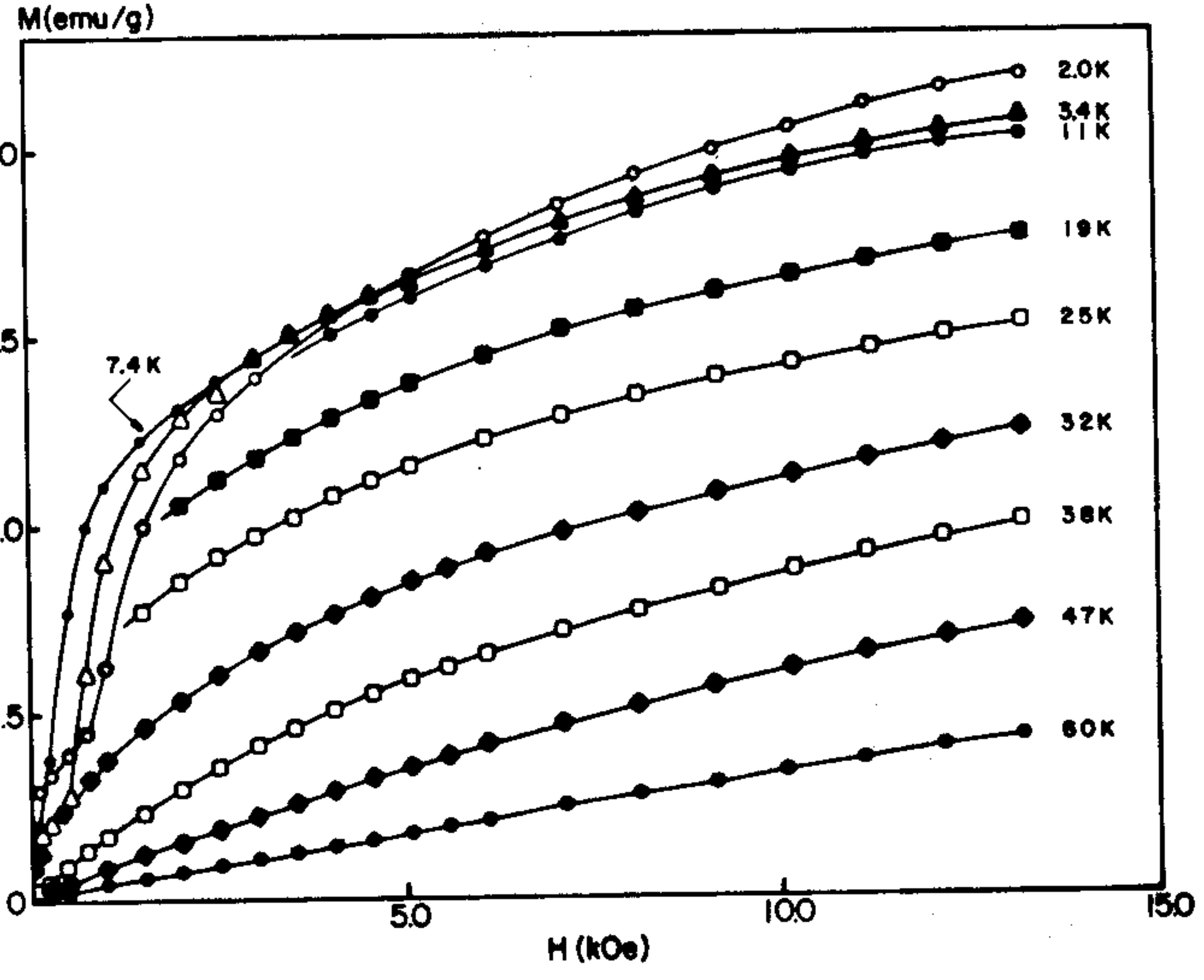
M(emu/g)
6.0
4.5
3.0
1.5
0
5.0
10.0
15.0
H (kOe)
2.0 K
3.4 K
11 K
19 K
25 K
32 K
38 K
47 K
60 K
7.4 K

Fig. 2

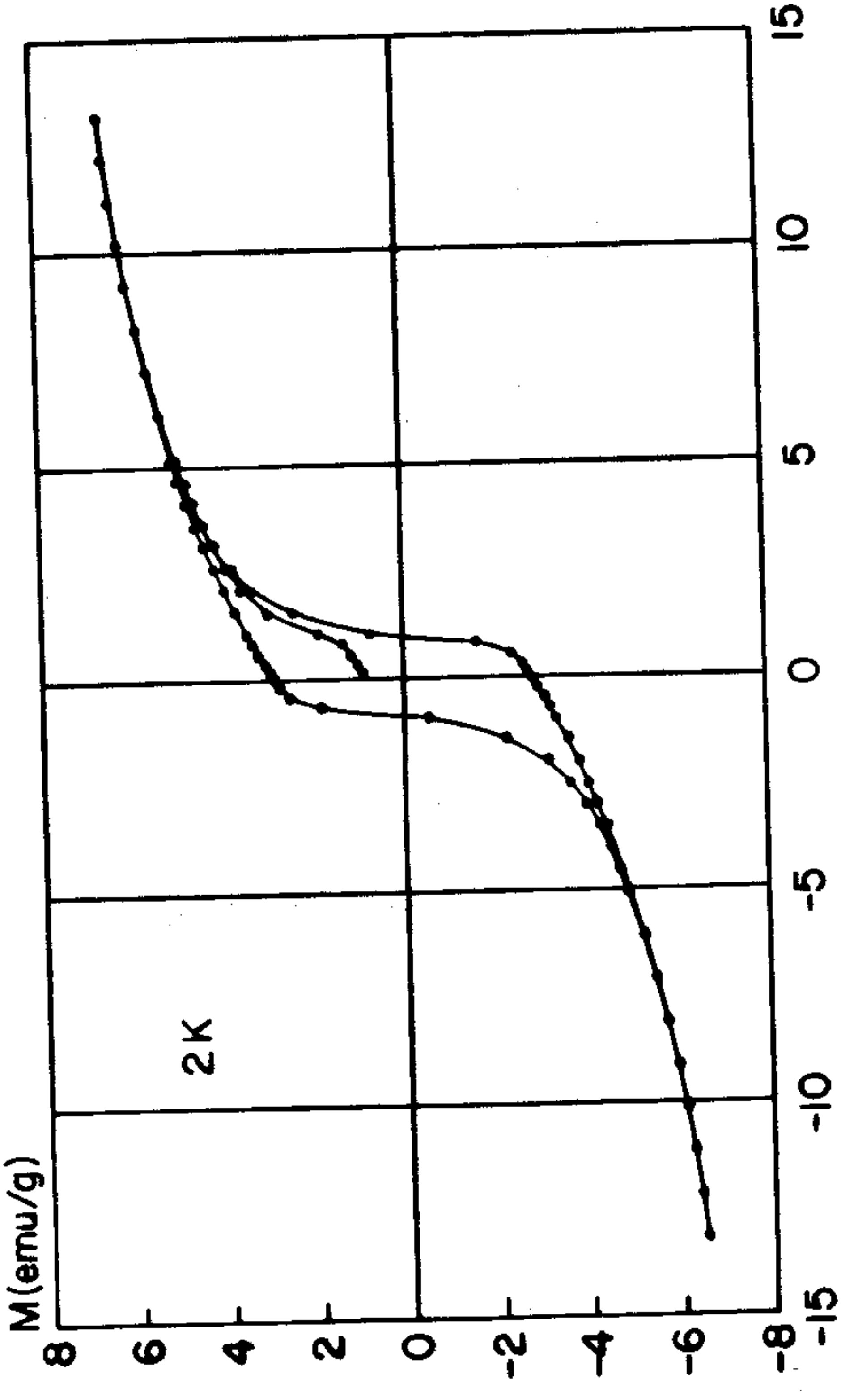

Fig. 3

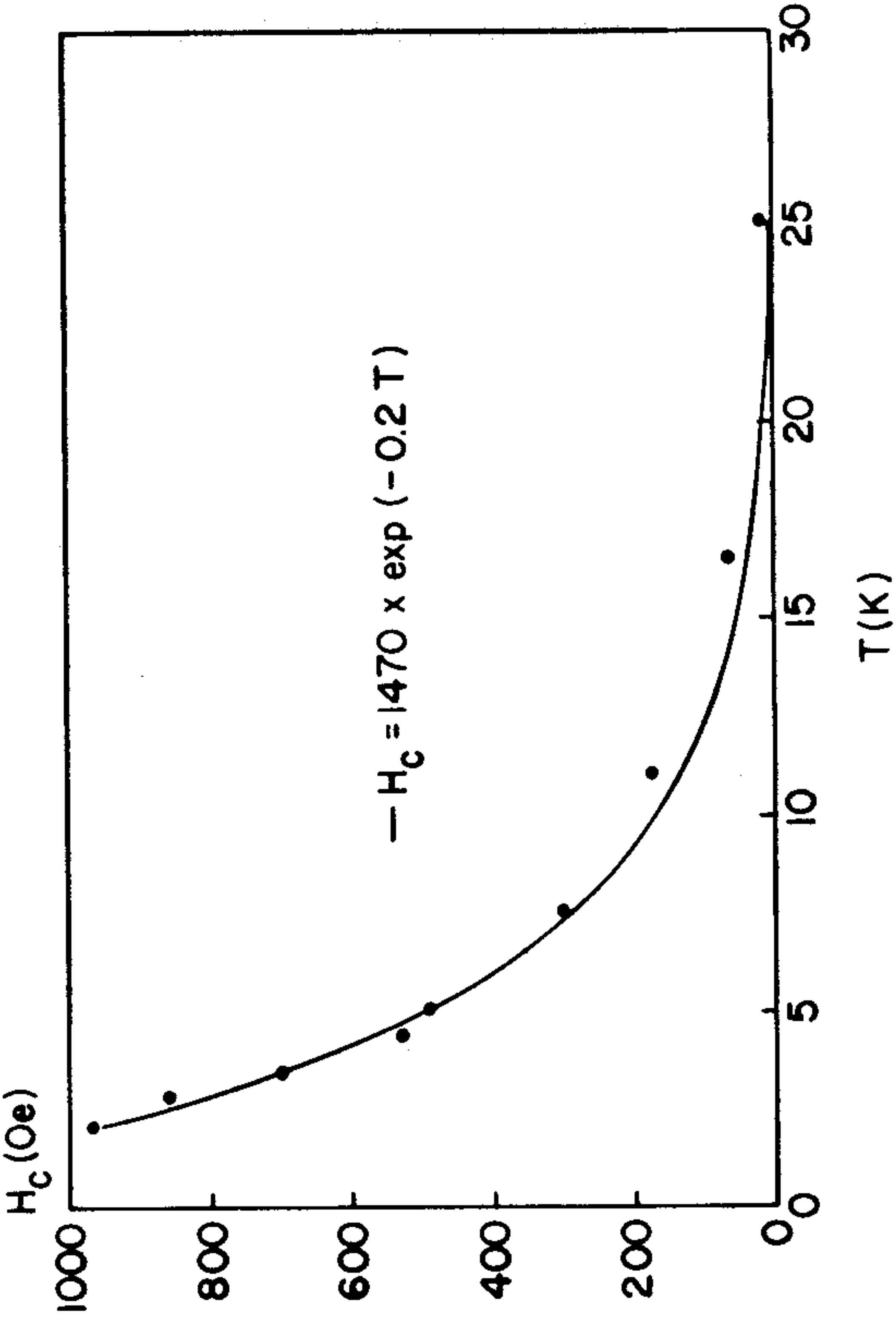

Fig. 4

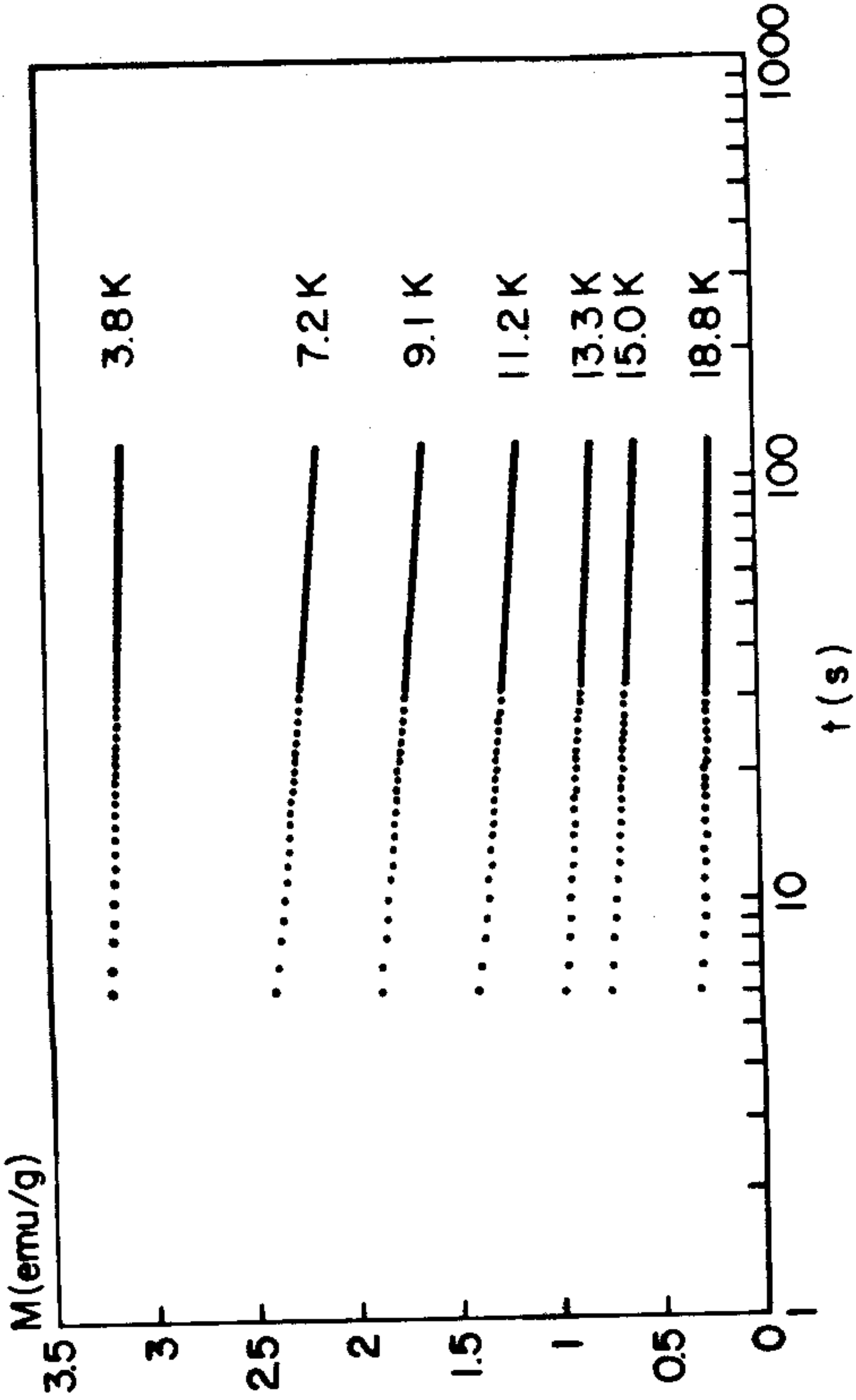
M(emu/g)
3.5
3
2.5
2
1.5
1
0.5
0
t (s)
1
10
100
1000
3.8 K
7.2 K
9.1 K
11.2 K
13.3 K
15.0 K
18.8 K

Fig. 5

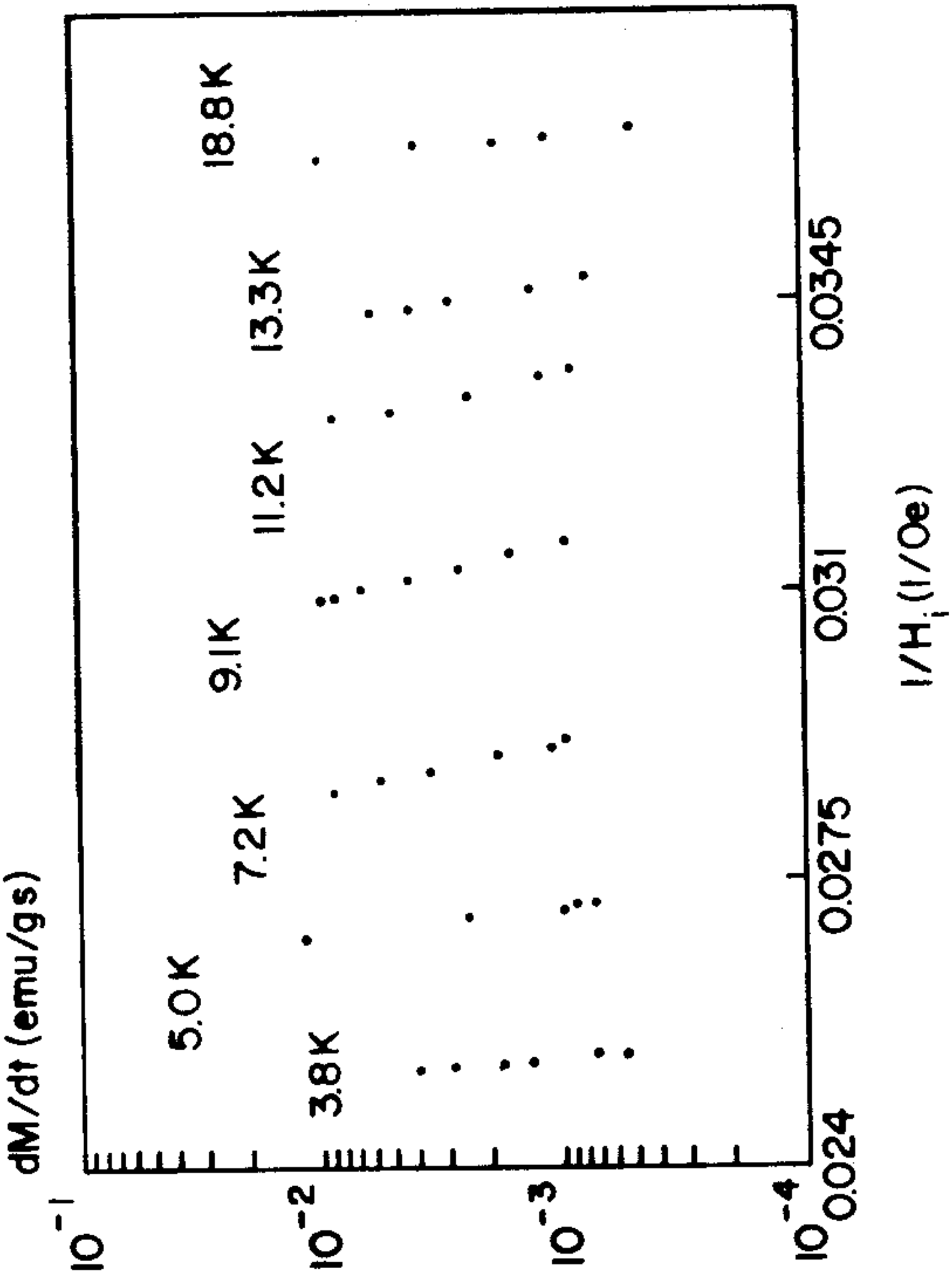
dM/dt (emu/gs)
10^{-1}
10^{-2}
10^{-3}
10^{-4}
0.024
0.0275
0.031
0.0345
$1/H_i$ (1/Oe)
3.8K
5.0K
7.2K
9.1K
11.2K
13.3K
18.8K

Fig. 6

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