

D.C. MAGNETIZATION MEASUREMENTS ON TWO
PtMn SPIN GLASSES

by

Wai Tong Ivan Yeung

A thesis
presented to the University of Manitoba
in fulfillment of the
thesis requirement for the degree of
Master of Science
in
Department of Physics

Winnipeg, Manitoba

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WAI TONG IVAN YEUNG

A thesis submitted to the Faculty of Graduate Studies of
the University of Manitoba in partial fulfillment of the requirements
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MASTER OF SCIENCE

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ABSTRACT

The magnetization of two very dilute spin glasses of Manganese in Platinum (Pt + 2400 ppm Mn and Pt + 1100 ppm Mn) has been measured with a SQUID magnetometer as a function of applied magnetic field and temperature. The magnetization is measured at fixed temperatures in a variable applied magnetic field up to 60 Gauss, for several temperatures above the glass temperatures, T_0 's. In addition, time effects were also studied for the 2400 ppm sample for up to 2 hours at several temperatures both above and below T_0 .

The samples were found to exhibit, as $T \rightarrow T_0$, critical behaviour which is an indication of a phase transition at T_0 . The temperature dependence of the cubic term in a small H expansion of the magnetization showed a 'divergence' as $T \rightarrow T_0$ from above. Moreover, the data also gave good scaling which also supports a thermodynamic phase transition hypothesis. The critical exponent γ was found to be 2.10 ± 0.1 and 2.31 ± 0.1 for the 2400 ppm and 1100 ppm samples, respectively, while β was determined to be 0.6 ± 0.1 and 0.8 ± 0.2 , respectively, by constructing the scaling function. Time effects were found to be present at temperatures above and below T_0 , but they diminished quickly for $T > T_0$. The time dependence of the magnetization was well described by a stretched exponential function.

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1. INTRODUCTION TO SPIN GLASS SYSTEMS

1.1 GENERAL BACKGROUND

Over the last decade, the term spin glass has become a popular topic of controversy in the literature on magnetism. A spin glass system may be defined as a magnetic alloy where the spins on the impurities become locked or frozen into random orientations below a characteristic freezing temperature, T_0 . The long range RKKY interaction is believed to be responsible for the cooperative freezing of the spins. All spin glass systems exhibit certain characteristic features, among which are the sharp peak at T_0 in low-field a.c. susceptibility measurements, a broad maximum in specific heat measurements, and the onset below T_0 of irreversibilities, remanences, and time dependences with the application of an external, static field.

The original materials, in which the characteristic properties of spin glasses were first observed, were dilute alloys of transition metal impurities in noble matrices. Indirect investigation of such materials was started as early as 1932 when Néel attempted to obtain a better understanding of the magnetic properties of pure transition metals. It was not until the early 70's, with Cannella and Mydosh's (1972, 1973) observations of a cusp-like peak in a.c. susceptibility measurements of AuFe alloys, that an explosion of interest in this subject arose.

The cusp in the low-field a.c. susceptibility measurement suggests that some type of phase transition occurs at the freezing temperature. In addition, the splitting of the Mössbauer spectrum and the measurement of the anomalous Hall effect show clear features at T_0 supporting this viewpoint. However, measurements of the electrical resistivity and specific heat, also the remanences, magnetic hysteresis, and time-dependent effects observed in spin glasses are difficult to reconcile with a phase transition approach. The debate over whether a spin glass actually undergoes a phase transition has been raging over the years and the question has not yet been resolved.

1.2 EXPERIMENTAL PROPERTIES

The most obvious way of determining the freezing temperature T_0 is by a.c. susceptibility measurements which give a cusp at T_0 . Cannella and Mydosh(1972, 1973) measured the a.c. susceptibility of several AuFe alloys and some of the results are shown in figures 1.1 and 1.2. The peak becomes rounded with the application of low external fields from 10 to 20 mT. It was also observed that T_0 is roughly linearly related to the concentration C below 1 at.% but it was proportional to $C^{2/3}$ otherwise. The data can be fitted to a Curie-Weiss Law above T_0 . Although only a very small dependence of T_0 on the frequency of the a.c. field was observed for AuFe alloys (Holtzberg et al., 1979), a stronger depen-

dence was observed in PtMn (Lecomte et al., 1983) and PdMn (Mulder et al., 1981). Moreover, magnetic semiconductors and rare earth systems have been recognized to have the strongest frequency dependence of T_0 (Fischer, 1985).

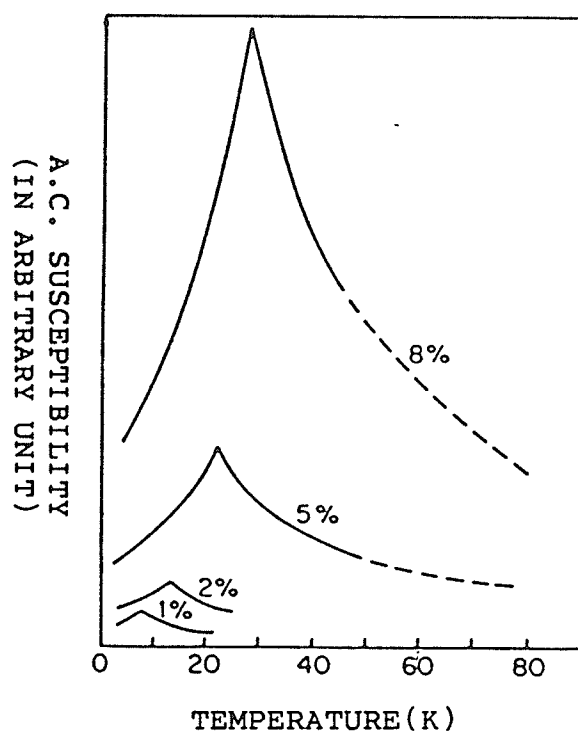


Figure 1.1. Low field a.c. susceptibility of four AuFe alloys with different concentration. (After Cannella and Mydosh, 1979).

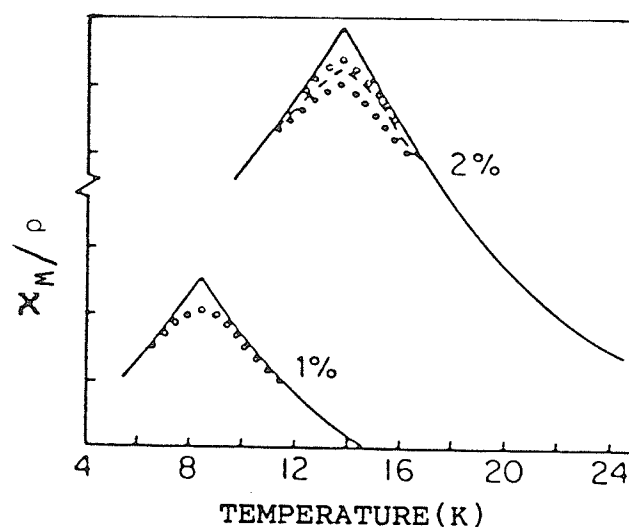


Figure 1.2. A.c. susceptibility of two AuFe alloys in zero field —; o o o, - - -, and • • • represent that in a field with flux density of 10mT, 20mT and 30mT respectively. (After Ford, 1982).

The presence of a phase transition may also be inferred from transport coefficients. Neither the direct measurement of electrical resistivity nor its derivative give anomalous features at T_0 . There exists a concentration dependent maximum which occurs at a temperature $T_{\max} \gg T_0$. Although a good correlation between the temperature corresponding to the maximum of $d(\Delta \rho)/dT$ ($\Delta \rho = \rho_{\text{alloy}} - \rho_{\text{pure metal}}$) and T_0 was found in AuFe (Mydosh et al. 1974), this relation breaks down for AuMn and AuCr systems (Ford and Mydosh, 1976). Fig-

ure 1.3 shows the result of resistivity measurement on AuFe by Mydosh et al. (1974). They found that the impurity resistivity obeyed the relation,

$$\Delta\rho(T,C) = C \Delta\rho_0 + A(C) T^{2/3}$$

at low temperatures, where C is the concentration, T , the temperature and $\Delta\rho_0$, the residual impurity resistivity. The coefficient $A(C)$ varies slowly with concentration. As the temperature increases to around T_0 , $\Delta\rho$ is found to be predominantly linear in T . At extremely low temperatures, less than 0.3K, Laborde and Radhakrishna(1973) have observed a T^2 dependence.

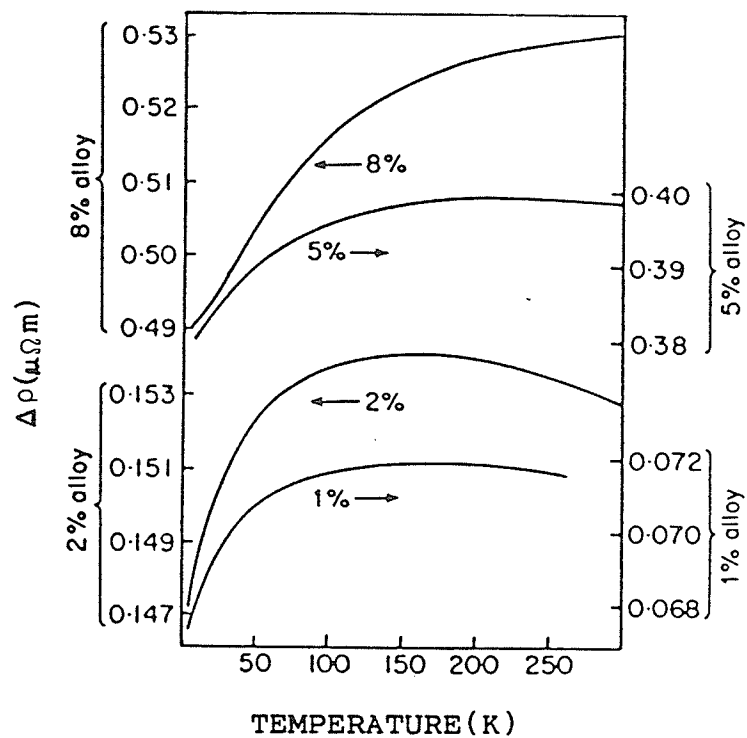


Figure 1.3. Impurity resistivity $\Delta\rho$ as a function of temperature for four AuFe alloys. (After Mydosh et al., 1974).

Again, if a phase transition does take place at T_0 , an anomaly at T_0 would be expected in a specific heat (molar heat capacity), C_m , measurement. Wenger and Keensom(1975, 1976) have examined the specific heat of AuFe and CuMn alloys and their results are displayed in figure 1.4. They found no anomaly at T_0 , as defined by a.c. susceptibility measurements on other portions of the same sample. At very low temperatures, C_m has a large, concentration independent contribution which is linear in T . A broad peak occurs at a temperature, T_{max} , above T_0 but no definite correlation between these two temperatures has been found; C_m then behaves as $1/T$ at higher temperatures. Nevertheless, some recent investigations have suggested that some sort of anomalous features exist at T_0 . Martin(1979, 1980a,b) has observed a knee at T_0 in C_m/T vs T plots for CuMn alloys. Fogle et al.(1981) have also examined CuMn alloys and found an anomaly at T_0 in $\Delta(C_m/T)/\Delta T$ vs T plots as shown in figure 1.5. In addition, the latter group have also discovered an anomaly at T_0 in the quadratic term, B , when C_m/T is expanded as a function of H , ie,

$$C_m/T = A + BH^2$$

Figure 1.6 shows the quadratic term, B , plotted against the temperature.

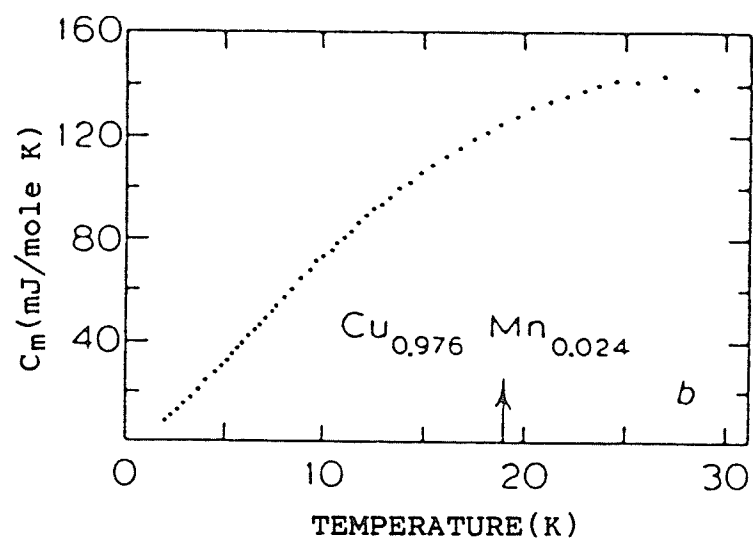
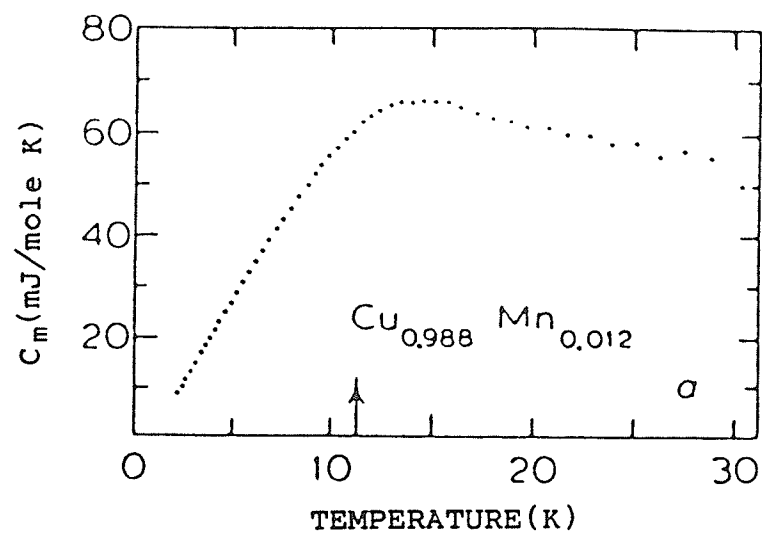


Figure 1.4. Temperature dependence magnetic specific heat for two CuMn spin glass alloys. The arrows indicate the glass temperatures defined by a.c. susceptibility. (After Wenger and Keenom, 1976).

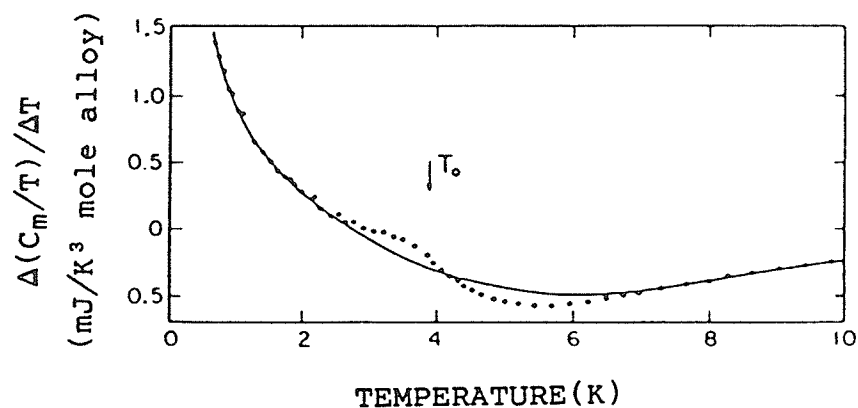


Figure 1.5. Derivative of C_m/T with respect to T in Cu + 0.279at.% Mn. (After Fogle et al., 1981).

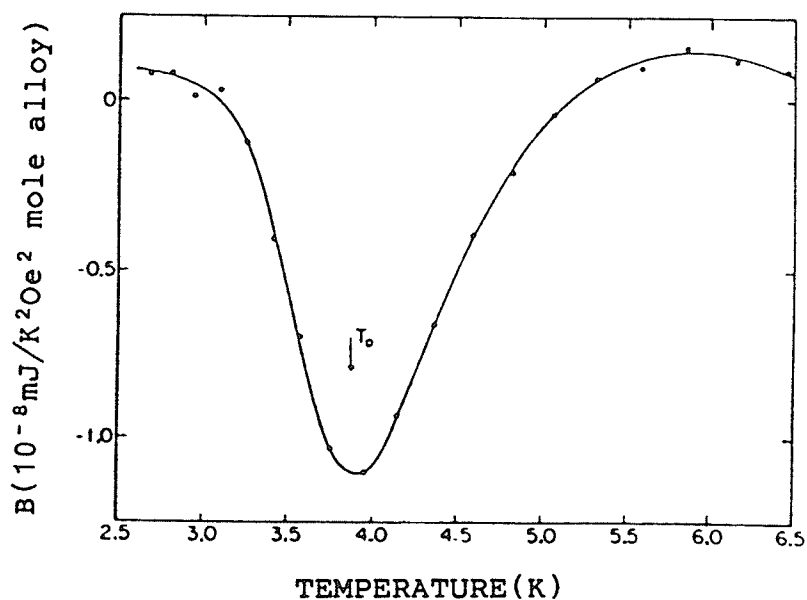


Figure 1.6. Temperature dependence of B in $C_m/T = A + BH^2$ of Cu + 0.279 at.% Mn. (After Fogle et al., 1981)

In studying the magnetic remanence of spin glasses, one can consider two different experimental situations, namely, the isothermal remanent magnetization (I.R.M.) and the thermoremanent magnetization (T.R.M.). In the I.R.M. case, a specimen is cooled in zero field, followed by an application of a field, H , and then the field is turned off slowly. In the T.R.M. situation the specimen is measured in zero field after cooling the sample from above to below T_0 in a field H . Figure 1.7 shows these two remanent magnetizations for AuFe, by Tholence and Tournier (1974). It is noted that both I.R.M. and T.R.M. saturate at the same value but T.R.M. reaches this value at a lower field.

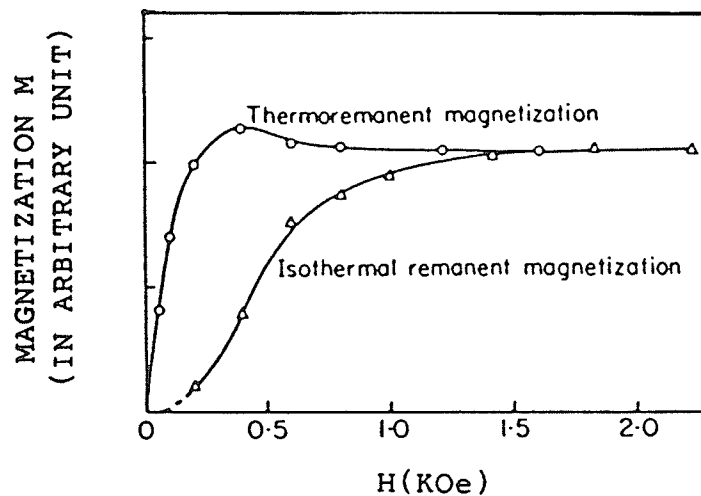


Figure 1.7. Field dependence of the thermoremanent magnetization (T.R.M.) and the isothermal remanent magnetization (I.R.M.) for a AuFe alloy at $T \ll T_0$. (After Tholence and Tournier, 1974).

The remanence and irreversible behaviour of spin glasses can also be seen in their hysteresis loop. Figure 1.8 is a diagram of typical hysteresis loops. Part (a) represents a zero field-cooled situation where the inner line is the reversible part. When the applied field H exceeds the critical field, an I.R.M. starts to develop which corresponds to the outer loop of part (a). Part (b) represents a field-cooled spin glass which has a distinct shifted hysteresis loop. The unusual character of a field-cooled spin glass can be compared with that of a typical ferromagnetic loop shown in (c).

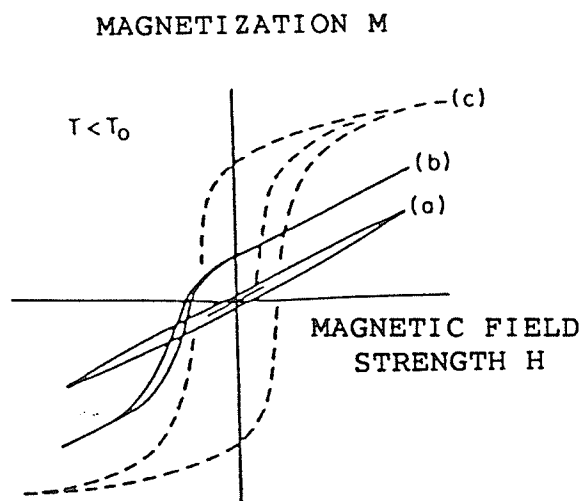


Figure 1.8. Hysteresis loops of magnetization M against magnetic field strength H . (a) spin glass cooled in zero field and (b) in a magnetic field. (c) is ferromagnet. (After Mydosh, 1975).

The time dependence of the remanence, both the I.R.M. and T.R.M., takes on logarithmic and/or exponential form. Figure 1.9 shows typical data due to Guy (1978) for the time decay of the T.R.M. for a 2 at.% AuFe alloy. For times greater than 10s, both I.R.M. and T.R.M. decay can be described by the form (Ford, 1982)

$$M = M_0 - S \ln(t)$$

where M_0 is a constant and S is also a constant which can depend on the field, temperature and material. Other authors, such as Chamberlin et al.(1984) have shown that a stretched exponential form,

$$M = M_0 \exp[-(t/t_p)^{1-n}]$$

is able to characterize the T.R.M. for CuMn and AgMn systems, where t_p depends on the temperature and the time in which the sample was left in a constant field at a constant temperature before the field was turned off (the 'wait time'). However, recent work by Nordblad et al.(1986) has demonstrated that the total relaxation of the magnetization for a CuMn alloy must be described by a pure logarithmic decay superimposed on a stretched exponential form:

$$M = S' H \ln(t) + M_1 + M_0 \exp[-(t/t_p)^{1-\eta}]$$

where S' is the relaxation rate at dynamic equilibrium, H the field, M_1 the intercept at $\log_{10}(t)=0$ for the logarithmic decay.

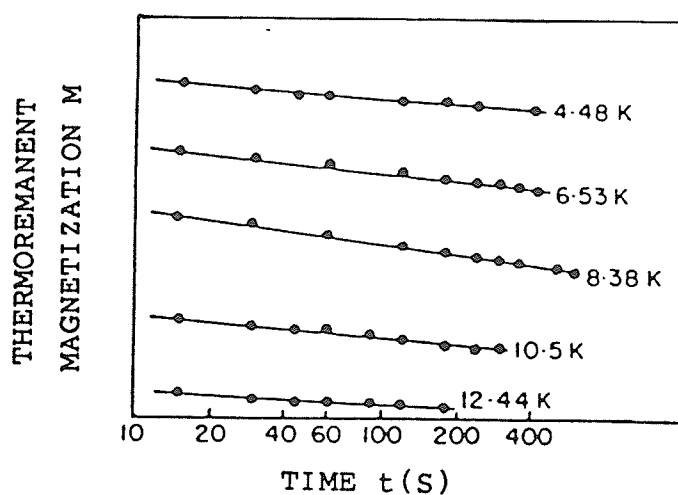


Figure 1.9. Thermoremanent magnetization decay as a function of time for a 2 at.% AuFe alloy which had been field cooled in 12.8 mT. (After Guy, 1978)

1.3 THE FORMATION OF A SPIN GLASS

At extremely low concentrations, the impurities are separated so far apart that they basically are non-interacting. The experimental properties of such systems can be explained in terms of the Kondo framework: below the Kondo temperature, which depends on concentration, the impurity becomes non-magnetic due to its interaction with the conduction electrons.

As the concentration increases, the local moments begin to interact via the RKKY mechanism which is an indirect interaction between impurity spins through the conduction electrons. A local spin polarizes the surrounding conduction electrons which subsequently polarize another impurity spin. Hence, an indirect coupling between two magnetic spins occurs which, as shown in figure 1.10, can be either ferromagnetic or antiferromagnetic depending on their separation. Because of the random positions of the impurity spins within the crystal, the magnetic interactions are also arbitrarily distributed. Therefore, the impurity spins become randomly 'locked' in place below T_0 and so the spontaneous magnetization vanishes. The RKKY interaction will be discussed in more detail in the next section.

There are two criteria in order to obtain 'good' spin glass systems (Ford and Mydosh, 1976). Firstly, the impurity should have a high solubility in the host, at least up to 10 at.%, to ensure that the impurity is evenly distributed in the host. Secondly, the alloy should have a low Kondo temperature, less than 1K, so that one is dealing with good moment systems. Tables 1.1 and 1.2 are extracted from Mydosh and Nieuwenhuys(1980) showing the 'grades' of spin glasses from some common combinations.

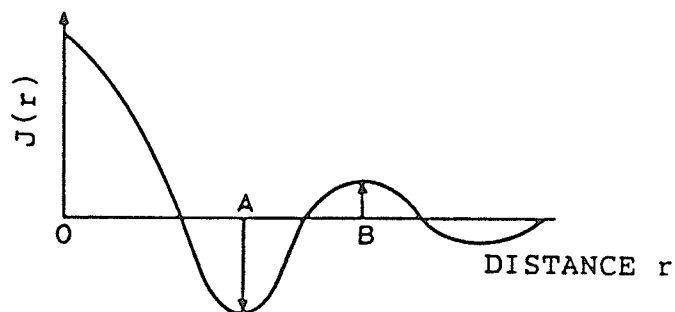


Figure 1.10. $J(r)$ is the RKKY exchange interaction between two spins. The negative sign of $J(r=A)$ implies an antiferromagnetic interaction between spins at o and at A . $J(r=B)$ is positive so that spins at o and at B have a ferromagnetic interaction. (After Ford, 1982).

Host	Impurity:	V	Cr	Mn	Fe	Co	Ni
Cu		XS	XS	GOOD	XS+T	XS	XT
Ag		XS	XS	GOOD	XS	XS	XS
Au		XT	GOOD	GOOD	GOOD	XS+T	XT

Table 1.1. Spin glass combinations of noble metal - transition metal. 'Good' represents the most favourable combinations. XS and XT indicates the spin glass behaviour is limited by the lack of solubility and too high a Kondo temperature respectively. (After Mydosh and Nieuwenhuys, 1980)

Host	Impurity	Cr	Mn	Fe	Co	
Mo		XT	SG	SG	XS+T	} simple
Rh		XT	SG	XT	XT	
Pd		XT	GM+SG	GM	GM	} exchange enhanced
Pt		XT	SG	GM	SG	

Table 1.2. Spin glass combination of transition metal - transition metal. SG and GM represent favourable spin glass combination and giant moment combination respectively. XS and XT denotes those combinations limited by solubility problem and high Kondo temperature. (After Mydosh and Nieuwenhuys, 1980)

1.4 THE RKKY INTERACTION

The exchange interaction, $-J\vec{S}\cdot\vec{\sigma}$, between an impurity spin $\vec{S}$ and the host induces a polarization of the conduction electrons spins $\vec{\sigma}$. The conduction spins in turn interact with another impurity electron and thus an indirect coupling between two impurities is developed via the conduction electron gas. This interaction is called the RKKY interaction due to the major contribution by Ruderman, Kittel, Kasuya and Yosida in developing its formulation. The following is an outline of the derivation of the RKKY interaction based on Kittel(1968) and Ho(1982).

Consider the spin polarization $\vec{\sigma}(\vec{r})$ of a free electron gas when a magnetic moment is placed in it. The basic assumptions of this analysis are (Ho, 1982):

1. free electron model at absolute zero
2. linear response approximation
3. static magnetic field
4. energy corrected to 2nd order only, within non-degenerate perturbation theory.

We have

$$\sigma(\vec{r}) = \frac{M(\vec{r})}{g \mu_B} \quad (1)$$

where the magnetization of the electron gas per unit volume $M(\vec{r})$ is related to the magnetic field $H(\vec{r})$ and the susceptibility $\chi_0(\vec{r})$ by (Kittel, 1968)

$$M(\vec{r}) = \int d^3r' \chi_0(\vec{r} - \vec{r}') H(\vec{r}') \quad (2)$$

By the linear response approximation, the three quantities can be written in terms of Fourier series:

$$M(\vec{r}) = \sum_{\vec{q}} M_{\vec{q}} \exp(i\vec{q} \cdot \vec{r}) \quad (3a)$$

$$H(\vec{r}) = \sum_{\vec{q}} h_{\vec{q}} \exp(i\vec{q} \cdot \vec{r}) \quad (3b)$$

$$\chi_0(\vec{r}) = \sum_{\vec{q}} \chi_{\vec{q}} \exp(i\vec{q} \cdot \vec{r}) \quad (3c)$$

where $\vec{q}$ is the reciprocal lattice vector

Substitute equations (3) into (2), we obtain,

$$M(\vec{r}) = \sum_{\vec{q}} \chi_{\vec{q}} h_{\vec{q}} \exp(i\vec{q} \cdot \vec{r})$$

and $M_{\vec{q}} = \chi_{\vec{q}} h_{\vec{q}}$

Therefore, the problem can be reduced to finding the static spin susceptibility χ_q of a free electron gas upon the application of the Fourier component $\vec{h}_q \cos(\vec{q} \cdot \vec{r})$.

The Hamiltonian at absolute zero is

$$H = \sum_i \frac{P_i^2}{2m} + \mu_B \sum_i \vec{\sigma}_i \cdot \vec{h}_q \cos(\vec{q} \cdot \vec{r}_i)$$

where the first term is the kinetic energy while the second,

$$H_1 = \mu_B \sum_i \vec{\sigma}_i \cdot \vec{h}_q \cos(\vec{q} \cdot \vec{r}_i)$$

is the perturbation. If $\vec{h}_q$ is taken to lie along the x-direction, then (Kittel, 1968),

$$H_1 = \frac{1}{4} \mu_B h_q \sum_i (\sigma_i^+ + \sigma_i^-) (e^{i\vec{q} \cdot \vec{r}_i} + e^{-i\vec{q} \cdot \vec{r}_i})$$

The first order correction of the energy, $\langle \vec{k} | H_1 | \vec{k} \rangle$, is equal to zero because of the orthogonality of the plane wave states (Kittel, 1968); the 2nd order correction of the energy for state k can be shown to be (Kittel, 1968)

$$\epsilon_k^{(2)} = \frac{1}{4} (\mu_B h_q)^2 \left[\frac{1 - f_{k+q}}{\epsilon_k - \epsilon_{k+q}} + \frac{1 - f_{k-q}}{\epsilon_k - \epsilon_{k-q}} \right]$$

where ϵ_k is the free electron energy for state $\vec{k}$ and f_k is the Fermi-Dirac distribution.

The total 2nd order energy correction is a summation over all occupied k states and it becomes (Kittel, 1968)

$$E^{(2)} = - \frac{1}{4} (\mu_B \hbar q)^2 F(q)$$

where $F(q) = \sum_k \frac{f_k - f_{k+q}}{\epsilon_{k+q} - \epsilon_k}$

The susceptibility for wave vector q is

$$\chi_q = - \frac{\partial E^{(2)}}{\partial (\text{magnetic field})^2}$$

$$\chi_q = \mu_B^2 F(q)$$

Hence, the susceptibility in real space is, from (3) (Kittel, 1968),

$$\begin{aligned} \chi_0(\vec{r}) &= \sum_q \mu_B^2 F(q) \exp(i\vec{q} \cdot \vec{r}) \\ &= \frac{m \mu_B^2}{(2\pi)^3 \hbar^2} \left[\frac{\sin 2k_F r - 2k_F r \cos k_F r}{r^4} \right] \end{aligned} \quad (4)$$

where k_F is the Fermi vector.

Assume the interaction between the impurity spin $\vec{S}_\alpha$ and the conduction electron spin $\vec{\sigma}_i$ is a delta function of the form:

$$- J \sum_i \vec{S}_\alpha \cdot \vec{\sigma}_i \delta(\vec{r}_i)$$

The magnetic field at the conduction electron site can then be written as (Ho, 1982)

$$H_{\text{eff}}(\vec{r}) = \frac{J}{g \mu_B} \vec{S}_\alpha \delta(\vec{r}_i)$$

with a Fourier component of

$$H_{\text{eff}}(q) = \frac{J}{g \mu_B} \vec{S}_\alpha$$

Using equations (1) to (3), the spin density can be expressed in the form (Ho, 1982):

$$\sigma(\vec{r}) = \frac{J}{g^2 \mu_B^2} \sum_q \chi_q e^{i\vec{q} \cdot \vec{r}} \vec{S}_\alpha$$

From (4)

$$\sigma(\vec{r}) = \frac{J m}{(2\pi)^3 g^2 \hbar^2} \left[\frac{\sin 2k_F r - 2k_F r \cos 2k_F r}{r^4} \right] \vec{S}_\alpha$$

In figure 1.11, the spin polarization is plotted as a function of distance from the impurity site, r . The function is largest for small r ; and at large distances from the $\vec{S}_\alpha$ site, when $k_F r \gg 0$, then (Ho, 1982)

$$\sigma(\vec{r}) \sim - \frac{2k_F r \cos 2k_F r}{r^4}$$

That is, the spin polarization oscillates with a period of πk_F^{-1} and its amplitude decreases as r^{-3} .

The above discussion assumes that the impurity is placed in a non-interacting free electron gas. However, for transition metal hosts, such as Pd and Pt, the conduction d-electrons are confined to a narrow band. The Coulomb interactions among those conduction electrons must be taken into account and the free electron susceptibility must be substituted with that for an interacting electron gas in order to obtain the appropriate spin polarization. If the Coulomb interaction is a δ -function with strength V on lat-

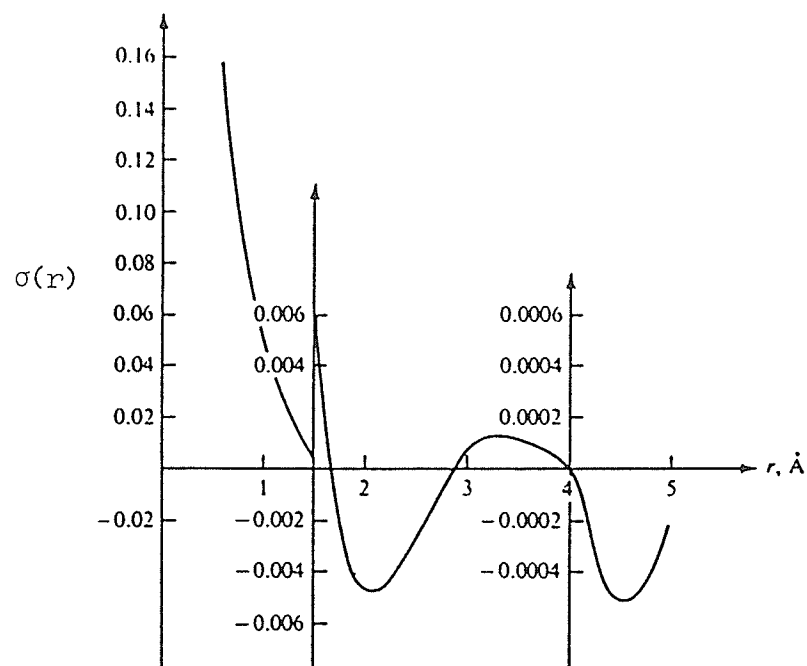


Figure 1.11. Conduction electron spin density $\sigma(r)$ as a function of distance from an impurity moment placed in a metallic host. (After White, 1970)

tice sites, the susceptibility would be modified as (Wolff, 1960):

$$\chi'(q) = \frac{3/4 \mu_B^2 E_F F(q)}{1 - 2/3 V E_F F(q)}$$

Therefore, $\chi'(q) > \chi_0(q)$ for all $\vec{q}$. Hence, this enhanced susceptibility provides a longer range for $\sigma(\vec{r})$ such that it pushes out the first zero of $\sigma(\vec{r})$. Both the non-interacting and enhanced cases are plotted in figure 1.12.

The conduction electron spin polarization provides the mechanism for the interaction between two impurity spins. For an impurity $\vec{S}_i$ located at $\vec{R}_i$, it induces a spin polarization $\sigma(\vec{r} - \vec{R}_i)$ on the surrounding conduction electrons. If another impurity spin $\vec{S}_j$ at $\vec{R}_j$ interacts with these polarized conduction electrons, the indirect interaction between these two impurity spins would be described by the Hamiltonian (Ho, 1982):

$$H_{\text{RKKY}} = \frac{-J^2 m}{(2\pi)^3 g^2 \hbar^2} \left[\frac{\sin 2k_F r - 2k_F r \cos 2k_F r}{r^4} \right] \vec{S}_\alpha \cdot \vec{S}_\beta$$

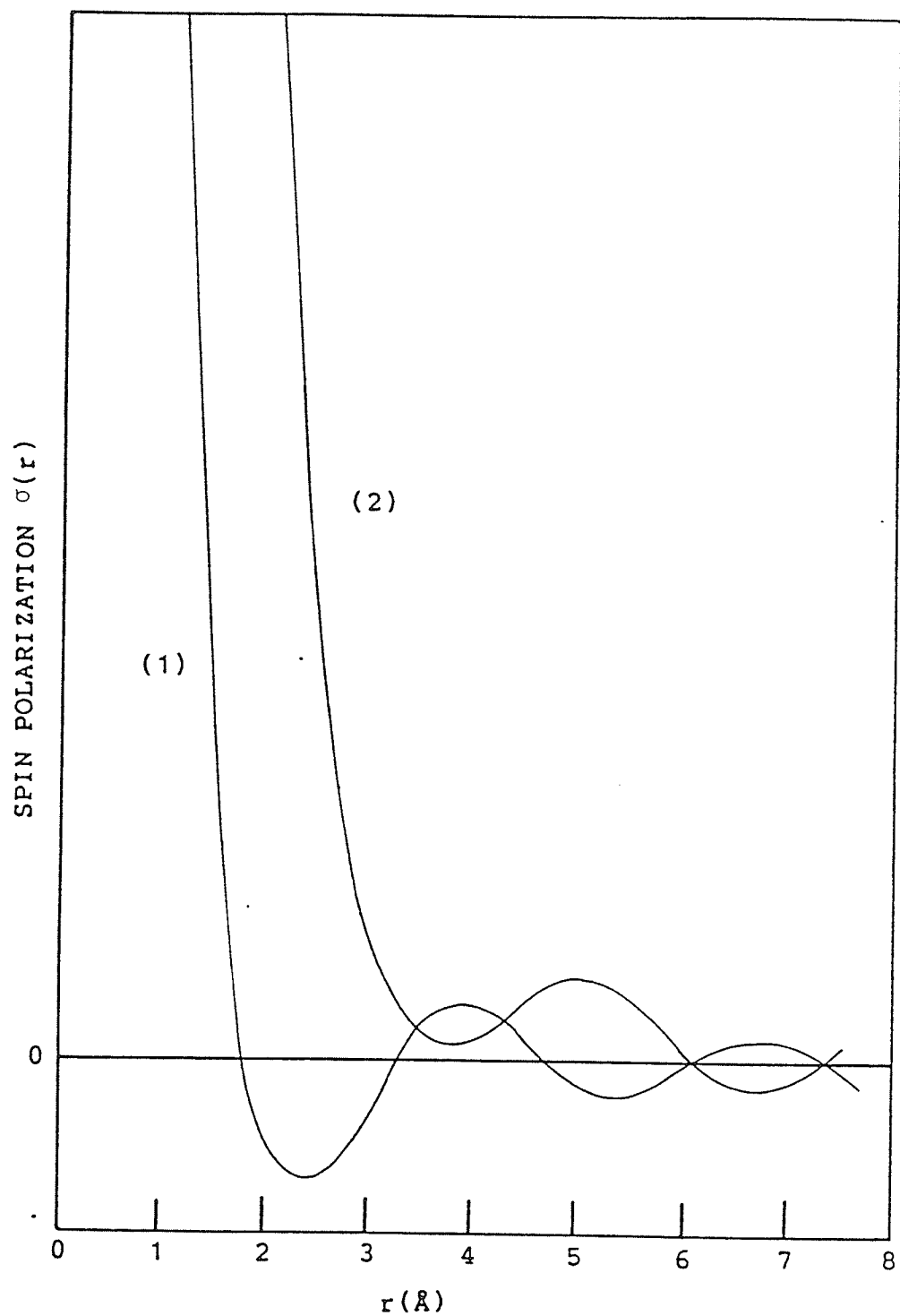


Figure 1.12. Comparison of the electron spin density as a function of distance from the impurity moment for (1) a non-interacting electron gas, and (2) an interacting electron gas. (After Roshko, 1979)

1.5 EDWARDS-ANDERSON MODEL

In describing the magnetic transition at T_0 , Edwards and Anderson(1975) postulated that although there is no mean ferro- or antiferromagnetic configuration in a spin glass system, there exists a ground state where each spin 'freezes' in a preferred direction which is randomly oriented and different at every site. The existence of this ground state, they showed, is sufficient to give rise to a phase transition and a cusp in the susceptibility. It is also assumed that if J_{ij} is the exchange interaction between spins i and j while ϵ_{ij} is the probability of finding a pair of spins at i and j , then

$$\sum J_{ij} \epsilon_{ij} = 0$$

on any scale of the alloy. Furthermore, if one observes a spin $\vec{S}_i(0)$ at $t=0$, the probability q that this spin $\vec{S}_i(t)$ will point at the same direction at any later time, even as $t \rightarrow \infty$, is nonvanishing. A new order parameter for characterizing the phase transition is then defined as (Moorjani and Coey, 1984):

$$q = \lim_{t \rightarrow \infty} \langle \vec{S}_i(0) \cdot \vec{S}_i(t) \rangle$$

where the inner set of angular brackets represents the thermal average while the outer set represents the average over all spins. Hence at $T=0$, one expects $q=1$, at $T \geq T_0$, $q=0$.

In setting up the spin glass problem, Edwards and Anderson adopted a classical approach on the following Hamiltonian and treated the spin vectors as magnetic dipoles which were able to orient in any direction:

$$H = -\frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j - \sum_i \vec{h}_i \cdot \vec{S}_i$$

where the second term is the energy contribution resulting from the application of an external magnetic field $\frac{h_i}{g \mu_B}$ to the spins. The first term is the exchange interaction J_{ij} between two spins i and j with J_{ij} having a probability given by the Gaussian distribution,

$$P(J_{ij}) = \frac{1}{\sqrt{2\pi} \bar{J}} \exp\left(-\frac{J_{ij}^2}{2\bar{J}^2}\right)$$

where $\bar{J}$ is the standard deviation.

Edwards and Anderson then evaluated the average free energy over the distribution $P(J_{ij})$ by the replica method in which $\ln(Z)$ is replaced by the approximation

$$\ln Z = \lim_{n \rightarrow 0} \frac{1}{n} (Z^n - 1)$$

where Z is the partition function. The replica method can be physically interpreted as an averaging process over n identical systems, $Y_1, Y_2, \dots, Y_n$. The average is over all the couplings of any two replicas, which represent the system at different times. Therefore, the replica-replica correlation-function (or coupling) behaves in the same manner as the long-time auto-correlation function, ie. the definition of the order parameter q (Moorjani and Coey, 1984).

They arrived at the following expression for the susceptibility (Moorjani and Coey, 1984),

$$\chi = \chi_p (1 - q)$$

where $\chi_p = N(g \mu_B s)^2 / 3k T = C/T$, is the usual paramagnetic susceptibility. χ obeys a Curie law above T_0 because $q=0$ for $T > T_0$. A kink occurs when χ starts to deviate from the Curie law at T_0 , as $q \neq 0$ for $T \leq T_0$. In non-zero magnetic fields, the cusp becomes rounded with a field twenty times stronger than that needed in experiments (Moorjani and Coey, 1984). The Edwards-Anderson model also gives rise to a cusp at T_0 in the specific heat, which is in serious disagreement with experimental data.

At low temperatures, one obtains (Moorjani and Coey, 1984)

$$q = 1 - \left(\frac{2}{3\pi}\right)^{\frac{1}{2}} \frac{T}{T_0}$$

and

$$\chi = \frac{A}{T} \left(\frac{2}{3\pi}\right)^{\frac{1}{2}} \frac{T}{T_0} \quad A - \text{a constant}$$

so $q \rightarrow 1$ and $\chi \rightarrow \text{a constant}$ as $T \rightarrow 0$.

1.6 SHERRINGTON AND KIRKPATRICK MODEL

Sherrington and Kirkpatrick (1975) presented a spin glass model which was based on the fact that one can exactly solve a ferromagnet using molecular field theory with an exchange interaction of infinite range. They used a similar approach for disordered systems to solve the spin glass problem. The Sherrington-Kirkpatrick model consists of an ensemble of N spins coupled by infinite-ranged Ising exchange interactions. The Hamiltonian is

$$H = -\frac{1}{2} \sum_{i \neq j} J_{ij} S_i S_j, \quad S_i = \pm 1$$

where J_{ij} has a Gaussian distribution,

$$P(J_{ij}) = \frac{1}{\sqrt{2\pi} \bar{J}} \exp \left[-\frac{(J_{ij} - \bar{J})^2}{2 \bar{J}^2} \right]$$

Because the interaction is of infinite range, extending to all spins in the lattice and not just nearest neighbours, one has to scale $\tilde{J}$ and $\tilde{J}_0$ as

$$J_0 = \frac{\tilde{J}}{N} \qquad \bar{J} = \frac{\tilde{J}}{N}$$

The order parameter and magnetization are defined as (Fischer, 1983)

$$q = \frac{1}{N} \sum_i \langle S_i \rangle_T^2 = \langle \langle S_i \rangle_T^2 \rangle_J$$

$$m = \frac{1}{N} \sum_i \langle S_i \rangle_T = \langle \langle S_i \rangle_T \rangle_J$$

where $\langle \rangle_T$ is the thermal average while $\langle \rangle_J$ the average over all sites, so that q is the average of the square of the local magnetization. They then solved the problem by evaluating the free energy with the replica trick and finally arrived at a pair of coupled equations for q and m (Moorjani and Coey, 1984):

$$q = \frac{1}{\sqrt{2\pi}} \int dZ \exp\left(-\frac{Z^2}{2}\right) \tanh^2\left[\frac{\tilde{J} q^{\frac{1}{2}}}{k T} Z + \frac{\tilde{J}_0 m}{k T}\right]$$

$$m = \frac{1}{\sqrt{2\pi}} \int dZ \exp\left(-\frac{Z^2}{2}\right) \tanh\left[\frac{\tilde{J} q^{\frac{1}{2}}}{k T} Z + \frac{\tilde{J}_0 m}{k T}\right]$$

As shown in figure 1.13, the magnetic phase of the system depends on the ratio $\tilde{J}_0/\tilde{J}$. If $\tilde{J}_0 > \tilde{J}$, there are not enough antiferromagnetic exchange interactions to produce a spin glass. For $\tilde{J}_0 < \tilde{J}$, the antiferromagnetic interactions dominate the ferromagnetic ones and so the spin glass state is preferred.

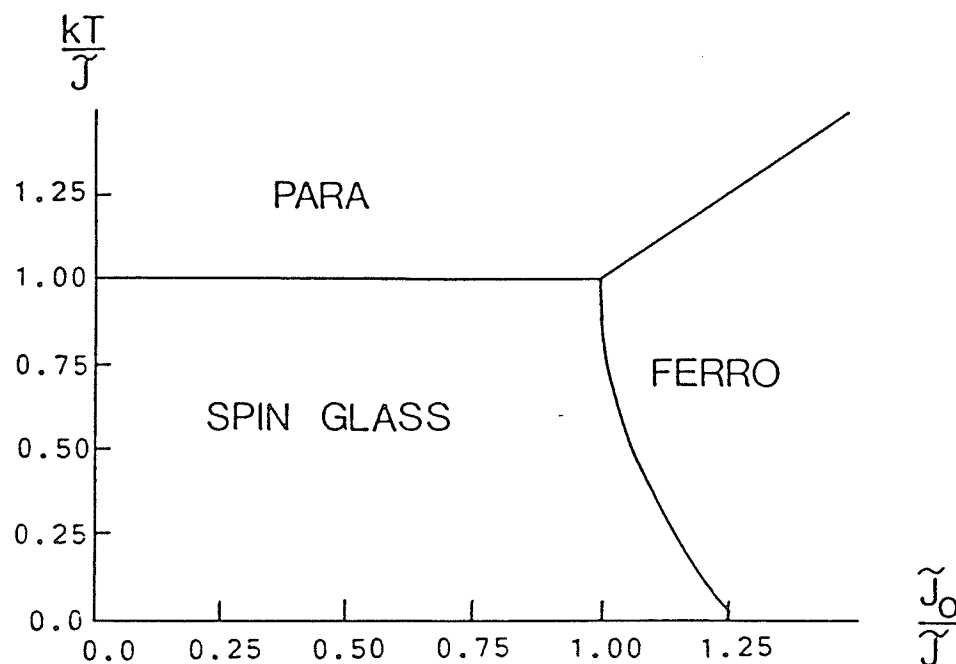


Figure 1.13. Phase diagram in the Sherrington-Kirkpatrick model. (Sherrington et al., 1975)

The susceptibility from the Sherrington-Kirkpatrick model takes on the form (Sherrington and Kirkpatrick, 1975)

$$\chi(T) = \frac{\chi_0}{1 - \mathcal{J}_0 \chi^0}$$

where χ^0 is the susceptibility for $\mathcal{J}_0=0$. The susceptibility obeys a Curie law above the phase transition which occurs at $T_0 = \mathcal{J}_0/k_B$. Figure 1.14 shows the susceptibility in both an applied field and zero field.

There is a flaw in the Sherrington-Kirkpatrick model: the entropy becomes negative as $T \rightarrow 0$. De Almeida and Thouless (1978) examined the model in the Ising version and found that this unphysical result is due to the instability of the solution below T_0 where the symmetry between replicas breaks down. This instability also occurs in non-zero fields and hence an instability line (AT line) can be drawn in the H-T plane. This line represents the transitions from paramagnetic to replica-symmetry-broken order.

Gabay and Toulouse (1981) investigated the Sherrington-Kirkpatrick model using a Heisenberg spin system and found two transition lines. The Gabay Toulouse (GT) line represents the onset of canting or transverse irreversibility while the AT line expresses the longitudinal irreversibility. Therefore, the system crosses over to regimes of weak and then strong irreversibility as it goes across the GT and AT lines respectively.

Figure 1.15 is a 3-dimensional diagram showing the AT and GT lines in the spin glass region in non-zero fields. The extension of these two lines to the ferromagnetic region in zero field is also drawn.

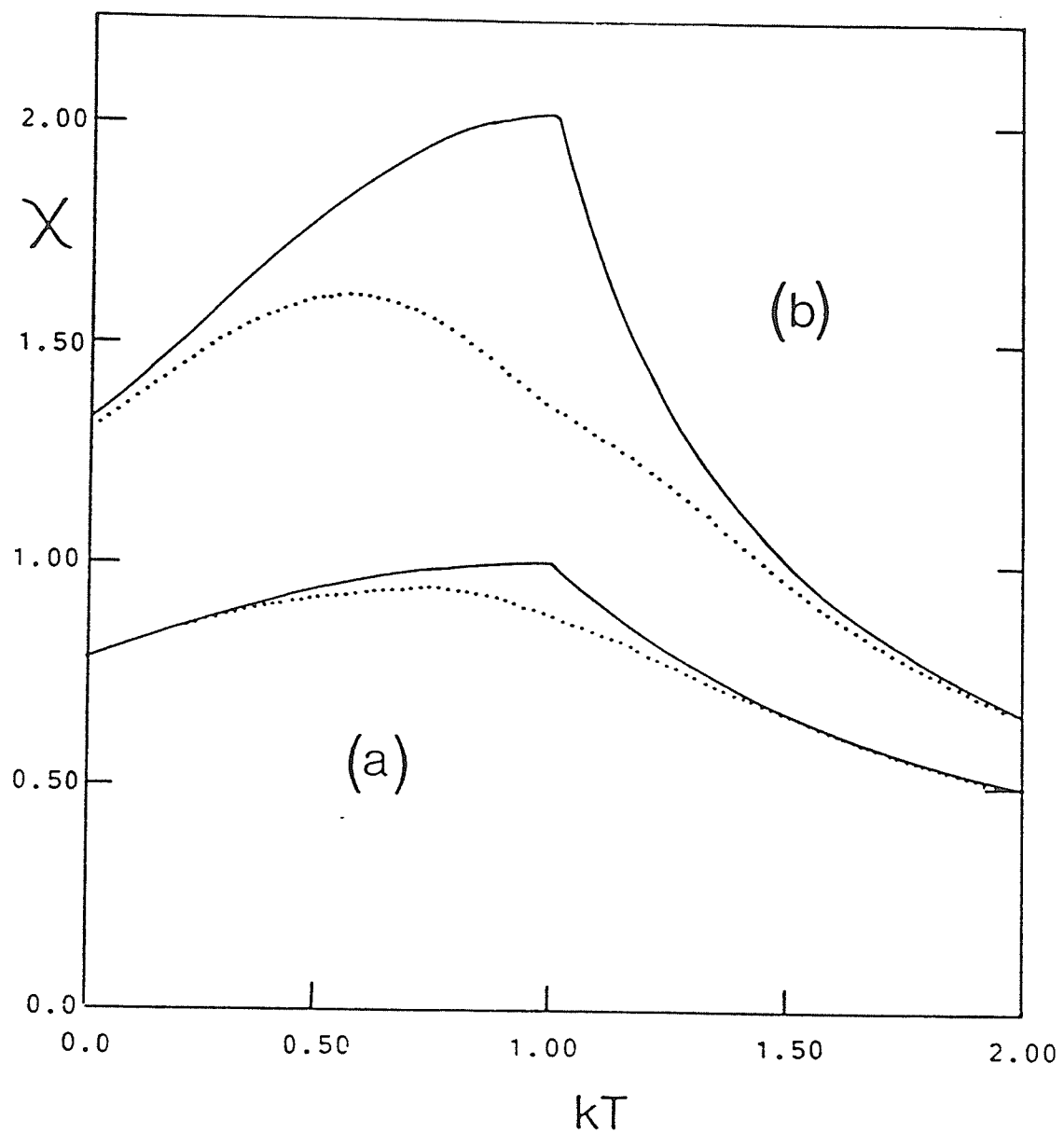


Figure 1.14. Predicted behavior of the differential susceptibility in the Sherrington-Kirkpatrick model. The solid lines represent the susceptibility in zero applied field, while the dotted lines are for an applied field of $H=0.1J$. (After Sherrington et al., 1975)

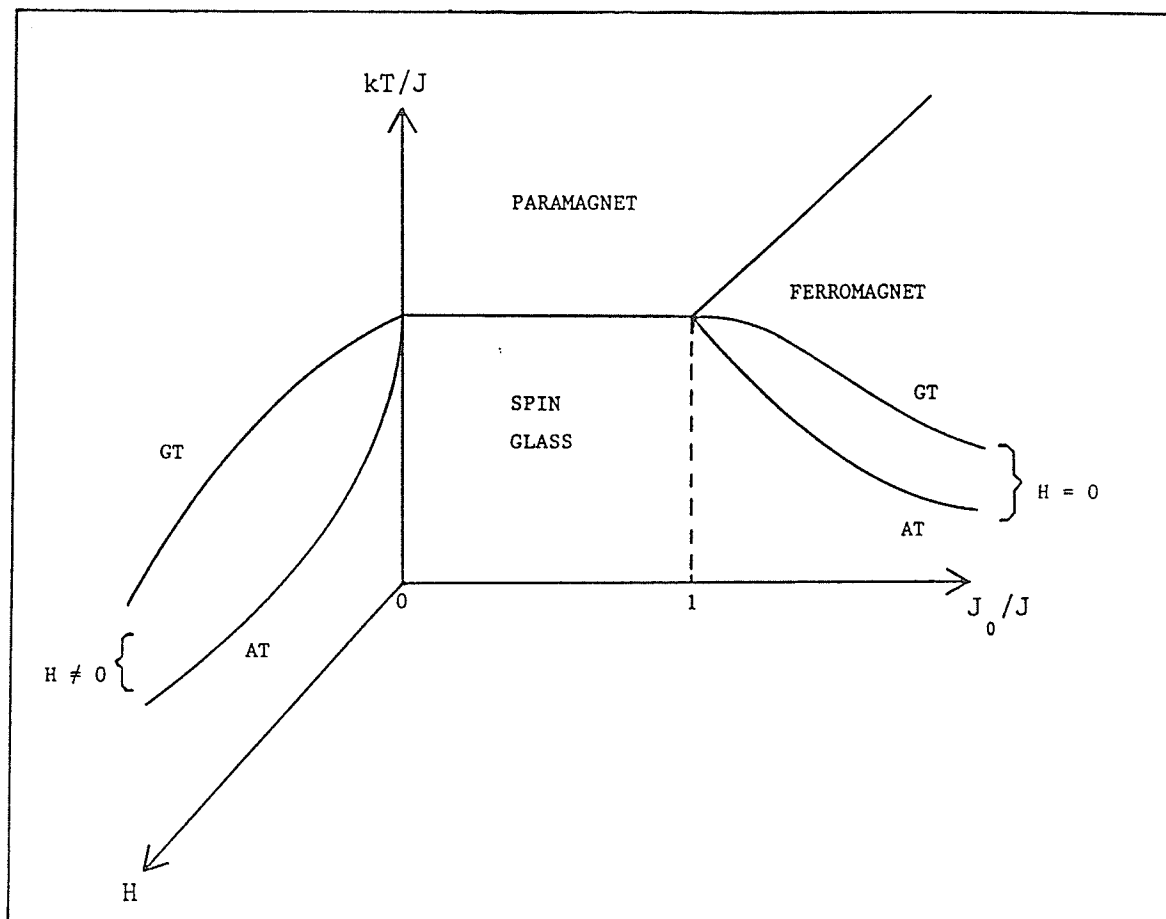


Figure 1.15. Phase diagram of the Sherrington-Kirkpatrick model in 3-dimensions.

1.7 PHASE TRANSITIONS AND SCALING LAWS

The central question in the study of a spin glass system is whether it actually goes through a phase transition at T_0 . If this is the case, one would expect to see a discontinuity in some physical properties of the system at T_0 , such as the susceptibility.

In thermodynamics, one can characterize a phase transition by the behaviour of the Gibb's function. The Gibb's function for a magnetic system is defined by:

$$G(T,H) = U - TS - MH$$

where U is the internal energy, S the entropy and T the temperature. Its differential form is given by (Elliott, 1983)

$$dG = -SdT - MdH$$

A phase transition occurs when there is a discontinuity in the Gibb's function. The order of the phase transition is the lowest order of derivative of the Gibb's function in which a discontinuity appears. Hence one can identify the order of the phase transition by observing the discontinuities in the thermodynamic quantities which are related to

the Gibb's function. For example, the susceptibility and specific heat are given by (Elliott, 1983):

$$\chi_T = - \frac{\partial^2 G}{\partial H^2} \bigg|_T, \quad C_H = - T \frac{\partial^2 G}{\partial T^2} \bigg|_H$$

If a magnetic system experiences a second order phase transition, the above two quantities would become discontinuous at T_0 while the first derivative of G would remain continuous throughout.

The cusp in the a.c. susceptibility of spin glasses suggests a phase transition which, however, is not consistent with the results of specific heat measurements. Therefore, the spin glass phase transition would be a more complex one than the usual thermodynamic phase transition outlined above.

The critical phenomena at a phase transition can be characterized by a set of indices called the 'critical-point exponents'. In examining the phase change, it is natural to define a reduced temperature:

$$t = \frac{T - T_0}{T_0}$$

Now, the critical-point exponent ϕ of a thermodynamic quantity $D(t)$ is defined as

$$\phi = \lim_{t \rightarrow 0} \frac{\ln D(t)}{t}$$

which means D goes as t^ϕ as $t \rightarrow 0$. For a ferromagnetic phase transition, the critical exponents are defined from the spontaneous magnetization, the susceptibility, and the specific heat in zero field as follows (Stanley, 1971):

$$\frac{M(T)}{M(0)} = B(-t)^\beta [1 + \dots] \quad \begin{matrix} H = 0 \\ T < T_0 \end{matrix}$$

$$\frac{\chi_T}{\chi_0} = \begin{bmatrix} C' (-t)^{-\gamma'} (1 + \dots) & T < T_0 & H = 0 \\ C (t)^\gamma (1 + \dots) & T > T_0 & H = 0 \end{bmatrix}$$

$$C_H = \begin{bmatrix} A' (-t)^{-\alpha'} (1 + \dots) & T < T_0 & H = 0 \\ A (t)^{-\alpha} (1 + \dots) & T > T_0 & H = 0 \end{bmatrix}$$

where $M(0)$ and χ_0 are normalization constants, A, A', B, C, C' are the coefficients of the leading term in the expressions and $\alpha, \alpha', \beta, \gamma, \gamma'$ are critical indices given by Stanley(1971).

The importance of these critical-point exponents is that they give a set of universal inequalities describing the phase transition, which transcends a particular magnetic system. For example, the Rushbrooke inequality is (Stanley, 1971)

$$\alpha^2 + 2\beta + \gamma' \geq 2$$

This set of inequalities can become equalities if the Scaling Law is invoked.

The Static Scaling Law for the ferromagnetic system is based on the assumption that the Gibbs function G is a generalized homogeneous function, ie. (Stanley, 1971)

$$G(\lambda^{a_t} t, \lambda^{a_H} H) = \lambda G(t, H)$$

This equation is satisfied with any value for λ , and two scaling parameters a_t and a_H . It can be shown that all the critical-point exponents can thus be written in terms of a_t

and a_H . It also follows that the set of inequalities mentioned above can be shown to be equalities. For instance, the Rushbrooke inequality becomes (Stanley, 1971)

$$\alpha^2 + 2\beta + \gamma' = 2$$

Another consequence of this homogeneity assumption of the Gibbs's function is the scaling equation of state which relates the magnetization of a ferromagnet to the magnetic field and reduced temperature (Ho, 1981):

$$M = t^\beta F\left(\frac{H}{t^{\alpha+\beta}}\right)$$

The corresponding susceptibility is (Yeung, 1987):

$$\chi(H, t) = \left(\frac{\partial M}{\partial H}\right)_t = t^{-\gamma} \left[1 + \left(\frac{H}{t^{\gamma+\beta}}\right)^2 g_2(t) + \left(\frac{H}{t^{\gamma+\beta}}\right)^4 g_4(t) + \dots\right]$$

where $g_i(t)$ are non-universal correction terms. All the expansion terms diverge as $t \rightarrow 0$. including the zero field susceptibility, $\chi(0, t) \sim t^{-\gamma}$.

The Scaling Law equation of state for the spin glass has not been developed as formally as that for the pure ferromagnet. No evidence exists for a critical divergence in the zero field susceptibility for a spin glass. However, a field expansion of the coupled equations of the Sherrington-Kirkpatrick model for a spin glass (Yeung, 1987) shows that while the coefficient of the linear term in the magnetization is finite for all t , the coefficient of the non-linear terms in the magnetization all diverge as $t \rightarrow 0$, suggesting that the non-linear terms in the magnetization could be described by a scaling law of the form (Omari et al., 1983):

$$M_{NL}(H, t) = M(H, t) - a(t) H = H t^{\beta^*} F^*\left(\frac{H^2}{t^{\gamma^* + \beta^*}}\right)$$

where M_{NL} denotes the non-linear magnetization, $a(t)$ the coefficient of the linear magnetization, and γ^* and β^* are critical exponents.

Such an expression of the Scaling Law for the spin glass system has been derived phenomenologically by Susuki(1977).

2. EXPERIMENTAL

2.1 SAMPLE PREPARATION

The two PtMn samples (Pt + 2400 ppm Mn and Pt + 1100 ppm Mn) were prepared from a master sample of Pt + 2.24 at.% Mn. The component materials used in preparing the master sample were 99.99% pure platinum wire from Leico Industries Inc. of New York and specpure grade manganese flake from Johnson Matthey Chemicals Limited of London, England. A solution of 50% nitric acid, 25% acetic acid and 25% water was used to etch the manganese flake to get rid of the oxide on the surface.

The Pt wire was melted into a pellet shape on the water cooled copper hearth of an argon arc furnace with a surrounding atmosphere of 200 torr pressure of 99.996% pure argon gas. The pellet was then rolled into a sheet.

The appropriate amounts of Pt and Mn were then melted in the arc furnace with the Pt sheet wrapped around the etched Mn flake in order to minimize the amount of Mn loss during melting. The melting process was repeated six times with the sample inverted each time to insure the homogeneity of the alloy. The final product was then annealed at a temperature of $650 \pm 20^{\circ}\text{C}$ for 34.5 hours to remove defects as much as possible. As a result, the master sample had a nominal concentration of 2.24 at.% Mn in Pt. The range of con-

centration, if melting loss was totally due to Mn or to Pt, was from 1.94 at.% to 2.24 at.% respectively.

The nominal 2400 ppm Mn in Pt sample was prepared by diluting the master sample with pure Pt. The master sample and the additional pure Pt were melted in the arc furnace under the same conditions as before. The melting was repeated seven times with the sample inverted every time. There was negligible loss during these melts. The alloy was then annealed at a temperature of 650°C for 35 hours. After the sample was spark cut into a cylindrical shape with average diameter of 0.360 cm and length of 1 cm, it was annealed again at the same temperature for about 24 hours. Finally, the cylindrical sample was electropolished with the solution of

60% saturated solution of calcium chloride in water
36% water
4% HCl

using 10 volts and an a.c. current of 3 amp.

Taking into account the melting loss in preparing the master sample (the melting loss in the preparation of the dilute sample was negligible), the concentration range of the sample was from 2060 ppm to 2400 ppm.

The nominal 1100 ppm Mn in Pt sample was prepared from the same master alloy using an identical technique. The

melting loss during the preparation of the 1100 ppm sample was also negligible. The concentration range for the sample was from 970 ppm to 1100 ppm.

1.2 DILUTION REFRIGERATOR

The magnetization measurements were performed in a He^3/He^4 Dilution Refrigerator system purchased from S.H.E. Corporation, San Diego. The system consisted of a model DRI-236 Dilution Refrigerator Cryostat, and a Pumping and Gas Handling System. These will be discussed in more detail later.

The operation of a He^3/He^4 dilution refrigerator is based on the fact that a mixture of approximately 30% helium³ and 70% helium⁴ separates into two phases at low temperatures. Figure 2.1 is a phase diagram of liquid $\text{He}^3\text{-He}^4$ mixtures. For temperatures above approximately 0.9K, the liquid is essentially a homogeneous mixture of the 2 isotopes; the λ line separates the normal and the superfluid phase. Phase separation starts to occur at a temperature of about 0.9K. At very low temperatures, one phase is virtually pure liquid He^3 and the other phase is the pure He^4 containing about 6.3 at.% He^3 even as the temperature approaches absolute zero (Rose-Innes, 1973). He^3 'floats' on top of the He^4 as it is lighter than the latter. The He^4 -rich phase has superfluid properties and hence He^3 moves through it unhindered. Therefore, the He^4 -rich phase is analogous to

a free space containing a gas; the He^4 provides a space for He^3 to move freely like the atoms of a gas.

A liquid can be cooled by evaporation when its vapour is pumped away. In the same way, if He^3 atoms are removed from the He^4 -rich phase, the concentrated He^3 liquid would be cooled down because of the 'latent heat of evaporation' as He^3 atoms 'evaporate' into the He^4 -rich phase.

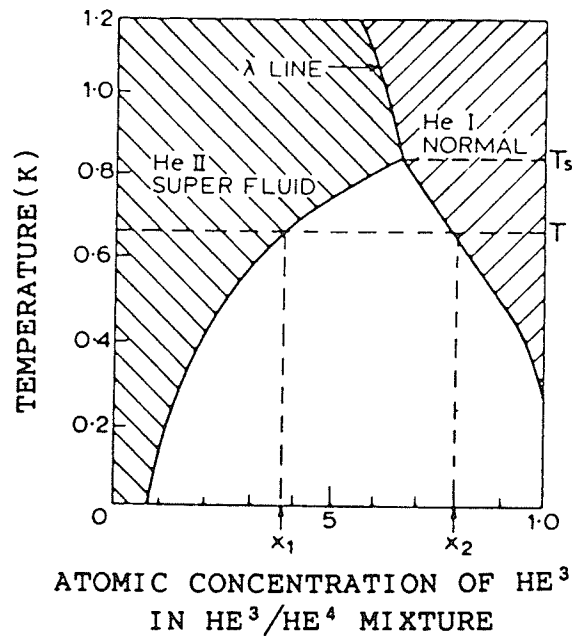


Figure 2.1. Phase diagram of liquid He^3/He^4 mixtures. At any temperature T less than T_s (separation temperature), two phases appear with the concentration of x_1 and x_2 . The λ line separates the superfluid phase from the normal phase. (After Stanley, 1971)

2.2.1 Cryostat

Figure 2.2 shows a diagram which contains the essential features of a He^3/He^4 dilution refrigerator cryostat. In the dilution cryostat, the cooling takes place in the mixing chamber. The proportion of He^3 and He^4 in the mixture is chosen so that, when phase separation occurs, the upper part of the mixing chamber is filled with concentrated He^3 liquid and the lower part with He^4 -rich phase.

The He^3 atoms are distilled from the He^4 -rich solution in the still which is connected with the mixing chamber by a capillary tube. A heater is installed in the still to provide heat for the evaporation of He^3 atoms which evaporate preferentially in the He^3 - He^4 mixture because of the lower boiling point of He^3 .

The 1°K cold plate is a small volume which has a capillary opening to the helium dewar. Helium flowing in from the helium dewar is pumped on, so that the temperature of the cold plate can be reduced to approximately 1°K. The cold plate condenses and cools the incoming He^3 - He^4 mixture which is mostly in gaseous form.

Each step on the six-step heat exchanger is made up of two copper tubes in close contact, each containing sintered copper. The close thermal contact of the two tubes enables the cool outcoming mixture to further cool the 'warm' incoming one before the latter reaches the mixing chamber.

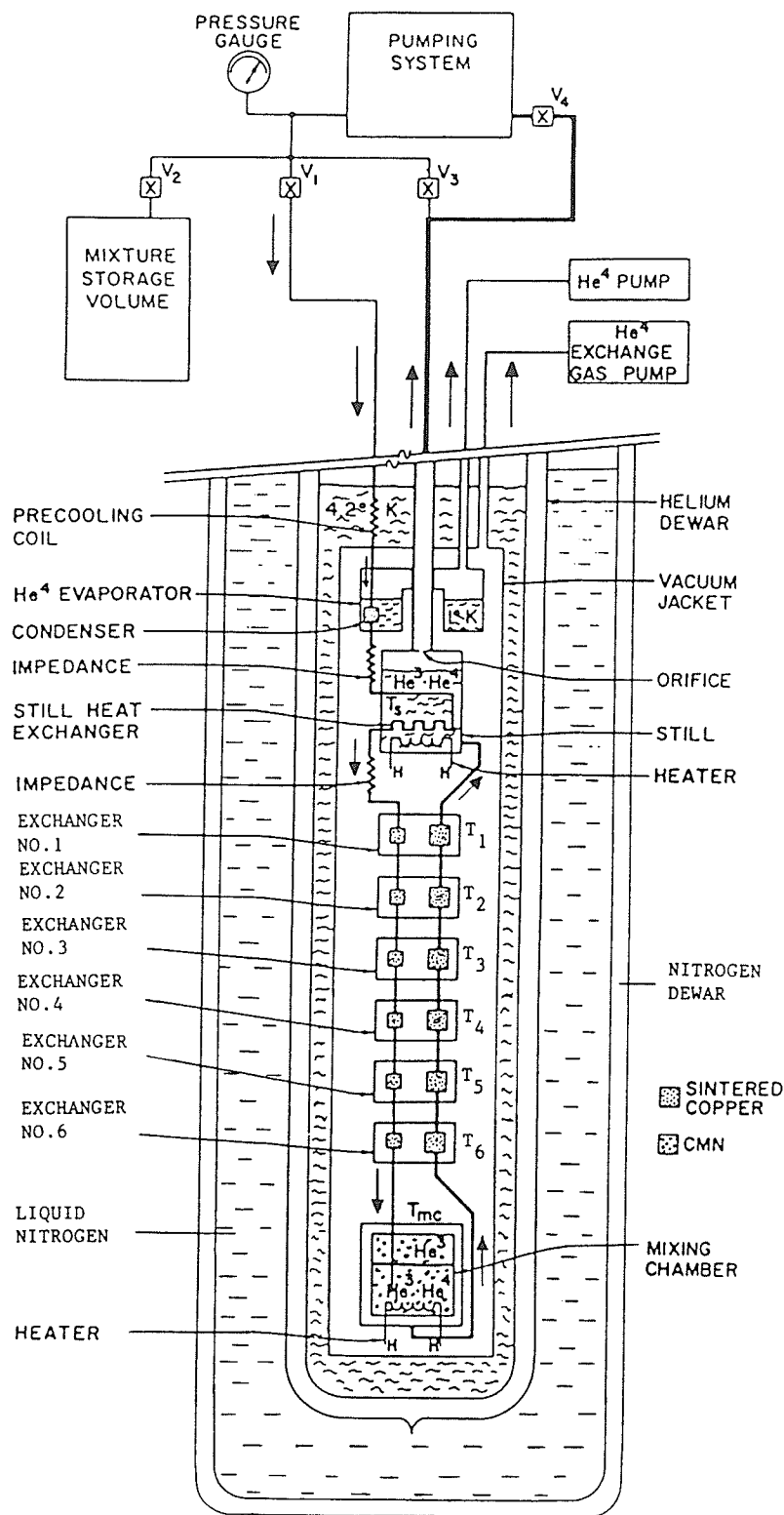


Figure 2.2. Schematic diagram of a He^3/He^4 Dilution Refrigerator Cryostat.

The $\text{He}^3\text{-He}^4$ phase separation occurs in the mixing chamber and cooling starts when He^3 atoms are removed to replace those 'evaporated' in the still. The 'evaporated' He^3 atoms are pumped out of the still and subsequently returned to the cryostat. The warm He^3 gas is initially cooled as it passes through the precooling coil in the helium dewar. The mixture condenses as it travels through the 1°K cold plate and is further cooled down as it is forced through the impedance and goes through the still. The final cooling of this incoming mixture takes place in the heat exchangers before it enters the mixing chamber to replenish the He^3 atoms removed.

2.2.2 Pumping and Gas Handling System

Figure 2.3 is a diagram of the pumping and gas handling system which has four mechanical pumps and two diffusion pumps. The Sargent Welch model 1374 mechanical pump is used to pump on the 1°K cold plate and the He^4 dewar. A Sargent Welch 1402 mechanical pump, along with a diffusion pump, evacuates the vacuum can. Another Sargent Welch 1402 pump is for pumping on the interconnecting tubes and cleaning the cold traps. An Edwards model 660 sealed mechanical pump is used for circulating the mixture in a closed system. The second diffusion pump, the booster, is to reinforce the circulation of the mixture but it was found to be not very useful in our system.

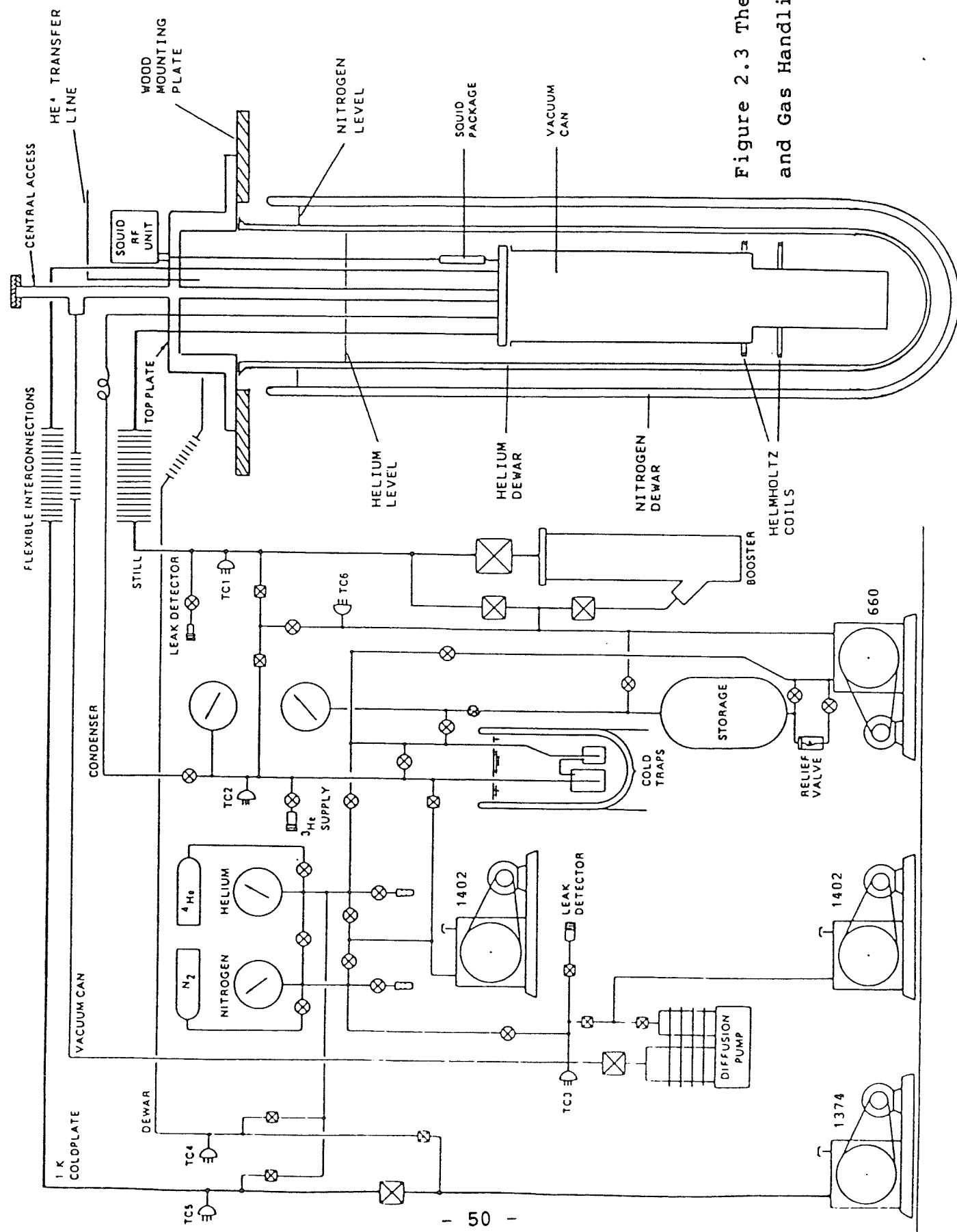


Figure 2.3 The Pumping and Gas Handling System.

2.3 MAGNETIZATION MEASUREMENTS

The magnetization was measured with a SQUID system which included a model MFP multi-function SQUID probe, a model 300 rf head and a model 30 SQUID control unit, all of which were manufactured by S.H.E. Corporation, San Diego.

The pick-up coil system consisted of two coils, each with 9 turns of 0.007" Niobium-Titanium wire, wound astatically (counterwound) on a coil former which was made of Emerson and Cuming Stycast 1266 epoxy. The coil radius was 0.170" and the separation of the coils was 0.750" centre to centre. The epoxy coil former was installed inside a copper chamber which was suspended from the bottom of the mixing chamber with screws. The NbTi leads from the pick up coils (called the flux transformer) were enclosed in PbSn tubing and connected to two screws on the SQUID probe. Figure 2.4 shows details of the sample chamber. Figure 2.5 shows the position of the SQUID sensor and the sample chamber relative to the dilution refrigerator unit. Figure 2.6 is a close-up of the sample chamber attachment.

If the pick-up coils were perfect in geometry, the astaticity of the two coils would give zero resultant signal when both of the coils were exposed to equal magnetic field. Therefore, only the signal due to the sample, which was situated in the lower coil, would be picked up. However, the coils were not perfectly constructed and so a background

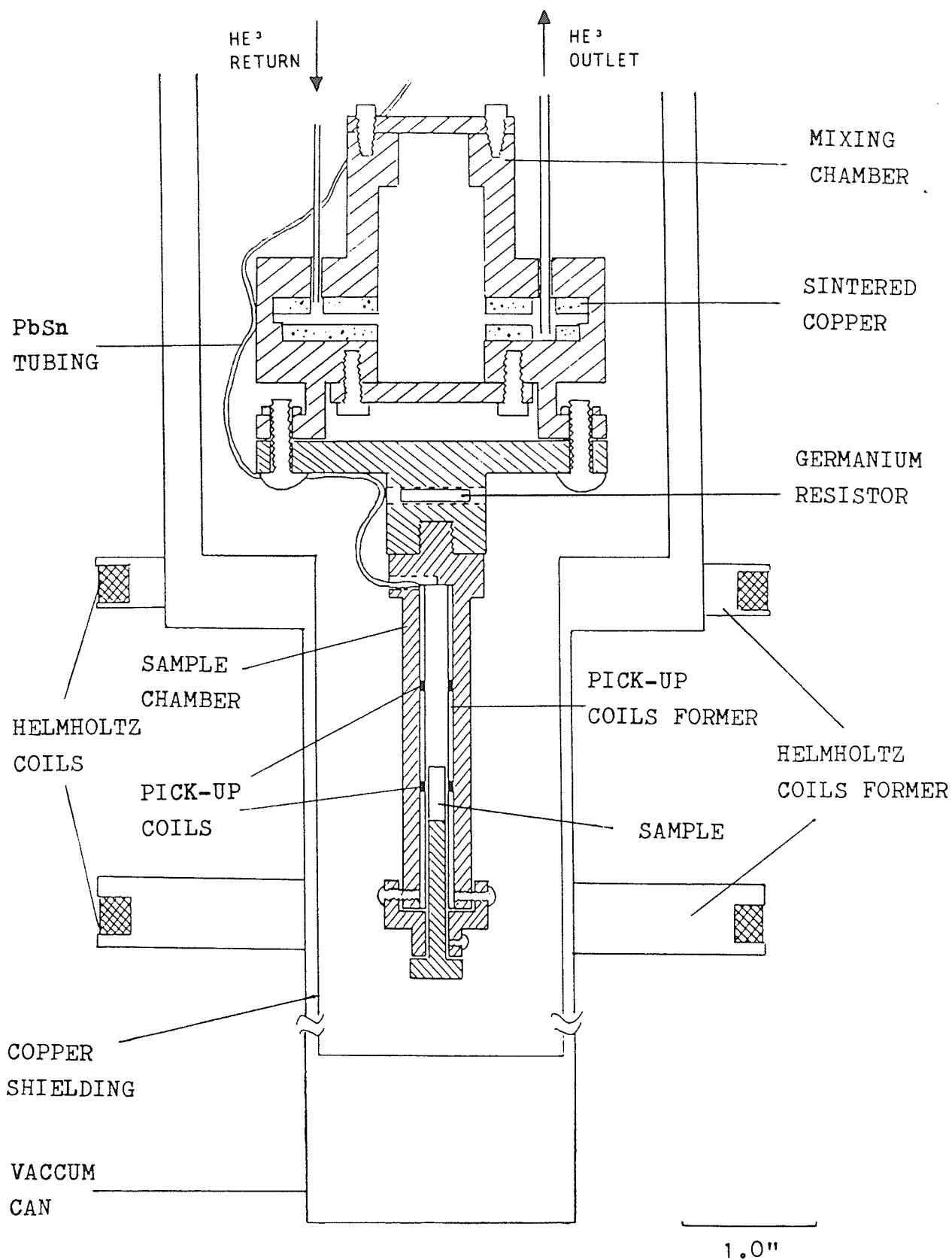


Figure 2.4. Scaled diagram of the lower part of the Refrigerator.

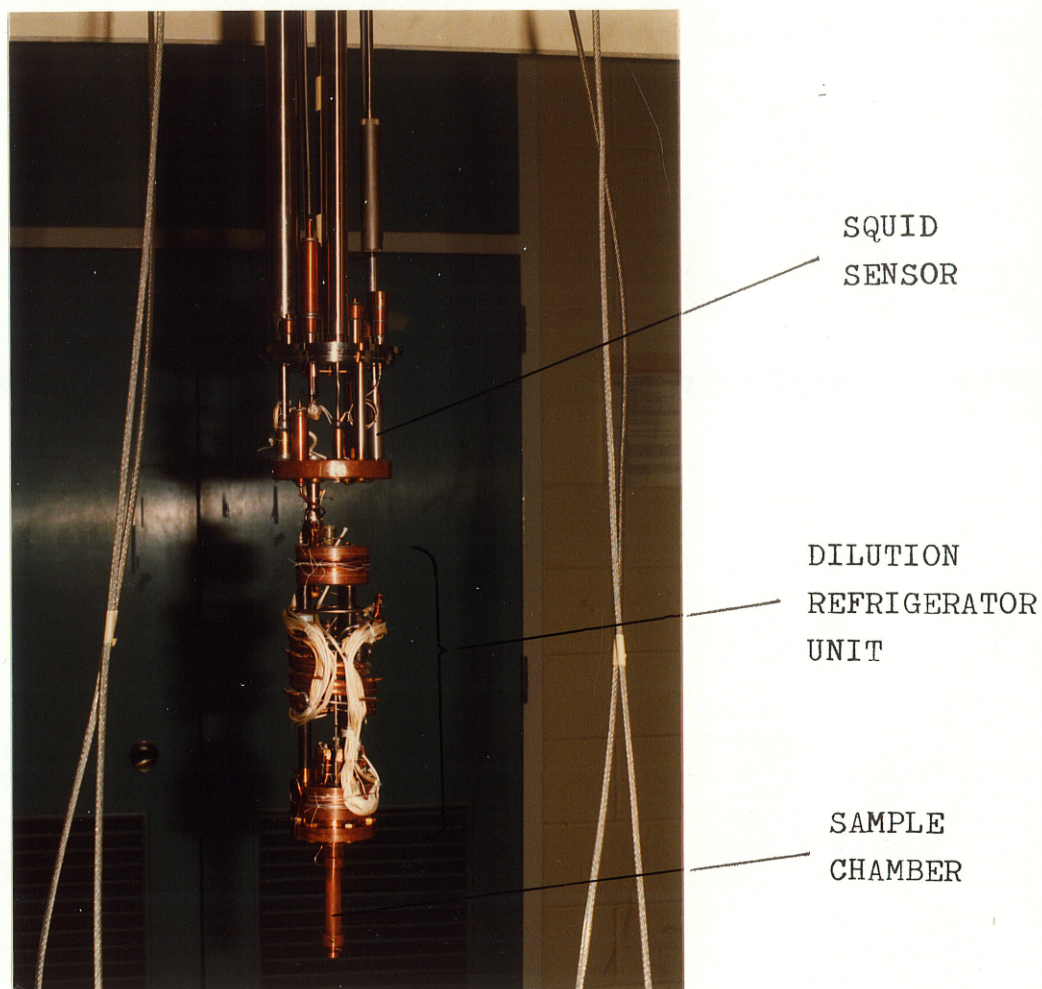
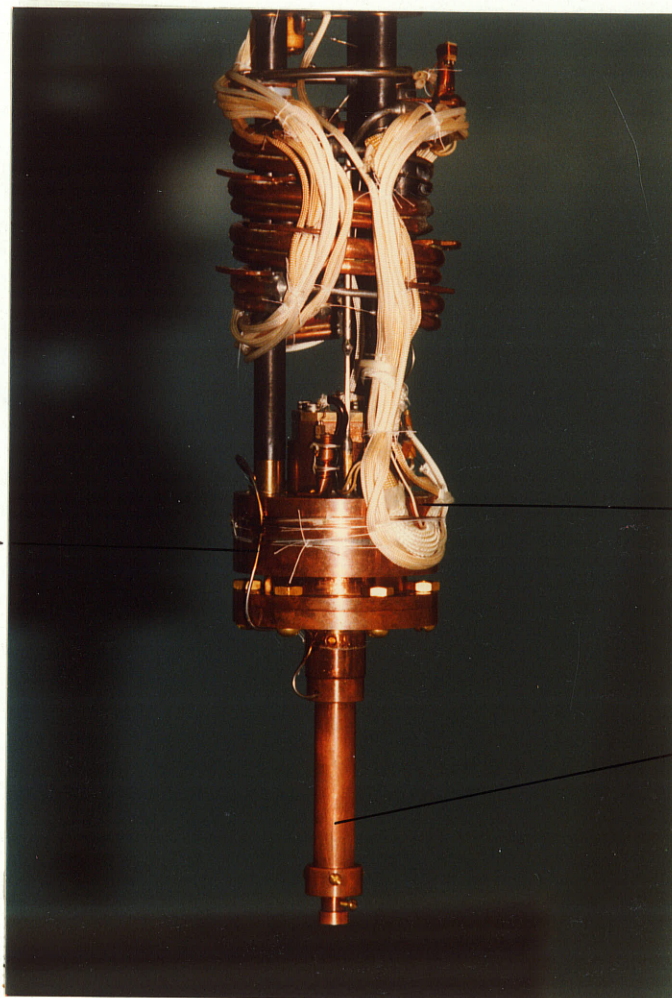


Figure 2.5. Picture of the Dilution Refrigerator Unit.

PbSn
TUBING



MIXING
CHAMBER

SAMPLE
CHAMBER

Figure 2.6. Close-up of the lower part of
the Dilution Refrigerator Unit.

signal was also registered along with the sample signal. Hence the background must be subtracted from the total signal for analysis. The magnetization data was read with a Racal-Dana model 5003 digital multimeter in volts.

The applied magnetic field was generated by a pair of Helmholtz coils and a constant current source. The latter was able to supply a current of up to 1 ampere and its circuit is shown in figures 2.7 through 2.9.

The Helmholtz pair consisted of two coils, one wound on a brass former and the other on a delrin former. The mean radius of the two coils, which was equal to their mean separation, was 6.033 cm. Each of the two coils was wound with 650 turns of #31 gauge copper wire and the Helmholtz pair provided a field of 96.81 gauss/ampere at the centre on the axis. A Racal-Dana model 5003 digital multimeter was used to read the current, by measuring the voltage across a 1Ω standard resistor. Figure 2.4 shows the position of the Helmholtz pair with respect to the sample chamber while figure 2.10 is a picture which indicates how the Helmholtz pair was mounted on the vacuum can.

A germanium resistor, (model GR-200A-30 by Lake Shore Cryotronics, Inc. of Westerville, Ohio), was mounted in good thermal contact above the sample holder (see figure 2.4) to measure the temperature of the sample. The resistance was read with a potentiometric conductance bridge (model PCB by

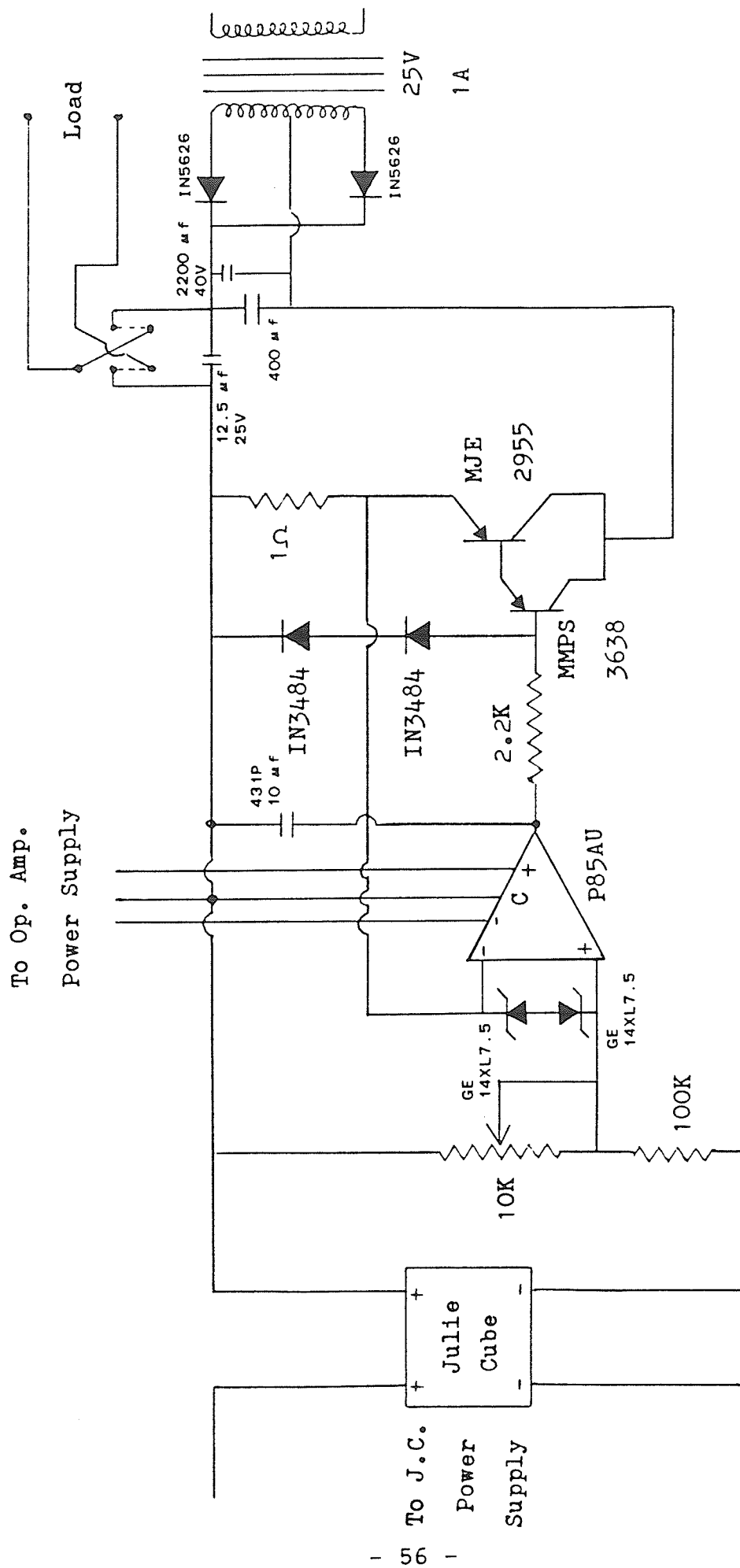


Figure 2.7. Circuit diagram of the Constant Current Supply.

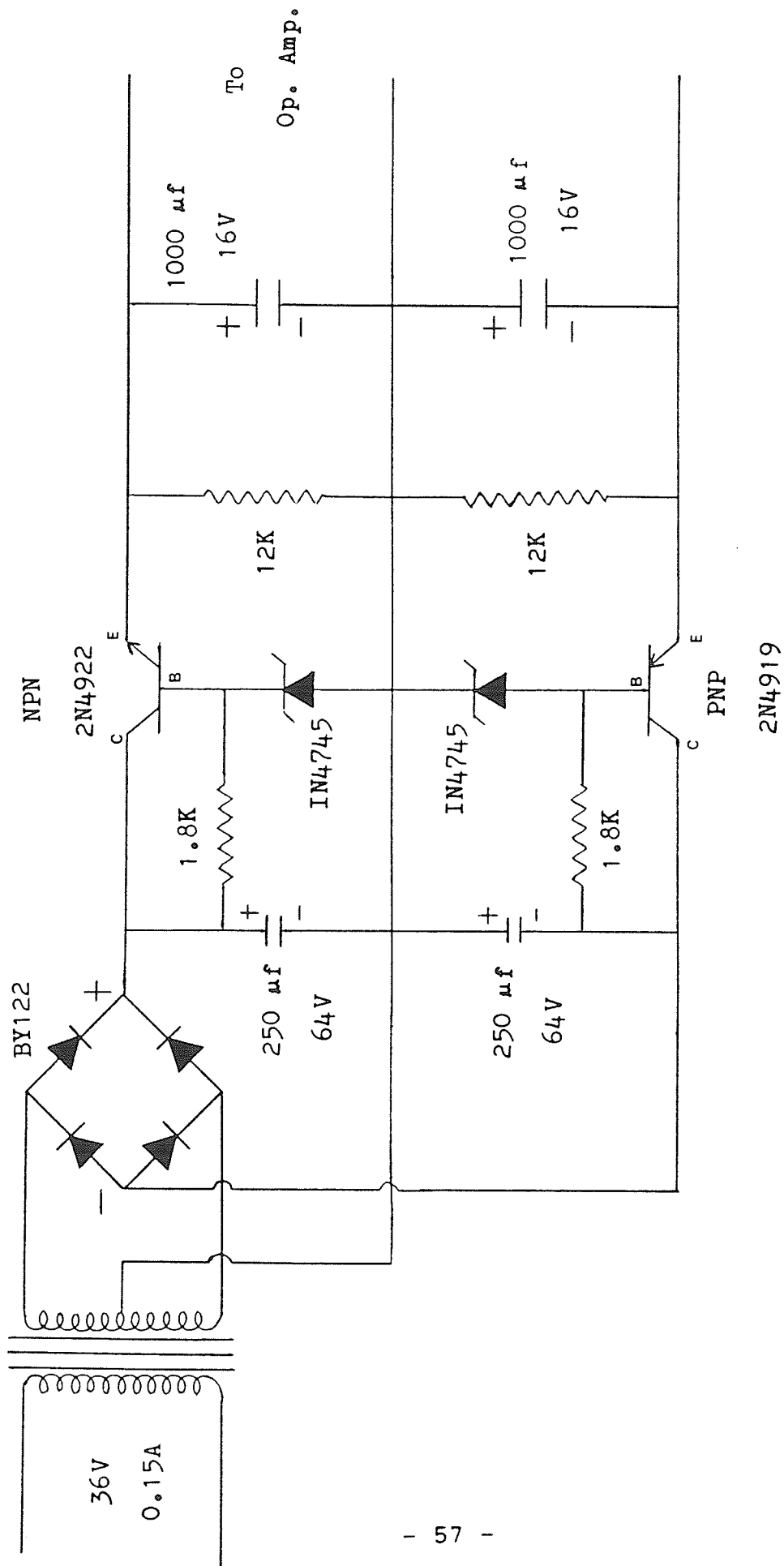


Figure 2.8. Power Supply for the Op. Amp.

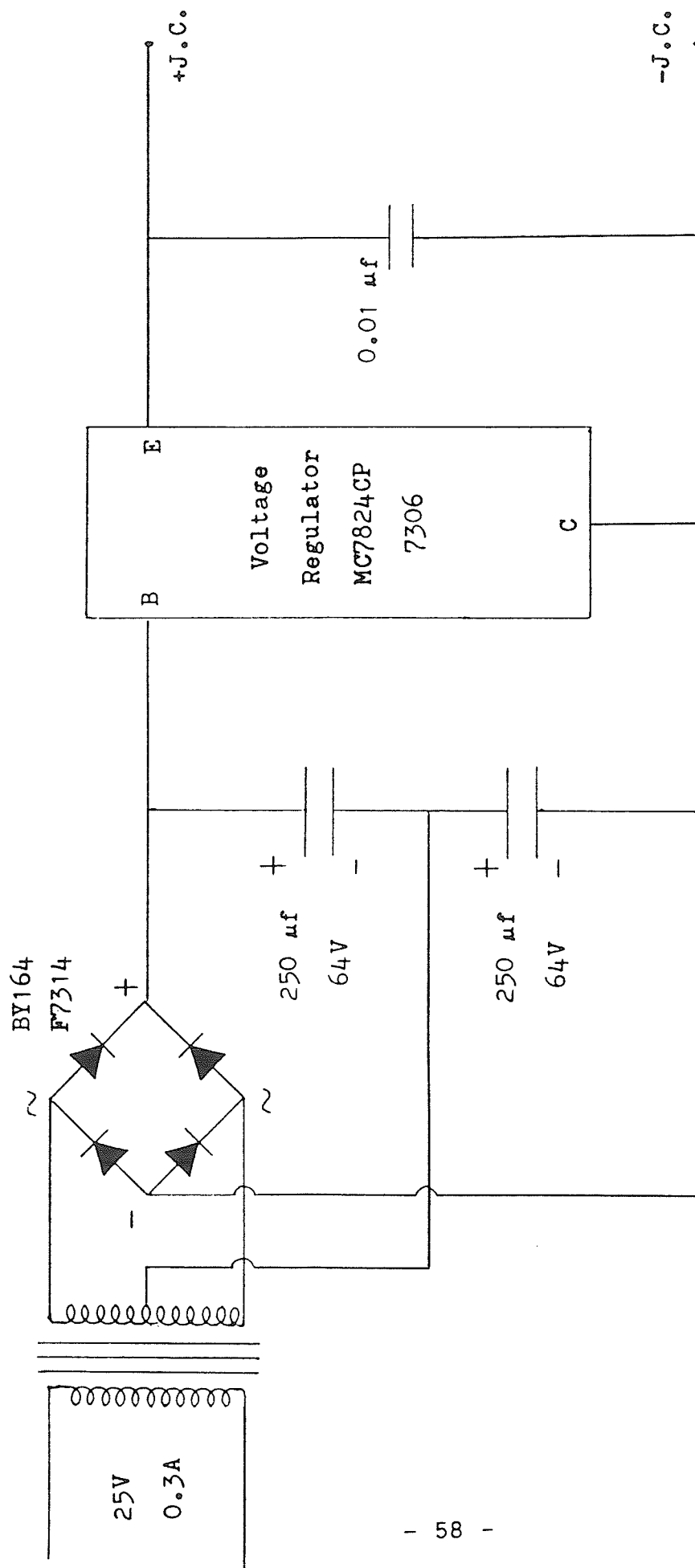


Figure 2.9. Power Supply for the Julie Cube.

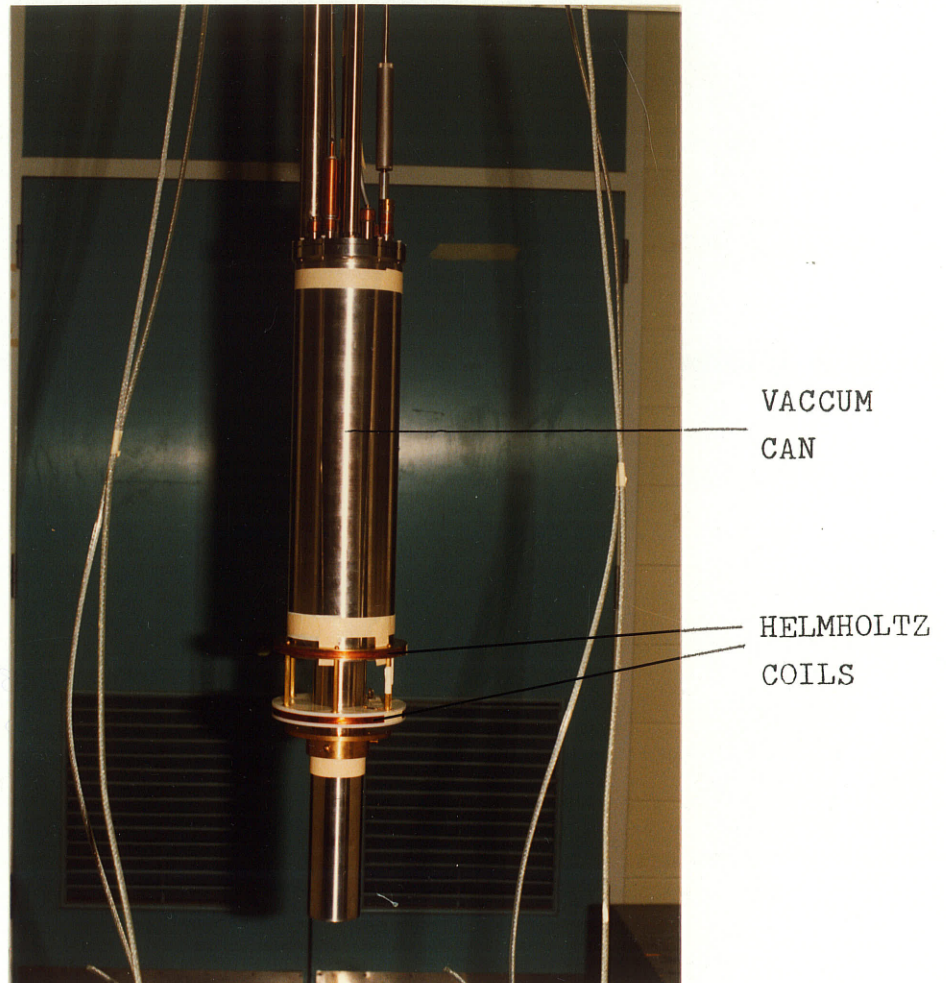


Figure 2.10. Picture of the Dilution Refrigerator Unit with the vaccum can mounted.

S.H.E., San Diego). A calibration of the germanium resistor was supplied by the manufacturer as shown in figure 2.11. A particular data point between two known points was determined by linear interpolation.

The temperature of the mixing chamber (and hence the sample) was controlled using a nominal 100Ω Speer carbon resistor and a heater, both mounted in close thermal contact on the mixing chamber. Figure 2.12 shows a block diagram of the temperature control system. A picowatt a.c. resistance bridge (model LR-10 by Linear Research, San Diego) was used to compare the resistance of the carbon resistor with the value set on a 4-disc concentric decade resistor, which corresponds to the desired temperature of the mixing chamber. The resistance bridge produced a signal which was proportional to the imbalance between the 2 resistors; the signal was fed into the temperature control unit (model ATC by S.H.E.) which in turn fed current to the mixing chamber heater. The system was found to be able to maintain a temperature stability of 5×10^{-4} Kelvin at 50 millidegree and 5×10^{-3} Kelvin at 800 millidegree.

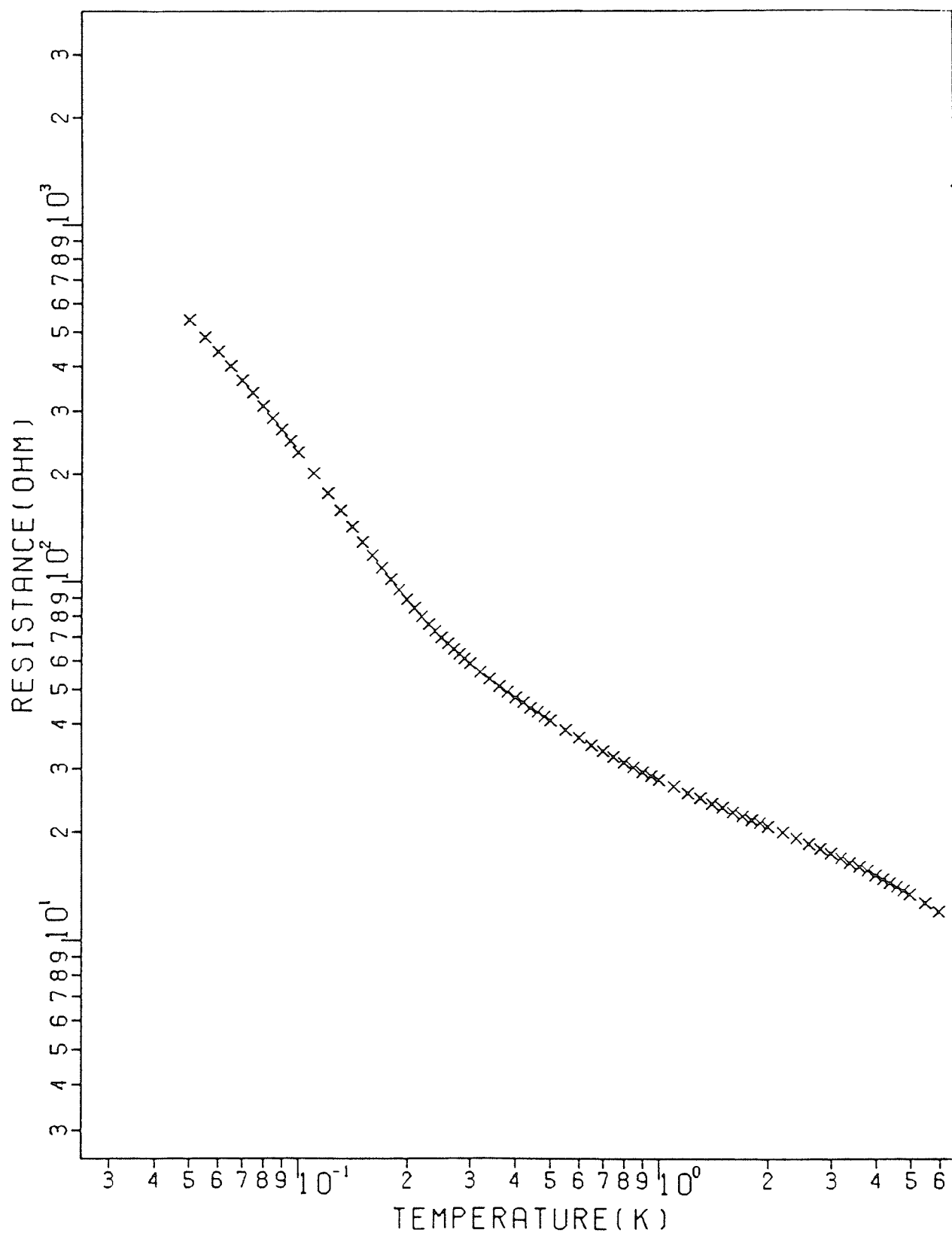


Figure 2.11. The calibration of the Germanium Resistor plotted as resistance vs temperature in logarithmic scale.

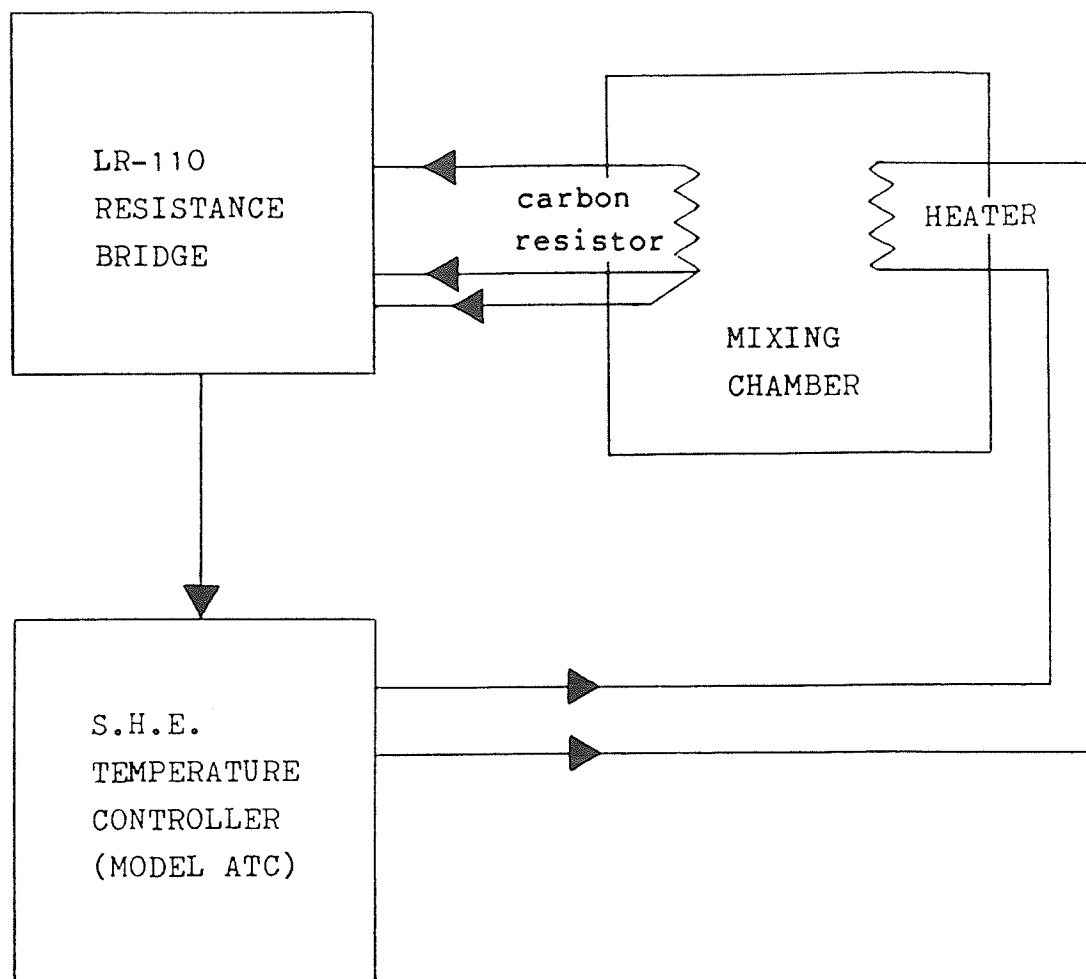


Figure 2.12. Schematic diagram of the Temperature Control System. The resistance bridge reads the resistance of the carbon resistor and compares the reading with its set resistance. The imbalance is sent to the temperature controller and then the latter feeds appropriate current to the heater.

3. RESULTS

3.1 MAGNETIC PHASE DIAGRAM FOR PTMN

Figure 3.1 is a magnetic phase diagram extracted from Wassermann(1982). At concentrations below 500 ppm, the Mn impurities are essentially noninteracting and their behaviour is dominated by the Kondo effect: the impurities become nonmagnetic as the temperature decreases below the Kondo temperature $T \approx 25$ mK for Mn in Pt. Samples with concentrations from 0.1 to about 2.5 at.% were found to show definite spin glass behaviour (Wassermann, 1982) with freezing temperatures $T_0 \propto$ impurity concentration c , and our samples, by no accident, fall into this concentration range. The RKKY interaction is believed to be the dominant mechanism at work in the spin glass regime.

From 2.5 to 9 at.% Mn, T_0 rises more rapidly than linearly with concentration, which is evidence for the onset of the formation of clusters which are antiferromagnetically ordered, and the system becomes a typical cluster glass. Between 10 and 15 at.% Mn, a ferromagnetic component develops, probably due to the presence of small regions of ferromagnetic short range ordered Pt_3Mn phase, and competes with the antiferromagnetic component, and the system enters a crossover regime.

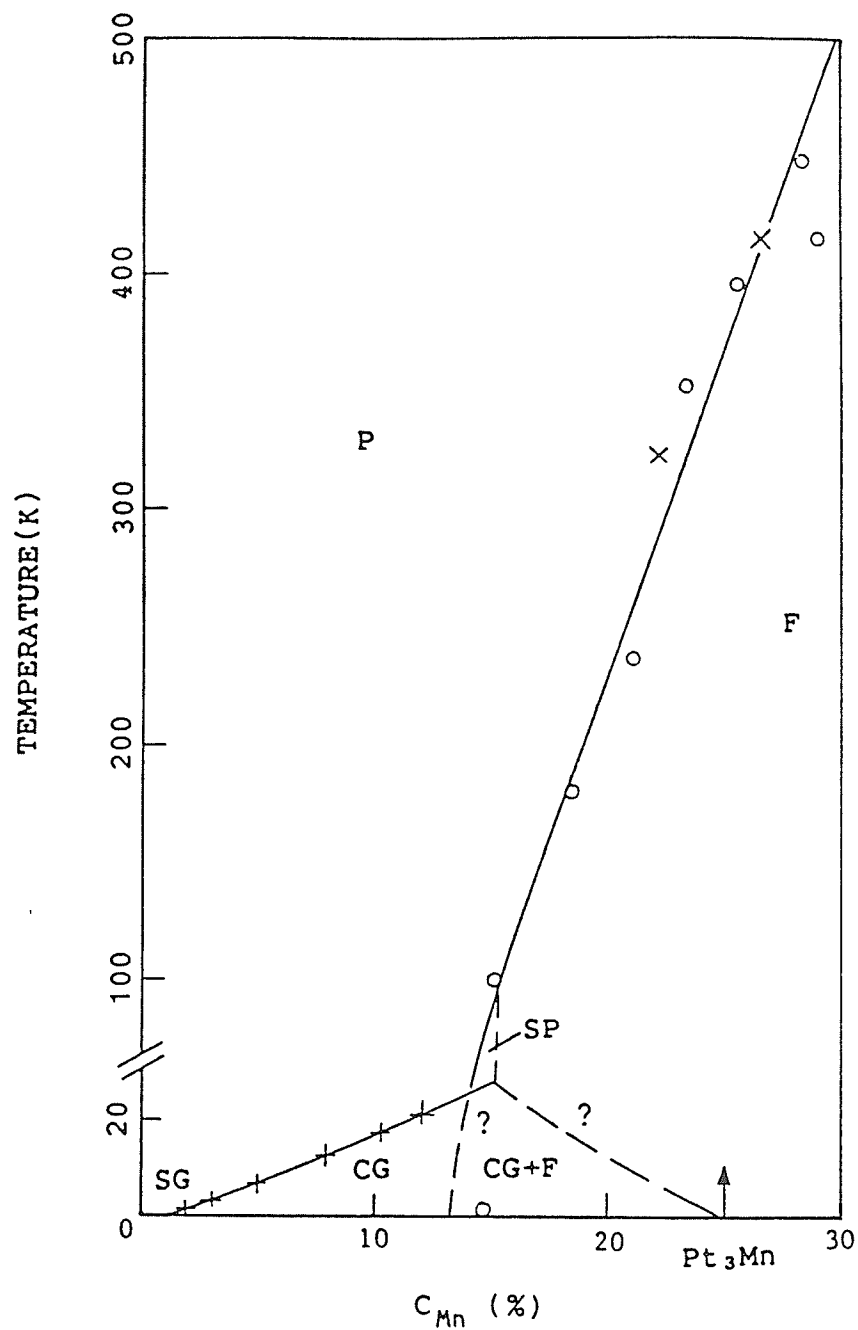


Figure 3.1. A magnetic phase diagram of PtMn plotted in temperature vs Mn concentration up to a value of 30% Mn. +, o, and x represent data from different authors. (After Wassermann, 1982)

Beyond this concentration, ferromagnetic ordering becomes dominant over the antiferromagnetic coupling; the reason may be that when three Mn atoms are located closely together during crystallization, one of the antiferromagnetic nearest neighbour (nn) Mn sites jumps to a ferromagnetic next nearest neighbour (nnn) site due to the lattice energy instability; hence, the number of nnn ferromagnetic couplings increases (Lecomte et al., 1983). Therefore, a mixed phase of cluster glass and ferromagnet exists in this concentration range up to a concentration of 25 at.% at which the stoichiometric composition Pt_3Mn is formed. Pt_3Mn is a ferromagnet and it has a face-centered cubic structure with the Mn atoms at the corners. All of the Mn atoms hence occupy the nnn sites and therefore only the nnn ferromagnetic coupling exists.

3.2 BACKGROUND SIGNAL

The two NbTi pick-up coils were wound astatically so that when they are located symmetrically about the centre of the Helmholtz pair and so exposed to equal magnetic field, the currents induced in these two pick-up coils should be equal in magnitude but opposite in direction. Therefore only the signal induced by the sample, which was situated in the lower pick-up coil, should be registered. However, due to the imperfect geometry of the pick-up coils and/or that of the Helmholtz coils, the cancellation was imperfect and a background signal was recorded along with the sample signal.

The background signal was found to be temperature independent in the temperature range we worked on. Figure 3.2 shows a typical set of background data (the x's) measured at a temperature $T = 361$ mK. The applied field and magnetization are expressed in units of amperes and volts respectively. The calibration for the applied field was ~ 98.5 Gauss/Ampere, while that for the magnetization ($\text{emu-cm}^{-3}/\text{volt}$) had not been determined at the time the thesis was written. However, the analyses are independent of the system of units chosen, and hence all subsequent plots are expressed in terms of volts and amperes.

We performed least square fits of polynomials of various order to the background and found that a fit with a linear and third power term represented the background most appropriately with coefficients as follows:

$$M = -85.20 H + 9.85 H^3$$

The solid line in figure 3.2 represents the fitted curve. Therefore, the PtMn sample data was obtained from the total data by subtracting the fitted background curve.

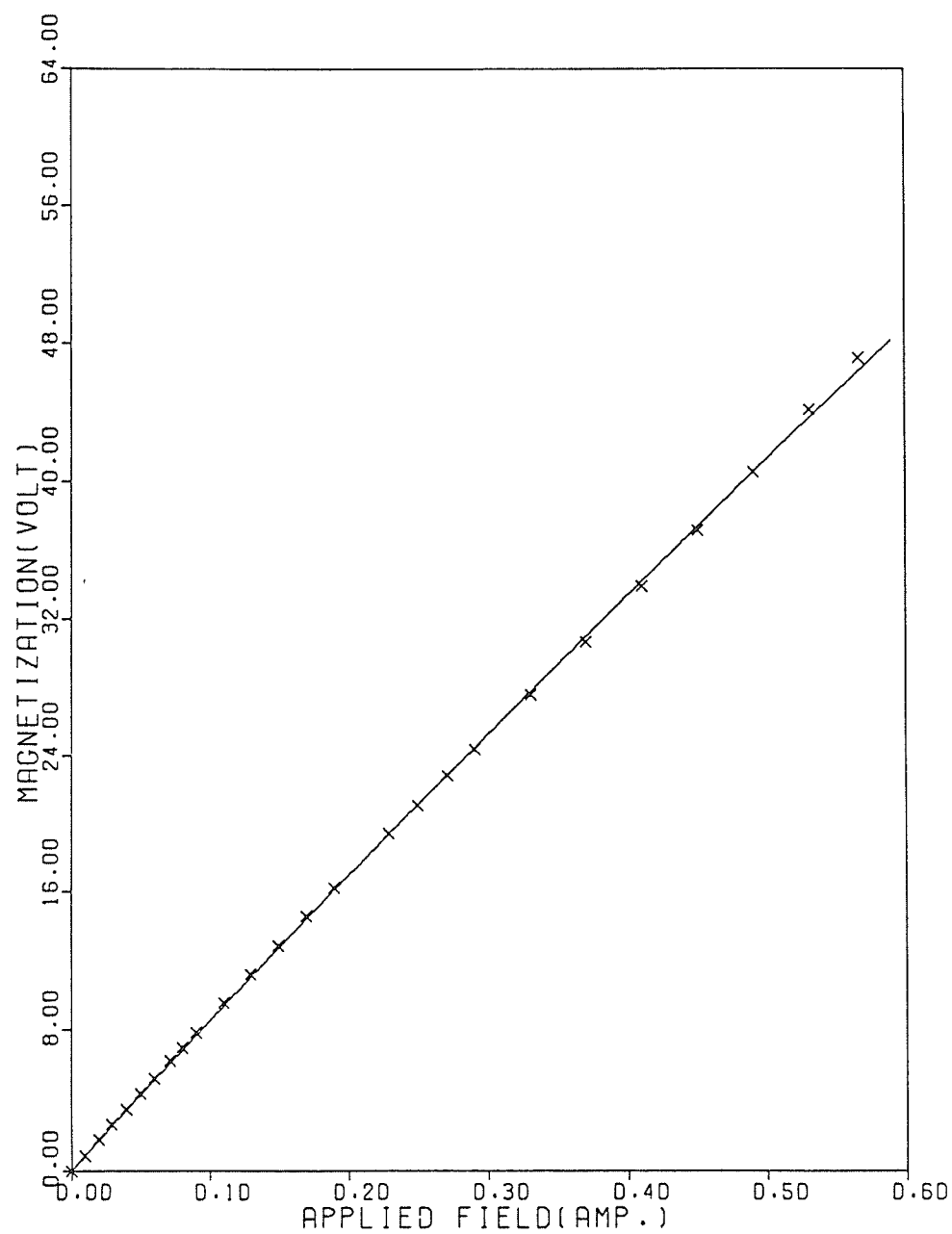


Figure 3.2. The x's represent a typical set of background at $T = 361$ mK. The solid line is a least square fit of the data to a linear plus a cubic term.

3.3 MAGNETIZATION MEASUREMENTS

3.3.1 Determination of the Freezing Temperature

A measurement of magnetization as a function of temperature at a field of 2 Gauss was carried out for each sample in order to determine the spin glass ordering temperatures. Both the 2400 ppm and 1100 ppm samples were cooled to the lowest temperature in zero field and then the magnetization was measured while the sample was warmed in a field of ~ 2 G. (A field of 0.0044 ampere, corresponding to ~ 0.44 Gauss was used to offset the local earth's magnetic field in order to achieve a zero field condition.) In addition, the magnetization of the more concentrated sample was measured again as it was cooled in the same field.

All these measurements were done with a SQUID magnetometer, which senses only changes in magnetization, and thus the original magnetization data was only recorded relative to an arbitrary zero. To determine the absolute magnitude, we assumed that the magnetization follows a Curie Law ($M \propto H/T$) at high temperatures. Therefore, by plotting the high temperature magnetization data versus $1/\text{temperature}$, we obtained the zero magnetization by extrapolating to $1/T = 0$. Figure 3.3 shows the curves of the adjusted (hence absolute) magnetization (in volts) plotted against temperature for both samples. Both of the zero field cooled curves exhibit a peak which defines the freezing temperatures. The freezing temperatures, T_0 , for the 2400 ppm and 1100 ppm samples were

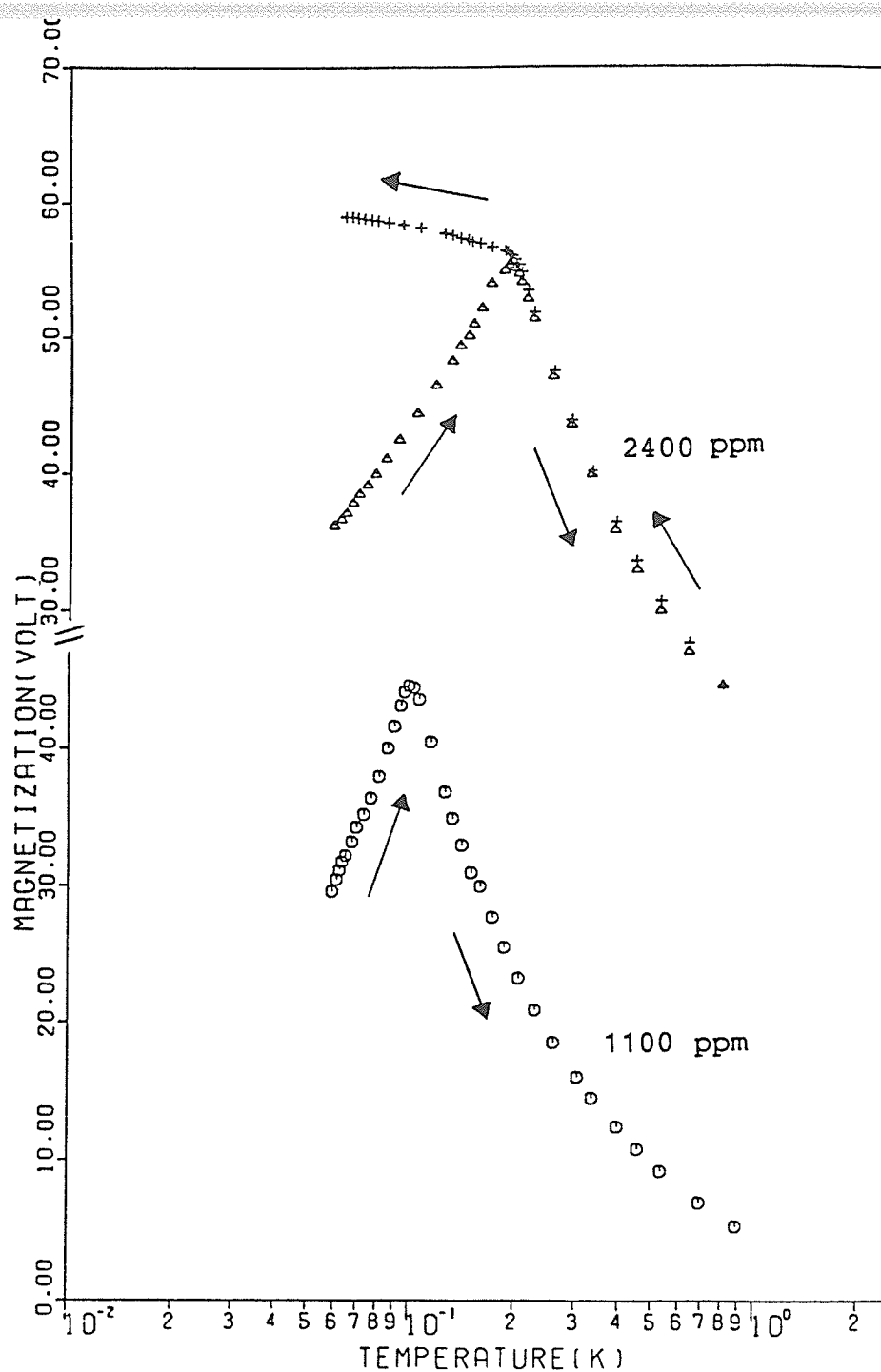


Figure 3.3. The magnetization as a function of temperature in a field of 2G. The curve for the 1100 ppm sample and the curve with a peak for the 2400 ppm sample are data of warming after cooling in zero field. The other curve for the 2400 ppm sample corresponds to cooling in a field.

found to be 200 ± 5 mK and 100 ± 3 mK respectively. The freezing temperature roughly scales with the Mn concentration, and the magnetization of the more concentrated sample is about double of that of the less concentrated one at high temperatures (well above both peaks) as expected.

The magnetization has a strong time dependence below T_0 in the zero field cooled case. The temperature of each data point was allowed to stabilize for approximately 10-15 minutes before changing to a new temperature. If we were to wait for an infinitely long time at any temperature, the magnetization would eventually reach its saturation value, represented supposedly by the field-cooled curve. We also found that time effects persisted even at temperatures above T_0 ; the time effects at temperatures above T_0 faded away quickly as the temperature increased.

3.3.2 Magnetization Data for the 2400 ppm Sample

Kaneyoshi(1975) and Southern(1976) developed a mean-field effective-field model for disordered systems with spin $1/2$ impurities, later modified by Roshko and Williams(1985) for arbitrary spin S , which yields the same coupled equations for the magnetization M and order parameter Q as those of the Sherrington-Kirkpatrick model. These coupled equations are:

$$q = \frac{1}{\sqrt{2\pi}} \int dZ \exp\left(-\frac{Z^2}{2}\right) \tanh^2\left[\frac{\gamma}{kT} q^{\frac{1}{2}} Z + \frac{\gamma}{kT} m\right]$$

$$m = \frac{1}{\sqrt{2\pi}} \int dZ \exp\left(-\frac{Z^2}{2}\right) \tanh\left[\frac{\gamma}{kT} q^{\frac{1}{2}} Z + \frac{\gamma}{kT} m\right]$$

If these equations are expanded for small values of the argument of the Brillouin function, the magnetization can be expressed as a power series in the applied field H above the ordering temperature $T_0 = J/K$:

$$M(H,t) = a(t)H + b(t)H^3 + c(t)H^5 + \dots$$

where

$$a(t) \sim \frac{1}{t + 1 - \bar{\eta}} \quad \text{with} \quad \bar{\eta} = \frac{\bar{J}}{\bar{J}}$$

$$b(t) \sim \frac{1}{t^{\gamma}} f_2(t) \quad \text{with} \quad \gamma = 1$$

$$c(t) \sim \frac{1}{t^{\gamma'}} f_4(t) \quad \text{with} \quad \gamma' = 3$$

and t is the reduced temperature defined as $(T-T_0)/T_0$. The zero field susceptibility $a(t)$ is finite for all temperatures $t \geq 0$, while the subsequent terms $b(t)$, $c(t)$ and so on

diverge as $t \rightarrow 0$ from above. $f_2(t)$ and $f_4(t)$ are non-universal correction terms: which represent non-critical contribution to M and which approach constants as $t \rightarrow 0$.

In order to verify the predictions of the field model, we measured the magnetization as a function of field at 11 different temperatures above $T_0 = 200$ mK for the 2400 ppm sample. Figure 3.4 is a M versus H plot for a few representative temperatures. The initial slope, which represents the coefficient of the linear term in the magnetization, (the zero field susceptibility), increases as the temperature approaches the freezing temperature. This behaviour agrees with the prediction for $a(t)$ which is expected to increase as the reduced temperature, t , decreases.

Figures 3.5(a) and (b) display M/H versus H^2 plots for all 11 temperatures measured. As expected, the initial slope in this case, which represents the coefficient of the cubic term in the magnetization, $b(t)$, increases as the temperature moves toward T_0 . Figure 3.6 shows the estimated cubic term in more detail for 3 out of the 11 temperatures. Notice that as the slope of the straight line increases, its range of validity in field decreases dramatically, so that as one approaches T_0 , the straight line is really only a tangent to the low field data and may represent a serious underestimate of the correct H^3 coefficient.

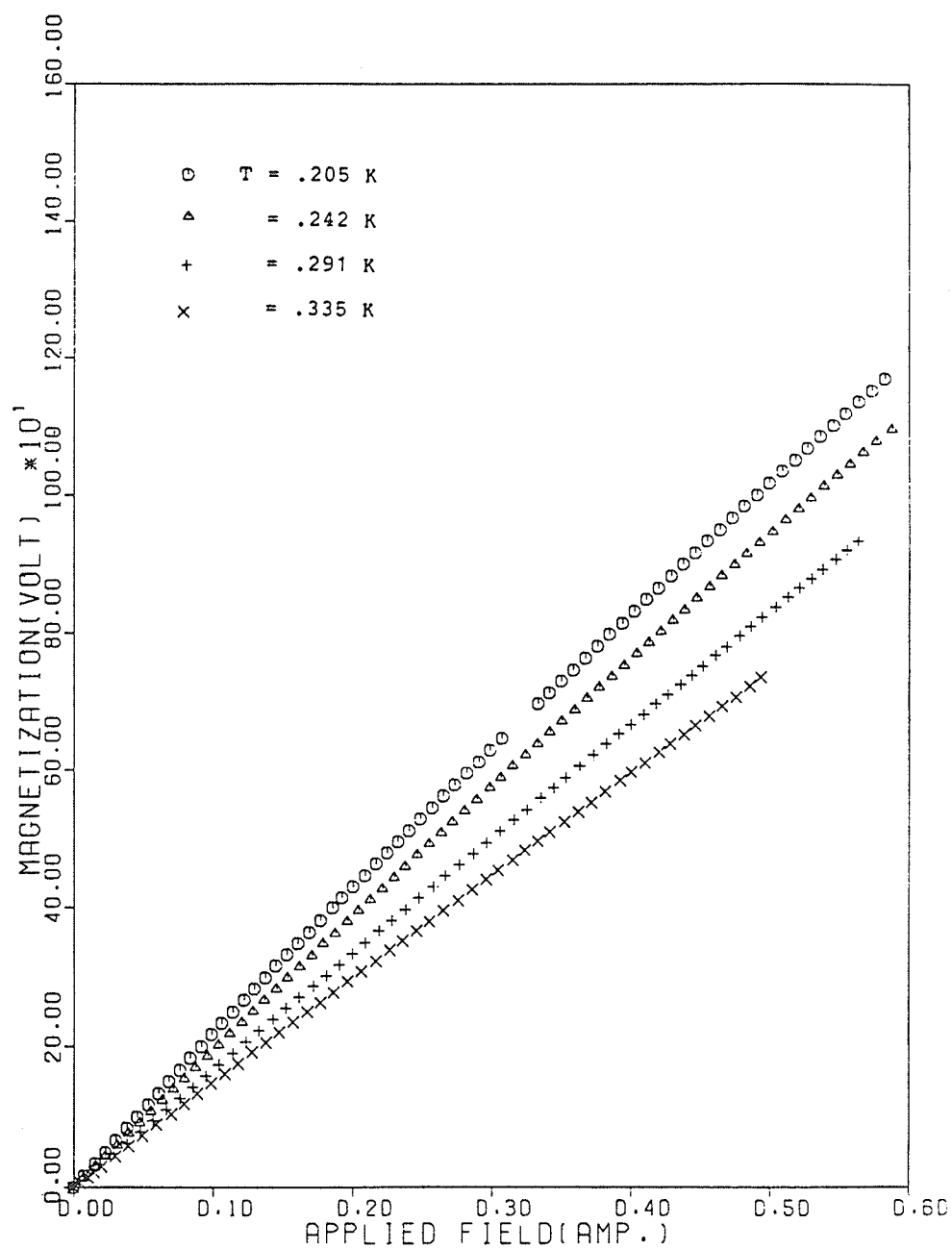


Figure 3.4 Magnetization vs applied field at 4 representative temperatures for the Pt + 2400 ppm Mn sample.

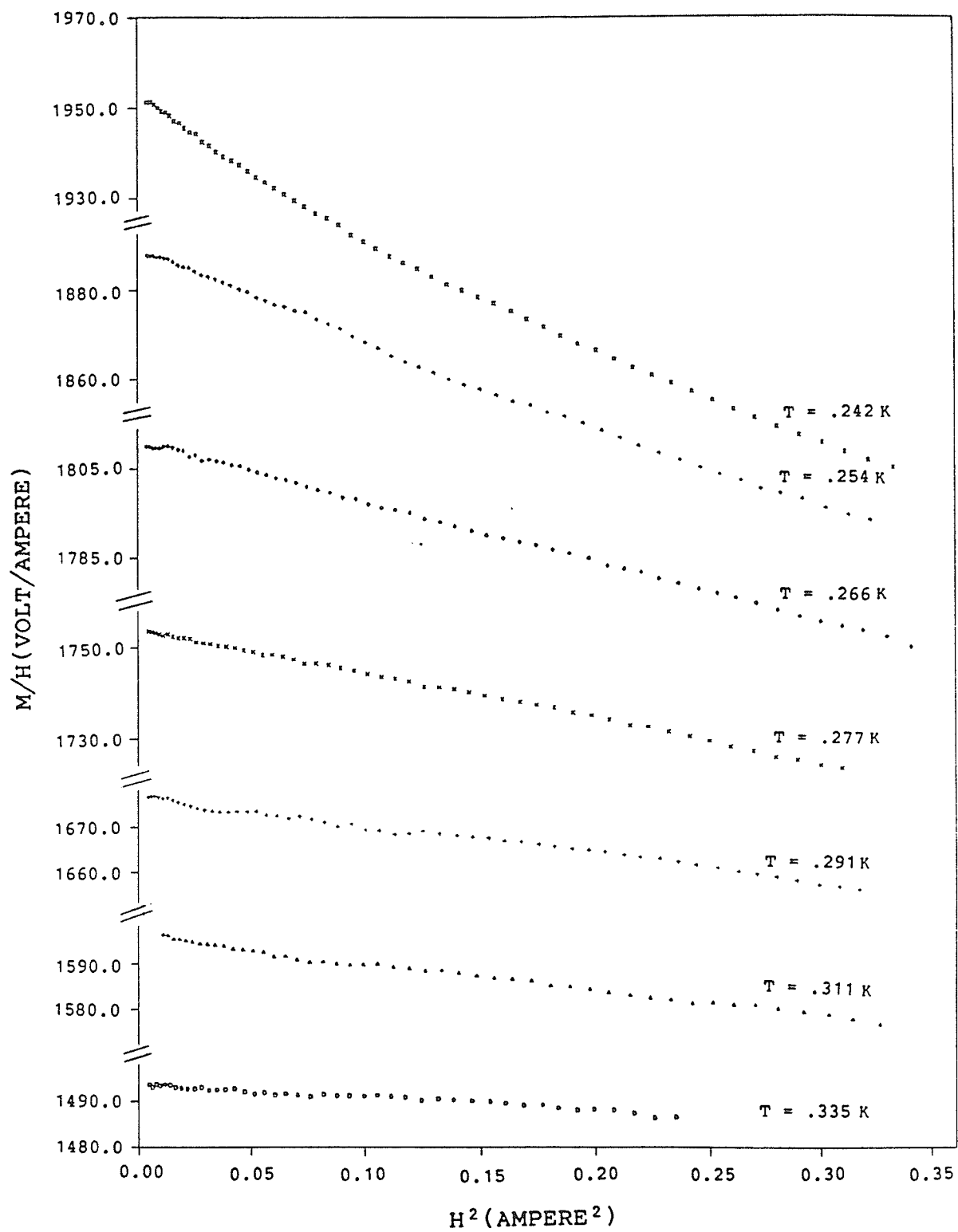


Figure 3.5(a)

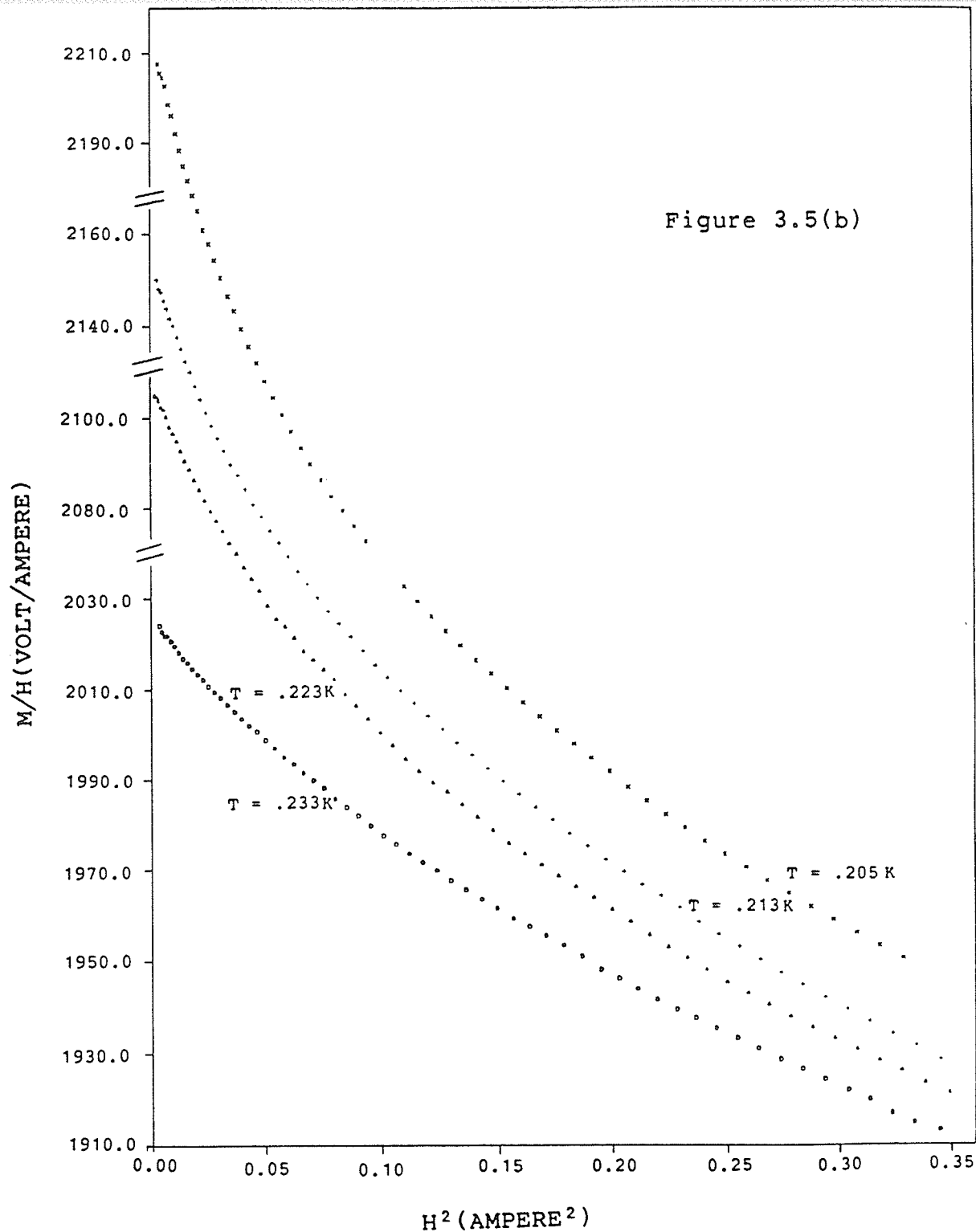


Figure 3.5(a) & (b). M/H vs H^2 at all 11 temperatures for the Pt + 2400 ppm Mn sample. It is shown that the initial slope increases as $T \rightarrow T_0 = 200$ mK.

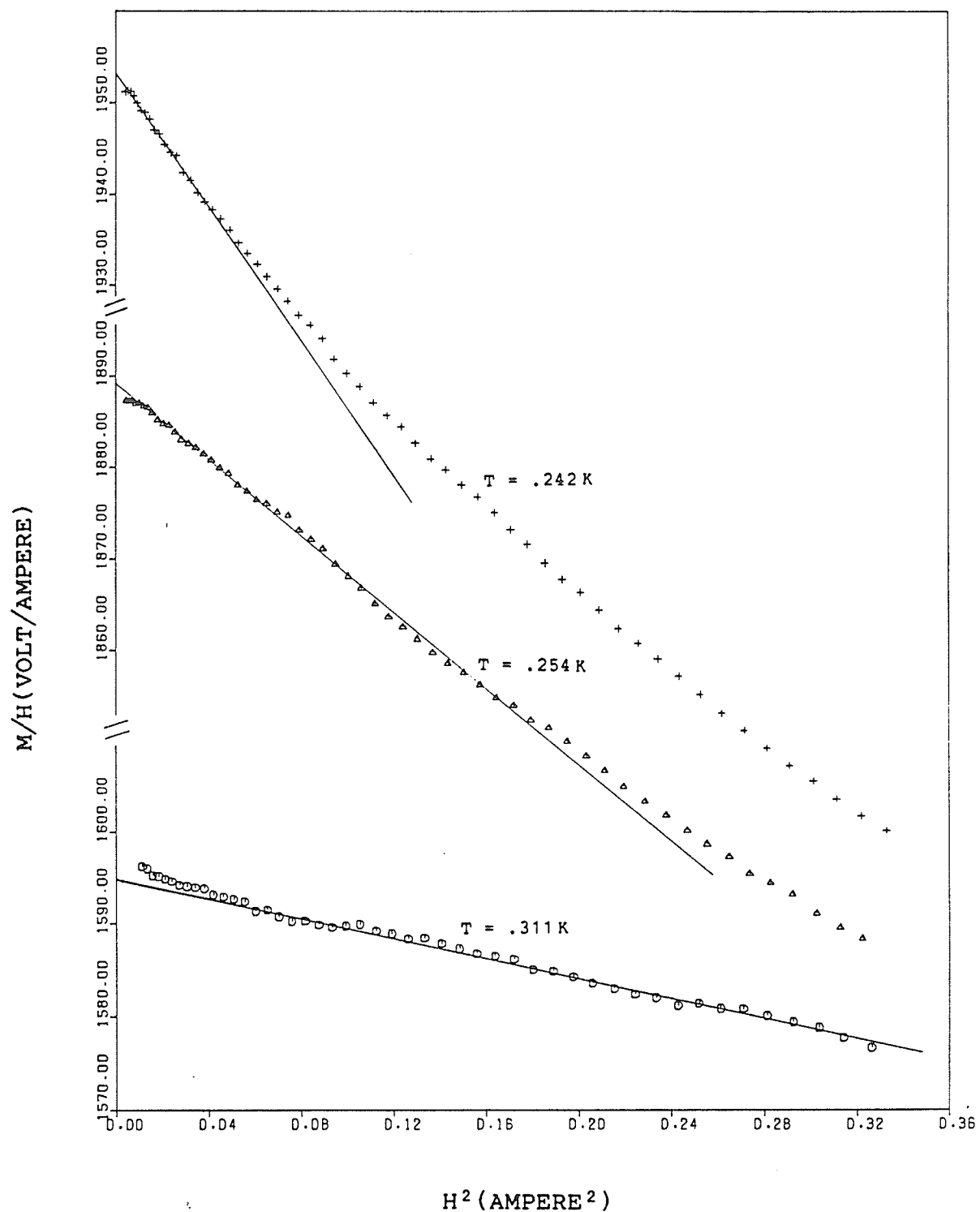


Figure 3.6. M/H vs H^2 plots at 3 representative temperatures for the 2400 ppm sample. The solid lines give the initial slopes of each curve.

Figure 3.7 is a plot of coefficient $b(t)$ against the reduced temperature, t , on a double logarithmic scale. The value of $b(t)$ at high temperatures can be fitted to a good straight line and the slope is the critical exponent γ which describes the divergence in the H^3 in M . However, the graph breaks away from linearity at low temperatures. The breakaway is probably due to the limitations of our measurement rather than the failure of the theory. One possible source is the underestimate of the slope mentioned above. Another possibility is the dynamics of the critical fluctuation: the characteristic time for the fluctuations in magnetization grows prohibitively long as $T \rightarrow T_0$ and exceed the time constant of the experimental measurements. This may mean that certain components of the magnetization may not have time to develop fully during the time constant of the measurement, so that some information is lost.

The vertical error bars on the γ -plot were determined by the slopes of the worst straight lines (which give the steepest and the shallowest slopes within the error bars) in the M/H vs H^2 plots. The horizontal error bars due to the temperature instability were negligible except for the data at the two lowest temperatures. From figure 3.7, the best value of γ was found to be 2.1 ± 0.1 .

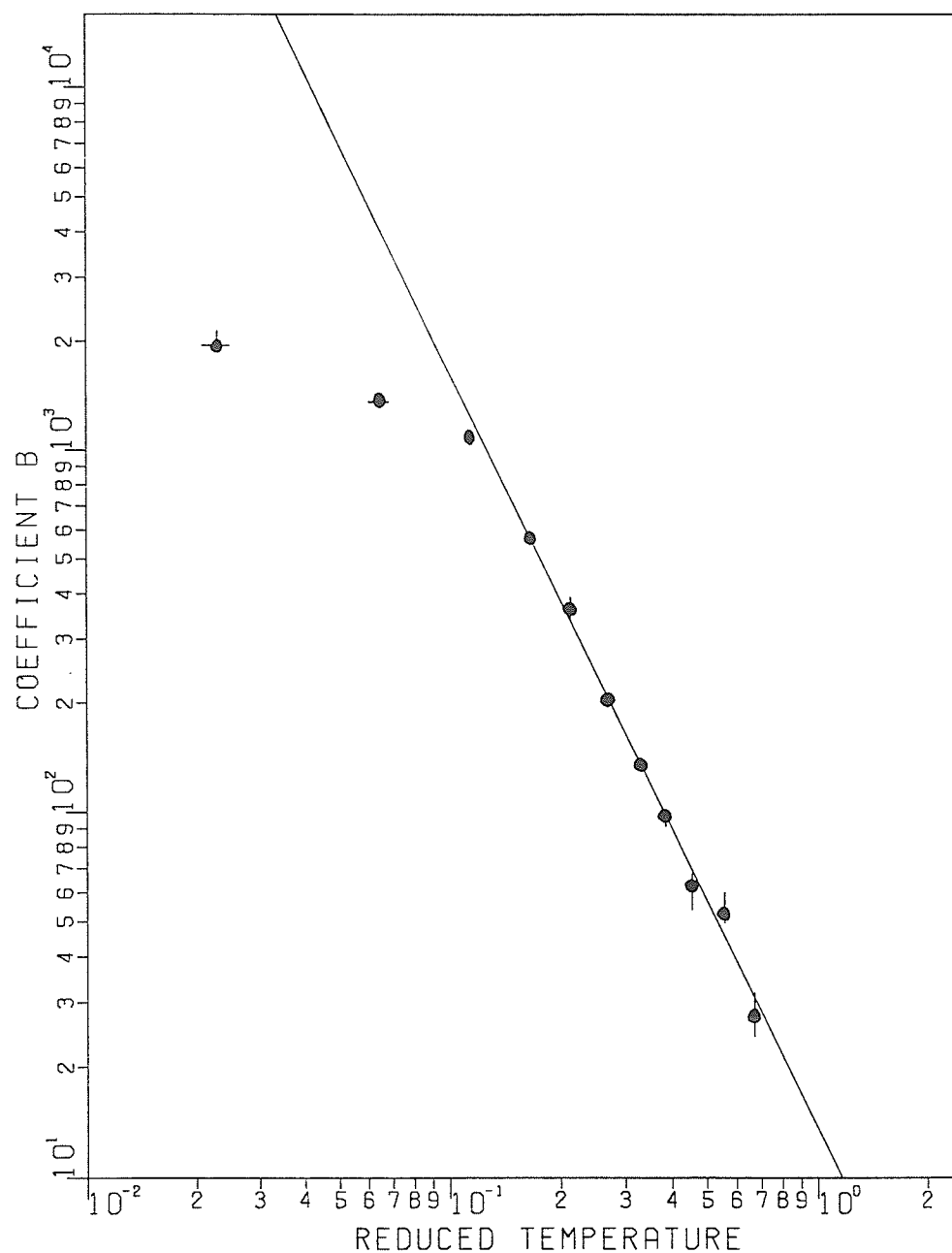


Figure 3.7. Coefficient B plotted against T in log-log scale gives rise a straight line with a slope of γ , the critical index by which the cubic term in the magnetization diverges. The breakaway of linearity for the lowest temperatures is likely due to limitation of the experimental technique.

3.3.3 Scaling Function

As discussed in chapter 1, the field expansion of the magnetization in equation 3.1 suggests that the non-linear magnetization can be represented as a universal scaling function of H and t :

$$M_{NL}(H,t) = M(H,t) - a(t)H = H t^{\beta} F\left(\frac{H^2}{t^{\gamma+\beta}}\right)$$

In the mean-field effective-field model, both γ and β take on a value of 1. Figure 3.8 shows a plot of the magnetization data for the 2400 ppm sample in the form $[M/H - a(t)]t^{-\beta}$ vs $H^2/t^{\gamma+\beta}$ with $\gamma = 2.1$ as determined in section 3.3.2 and with $\beta = 1.0$ predicted by the theory as well as determined in other systems (for example, in AgMn, Bouchiat 1986; in CuMn, Omari et.al. 1983). With the exception of the two lowest temperatures, the data scale very well. The non-universal behaviour for the data at the two lowest temperatures is not surprising since the breakaway from linearity on the γ -plot occurs at these very temperatures. We therefore expect to see the data at these temperatures behave differently from the rest. If we ignore the data from the two lowest temperatures, the scaling function defined by the remaining temperatures is found to be rather insensitive to the change of β . However, we have determined that $\beta = 0.6 \pm 0.1$ produces the best scaling and figure 3.9 is a

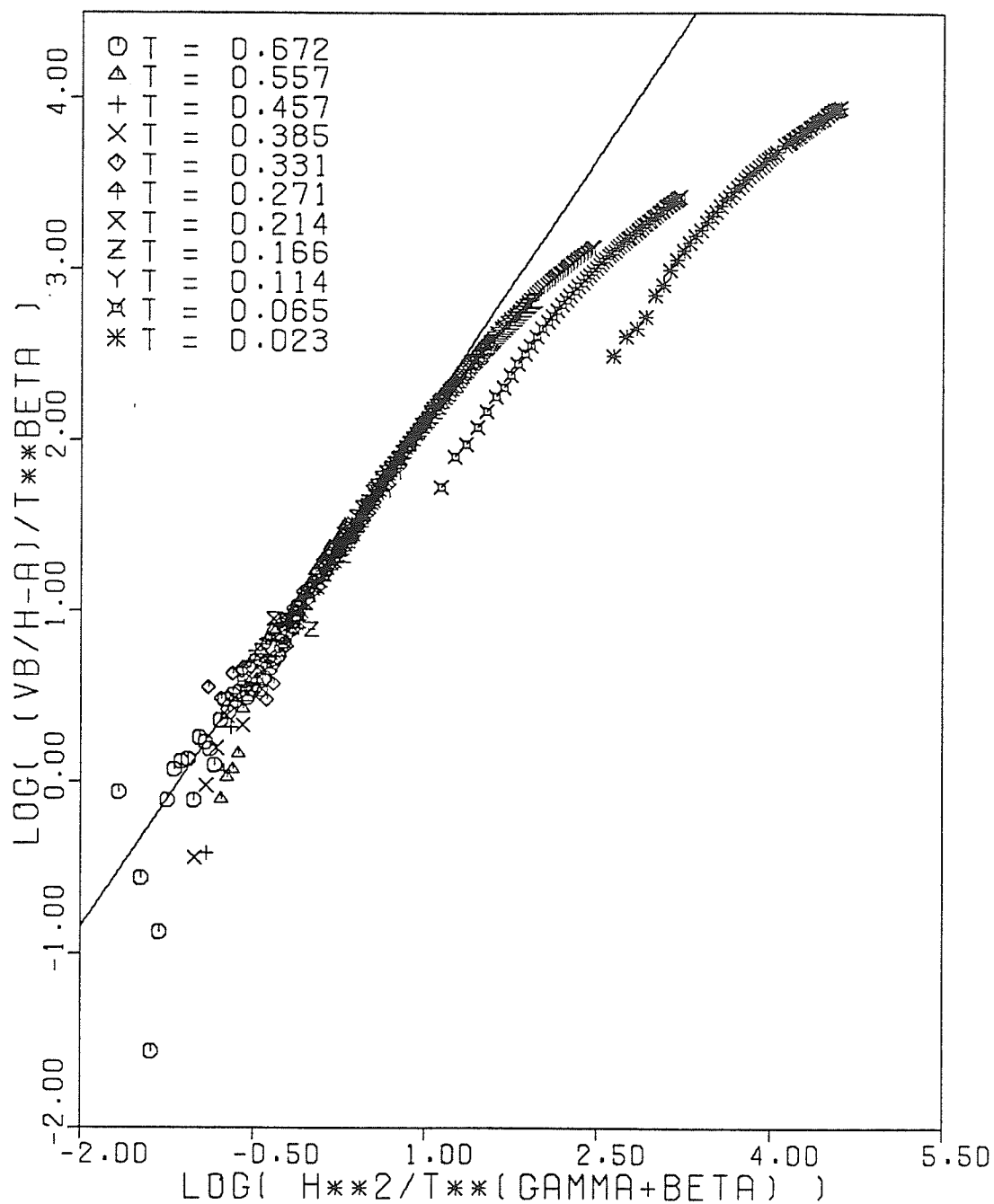


Figure 3.8. Scaling plot of the 2400 ppm sample with $\gamma = 2.1$ and $\beta = 1.0$.
Temperatures shown are reduced temperatures.

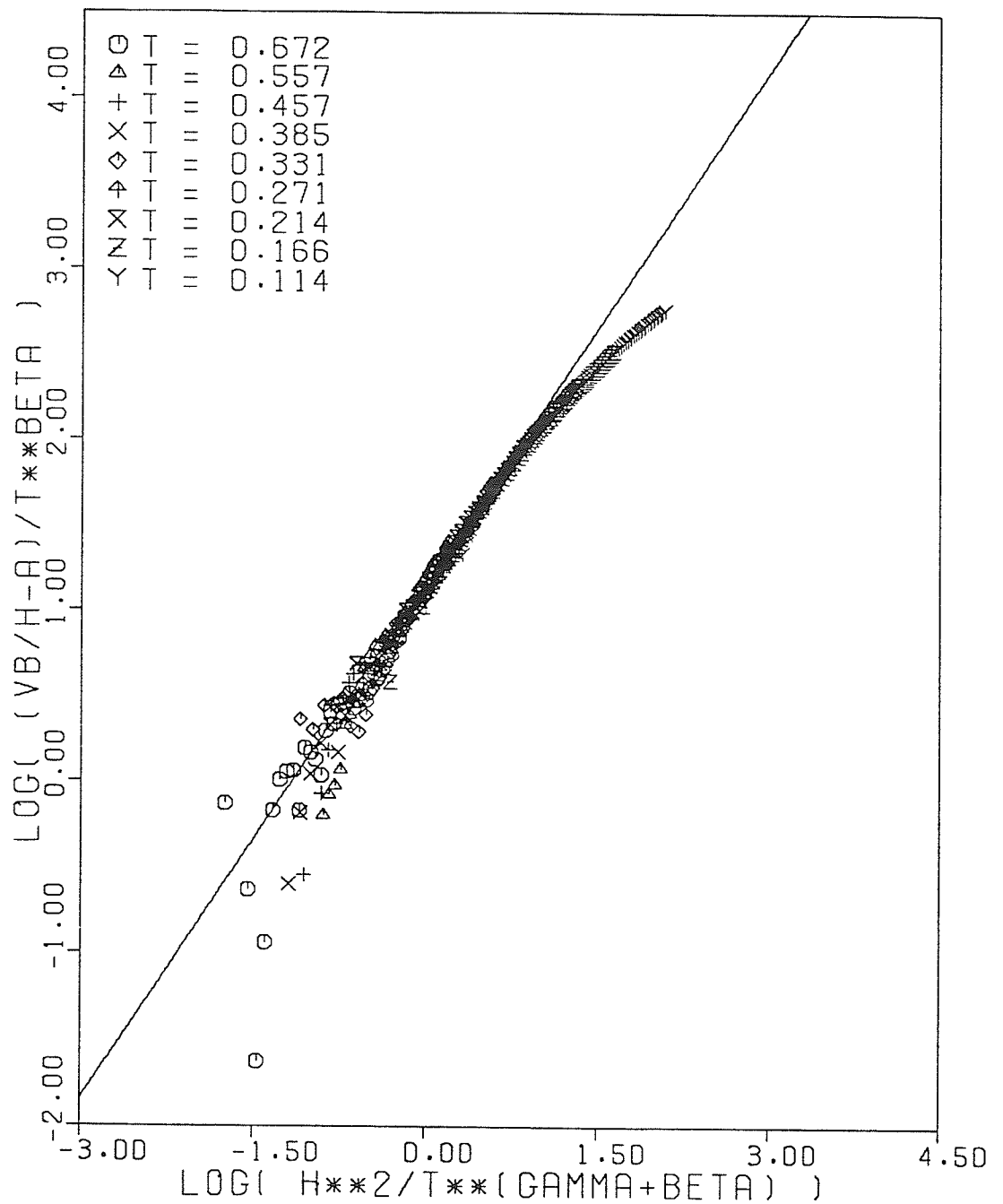


Figure 3.9. Scaling plot for the 2400 ppm sample excluding the data of the lowest 2 temperatures. The values of γ and β are 2.1 and 0.6 respectively and this value of β produces the best scaling for these temperatures.

scaling plot with $\gamma = 2.1$ and $\beta = 0.6$. The straight line in both figures 3.8 and 3.9 corresponds to the predicted asymptotic behaviour $\lim_{x \rightarrow 0} F(x) \propto x$. With perhaps the exception of the lowest temperatures, the data is well represented by the scaling function.

3.3.4 Analysis for the 1100 ppm sample

We have performed an identical analysis on the data for the 1100 ppm sample. Figure 3.10 shows the data for four representative temperatures plotted as magnetization M against applied field H , while figures 3.11(a) and (b) are plots of M/H vs H^2 . Again, we observe that the initial slope of all three plots increases as $T \rightarrow T_0$ from above. That is, both the linear term $a(t)$ (initial slopes in figure 3.10) and the cubic term $b(t)$ (initial slope in figures 3.11(a) and (b)) increase as $T \rightarrow T_0$ from above, as predicted by the mean-field effective-field theory. Furthermore, the theory also predicts a divergence in $b(t)$ of the form $1/t$ as $t \rightarrow 0$ from above. Figure 3.12 is a plot of the coefficients $b(t)$ against the reduced temperature on a double logarithmic scale. With the exception of the two lowest temperatures, the data defines a good straight line which has a slope, $\gamma = 2.31 \pm .10$. The breakaway from linearity for data at the lowest temperatures is probably of the same origin as that for the 2400 ppm sample.

Figure 3.13 shows a scaling plot for all the data from the 1100 ppm sample with $\gamma = 2.31$ determined from the γ -plot and with $\beta = 1.0$ predicted in the mean-field effective-field theory. The plot shows fairly good universal behaviour except at the lowest temperatures. Again, a change in β was found to have only a very subtle influence on the universal behaviour of the high temperature data. The best scaling was found for data at high temperatures with $\beta = 0.8 \pm 0.2$. Figure 3.14 is a scaling plot for the high temperature data with $\gamma = 2.31$ and $\beta = 0.8$.

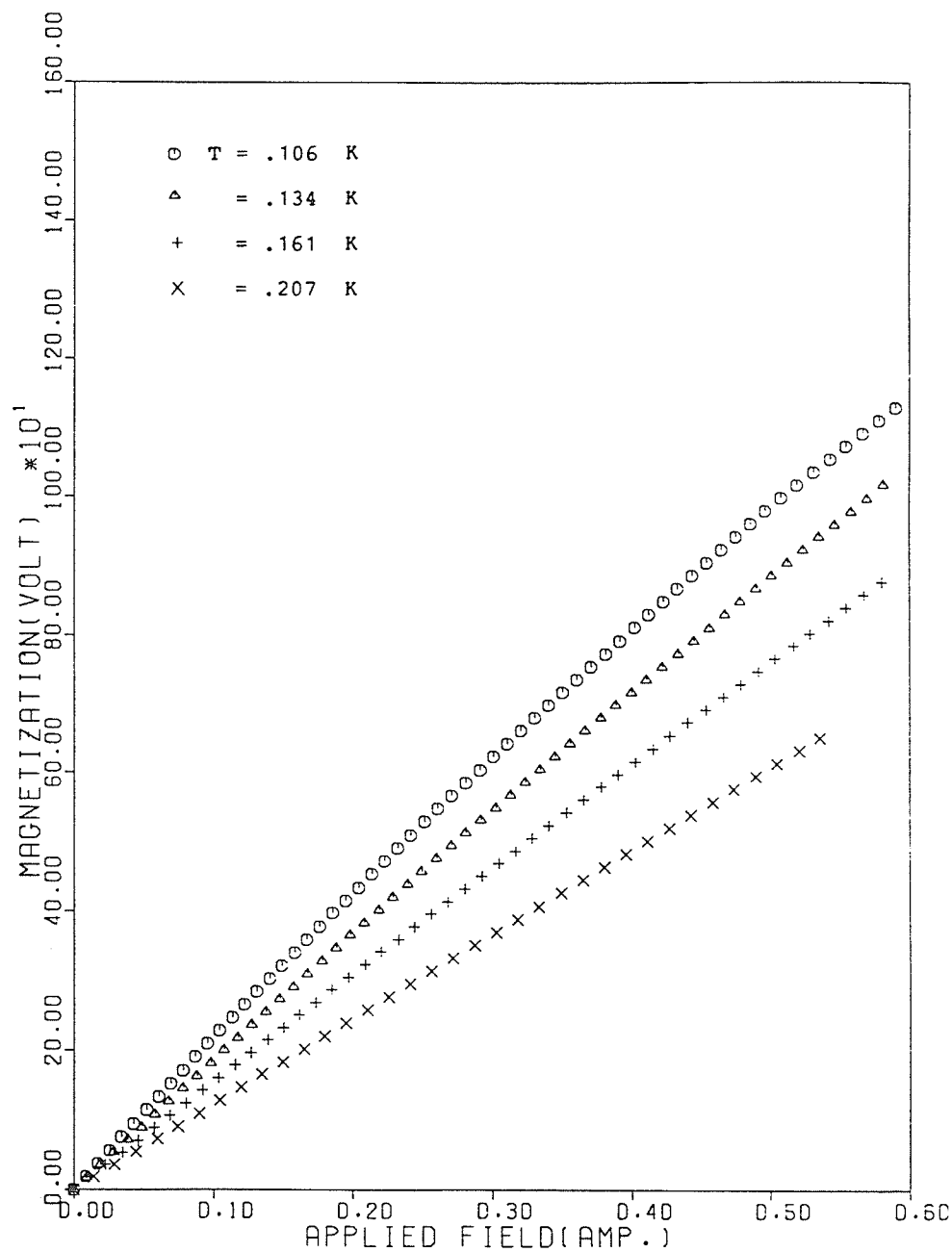


Figure 3.10. Plots of magnetization vs applied field for 4 representative temperatures for the Pt + 1100 ppm Mn sample.

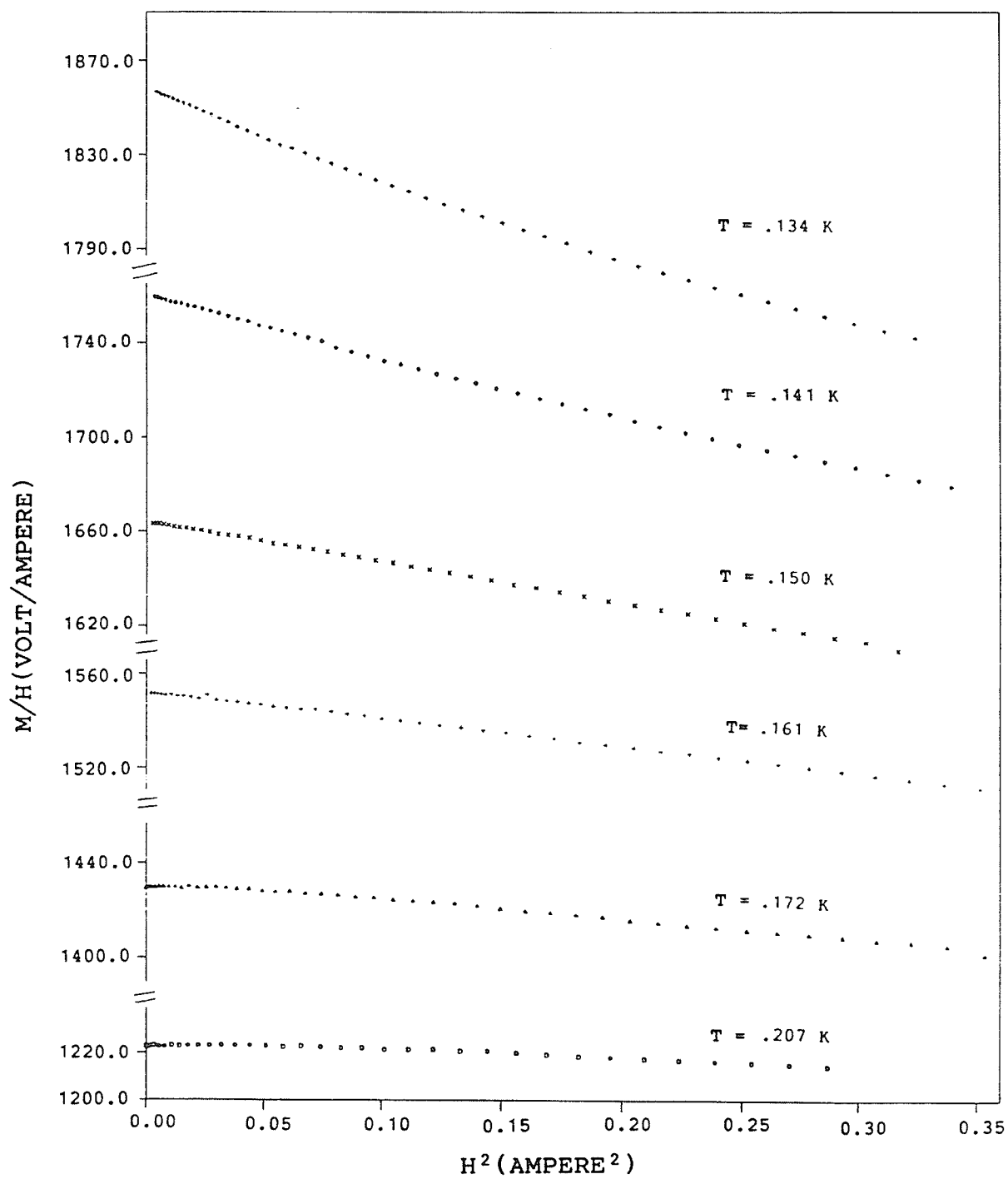


Figure 3.11(a)

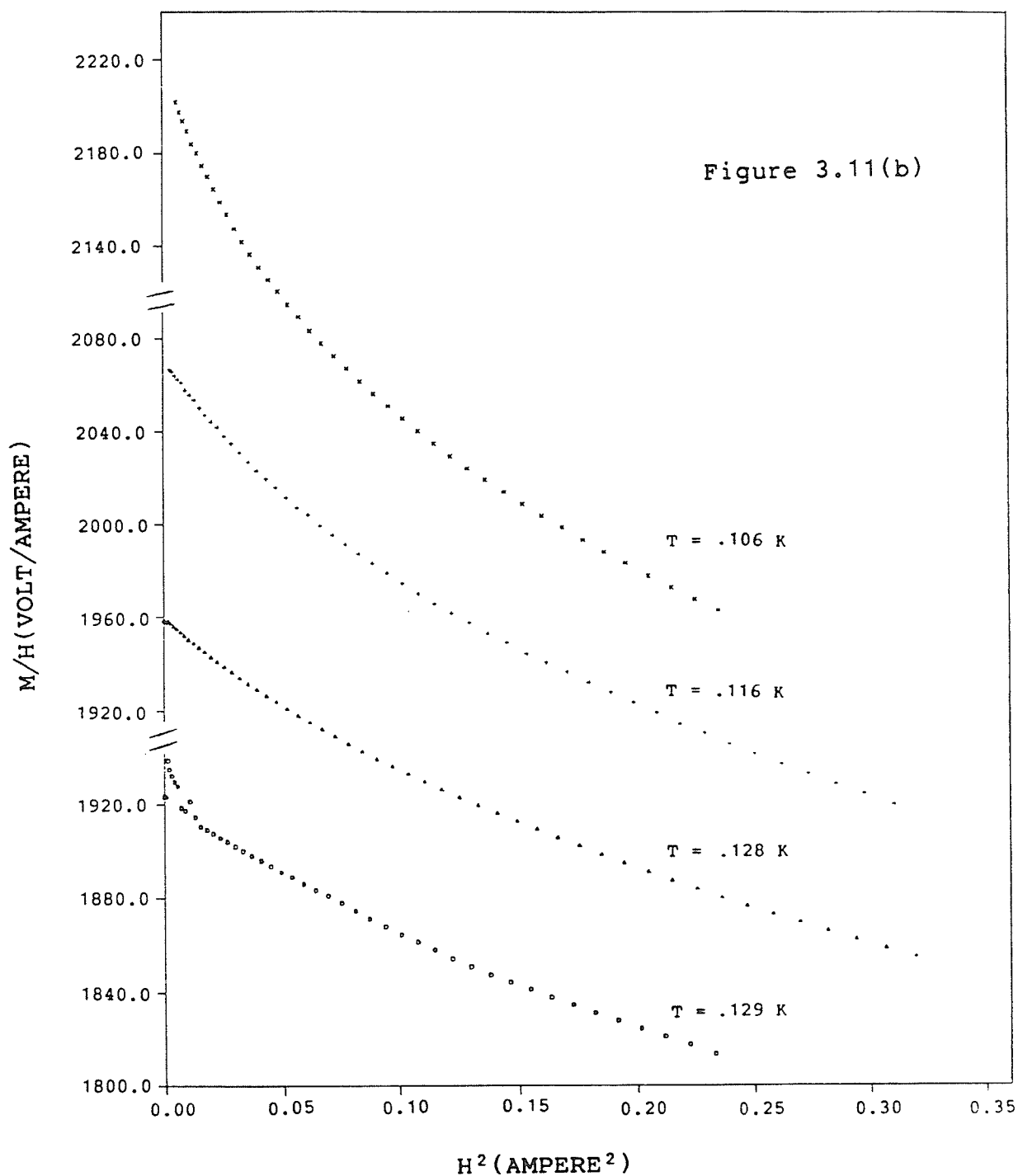


Figure 3.11(a) & (b). Plots of M/H vs H^2 for all 10 temperatures for the Pt + 1100 ppm Mn sample.

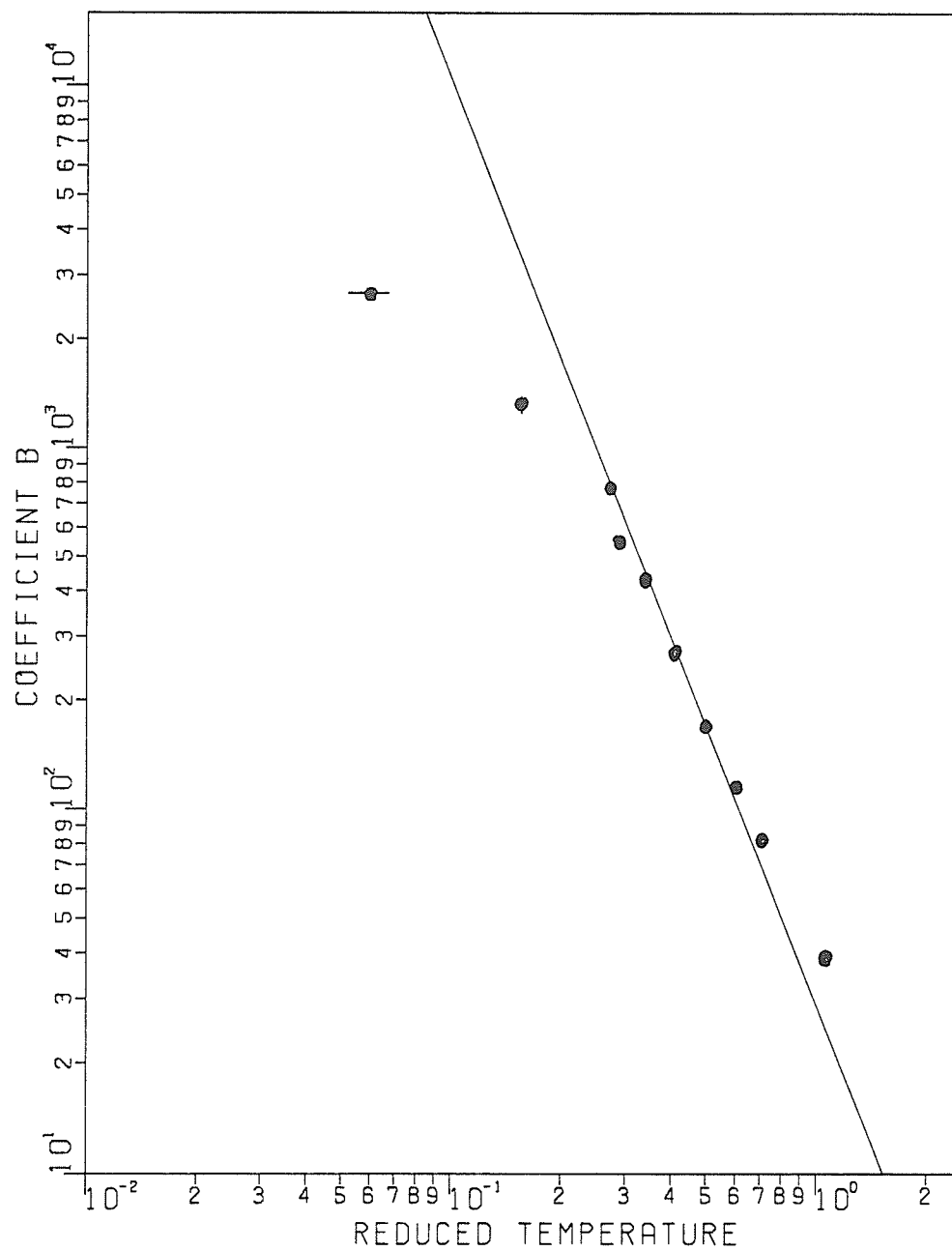


Figure 3.12. The cubic term vs log T for the 1100 ppm sample. The slope of the straight line gives the value of γ .

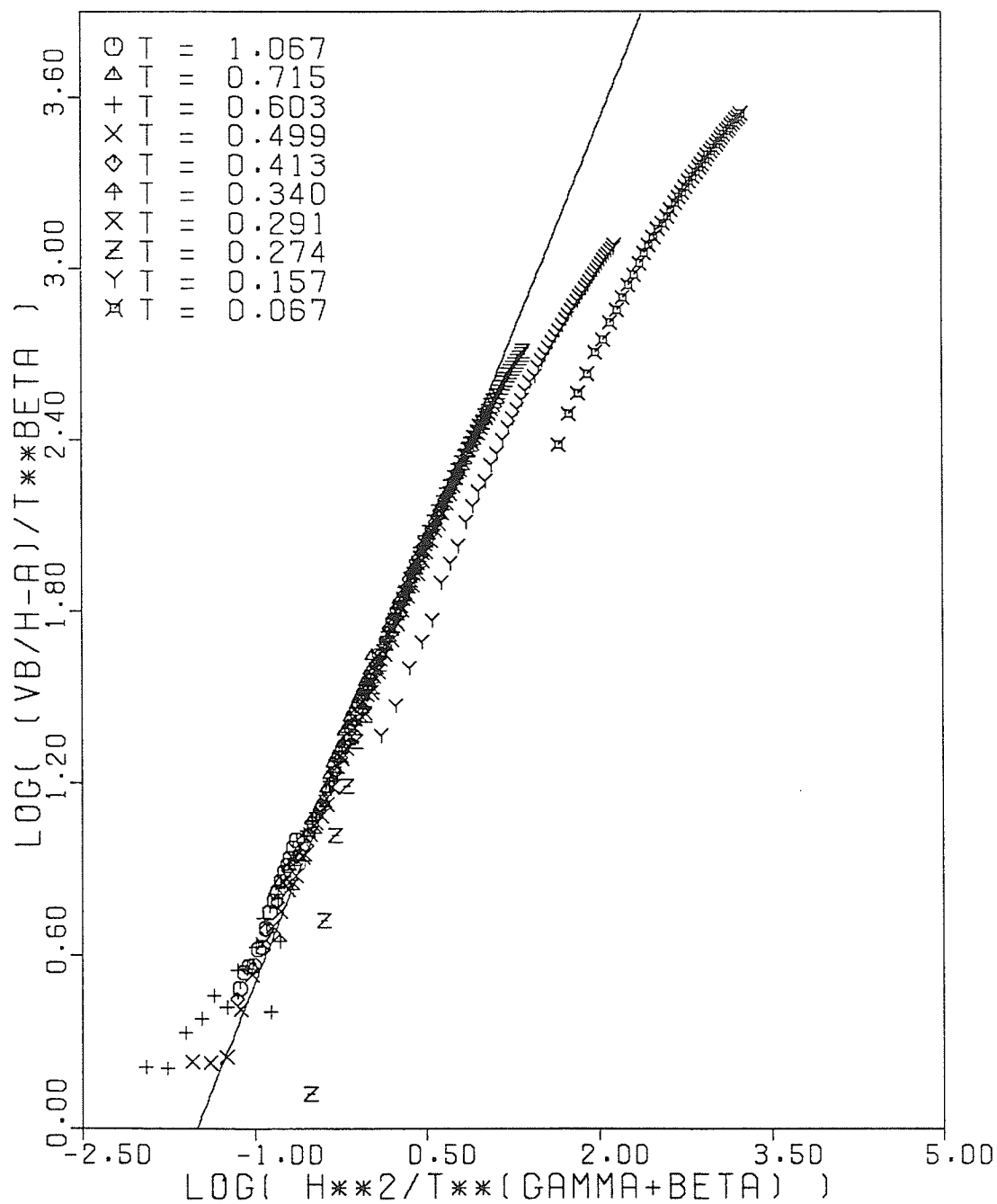


Figure 3.13. Scaling plot for the 1100 ppm sample with $\gamma = 2.31$ and $\beta = 1.0$ which is the mean-field value.

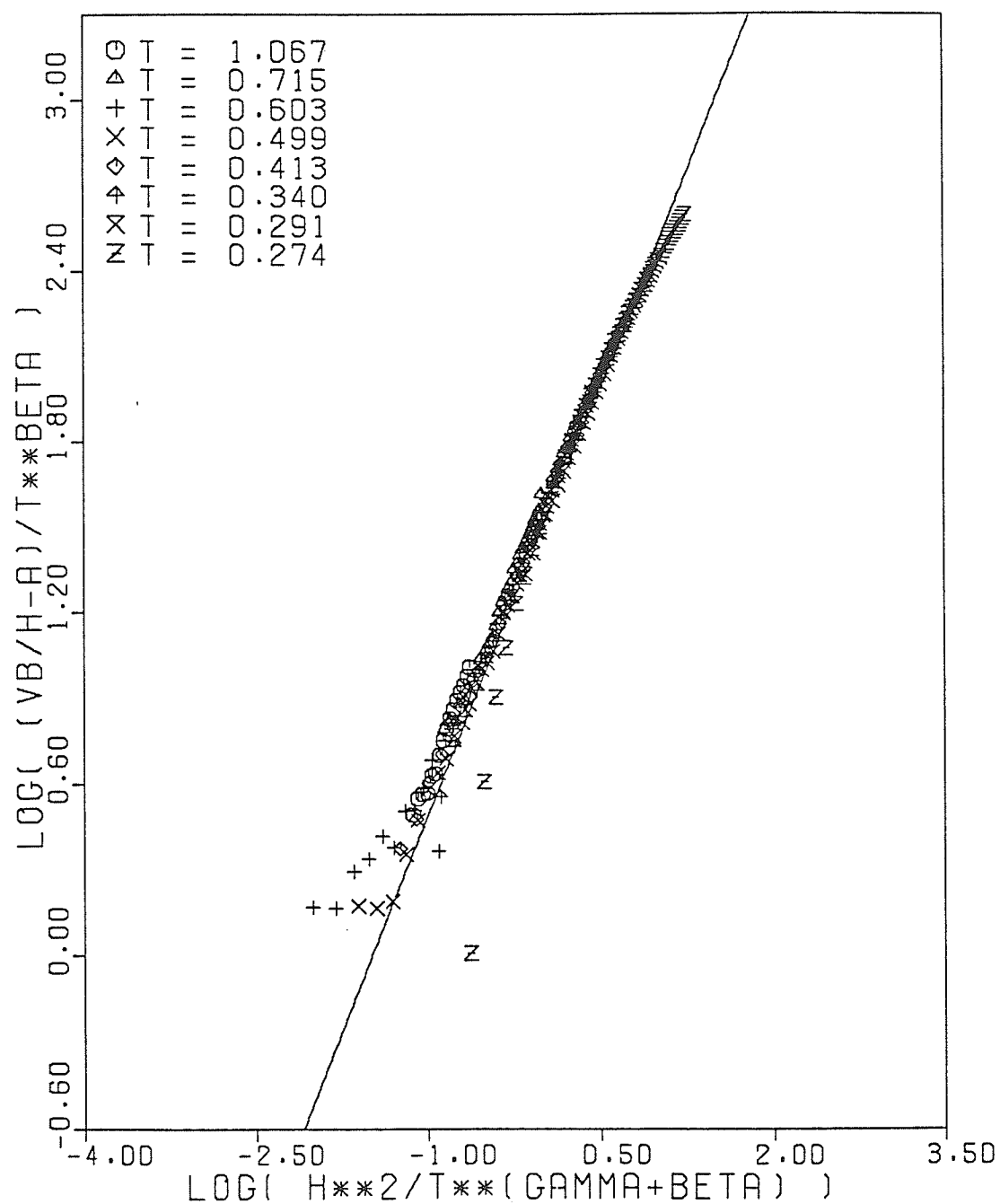


Figure 3.14. Scaling plot for the 1100 ppm sample without the data for the lowest 2 temperatures. $\gamma = 2.31$ and $\beta = 0.8$ which gives the best scaling.

3.4 TIME EFFECTS

We also investigated time effects for the 2400 ppm sample. As mentioned in section 3.3.1, the magnetization exhibited a strong time dependence and these time effects persisted even at temperatures above. To estimate the temperature (above T_0) at which time effects first become significant, we first heated the sample to a temperature $T \gg T_0$, applied a magnetic field of 2 Gauss, and then turned off the field and observed the decay of the remanent magnetization as a function of time. If it was instantaneous, we turned on the field again, lowered the temperature to another value, and repeated the procedure. After a sequence of such measurements we eventually reached a temperature at which the sample first showed noticeable time effects; this temperature was approximately 262 mK ($= 1.31 T_0$), and a reference temperature (T_R) was defined to be 262 mK.

We measured the decay of the thermoremanent magnetization for the 2400 ppm sample as a function of temperature. The sample was cooled in a field of 2G from T_R to a desired measuring temperature T_M . The time to cool from T_R to T_M was called t_w^1 (wait time 1). Once the desired temperature T_M was attained, we waited for an additional period of time t_w^2 (wait time 2), to ensure temperature stability, before the magnetic field was turned off. We then recorded the thermoremanent magnetization (in volts) as a function of time for a period of up to 2 hours (7200 sec). At the end of

this time, we heated the sample back to the reference temperature T_R , so the sample was well into the paramagnetic regime. The magnetization reading in this regime should correspond to that in zero field and hence the absolute value of the magnetization could be obtained by subtracting this zero magnetization reading.

We studied the time effects at seven different temperatures, 3 above T_0 and 4 below. The cooling time t_w^1 from T_R to T_M was clearly a variable depending on T_M , but the temperature equilibrium time t_w^2 was chosen consistently to be 60 minutes.

As mentioned in section 1.3, various forms have been proposed by different authors to describe the dependence of the remanent magnetization of a spin glass. The more common forms suggested are the logarithmic and stretched exponential form and combination of them. To determine the form which fits our data, we have plotted the data in several different ways.

The remanent magnetization was first plotted on a linear scale against time on a logarithmic scale in order to test for a logarithmic time dependence. As shown in figure 3.15, although restricted portions of each curve for different temperatures might be consistent with a straight line, overall the data exhibits definite curvature. Hence, a logarithmic form is not sufficient to completely describe the data at any one of the temperatures.

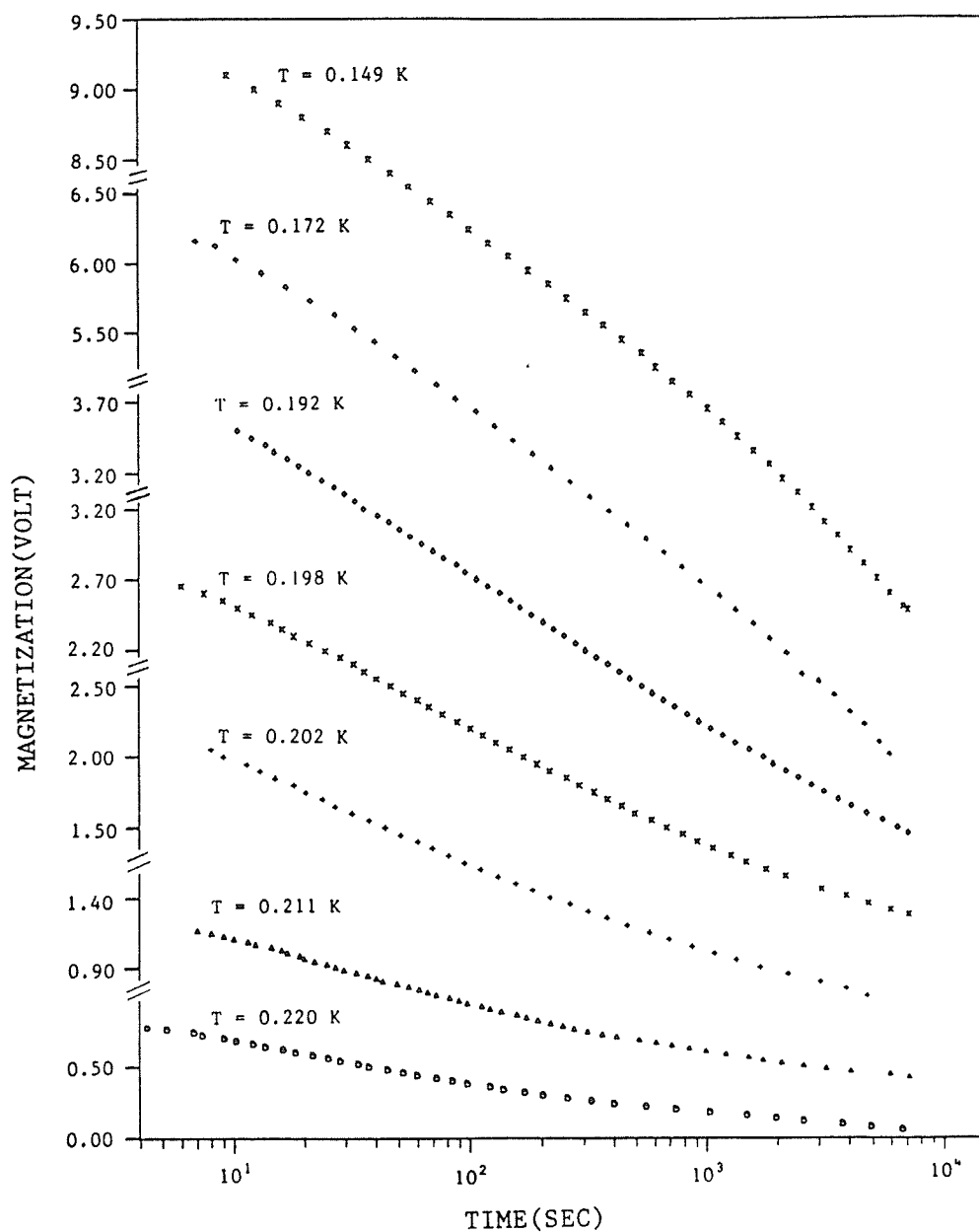


Figure 3.15. The remanent magnetization plotted against $\log t$. A logarithmic time dependence would produce a straight line for each temperature.

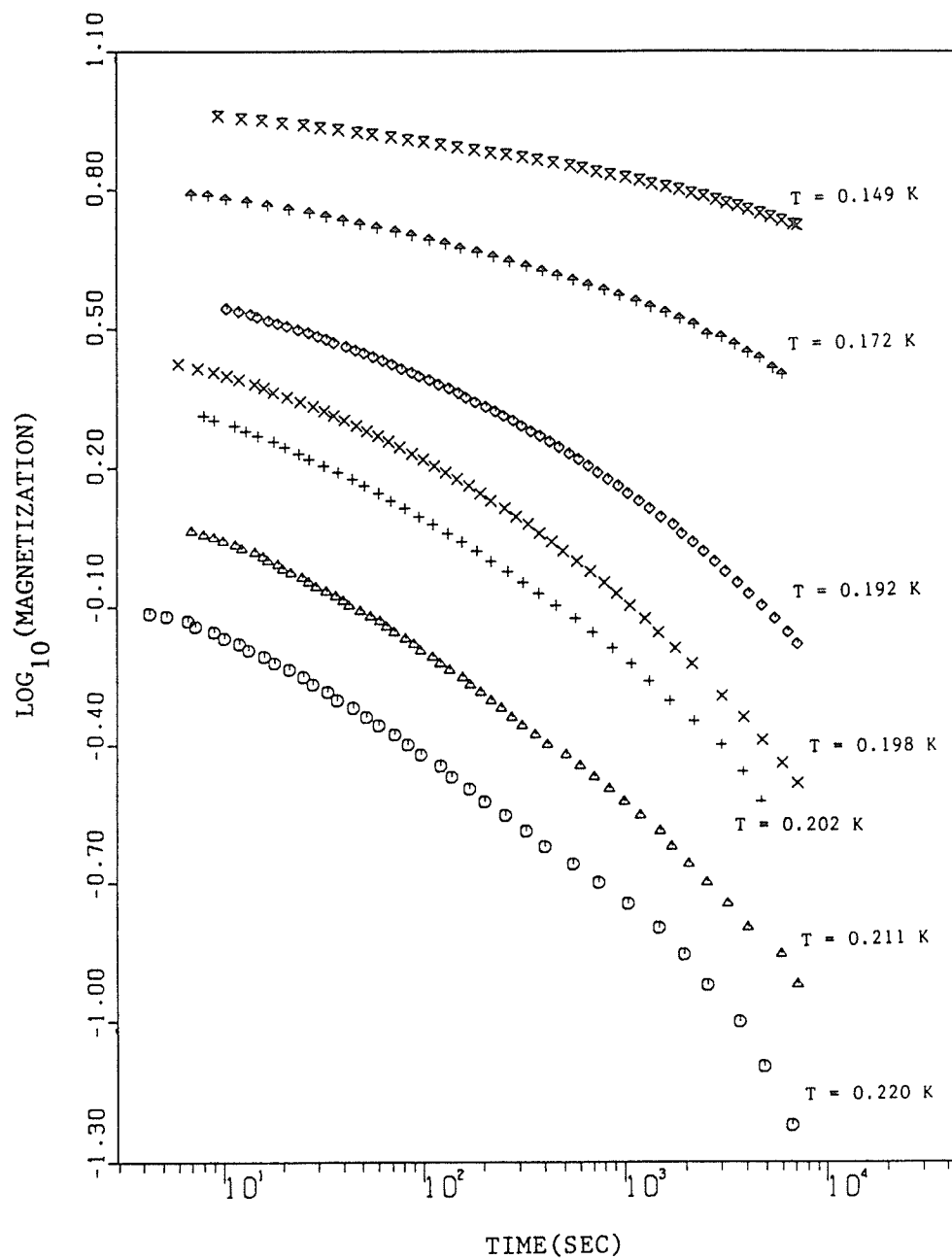


Figure 3.16. The remanent magnetization plotted against t in a log-log scale. A power law decay would give a straight line for every temperature.

Figure 3.16 shows a plot of the remanent magnetization against time on a double logarithmic scale, as a test for a power law dependence. A simple power law decay of the remanent magnetization with time (t^{-n}) would yield a straight line with a slope of $(-n)$. Clearly, the data for all temperatures is convex downward with increasing curvature as the temperature increases. Hence a power law decay is also not appropriate to describe the data.

Another possible form for the decay of the remanent magnetization is a stretched exponential:

$$M_R = M_0 \exp\left[-\left(\frac{t}{t_p}\right)^{1-n}\right]$$

From this expression, it follows that:

$$\log\left[\frac{-d \ln(M_R)}{dt}\right] = -n \log(t) + [\log(1-n) + (n-1) \log(t_p)]$$

so that, a plot of $\log[-d \ln(M_R)/dt]$ vs $\log t$ should yield a straight line with a slope of $-n$ and an 'intercept' of $t = 1$ sec of $(1-n)/t_p^{1-n}$. Figure 3.17 is a plot of the data in this form; the solid lines represent the best-fit straight lines using a least square method. Despite the scattering (which is expected when the derivative is numerically per-

formed), the data are well described by straight lines. Moreover, there does not exist any of the systematic curvature which was observed in both of the previous two plots. As a result, it is concluded that the data can be well described by a stretched exponential form, at least within the time range of the measurements.

From the least square fits performed on the data for different temperatures, t_p and n were evaluated from the intercept and slope respectively and table 3.1 lists their values for all the temperatures. Figures 3.18 and 3.19 are the plots of n and t_p vs T/T_0 on a linear-linear and on a logarithmic-linear scale respectively. As shown in figure 3.18, the n 's for $T > T_0$ have a slightly higher value than those for $T < T_0$, but no definite correlation could be drawn from the limited number of data points with large error bars. Therefore, n is basically constant with a value of approximately 0.82 within experimental error. On the other hand, figure 3.19 shows the drastic change in the value of t as a function of temperature; t_p is very small (< 10 sec) for $T > T_0$ and increases rapidly by orders of magnitude as the temperature falls below T_0 , approximately exponentially.

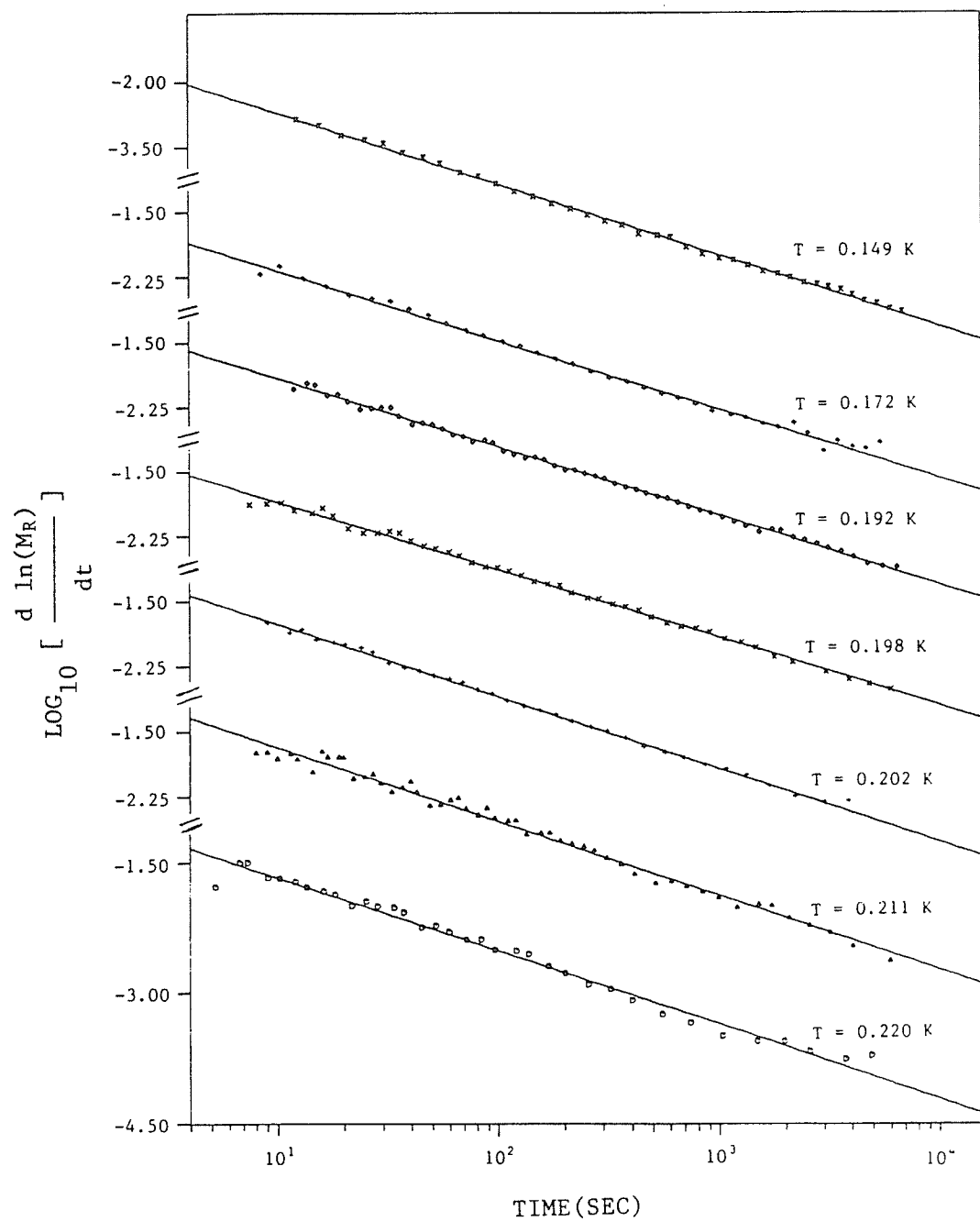


Figure 3.17. $\text{Log} \left[- \frac{d \ln(M_R)}{dt} \right]$ vs $\log t$. A stretched exponential form dependence of time would give rise straight lines. The solid lines are the least square fits of the data.

TEMPERATURE	t_p (sec)	n
220 mK	$1.8 \pm .2$	$.842 \pm .018$
211 mK	$1.2 \pm .2$	$.852 \pm .014$
202 mK	$8.8 \pm .4$	$.833 \pm .006$
198 mK	84 ± 15	$.778 \pm .008$
192 mK	134 ± 19	$.795 \pm .005$
172 mK	2672 ± 1112	$.797 \pm .009$
149 mK	27708 ± 9923	$.816 \pm .006$

Table 3.1. A list of n and t_p , parameters of the stretched exponential form for the remanent magnetization at different temperatures.

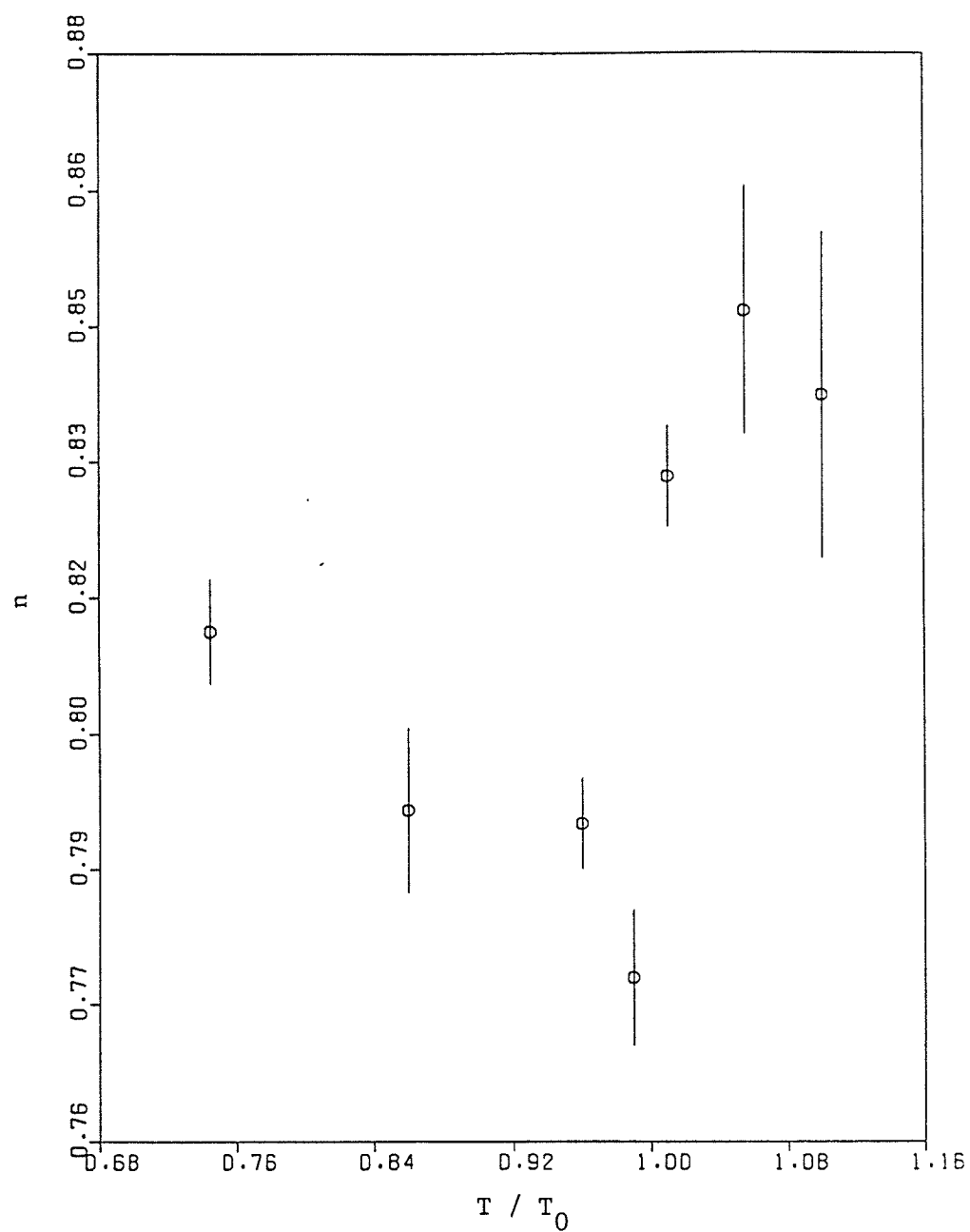


Figure 3.18. A parameter of the stretched exponential form, n , is plotted against T/T_0 and it appears to be constant for all the temperatures measured in the experiment.

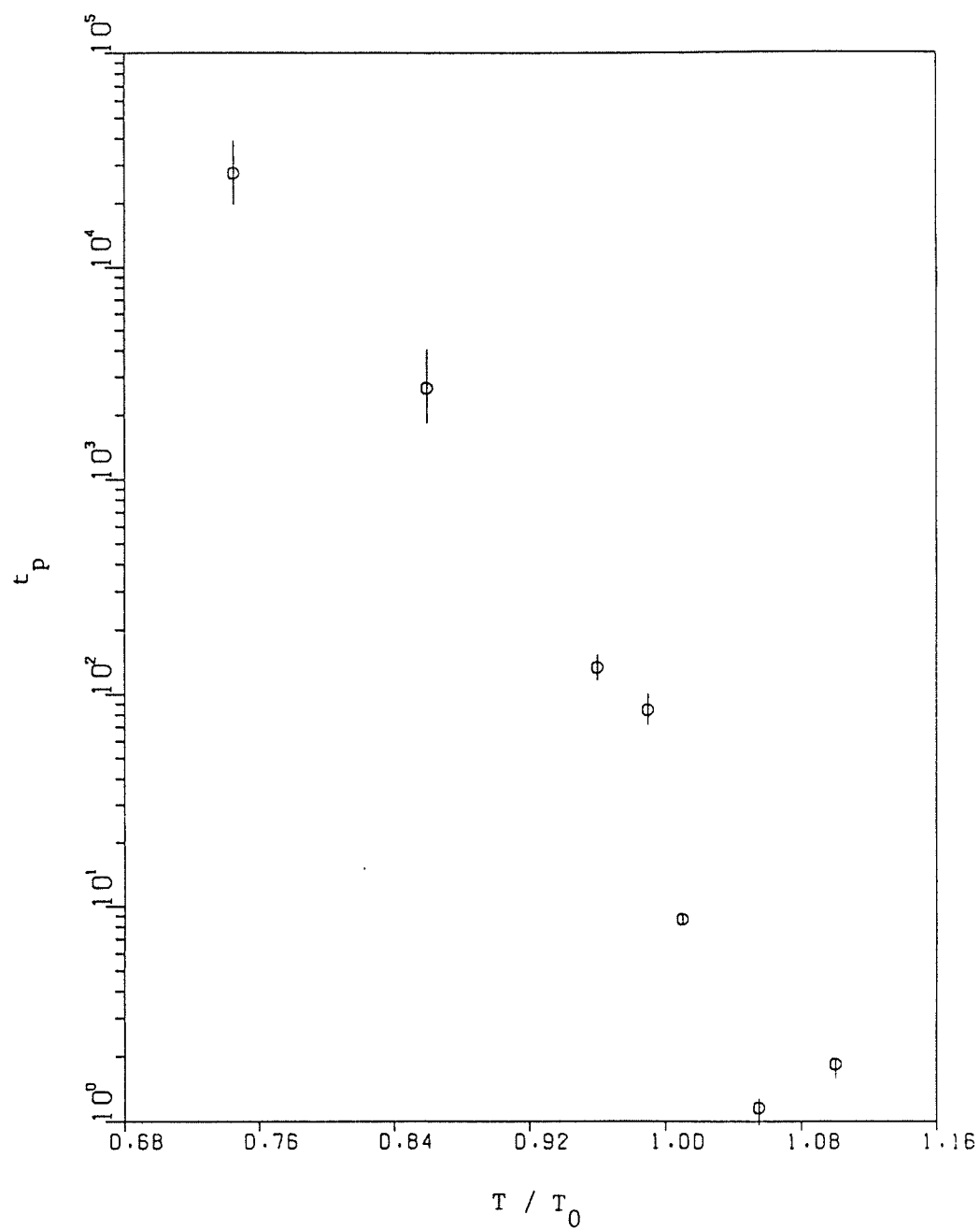


Figure 3.19. Another parameter of the stretched exponential form, t_p is plotted against T/T_0 in logarithmic-linear scale. For $T/T_0 < 1$, t_p increases exponentially as T decreases.

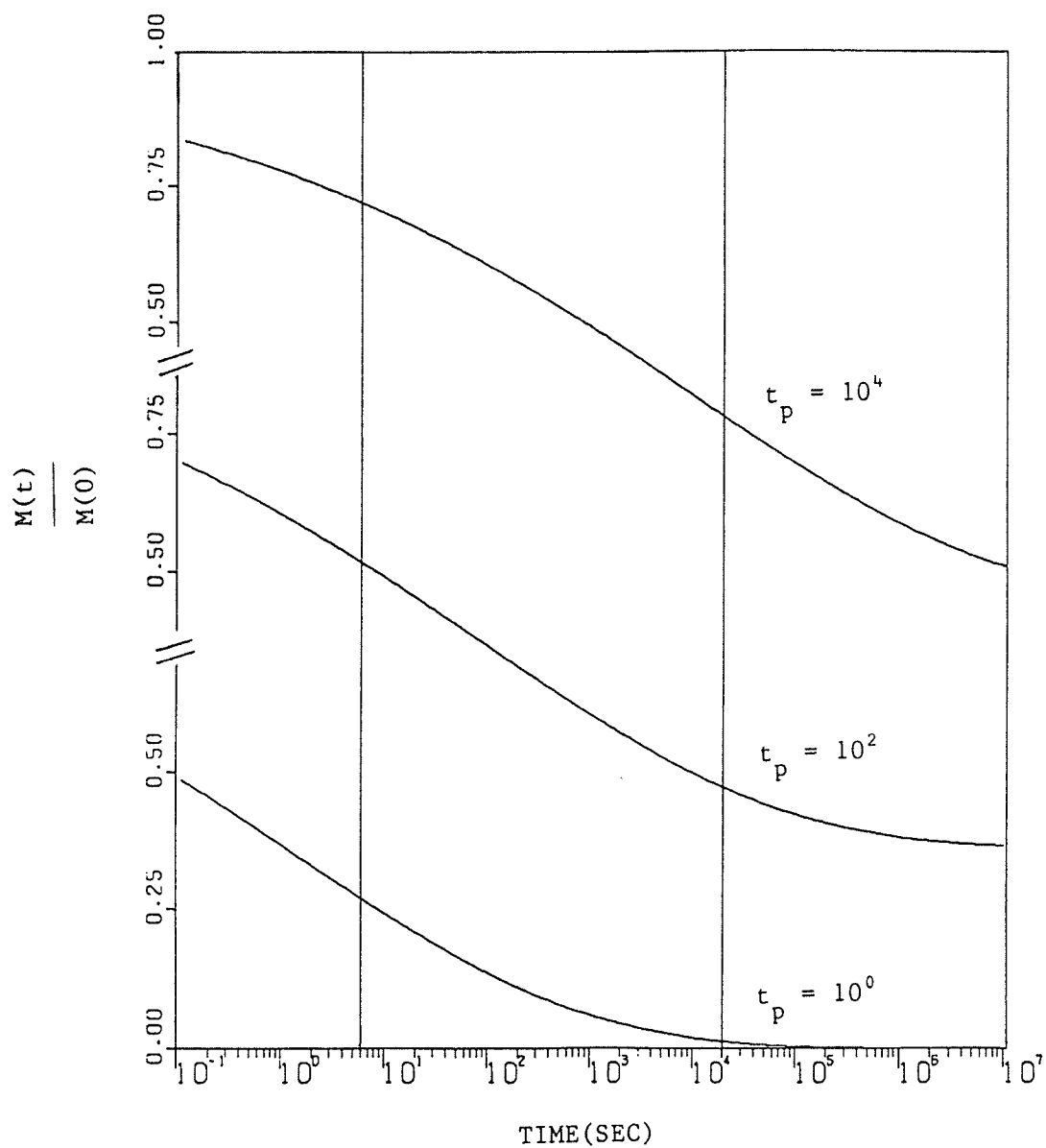


Figure 3.20. Generated data of a stretched exponential form with $n = 0.85$ and $t_p = 1, 10^2$ and 10^4 sec. The time window is the time range in which the remanent magnetization was recorded.

Figure 3.20 shows numerically generated plots of a stretched exponential function with $n = 0.85$ and with three different values of t_p ($t_p = 1, 10^2$, and 10^4 sec). These three values of t_p were chosen to be of the same order of magnitude as those obtained experimentally for $T = 0.211\text{K}$, $T = 0.198\text{K}$ and $T = 0.149\text{K}$ respectively. The time window $4 \leq t \leq 10^4$ sec enclosed by vertical lines represents the time range over which the experiments were carried out. Notice that the value of t_p is closely related to the position of the inflection point in the curves. As a result, the shape of the curves within the time window depends on the value of t_p relative for the window. Hence we may understand the gradual change in curvature from concave down to concave up behaviour in the data in figure 3.15: the curves for different temperatures are characterized by different values of t_p which in turn determines the position of the inflexion point, so that different portions of the stretched exponential curves are captured by the fixed time window used experimentally.

The stretched exponential form of the time dependence can be shown to be physically interpretable. There are two models discussed by Chamberlin(1985) which yield a fractional power of time in the exponent. The first theory "considers a cooperative relaxation of a primary system of dipoles perturbatively coupled to a secondary continuum of low-energy excitation whose density of available levels is linear in

energy" (Chamberlin, 1985). As the dipoles relax, the primary system couples with the secondary continuum which in turn influences the relaxation rate. The second theory is based on a physical picture of a parallel ensemble of independently relaxing constituents. The system is based on a hierarchical constraint that "the degrees of freedom of a relaxing system are ordered into a hierarchy of levels so that level $k+1$ cannot begin to relax until the degrees of freedom in level k have found a combination that releases them" (Chamberlin, 1985)

3.5 CONCLUSION

The two very dilute spin glass samples of Pt + 2400 ppm Mn and Pt + 1100 ppm Mn were found to exhibit critical behaviour as $T \rightarrow T_0$ from above. The cubic term of the magnetization, in the expansion in small H , shows a 'divergence' as $T \rightarrow T_0$ from above predicted in the mean-field effective-field theory. Such behaviour is an indication of a phase transition at T_0 , as determined by the peak in the zero field cooled magnetization as a function of temperature. The coefficient of the cubic term clearly demonstrates the divergence as shown by the γ -plots. The flattening of the γ plots at low temperature is likely due to limitation of the experimental technique rather than a failure of the theory. The critical index γ was determined from the γ -plots to be 2.10 ± 0.1 and 2.31 ± 0.1 for the 2400 ppm and 1100 ppm sam-

ples, respectively, and these values are similar to those in other systems investigated by other authors. The other critical index β was estimated to be 0.6 ± 0.1 and 0.8 ± 0.2 from the best fit to a universal scaling function. The well behaved universal scaling function is also another evidence for a phase transition. However, the determination of a phase transition is obscured by the time effects above and below T_0 , which probably influence the position of the peak in the magnetization as a function of temperature.

It has been discovered that time effects occur not only below T_0 but also above T_0 ; nevertheless, the magnetization decay quickly becomes insignificant as the temperature increases in the paramagnetic regime. Both the logarithmic and power law forms were found to be insufficient to describe the time effect data for the 2400 ppm sample. Conversely, a stretched exponential form represents the data well at least within the time window of the experiments. A parameter of the stretched exponential form, n , has a value of ~ 0.82 for all temperatures while another parameter, t_p , increases exponentially as the temperature decreases below T_0 .

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