INDUCED MAGNETISM VIA CONDUCTION ELECTRON EXCHANGE: APPLICATION TO $PrAl_2$

by

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ABSTRACT

The magnetization, susceptibility and magnetic specific heat of $PrAl_2$ versus temperature are studied within a model where the onset of magnetism is due to mutual magnetization of the rare-earth ions and the conduction electrons, the ionic magnetization process being van Vleck-like. The crystal field, bandwith and exchange parameters of the model which fit the available experimental data are 2.5 meV, 7000 meV and 341 meV respectively.

On étudie la magnetization, la susceptibilité et la chaleur specifique magnetique de $PrAl_2$ versus temperature dans un modèlle où l'ordre magnetique est due à par la magnetization mutuelle des ions et des electrons de conduction, la magnetization ionique étant du type van Vleck. Le champ cristallin, la larguer de bande et le paramètre d'échange du modèlle en accord avec les données experimentaux sont 2.5 meV, 7000 meV et 341 meV respectivement.

INTRODUCTION

In a previous paper [1], from now on to be referred as I, we have studied the magnetic behaviour of a model consis - ting of van Vleck's ions coexisting with a paramagnetic electron gas. Each ion is under the action of a crystal field which splits the ground state into two non degenerated levels and interacts, via exchange with the conduction electrons. It was shown that the model may exhibit ferromagnetism in the band and in the ionic counterparts. The magnetizations and susceptibilities versus temperature were obtained as a function of the band width, crystal field and exchange parameters.

In this paper we propose that the model discussed in I can be applied to the magnetism of $PrAl_2$. In fact, the crystal field level structure of Pr^{+++} in $PrAl_2$ may be well approximated by a two non-degenerated levels [2]; also the metallic nature of $PrAl_2$ suggests, similarly to what occurs in the heavy rare-earth intermetallics 3 , that the conduction band should play a significant role in the onset of the magnetic order. Of interest in this work is the temperature dependence of the magnetization, magnetic susceptibility and the magnetic specific heat of the $PrAl_2$, which are experimentally studied in the literature 4,5,6 . Using a same set of model parameters, a good fitting is obtained in a temperature range which includes the ferro and the paramagnetic regions.

The paper is organized as follows: in section 2 the integral equations relating the magnetizations and the chemical potential as a function of the temperature, obtained in I, are, taking into account the Curie temperature and bandwidth of the $PrAl_2$, approximated by transcendental equations which are numerically solved. In section 3, an expression for the magnetic specific heat as a function of the temperature is obtained. Finally, in section 4, the fitting between experimental and theoretical results is discussed.

2. Ionic and band magnetization and susceptibilities for $kT_c/\varepsilon_o \ll 1$.

Making use of the approximation

$$F(\eta) = \frac{2}{3} \eta^{3/2} (1 + 1.23\, \eta^{-2}) \tag{1}$$

which applies for $\eta \gg |7|$, equations 7,8 and 11 of I lead to

$$\frac{\varepsilon_o}{J} \frac{\left[\zeta^2 + 4\left(\frac{\Delta}{J}\right)^2\right]^{1/2}}{\zeta} \left[(1+\zeta)^{3/2} - (1-\zeta)^{3/2}\right] = \tanh \frac{J}{4kT} \left[\zeta^2 + 4\left(\frac{\Delta}{J}\right)^2\right]^{1/2} \tag{2}$$

$$\langle S^z \rangle = \frac{\zeta}{\left[\zeta^2 + 4\left(\frac{\Delta}{J}\right)^2\right]^{1/2}} \tanh \frac{J}{4kT} \left[\zeta^2 + 4\left(\frac{\Delta}{J}\right)^2\right]^{1/2} \tag{3}$$

In obtaining (2) and (3) only the first term of (1) was used what is quite reasonable in the case of $PrAl_2$, since taking ε_o = 7000 MeV and T_c = 34K and using equation 15 of I and (1) one obtains $\eta_c \simeq 2389$.

Adopting a similar procedure we obtain, from equations (17) and (18) of I, the ionic and electronic susceptibilities

$$\chi_i = 2 \frac{\mu_B^2 (g_i \alpha)^2}{\varepsilon_o} N_o C \left[\frac{\frac{3}{4}\frac{J}{\Delta} + 4 g_i \alpha \frac{\varepsilon_o}{\Delta} \tanh \frac{\Delta}{2kT}}{1 - \frac{3}{8}\frac{J^2}{\Delta \varepsilon_o} \tanh \frac{\Delta}{2kT}}\right] \tag{4}$$

$$\chi_\ell = \frac{\mu_B^2}{\varepsilon_o} N_o \left[\frac{\frac{3}{2}(1 + 2g_e \alpha \frac{J}{\Delta} \tanh \frac{\Delta}{2kT})}{1 - \frac{3}{2}(\frac{1}{4} \frac{J^2}{\Delta \varepsilon_o} \tanh \frac{\Delta}{2kT})} \right] \tag{5}$$

where N_o is the Avogrado number and $C(\simeq 0.72)$ is the Pr molar fraction in the $PrAl_2$ which contributes for the ionic susceptibility; the other symbols are the same as I.

3. Magnetic specific heat

In I the Helmboltz free energy of the ionic subsystem was obtained. From this quantity we can compute the ionic magnetic specific heat

$$C_H = - T \left(\frac{\partial^2 F}{\partial T^2}\right)_h = RC \frac{J^2 \zeta^2 + 4\Delta^2}{16kT^2} \operatorname{sech}^2 \frac{J}{4kT} \left[\zeta^2 + 4\left(\frac{\Delta}{J}\right)^2\right]^{1/2} \tag{6}$$

where R is an ajustable parameter defined as $C_H^{exp} = R\, C_H^{th}$.

The electronic specific heat is much lower than the ionic one and does not contribute to the total magnetic specific heat.

4. Numerical results and comparison with experimental data.

Taking the experimental data from the magnetization [3], susceptibility [4] and magnetic specific heat [5] versus temperature, we try to obtain a same set of parameters (Δ, ε_o, J) which allows a fitting between the experimental and theoretical results both in the ferro and paramagnetic temperature range. This is done in the subsections 4-1 to 4-3.

4.1 - Magnetization versus temperature.

Using the equations 2 and 3 and the relation (equation 16 of I)

$$\frac{M}{\mu_B} = \zeta + 2g_i\alpha\langle S^z\rangle \qquad (7)$$

a theoretical curve of the magnetization is obtained, which is compared to the experimental one in figure 1. The fitting is obtained using J = 341 meV, Δ = 2,5 meV, ε_o = 7000 meV and $g_i\alpha$ = 3.2.

4.2 - Susceptibility versus temperature

Using equations 4 and 5 and same values of J, Δ and ε_o of subsection 4.1, but now taking $g_i\alpha$ = 1.25 the theoretical curve of the susceptibility, shown in figure 2 is obtained and fitted with the experimental data. The use of different values of $g_i\alpha$ in the ferro and paramagnetic phases is attributed mainly to the variations of g_i. Actually the factor g_i of Pr^{+++} in intermetallic compounds depends on the temperature and also on impurities concentration of the sample |8|. The experimental data of ferromagnetic (fig. 1) and paramagnetic phase (fig. 2) are from different authors.

4.3 - Specific magnetic heat versus temperature

Using equation 6 and taking for Δ and J the values of subsection 4.1 and 4.2, with R = 5,4 the theoretical curve of the magnetic specific heat was obtained. This curve is compared with the experimental one in figure 3. Except for $T \sim T_c$, where fluctuations are important and the molecular field approximation does not apply, the fitting is satisfactory.

4.4 - Conclusion

The theoretical results obtained in this work, which emphasizes the role of the conduction band in the process of magnetic induction, allows a fitting with magnetic and specific heat data as good as that obtained by Frauenheim et al. [9] using a model Hamiltonian which contains besides a crystal field term an exchange interaction Heisenberg like between the rare earth pseudo-spins.

References

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FIGURE CAPTIONS

Fig. 1 : Spontaneous magnetization (in μ_B per Pr^{+3}) of $PrAl_2$ versus temperature (in K). The experimental points are from |3|. The curve was calculated with the parameters given in the text.

Fig. 2 : Inverse of the susceptibility (in mol/emu) of $PrAl_2$ versus temperature (in K). Curve 1 is computed from the parameters given in the text; curve 2 is taken from |4|.

Fig. 3 : Magnetic specific heat of $PrAl_2$ (in J/mol K) versus temperature (in K). The experimental points are from |5| and the curve is computed as told in the text.

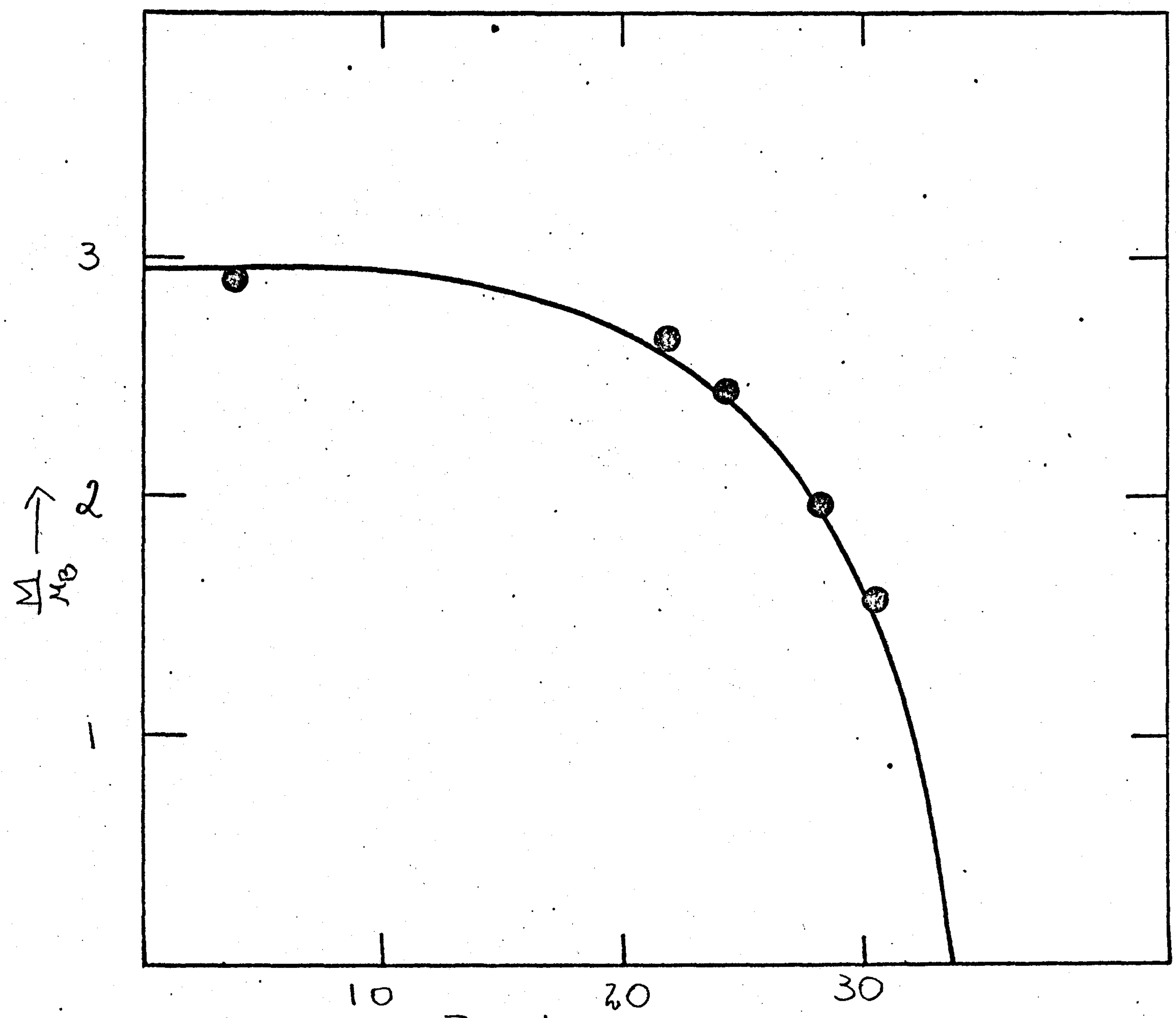

Fig. 1

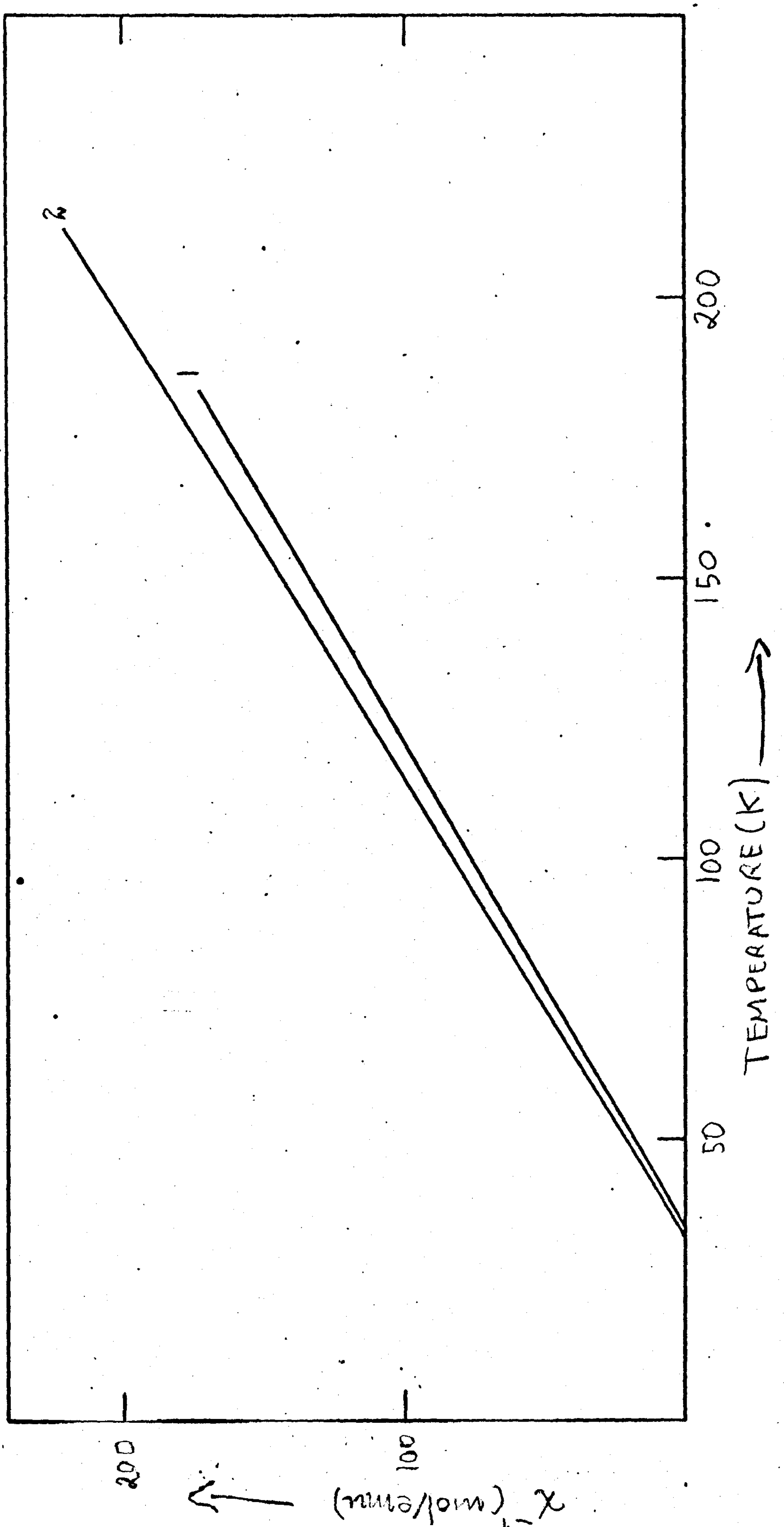

Fig. 2

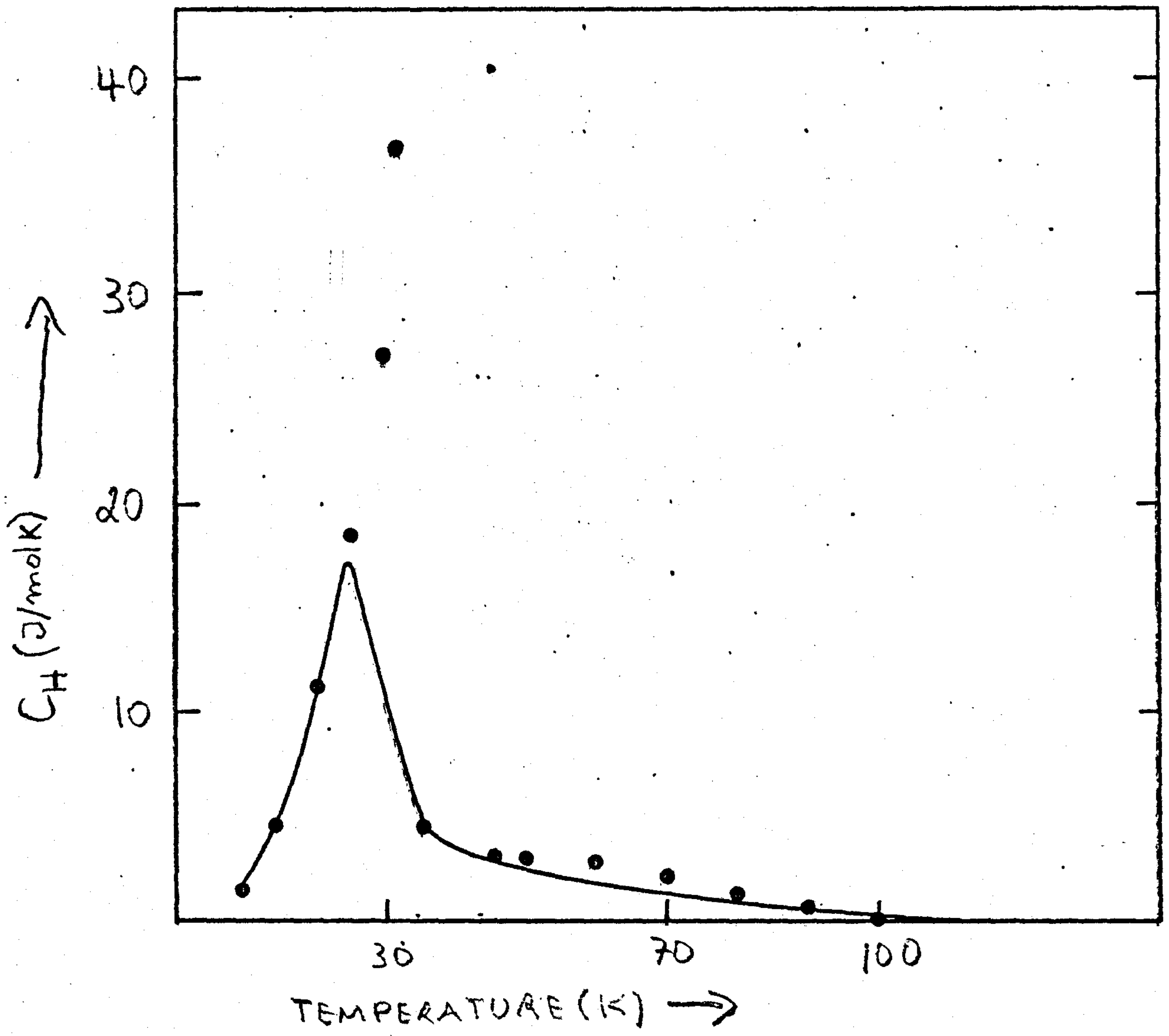

Fig. 3