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FERMICONDENSATION - AN ALMOST IDEAL GLASS TRANSITION

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ABSTRACT

A fermisystem in a thermodynamic density of states evaporates into a classical gas at a finite temperature. The converse process, "fermicondensation", is shown to be close to an ideal glass transition (a transition to a state of zero entropy at a positive temperature). The fermicondensation phenomenon has a number of properties in common with Derrida's random energy model. The present study of the two models weakens the Adam-Gibbs hypothesis concerning a diverging average relaxation time at the ideal glass transition.

1. INTRODUCTION

Most liquids are able to form glasses when cooled sufficiently fast to avoid crystallization [1,2]. Examples of glass forming liquids include polymeric liquids, ionic liquids, molten salts or metals, simple organic liquids, and the various oxides forming ordinary glass. The glass transition is not an ordinary phase transition, but a kinetic phenomenon. The transition is not sharp and its characteristic temperature, T_g , depends somewhat on the cooling rate. There are, however, speculations that the experimental glass transition is a manifestation of an underlying genuine second order phase transition to a state of zero entropy, an "ideal glass transition". This idea was proposed by Gibbs and DiMarzio in 1958 [3]. It resolves the so-called Kauzmann paradox [4,5], i. e., the fact that ^{the} excess entropy of the supercooled liquid upon cooling extrapolates to zero at a positive temperature T_0 . The excess entropy is the entropy of the liquid in excess of the purely vibrational contribution (estimated by the entropy of the crystal at the same temperature). This quantity represents the configurational entropy of the liquid and must be positive. Thus, either the extrapolation is incorrect or there is a phase transition at T_0 to a state of zero configurational entropy, an "ideal glass". The real glass transition intervenes before T_0 is reached, though, and the existence of an underlying ideal glass transition remains a conjecture.

Henceforth attention is focussed on the thermodynamics of the configurational degrees of freedom only; the vibrational degrees of freedom are ignored since they undergo little change at the

glass transition. Any thermodynamic system is characterized by the entropy as function of energy, $S(E)$ [6]. The temperature is defined by

$$\frac{1}{T} = \frac{\partial S}{\partial E} \quad (1)$$

The requirements of a positive temperature and of a non-negative specific heat lead to

$$\frac{\partial S}{\partial E} > 0, \quad \frac{\partial^2 S}{\partial E^2} \leq 0. \quad (2)$$

For an ordinary system, the state of zero entropy is reached only at $T=0$. Thus, if $E=0$ is the ground state energy, one has $\frac{\partial S}{\partial E} \rightarrow \infty$ for $E \rightarrow 0$. For a system exhibiting an ideal glass transition, by definition, $\frac{\partial S}{\partial E}$ is finite as $E \rightarrow 0$. These two possibilities are illustrated in Fig. 1. The ideal glass transition of Fig. 1b takes place at $T=T_0$ where

$$\frac{1}{T_0} = \lim_{E \rightarrow 0} \frac{\partial S}{\partial E} \quad (3)$$

Gibbs and DiMarzio [3] originally argued for the existence of an ideal glass transition in polymeric liquids from a study of a lattice model. Several years later it was shown by Gujrati and Goldstein [7] that the mean field approximations made by Gibbs and DiMarzio are incorrect and that, in fact, the lattice model has a positive entropy at any nonzero temperature. But despite this negative result, the Gibbs-DiMarzio idea remains attractive since it offers a simple resolution of the Kauzmann paradox.

The standard example of a system exhibiting an ideal glass transition is Derrida's random energy model (REM) from 1980 [8] (sec. 2). It seems that few other examples exist. This paper studies the "fermi condensation" phenomenon (sec. 3), which has a number of properties in common with the REM. Both models exhibit an ideal glass transition only in a certain limit. The two models also have similar relaxation behavior, as shown in sec. 4.

2. THE RANDOM ENERGY MODEL

This section briefly reviews the random energy model. To motivate this model, expand $S(E)$ to second order in E for a system exhibiting an ideal glass transition (where again it is assumed that the ground state is $E=0$):

$$S(E) = \alpha_1 E - \frac{1}{2} \alpha_2 E^2 + \dots \quad (4)$$

The function $S(E)$ is the logarithm of the density of states $n(E)$ [6]. Thus, ignoring the higher order terms in Eq. (4) corresponds to assuming a gaussian density of states,

$$n(E) \propto \exp \left[\alpha_1 E - \frac{1}{2} \alpha_2 E^2 \right] \quad (5)$$

The random energy model is now defined simply by choosing N

energy levels randomly from the gaussian [8]. These N energies completely define the system. It is important that N is finite, only after the thermodynamics is worked out is the $N \rightarrow \infty$ limit taken. In letting N go to infinity care must be taken to assure that the lowest energy remains close to zero; this is done by varying α_1 appropriately with N [8]. It is easy to see that the resulting model exhibits an ideal glass transition: Close to $E=0$ the density of states is approximately exponential,

$$n(E) \propto \exp[\alpha_1 E], \quad (E \approx 0). \quad (6)$$

For $T > T_0$ where $T_0 = 1/\alpha_1$, the maximum of the canonical probability, $n(E) \exp(-E/k_B T)$, increases with increasing T . The average energy, $\bar{E}(T)$, is an increasing function of temperature so the specific heat, and thereby the entropy, is positive. Below T_0 , on the other hand, the Boltzmann factor dominates over $n(E)$, driving the system to the lowest energy state. Thus, the entropy is zero. Actually, the entropy is only strictly zero in the $N \rightarrow \infty$ limit, but even for a finite system there is a sharp decrease in entropy below T_0 .

3. FERMICONDENSATION

A system of fermions is usually thought of in terms of occupied and unoccupied states, where it is implied that the

majority of the particles are to be found in occupied states. For some fermisystems, however, this picture is adequate only at low temperatures whereas at higher temperatures most particles are classical, i. e. , have energies for which the average occupation number is $\ll 1$. Consider a system of non-interacting fermions in a "thermodynamic" density of states $g(\epsilon)$, i. e. , where $S(\epsilon) = \ln(g(\epsilon))$ obeys Eq. (2). Expanding $S(\epsilon)$ around the zero temperature chemical potential (the fermi energy), μ_0 , leads to

$$g(\epsilon) = g_0 \exp \left[c_1 (\epsilon - \mu_0) - \frac{1}{2} c_2 (\epsilon - \mu_0)^2 + \dots \right] . \quad (7)$$

At low temperatures most particles have energies close to μ_0 . But at higher temperatures the Boltzmann tail of the fermifunction is not able to suppress the $\exp[c_1(\epsilon - \mu_0)]$ part of $g(\epsilon)$. The result is that most particles move to higher energies. The particles become classical in the sense that, for the majority of particles, the average occupation number is much less than one. The transition from a fermi to a classical system is continuous and not a sharp transition. It takes place around $T=T_0$ where T_0 is given by

$$T_0 = \frac{1}{k_B c_1} . \quad (8)$$

The transition upon cooling from high temperatures may be termed a "fermicondensation", since here the fermisea is formed from the classical gas. The term "gas" is relevant for the high temperature state where the particles effectively do not "interact" via the fermi exclusion principle.

The fermion condensation is signalled by a decrease in specific heat below T_0 . In Fig. 2 the specific heat per particle is shown for two different values of $\lambda = c_1/\sqrt{c_2}$ for a system with a gaussian density of states (Eq. (7)). Above T_0 most of the fermions has "evaporated". The particles move independently and each particle has a specific heat of $1/(2k_B T^2 c_2)$, as is easy to show. Below T_0 the specific heat per particle is small as in an ordinary fermion system. For large λ the transition is close to a phase transition. For $\lambda \rightarrow \infty$ an ideal glass transition takes place since in this limit the specific heat, and thereby the entropy, vanishes below T_0 .

We now briefly discuss relaxation in the model. Assume the simplest possible realistic dynamics, namely that where the jump rate of a fermion depends only on the energy of the initial state [9]. This corresponds to the case where the fermion, once it is excited into the transition state, has forgotten where it came from and may end up in any other (unoccupied) state. If the energy of the transition state is ϵ_0 , the relaxation time for jumps from a state of energy ϵ is

$$\tau = \tau_0 e^{\frac{\epsilon_0 - \epsilon}{k_B T}} \quad (\epsilon < \epsilon_0) \quad (9)$$

where τ_0 is a microscopic time. A cut-off at ϵ_0 has been introduced to ensure that no state has larger energy than the transition state. Now, below T_0 most particles have energy close to μ_0 since the density of states is exponentially increasing. Thus, relaxation is Arrhenius with an activation energy given by

$$\Delta \varepsilon = \varepsilon_0 - \mu_0 \quad (10)$$

Above T_0 , the average energy of the particles, $\bar{\varepsilon}$, depends on temperature. Since the particles here move independently, $\bar{\varepsilon}(T)$ is determined by maximizing the probability for one particle having energy ε :

$$\left. \frac{d}{d\varepsilon} \left[g(\varepsilon) e^{-\frac{\varepsilon}{k_B T}} \right] \right|_{\varepsilon = \bar{\varepsilon}(T)} = 0 \quad (11)$$

Because $g(\varepsilon)$ is by assumption a thermodynamic density of states, $\bar{\varepsilon}(T)$ is an increasing function of temperature so the activation energy of τ ,

$$\Delta \varepsilon = \varepsilon_0 - \bar{\varepsilon}(T), \quad (12)$$

decreases with increasing temperature.

Figure 3 shows a so-called Arrhenius plot of τ , i. e., $\ln(\tau)$ as function of T^{-1} . The behavior of Fig. 3 is like that seen in experiments on the real glass transition. Above the glass transition one observes an average relaxation time which is more than Arrhenius temperature dependent, while in the glassy phase τ is Arrhenius [10]. In experiment, there is furthermore a sudden decrease in the slope of the Arrhenius plot at T_g , just like in Fig. 3. However, one should not attach too much significance to the similarity between fermion condensation and experiments on the real liquid-glass transition. The real glass transition is a kinetic phenomenon and, for instance, the activation energy of in the glassy phase depends on the cooling rate at which the glass

was formed.

4. DISCUSSION

Fermi condensation is an alternative to the random energy model as a model exhibiting an almost ideal glass transition. Unlike the REM, no randomness is invoked in fermi condensation. Therefore, no ensemble averaging is needed to evaluate physical properties like the specific heat, as is necessary for a more accurate treatment of the REM than given here [8]. Since both models involve a gaussian density of states and since the fermions above T_0 move independently, the fermi system behaves like the REM above T_0 . In particular, if the assumption of transition rates being function of the initial energy only is made also for the REM [11], the two models have similar relaxation behavior. Note that the behavior of τ shown in Fig. 3 is valid for the REM even below T_0 : Here the REM is in its ground state so the activation energy of τ becomes constant. Thus, the two models are similar both as regards their thermodynamics and their relaxation behavior.

Adam and Gibbs have argued that at an ideal glass transition the average relaxation time becomes infinite because of the few available states to jump into [12]. This argument is still controversial, but this study certainly weakens the Adam-Gibbs hypothesis. In both models relaxation in the glassy phase takes place with a finite relaxation time which is, as a matter of fact, smaller than expected by extrapolating the behavior above T_0 .

To summarize, the fermicondensation phenomenon provides an abstract model for the ideal glass transition. No randomness is invoked in the model, but otherwise the model has a number of features in common with the random energy model. The present study of the two models does not confirm the Adams-Gibbs hypothesis concerning a diverging average relaxation time at T_0 .

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FIGURE CAPTIONS

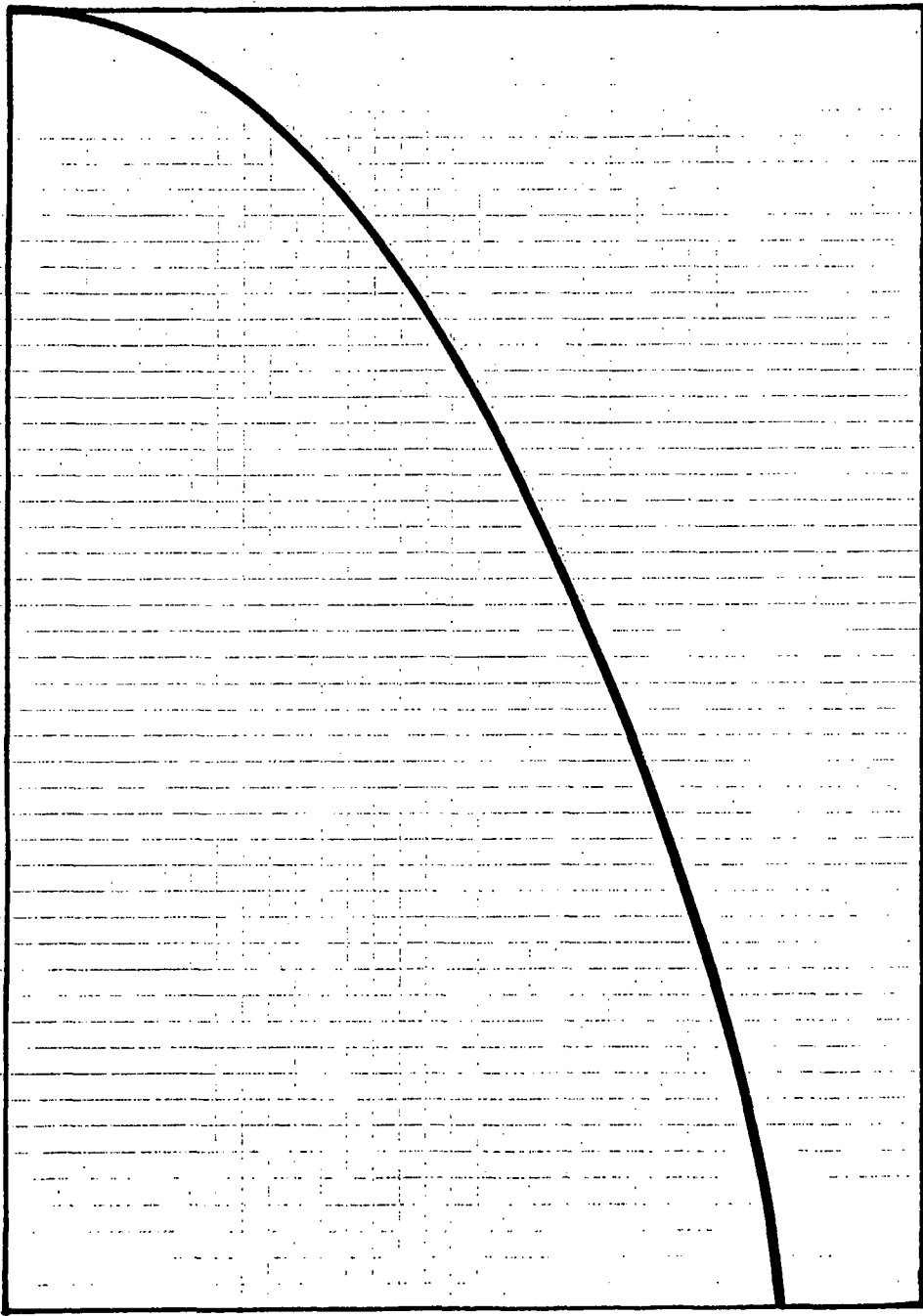
Fig. 1: Entropy as function of energy for an ordinary system (Fig. 1a) and for a system exhibiting an ideal glass transition (Fig. 1b). Temperature is defined by $\frac{1}{T} = \frac{\partial S}{\partial E}$ [6] and thermodynamics requires $S(E)$ to be an increasing function of energy with a non-positive second derivative. If $S(E)$ has a finite slope at $E=0$, there is a transition to a state of zero entropy at a positive temperature (Fig. 1b). This is the ideal glass transition, which is a second order phase transition.

Fig. 2: Specific heat per particle as function of temperature for a fixed number of fermions in a gaussian density of states (Eq. (7)). The two curves show the case of (a) $\lambda = 8$ and (b) $\lambda = 20$ where $\lambda = c_1/\sqrt{c_2}$. The specific heat is given in units of $k_B c_1^2/2c_2$. Around $T=T_0$ where T_0 is given by Eq. (8) the fermisea "evaporates". Above T_0 the fermions behave as independent classical particles, each with a specific heat $\propto T^{-2}$. Upon cooling, the classical gas "condenses" into the fermisea, the fermicondensation.

Fig. 3: Logarithm of the average relaxation time τ plotted as function of inverse temperature in the fermicondensation model where τ is given by Eq. (9) (schematic drawing). Above T_0 τ is more than Arrhenius temperature dependent while below T_0 τ becomes Arrhenius, much like for the real laboratory glass transition. The activation energy of τ is the slope of the secant

marked by dots (and not the slope of the tangent marked by the punctuated line, as is often assumed). Upon cooling the activation energy of τ increases until T_0 is reached. The activation energy then becomes constant. As discussed briefly in sec. 4, the random energy model has a similar relaxation behavior.

S(E)



E

Fig. 1a

$S(E)$

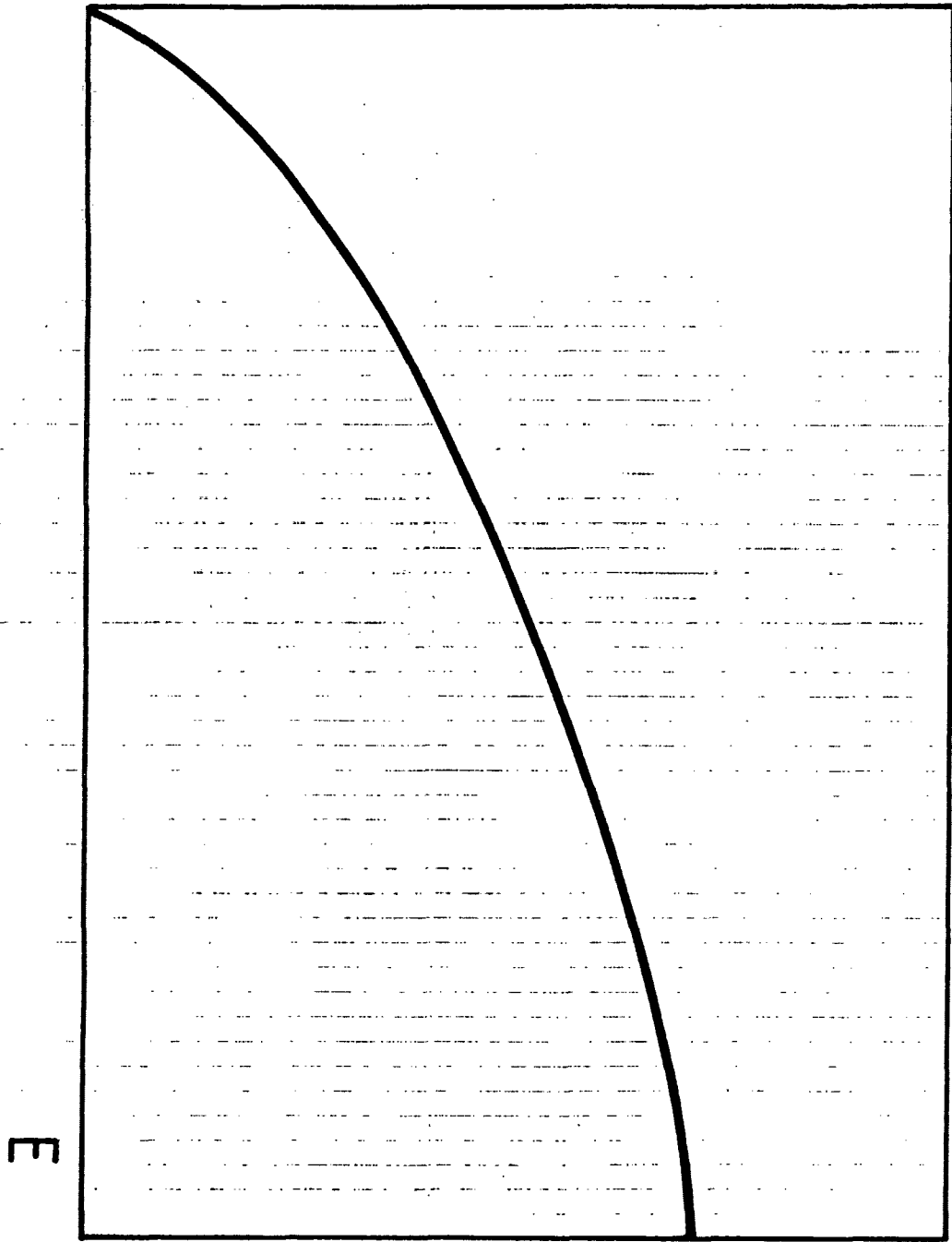


Fig. 1b

Fig. 2

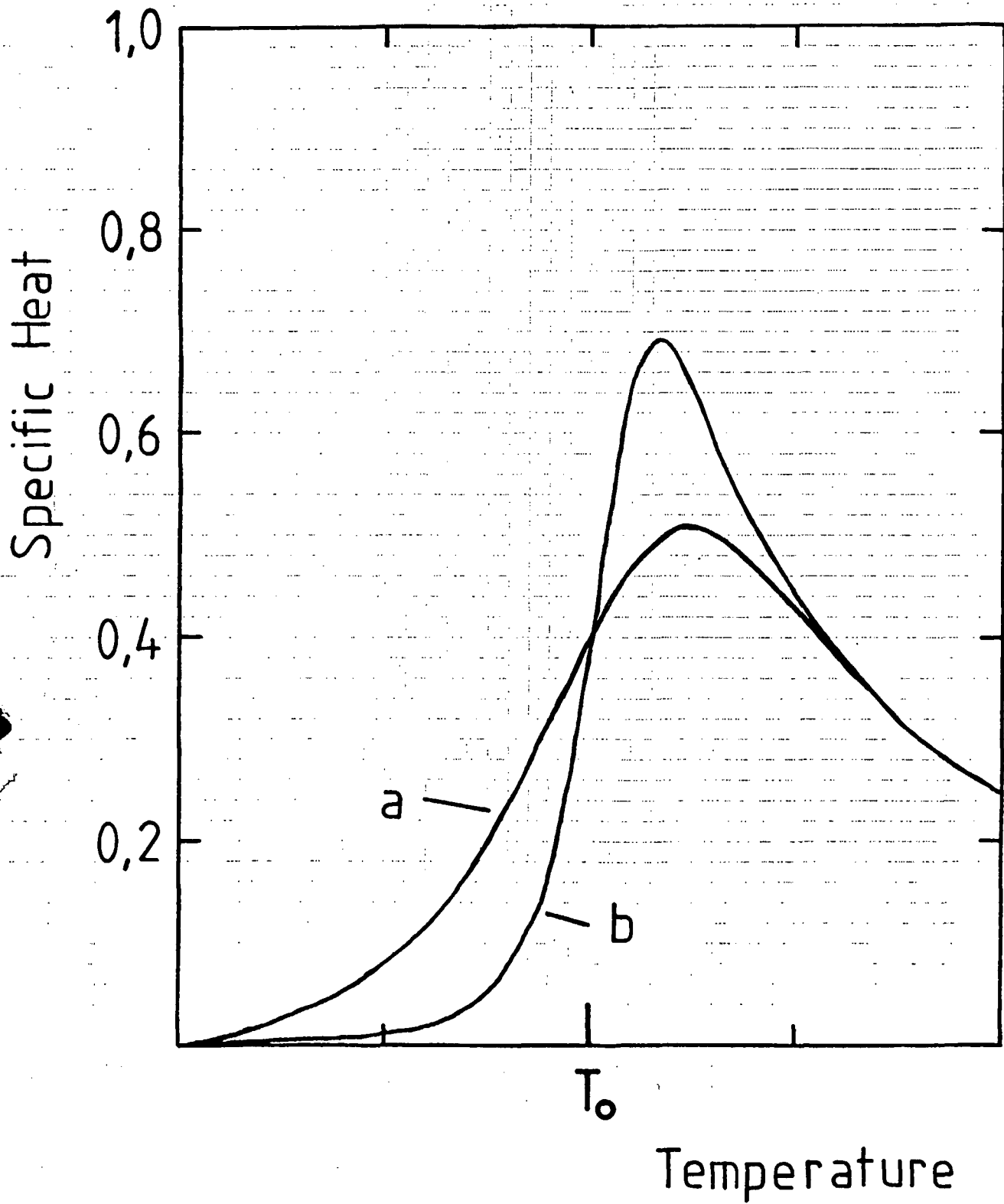
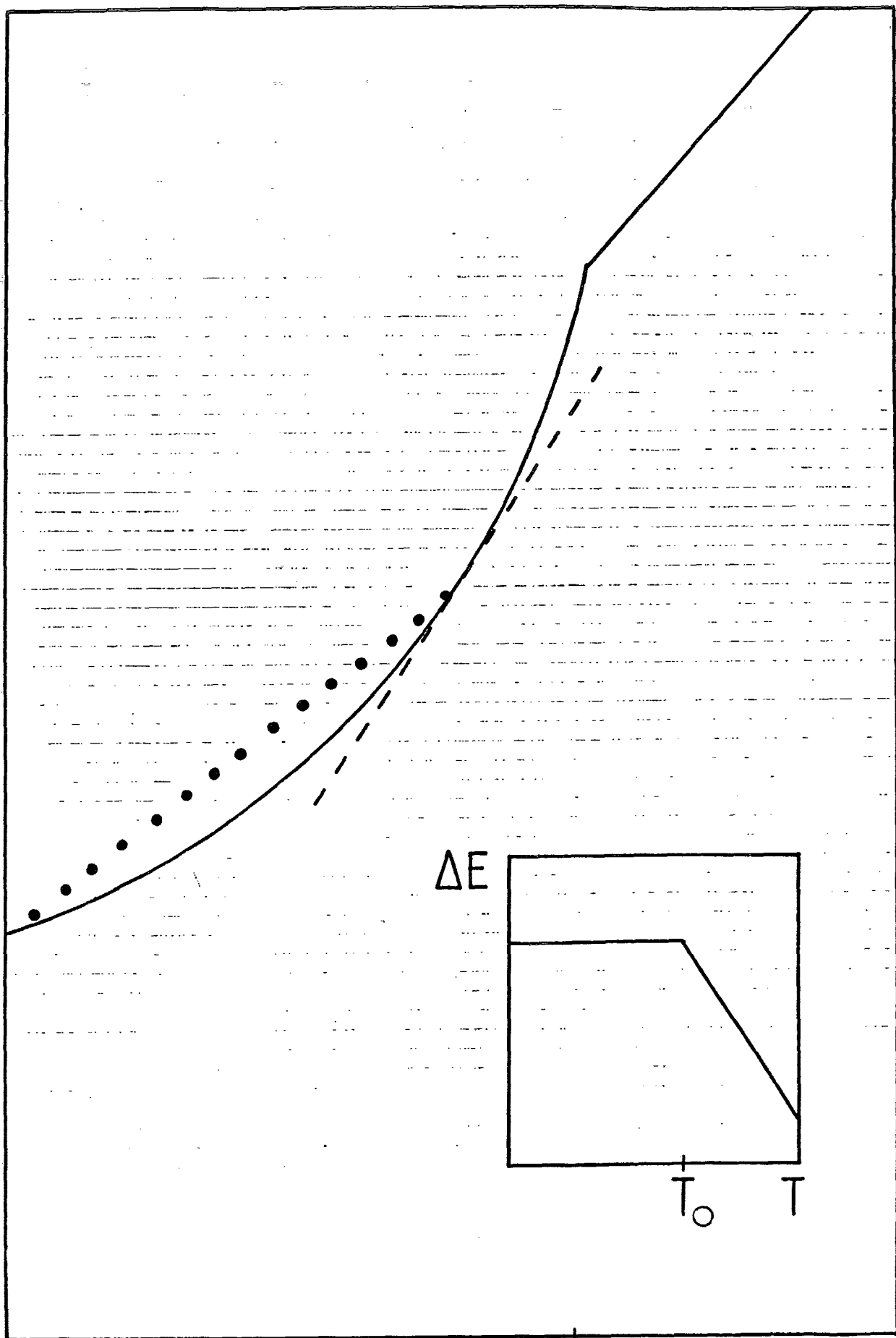


Fig 3

Fig.3

$\ln(\tau)$



ΔE

T_0

T

T_0^{-1}

T^{-1}

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