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LETTER TO THE EDITOR

Thermodynamic description of a dynamical glassy transition

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Abstract. For the dynamical glassy transition in the p-spin mean-field spin-glass model a thermodynamic description is given. The often considered marginal states are not the relevant ones for this purpose. This leads us to consider a cooling experiment on exponential timescales, where lower states are accessed. The very slow configurational modes are at quasi-equilibrium at an effective temperature. A system-independent law is derived that expresses their contribution to the specific heat. t/t_w-scaling in the ageing regime of two-time quantities is explained.

The structural glass transition occurs only in an idealized adiabatic cooling procedure at the Kauzmann temperature T_K. In a realistic experiment with finite cooling rate a gradual freezing transition takes place in a small interval centred around a higher temperature T_f, that depends on the cooling rate. Although the freezing is not a sharp thermodynamic transition, there can be some 15 decades in time involved, from picoseconds to many hours. By extrapolation from the high- and low-temperature sides, one may define jumps in quantities such as the specific heat and the compressibility. It has been pointed out by Jäckle [1] and Palmer [2] that the freezing transition can be described as a smeared thermodynamic transition, on a thermodynamic basis, with ensemble averages replacing time averages. The observation time sets the timescale that separates 'fast' processes (timescale less that the observation time) from 'slow' ones. The latter are essentially frozen.

Upon cooling, a liquid freezes dynamically in a glassy state with extensively smaller entropy. The free energy of the glassy state is then much larger, and it is not obvious why the system can become captured in such a state. The point is that the condensed system has lost the entropy of selecting one out of the many equivalent states; this part of the entropy is called the *complexity*, *configurational entropy*, or *information entropy* $\mathcal{I}$ [1, 2]. This can be understood as follows. For long times the system is stuck in states with long (but finite) lifetimes. These states are called 'components' by Palmer [2] and 'states', 'Thouless–Anderson–Palmer states' or 'TAP-states' in spin-glass theory. When the Gibbs free energy $F_{\bar{a}}$ of the relevant state $\bar{a}$ has a large degeneracy $\mathcal{N}_{\bar{a}} \equiv \exp(\mathcal{I}_{\bar{a}})$, the partition sum yields $Z = \sum_a \exp(-\beta F_a) \approx \mathcal{N}_{\bar{a}} \exp(-\beta F_{\bar{a}})$, so $F = F_{\bar{a}} - T\mathcal{I}_{\bar{a}}$ is the total free energy of the system. The entropy loss arises when the system chooses the state to condense into, since from then on only that single state is observed†. As the total entropy $S = S_{\bar{a}} + \mathcal{I}_{\bar{a}}$ is continuous, so is the total free energy.

† This sudden loss of entropy is reminiscent of the collapse of the wavefunction in the quantum measurement.

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The aim of this work is to analyse this thermodynamical picture of the dynamical freezing transition in a well understood mean-field model. The difficulty is to extract the information that is not due to the mean-field limit. The advantage is that we find strong constraints satisfied by the dynamics, without having solved it. We first discuss the present status of marginal replica theory, its relation with dynamics and its fundamental flaws. Then we shall propose a solution to these paradoxes.

We consider the mean-field p-spin interaction spin-glass model of N coupled spins in a field with Hamiltonian

$$\mathcal{H} = -\sum_{i_1<i_2<\cdots<i_p} J_{i_1 i_2 \ldots i_p} S_{i_1} S_{i_2} \ldots S_{i_p} - H \sum_i S_i. \tag{1}$$

The independent Gaussian random couplings $J_{i_1 i_2 \ldots i_p}$ have average zero and variance $J^2 p!/2N^{p-1}$. The spins are subject to the spherical condition $\sum_i S_i^2 = N$.

This model has a close analogy with models for the structural glass transition [3]. On a static level there occurs a transition to a state with one-step replica symmetry breaking (1RSB) at a temperature T_K, comparable with the ideal glass transition in an adiabatic cooling experiment. The 1RSB calculation involves three parameters. The overlap of spin configurations in two states can equal the Edwards–Anderson parameter or self-overlap q_1, or have the smaller value q_0 ($= 0$ for $H = 0$); these values occur with probability $1 - x$, and x, respectively. The free energy reads

$$\frac{F}{N} = -\frac{\beta J^2}{4}(1 - \xi q_1^p - x q_0^p) - \frac{\beta H^2}{2} Q - \frac{T}{2x} \log Q + \frac{T\xi}{2x} \log(1 - q_1) - \frac{T q_0}{2Q} \tag{2}$$

where $\xi = 1 - x$ and $Q = 1 - \xi q_1 - x q_0$. Unless stated otherwise, we shall take $H = 0$. q_0 and q_1 are determined by optimizing F. For x the situation is not unique, but depends on the timescale considered. Setting $\partial F/\partial x = 0$ yields the static phase transition at $T \equiv T_K$†. When considering the Langevin dynamics of this model, one may derive dynamical equations for correlation and response functions by taking first $N \to \infty$. [5] Solving for large t leads to a sharp phase transition at larger temperature $T_A = J\{p(p-2)^{p-2}/2(p-1)^{p-1}\}^{1/2}$, and a different form $x(T)$. This dynamical value for x can be simply rederived from a replica calculation in which the 'replicon' or 'ergodon' [5, 6] fluctuation mode is taken to be massless. This leads to the marginality criterion $p(p-1)\beta^2 J^2 q_1^{p-2}(1 - q_1)^2/2 = 1$. At the dynamical transition there is a sharp jump in the specific heat [5].

To discuss the situation, we must first consider the TAP states. A state is labelled by a and has local magnetizations $m_i^a = \langle S_i \rangle^a$. Its free energy $F_a(T)$ is a thermodynamic potential that determines the internal energy and the entropy by its derivatives. In the present model $F_a = F_{TAP}(m_i^a)$ is known explicitly. It is a minimum of the 'TAP' free-energy functional [7–9]

$$F_{TAP}(m_i) = -\sum_{i_1<\cdots<i_p} J_{i_1 \ldots i_p} m_{i_1} \ldots m_{i_p} - H \sum_i m_i - \frac{NT}{2} \log(1 - q)$$
$$-\frac{N\beta J^2}{4}(1 + (p-1)q^p - p q^{p-1}) \tag{3}$$

where $q = (1/N)\sum_i m_i^2$ is the self-overlap. Below we shall argue that the commonly used Gibbs weight, $p_a = \exp(-\beta F_a(T))/Z$ is the relevant one. Given the p_a's one can define the component averages such as $\overline{F} = \sum_a p_a F_a$, $\overline{U} = \sum_a p_a U_a$, $\overline{C} = \sum_a p_a C_a$, and even

† On a static level the system condenses in the temperature range $T_K < T < T_A$ into a state with higher free energy but with complexity such that the total free energy equals the would-be paramagnetic free energy [3].

the complexity [1, 2] $\mathcal{I} = -\sum_a p_a \ln p_a$. For observables the direct evaluation from the ordinary partition sum should coincide with the outcome of the TAP analysis: $U = \overline{U}$, $M = \overline{M}$. They need not be derivatives of $\overline{F}$. The role of the complexity in dynamical transitions was first pointed out by Kirkpatrick and Wolynes for Potts models [3] and for the p-spin model by Crisanti and Sommers [9]

For all $T < T_A$ the mode-coupling equations come in due to the presence of a zero mode. On a quasi-static level, this dynamical zero mode is related to a massless fluctuation eigenvalue (the 'ergodon') in the related replica calculus [5, 3]. We recently assumed that this is a very general phenomenon. It will automatically become trapped in a state with diverging timescale, whenever present. The marginal replica free energy has the form

$$F = \overline{F} - \frac{T\mathcal{I}_c}{x} \tag{4}$$

where

$$\mathcal{I}_c = N\left(\frac{1}{2}\log(p-1) + \frac{2}{p} - 1\right) \qquad \text{(marginality)} \tag{5}$$

is the complexity of the marginal states. Below T_A the free energy lies below the one of the paramagnets and has a larger slope. This would naively imply a latent heat.

There is, however, another prediction for the free energy [5]. It involves the internal energy and an entropy obtained by integrating $(1/T)\partial U/\partial T$ from a temperature in the glassy phase up to some large temperature. The resulting 'experimental' glassy free energy [5]

$$F_{\text{exp}} = \overline{F} - T\mathcal{I}_c \tag{6}$$

exceeds the paramagnetic free energy quadratically and is, by construction, a thermodynamic potential. It was reproduced by analysis of the TAP states [9].

The difference between (4) and (6) led us to question fundamentally the validity of replica calculations for dynamical 1RSB transitions. Our aim was to find the meaning of the logarithm of the dynamical replica free energy (equation (2) with $\partial F/\partial q_0 = \partial F/\partial q_1 = 0$ but with $\partial F/\partial x \neq 0$).

By doing the full thermodynamic analysis of the TAP partition sum $\sum_a \exp(-\beta F_a)$ at $H = 0$, we found that that the replica free energy (4) is reproduced [11]. As the glassy free energy lies below the continuation of the paramagnetic one, we considered this as proof that the complexity is the driving force for the dynamical phase transition [11]. In the doing the analysis we realized that the calculation of the dynamical complexity cannot be separated from the calculation of the free energy, and that replica symmetry breaking is essential. The problem boils down to a replicated TAP free energy that has six replicated order parameters. We have now extended this analysis to $H > 0$. For 1RSB with a common breakpoint $\tilde{x}$, each replica order parameter now brings three parameters. We thus obtain an optimization problem in 18 variables, that we can partly solve using an algebraic manipulation program. We have verified that the total free energy, the internal energy and the magnetization, calculated within the TAP approach, coincide with their replica values. For the magnetization this is particularly satisfying, as M_a of a given marginal TAP state is temperature independent. (It then holds that $\partial M_a/\partial T = \partial S_a/\partial H = 0$. Nevertheless, the component average $\overline{M} = \sum_a p_a M_a$ is temperature and field dependent, and equal to the replica value $M = \beta H(1 - \xi q_1 - x q_0)$).

For interpreting equation (4) one might be tempted to consider $\mathcal{I}_c/x$ as the full complexity. Since $x \to 0$ for $T \to 0$, this is hard to explain on a physical basis, however. The $1/x$ dependence in (4) does not disappear after the quantization of the spherical model,

recently proposed by us [12, 6]. Analysis of the equations for quantized spherical spins, or for Ising spins, teaches us that, though T_A shifts, the term $T\mathcal{I}_c/x(T)$ survives for marginal states.

The other interpretation of equation (4) is that $T_e \equiv T/x$ is an effective temperature at which the slow processes leading to $\mathcal{I}_c$ are in quasi-equilibrium. This interpretation is promising, since the same effective temperature shows up in the fluctuation-dissipation relation in the ageing regime of the mode-coupling equations [13].

There remain some paradoxes connected to the marginal states. For large p one has $\mathcal{I}_c \sim (N/2)\log p$, which (for quantized spherical spins or for Ising spins) already exceeds the total entropy available. This shows that *the dynamical transition at T_A has no thermodynamic counterpart in short-range systems*, at least for large enough p. In fact, T_A may be identified with the critical temperature of mode-coupling theory, which lies well above the freezing temperature T_f. Even more cumbersome is that for marginal states with $H \neq 0$ we have shown violation of the inequality $C \geqslant \overline{C}$ [11]. This says that marginal states are intermediate-time states, from which the system must escape at longer times.

The implication of these arguments is rather dramatic: for comparing with realistic short-range systems *we must abandon the assumption that the marginal states are the physically relevant states*. Let us see how this could happen. Marginality arises automatically in the dynamical equations after taking first the limit $N \to \infty$ and then $t \to \infty$. This order of limits, however, prevents all activated processes, which in the mean-field model would need a time $\sim \exp(N)$. As a result, all dynamics is confined to the highest TAP states, which are marginal. The lower states are never reached. In a realistic short-range glassy system, however, there is no sharp distinction between slow and activated processes, and the latter can certainly not be omitted. So in a realistic experiment we do expect to reach lower states.

In order to compare with realistic systems and to avoid thermodynamic paradoxes, we propose another look at the mean-field system. We consider the system at fixed large N under such conditions that a range of TAP states below the highest (marginal) ones are accessed. We thus focus our attention more on the free energy of the state than the timescale needed to reach it. At $H = 0$ the free energy of the TAP states can be characterized by a parameter η ($\eta_{\rm st} \leqslant \eta \leqslant 1$), that enters the condition

$$\beta^2 p(p-1)q^{p-2}(1-q)^2/2 = \eta. \tag{7}$$

For $\eta = 1$ this would be the marginality condition, while the static equations can also be put in this form [4] with $\eta = \eta_{\rm st} < 1$ independent of T. One now finds the breakpoint $x = (1-q)(p-1-\eta)/q\eta$, so choosing η between $\eta_{\rm st}$ and 1 can alternatively be seen as a way of fixing the mysterious parameter x, for which no obvious criterion was present. In the present approach it is directly related to the timescale $t_\eta \equiv \exp(N\tau_\eta)$ at which the η-states are reached. Unfortunately, the precise relation between the logarithmic time variable τ_η and the lowest reachable TAP free energy at that scale, F_η, is unknown.

We assume that at a given exponential timescale τ_η the dominant states are determined by a saddle point. Such behaviour was seen in a solvable glassy transition in a directed polymer model, recently proposed by us [15]. This allows us to restrict the discussion to TAP states with a common free energy.

We have verified that the equivalence between the replica and the TAP analysis also holds for $\eta < 1$. The η-states have a complexity that can be calculated in a straightforward manner from their replica free energy

$$\mathcal{I}_c(\eta) = \frac{N}{2}\left\{\log\frac{p-1}{\eta} - \frac{(p-1-\eta)(\eta+1)}{p\eta}\right\}. \tag{8}$$

As η decreases from 1 to $\eta_{\rm st}$, $\mathcal{I}_c(\eta)$ goes down from the marginal value $\mathcal{I}_c(1)$ to the static value $\mathcal{I}_c(\eta_{\rm st}) = 0$. A sensible cooling experiment must have a temperature-dependent value of η. If η does not approach $\eta_{\rm st}$ fast enough for $T \to 0$, then the system will reach a $T = 0$ state with finite configurational entropy, from which it may relax at later times. In hindsight this is just what is needed to describe a realistic cooling experiment in the mean-field p-spin model. Let us assume some logarithmically slow cooling traject $T(t)$ where also $\eta = \eta(t)$ depends on time in a still unknown, but determined fashion. We can then eliminate t and construct the function $T(\eta)$ (and its inverse $\eta(T)$) that characterizes our cooling traject. A dynamical freezing transition will occur when the lowest state reached at time t freezes at the temperature $T(t)$. This occurs at $T_f = T(\eta_f)$ where η_f is the solution of

$$T(\eta) = J\left\{\frac{p\eta(p-1-\eta)^{p-2}}{2(p-1)^{p-1}}\right\}^{1/2}. \tag{9}$$

As $T(t)$ and $\eta(t)$ will depend on the cooling procedure, we expect $T(\eta)$ to do the same, so that T_f will not be universal but depend on the specific traject.

Also the assumption of Gibbs weights is now justifyable. TAP states with free energy larger than or equal to the ones fixed by η are now effectively in thermodynamic equilibrium, and may be described by the Gibbs weight. Lower ones play no role.

The paradoxes related to marginal states, signalled here, occur much more widely. At present it is a whole field of research to consider dynamics of mean-field models by first taking the mean-field limit and then considering large times. Though the approach has relevance for short and intermediate time dynamics, its long-time regime is a result of 'squeezing the system into marginal states', which has no bearing on the long-time relaxation of short-range models. This is already expressed by the unique sharp dynamical transition temperature T_A, found in the dynamical approach. It disappears on exponential timescales, and is replaced by a cooling-rate dependent freezing temperature.

The basic problem with our approach is the complexity. An important question is whether it can be measured in the glassy phase. If so, it should be related to the specific heat or the temporal energy fluctuations. When monitoring the internal energy as function of time, as is easily achieved in a numerical experiment, one essentially obtains a noisy telegraph signal. Each plateau describes trapping in a TAP state for some definite time. The variance of the noise in the internal energy on this plateau is equal to $T^2\overline{C} = T^2\sum p_a C_a = \sum_a \langle(\delta U_a)^2\rangle = T^2\sum p_a \mathrm{d}U_a/\mathrm{d}T$. From time to time the system moves to another TAP state, causing additional noise. The variance of the total noise equals T^2C, and should exceed $T^2\overline{C}$ [2]. In [11] it was pointed out that there can be an extensive difference between the specific heat $C = \mathrm{d}U/\mathrm{d}T = \mathrm{d}\overline{U}/\mathrm{d}T = \sum_a \mathrm{d}(p_a U_a)/\mathrm{d}T$ and the component average energy fluctuations $\overline{C} = T\mathrm{d}\overline{S}/\mathrm{d}T$. We can now consider their difference at $H = 0$ in a cooling experiment of the type introduced above. We find the *excess specific heat*

$$\Delta C \equiv C - \overline{C} = N\frac{q(1-\eta)}{2p\eta(1-q)}\,T\,\frac{\mathrm{d}\eta}{\mathrm{d}T}. \tag{10}$$

Using this we can easily verify the fundamental relation

$$\frac{\mathrm{d}}{\mathrm{d}T}\mathcal{I}_c(T) = \frac{1}{T}x(T)\Delta C(T). \tag{11}$$

It expresses the complexity in terms of measurable quantities, namely the excess specific heat and the ergodicity-breaking parameter x. Equation (11) holds equally well for $H \neq 0$ but fixed. In the generalization of (10) one now encounters parameter $\eta(T; H)$ and its

derivative $\partial\eta/\partial T$. The proof of equation (11) for $H \neq 0$ is lengthy but could be verified using an algebraic manipulation program†.

The complexity can also be measured along the transition line in the (T, H)-plane. This issue is related to the Ehrenfest relations, and has been discussed elsewhere [16].

With $T_e = T/x$ equation (11) can also be written as

$$\frac{\mathrm{d}U}{\mathrm{d}T} = T\frac{\mathrm{d}\overline{S}}{\mathrm{d}T} + T_e\frac{\mathrm{d}\mathcal{I}_c}{\mathrm{d}T}. \tag{12}$$

Our interpretation of the replica results leads to an effective temperature $T_e(t) = T(t)/x(t)$. The slowest active modes are at quasi-equilibrium at this effective temperature. This explains why T_e also shows up in the fluctuation-dissipation relation [13, 14]. As they set the slowest timescale, they must also dominate the dynamical free energy. This is why the change of structural modes $\mathrm{d}\mathcal{I}_c/\mathrm{d}T$ in equation (12) has prefactor T_e.

In numerics on the fluctuation-dissipation relation in spin glasses and even binary soft spheres [17] it has been observed that the factor x is linear in T. Let us give a simple explanation for that behaviour. As the effective temperature must exceed the Kauzmann temperature, we can estimate $T_e(T) \approx$ constant, implying indeed $x = T/T_e \sim T$.

If we quench the system from high temperatures deep into the glassy state, and let it evolve freely during a long waiting time t_w, T_e will be set by equating the equilibrium relaxation time τ_{eq} at the instantaneous T_e to t_w: $\tau_{eq}(T_e) = t_w$. Naively, one then expects two-time quantities in dynamics to be a function of $(t - t_w)/\tau_{eq}(T_e) = t/t_w - 1$, explaining immediately the often observed t/t_w scaling in the ageing regime. There could be logarithmic corrections to this behaviour.

Replacing $x \to T/T_e$ we can write equations (2) and (4) as

$$F(T, T_e) = U - T\overline{S} - T_e\mathcal{I}_c. \tag{13}$$

It is a dynamical free energy determining $\overline{S} = -\partial F/\partial T$ and $\mathcal{I}_c = -\partial F/\partial T_e$, while $U = F + T\overline{S} + T_e\mathcal{I}_c$. The system-independent laws (12) and (13) are the cornerstone for our thermodynamic description of the dynamical glassy transition, and expected to be valid in general.

We have verified equation (12) for a cooling procedure in the Ising chain with Glauber dynamics. At $T = 0$ it is a coarsening problem of alternating up and down clusters of average length $\xi(t) = \sqrt{4\pi t}$ and energy $U(t) = NJ(-1 + 2/\xi)$ [18]. The number of configurations with this energy can be counted by standard methods. Its logarithm reads $\mathcal{I}_c = N(1 + \ln\xi)/\xi$. From the internal energy one may introduce an effective temperature: $1/\xi = \mathrm{e}^{-2\beta_e J} \to T_e = 2J/\ln\xi(t)$. The same value would follow when defining T_e from the complexity $\mathcal{I}_c$. Both results can thus be combined in terms of a dynamical free energy $F = -NT_e\ln(2\cosh\beta_e J)$. This is a special case of equation (13) with $\overline{S} = 0$.

As above, we then consider a cooling experiment where $T(t) = 2x(t)J/\ln\xi(t)$ with a smooth decreasing function $x(t)$. In the initial regime $x(t) > 1$ the system will achieve equilibrium at the instantaneous temperature $T(t)$. For $x(t) < 1$ this will not happen. The system falls out of equilibrium and behaves as at $T = 0$: it is at quasi-equilibrium at $T_e(t) = T(t)/x(t)$. The freezing transition occurs around $T_f = T(t_f)$ where $x(t_f) = 1$. In the frozen phase equations (12), (13) are valid with $\overline{S} = 0$. At $T = 0$ (and for all $T < T_f$) the on-site correlation function is found to be a scaling function of t/t_w,

Summarizing, we have shown that a thermodynamic description of a dynamical freezing transition can be given‡. We have been led to discard the whole issue of marginal states.

† The quantity $C - \overline{C} = -a(T, H; \eta)H^2 + b(T, H; \eta)\partial\eta/\partial T$ is negative when $\eta(T; H) = 1$ (marginal states). In the present approach with $\eta < 1$, $\partial\eta/\partial T > 0$ the equilibrium inequality $C \geqslant \overline{C}$ can be satisfied by the dynamics.

‡ Equations (12) and (13) with $\overline{S} = 0$ also hold for cooling procedures in the backgammon model of [19].

For comparison with realistic short-range systems, we consider the mean-field system at exponential timescales, where lower states are accessed. They contribute to the partition sum of Gibbs weights over dynamically relevant states. This approach naturally leads to slow cooling procedures where a dynamical freezing transition occurs at a tunable temperature. This dynamical transition is described by a free energy that depends on the real temperature and on an effective temperature.

Within the framework of the present approach the effective temperature shows up in equation (12), that is to say, in the formulation of the second law of thermodynamics. Since the submission of this work we have shown that, at least within the dynamics of a toy-model that does not rely on the mean-field approximation, the effective temperature depends slowly on time. It also shows up in fluctuation formulae and in the fluctuation-dissipation relation [20]. This approach also explains the confusion that has existed for half a century concerning the validity of the Ehrenfest relations along the freezing line, and the value of the Prigogine–Defay ratio [16].

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