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Relaxation dynamics in a reentrant CrFe ferromagnet: a random trap analysis

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Abstract. The decay of the thermoremanent magnetization of a reentrant $Cr_{79}Fe_{21}$ ferromagnet with $T_c \simeq 70$ K has been measured as a function of temperature T and wait time t_w, over an observation time interval $1\ s < t < 10^4$ s. A characteristic temperature $T_g \simeq 25$ K is identified, below which the relaxation dynamics show the non-equilibrium age dependence and thermal cycling effects typical of glassy materials, and above which the dynamics are 'stationary'. Fits to the predictions of a model based on anomalous diffusion among random free energy traps, with lifetimes τ distributed according to $\psi(\tau) \sim \tau^{-(1+x)}$, show that the dynamic crossover is consistent with a change in the structure of phase space, from a regime $T < T_g$ where $x < 1$, ergodicity is broken for all finite system ages, and the system configurations resemble those of the random energy model, to a regime $T > T_g$ where $x > 1$, and the system ergodically probes every available trap, resulting in a unique relaxation response. This analysis provides a quantitative link between 'reentrancy' and the formation of a glassy spin structure below T_g, as well as support for a particular aging mechanism of general applicability to all disordered systems.

1. Introduction

The relaxation dynamics of structurally disordered materials exhibits a remarkable 'universality'. In particular, the dielectric, mechanical (viscoelastic) and magnetic properties of 'glassy' materials as diverse as ordinary glasses [1], amorphous polymers [2], random magnets [3], ferroelectrics [4], glass-forming liquids [5], and even high-T_c superconductors [6], all feature an anomalously slow, non-Debye, temporal response to a step-function excitation, which has been described analytically by a variety of empirical functions, of which the two-parameter stretched exponential, $q(t) = \exp[-(t/\tau)^\beta]$ with $\beta < 1$, is perhaps the most popular. Furthermore, many of these systems exhibit aging effects, which refers to the experimental observation that the relaxation response is not unique, but rather depends on the time the material has been kept below the glass (or gelation or freezing) temperature before the excitation is applied, thus suggesting that thermodynamic equilibrium may never be reached on normal laboratory time scales. The generic character of the glassy relaxation response suggests an underlying physical mechanism of considerable generality, which is capable of transcending the diversity of system-specific details, like material, physical state, bonding type, polarizing species, and geometrical configuration.

Of the more recent theoretical attempts to replicate slow relaxation and non-equilibrium dynamics in spin systems, phenomenological models of domain growth, like those of Fisher and Huse [7], and Koper and Hilhorst [8], offer a physically appealing interpretation of the aging phenomenon and some of its experimental systematics, but are not readily adaptable to other physical systems, and predict analytical forms for the relaxation response which are

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not compatible with the measured decay, while alternative approaches of potentially greater generality, such as the de Dominicis, Orland and Lainée [9] treatment of random energy level dynamics based on the Sherrington–Kirkpatrick (SK) model [10], or the Chamberlin and Haines model [11] of finite-size quantized dispersive excitations within a percolation distribution of fixed domains, include no explicit aging mechanism.

However, recently, Bouchaud [12] has formulated a phenomenological 'random-trap' theory of aging, which appears to be applicable to all disordered systems, and is predicated on the general notion of a very rough energy landscape with many local minima corresponding to metastable configurations, surrounded by high energy barriers, each of which can trap the system for a time τ. For a sufficiently broad distribution $\psi(\tau)$, such as that associated with the random energy model (REM) [13] or the standard mean field SK model [14]:

$$\psi(\tau) \sim \tau^{-(1+x)} \text{ with } 0 < x < 1 \tag{1}$$

$\langle\tau\rangle$ diverges, and ergodicity is broken, in the sense that the system is essentially never able to probe the deepest traps as it evolves towards equilibrium. In fact, the deepest trap τ_{max} actually encountered during the aging process is of the order of (and thus limited by) the wait time t_w that the system is held at constant temperature $T < T_g$ before the excitation is applied, plus the elapsed observation time t after excitation, the latter to recognize the possibility of aging during relaxation. In the extreme non-ergodic limit of 'short' wait times, the model predicts a decay of the form:

$$m(t) = m_0 \exp\left[-\gamma \int_0^t t^{-x}(t + t_w)^{x-1}\, dt\right] \tag{2}$$

with a crossover from stretched exponential behaviour for $t \ll t_w$, to power law behaviour for $t \gg t_w$.

When $x > 1$, the distribution of lifetimes $\psi(\tau)$ in (1) decays rapidly enough for $\tau \cdot \psi(\tau)$ to become normalizable, $\langle\tau\rangle$ is finite, and the model predicts that the deepest trap encountered during the waiting time t_w is $\tau_{max} \ll t_w$, so that the system equilibrates essentially instantaneously after the quench. In fact, by following a procedure analogous to that described by Bouchard for $x < 1$, we have been able to show that the relaxation dynamics in the $x > 1$ regime are determined by a microscopic cut-off time τ_0 (rather than by the macroscopic waiting time t_w), and obey a simple power law:

$$m(t) = m_0\left(\frac{\tau_0}{t}\right)^{x-1} \quad (x > 1). \tag{3}$$

In this paper, we present measurements of the low field thermoremanent decay of a polycrystalline $Cr_{79}Fe_{21}$ ferromagnet over four decades of observation time ($1\text{ s} < t \leqslant 10^4$ s), as a function of both temperature and system age, under two thermal protocols: a simple temperature 'quench', and a temperature 'quench' followed by thermal cycling. The system exhibits two thermally distinct relaxation regimes, and these relaxation dynamics are analysed and interpreted within the framework of the random trap model outlined above.

2. Experimental details

An alloy of $Cr_{1-y}Fe_y$ containing nominally $y = 0.21$ was prepared by arc melting the appropriate amounts of 99.99% pure Cr chunks and 99.99% pure Fe wire on the water-cooled copper hearth of a titanium-gettered argon arc furnace with a tungsten electrode. The

ingot was repeatedly inverted and remelted in order to achieve a homogeneous consistency, and melting losses placed the true composition within ±0.005 of the nominal value. A rod-shaped sample with rectangular cross-section, of approximate dimensions 8.0 mm × 0.4 mm × 0.4 mm, was spark cut from the ingot, encapsulated in a quartz tube under a partial argon atmosphere, annealed for four days at 1160 °C, and then quenched rapidly into water. The magnetization measurements were performed with a superconducting quantum interference device (SQUID) susceptometer, which has been described in detail elsewhere in the literature [15].

3. Data analysis and interpretation

The principal features of the magnetic phase diagram of polycrystalline $Cr_{1-y}Fe_y$ are well established [16]: for Fe concentrations $y < 0.16$ the system is an itinerant antiferromagnet, with a crossover from incommensurate to commensurate spin density wave order above $y \simeq 0.023$, a spin glass in the narrow range between $0.16 < y < 0.19$, and a ferromagnet for $y \geqslant 0.19$, with a low temperature 'reentrant' phase between $0.19 < y < 0.25$. The composition $Cr_{79}Fe_{21}$ is thus expected to be a ferromagnet with sequential transitions, and measurements of the temperature dependence of the static magnetization under both field-cooled (FC) and zero field-cooled (ZFC) conditions, shown in figure 1, are indeed consistent with the anticipated [16] paramagnetic–ferromagnetic transition near $T_c \simeq 70$ K, and with the possibility of a ferromagnetic collapse below about $T_R \simeq 25$ K (vertical arrows in figure 1). Below T_c, the FC and ZFC curves bifurcate, indicating the presence of a significant viscous component in the magnetization throughout the ordered phase.

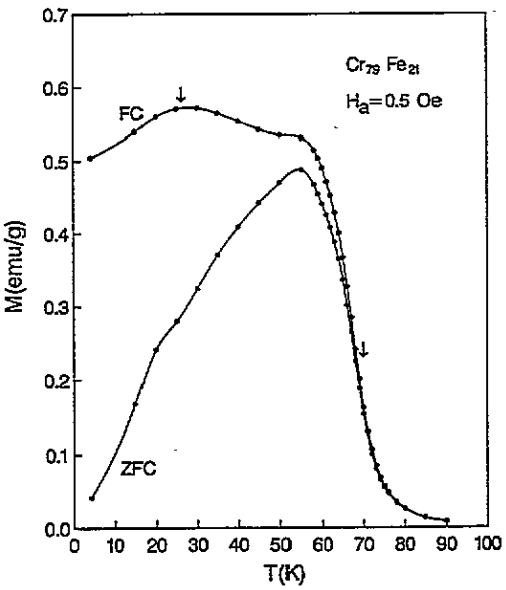

Figure 1. The temperature dependence of the static magnetization $M(T)$ of $Cr_{79}Fe_{21}$ measured under both FC and ZFC conditions in an applied field $H_a = 0.5$ Oe.

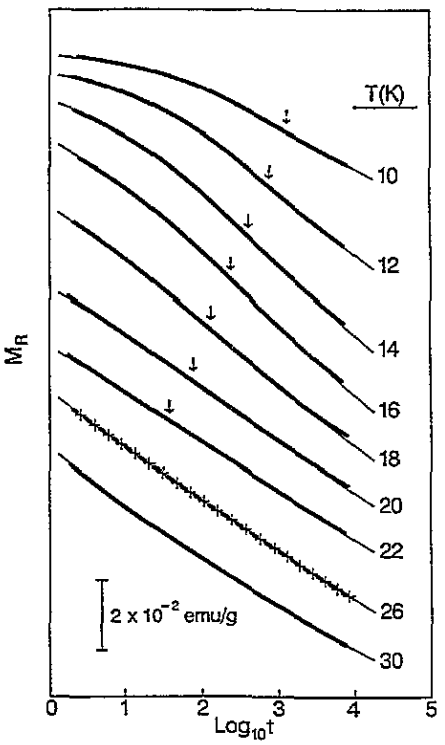

Figure 2. *Thermoremanent relaxation isotherms (solid circles ●) at various temperatures $T < T_c$, all corresponding to a wait time $t_w = 60$ s. The vertical arrows mark the inflection points, and the solid curves are fits to theoretical expressions (2) and (3) discussed in the text. The crosses (+) at $T = 26$ K correspond to a wait time of $t_w = 7200$ s, and illustrate the absence of aging above T_g.*

The relaxation response of the system was probed by cooling the sample in an applied field $H_a = 1.0$ Oe from a reference temperature $T_{ref} = 90$ K in the paramagnetic regime to the measurement temperature $T < T_c$ (cooling times were essentially temperature independent and typically $t_c \simeq 10$ m), waiting for a time t_w at constant T, then abruptly reducing the field to zero and recording the decay of the thermoremanent magnetization over four decades of observation time $1 \text{ s} < t \leqslant 10^4$ s. Figure 2 shows a sequence of relaxation isotherms measured at various temperatures T in the ordered phase, all corresponding to a common wait time $t_w = 60$ s, which was the shortest experimental time required to achieve thermal stability. In spite of their common experimental age $t_a = t_c + t_w$, the isotherms exhibit a systematic change in curvature, from concave down to concave up with increasing temperature, and, at intermediate temperatures, an inflection point (vertical arrows in figure 2) which shifts monotonically from longer to shorter observation times as T increases. Isotherms for $T \leqslant 25$ K are all characterized by a pronounced wait time dependence symptomatic of non-equilibrium dynamics. The age dependence of the response is illustrated in figure 3 for a representative measurement temperature $T = 14$ K and for a sequence of five wait times t_w. Figure 3(b) shows that the maximum in the relaxation rate $S(t) = -\partial M_R(t)/\partial \ln t$ (or equivalently, the inflection point in $M_R(\log t)$) propagates systematically towards longer observation times as the system ages. For $T > 25$ K, the relaxation isotherms are uniformly concave up and the relaxation dynamics

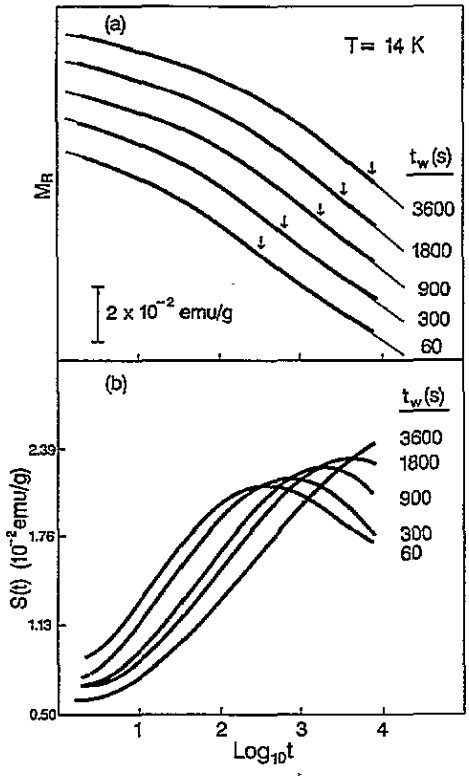

Figure 3. (a) The wait time (t_w) dependence of the thermoremanent decay at a typical temperature $T < T_g$. The vertical arrows mark the inflection points, and the solid curves are best fits to (2). (b) The relaxation rate $S = -\partial M/\partial \ln t$ for the isotherms in part (a).

become 'stationary', in the sense that the thermoremanent decay exhibits no experimentally detectable age dependence for all $t_w \leqslant 10^4$ s (compare the solid circles and the crosses at $T = 26$ K in figure 2, which correspond to $t_w = 60$ s and $t_w = 7200$ s, respectively). Thus $Cr_{79}Fe_{21}$ features a dynamic crossover from a high-temperature regime of equilibrium dynamics, which is roughly coincident with the ferromagnetic phase, to a low-temperature regime of glassy, non-equilibrium dynamics, where the relaxation response is not unique, and where the system becomes 'stiffer' and responds more sluggishly to an excitation with increasing age (in much the same manner as amorphous polymers [2], which become elastically less responsive and more 'brittle').

Evidence for the glassiness of the low temperature 'reentrant' phase extends to more complicated thermal protocols. If the sample is cooled in a field from T_{ref} to $T < T_g$, aged for $t_w = 10^4$ s at T, and then thermally cycled from $T \rightarrow T + \Delta T \rightarrow T$, which consumes typically $t_{cycle} \simeq 200$ s, after which the field is immediately reduced to zero, the relaxation rate $S(t)$ acquires multiple structure, as illustrated in figure 4. When $\Delta T = 0$, S has a single maximum at $t_{m1} \simeq t_w = 10^4$ s. However, increasing ΔT systematically suppresses this maximum, and, simultaneously, creates a second maximum in the vicinity of $t_{m2} \simeq t_{cycle} = 200$ s, which eventually dominates the first. This capacity to interrupt and then reverse the aging process with small *positive* temperature shifts appears to be another constitutive feature of glassy materials [2]. In spin glasses, it has been attributed [17] to a fundamental thermal fragility of the spin–spin correlations in the ordered state, which causes

some of the larger domains (more precisely, those which exceed a certain characteristic length scale which diverges inversely with ΔT) to fracture into small domains, leading to a coexistence of two aging states.

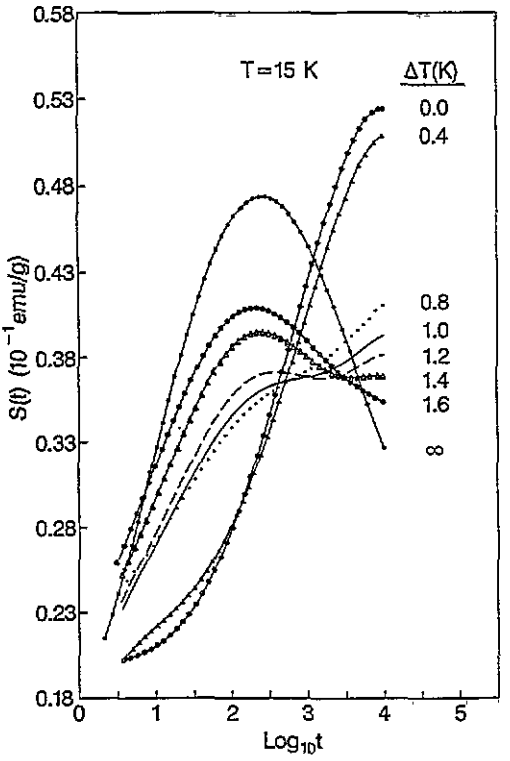

Figure 4. The relaxation rate S measured after cooling in a field $H_c = 1.0$ Oe to $T = 15$ K, waiting for $t_{w1} = 10^4$ s, cycling from $T \rightarrow T + \Delta T \rightarrow T$, with $t_{cycle} \simeq 200$ s, and then removing the field (which defines $t = 0$). $\Delta T = \infty$ corresponds to warming above T_g.

Table 1. Best fit parameters to equation (2) for $T < T_g$.

T (K)	t_w (s)	m_0 (10^{-2} emu g^{-1})	γ	x
10	60	44.1±0.2	0.012±0.003	0.46±0.02
12	60	44.2±0.2	0.019±0.003	0.55±0.02
14	60	42.9±0.2	0.023±0.002	0.72±0.02
14	300	42.9±0.2	0.026±0.002	0.79±0.02
14	900	43.9±0.2	0.027±0.002	0.78±0.02
14	1800	44.8±0.2	0.028±0.002	0.78±0.02
14	3600	42.9±0.2	0.029±0.002	0.77±0.02
16	60	43.5±0.2	0.024±0.002	0.82±0.02
18	60	47.0±0.2	0.021±0.002	0.91±0.01
20	60	52.1±0.2	0.015±0.002	0.92±0.01
22	60	45.6±0.2	0.018±0.002	0.93±0.01

The relaxation isotherms above and below the crossover temperature $T_g \simeq 25$ K are analytically distinct. However, both regimes are accommodated quite naturally within the

random trap formalism: (a) isotherms for $T < 25$ K are all compatible with equation (2), and the solid curves in figures 2 and 3 are the best fits to this expression, with the best fit parameters m_0, γ and x listed in table 1, and with t_w assigned its *experimental* value. The shape of the curves is parametrized exclusively by x, which is less than unity and increases monotonically at $T \to T_g$. Its physical significance will be discussed shortly. All three fitting parameters are only weakly dependent on system age, thus lending considerable credibility to the proposed aging mechanism, which is manifested explicitly through a single, experimentally-defined parameter t_w. The systematic deviations apparent in some of the isotherms at long observation times, particularly in younger versions of the system ($t_w < 300$ s) where the data tend to exhibit more curvature than the theoretical expression, are qualitatively similar to those encountered in pure spin glasses, and may be a consequence of some of the simplifying approximations (such as the assumption of constant $G(\tau/t_w)$ in equation (10) of reference [12], or perhaps the specific form of the cut-off function $\exp(-t/\tau)$ introduced in equation (9) of the same reference). Nevertheless, the essential features of the time dependence of the thermoremanent decay in the low-temperature glassy phase, including its variation with temperature and system age, are replicated remarkably well by a picture in which ergodicity is broken by a *divergent* mean trapping time, which prevents a system of *finite* experimental age t_w from exploring *all* of the available configuration space. (ii) For temperatures $T > 25$ K, the relaxation isotherms are accurately described by the power law decay in equation (3), superposed on a constant baseline, $m(t) = m_0' t^{1-x} + c$, as shown by the solid curves in figure 2. An inspection of the best fit parameters in table 2 shows that x is greater than unity, and increases monotonically with increasing temperature. Thus, the relaxation response in the high-temperature phase, where the dynamics are observed to be *stationary* (age-independent), is analytically consistent with the model prediction in the extreme equilibrium limit where the system ergodically probes a phase space for which the deepest traps of any significance are $\tau_{max} \ll t_w$, due to the form of the trapping distribution.

Table 2. Best fit parameters to $m(t) = m_0' t^{1-x} + c$ for $T > T_g$.

T (K)	t_w (s)	c (10^{-2} emu g^{-1})	x
26	60	1.1±0.3	1.02±0.01
26	7200	1.1±0.3	1.02±0.01
30	60	12.4±0.2	1.04±0.01

The preceding analysis offers new insight, from a dynamical perspective, into the phenomenon of sequential transitions in ferromagnets, as well as empirical support for a recent model of glassy dynamics based on a mechanism of anomalous diffusion in a disordered energy landscape. More specifically: (*a*) strongly bond-disordered ferromagnets like $Cr_{79}Fe_{21}$ ((and also $Ni_{76.5}Mn_{23.5}$ [18] and $(Fe_{0.65}Ni_{0.35})_{88.2}Mn_{11.8}$ [19]), which are close to the percolation threshold for ferromagnetic stability, possess a characteristic temperature $T_g < T_c$, below which the relaxation processes responsible for reconfiguring the spin structure become so slow that the system falls 'permanently' out of thermal equilibrium, in much the same manner as a pure spin glass below the freezing temperature T_f, or a supercooled liquid below the structural glass temperature. In $Cr_{79}Fe_{21}$, $T_g \simeq 25$ K and, in all systems where this effect has been observed, T_g lies close to (but perhaps slightly below) the temperature where the magnetic response function $\chi(T)$ shows the abrupt decline customarily identified with the onset of the 'reentrant' phase. (It should be noted, however,

that the location and articulation of this 'breakpoint' in $\chi(T)$ depend very much on sample geometry, or equivalently, on the demagnetizing factor.) The appearance of these anomalous relaxation dynamics below T_g, with all the attendant aging and thermal cycling effects (figures 3 and 4), is compelling evidence that 'reentrancy' is indeed linked to the formation of a glassy spin structure, where many energetically equivalent spin configurations compete to produce high metastability, although the 'hidden' interactions responsible for this 'spin collapse' have still not been identified [20]. (*b*) The power law distribution (1) for the lifetimes of the metastable states follows directly from an exponential distribution of free energy wells, $P(f) = (x/T)\exp[x(f-f_0)/T]$, which is a characteristic of both the standard SK model [14] and the REM [13] (f_0 is a reference level). Thus the parameter $x(T)$ provides information on the structure of phase space. In the REM, $x = T/T_g$, the energy landscape is temperature independent, and configuration space consists of many completely uncorrelated, but perfectly frozen metastable valleys at T_g, while, in the standard SK model, x has a non-trivial temperature dependence and decreases towards zero as $T \to T_g$, meaning that only a relatively *few* states out of the many available in Parisi's hierarchical replica symmetry breaking scheme [21] *dominate* the properties of the system in this limit. In $Cr_{79}Fe_{21}$, x *increases* with temperature throughout the glassy phase ($T < T_g$), thus offering some preliminary support for the REM picture, although the temperature dependence is *not* linear, in agreement with the recent observations of Bouchaud *et al* [22]. (More specifically, $x \propto T^{1.2\pm0.1}$ for $T < 18$ K, and then flattens abruptly above 18 K.) (*c*) Above 25 K, the data are no longer compatible with the extreme non-ergodic expression in (2) and, more importantly, *aging ceases to be observable experimentally*. However, the random trap formalism is capable of replicating the time dependence of the decay in *this* regime as well, provided that x is allowed to *exceed* unity. Moreover, this is precisely the condition which reduces aging effects to negligible proportions in the model, and which guarantees an equilibrium relaxation response for *any macroscopic* wait time $t_w \gg \tau_0 \sim 10^{-12}$ s. We emphasize that this situation is physically quite distinct from the scenario of 'interrupted aging' for $x < 1$, according to which equilibrium is achieved only when the wait time is long enough to exceed an ergodic time $t_{erg} \sim \tau_0 S^{1/x}$, where S is the total number of metastable states [12, 22]. Although the model offers little guidance concerning the types of frozen spin configurations which are expected to yield an energy landscape with $x > 1$, the current analysis, coupled with the observation of 'stationary' power law decay, with similar exponents, in the thermoremanence of 'good' (non-reentrant) random-exchange ferromagnets like $Pd_{0.986}Fe_{0.014}$ [23], suggests that this may be a constitutive feature of a state with predominantly ferromagnetic order. (*d*) Finally, we note that the dichotomy in the relaxation response of reentrant $Cr_{79}Fe_{21}$, which we have attributed to a fundamental change in the energy landscape as x crosses unity, is also apparent in other methods of analysis commonly employed in connection with structural relaxation in glassy materials. For example, if all the data in figure 2 are recast, in the form of a scaling plot, as a function of $T\ln(\nu t)$, where ν is a temperature-independent attempt frequency [24], then some rough degree of 'universality' can only be achieved among the *low* temperature isotherms $T < T_g$, with a value of $\nu \simeq 10^5\ s^{-1}$ which is comparable to that quoted for structural relaxation [24], while the high temperature isotherms $T > T_g$ have shapes which are clearly inconsistent with the 'universal' curve, and also clearly fail to extrapolate into the low temperature 'cluster'.

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