

## RIGIDITY AND MEMORY IN A SIMPLE GLASS

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### MOTIVATION

Why is a glass rigid despite its amorphous structure? In a rigid system, a local disturbance leads to a bulk response. For example, application of a small force to one end of a crystal will lead to its collective motion; energetically it is favorable for its constituent atoms to maintain their relative positions. By contrast a similar force applied to a liquid will lead to local rearrangements, and all memory of its prior configuration will be soon forgotten. Rigidity implies slow relaxation processes, long time-scales and thus long-range temporal correlations which are usually associated with long-range spatial ordering. So why does a glass have mechanical properties at all similar to those of a crystal?

As a point of reference, let us consider the response of an ideal crystal to application of shear. We note that its low-temperature state is uniquely defined (modulo transformations of the crystal group). The system is cooled into its crystalline phase and after a waiting time,  $t_w$ , a shear deformation is applied. For all practical measurement times,  $t_m = t_w + t$ , it recovers its initial state; it displays perfect memory of its original configuration independent of specifics associated with the shearing and measurement processes.

At low temperatures a crystal has long-range spatial order. By contrast the low-temperature structure of glasses is determined by the incompatibility between local and global ordering, and is *not* unique on any spatial scale.<sup>1, 2</sup> Though such frustration certainly plays a key role in glass formation, purely structural approaches do not address the thermal or dynamical aspects of glassy behavior. For example, unlike a liquid or a crystal, the low-temperature state of a glass is dependent on its sample history. Therefore the response of a glass to shear is not simple. First of all, its initial low-temperature state is a function of sample history; the number of such configurations

Table I.

	Liquid $\rightarrow$ Solid	Metal $\rightarrow$ Superconductor
Conductivity Equation	$\Pi = \eta \dot{u}$	$J = \sigma E = \sigma \dot{A}$
Drude Relation	$\eta = \eta_0 \frac{1}{1 - i\omega\tau}$	$\sigma = \frac{ne^2\tau}{m} \frac{1}{1 - i\omega\tau}$
London Kernel	$\Pi = Gu$	$J = -QA$
Kubo Relation	$G = -i\omega\eta$	$Q = -i\omega\sigma$

scales exponentially with the number of sites. Furthermore the response is dependent on both  $t_w$  and  $t_w + t$ , and not simply on their difference,  $t$ . On short time-scales ( $t < t_w$ ), a glass exhibits perfect memory of its original configuration; by contrast on longer ones ( $t \gg t_w$ ) a glass “flows” and thus forgets its past, a phenomenon often referred to as “ageing”.

Maxwell was one of the first physicists to address the crossover between viscous and elastic behavior in supercooled liquids.<sup>3</sup> Conceptually his treatment is amusingly similar to later studies of the metal-superconductor transition;<sup>4</sup> it also provides a nice study of “generalized rigidity”, an idea that has been emphasized in conjunction with broken symmetry by Anderson.<sup>5</sup> The analogy is presented in Table I. In a metal the flow equation is Ohm’s Law: the current ( $J$ ) is proportional to the field ( $E$ ); alternatively the flow of charge is proportional to the time derivative of the vector potential ( $\dot{A}$ ). Analogously in a liquid the shear ( $\Pi$ ) is proportional to the time derivative of the strain ( $\dot{u}$ ); the shear can be viewed as the flow of momentum. At finite frequency charge currents in a simple metal relax on a characteristic time-scale  $\tau$  leading to a Drude relation for the conductivity  $\sigma$ ; momentum currents in a liquid decay in a similar fashion with an analogous relation for the viscosity  $\eta$ . Then, for example at a particular temperature, this characteristic time-scale  $\tau$  diverges and the system displays rigidity. In the resulting solid, the shear is proportional to the strain. In the superconductor the supercurrents are proportional to the vector potential as described by the London equation. In the superconductors and the solids, the rigidity is associated with the development of supercurrents of charge and momentum respectively. The analogue of the London kernel in the superconductor is the shear modulus, and there is a Kubo relation for both of these quantities.

At this point we have only discussed the development of a solid, but have not distinguished between a crystal and a glass. This distinction is summarized in Table II. As we have already mentioned above, a crystal breaks translational symmetry and

Table II.

	Glass	Crystal
Long-Range Spatial Ordering	No	Yes
Number of States	$\sim \exp N$	$O(1)$
Fluctuation-Dissipation Theorem	Violated	Obedyed
Memory Effects	$\lim_{t \rightarrow \infty} \lim_{t_w \rightarrow \infty} G_{t_w+t, t_w} \neq 0$	
Ageing	$\lim_{t \rightarrow \infty} G_{t_w+t, t_w} = 0$	$\lim_{t \rightarrow \infty} G_{t_w+t, t_w} \neq 0$ (fixed $t_w$ )

has long-range spatial ordering; by contrast there is *not* any such simple long length-scale associated with a glass. A crystalline state is unique up to transformations of the crystal group. In a glass, there is no preferred order so that local changes can lead to new metastable states. As a result, the number of states in a glass scales exponentially with the number of sites in the system. Characterization of the low-temperature glassy state then becomes rather difficult. Conventionally, for example in a crystal, one applies a small symmetry-breaking field to restrict one's statistical trace to the states in a single well. Then, using this restricted trace, one can proceed with equilibrium statistical mechanics as usual in the low-temperature phase. However because there are an exponential number of states in a glass, it is not possible to distinguish different metastable configurations by application of a simple field. As a result one cannot determine properties of the glass using a conventional Gibbs approach; more specifically, one cannot employ the fluctuation-dissipation theorem.

As previously mentioned, the similarities and differences between a glass and a crystal are best displayed by considering the behavior of the shear modulus (e.g. Table II). The memory of a glass improves with increasing waiting time ( $t_w$ ); indeed if one takes the waiting time to infinite first and then lets the measurement time ( $t_w + t$ ) diverge it has perfect memory just like a crystal. Here the order of limits is crucial. By contrast for fixed waiting time in the limit of infinite measurement time, a glass loses its rigidity; by contrast the response of a crystal is invariant under time-translation, and thus its memory is perfect and independent of the order of limits associated with  $t_w$  and  $t$ .

In a nutshell, glassiness is solidification without crystallization. It occurs in a



large number of materials of diverse microscopic character; therefore one feels that there should exist an underlying reductionist picture, the analogue of the Ising model for second-order phase transitions, which has not yet been found. In particular, how is a system simultaneously out-of-equilibrium and stable on very long (sometime millenium) time-scales? How does one keep an intrinsically non-random system away from its thermodynamic ground-state? There has been tremendous technical progress in our theoretical understanding of disordered models; is it possible to link "dirty" and "clean" glasses? There have been several studies of such mappings, which seem to exist, and we refer the interested reader to an excellent review on this subject.<sup>6</sup> Here the strategy is similar to that associated with energy level spectra in complex nuclei, where progress was made through studies of random Hamiltonians. However in what follows, we shall discuss a complementary approach: a direct study of a specific non-random system that displays glassiness. We have taken a somewhat "Galilean" approach to the glass problem: we have abstracted it to a point that we can manage, hoping to have gleaned its essential features. Then we have characterized it with the available tools at our disposal to check whether it has the desired properties. Finally we have tried to make contact with experiment by making predictions for measurements to check whether we are on the right track! In the next section we hope to give the reader a flavor of the model of study and for our main analytic results. Next we'll discuss some early experiments on this "simple glass" and some predictions for measurements that we'd like to see. We'll mention briefly how this "Galilean" system, with both memory and a large number of states, can be used for information storage. Finally we'll end with a summary and everpresent open questions which should be addressed in the future.

## A "GALILEAN" MODEL FOR A SIMPLE GLASS

In the spirit of the Maxwellian analogy between liquids and metals discussed above, we have studied the glass transition in a long-range superconducting array. The physical system of study is a stack of two mutually perpendicular sets of  $N$  parallel thin wires with Josephson junctions at each node (Figure 1) that is placed in an external transverse field  $H$ . The thermodynamic variables of this array are the  $2N$  quantum mechanical phases of the superconducting order parameter associated with each wire; these play an analogous role to that of the atomic positions or density waves in a real glass. We note that in this network each horizontal/vertical wire is coupled to each vertical/horizontal one by a Josephson junction, so that in the thermodynamic limit ( $N \rightarrow \infty$  for fixed array area) the number of neighbors diverges.

The energy scales associated with this long-range array are associated with the individual superconducting wires and with the Josephson junctions. In the absence of a field the macroscopic phase of a superconductor is constant in equilibrium, assuming that its thickness  $l$  is larger than the coherence length  $\xi_0$  so that phase slips are energetically unfavorable. Application of a field results in the precession of this macroscopic phase, where the rate of precession is determined by the amplitude of the applied field. In the absence of a field the phase differences would be zero at each Josephson junction, but this is no longer possible at finite  $H$ . Thus frustration in this network can be "tuned" by application of a transverse field; the system is then overconstrained since there are  $2N$  phases and  $N^2$  Josephson junctions with competing energetic requirements.

Here we assume that the Josephson couplings in the array are sufficiently small so that the induced fields are negligible in comparison with  $H$ ; we shall return to this issue when discussing the experimental realization of this network. We can therefore

describe the array by the Hamiltonian

$$\mathcal{H} = -J \sum_{jk} \cos \left( \phi_j - \phi_k - \frac{2\pi\Phi_{jk}}{\Phi_0} \right) = \text{Re} \sum_{jk} s_j^* J_{jk} s_k \quad (1)$$

with  $1 \leq (j, k) \leq N$  where  $j(k)$  is the index of the horizontal (vertical) wires and  $\Phi_0$  is the flux quantum; the effective spins  $s_k = e^{i\phi_k}$  where the  $\phi_k$  are the superconducting phases of the  $2N$  wires. The couplings are site-dependent and are related to the enclosed flux at a given node such that

$$J_{jk} = J \exp \frac{2\pi i \Phi_{jk}}{\Phi_0} = J \exp \frac{2\pi \alpha k}{N} \quad (2)$$

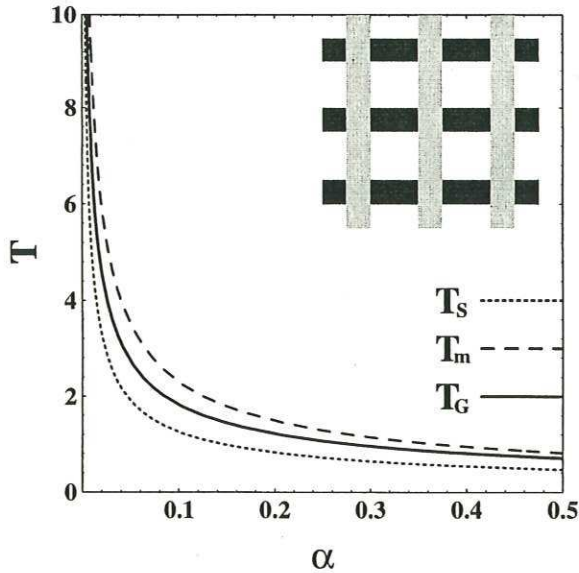
where we have introduced the flux per unit strip  $\alpha = \frac{NHl^2}{\Phi_0}$ , with  $l$  is the inter-node spacing; here we note that the sign of the coupling depends on the enclosed flux and can be both positive and negative.

Because every horizontal (vertical) wire is linked to every vertical (horizontal) wire, the number of nearest neighbors in this model is  $N$  and it is accessible to an analytic mean-field treatment. For the same reason, the free energy barriers separating its low-temperature solutions scale with  $N$ . This situation is in marked contrast to that in conventional 2D arrays where the coordination and hence the barriers are low.<sup>7</sup> A similar long-range network with positional disorder was studied previously.<sup>8</sup> For  $\alpha \gg 1/N$  that system displays a spin glass transition which was mapped onto the Sherrington-Kirkpatrick model<sup>9</sup> for  $\alpha \gg 1$ ; in this field regime there is no residual "ferromagnetic" phase coherence between wires. Physically this glassy behavior occurs because the phase differences associated with the couplings  $J_{jk}$  acquire random values and fill the interval  $(0, 2\pi)$  uniformly for  $\alpha \gg 1/N$ . More specifically, there will be no commensurability if the sum

$$\sum_{k=1}^N J_{jk} J_{kl}^\dagger = J_0^2 \sum_{k=1}^N \exp 2\pi i \left\{ \frac{\alpha k(j-l)}{N} \right\} \quad (3)$$

is smooth where  $\{j, l\}$  ( $k$ ) are indices labelling horizontal (vertical) wires; this will occur only if the expression in curly brackets on the r.h.s. of (3) is *not* an integer, a condition always satisfied for the disordered array. For the periodic case, this situation is realized in the "incommensurate window"  $1/N \ll \alpha \leq 1$ ; here the phase-ordering unit cell is larger than the system size so that the "crystalline" phase is inaccessible. There are thus no special field values where the number of low-temperature solutions are finite, in contrast to the situation for  $\alpha > 1$ . In the thermodynamic limit of  $N \rightarrow \infty$  (with fixed array area), the high-temperature approach to the glass transition in this system has been studied<sup>10, 11</sup> using a modified Thouless-Anderson-Palmer (TAP) method.<sup>12</sup> Here we discuss the qualitative picture that emerges from these results (cf. Fig. 1), referring the interested reader elsewhere for a more detailed quantitative treatment. As  $T$  approaches  $T_m^+$ , where  $T_m^+ \sim T_0 \approx \frac{J\sqrt{N}}{2\sqrt{\alpha}}$ , there appear a number of metastable states in addition to the paramagnetic free-energy minimum; most likely they are energetically unfavorable and thus do not "trap" the system upon cooling from high temperatures. As  $T \rightarrow T_G^+$ , the paramagnetic minimum is "subdivided" into an extensive number of degenerate metastable states separated by effectively infinite barriers, and the system is dynamically localized into one of them. Qualitatively, in the interval  $T_m > T > T_G$  there appear many local minima in the vicinity of the paramagnetic state separated by *finite* barriers; these barriers increase continuously and become infinite at  $T = T_G$ . Each of these minima is characterized by a finite "site magnetization"  $m_i = \langle s_i \rangle_T$  where "site"

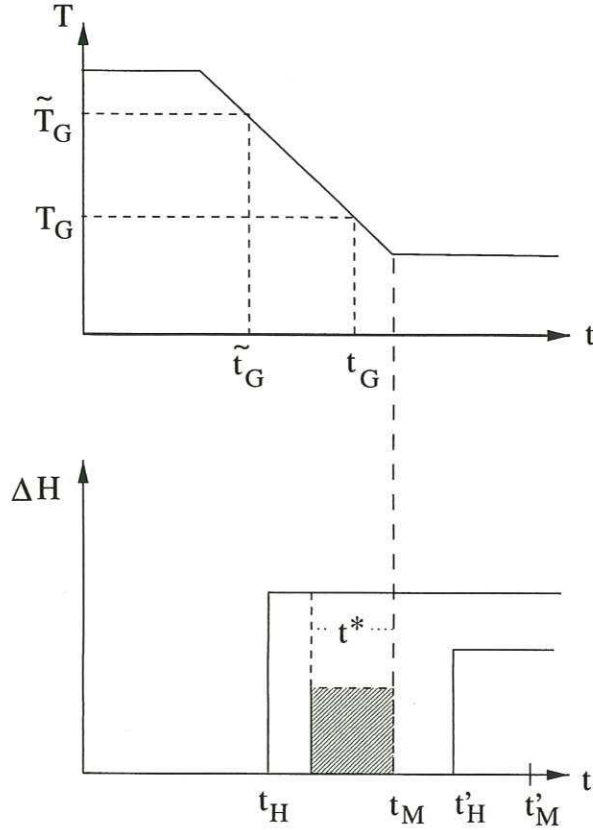




**Figure 1.** The phase diagram of the array (inset) where  $T_G$  indicates the temperature associated with the dynamical instability discussed in the text,  $T_S$  is the speculated equilibrium transition temperature and  $T_m$  is the “superheating” temperature where the low-temperature metastable states cease to exist.

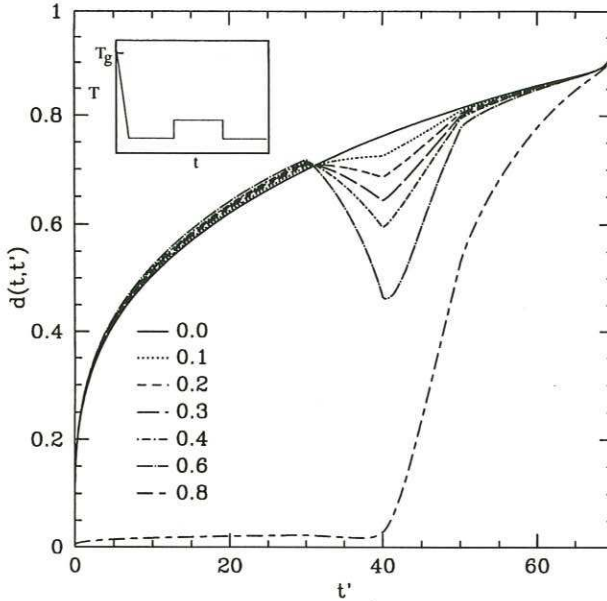
refers to a wire. When  $T > T_G$  thermal fluctuations average over many states so that  $\langle m_i \rangle \equiv 0$ . At  $T = T_G$  the system is localized in one metastable state and there is an associated jump in the Edwards-Anderson order parameter,  $(q = \frac{1}{N} \sum_i \langle m_i \rangle^2)$ . The low-temperature phase is characterized by a finite  $q$  and by the presence of a memory,  $\lim_{t' \rightarrow \infty} \Delta(t, t') \neq 0$  where  $\Delta(t, t')$  is the anomalous response, defined as  $\Delta(t, t') \equiv \langle \frac{\partial m(t)}{\partial h(t')} \rangle$ . At  $T = T_G$ , the metastable states are degenerate and thus there can be no thermodynamic selection. However at lower temperatures interactions will probably break this degeneracy and select a subset of this manifold; then we expect an  $(t \rightarrow \infty)$  equilibrium first-order transition ( $T_S$ ) which should be accompanied by a jump in the local magnetization. In order to observe this transition at  $T_S$  the array must be equilibrated on a time-scale ( $t_E$ ) longer than that ( $t_A$ ) necessary to overcome the barriers separating its metastable states;  $t_A$  scales exponentially with the number of wires in the array. Thus the equilibrium transition at  $T_S$  is observable *only* if  $t_E \rightarrow \infty$  *before* the thermodynamic limit ( $N \rightarrow \infty$ ) is taken; in the opposite order of limits only the dynamical transition occurs.

The periodic array thus exhibits a first-order thermodynamic transition *preceded* by a dynamical instability; the glass transition at  $T_G$  is characterized by a diverging relaxation time and an accompanying jump in the Edwards-Anderson order parameter. In general the dynamical behavior of this network is described by coupled integral-differential equations for the correlation ( $D_{tt'} = \langle s(t)s(t') \rangle$ ) and the response ( $G_{tt'} = \langle \frac{\partial s(t)}{\partial h(t')} \rangle$  where  $t > t'$ ) functions. At temperatures above the glass transition  $T > T_G$  the response and the correlation functions are related by the fluctuation-dissipation theorem and there is time-translation invariance; the resulting equation



**Figure 2.** Schematics of (a) Temperature (T) (b) Applications of an additional field ( $\Delta H$ ) vs. time (t) indicates the time-scales involved in a finite-cooling experiment as discussed in the text;  $\tilde{T}_G$  and  $\tilde{t}_G$  refer to the “effective glass transition” when the system goes out of equilibrium, and  $t_H$ ,  $t_M$  and  $t^*$  are the time-scales associated with the onset of an additional field, a subsequent measurement and ageing.

for the system’s dynamical evolution is structurally similar to that derived for density-density correlations in the mode-coupling approach to the liquid-glass transition.<sup>13</sup> Furthermore the general coupled equations describing the dynamical behavior of this *non-random* network in the regime ( $1/N \ll \alpha < 1$ ) are *identical* to those obtained for the  $p = 4$  (disordered) spherical Potts model.<sup>14</sup> The possible connection between non-random glasses and  $p \geq 3$  (disordered) spherical models has been previously suggested in the literature,<sup>15, 16, 17</sup> and these results give support to those conjectures. Furthermore these long-range Josephson arrays can be built in the laboratory, allowing for parallel theoretical and experimental studies of the “simplest spin glasses” that have previously been an abstraction. Several physical properties of the array have been characterized<sup>18</sup> in its non-ergodic regime ( $T < T_G$ ). We note that the glass transition temperature  $T_G$  described above corresponds to the system going out of equilibrium as it is cooled infinitesimally slowly from high temperatures. In practice any physical cooling process occurs at a finite rate, and the effective glass transition occurs when the system drops out of equilibrium; thus the observed glass transition temperature is a function of the time-scale of the experimental probe. If we define the reduced temperature  $\Theta \equiv \left( \frac{T - T_G}{T_G} \right)$  then the effective glass transition will occur when the time associated with cooling,  $t_c \approx \frac{\Theta}{d\Theta/dt}$  is equal to the relaxation time at that tempera-



**Figure 3.** The rescaled correlation function  $d(t, t')$  for the cooling-heating regime. Here the temperature was reduced infinitely fast to a temperature  $T_i < T_g$  at  $t' = 0$ ; the system was equilibrated for  $0 < t' < 30$ , then heated linearly for  $30 < t' < 40$ , then cooled linearly back to its original temperature  $T_i$  for  $40 < t' < 50$ , and then measured at  $t = 70$ . Different curves correspond to different amplitudes of the heat pulse as shown. We see that for heat pulses of amplitude less than 0.6 the system recovers its original state; however, for larger heat pulses all memory is lost.

ture,  $\tau_R \approx t_0 \theta^{-\nu}$  where  $t_0$  and  $\nu$  are determined from the high-temperature analysis elsewhere.<sup>11</sup> There is evidence for history-dependence; the system's response is very different if an additional field is turned on during or after the cooling process, reminiscent of the zero-field cooled vs. field-cooled susceptibility observed in spin glasses. Furthermore there exists an "ageing" time-scale  $t^*$  this system (cf. Fig. 2); if the field is turned on during the cooling process at time  $t_H$  and a measurement is taken at time  $t_M$  such that  $t_H - t_M < t^*$  the system "remembers" the cooling process; otherwise ( $t_H - t_M > t^*$ ) it "forgets" it completely.

The low-temperature "memory" of the superconducting array is perhaps best exhibited by its response to an applied heat pulse<sup>18</sup>. The correlation function  $D_{tt'}$  as a function of  $t'$ , displayed in Figure 3, exhibits the system's overlap between its state at time  $t$  and time  $t'$  after a fast temperature quench. Heat pulses of varying amplitudes (c.f. Fig. 3) are applied after the quench, and the system's memory is probed. For all but the largest amplitude pulse, the system recovers its original dynamical trajectory indicating the presence of long-range temporal correlations. Such measurements have been performed experimentally on real spin glasses to determine the structure of their low-temperature states.<sup>19</sup> An analogous numerical analysis of the superconducting array indicates that *all* metastable states appear at the glass transition; this absence of further subdivision of states is consistent with analytic work on the structure of states in the  $p = 4$  (disordered) spherical model.<sup>20</sup>



## DISCUSSION

Since the experimental realization of these arrays is important, we would like to identify several assumptions in the theoretical treatment; in order to test its predictions and probe beyond its realm (e.g. finite-size effects) the fabricated system must satisfy certain physical requirements<sup>21, 22</sup> In particular the effects of everpresent screening currents must be minimized to ensure that the external field frustrates the superconducting phases effectively. More specifically the induced flux in the array should be less than a flux quantum so that all fields and phase gradients produced by diamagnetic currents are negligible. The resulting condition on the array limits the number of wires; there is thus a delicate balance between the need for many neighbors (and high free-energy barriers) and an external field that effectively frustrates the phases. Furthermore the model has only been considered in the classical limit where quantum mechanical fluctuations of the phases are negligible. These constraints put some strong restrictions on the choice of array parameters, which have been elaborated elsewhere.<sup>21, 22</sup>

Once the arrays are fabricated to specification, what experiments should be performed? Recently there have been some measurements probing the static properties of such long-range arrays.<sup>22</sup> Commensurability effects were studied in a small network as a function of field; the results were in good agreement with mean-field theory and in particular indicated the absence of long-range spatial phase ordering for field strengths  $\alpha < 1$ . What other experimental measurements would we like to see? History-dependence of the critical current  $j_c$  should be studied to establish the presence of large barriers crucial for the development of glassiness; more specifically the dependence of  $j_c$  on path in the  $T - H$  plane should be investigated. The critical current  $j_c$  should be measured for a large number of thermal cyclings; if the system is glassy it should display a well-defined distribution  $P(j_c)$ . Finally the diverging relaxation time at  $T_g$  should be accessible via the a.c. response to a time-varying field  $H(t)$ ; the associated a.c. susceptibility is

$$\chi_\omega = \frac{\partial M_\omega}{\partial H_\omega} = \frac{C(\alpha)\omega}{\omega + i/\tau_R(T)} \quad (4)$$

where  $\tau_R$  is the longest response of the system that diverges at  $T = T_g$  and  $C(\alpha)$  is to be found elsewhere.<sup>11</sup> The  $\omega \rightarrow 0$  limit of the a.c. susceptibility jumps to a finite value at  $T = T_g$ , indicating the development of a finite superconducting stiffness at the transition. Therefore measurement of this a.c. response in a fabricated array would be a direct probe its predicted glassiness.

This array has long-range temporal correlations (memory) and has an extensive number of metastable states;<sup>23</sup> can it be used for information storage? Indeed high connectivity and nonlinear elements (here the Josephson junctions) are key features required for the construction of associative memories.<sup>24</sup> Here one would like to store  $p$  patterns in such a way that if the memory is exposed to a slightly different one, it produces the stored pattern most similar to it. A simple model for such a memory is based on an array of McCulloch-Pitts neurons.<sup>24</sup> The patterns are stored in couplings which are chosen to minimize an energy function. Each nonlinear element has multiple inputs  $n_i = 0, 1$ ; in the simplest model the couplings can have arbitrary sign and the output is "computed" by each element using the simple formula  $n_i = \Theta(\sum_j J_{ij}n_j)$ . Clearly the output is robust to errors in the input due to the multiple connections present. An array of such artificial neurons is thus content-addressable and fault-tolerant.

It is quite straightforward to adapt the long-range array described above to become a superconducting analog of a McCulloch-Pitts networks.<sup>25</sup> The couplings at each

Josephson node can be “written” by a superimposed array of superconducting quantum interference devices (SQUIDS). The inputs and outputs are rapid single flux quantum (RSFQ) voltage pulses.<sup>26</sup> For each stored pattern there is an input-output dictionary associated with the pulses, and the system would be tolerant to input errors. For an array of  $N = 1000$  wires with internode spacing  $l = 0.5\mu$ , the capacity would be  $C = 0.1N^2 = 10^5$  bits with an access time per bit of  $\tau_a \sim 10^{-12}$  sec. Because the  $N = 1000$  bits in a given pattern could be accessed in parallel, the access time per image could be  $\tau_{DT} = \frac{\tau_a}{N} = 10^{-15}$  seconds. Thus it is a candidate for nonlocal information storage; it is faster but has lower capacity per unit area than the best optical holographic memories.<sup>28</sup>

In conclusion, we have presented a summary of recent work on a periodic “Galilean” glass that displays rigidity in the absence of long-range spatial order. This glass transition is characterized by a diverging relaxation time and an accompanying jump in the Edwards-Anderson order parameter. At temperatures above the glass transition its dynamical equation is similar in structure to that studied in mode-coupling approaches to the liquid-glass transition. More generally, its evolution is described by coupled integral-differential equations which are identical to those of a well-studied disordered model. Preliminary experiments have probed the static structure of the array, indicating the absence of commensurability except for special field values. Predictions for further dynamical measurements have been made, including a suggestion for probing the diverging relaxation time at the glass transition.

Naturally there remain many open questions. How many of the extensive number of metastable states are actually physically accessible? Recent work<sup>27</sup> suggests that a small number of these states have basins of attraction far exceeding the average. If true, this result could have more general implications for other complex systems, particularly for the problem of protein-folding. Quantum effects in this array remain to be studied and could have interesting consequences. The memory discussed above is completely passive; in principle one could construct an adaptation of this array that both remembers and learns. Finally how robust are the properties of this long-range glass to spatial inhomogeneities (finite-size effects)? There is still much to be learned in the study of rigidity, even in the simplest systems.

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