Derivation of a stretched-exponential time relaxation

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Abstract. On the basis of a simple model of time relaxation in disordered systems (such as glasses or spin glasses), we study the diffusion on clusters built on high-dimensional hypercubes. We show there exist two particular concentrations of available sites on the hypercube. The first one defines two distinct behaviours of relaxation: above $p = \frac{1}{2}$, the relaxation is purely exponential; below $p = \frac{1}{2}$, the relaxation is no longer exponential. The other case is of more interest: it is the percolation concentration. Using the unproven but argued assumption that, because of the high dimensionality of the considered space, the calculation may be mapped onto the problem of diffusion on a standard mean-field percolation cluster, we show that the relaxation should follow a stretched exponential law there with exponent $\frac{1}{3}$. We briefly discuss the physical implications of this approach.

1. Introduction

For a long time [1], a broad class of physical systems [2] has been recognised to exhibit non-exponential time relaxation at low temperatures. Dielectric materials [3], polymers [4], glasses [5] and spin glasses [6] show a similar critical slowing down of some pertinent macroscopic quantity (polarity, magnetisation, etc) to its equilibrium value near a critical temperature T_g. These similarities suggest a universal physical origin, related to randomness, which is present in all these materials. Experimentally, most of these relaxations are well fitted by a stretched exponential with time:

$$F(t) \sim \exp[-(t/\tau)^\beta] \qquad (1)$$

where $\beta = 1$ when the temperature is larger than a value T_c, and $\frac{1}{3} \leqslant \beta \leqslant 1$ for $T_g \leqslant T \leqslant T_c$. Some authors (see, for example, [7]) have suggested different mechanisms to explain this empirical result but none of them gives a clear explanation of the observed values of β, and especially of its tending to $\frac{1}{3}$ [8] as the temperature approaches T_g. Recently, Campbell [9] has argued that this phenomenon is related to the topology of the diffusion path of the system in its configuration space. The aim of this paper is to show that, under reasonable assumptions, this argument leads to a stretched-exponential time relaxation of the form (1), with $\frac{1}{3} \leqslant \beta \leqslant 1$. For convenience, we concentrate on Ising spin systems, but the following scheme remains quite general.

Consider a system of N spins $\frac{1}{2}$. Its configuration space can be represented by the vertices of a N-dimensional hypercube. Two states of the system differing from the flipping of one spin are near-neighbouring vertices on the hypercube. The Hamiltonian H, which governs the thermodynamics of the system, allows for associating an energy to each vertex. We call E_0 the lowest energy and $A(T)$ the subset of vertices whose

energies are smaller than $E_0 + \frac{1}{2} N k_B T$. The proportion, p, of configurations which belong to $A(T)$ is indeed a non-decreasing function of the temperature.

The relevant parameter for spin glasses is

$$q(t) = \overline{\langle S(0) S(t) \rangle_T} \tag{2}$$

where $\langle \rangle_T$ means thermal average and the bar means the average over sites. There is a corresponding parameter in the configuration space. To show it, let us first define the Hamming vector r between two configurations α and β: the ith component of r is 0 if the ith spin is in the same state in both configurations and 1 if one is $+\frac{1}{2}$ and the other $-\frac{1}{2}$. So r^2 is just the minimum number of spins which have to be flipped to change from configuration α to configuration β. If $S_{i,\alpha}$ is the state of the ith spin in the configuration α, we have exactly $N - r^2$ products $S_{i,\alpha} S_{i,\beta}$ equal to $+\frac{1}{4}$, and r^2 products equal to $-\frac{1}{4}$. Imagine now that the system is prepared in any state at time $t = 0$. As the time increases, its representative point in the configuration space jumps from one vertex to other. The jump probability is simply related to the Boltzmann factor: $\exp(-\Delta E / k_B T)$, where ΔE is the difference in energy between two neighbouring sites (here, we assume that just one spin can flip at the same time). This kind of random walk is thermally averaged by the choice of the jump probability factor, so that one can write

$$q_N(t) = \frac{1}{4} \left(1 - \frac{2}{N} \langle r^2(t) \rangle \right). \tag{3}$$

$r^2(t)$ is now the average value of the Hamming distance r^2 over all possible walks between the configuration at time t and the initial time. Moreover $\langle \rangle$ means the average over the quenched disorder of interactions between spins. In practice, we are neither able to calculate the exact distribution of energies on the hypercube of configurations, nor, of course, the statistical weight of all the walks for a given Hamiltonian. Therefore, some drastic simplifications are needed to pursue the calculation. Though the approximations are quite crude, this approach is still based on physical grounds because the result seems to be rather universal.

In this way, we suppose that energies are randomly distributed on the hypercube, according to some probability law: $\rho(E)$. Moreover, we approximate the Boltzmann factor by 1 if the two vertices belong to $A(T)$, and 0 if not.

Let us now introduce some notation: the N-dimensional hypercube is called Q_N and the initial configuration at $t = 0$ is called O. We suppose that O belongs to $A(T)$ for simplicity, but this is not important. Finally, we call $P_Q(r, t)$ the probability that the representative point of the system is at distance r from O at time t.

So the quantity to be calculated is

$$\langle r^2(t) \rangle = \left\langle \sum_{r \in A(t)} r^2 P_Q(r, t) \right\rangle. \tag{4}$$

2. Relaxation for $\frac{1}{2} < p \leqslant 1$

For each direction i $(1 \leqslant i \leqslant N)$, we define the subset $A_i(T)$ of vertices which belong to $A(T)$, and whose vector r is such that $r_i = 1$. Symmetrically, we define the set $\bar{A}_i(T)$ of vertices which belong to $A(T)$, with $r_i = 0$. Since p is supposed to be strictly larger than $\frac{1}{2}$, the two disjoint subsets A_i and $\bar{A}_i$ are non-empty, and their reunion is $A(T)$.

The definition of $r^2(t)$ is

$$r^2(t) = \sum_{i=1}^{N} \sum_{r \in A_i(T)} P_Q(r, t).$$ (5)

This can be seen as the sum of the temporal evolution of the quantities:

$$r_i^2(t) = \sum_{r \in A_i(T)} P_Q(r, t)$$ (6)

which is just the behaviour of the random walk projected on an edge of Q_N.

If n_i is the number of sites which belong to $A_i(T)$, and m_i the number of links connecting sites of $A_i(T)$ to sites of $\bar{A}_i(T)$, the characteristic jump time from $r_i = 0$ to $r_i = 1$ can be written as

$$\frac{1}{\tau_{0,i}} = \frac{1}{N} \frac{m_i}{n - n_i}$$ (7)

where $n = p2^N$. Similarly, the characteristic jump time from $r_i = 1$ to $r_i = 0$ is such that

$$\frac{1}{\tau_{1,i}} = \frac{1}{N} \frac{m_i}{n_i}.$$ (8)

Then we have

$$r_i^2 = \frac{\tau_{0,i}}{\tau_{0,i} + \tau_{1,i}} \left\{ 1 - \exp\left[-\left(\frac{1}{\tau_{0,i}} + \frac{1}{\tau_{1,i}} \right) t \right] \right\}.$$ (9)

The problem is solved if we are able to average this quantity over all possible couples (m_i, n_i), with the two constraints: vertices belong to Q_N and $n = p2^N$ (where p is fixed).

When one disposes n sites on the vertices of Q_N, the probability that n_i sites belong to a given $(N-1)$-dimensional hypercube is just a hypergeometric law centred in $n/2$. Once one knows that $A_i(T)$ contains exactly n_i sites, the probability of constructing m_i bonds between sites of $A_i(T)$ and sites of $\bar{A}_i(T)$ is also given by a hypergeometric law, centred in $n_i(n - n_i)/2^{N-1}$.

More precisely:

$$\text{prob}[n_i; m_i] = \left[\binom{2^{N-1}}{n_i} \binom{2^{N-1}}{n - n_i} \binom{2^N}{n}^{-1} \right] \left[\binom{n - n_i}{m_i} \binom{2^{N-1} - n + n_i}{n_i - m_i} \binom{2^{N-1}}{n_i}^{-1} \right]$$ (10)

with the constraints

$$\max\{0, (2p - 1)2^{N-1}\} \leq m_i \leq \min\{n_i, n - n_i\}.$$ (11)

Now, we can write $q_N(t)$ in the more pleasant form:

$$q_N(t) = \left\langle \frac{1}{2n} \sum_{n_i, m_i} \text{prob}[n_i; m_i] n_i \exp\left(-\frac{m_i}{n_i(n - n_i)} \frac{nt}{N} \right) \right\rangle$$ (12)

with the constraints (11).

Replacing prob$[n_i; m_i]$ by a product of two normal laws allows us to find $q_N(t)$ as

$$q_n(t) \sim \tfrac{1}{4} \exp\left(-2p\frac{t}{N}\right) \exp\left(\frac{4}{2^N}\frac{(1-p)^2 t^2}{N^2}\right)$$

$$\times \left(1 + \frac{8}{2^{2N}}\frac{(2p-1)(1-p)}{p}\frac{t^2}{N^2}\right)^{-1/2} \qquad \text{for } t < \frac{N2^N}{4p}. \qquad (13)\dagger$$

In practice (this means, in experiments or numerical simulations), one has $t \sim N \ll N2^{N/2}$, so that formula (13) can be well approximated by

$$q_N(t) \sim \tfrac{1}{4} \exp(-2pt/N) \qquad (14)$$

for $\tfrac{1}{2} < p < 1$. This formula remains valid for $p = 1$, as can be easily seen from equation (12). This result (14) is not very surprising since it just corresponds to the most probable value of exponent

$$\frac{m_i}{n_i(n-n_i)}\frac{nt}{N}$$

appearing in (12). It agrees quite well with numerical simulations published earlier [10]‡. For $p \leq \tfrac{1}{2}$, the failure of the method comes from the non-zero probability that n_i vanishes. In this case, the method has obviously no more meaning.

3. Relaxation at the percolation threshold

In our model, two values of p play a particular role: $p = \tfrac{1}{2}$, above which the relaxation is exponential, and $p = p_c = 1/N$, which is the percolation threshold in site percolation on the hypercube [10]. These values arise from graph theory and the main results about the site percolation problem on a hypercube are summarised below [12].

   (a) $\tfrac{1}{2} < p \leq 1$: there is almost surely a single cluster, the size of which is obviously $p2^N$.

   (b) $1/N < p < \tfrac{1}{2}$: there is almost surely a main cluster of size of order $p2^N$, and many small clusters, the largest of which has almost surely a size of order $\{\log_2[1/(1-p)]\}^{-1}$.

   (c) $p < 1/N$: the largest cluster is asymptotically of size N.

   For spin glasses, it is tempting to associate these two thresholds, respectively, to the appearance of Griffiths singularities [13] and to the spin-glass transition.

   If this assumption is true, a stretched exponential (1) with $\beta = \tfrac{1}{3}$ is expected at the percolation threshold. This is the aim of the following derivation. The idea of the analytical calculation consists in mapping a random walk on the hypercube into a random walk in the whole space of the same Euclidean dimension. This is made possible by assuming that, at each Monte Carlo step, the random walker chooses at

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† If we take $\tau \sim 10^{-13}$ s as the elementary jump time, this limit is obtained for $t \sim 10^{12}$ yr with an assembly of 100 spins! This limit is even larger for macroscopic systems. For times larger than this limit, one finds

$$q_N(t) \propto \exp\left(-\frac{2}{p}(2p-1)\frac{t}{N}\right).$$

‡ In the case $p = 1$, an exact result may be found for discrete time. One finds [11]

$$q_N(t) = \tfrac{1}{4}\{1 - \exp[t\ln(1-2/N)]\}.$$

It appears clearly that one first has to make $N \to +\infty$ before $t \to +\infty$, to find equation (14).

random any available direction in space filled with identical hypercubes, instead of choosing just one available bond (see figure 1).

This mapping allows us to write $P_Q(r, t)$ in terms of $P_Z(r, t)$, the probability for the random walker on the Euclidean space to be at a distance r from its starting point:

$$P_Q(r, t) = \sum_{R \in \mathbb{Z}^N} P_Z(r + R, t) \tag{15}$$

where $\mathbb{Z}^N$ is the set of all N-dimensional vectors with integer coordinates (translations of the Bravais lattice).

The crucial assumption now lies in replacing all the hypercubes by statistically similar hypercubes (i.e. the same value of p, but different configurations). The whole calculation rests on this assumption which remains unproven, but may be argued as follows. Though at the percolation threshold the random walker has only a very few actually possible jumps from a given vertex, these jumps will be done in directions that always differ from one vertex to another because of the high dimensionality of the space: after a few steps, the random walker should lose the memory of its former path. Moreover, as the dimension N is larger than the upper critical dimension $d_c = 6$, we may use the mean-field results of percolation theory. So we have linked a problem of a random walk on the hypercube at the percolation threshold to a random walk on a percolation cluster above the upper critical dimension. In this case, P_Z is given by the scaling formula [14]:

$$P_Z(r, t) \sim t^{-\tilde{d}/2} f(r/t^{\tilde{d}/2D}) \tag{16}$$

with $\tilde{d} = \frac{4}{3}$ and $D = 4$.

Formula (15) allows us to extend the function P_Q in the space $\mathbb{Z}^N$. It is then a periodic function and can be written as a Fourier series:

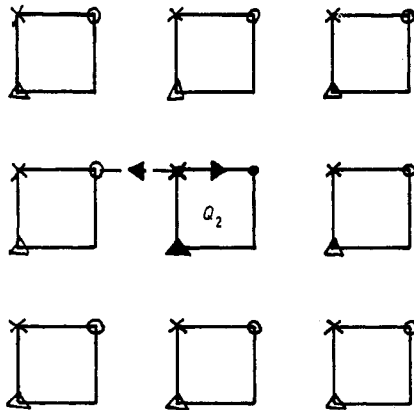$$P(r, t) = \sum_{k \in BZ} a_k \exp(i\pi k \cdot r). \tag{17}$$

Figure 1. Sketch of the mapping of the diffusion on hypercube Q_2 to the diffusion in the whole space $\mathbb{Z}^2$ considered as an assembly of identical hypercubes. The random walker is initially on the site ×. Then it jumps onto the available site ●, following the full line with arrow. In the whole space, it is identical to consider that the random walker can jump with equal probability either on ● or on the site denoted ○ following the broken line with arrow. In the latter description, all the sites ×, ○, △ are respectively identical to sites ×, ●, ▲.

Here the vectors k are restricted to the first Brillouin zone: $k = (\varepsilon_1, \ldots, \varepsilon_N)$ where all the ε_i are 0 or 1 and a_k is given by

$$a_k = \frac{1}{2^N} \sum_{r \in \mathbb{Z}^N} P_Z(r, t) \exp(-i\pi k \cdot r). \tag{18}$$

This representation in terms of a_k is useful because of the relations:

$$\sum_{r \in Q_N} r_i^2 \exp(i\pi k \cdot r) = \begin{cases} 2^{N-1} & \text{if } k = 0 \\ -2^{N-1} & \text{if } k_i = 1 \text{ and } k_j = 0 \text{ for } j \neq i \\ 0 & \text{if } k^2 \geq 2 \end{cases} \tag{19}$$

for every value of $i = 1, 2, \ldots, N$. This implies the value of $r^2(t)$:

$$\langle r^2(t) \rangle = \tfrac{1}{2} N - 2^{N-1} \sum_{\substack{k \in \text{BZ} \\ \|k\| = 1}} a_k \tag{20}$$

which is the key step to evaluate $q_N(t)$ via equation (3).

Since P_Z vanishes beyond the percolation cluster, the coefficient a_k can be estimated as

$$a_k \simeq \frac{1}{2^N} \int_{\mathbb{R}^N} \rho(r) P_Z(r, t) \exp(-i\pi k \cdot r) \, dr \tag{21}$$

where $\rho(r)$ is the two-point correlation function (probability of finding a particle at r from a given particle). Since $\rho(r)$ behaves as r^{D-N}, standard spherical transformations lead to

$$a_k \sim t^{-\bar{d}/2} k^{1-N/2} \int_0^\infty r^{D-N} f\left(\frac{r}{t^{\bar{d}/2D}}\right) J_{N/2-1}(\pi k r) \, dr \tag{22}$$

where $J_{N/2-1}$ is the spherical Bessel function of order $N/2 - 1$. The function $f(x)$ must be regular near $x = 0$ and is an even function of its arguments; a simple form for $x \ll 1$ is

$$f(x) \sim \exp(-\alpha^2 x^2 / \mathcal{D}) \tag{23}$$

where $\mathcal{D}$ is the diffusion constant and α^2 is a constant of order unity (this is exact on Euclidean lattices). This gives the following formula for a_k:

$$a_k \sim \varphi\left(\frac{D}{2}, \frac{N}{2}, -\frac{\pi^2 k^2 \mathcal{D} t^{\bar{d}/D}}{4\alpha^2}\right) \tag{24}$$

where φ is the confluent hypergeometric function. The proportionality factor omitted in (24) is just $a_0 = 2^{-N}$.

Finally, (20) and (24) give an explicit expression for $q_N(t)$:

$$q_N(t) = \tfrac{1}{4} \varphi\left(2, \frac{N}{2}, -\pi^2 \frac{t^{1/3} \mathcal{D}}{4\alpha^2}\right). \tag{25}$$

This is not exactly a stretched exponential. But if the second argument of φ is large ($t \ll N^3 \mathcal{D}^{-3}$) one finds the asymptotic expansion:

$$q_N(t) \sim \tfrac{1}{4} \exp\left(-\frac{\pi^2 \mathcal{D} t^{1/3}}{N\alpha^2}\right) \tag{26}$$

which is of the form (1) with $\beta = \tfrac{1}{3}$.

So, for large times, a stretched exponential with exponent $\tfrac{1}{3}$ is derived, but note that for even larger times ($t \gg N^3 \mathcal{D}^{-3}$), one finds a simple power law:

$$q_N(t) \sim t^{-2/3}.$$

Indeed both stretched exponential and power law seem to be two natural aspects of these kinds of relaxation processes in disordered systems, as pointed out in many articles [15].

5. Conclusion

This paper presents an analytic derivation of relaxation phenomena for disordered systems with stretched-exponential behaviour (and $\beta = \frac{1}{3}$) at the glass transition. Some analytical results have been obtained under an unproven assumption of irrelevant disorder, which is supposed to be true because of the large number of degrees of freedom of the system. The example which has been treated (random energies in the configuration space) is obviously too naive to be realistic. But we can argue that the scheme is quite general if it is explained as follows: instead of energies, consider energy valleys. It is well known that disordered materials have many valleys of almost the same minimum energy. Consider now the configuration space as an assembly of valleys in a large dimensional space. The topology of the ensemble of valleys is more complicated than simple vertices of an hypercube, but it keeps the same important features: it is a closed subset of a N-dimensional space, whose proportion of available valleys is an increasing function of temperature. So all the relaxation behaviours found above are expected to be valid when this description of the configuration space can be done.

Acknowledgments

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