Relaxation in disordered systems

M Ghosh and B K Chakrabarti

Saha Institute of Nuclear Physics, 92, Acharya Prafulla Chandra Road,
Calcutta-700 009, India

Abstract : The relaxation behaviour of various statistical and thermodynamic many body systems like percolating networks, magnetic systems, spin glasses, ordinary glasses, polymers, powders or sand-pile, neural network etc. are reviewed. Various theoretical models to explain these behaviours are briefly sketched

Plan of the Article

I. Introduction

Any dynamical system in equilibrium, when disturbed or perturbed, evolves in time to its new equilibrium state according to the dynamical equations of motion. After some typical time, characteristic of the system (of the order of the relaxation time), the system reaches its new equilibrium. For example, consider an LCR circuit.

This simple system involves only a few degrees of freedom. The evolution of charge on the capacitor follows a precise equation of motion and the relaxation behaviour and relaxation time is known exactly, solving the (first order) differential equation of evolution. For a thermodynamic or statistical system, which involves many degrees of freedom, the equations of motion become coupled and the dynamical evolution behaviour, of some macroscopic average thermodynamic quantity, become nontrivial involving statistical averaging over various initial conditions. The study of relaxation behaviour and the relaxation time of such statistical or thermodynamical systems have attracted a lot of attention recently. In particular, near some (cooperative) phase transition points of such many body systems, the average macroscopic dynamics become extremely slow and also some anomalous relaxation behaviour are seen.

In many-body systems involving interactions between the various degrees of freedom, for example, in magnetic system with Heisenberg interaction between spins, the system gets ordered at lowest temperature because of the cooperative interactions and the order gradually disappears with increase in temperature (increasing thermal noise). Finally at a transition point, the cooperative order disappears (e.g. in the magnetic case the ferromagnetic order disappears at the Curie point above which the spin system becomes paramagnetic) and the correlation length (denoted by ξ), over which fluctuations are correlated, diverges at the transition point (Stanley 1971). We will consider dynamics of average thermodynamic quantities like magnetization etc., before the system reaches equilibrium corresponding to various thermodynamic fields (temperature, external magnetic field etc.), in various magnetic (spin glass etc.) systems. Similarly, we will consider the dynamics (of, say, dispersive strain modes etc.) in purely statistical systems like percolating systems. Such systems may be ideally defined on a lattice where randomly some bonds are removed with a concentration c ($= 1 - p$). Due to fluctuation, the various kinds of vaccancy clusters will be formed and at a typical concentration $c_0 (= 1 - p_0)$, called the percolation threshold, the occupied bonds cease to percolate ; thereby loosing macroscopic connection. The correlation length ξ, defined as the typical cluster size, diverges at the percolation point (Stauffer 1985).

Normally, the relaxation behaviour in a thermodynamic system follows the common Debye type form with a single relaxation time. The standard (Debye) form for any response function $\eta(t)$ is

$$\eta(t) \sim \eta(\infty) - A \exp(-t/\tau) \tag{1.1}$$

where τ is the relaxation time, $\eta(\infty)$ denotes the new equilibrium value. A is a constant. As the critical temperature is approached τ shows a slowing down ; τ diverges at T_0 :

$$\tau \sim \xi^z \sim (T - T_0)^{-\nu z}, \tag{1.2}$$

where z is the dynamic exponent and ν is the correlation length exponent (Stanley 1971). For percolation model the T will be replaced by p (and T_0 by p_0).

Experimental observations suggest a completely different relaxation phenomena in glasses. Glassy system under stress or any perturbation, relaxes to its equilibrium value with an altogether different behaviour. In case of ordinary glasses under (constant) stress, it is observed (Douglas 1965) that the growth of strain $\epsilon(t)$ is given by the form

$$\epsilon(t) \sim \epsilon(\infty) - A \exp\left[-(t/\tau)^\alpha\right] \tag{1.3}$$

where the exponent α is less than 1. This form is commonly known as the Kohlrausch's stretched exponential form. From now on, we will refer this form as stretched exponential relaxation. The relaxation time τ also shows a completely new behaviour known as the Vogel-Fulcher law (Vogel 1921 and Fulcher 1925) :

$$\tau \sim \exp\left[1/(T - T_0)\right] \tag{1.4}$$

where T_0 is a fitting parameter, and the relaxation time diverges usually at a temperature T_0 different from T_g, the glass melting point ($T_0 < T_g$).

Very recently, however, stretched exponential relaxation behaviour ($\alpha < 1$ in expression (1.3)) has been seen in pure magnetic (Takano et al 1988, Ogielski 1987, Ghosh and Chakrabarti 1990) or percolating systems (Ghosh et al 1989), which are very precisely characterized by their respective (single) correlation length ξ, diverging near the respective critical points, with well-known power law behaviour. Although, the stretched exponential behaviour is observed (as originally observed in glasses), none of these systems show Vogel-Fulcher behaviour (expression (1.4) for τ), rather the normal critical slowing down (expressions (1.2) for τ) is observed. The same behaviour was observed (Ogielski 1985) earlier for spin glasses or for magnetic alloys with random competing interactions (frustrations) (Binder and Young 1986, Chowdhury 1986). These observations have recently established that stretched exponential relaxation behaviour is not any characteristic of glasses and that such behaviour are, in fact, quite common ; rather the Debye relaxation behaviour (eq. (1.1)) is an exception for the response behaviour of many-body systems. However, the Vogel-Fulcher behaviour for average relaxation time remains still a characteristic of (some) glasses.

In the next section we will give some typical results of relaxation phenomena observed experimentally (by experiment we mean both real experiments and computer simulations) in various system. In Section 3 some models, which have been proposed to explain such relaxation behaviour will be discussed.

2. Experimental results

2.1. *Relaxation in magnetic systems* :

Extensive studies on magnetic relaxations have been made using the Monte Carlo computer simulation technique. In the single spin flip kinetic Ising model the details of the Monte Carlo procedure (for studying the Glauber dynamics) are as follows (see Binder 1978). One starts with an initial spin configuration (usually the completely ordered ferromagnetic), selects a spin S_i at random and

determines the transition probability $W(S_j \to -S_j)$. Let E_j be the energy of the J-th spin due to the local (say, nearest neighbour) spin configuration. Then, flipping the spin would need energy $\Delta E = 2E_j$ and one can take the flipping (transition) probability W for the S_j spin as $W = \exp(-\Delta E/KT)/(1 + \exp(-\Delta E/KT))$, which gives the same Boltzmann equilibrium distribution. This spin is then flipped if the random fraction between 0 and 1 exceeds normalised W. If the spin is not flipped the old configuration is counted as a new configuration. The process is repeated many times. Thus a sequence of new spin configuration is generated, where the number of computer iteration steps correspond to the time lapse. The system now relaxes to the thermal equilibrium configuration appropriate to the chosen external variable (temperature, external field etc.). Equilibrium is reached when the average macroscopic quantities saturate with time.

The critical dynamics of Ising systems was studied by Chakrabarti *et al* (1981) in 3 and 4 dimension (Figure 1). The system sizes were up to 360^3 and 40^4 in $d = 3$ and 4, and they used multispin coding technique. They fitted their
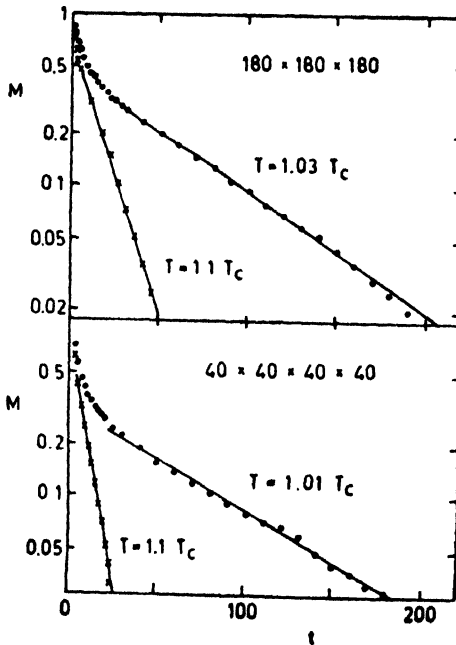

Figure I. Semi-logarithmic plot of magnetisation versus time in three and four dimensions. The straight-line fits give the asymptotic decaytime τ as the reciprocal slope (Ref. Chakrabarti *et al* 1981).

data to an exponential form (eq. (1.1)). There was infact some indications of a systematic deviation in the relaxation data when fitted to the above form. These deviations were, however, attributed as nonlinear relaxation. They observed the relaxation time τ to diverge with an exponent $\nu z = 1.05 \pm 0.04$ in $3-d$. Brower *et al* (1988) have studied this same critical slowing down in microcanonical Ising

dynamics in a three dimensional system ($128 \times 129 \times 128$ spins). Assuming again the relaxation to be exponential, they obtained the value of dynamic exponent z to be 2.26 ± 0.05 in $3-d$ ($\nu z \simeq 1.42$).

As mentioned before, very recently the failure of simple exponential relaxation (Debye relaxation behaviour) has been observed for simple Ising dynamics. Simulation by Takano *et al* (1988) suggests the magnetisation (Takano *et al* studied the time development of autocorrelation function) at $T < T_o$ relaxes to its equilibrium value, following a stretched exponential behaviour (eq. 1.3), with $\alpha \simeq 1/3$ and $1/2$ in $d = 2$ and 3. Ogielski (1987) observed similar dynamics at $T < T_o$ and in the short time scale (long time behaviour is observed to be normal Debye like). He found $\alpha = 0.33, 0.4, 0.5$ for $d = 2, 3$ and 4 (Figure 2). Ghosh and Chakrabarti
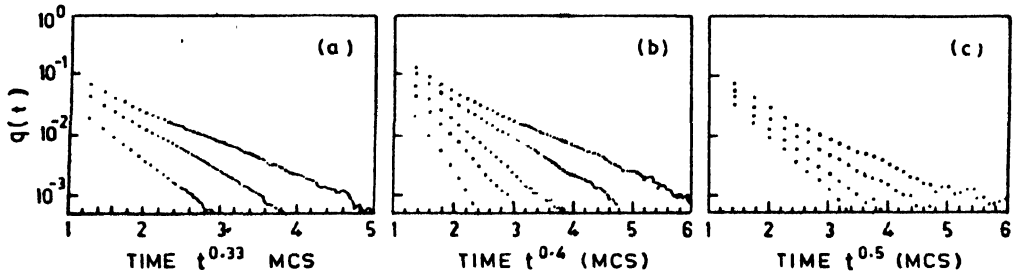

Figure 2. Numerical solutions for the autocorrelation function $q(t)$; (a) $d = 2$, at temp. $T = 2.15$, 2.10 and 2.00 ; (b) $d = 3$, $T = 4.45, 4.40, 4.30, 4.20$ and 4.00 ; (c) $d = 4$, $T = 6.60$, 6.50, 6.40 and 6.30 from top to bottom. The critical temperatures are $T_c = 2.27$, 4.51, 6.7 for $d = 2, 3$ and 4 respectively (Ref. Ogielski 1987).

(1990) repeated the Ising dynamics study by Chakrabarti *et al* (1981) for 1000^3 and 100^3 spins (Figure 3). They observed in the para phase ($T > T_o$), magnetic relaxation have a Kohlrausch-type behaviour (eq. (1.3)) for $t < t_o$, where t_o is a typical crossover time dependent on the amount of disorders : $t_o \to \infty$ as $T \to T_o$ with $\alpha \simeq 0.33$ and 0.4 for $d = 2$ and 3. $\alpha = 1$ for $t > t_o$. The average relaxation time τ diverges with an exponent $\nu z \simeq 1.8$ and 1.1 in 2 and $3-d$. It may be noticed that since $\tau \sim \sum_t m(t)$ the stretched exponential behaviour does not affect the dynamic exponent values ; z is identical in magnitude for both exponential and stretched exponential relaxation regions (Chowdhury 1990).

2.2. *Relaxation in spin glasses :*

As mentioned before, spin glasses are random magnetic alloys with competing (ferromagnetic as well as antiferromagnetic) interactions between the magnetic moments. Infact, the magnetic interaction in metals can be oscillatory of the RKKY type. In dilute magnetic alloys, therefore, the above random competition of interactions occur. Such systems have got interesting (static) phase transition behaviour due to microscopically degenerate ground states (Binder and Young 1986,

Chowdhury 1986). We will discuss here some experimental details of the spin glass relaxation behaviour.

Let a field H be applied to a sample spin glass (1.0% Cu : Mn and 2.6% Ag : Mn) in the paramagnetic phase $T > T_g$. The sample then aquires a magnetisa-
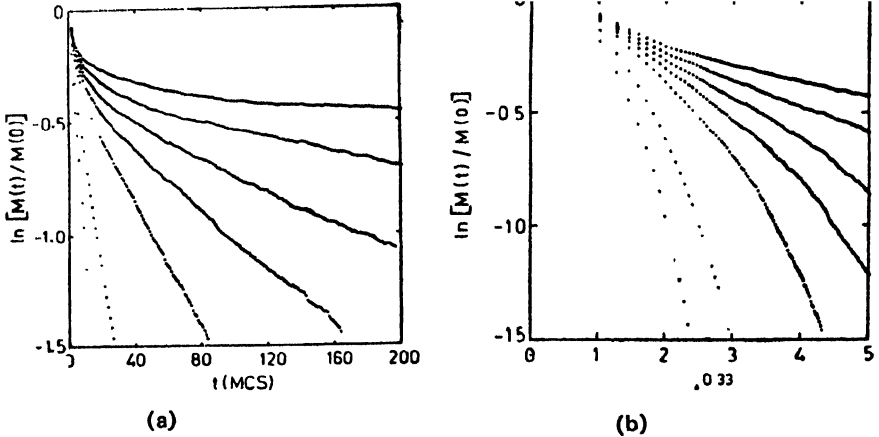
(a)   (b)

Figure 3. (a) Time development of magnetisation $M(t)/M(0)$ for different temperatures. $T/T_c = 1.01$, 1.03, 1.05, 1.07, 1.10, 1.20 and 1.30, respectively (from top to bottom). Lattice size 1000^2. (b) A possible fitting of $M(t)/M(0)$ vs t^α to an exponent value $\alpha = 0.33$ in $d = 2$. (Ref. Ghosh and Chakrabarti 1990)

tion. The sample is then field cooled through the transition temperature T_g, and the applied field H be removed. There appears, then, a field cooled remanent magnetisation, called thermoremanent magnetisation, (σ_{trm}). This was measured
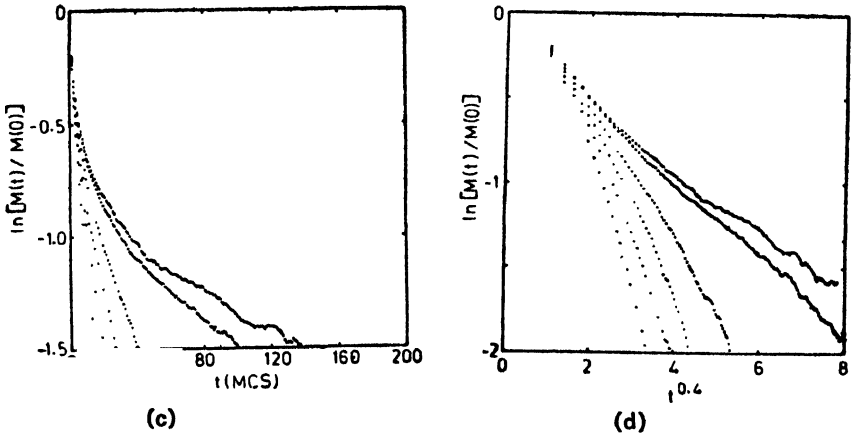
(c)   (d)

Figure 3. (c) Time development of magnetisation $M(t)/M(0)$ for different temperatures. $T/T_c = 1.005$, 1.010, 1.030, 1.070, 1.100, 1.20, respectively (from top to bottom). Lattice size 100^3. (d) A possible fitting of $M(t)M(0)$ vs t^α to an exponent value $\alpha = 0.4$ in $d = 3$ (Ref. Ghosh and Chakrabarti 1990).

(Chamberlin et al 1984) as a function of time after the field is removed. The measurement was made in the interval from 0.2 to 1000 seconds after removing H. They found the time dependance of σ_{trm} as

$$\sigma_{trm} = \sigma_0 \exp\left[-C(t/\tau)^\alpha\right]$$

The exponential factor (C) and relaxation time (τ) can be chosen to be independent of temperature throughout the spin-glass region, whereas the prefactor (σ_0) and time-stretched exponent (α) are observed to become temperature-dependent constants. For $T < 0.75 \, T_g$, $\alpha = 1/3$ independent of temperature and for $T > 0.75 \, T_g$, α decreases while σ_{trm} decreases more rapidly than at lower temperatures (See however, Chamberlin and Haines 1990 for recent developments).

De Fotis *et al* (1988) studied the magnetic relaxation of the insulating spin glass $Co_{1-x}Mn_xCl_2.2H_2O$ for $x = 0.452$. The thermoremanent magnetisation exhibits the features characteristic of spin glass i.e. a rather slow (viscous) decay extending over a large time interval. Of several theoretical decay expressions tested, they found the stretched exponential with a power law prefactor to be the most satisfactory :

$$\sigma_{trm} = At^{-s} \exp\left[-(t/\tau)^\alpha\right] \tag{2.1}$$

The best fit gives $x \simeq 0.06$ and $\alpha \simeq 0.45$.

Ogielski (1985) presented an extensive study of the dynamic behaviour of short range Ising spin glasses, as observed in stochastic (Monte Carlo) simulation. The time dependence of the order parameter $q(t) = \langle S_\alpha(0) S_\alpha(t) \rangle$, and dynamic correlation functions have been recorded. A wide range of temperature $(0.7 \leq kT/J \leq 5.0)$ and lattice sizes $(8^3, 16^3, 32^3, 64^3)$ were investigated. He found that
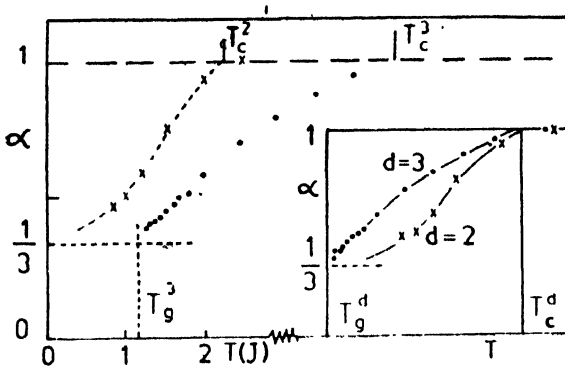

Figure 4. Stretched exponential exponent α for relaxation above T_g in near-neighbour $\pm J$ Ising spin glasses. Circles : data of Ogielski for $d = 3$. Crosses : results from McMillan's data. T_c^d and T_g^d are the ferromagnetic Curie temperature and spin-glass temperature for dimension d. Inset : the same results plotted with the temperature scale going from T_c^d to T_g^d (Ref. Campbell et *al* 1988).

the emperical formula for $q(t)$ to be similar to eq. (2.1) with temperature dependent exponents $x(T)$ (approximate value 0.065) and $\alpha(t)$ (nearly 1/3 as $T \rightarrow T_g$) describes the decay very well at all temperatures above the spin glass transition. Infact, the analysis by Campbell *et al* (1988) suggests that the effective (temperature dependant) estimate of α approaches the terminal value $\alpha = 1/3$ as T approaches T_g. They analysed Ogielski's (1985) and McMillan's (1983) data by fitting them to the

Ogielski's parametrization equation (Figure 4). In the spin glass phase, only the algebric decay $q(t) = At^{-s}$ could be observed with different temperature dependence of the exponent $x(T)$. Power-law fits for $\tau(T)$ are found. The fitting yields $zv = 7.9 \pm 1.0$. The estimated error for the exponents include the uncertainty of the estimate of T_g. Although many previous simulation studies indicated Vogel-Fulcher type behaviour for the spin glass relaxation time variation with temperature, extensive simulation study by Ogielski has ruled it out for spin glass. However, in all cases for spin glass systems where power laws are observed, the exponent vz turns out to be usually large (Binder and Young 1986).

2.3. Relaxation in percolating systems :

The elastic behaviour obeying Hooke's law is an ideal case. In single crystals we expect the strain to be a function of stress alone. In anelastic systems (Zener 1948, Balakrishnan 1985) such linearity is maintained but the strain is also a function of time. Kè (1947) observed the anelastic effects in polycrystalline aluminium (Figure 5). The strain was measured in the sample under constant
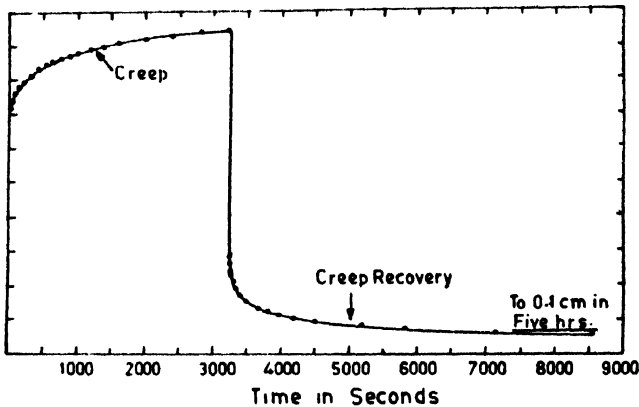

Figure 5. Creep under constant stress and recovery at 175 C in polycrystalline aluminium (Ref. Kè 1947).

stress. To measure the anelastic effects (see e.g. Dattagupta 1987), the applied stress is made small enough so that the effects are recoverable and are linear. The strain grows to a constant value (linearly proportional to the stress ; Hooke's law) exponentially and thus reaches its equilibrium. This is also known as creep relaxation. The viscous slipping at grain boundaries are identified to be responsible for the creep. As mentioned earlier, with random voids in a system, the (material) connectivity is lost beyond the percolation threshold of void concentration. At the threshold the system becomes marginally connected and the structures of the network becomes scale invariant self similar fractals (Stauffer 1985). Ghosh et al (1989) have investigated experimentally the strain relaxation behaviour of two .dimensional random percolating networks.

The samples were anelastic to start with and the growing (relaxing) strain $\epsilon(t)$ in time t was recorded in the linear region. The relaxation process becomes extremely slow near the percolation point. The elongation was measured (Benguigui 1984) with a displacement transducer (LVDT) and the growth of the strain under a constant load was recorded by a chart recorder. Polycrystalline aluminium foils of thickness 0.10 mm and copper foils of thickness 0.09 mm were used. A sheet of 20×20 cm was used and holes were punched regularly on it with diameter 0.8 cm on a square lattice of 1 cm unit cell. Defects were then introduced by cutting inter-hole bonds at random with a concentration $c \, (= 1 - p)$ (following a Monte Carlo generated random number sequence). For each sample (configuration), loads were varied within elastic limit and the linearity and reversibility of the growing strain $\epsilon(t)$ was checked. The normalised strain, which was found to be independent of load, was obtained for different c values (Figure 6). The
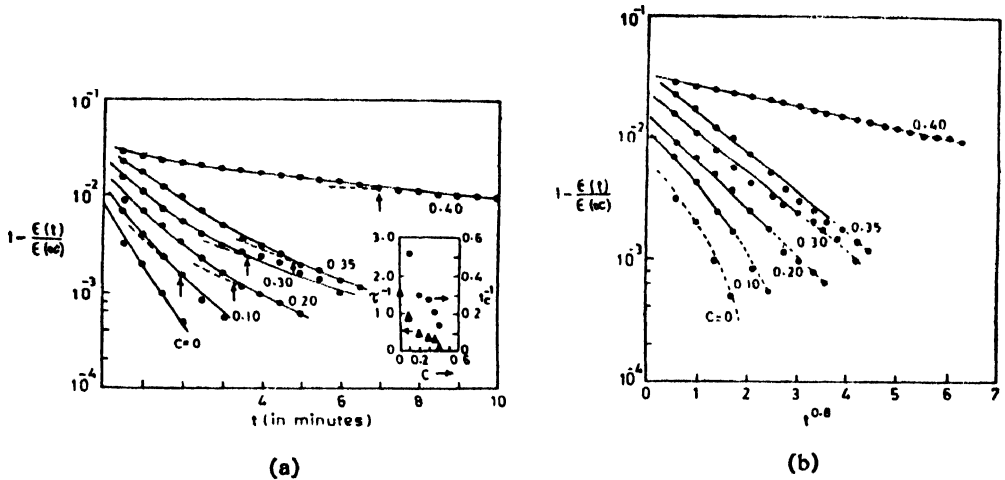

(a)   (b)

Figure 6.   (a) $[1 - \epsilon(t)/\epsilon(\infty)]$ against t, showing the crossover from stretched-exponential (for $t < t_c$; t_c indicated by vertical arrows). The insets show τ^{-1} and t_c^{-1} against c in square lattice for copper.   (b) A possible fitting to an exponent value $\kappa = 0.8$ for the stretched-exponential region in square lattice for cooper (Ref. Ghosh et al 1990a).

strain growth $\epsilon(t)$ in time t was recorded in the linear region. A crossover from a simple exponential behaviour to a stretched exponential behaviour was observed.

$$\epsilon(t) \, , \, \epsilon(\infty) - A \exp\left[-(t/\tau)^\alpha\right] \quad \text{for} \quad t < t_0 \quad \kappa \simeq 0.8$$

and

$$\epsilon(t) \sim \epsilon(\infty) - A \exp\left(-t/\tau\right) \quad \text{for} \quad t > t_0$$

$t_0 = 0$ for the perfect network. Both the crossover time t_0 and the relaxation time τ increases and tend to diverge near the percolation threshold (c_0) of the dilution c. $\epsilon(\infty)$ and A are constants dependent on c.

2

The molecular dynamics simulation (MDS) has recently been performed (Ghosh and Ray 1990) solving dynamical equation in the difference form for all the lattice sites in the system. The strain relaxation behaviour of a two dimensional (20 × 20) randomly diluted elastic network was studied which involves both central and rotationally invariant bond-bending forces was studied. The potential energy of the system is given by (Kantor and Webman 1984)

$$V = a/2 \sum_{<ij>} (\Delta r_{ij})^2 g_{ij} + b/2 \sum_{<ijk>} (\Delta \theta_{ijk})^2 g_{ij} g_{jk}$$

The first part is for bond stretching force as is considered in the central force system ; each bond is replaced by a spring with force constant a. Δr_{ij} is the change in the bond-length between nearest neighbour sites $\langle ij \rangle$ and $\Delta \theta_{ijk}$ is the change in the bond angle between two adjacent bonds $\langle ij \rangle$ and $\langle jk \rangle$. The second part represents the bond-bending force with force constants b. In the present simulation $b/a = 0.1$. $g_{ij} = 1$ for the bond occupied with a probability $(1-c)$ and $g_{ij} = 0$ otherwise. The network is subjected to a constant tensile force and Verlet's algorithm (Dienes and Paskin 1983, Ray and Chakrabarti 1985) of MDS is used. The dynamics minimizes the energy of the system and the system reaches equilibrium when the force on individual lattice site becomes balanced. The strain is measured after every 10 iterations and the process is continued till the strain
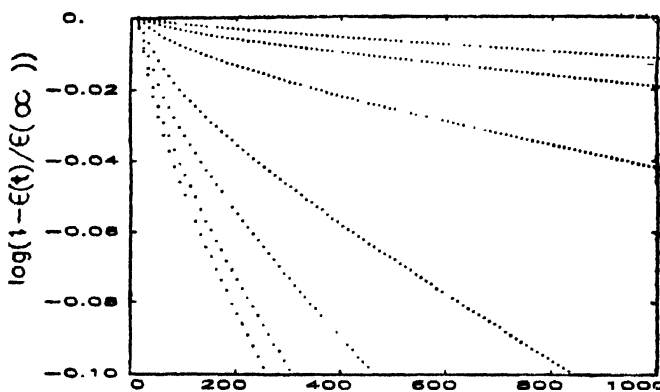
(a)

Figure 7. (a) $[1 - \epsilon(t)/\epsilon(\infty)]$ against t in a 20 × 20 square lattice for different c values. $c = 0.05, 0.10, 0.15, 0.20, 0.25, 0.30$ and 0.35 (from bottom to top) from the data obtained by molecular dynamics.

reaches its final value. The process is repeated for different dilution concentration c. They have determined $\epsilon(t)$ for $c = 0.05$ to 0.35. The $\epsilon(t)/\epsilon(\infty)$ vs t (iteration steps) plot shows that there is a significant increase in relaxation time τ as c_0 is approached. The more the system is diluted the more slowly it reaches equilibrium

value. The behaviour observed is similar to that observed experimentally but with an exponent value $\alpha = 0.65 \pm 0.02$ (Figure 7). τ was measured from the slope of
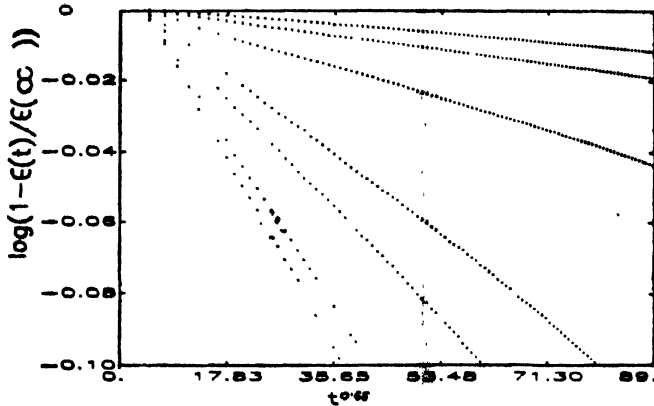
(b)

Figure 7. (b) A possible fitting to an exponent value $\alpha = 0.65$ for the stretched-exponential region in 20×20 square lattice (Ref. Ghosh and Ray 1990).

[ln $\epsilon(t)/\epsilon(\infty)$] vs t plot for t $>$ t_θ. It show a typical critical slowing down as $c \to c_c$ with an exponent value 1.2 for z. Similar behaviour was observed for t_θ with exponent $\sim$ 2.0. The Young's modulus shows a rapid fall as $c \to c_\theta$ with exponent 3.7.

2.4. Relaxation in polymers and glasses :

Recently, there has been a lot of studies on Ising dynamics on percolation cluster (at the percolation point) and interesting non Debye relaxation behaviour are observed (Jain 1986, for $d = 2$ and Chowdhury and Stauffer 1986). However, since non Debye relaxations are already observed in pure Ising systems (see Section 2.1), the additional modifications in Ising relaxations on percolating need further consideration. However, the change in dynamic exponent z on percolation fractals can be straightforwardly identified as the percolation fractal effect, since the non Debye relaxation observed in pure Ising system does not affect the dynamic exponents (see Section 2.1). In the glassy systems only stretched exponential functions were observed in 1966 by Douglas (1966), Scott (1925) and Jones (1944) suggested that this creep data in inorganic glasses were in accord with stretched exponential form (eq. (1.3)) with a temperature independent exponent $\alpha \simeq 0.5$. There is also data for extensive study of creep in polymers. A few examples analysed by Ngai (1987) are given here. Creep was studied in polyvinylchloride. In fact, a decrease in α with increase in annealing time was reported (Turner 1964). Chai and McCrum (1980) measured the creep in isotatic polypropylene with different aging times. They found the similar stretched exponential behaviour with $\alpha \simeq 0.226, 0.180, 0.142$ for annealing times 0.72, 11.5, and 191 Ksec respectively. These observations indicate, although the exponent α

is time dependent and varies as annealing or physical aging proceeds, relaxation function in eq. (1.3) is still a good description.

For stress relaxation in ordinary (silica) glasses, the stretched exponential behaviour, say, stress relaxation etc. has been observed and established long back. Recently, in comparison with spin glass relaxation (see Section 2.2), the analysis of Campbell et al (1988) for the relaxation data of glasses suggests $\propto \rightarrow 1/3$ as the temperature approaches the glass transition point. Lyons et al (1986) directly observed fluctuating dipolar crystals $KTa_{0.991}Nb_{0.009}O_3$ (dipolar glass) $KTa_{0.991}$ using inelastic light scattering techniques for temperatures between 1.8 and 2.5 K and for electric fields up to 2 KV/cm. By combining results of inelastic light scattering with earlier dielectric-relaxation data, they obtained a quantitative measure of the dipolar dynamics spanning more than nine decades in frequency. They found clear evidence for a cooperative dynamic regime, suggesting a transition at a finite transition temperature to a glassy dipolar state. Their dielectric relaxation data obey a Vogel-Fulcher relaxation model over the temperature range 6-15 K with an extrapolated transition at 3.0 K to a glassy dipolar state

$$\tau^{-1} = \omega \exp\left[-E_a/(T-T_0)\right]$$

τ is the average relaxation time, E_a activation energy, ω an attempt frequency, T_0 the temperature where all relaxation times diverge.

Alder et al (1970) studied the hard sphere model system in its stable fluid range and showed that the diffusivity

$$D = A(V/V_0 - 1)$$

Woodcock and Angel (1981) extended the study using essentially the same algorithm $\rho\sigma^3 = 0.95$ to 1.08 in the metastable range. In this region the diffusion coefficient behaves as in the laboratory fluids following the equation

$$D = A' \exp\left[-B/(V-V_0)\right]$$

The diffusing, internally equilibrated, metastable fluid can be arrested at different densities by sudden quenching to obtain the glassy state. It is found that this limit of amorphous-phase also gives the above equation.

2.5. Relaxation in sand-pile :

Very recently, there has been considerable upsurge of interest in the (statistical) dynamics of granular systems like sand-piles. Many previous observations on sand-pile instabilities by chemical engineers (Bagnold 1966) are being recently repeated with a view to observe and establish the self-organised critical phenomena (Tang and Bak 1988) in such systems.

Consider a pile of dry sand. If left to itself it can sustain, under the influence of gravity, a finite 'angle of repose' θ_r when additional sand grains are added. This angle is the angle between the horizontal and the free surface of the sand-pile. With addition of sand grains, this angle is restored after successive slides (avalanches) and the pile is again brought to a metastable (self organised) state of equilibrium slope. Obviously for $\theta < \theta_r$ there will be no such flow (avalanche).

Bak *et al* (1987) introduced the idea of self-organised criticality. Their idea rests on the assumption that θ_r is the critical angle. If θ is made greater than θ_r continuously, by adding more material to the top or tilting the base of the pile, the pile will organise itself such that its average slope will be θ_r (Figure 8(a)) by unloading excess material through avalanches. Thus, suggested an analogy between the dynamic behaviour of sand-pile and traditional critical phenomena (power-law behaviour for $\theta \geqslant \theta_r$). In a cellular automata model introduced by Tang and Bak, the angle variable at any site can be bounded such that when, with addition of grains, 'height' h_i at any site i increases beyond h_o, then the additional materials flows to and are uniformly shared by the nearest neighbours $i \pm \delta$ of i and $h_{i+\delta}$ increases to accommodate this flow. Further avalanche occurs if any of $h_{i+\delta}$ also exceeds h_o. Power law behaviour for the growth of avalanche mass, similar to critical phenomena, are observed in such automata models. The time trace of a typical sequence of avalanches gives the relaxation behaviour in the sand-piles. Thus a power law dependence for the relaxation from supercritical state $\theta > \theta_r$, back to the critical state $\theta = \theta_r$, is also expected in such models.

Instability is introduced in a cohesionless granular material when submitted to vertical vibrations (say by a loud-speaker) beyond a certain threshold ; the horizontal free surface becomes unstable and exhibits a slope at an angle $\theta'_r (< \theta_r)$ with the horizontal. It simultaneously appears as a permanent flow of avalanches on the free surface, and there is a convective transport of particles in the bulk from the bottom to the top. Such a dynamic (liquid-like convective) equilibrium then maintains the (solid-like) finite slope θ'_r of the free surface with the horizontal. If the amplitude and frequency of vibration of the platform is increased, θ'_r decreases and the loss of extra mass through successive avalanches follow a typical relaxation behaviour.

An experimental set-up to study such sand-pile behaviour is the following (Evesque and Rajchenbach 1989). A parallelepipedic cell is partially filled-up with very small glass spheres and the free surface is made horizontal. The box is fixed on a loudspeaker so that the bead heap can be vertically shaken, in the range of frequencies 10-1000 Hz. The exact displacement of the cell is precisely evaluated by photoelectric measurements. At low amplitudes of vibrations, the beads remain motionless in the box reference frame. Beyond a given threshold of amplitude, which appears to depend on frequency, a relative motion of beads is allowed which leads to internal convection transport together with a new stationary profile (Figure 8(a)).

Jaeger *et al* (1989) have studied the nature of the particle flow by investigating the dependence of the average slope on vibration intensity. Switching on the vibrations is a way to prepare the system in a supercritical state, and one can then

observe the relaxation to the steady-state corresponding to the vibrations. In a supercritical state, the slope of a sand-pile decays as log (t) where vibrations were introduced (Figure 8(b)).
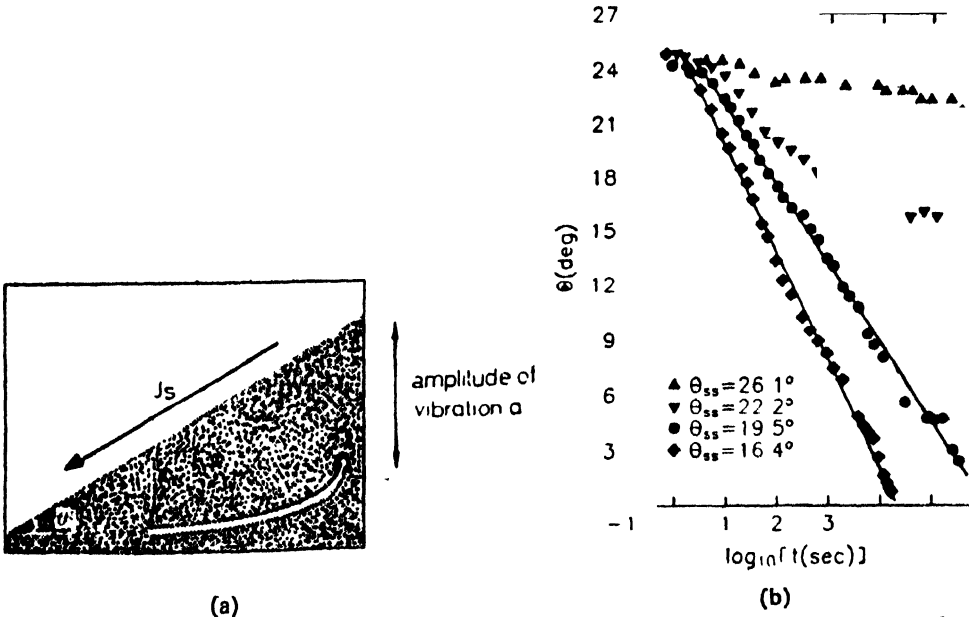

(a)   (b)

Figure 8. (a) Beyond a certain threshold of amplitude of vibrations, the free surface of a bead packing becomes inclined at θ to the horizontal. Simultaneously, there is a convective transport of matter from the bottom to the top and a permanent current J_s rolling down the surface (Ref. Evesque and Rajchenbach 1989). (b) The relaxation of θ in a stationary drum with glass beads. Vibration intensities increase from top to bottom. Straight lines indicate log (t) behaviour. θ_{cj} is the steady state angle (Ref. Jaeger et al 1989).

2.6. Relaxation in neural network models :

Models of neural networks which exhibit features of associative memory have been the subject of intense theoretical activity, starting with the Hopfield model of neural network (see e.g. Amit 1989). In such a spin (glass) neural network model, each neuron is modelled as a two state (Ising spin) unit representing the 'active' or 'passive' states of the neuron. Each recognisable pattern can be represented by a set of values of each neuron or Ising spin (a network of N neurons or spins can have 2^N possible states), and each learned pattern of the network is represented in such models of neural network as a local attractor state or metastable (degenerate) ground state. Following Hopfield's work, attentions were focussed on networks that possess a global energy function. Assuming for simplicity a system of N two state neurons, their energy function is given by

$$H = -1/2 \sum_{i,j=1}^{N} J_{i,j} S_i S_j$$

here $S_i (=\pm 1)$ denotes the state of the *i*-th neuron, and $J_{ij} = J_{ji}$ is the coupling constant (synaptic efficacy) of the pair (i, j).

Information coded in the system as a set of patterns (or 'memories') which are *N*-dimensional binary vectors. Storage of information is achieved by constructing the J_{ij}'s so that the stored patterns become the local (metastable) ground state. Hopfield's model imply the Hebb's rule. A set of p learned patterns are denoted by $\{\xi_i^\mu\}$ $i = 1, 2 \cdots N$, $\mu = 1, 2 \cdots p$ where each ξ_i^μ takes the value $+1$ or -1. Hopfield's version of the Hebb's rule then gives symmetric synaptic strengths :

$$J_{ij} = 1/N \sum_{\mu=1}^{p} \xi_i^\mu \xi_j^\mu, \quad i \neq j$$

After constructing the model (constructing the J_{ij}), following the above procedure, for some random $P = \alpha N$ learned patterns, one can study the recall process. Any arbitrary pattern, given by a set of *N* values of S_i's, then develop according to the (zero temperature Monte Carlo) dynamics :

$$S_i(t+1) = sgn \left(\sum_{j \neq i} J_{ij} S_j(t) \right)$$

Any 'corrupted' pattern will evolve following the above dynamics. If the initial overlap, $m^\mu(0) \equiv (1/N) \sum_j \xi_j^\mu S_j(t)$, is within the domain of attraction of the learned

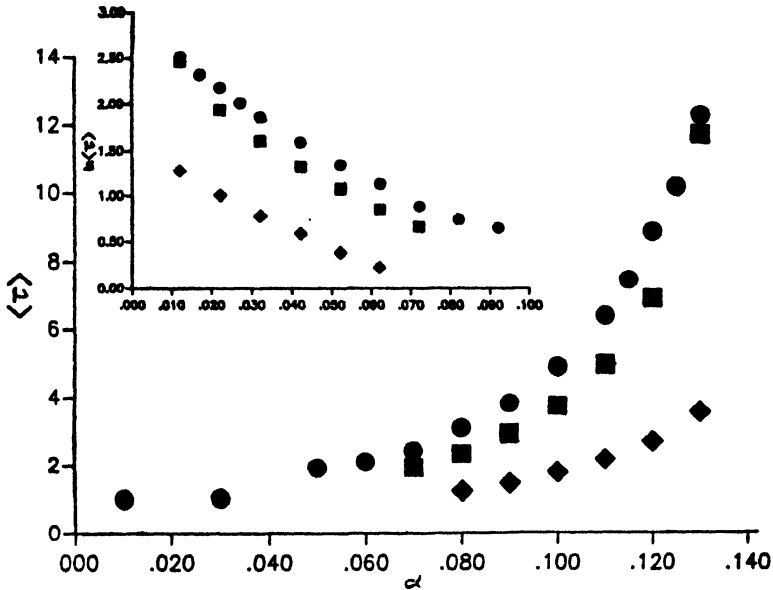**Figure 9.** Plot of average convergence time as a function of α for $m(0) = 0.80$ (circles) and $m(0) = 0.95$ (squares) at $N = 16,000$ and $m(0) = 0.95$ (diamonds) at $N = 1,000$. The inset shows the best fit (Ref. Ghosh *et al* 1990b).

pattern μ, then either the network recognises the stored pattern perfectly as the dynamics brings the corrupted pattern back to the learned pattern (for fine number of

patterns : $\alpha = 0$) ; or it recognises the pattern somewhat imperfectly as the dynamics brings it to a state with significant overlap with the stored pattern (for $0 < \alpha < 0.14$) (Amit 1989). Ghosh et al (1990b) studied the retrieval time (number of itera- tions required to reach from a 'corrupted' patterns with fixed distortion to the corresponding learned pattern) near the phase transition point driven by the storage capacity (the critical storage capacity α_o is taken as 0.142). From their simulations on fully connected networks of N upto 16,000 neurons they obtained that the average relaxation time behaves like $\tau \sim \exp\{-A(N)(\alpha_o - \alpha)^n\}$, with n of the order of unity.

3. Theoretical models

3.1. Models for explaining stretched exponential relaxation behaviour :

There has been many attempts to explain the slow relaxation behaviour in disordered solids or glasses. As discussed in Section 2, the stretched exponential behaviour (eq. (1.3)) is very common in thermodynamic and statistical systems, and can be explained in many ways, while the divergence of the relaxation time following a Vogel-Fulcher like behaviour (eq. (1.4)), is rather rarely observed and also quite difficult to explain. In fact the stretched exponential relaxation behaviour, has been explained using various models for the dynamics of microscopic variables :

(a) Distribution of relaxation times :

It has been suggested (see e.g. Majumdar 1971) that this non Debye relaxation can be implemented as an effect of a distribution of relaxation times.

In glass the stress field at any point can be decomposed into elementary stress relaxation modes q. Each mode decays exponentially with a relaxation time $\tau(q)$. The stretched exponential form $(\exp - (t/\tau)^\alpha)$ may be considered as the result of many exponential decays :

$$\exp - (t/\tau)^\alpha = \int \exp[-(t/\tau)]f(\tau)d\tau, \quad \text{with} \quad f(\tau) \sim \exp(-\tau^{-\beta}) \qquad (3.1)$$

The saddle point equation then gives $\alpha = x/(x+1)$. Majumdar justified the above distribution, by considering diffusion of relaxation mode of wavelength $\lambda = 1/q$; $\tau \sim Dq^2 \sim D\lambda^{-2}$; where D is the diffusion constant. For short time $t \ll \tau$, $f(\tau)$ for a mode λ may be taken as (comparing with the decay of correlation functions near the critical point) $\exp(-\lambda) \sim \exp(-\tau^{-1/2})$ which leads to a value 1/3 for α. At longer times, when regions already relaxed adjust mismatches by slipping along surfaces, $f(\tau) \sim \exp(-\lambda^2)$ which keeps $\alpha = 1/2$. Considering volume relaxation i.e. $f(\tau) = \exp(-\lambda^3)$ one gets $\alpha = 3/5$, as $t \to \infty$ this become more and more relaxed and only the longer mode contributes $f(\tau) = \delta(q - 1/\lambda_{max})$ which gives the normal relaxation (expression (1.1)).

(b) Hierarchical models :

A number of recent theoretical papers considered a model involving hierarchically organised set of free energy barriers (Figure 10) to explain the anomalous dynamics.

Consider the thermalisation of a building partitioned by thick walls, the large rooms in term contain room partitioned by thinner ones. If an initial temperature gradiant is established in the building, the approach to equilibrium through partitions with different thermal conductivities will lead to diffusion coefficients whose actual
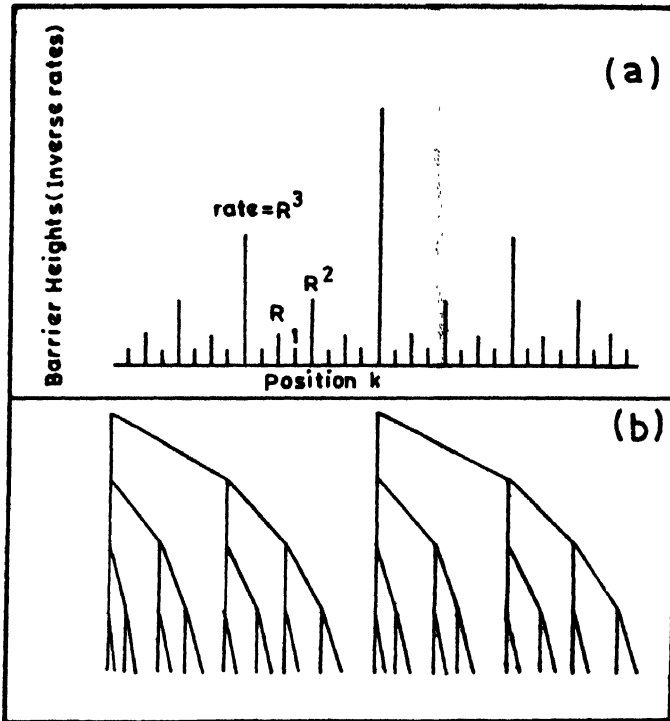

Figure 10. (a) Hierarchical barrier structure. The height of a barrier is inversely proportional to the transition rate. The largest rate (smallest barrier) is equal to 1. The other rates are given by R^l ($0 < R < l$) l integer (where l denotes the level of the hierarchy, as illustrated. (b) A sketch of the hierarchical structure with branching index 2 (Ref. Tietel *et al* 1987).

value depend on time. The process is reminiscent of the non-ergodic behaviour established by system with a hierarchy of energy barriers, as in spin glass system investigated by Palmer *et al* (1984). They considered hierarchically constrained glassy dynamics. They gave the idea that there is a distribution of relaxation times but it is not parallel relaxation as in (a) but series relaxation in which slow degrees of freedom can relax only after the faster processes have taken place.

Huberman and Kerszberg (1985) considered diffusion on a linear chain, with a hierarchically assigned set of barriers (heights) between neighbouring sites. Low temperature diffusion patterns were studied by Blumen *et al* (1986) and extension to higher dimensions were done by Kumar and Shenoy (1986).

(c) Droplet model :

As discussed in Section 3.1, even pure magnetic systems show stretched exponential behaviour.   Takano *et al* (1988) studied below T_c the autocorrelation function in the kinetic Ising model.   Huse and Fisher (1987) argued that in the para phase a droplet of size r will relax as exp $(-r^{d-1}/KT)$, the barrier height being determined by the surface area $\sim r^{d-1}$.   However, as r grows with time in a random walk fashion, $r \sim t^{1/2}$.   This gives,

$$\langle S_i(0)S_i(t)\rangle_o \sim \exp [-Dt^{(d-1)/2}] \tag{3.3}$$

As mentioned in Section 2.1, Ogielski's study of the correlation functions, using computer simulation indicate such behaviour only in the pre-assymptotic region.

(d) Anomalous diffusive origin of the stretched exponential form :

The diffusion equation followed by the strain field (linear to the stress field) in linear solid (metal and glass) is (Ghosh *et al* 1989)

$$d\epsilon/dt = D\nabla^2\epsilon \tag{3.4}$$

The strain modes can be decomposed into elementary modes

$$\epsilon_q(t) = \int_0^\infty \epsilon_r(t) \exp (iqr)dr$$

In Euclidean lattice eq. (3.4) can be solved to get

$$\epsilon_q(t) \sim \epsilon_q(\infty) - A \exp (-Dq^2t),$$

where $q \sqrt{t}$ is the average Brownian spread of the diffusing mode q in time t and A is constant.   The solution can be generally written as

$$\epsilon_q(t) \sim \epsilon_q(\infty) - A \exp [-B(t)q^2],$$

where

$$B \sim \partial^2\epsilon/\partial q^2 \mid_{q=0} = \int_{-\infty}^\infty \epsilon_r(t)r^2 dr \equiv G\langle r^2(t)\rangle ; G \sim D$$

$\langle r^2(t)\rangle$ being the average end to end distance squarred of a random walker in time t.   Then the general solution of diffusion equation may be written as

$$\epsilon_q(t) \sim \epsilon_q(\infty) - A \exp [-Gq^2\langle r^2(t)\rangle] \tag{3.5}$$

where, in general, $\langle r^2(t)\rangle \sim t^{2/d_w}$ in percolating fractals. d_w, the walk dimension, has the value 3 and 5 in two and three dimensions, respectively.   But when $\langle r(t)\rangle > \xi$ the diffusion does not see the fractal and in that case $\langle r^2(t)\rangle \sim t$ ($d_w=2$ for a regular lattice).   Since $\xi \sim (p-p_c)^{-\nu}$.   So for $\langle r(t)\rangle > \xi$, i.e. $t^{1/d_w} > (p-p_o)^{-\nu}$ or at $t > (p-p_o)^{-\nu d_w}$, we see the general solution takes the form of normal relaxation (with single relaxation time)

$$\epsilon_q(t) \sim \epsilon_q(\infty) - A \exp [-Gq^2t] \tag{3.5a}$$

but for $\langle r(t) \rangle \langle \xi$ or for $t \langle (p - p_o)^{-\nu d_w}$ the general solution takes the non-Debye form

$$\epsilon_q(t) \sim \epsilon_q(\infty) - A \exp [- G q^2 t^{2/d_w}] \qquad (3.5b)$$

comparing with eq. (1.3) of text $\alpha = 2/d_w$ and $\tau \sim G^{-1/\alpha} \sim D^{-1}$. Putting the value of d_w, one gets $\alpha = 0.67$ and 0.4 in two dimensions, respectively, and the value of crossover time $t_o = \xi^d{}_w = (p - p_o)^{-\nu d}w$, $d_w = 2$ (for normal relaxation). A similar solution for the dilute magnetic relaxation is expected. It may be noted that the expression (3.5) is not the rigorous solution in a fractal, since the exact form of the diffusion eq. (3.4) there is more complicated.

In an alternative formulation by Dhar (1987) a somewhat similar crossover from the evolution of a master equation in one dimension was suggested. He argued that exp $(-t^\alpha)$ relaxation $(0 \langle \alpha \langle 1)$ encountered in many disordered material may be understood in terms of the spectral density of the Lifshitz states near the band edges.

(e) Relaxation in sand heap :

de Gennes (Evesque and Rajchenbach 1989) had proposed a possible scenario to account for the instability of sand heap based on a series of alternative passive and active regimes.

(i) Passive regime—When the cell is raised up, the beads are compacted so that no possible intergranular motion is allowed. The bead heap can be considered as a solid.

(ii) Active regime—When the cell is carried down with an acceleration γ of the cell larger than the acceleration g of gravity, the system of beads is fluidised (for $\gamma \rangle g$).

During active regime beads are submitted to an apparent gravity γ-g reversed upward. The surface tension being zero, surface of the fluidised beads becomes unstable and exhibits a viscous finguring. The excess of matter which has been raised up on bumps during the active lapse of time will flow as avalanches downwards during the passive regime. The model predicts the existence of a permanent surface flow of particles as is observed experimentally.

According to such picture, the plausible relevant parameters are the amplitude a and the frequency ω of the alternative displacement of the cell, the relative size of particles and the dimensions of the container. In the scheme of gravitational instability of the free surface the important parameter is the acceleration $\gamma = a\omega^2$ of the cell compared to g. The model of self-organised criticality predicts a power law dependence on t while their data cannot be fitted by a power law with reasonable parameters over any wide interval of time. Instead the data for high vibration intensity are consistent with a log t dependence over many decades (see Section 2.5).

To explain such dependence, a different scenario was proposed (Jaeger *et al* 1989) where the vibration intensity ($a^2\omega^2$) plays the role of an effective temperature.

The motion of particles is impeded by the barriers posed by neighbouring beads and the corresponding random potential will be a complicated function of the local configurations. However, we are interested in the average effective barrier height U as the angle θ is varied. Assuming an effective temperature T given by $T = a^2\omega^2$ due to mechanical vibrations the rate of escape over barriers exponentially dependent on the ratio exp $(-U/T)$. This leads to

$$d\theta/dt = -A\theta \exp [(\theta - \theta_r)/a^2\omega^2] \tag{3.6}$$

The equation can be solved analytically, and they get a logarithmic dependence on amplitude and frequency as observed experimentally (Jaeger *et al* 1989). Note, the change in the relevant variables (from $\gamma = a\omega^2$ in de Gennes explaination (a) to $T = a^2\omega^2$ here).

3.2.   Models for explaining Vogel-Fulcher behaviour :

(a)   Model with random distribution of energy and coordination numbers :

Vilgis (1990), noted that two stochastic quantities in random systems, viz. the energy and the coordination number are responsible for the appearance of the law, which shows an unusual essential singularity. He considered both the energy and coordination number to be random variables having a Gaussian distribution.

The characteristic time scale for the simple hopping of arbitrarily chosen test particle of an energy valley with depth E is given by Arhenius law,

$$\tau(E) \sim \exp (E/T)$$

E depends on the coordination number z. Thus, assuming a linear relationship $E(z) = zE$,

$$\tau(E) \sim \exp (zE/T)$$

Considering a Gaussian distribution for both z and E, i.e.

$$P(E) = (1/E_0^2)^{1/2} \exp (-E^2/2E_0^2)$$

and

$$P(z) = (1/(\Delta z)^2)^{1/2} \exp -[(z - z_0)^2/2(\Delta z)^2]$$

The characteristic time scale, which is relevant for macroscopic purpose is the average

$$\tau(T) = \langle \tau(z, E) \rangle = \int_{0 < E < 8} dP(z) \int_{-\infty < E < \infty} dP(E) \exp (zE/T)$$

which on simplification gives the Vogel-Fulcher law :

$$\tau(T) \sim \exp [z_0/2(\Delta z)^2)/(1 - T_0/T)]$$

where

$$T_0 = \Delta z E_0$$

(b) Random barrier heights and thermally activated hopping :

Ghosh and Chakrabarti (1990) argued that in glass, where the free energy has many metastable (local) minima, the relaxation time $\tau \sim$ (hopping diffusion constant)$^{-1}$, comes from thermally activated hopping over 'typical' barrier heights h_0 [$\tau \sim \exp$ $(-h_0/T)$]. In cases where there is a thermodynamic rearrangement of the barrier heights due to cooperative structural rearrangements, the typical barrier height may diverge as $h_0 \sim \xi' \sim (T-T_0)^{-\nu'}$ near the structural-rearrangement transition point T_0. This would give a Vogel-Fulcher-like relaxation behaviour (Edwards and Mehta 1989) ($\tau \sim \exp [A/(T-T_0)^{\nu'}]$). Observation of such behaviour (in standard glass, for example) would then indicate the existence and divergence of another correlation length (with exponent ν') near the barrier-height rearrangement transition point.

4. Outlook

In Section 2 we have described some of the experiments which studied the relaxation phenomena in different many body systems near the critical point (thermal or statistical). We find that the stretched exponential behaviour (eq. (1.3)) for the relaxation of some average macroscopic variables of the many body system is quite common and nothing unique of glass. Among the cases discussed above, except for the cases of sand-pile and the neural-network (Sections 2.5 and 2.6) all show stretched exponential behaviour. The relaxation time behaviour represented by Vogel-Fulcher law seems to be a characteristic of (non-metallic) glass only. We observe critical slowing down of the relaxation time as critical point is approached. However, the exponent is found to be quite large in some cases (as in spin-glass systems).

Two established different kinds of relaxation behaviours are thus observed commonly in many body systems :

(a) Kohlrausch stretched exponential relaxation with critical slowing down :

$\eta_b(T) \sim \exp -(t/\tau)^\alpha$; $\alpha < 1$ and $\tau(T) \sim \xi^z \sim (T-T_0)^{-\nu z}$, $\alpha < 1$ and νz depending on dimension and symmetry of order parameter. For Ising systems $\alpha \simeq 0.33, 0.4$. 0.5 for $d=2$, 3 and 4 respectively (Takano et al 1988, Ogielski 1987), with $\nu z = 2.0$, 1.4 and 1.0 (exact) (Brower et al 1988). For percolating systems (Ghosh et al 1990a,) $\alpha \simeq 0.6$ and $\nu z = 4.0$ for $d=2$. Above the lower critical dimensions, $\alpha = 1/3$ and $\nu z \simeq 7.9$ and 8.54 for Ising spin glass and XY spin glass respectively in $d=3$ (Ogielski 1985 and McMillan 1983).

(b) Kohlrausch stretched exponential relaxation Vogel-Fulcher behaviour for relaxation time :

$\eta_b(T) \sim \exp -(t/\tau)^\alpha$, $\alpha < 1$ and $\tau(T) \sim \exp (1/(T-T_0))$. This type of behaviour seems now to be ruled out for spin glass dynamics, although, for some dipolar glass this Vogel-Fulcher like behaviour for τ is traditionally being discussed (Lyons et al 1986).

In Section 3, some of the models have been reviewed to understand the mechanisms, responsible for this anomalous behaviour of relaxation. As discussed there, we believe, stretched-exponential behaviour essentially indicates a classical localization or anomalous diffusion originating due to the appearance of fractals (dynamic or otherwise) at length (time) scales lower than the (percolation) correlation lengths. For these time scales, the diffusion being anomalous, stretched-exponential behaviour occurs, which crossovers to normal behaviour for large time scales where the diffusion spread exceeds the correlation length size (see Section 3.1). We also believe thermally activated hopping over barrier heights, diverging near some cooperative structural rearrangement point (see Section 3.2.b), seems to be a plausible explanation of Vogel-Fulcher law.

Acknowledgment

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About the Authors :

M Ghosh, born in 1962, got her M.Sc. in 1986 from Calcutta University. Doing PhD work on Dynamic Properties of Statistical Systems, in Saha Institute of Nuclear Physics, Calcutta, since 1986.

B K Chakrabarti, born in 1952, got his academic degrees from Calcutta University (PhD in 1979). Visited Department of Theoretical Physics, University of Oxford, UK, during 1979-80, 1985 and 1987 ; University of Cologne, W Germany, during 1980-81 and 1984-85 ; University of Paris, France, during 1988, etc. Working on Statistical Physics of Disordered Systems. Presently employed as Associate Professor in Saha Institute of Nuclear Physics, Calcutta. Awarded INSA Medal for the Young Scientists (1984) and presently an INSA Research Fellow (since 1989).