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LETTER TO THE EDITOR

Anomolous dynamics of interacting particles in random systems

T Ohtsuki and T Keyes

Department of Chemistry, Boston University, Boston, MA 02215, USA

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Abstract. Anomalous behaviour of interacting particles in random systems such as percolation clusters and polymers is discussed from a geometrical point of view. For diffusion-controlled reactions in random systems, it becomes evident that attractive interparticle interactions cause trapping of particles and inhibition of reactions. The existence of a glass transition due to combined effects of randomness and interactions is indicated. Criteria for the trapping and the glass transition are proposed.

In recent years, the study of percolating systems has been one of the most exciting fields of physics. The essence of their geometrical structure is self-similarity and associated scaling relations (Stauffer 1979). All static critical exponents are expressed in terms of fractal dimensionalities (Ohtsuki and Keyes 1984d). The self-similarity also causes various types of anomalous dynamics (Gefen *et al* 1983, Rammal and Toulouse 1983, Harris and Stinchcombe 1983, Ohtsuki and Keyes 1984a, b). Extensive studies by many authors have shown that these anomalies are described by two 'dynamic' fractal dimensionalities: a thickness (fracton or spectral) dimensionality and a length dimensionality (Alexander and Orbach 1982, Keyes and Ohtsuki 1984, Ohtsuki and Keyes 1984e). Recently, another unusual behaviour belonging to a completely different universality class has been suggested (Ohtsuki 1982, Bottger and Bryksin 1982, Barma and Dhar 1983). In the presence of a strong external field, a particle diffusing in percolating systems is captured by dead ends and mobility decreases. On the basis of a real space renormalisation group technique, Ohtsuki and Keyes (1984c) derived criteria for the trapping and clarified the breakdown of a linear response theory near the percolation threshold. In ordinary systems, the velocity of a particle is a monotonically increasing function of the strength of an external field. In random systems, in contrast, the stronger field may lead to a smaller velocity (Pandey 1984, Seifert and Suessenbach 1984). This phenomenon can be regarded as an ergodic-nonergodic phase transition. The same mechanism is considered to work on other dynamical properties of random systems. The purpose of this letter is to discuss some examples of such phenomena and to elucidate their unusual nature.

First, we investigate the effects of attractive interactions between particles on diffusion-controlled reactions in random systems such as percolation clusters and polymers. In usual systems, attractive interactions obviously promote reactions, whereas in random systems, the trapping of particles happens and reactions are suppressed as illustrated schematically in figure 1. Here particles can move only along a random structure. When an attractive interaction is much stronger than a thermal

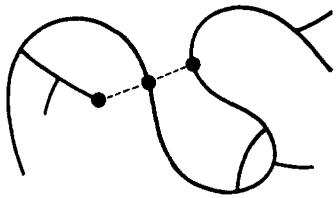

Figure 1. Schematic illustration of the trapping of diffusing particles (●) in a random structure (——) due to an attractive interaction (-----).

energy $k_B T$, particles are trapped and cannot meet each other. Then the reaction rate becomes exponentially small and reactions are substantially inhibited. Note that particles are captured not only by dead ends but also at backbones.

We now discuss a criterion for the trapping. As an example, absorption of diffusing particles by a static absorber in an infinite percolation cluster near the percolation threshold is considered. We deal with two different types of interactions Ψ of the form

$$\text{(a)} \qquad \Psi(r) = \begin{cases} \Psi_0(r/\Lambda - 1) & (r \leq \Lambda), \\ 0 & (r > \Lambda), \end{cases} \tag{1}$$

$$\text{(b)} \qquad \Psi(r) = \begin{cases} -\Psi_0 & (r \leq \Lambda), \\ 0 & (r > \Lambda). \end{cases} \tag{2}$$

In case (a), a particle feels a constant force Ψ_0/Λ in the hypersphere of radius Λ centred at the absorber. When $\Lambda \gg a$, where a is a lattice constant, we can apply the same arguments as those for the trapping by an external field (for details, see Ohtsuki and Keyes 1984c). In the hypersphere, the force attempts to impose a deterministic motion on a particle whose mean displacement $\langle R \rangle$ varies as $\langle R(t) \rangle \propto \gamma t^{\chi}$, where $\gamma = \Psi_0/\Lambda k_B T$ and χ is a critical exponent less than unity. The characteristic time τ necessary to travel a distance Λ from the perimeter to the centre (absorber) is expressed as $\tau \sim (\Lambda/\gamma)^{1/\chi}$, because $\langle R(\tau) \rangle \sim \Lambda$. In this time interval, a particle also makes a random motion (walk) with a root mean square displacement $L = \langle (R - \langle R \rangle)^2 \sim \tau^{\chi/2} = (\Lambda/\gamma)^{1/2}$. In random systems, a particle has to find a microscopic path which will avoid the trapping in order to meet the absorber. Since a particle makes a compact search (Rammal and Toulouse 1983), the criterion for the successful search is given by $L \gtrsim \langle R \rangle$. In this case, therefore, we have the criteria for the trapping

$$\gamma\Lambda = \Psi_0/k_B T \gg 1 \qquad (\xi \gtrsim \Lambda \gg a), \tag{3}$$

$$\gamma\xi = \xi\Psi_0/\Lambda k_B T \gg 1 \qquad (\Lambda \gtrsim \xi), \tag{4}$$

where ξ is the percolation correlation length. Equation (4) comes from the fact that the system is homogeneous in the length scale larger than ξ and the search in each block of linear size ξ is independent. It is worthwhile noting that the criteria (3) and (4) can be derived on the basis of a real space renormalisation group method in much the same way as for the trapping by an external field (Ohtsuki and Keyes 1984c).

In case (b), the probability that a particle passes over the potential barrier at the perimeter of the hypersphere from inside to outside is estimated from $\exp(-\Psi_0/k_B T)$. If $\Psi_0/k_B T \gg 1$, therefore, a particle once entering the hypersphere cannot return to the outside. When $\xi \gtrsim \Lambda \gg a$, the probability that a path connecting one site of the perimeter to the centre is wholly contained in the hypersphere, i.e. the probability that a particle just inside the perimeter can diffuse to the centre without getting out of the hypersphere, is thought to be zero substantially. At $\Lambda \gtrsim \xi$, on the contrary, such a probability is unity and the trapping does not occur because of the homogeneity of the system in this length scale. Hence, the criterion for the trapping is given by

$$\Psi_0/k_B T \gg 1 \qquad (\xi \gtrsim \Lambda \gg a). \tag{5}$$

From (3)-(5), we propose a general criterion for the trapping of particles and the inhibition of reactions in random systems due to attractive interparticle interactions,

$$|\Psi_{max}(\xi \gtrsim r \gg a) - \Psi_{min}(\xi \gtrsim r \gg a)|/k_B T \gg 1, \tag{6}$$

where Ψ_{max} and Ψ_{min} represent the maximum and minimum value of an interaction potential Ψ at $\xi \gtrsim r \gg a$. Here ξ is the general correlation length of a random structure and in the case of polymers, ξ is the radius of gyration.

The physical meaning of the criterion (6) is the following. Consider particle migration in a random coil polymer. Since a particle can move only along the polymer, the position of a particle is described by a contour length s along the chain. The distance $r = r(s)$ measured in the Euclidean space is a random function of s, because the conformation of the polymer is random. Even if an interaction potential $\Psi = \Psi(r)$ is a monotonic function of r, therefore, $\Psi(s) = \Psi(r(s))$ becomes random as a function of s. A particle feels this random potential along the chain and is trapped under the condition that the amplitude of fluctuations in $\Psi(s)$ is much larger than $k_B T$, which is just described by (6). A similar situation is realised generally in random systems. Then the criterion (6) is thought to be irrelevant to the detailed form of an interaction potential $\Psi(r)$ and to hold universally. It should be noted that this type of trapping also arises from a repulsive interaction. In this case, however, reactions are suppressed even in ordered systems and randomness does not play an essential role.

The effective random potential $\Psi(s)$ also gives rise to unusual behaviour of interacting particles in random systems. Here the discussion is limited to infinitely extended systems such as percolation lattices and finite systems like polymers are excluded. Consider a system of particles interacting with each other via a simple repulsive interaction, e.g., a soft core potential $\Psi(r) = \Psi_0(r/a)^{-m}$. The system exhibits a fluid-solid phase transition at a certain number density $n = N/V$. In normal systems, the criterion for this phase transition is approximately given by

$$\phi \equiv n\Lambda^d \sim 1, \tag{7}$$

where ϕ is the effective volume fraction and Λ is the range of the interaction defined by $\Psi(\Lambda)/k_B T = 1$. In random systems, as mentioned before, particle motion is governed by the effective random potential $\Psi(s)$. The range of the interaction is of the order of ξ and its strength is given by $\Psi_{max}(\xi \gtrsim r \gg a) - \Psi_{min}(\xi \gtrsim r \gg a)$. When $\xi \gg \Lambda$ and $\Psi_{max}/k_B T \gg 1$ ($\Psi_{min} = 0$), therefore, an effective 'dynamic' volume fraction ϕ^* becomes singularly larger than the 'thermal' volume fraction ϕ,

$$\phi^* = n\xi^d \gg \phi = n\Lambda^d. \tag{8}$$

At $\phi^* \gtrsim 1$, particle migration over a distance larger than ξ is inhibited and, for instance, the self-diffusion coefficient is almost zero. On the other hand, thermodynamic properties of the system are described by ϕ and have nothing unusual when $\phi \lesssim 1$. Thus, the system can be regarded as showing a 'glass' transition at

$$n^* \sim \xi^{-d}. \tag{9}$$

In the region

$$\Lambda^{-d} \gg n \gg \xi^{-d}, \tag{10}$$

the system is in a glassy state in the sense that the global motion of the particles is frozen in spite of the absence of anomalies in the thermodynamic properties. Similarly to the case of the trapping, it seems that detailed properties of interactions are not responsible for the glass transition. Quite recently, Gefen and Halley (1984) made computer simulations of charged particles moving in percolation clusters and indicated that Coulomb interactions cause a metastable configuration near the percolation threshold. This metastable state is thought to correspond to the glassy phase discussed here.

As mentioned first, self-similarity plays an essential role in the anomalous behaviour of percolating systems such as density of states and diffusion. In practice, however, ideal self-similarity is realised in a limited region. As for the trapping and the glass transition discussed here, on the other hand, the essential point is that a particle cannot move along the direction in which a force due to an external field or an interaction attempts to drive. Then these phenomena are believed to be observed widely in real disordered systems. In Anderson localisation problems, it is well known that electron–electron interactions change qualitative properties of the system (Lee 1982, Fukyama 1982). Laibowitz and Gefen (1984) suggested the importance of Coulomb interactions in real percolating systems (thin Au films). We consider that in random systems, interparticle interactions generally exert a serious influence on the physical properties of the system. Although the present arguments are rather heuristic, we hope that this work will stimulate further researches of disordered interacting systems.

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