Relaxation in Self-Similar Hierarchical Spaces

E. W. Knapp
Physik Department, Technische Universität München, D-8046 Garching, Federal Republic of Germany
(Received 16 November 1987)

A new model of a hierarchical space with arbitrary transfer rates is introduced. The topology is determined by a self-similar coupling scheme. It is closely related to the topology of a corresponding ultrametric space. The model allows exact solutions to the random-walk problem, the intermediate scattering function, and the autocorrelation function. The solutions are identical in structure to the exact solution of the problem of energy transfer in disordered media which corresponds to parallel rather than to sequential coupling schemes.

PACS numbers: 61.41.+e, 75.40.−s, 87.10.+e

Complex systems with many nearly degenerate low-energy states which are separated by high energy barriers often exhibit nonexpontial decay patterns. Among these systems are glasses,[1] spin-glasses,[2] proteins,[3,4] neural networks,[5] and also problems of optimization like the one of the traveling salesman.[6] Diffusion in such systems is slowed down or accelerated. The latter can occur in turbulent flow.[7] Nonexponential decay patterns can also be observed by the measurement of the intermediate scattering function with neutron scattering or Mössbauer spectroscopy. Mössbauer spectra of iron containing proteins may contain broad quasielastic lines[8] which are indicative of nonexponential decay.

To account for nonexponential time decay, models of hierarchically constrained dynamics have been introduced.[9] Ultrametric spaces support such hierarchical structures and provide the topology for the mean-field spin-glass.[10] Though the spin-glass topology does not directly relate to its dynamics, ultrametric spaces are considered to yield suitable model structures for the dynamics of many complex systems. The dynamics in a ultrametric space follows a sequential coupling scheme where relaxation or transport processes involve many intermediate states. But also relaxation processes with a parallel coupling scheme give rise to nonexponential decay patterns. An example is the problem of energy transfer from a donor to acceptors in disordered media.[1]

In this Letter dynamics of complex systems are studied with use of a general model with a hierarchical coupling scheme which exhibits a nearly ultrametric topology. Exact general solutions for the random-walk problem, the intermediate scattering function, and the autocorrelation function are provided. The structure of the solution agrees with that of the energy-transfer problems in disordered media.

Discrete states in spaces with an ultrametric topology form clusters on various hierarchies. An example of dimer clusters on three hierarchies is depicted in Fig. 1. At a low hierarchy (energy) level there are many distinct clusters. At a higher hierarchy level these clusters are connected by pathways involving a high energy barrier and can merge into a single cluster. In Fig. 1 two distinct coupling schemes of dimer clusters are shown. In part (a) the coupling scheme is such that at a given hierarchy transitions occur with equal probability between all states of the corresponding cluster. This cou-

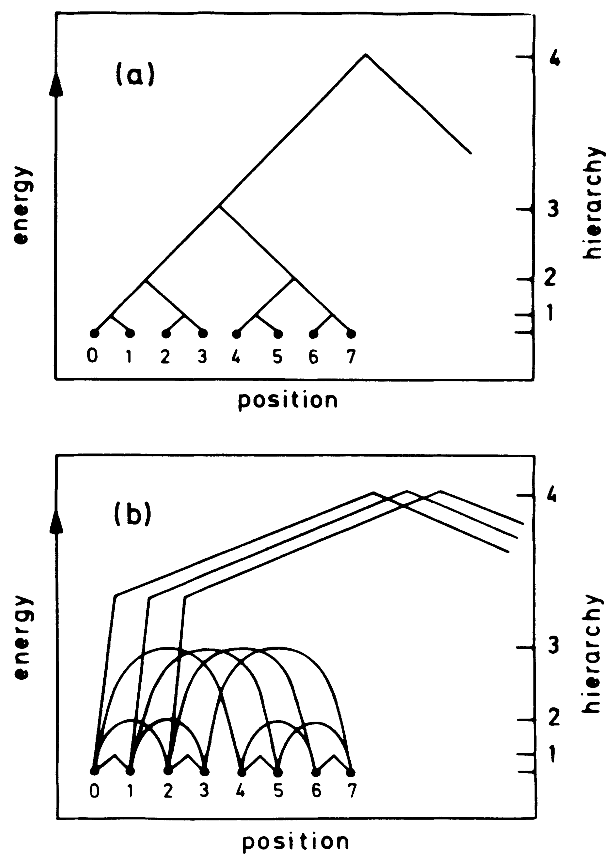

FIG. 1. Two coupling schemes involving dimer clusters. Each dot represents a state. Pairs of states are connected by pathways involving energy barriers which determine the distance. (a) An ultrametric topology where all states within a given hierarchy level can be reached with equal probability. (b) The coupling scheme is approximately ultrametric. At each hierarchy level only pairs of states are connected. The set of pathways on hierarchy level four is not completed.

pling mechanism wipes out the cluster structures prevailing at lower hierarchy levels. This space with an ultrametric topology is normally used.[11] In the other coupling scheme [depicted in Fig. 1(b)] states are connected pairwise only. Its topology is approximately ultrametric. Dynamics on the two different spaces are equivalent if distances (energies) of subsequent hierarchy levels are well separated from each other.

All relaxation processes in a space with a discrete number of states are governed by a master equation:

$$d\mathbf{p}(t)/dt = \mathbf{p}(t)\tilde{\mathbf{R}}_L, \quad \mathbf{p}(0) = \mathbf{p}_0. \tag{1}$$

A finite ultrametric space of binary clusters involving L hierarchy levels contains 2^L states. The component $p_n(t)$ of the state vector $\mathbf{p}(t)$ yields the probability to meet the system in the local state at site n. Hence, $\sum p_n(t) = 1$. The rate matrix $\tilde{\mathbf{R}}_L$ corresponding to the coupling scheme of Fig. 1(b) can be defined by the following recursion relation:

$$\mathbf{R}_{j+1} = \begin{bmatrix} \mathbf{R}_j & R_{j+1}\mathbf{I}_j \\ R_{j+1}\mathbf{I}_j & \mathbf{R}_j \end{bmatrix}, \quad j = 0,1,\ldots,L-1, \tag{2}$$

where $\mathbf{R}_j$ and $\mathbf{I}_j$ are $(2^j \times 2^j)$-dimensional matrices and $\mathbf{I}_j$ is a unit matrix ($\mathbf{I}_0 = 1$). The recursion (2) starts with

$$\mathbf{R}_1 = \begin{bmatrix} R_0 & R_1 \\ R_1 & R_0 \end{bmatrix}, \quad R_0 = 0. \tag{3}$$

The parameters R_j determine the transfer rate on the individual hierarchy levels. The rate matrix given by

$$\tilde{\mathbf{R}}_L = \mathbf{R}_L - \mathbf{I}_L \sum_{j=1}^{L} R_j \tag{4}$$

accounts for detailed balance, i.e.,

$$\sum_{n=0}^{2^L-1} \tilde{\mathbf{R}}_{mn} = 0.$$

The rate matrix can be diagonalized by a similarity transform $\tilde{\mathbf{R}}_{Ld} = \mathbf{D}_L \tilde{\mathbf{R}}_L \mathbf{D}_L^{-1}$ where

$$\mathbf{D}_{j+1} = \tfrac{1}{2}\sqrt{2} \begin{bmatrix} \mathbf{D}_j & \mathbf{D}_j \\ -\mathbf{D}_j & \mathbf{D}_j \end{bmatrix}; \quad j = 0,1,\ldots, \quad \mathbf{D}_0 = 1. \tag{5}$$

The eigenstates of the rate matrix are labeled by $l = \sum_{j=1}^{L} l_j \times 2^{j-1}$ whose binary representation can be abbreviated by $[l] = (l_1,\ldots,l_L)$. The eigenvalues of the rate matrix $\tilde{\mathbf{R}}_L$ are

$$R_{[l]} = -2\sum_{j=1}^{L} l_j R_j, \tag{6}$$

the corresponding eigenstates are

$$|[l]\rangle = 2^{-L/2} q_L, \tag{7}$$

where q_L is defined by the following recursion relation:

$$q_{j+1} = [q_j, (1-2l_j)q_j], \qquad q_1 = 1, \quad j = 1,2,\ldots,L-1. \tag{8}$$

The stationary state with eigenvalue $R_{[0]} = 0$ is for instance given by

$$|[0]\rangle = 2^{-L/2}(1,1,\ldots,1).$$

We are now ready to write down exact solutions. The autocorrelation function describing the time decay of a state localized at site n is

$$\Phi(t) = \langle n | \exp(t\tilde{\mathbf{R}}_L) | n \rangle = \sum_{[l]} \prod_{j=1}^{L} \tfrac{1}{2} \exp(-2R_j l_j t). \tag{9}$$

All combinations of the factors $\frac{1}{2}\exp(-2R_j t)$ appear in the products of the above expression. Thus the time decay can also be written as

$$\Phi(t) = \prod_{j=1}^{L} [\tfrac{1}{2} + \tfrac{1}{2}\exp(-2R_j t)]. \tag{10}$$

This expression is identical to the time decay function for energy transfer from a donor to randomly distributed acceptors where the probability that an acceptor resides at a lattice site is given by $p = 0.5$.[1] The value $p \neq 0.5$ corresponds to asymmetric dimer clusters. The more general case of n different acceptor molecules corresponds to ultrametric spaces built with $(n+1)$-mer clusters. The detailed analytical expressions worked out for the energy-transfer problem[12] facilitate the evaluation of the time correlation function. A rectangular distribution of energy barriers with rates

$$R_{j+1} = R_{\max} \exp(-j\Delta\epsilon/k_B T), \qquad j = 0,1,\ldots,L-1, \tag{11}$$

corresponds to energy transfer by exchange interaction yielding an algebraic long-time decay

$$\Phi(t) \sim t^{-\alpha}, \quad \alpha = (k_B T/\Delta\epsilon)\ln(2).$$

The other important case corresponds to energy transfer by multipolar interactions:

$$R_{j+1} = R_{\max} j^{-1/\beta}, \qquad \beta = k_B T/\Delta\epsilon, \quad j = 1,2,\ldots,L, \tag{12}$$

where the long-time decay becomes a stretched exponential Kohlrausch law,

$$\Phi(t) \sim \exp(-ct^{\beta}), \quad \beta < 1.$$

In an infinite space ($L \to \infty$), $\Phi(t) = 0$ for $t > 0$ and $\beta > 1$. For finite L and $\beta > 1$, $\Phi(t) \sim \exp(-ct)$.

Next we consider diffusion. The average distance the system has moved at time t starting from the local site j

is

$$x_j(t) = \Delta x \sum_{n=0}^{2^L-1} (n-j) p_{jn}(t), \quad (13)$$

where

$$p_{jn}(t) = \langle j | \exp(\tilde{\mathbf{R}}_L t) | n \rangle$$

is the conditional probability that the system is at $t=0$ at site j and at time t at site n. A generating function is introduced:

$$A_j(t) = \langle j | \exp(\tilde{\mathbf{R}}_L t) | \alpha_j \rangle,$$

where

$$|\alpha_j\rangle = e^{-j\alpha}(1, e^{\alpha}, e^{2\alpha}, \ldots, e^{(2^L-1)\alpha})$$

such that

$$x_j(t) = \Delta x [dA_j(t)/d\alpha]_{\alpha=0}. \quad (14)$$

After some algebra one obtains with the binary representation $j = \sum_{i=1}^{L} j_i 2^{i-1}$ for the generating function

$$A_j(t) = e^{-j\alpha} 2^{-L} \prod_{i=1}^{L} [(1+e^{\alpha 2^{i-1}}) + (1-2j_i) e^{-2R_i t}(1-e^{\alpha 2^{i-1}})] \quad (15)$$

and with Eq. (14)

$$x_j(t)/\Delta x = -j + \sum_{i=1}^{L} 2^{i-2}[1-(1-2j_i)e^{-2R_i t}].$$

With use of the generating function $A_j(t)$, arbitrary moments of the distance distribution can be calculated. With a rectangular distribution of energy barriers (11) the average distance for diffusion at long times behaves as

$$x_0(t) \sim t^{\alpha} \text{ for } \alpha = (k_B T/\Delta\epsilon)\ln(2) < 1$$

and as

$$x_0(t) \sim t \text{ for } \alpha > 1.$$

At $\alpha < 1$, $x_0(t) \sim 1/\Phi(t)$ as is typical for compact exploration.[1] For $\alpha > 1$ the character of the diffusion process changes to an open exploration of the ultrametric space. The long-time behavior of $x_0(t)$ corresponding to the Kohlrausch law (12) is for a large but finite number of hierarchy levels $x_0(t) \sim t$. Thus the exploration is open for all values of β.

Finally the intermediate scattering function is evaluated which can be defined as

$$I(k,t) = \langle f | \exp(\tilde{\mathbf{R}}_L t) | f \rangle \quad (16)$$

with states

$$|f\rangle = 2^{-L/2}(1, f, f^2, \ldots, f^{2^L-1})$$

and a phase factor $f = \exp(i\Delta x k)$. It can be cast into a form which is reminiscent of Eqs. (10) and (15),

$$I(k,t) = \prod_{i=1}^{L} [1 - \sin^2(2^{i-2}\Delta x k) + \sin^2(2^{i-2}\Delta x k) e^{-2R_i t}]. \quad (17)$$

It is analogous to the problem of energy transfer from a donor to acceptors where the acceptor distribution is not completely at random but exhibits a periodic structure. It is determined by the probability $p_i = \sin^2(2^{i-2}\Delta x k)$ to meet acceptors at a given hierarchy level i. The effect of this hierarchy dependence is to reduce the effective number of hierarchies which are involved. In the special case where $2^i \Delta x k = \pi$ at a certain i, all hierarchy levels greater than i do not contribute to the intermediate scattering function (17). Thus unless this effect reduces the number of hierarchies dramatically the long-time behavior of the intermediate scattering function (17) and of the autocorrelation function are equal.

In summary, a new type of hierarchical space has been considered. Its self-similar coupling scheme imposes an approximate ultrametric topology. A major difference from other ultrametric spaces is the pairwise connection of states on each hierarchy level such that the state distribution enforced by dynamics on a given set of hierarchy levels is not wiped out by dynamics on the next higher hierarchy level.

Exact solutions are available. For energy barriers increasing linearly or logarithmically with the hierarchy level, the time decay pattern is algebraic or stretched exponential, respectively. Diffusion processes on such a space correspond for a stretched exponential decay law always to an open exploration. For an algebraic decay the diffusion process is an open exploration at high temperatures $[\alpha = (k_B T/\Delta\epsilon)\ln(2) > 1]$ and a compact exploration with a slowed down diffusion at lower temperatures $[\alpha < 1]$. The present model of an ultrametric space does not exhibit anomalous accelerated diffusion which is observed in the model of Wegner and Grossmann.[7] This major difference is due to the influence of intermediate states at higher hierarchies which are responsible for the richer dynamical behavior of their model. In the present model such intermediate states are absent.

It is a surprising fact that relaxation processes in hierarchical spaces which posses a sequential coupling scheme and the energy-transfer process from a donor to randomly distributed acceptors which is based on a parallel coupling scheme lead to the same expressions for time decay. With the simple analytical expressions at hand decay patterns at intermediate times and the range of validity of the long-time decay laws can be studied in more detail. Furthermore, it becomes possible to construct hierarchical spaces whose topologies give rise to different decay laws in different time regimes, a situation which is often met in complex systems.

The author would like to thank Professor S. F. Fischer for discussions and generous support. Financial support by the Deutsche Forschungsgemeinschaft with a Heisenberg Fellowship and the project Sonderforschungsbereich No. 143 C3 are gratefully acknowledged.

[1]A. Blumen, J. Klafter, and G. Zumofen, in *Optical Spectroscopy of Glasses,* edited by I. Zschokke (Reidel, Dordrecht, 1986), p. 199.

[2]K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).

[3]R. H. Austin, K. W. Beeson, L. Eisenstein, H. Frauenfelder, and I. C. Gunsalus, Biochemistry **14**, 5355 (1975).

[4]A. Ansari, Y. Berendzen, S. F. Bowne, H. Frauenfelder, I. E. T. Iben, T. B. Sauke, E. Shyamsunder, and R. D. Young, Proc. Nat. Acad. Sci. U.S.A. **82**, 5000 (1985).

[5]J. Hopfield and D. W. Tenk, Science **233**, 625 (1985).

[6]S. Kirkpatrick, C. D. Gelatt, Jr., and M. P. Vecchi, Science **220**, 671 (1983).

[7]F. Wegner and S. Grossmann, Z. Phys. B **59**, 197 (1985).

[8]E. W. Knapp, S. F. Fischer, and F. Parak, J. Chem. Phys. **78**, 4701 (1983).

[9]R. G. Palmer, D. L. Stein, E. Abrahams, and P. W. Anderson, Phys. Rev. Lett. **53**, 958 (1984).

[10]M. Mezard, G. Parisi, N. Sourlas, G. Toulouse, and M. Virasoro, Phys. Rev. Lett. **52**, 1156 (1984).

[11]A. T. Ogielski and D. L. Stein, Phys. Rev. Lett. **55**, 1634 (1985).

[12]A. Blumen and J. Manz, J. Chem. Phys. **71** 4694 (1979); A. Blumen, J. Chem. Phys. **72**, 2632 (1980).