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Anomalous temperature dependence of diffusion coefficient

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A model is proposed which yields an anomalous diffusion coefficient, with a maximum in its temperature dependence, in contrast to the linear temperature dependence of simple diffusion processes. It is found that the anomalous temperature dependence of this diffusion coefficient is obtained by taking into account "memory effects" or "correlation" in the usual random walks. As an example, random motions of paramecium, which show the anomalous temperature dependence of this diffusion coefficient, are considered.

I. INTRODUCTION

For many problems it is important to use a stochastic approach. The basic equations describe Markovian processes in which memory or correlation effects are not taken into account. Recently, however, non-Markovian character has been observed in many fields.[1] Specifically, in rapidly relaxed systems,[2] rapidly quenched systems,[3] disordered lattice systems,[4] and ecological systems,[5] the processes show anomalous behaviors which cannot be described by the usual random walks. The anomalous behaviors arise from nonlinear, non-Markovian diffusing elements.

Kawakubo and Tsuchiya[6] observed interesting behavior in the random motions of paramecium, which show an anomalous temperature dependence of their diffusion coefficient: The diffusion coefficient has a maximum value at the temperature at which they were cultivated. The anomalous behavior seems to come from memory effects or correlations between elements, proper to biological species.

So far, the random-walk theory has been applied to various problems and valuable results have been obtained.[7] For the nonlinear, non-Markovian processes, however, the usual random walks have to be generalized. For this purpose a generalization of the random walks has been proposed in previous papers.[8,9]

In this paper, the generalized random walk (GRW) having memory or correlation effects between the walker's behaviors is applied to setting up a model yielding the anomalous behavior mentioned above.[10] Here we focus our attention on the "random motions" of paramecium (the walker), which are studied as diffusion processes.

II. GENERALIZED RANDOM WALKS

We review the basic expressions of the generalized random walk.[8–10] The recursion relation for the GRW reads

$$W(m,N)=\sum_{\alpha=\pm,0}\tilde{P}^{\alpha}_{N-1}(m\mid m-\alpha\cdot 1)W(m-\alpha\cdot 1,N-1)\ , \tag{1}$$

where

$$\sum_{\alpha}\tilde{P}^{\alpha}_{N}(m+\alpha\cdot 1\mid m)=1 \tag{2}$$

and $W(m,N)$ is the probability that a walker starting at the origin arrives at the site m after N steps. The symbol $\tilde{P}^{\alpha}_{N-1}(m\mid m-\alpha\cdot 1)$ represents the probability of jumping from the site $m-\alpha\cdot 1$ to the site m at the $(N-1)$th step. The summation over α in Eqs. (1) and (2) goes over $\alpha=+,-$, and 0. Memory effects in the processes are considered in terms of a function f connecting the jumping probabilities with a set of the corresponding ones at previous steps;

$$\tilde{P}^{\alpha}_{N}=f(\{\tilde{P}^{\alpha}_{N-k}\})\quad (k=1,2,\ldots,N)\ . \tag{3}$$

Here, for simplicity we have omitted the m dependence of $\tilde{P}^{\alpha}_{N}$. The continuum limit of the recursion relation specified by Eqs. (1) and (3) leads us to a nonlinear Fokker-Planck (FP) equation

$$\frac{\partial w}{\partial t}=-\frac{\partial}{\partial x}\frac{a}{t_0}[\tilde{p}^{+}(t)-\tilde{p}^{-}(t)]w+\frac{\partial^2}{\partial x^2}\frac{a^2}{2t_0}[1-\tilde{p}^{0}(t)]w\ , \tag{4}$$

where the jumping probabilities satisfy the relation $\sum_{\alpha}\tilde{p}^{\alpha}(t)=1$. In the derivation of Eq. (4), we have set

$$x=ma,\quad t=Nt_0 \tag{5}$$

(a equals the unit distance, t_0 equals the unit time) in Eq. (1) and expanded the corresponding quantities around $a=0$ and $t_0=0$ while keeping a^2/t_0 constant.[8,9] Note that the quantities w and $\tilde{p}^{\alpha}(t)$ are continuous ones, corresponding to W and $\tilde{P}^{\alpha}_{N}$, respectively.

A. Simple memory case

Firstly, we consider a simple case for the memory, in which the jumping probabilities are specified by the walker's "memory box": The f in Eq. (3) for $\tilde{P}^{\alpha}_{N}$ is given in the form[9]

$$\tilde{P}^{\alpha}_{N}=C_{N-1}(\alpha\mid\alpha)\tilde{P}^{\alpha}_{N-1}+C_{N-1}(\alpha\mid-\alpha)\tilde{P}^{-\alpha}_{N-1} \tag{6}$$

(P^{0}_{N} represents N-independent P^{0}), for $\alpha=\pm$ and

$C_{N-1}(\alpha\,|\,\alpha')$ is a correlation factor (or transition rate) between $\widetilde{P}_N^{\alpha}$ and $\widetilde{P}_{N-1}^{\alpha}$ or $\widetilde{P}_{N-1}^{-\alpha}$. The correlation factors satisfy

$$\sum_{\alpha=\pm}{}' C_{N-1}(\alpha\,|\,\alpha')=1\ . \tag{7}$$

A prime on the summation sign means that the case $\alpha=0$ is excluded. The continuum limit of the relation (6) leads to

$$\frac{\partial\widetilde{p}^{\,\alpha}(t)}{\partial t}=c(\alpha\,|\,-\alpha)\widetilde{p}^{\,-\alpha}(t)-c(-\alpha\,|\,\alpha)\widetilde{p}^{\,\alpha}(t) \tag{8}$$

[$\widetilde{p}_0(t)$ equals t independent p_0], where $c(\alpha\,|\,\alpha')=C_{N=t/t_0}(\alpha\,|\,\alpha')/t_0$. With the aid of $\sum_{\alpha=\pm,0}\widetilde{p}^{\,\alpha}(t)=1$, where $\widetilde{p}^{\,0}(t)=p_0$, the solution of Eq. (8) reads

$$\widetilde{p}^{\,\pm}(t)=\widetilde{p}^{\,\pm}(0)e^{-2c_1t}+\widetilde{p}^{\,\pm}(\infty)(1-e^{-2c_1t}) \tag{9}$$

where

$$c_1=[c(+\,|\,-)+c(-\,|\,+)]/2\ , \tag{10a}$$

$$\widetilde{p}^{\,\pm}(\infty)=\frac{c(\pm\,|\,\mp)(1-p_0)}{c(+\,|\,-)+c(-\,|\,+)}\ , \tag{10b}$$

and the $\widetilde{p}^{\,\pm}(0)$ are initial values satisfying $\widetilde{p}^{\,\pm}(0)=(1-p_0)/2$. A quantity defined by

$$Q(t)=\widetilde{p}^{\,+}(t)-\widetilde{p}^{\,-}(t) \tag{11}$$

is therefore written from Eq. (9) as follows:

$$Q(t)=Q(0)e^{-2c_1t}+Q(\infty)(1-e^{-2c_1t})\ . \tag{12}$$

For a homogeneous system expressed by $c(+\,|\,-)=c(-\,|\,+)$, the $Q(t)$ given in Eq. (11) is reduced to $Q(t)=Q(0)\exp(-c_0t)$, where $c_0=c(+\,|\,-)$ or $c(-\,|\,+)$. It is important to note that the recursion relation (6) itself is Markovian but the recursion relation (1) [or (4)] specialized by Eq. (6) [or Eq. (8)] is non-Markovian.

B. More general case

Here we consider the more general case that the jumping probabilities are determined by the walker's n memory boxes. This model yields the anomalous temperature dependence of the diffusion coefficient. In this model, each memory box can store a direction denoted by α_i ($i=1,2,\ldots,n$), and the direction is correlated with other ones. To represent the states of the n memory boxes, we use a set of directions α_i ($i=1,2,\ldots,n$) and express it by new probability functions $P_N^{\alpha_1,\alpha_2,\ldots,\alpha_n}$. The $P_N^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ is a generalization of $\widetilde{P}_N^{\alpha}$ used in Sec. II A.

Step (time) evolution of the $P_N^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ is supposed to be expressed by the corresponding probabilities $P_{N-1}^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ and the correlation factors $C_{N-1}(\alpha_i\{\alpha_j\}\,|\,\alpha_i'\{\alpha_j\})$ in a recursive form; cf. Eq. (6). The $C_{N-1}(\alpha_i\{\alpha_j\}\,|\,\alpha_i'\{\alpha_j\})$ means a correlation factor (or transition rate) of the ith memory box from α_i' to α_i, with the other memory boxes denoted by α_j fixed.

In the continuum limit, the time evolution of $p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)$ corresponding to $P_N^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ reads

$$\begin{aligned}\frac{\partial p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)}{\partial t}=&\sum_{i=1}^{n}c(\alpha_i\{\alpha_j\}\,|\,-\alpha_i\{\alpha_j\})p^{\alpha_1,\alpha_2,\ldots,-\alpha_i,\ldots,\alpha_n}(t)\\&-\sum_{i=1}^{n}c(-\alpha_i\{\alpha_j\}\,|\,\alpha_i\{\alpha_j\})p^{\alpha_1,\alpha_2,\ldots,\alpha_i,\ldots,\alpha_n}(t)\end{aligned} \tag{13}$$

[$p^{0,0,\ldots,0}(t)$ equals t-independent p_0], for α_i ($i=1,2,\ldots,n$)$=\pm$. The correlation factor

$$c(\alpha_i\{\alpha_j\}\,|\,\alpha_i'\{\alpha_j\})$$

is

$$C_{N=t/t_0}(\alpha_i\{\alpha_j\}\,|\,\alpha_i'\{\alpha_j\})/t_0\ .$$

Summation of the $p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)$ with respect to $\{\alpha\}$ leads to a t-independent quantity:

$$\sum_{\{\alpha\}}p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)=1-p_0\ .$$

The summation denotes a sum taken over all 2^n configurations for n directions. Also, the summation over $\{\alpha\}$ of $c(\alpha_i\{\alpha_j\}\,|\,\alpha_i'\{\alpha_j\})$ gives a t-independent result, normalization

$$\sum_{\{\alpha\}}c(\alpha_i\{\alpha_j\}\,|\,\alpha_i\{\alpha_j\})=1\ .$$

If we regard the $c(\alpha_i\{\alpha_j\}\,|\,\alpha_i'\{\alpha_j\})$ as a probability of "flipping of directions" from α_i' to α_i, the present model leads to a version of the Glauber model for Ising spin.[11]

Let $\alpha_i^k(t)\rangle$ be an expectation value[12] defined by

$$\langle\alpha_i^k(t)\rangle=\sum_{\{\alpha\}}\alpha_i^k p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)\quad(k=1,2)\ . \tag{14}$$

Here we restrict the consideration to a case where the expectation values $\langle\alpha_i(t)\rangle$ and $\langle\alpha_i^2(t)\rangle$ specialize the jumping probabilities $\widetilde{p}^{\,\pm}(t)$ used in Eq. (4) as follows:

$$\begin{aligned}\frac{1}{n}\sum_{i=1}^{n}\langle\alpha_i^k(t)\rangle&=\widetilde{p}^{\,+}(t)+(-1)^k\widetilde{p}^{\,-}(t)\\&\equiv\langle\langle\alpha^k(t)\rangle\rangle\quad(k=1,2)\ ,\end{aligned} \tag{15a}$$

or

$$\begin{aligned}&\widetilde{p}^{\,\pm}(t)=\tfrac{1}{2}[(1-p_0)\pm\langle\langle\alpha(t)\rangle\rangle]\ ,\\&\widetilde{p}^{\,0}(t)=p_0\ ,\end{aligned} \tag{15b}$$

where we have used $\widetilde{p}^{\,+}(t)+\widetilde{p}^{\,-}(t)+p_0=1$. The statement (15a) or (15b) means that $Q(t)$ in Eq. (11) is expressed by

$$Q(t)=\langle\langle \alpha(t)\rangle\rangle \ . \tag{16}$$

The time evolution of $Q(t)$ is written in the form

$$\langle\langle \dot{\alpha}(t)\rangle\rangle=\frac{1}{n}\sum_{i=1}^{n}\langle \dot{\alpha}_i(t)\rangle \ , \tag{17a}$$

where

$$\langle \dot{\alpha}(t)\rangle=-2\langle \alpha_i c(-\alpha_i\{\alpha_j\}\,|\,\alpha_i\{\alpha_j\})\rangle \tag{17b}$$

from Eqs. (13) and (14). For the derivation of Eq. (17b), we have used the relation $\sum_i \alpha_i\cdot 1=0$ and replaced α_i by $-\alpha_i$ in the summation for $\alpha_i c(\alpha_i\{\alpha_j\}\,|-\alpha_i\{\alpha_j\})$.

In the following, we consider processes in which the correlation factor $c(-\alpha_i\{\alpha_j\}\,|\,\alpha_i\{\alpha_j\})$ is given by

$$c(-\alpha_i\{\alpha_j\}\,|\,\alpha_i\{\alpha_i\})=\frac{c_0}{2}\left[1-\frac{\gamma}{2}\alpha_i(\alpha_{i-1}+\alpha_{i+1})\right] , \tag{18}$$

where c_0 is $1/t_0$ and γ is a constant. With Eq. (18) and $\alpha_i^2\cdot 1=1$, the expression (17b) can be rewritten as follows:

$$\frac{1}{c_0}\langle \dot{\alpha}_i(t)\rangle=-\langle \alpha_i(t)\rangle+\frac{\gamma}{2}[\langle \alpha_{i-1}(t)\rangle+\langle \alpha_{i+1}(t)\rangle] \ . \tag{19}$$

The specialization $\gamma=0$ leads to the case considered previously below Eq. (12).

III. EVALUATION OF $\langle\langle \alpha(t)\rangle\rangle$

In this section, to calculate the $\langle\langle \alpha(t)\rangle\rangle$ explicitly, we must consider some parameters specifying the model. Firstly, we introduce a coupling constant between nearest-neighbor directions in the walker's n memory boxes. Secondly, we specify the temperature of the environment of the walker and introduce a probability that the ith box stores a direction α_i.

The coupling constant J specializes "correlations" of the directions stored in the n memory boxes in the form

$$J\alpha_i\alpha_j \ . \tag{20}$$

Let the probability $\rho(\alpha_i)$ that the direction of the ith box is α_i be

$$\rho(\alpha_j)\propto e^{-\beta\mathscr{H}_j} \quad (\beta=1/k_BT) \ , \tag{21}$$

where k_B is the Boltzmann constant and T is the temperature mentioned above. The quantity $\mathscr{H}_j$ characterizes the memory in terms of the correlation between directions,

$$\mathscr{H}_j=-\frac{J}{2}\alpha_j(\alpha_{j-1}+\alpha_{j+1}) \ . \tag{22}$$

The statement (21) with Eq. (22) implies that the nearest-neighbor directions show behaviors similar to that of ferromagnet when $J>0$. The function $\mathscr{H}\,(=\sum_{j=1}^{n}\mathscr{H}_j)$ is a "Hamiltonian," which determines the correlation between directions in the n memory boxes.

Steady state of $p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)$ in Eq. (13) leads us to the detailed balance expressed by the form

$$\frac{c(-\alpha_i\{\alpha_j\}\,|\,\alpha_i\{\alpha_j\})}{c(\alpha_i\{\alpha_j\}\,|-\alpha_i\{\alpha_j\})}=\frac{p_{\rm st}^{\alpha_1,\alpha_2,\ldots,-\alpha_i,\ldots,\alpha_n}}{p_{\rm st}^{\alpha_1,\alpha_2,\ldots,\alpha_i,\ldots,\alpha_n}} \ , \tag{23}$$

where $p_{\rm st}^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ denotes the steady state of $p^{\alpha_1,\alpha_2,\ldots,\alpha_n}(t)$. Here we impose the periodicity condition, $\alpha_{n+1}=\alpha_1$, and rewrite the $p_{\rm st}^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ by $\exp(-\beta\mathscr{H})$. Then the $p_{\rm st}^{\alpha_1,\alpha_2,\ldots,\alpha_n}$ in Eq. (23) becomes the $\rho(\alpha_i)$ given by Eq. (21), and algebraic manipulation of the result leads to an expression for γ,

$$\gamma=\tanh\beta J \ , \tag{24}$$

where the parameters J and T are defined by Eqs. (20) and (21), respectively. Here note that the expression (19) is obtained from the equation[13]

$$\frac{d}{dt}\langle \alpha_i(t)\rangle=-\frac{1}{\tau_0}[\langle \alpha_i(t)\rangle-\langle \alpha_i(t)\rangle_e] \ , \tag{25}$$

where

$$\tau_0=\frac{1}{c_0} \ , \tag{26}$$

$$\langle \alpha_i(t)\rangle_e=\frac{\sum\limits_{\alpha_i}\alpha_i\exp\{-\frac{1}{2}\beta J\alpha_i[\langle \alpha_{i-1}(t)\rangle+\langle \alpha_{i+1}(t)\rangle]\}}{\sum\limits_{\alpha_i}\exp\{-\frac{1}{2}\beta J\alpha_i[\langle \alpha_{i-1}(t)\rangle+\langle \alpha_{i+1}(t)\rangle]\}} \ . \tag{27}$$

The nonlinear FP equation (4) having the memory effects specialized by Eqs. (13), (15a), and (15b) is reduced to

$$\frac{\partial w}{\partial t}=-\frac{\partial}{\partial x}\frac{F_0}{\eta}w+D\frac{\partial^2}{\partial x^2}w \ , \tag{28}$$

where

$$F_0=M\frac{a}{t_0^2} \ , \tag{29}$$

$$D=\frac{a^2}{2t_0}[1-p_0(T)] \ , \tag{30a}$$

$$\frac{1}{\eta}=\frac{t_0}{M}[\tilde{p}^{\,+}(t)-\tilde{p}^{\,-}(t)]=\frac{1}{\eta_0}\langle\langle \alpha(t)\rangle\rangle \quad (\eta_0=M/t_0) \ , \tag{30b}$$

and M is the mass of the diffusion particles.

In Eq. (30a), we rewrite the constant p_0 in Eq. (15b) as $p_0(T)$, which indicates the temperature dependence. From the expression (25) with Eqs. (26) and (27), we note that for $t>\tau_0$ we can use a simplified relation for $\langle\langle \alpha(t)\rangle\rangle$ in the steady state; that is,

$$\langle\langle \alpha\rangle\rangle=\tanh(\beta J)\langle\langle \alpha\rangle\rangle \ , \tag{31a}$$

where $\langle\langle \alpha\rangle\rangle$ denotes the value of $\langle\langle \alpha(t)\rangle\rangle$ $[=(1/n)\sum_{i=1}^{n}\langle \alpha_i(t)\rangle]$ in the steady state and leads to the relation

$$\langle\langle \alpha\rangle\rangle=[\tilde{p}^{\,+}(t)-\tilde{p}^{\,-}(t)]_{t\to\tau} \quad (\tau>\tau_0) \ . \tag{31b}$$

Graphical consideration for Eq. (31a) leads us to a familiar curve for $\langle\langle \alpha\rangle\rangle/\langle\langle \alpha\rangle\rangle_0$ as shown in Fig. 1, where

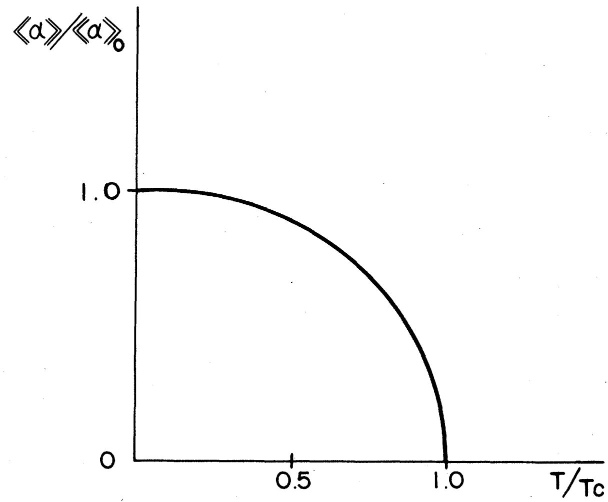

FIG. 1. Qualitative behavior of $\langle\langle\alpha\rangle\rangle/\langle\langle\alpha\rangle\rangle_0$.

$\langle\langle\alpha\rangle\rangle_0$ is an expectation value independent of T. In Fig. 1, T_c $(=J/k_B)$ is a critical temperature defined by the parameter J which specializes the coupling or the correlation between the directions α_i and α_j stored in the ith and jth memory boxes.

Here we postulate that the steady state of $w(x,t)$ and $w_{\rm st}(x)$ is expressed in the form

$$w_{\rm st}(x)=e^{-\lambda\phi(x)/k_BT}\,, \tag{32}$$

where ϕ is a potential and its derivative is a force $F_0(=-\partial\phi/\partial x)$, and the λ is a numerical factor adjusting the behavior of $w_{\rm st}(x)$ in the interval $[0,L]$ under consideration. The steady state then becomes

$$w_{\rm st}(x)=e^{\lambda F_0 x/k_BT}\,. \tag{33}$$

The condition that the local flux is balanced in the steady state gives the Einstein relation

$$D=\frac{k_BT}{\lambda\eta_0}\langle\langle\alpha\rangle\rangle\,. \tag{34}$$

From substitution of the solution determined by Eq. (31a) into Eq. (34), we are able to get behavior as shown in Fig. 2. If we neglect the temperature dependence of $\langle\langle\alpha\rangle\rangle$ arising from the memory effects, $\langle\langle\alpha\rangle\rangle$ becomes a constant. Relation (34) then leads to the usual Einstein relation, $D=k_BT/\lambda\eta_0$. A physical reason for why $\langle\langle\alpha\rangle\rangle$ exhibits a maximum in its temperature dependence is that, by considering the walker's memory, we were able to introduce correlation effects, which are strongly influenced by the environment-specified temperature T. The memory effects, mathematically, lead to non-Markovian processes. The strongly influenced motions yield an anomaly in the diffusion coefficient which is not expressed by the usual linear-T dependence of the environment.

IV. RANDOM MOTIONS OF PARAMECIA

With the aid of Eq. (34), we can study the random motions of paramecia. Figure 3 shows the diffusion coefficients for the paramecia observed by Kawakubo and Tsuchiya.[6] They find that the behavior of $w(x,t)$ quickly approaches "uniform distribution" after the initial distribution; see Fig. 4.

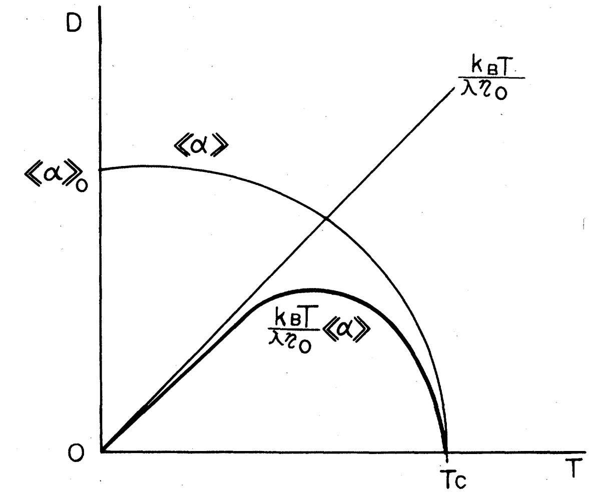

FIG. 2. Temperature dependence of diffusion coefficient D. Linear curve $(k_BT/\lambda\eta_0)$ denotes a usual temperature dependence of diffusion coefficient. The linear curve multiplied by $\langle\langle\alpha\rangle\rangle$ leads to the D showing a maximum in its temperature dependence.

The temperature dependence of the diffusion coefficient obtained with Eq. (34) shows a behavior similar to the observed ones. For numerical evaluation, we take the following units:

$$t_0\sim10^{-2}\ \text{sec},\quad a\sim2\times10^{-2}\ \text{cm}\,. \tag{35}$$

These values are estimated from the diffusion coefficient $(\sim4\times10^{-2}\ \text{cm}^2/\text{sec})$ and the velocity of paramecia $(\sim1$ mm/sec) observed experimentally.[6,14] Furthermore, we

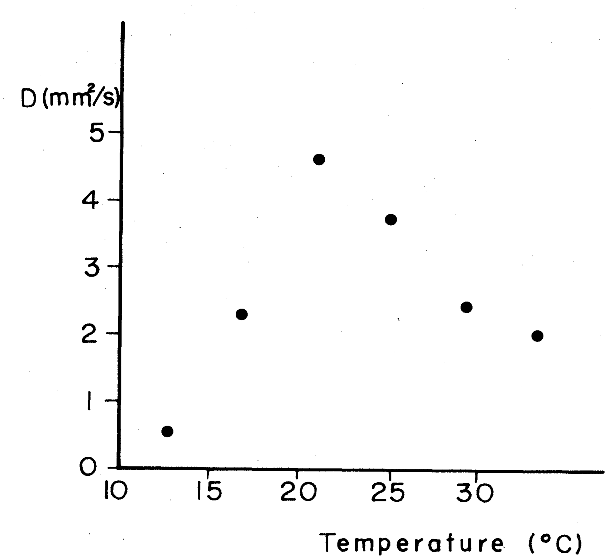

FIG. 3. Observed temperature dependence of the diffusion coefficient for paramecium (Ref. 6).

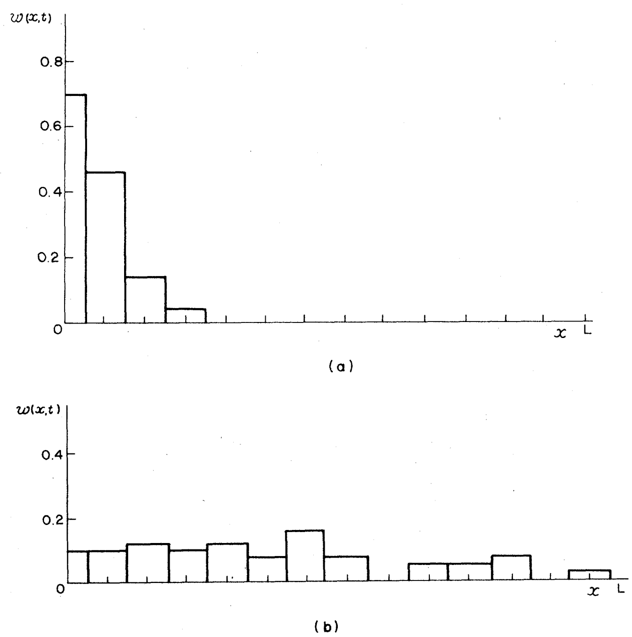

FIG. 4. Distributions observed for $w(x,t)$ due to Kawakubo and Tsuchiya (Ref. 6). (a) An initial distribution of $w(x,t)$ at $t=0$. (b) A "uniform distribution" observed after 7.5 sec; we regard it as a steady state in Eq. (33).

set $M \sim 10^{-4}$ g, $L \sim 10$ cm, $J=5.5\times10^{-15}$ ergs ($T_c=40\,^{\circ}\mathrm{C}$), $\lambda \sim 10^{-11}$, and $\langle\langle \alpha \rangle\rangle \sim \langle\langle \alpha \rangle\rangle_0 \sim 5\times10^{-2}$. Here we regard the uniform distribution shown in Fig. 4(b) as the distribution in the steady state $w_{st}(x)$. Agreement of the distribution (33) with the observed ones is not clear. However, it is important to note that the values taken above are not essential to the present qualitative temperature dependence of D. In the present model, the memory effects are specified by Eqs. (13), (15a), (15b), (21), and (22), and we consider a single walker, but we can treat many walkers by using the coupled random walks.[15] If we use the two-dimensional (2D) GRW and the Work-Fujita model[16] for the jumping probabilities, the present treatment leads to a more realistic 2D model for the random motions of paramecia.

Finally we note that there are other interesting phenomena, similar to the present, with anomalous temperature dependence of the diffusion coefficient: The longitudinal diffusion coefficient of a spherical particle shows a maximum with respect to the radius,[17] diffusion processes in a magnetic field yield diffusion coefficients showing a maximum with respect to the field parameter.[18] The anomalous behavior of memory effects in solid-state tracer diffusion processes have been extensively studied.[19]

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