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Statistical Properties of the 2D Attached Rouse Chain

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We study various dynamical properties (winding angles, areas) of a set of harmonically bound Brownian particles (monomers), one endpoint of this chain being kept fixed at the origin 0. In particular, we show that, for long times t, the areas $\{A_i\}$ enclosed by the monomers scale like $t^{1/2}$, with correlated gaussian distributions. This has to be compared to the winding angles $\{\theta_i\}$ around fixed points that scale like t and are distributed according to independent Cauchy laws.

KEY WORDS: Brownian motion; Rouse chain; path integrals; perturbation theory.

In this paper, we will study the planar motion of a chain of n harmonically bound Brownian particles. This model is usually referred to in the literature as the Rouse chain[(1)] and has shown to be historically very important in polymer science.[(2, 3)]

Physically, such a model can describe the motion of a polymer adsorbed on a solid substrate, the Brownian dynamics resulting from thermal activation induced by lattice vibrations.[(4)]

On another hand, a planar random walk can be considered as the projection of a 3D directed polymer. For instance, in ref. 5, windings of such polymers around a rod have been related to localization properties. Following this approach, a given trajectory of the Rouse chain will correspond to a conformation of n quasi-aligned *interacting* polymers; so, the chain dynamics will give access to static properties of such polymers.

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In the sequel, we will consider such a chain attached at the origin 0 and examine some of its winding properties. Describing a given configuration of the chain by a complex n-vector z (the components z_i, $i = 1,..., n$, are the complex coordinates of the particles), we consider the set of all the closed trajectories of length t, i.e., $z(t) = z(0)$, and this for all the starting configurations $z(0)$. Practically, we will not weight the starting configurations with any thermodynamical factor. We are aware that this approach is quite different from the one taken in polymer physics[6] where, at $t = 0$, the chain is supposed to be in equilibrium with the environment at some finite temperature T.

A_j and θ_j being the area enclosed by the jth particle's trajectory and its winding angle around 0, our goal is to compute the joint probability distributions $P(\{A_i\})$ and $P(\{\theta_i\})$ in the limit $t \to \infty$, n being kept fixed. In order to make comparisons, we now recall some of the results concerning the planar Brownian motion.

We first quote the area and winding angle distributions, respectively $P(A)$ (Lévy's law[7]) and $P(\theta)$ (Spitzer's law[8]) for a particle allowed to wander everywhere in the plane:

$$P(A) = \frac{\pi}{2t} \frac{1}{\cosh^2 \pi A/t} \tag{1}$$

$$P(\theta) = \frac{2}{\pi \ln t} \frac{1}{1 + (2\theta/\ln t)^2} \tag{2}$$

(the last one holds, in the limit $t \to \infty$, for open curves, the final point being left unspecified).

Those two laws were obtained more than 40 years ago and since that time many refinements have been brought. For instance, in ref. 9, the authors pointed out the importance of the small windings occuring when the particle is close to 0. Excluding an arbitrary small zone around 0, they showed that the variance $\langle \theta^2 \rangle$ becomes finite in contrast with the Spitzer's result, Eq. (2).

On the other hand, for Brownian motion on bounded domains,[10, 11] the scaling variables in the limit $t \to \infty$, become, resp., $A/\sqrt{t}$ and θ/t with still an infinite variance $\langle \theta^2 \rangle$. We close here this brief recall and start our chain study with the following set of coupled Langevin equations:

$$\begin{aligned} \dot{z}_1 &= k(z_2 - 2z_1) + \eta_1 \\ \dot{z}_l &= k(z_{l+1} + z_{l-1} - 2z_l) + \eta_l, \qquad 2 \leqslant l \leqslant n-1 \\ \dot{z}_n &= k(z_{n-1} - z_n) + \eta_n \end{aligned} \tag{3}$$

where k is the spring constant and η_m $(\equiv \eta_{mx} + i\eta_{my})$ a gaussian white noise:

$$\langle \eta_m(t) \rangle = 0$$

$$\langle \eta_m(t)\, \eta_{m'}(t') \rangle = 2\delta_{mm'}\, \delta(t - t') \tag{4}$$

Introducing the complex n-vector η, Eq. (3) can be written in a matrix form:

$$\dot{z} = -k\mathbf{M}z + \eta \tag{5}$$

where $\mathbf{M}$ is the tridiagonal $(n \times n)$ matrix:

$$\mathbf{M} = \begin{pmatrix} 2 & -1 & 0 & \cdots & 0 \\ -1 & 2 & -1 & \cdots & 0 \\ 0 & -1 & 2 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \cdots & 1 \end{pmatrix}$$

with an inverse given by:

$$\mathbf{M}^{-1} = \begin{pmatrix} 1 & 1 & 1 & \cdots & 1 \\ 1 & 2 & 2 & \cdots & 2 \\ 1 & 2 & 3 & \cdots & 3 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 1 & 2 & 3 & \cdots & n \end{pmatrix}$$

The eigenvalues of $\mathbf{M}$ are:

$$\omega_j = 2\left(1 - \cos\frac{\pi(2j-1)}{2n+1}\right), \qquad 1 \leqslant j \leqslant n \tag{6}$$

With the matrix $\omega = \mathrm{diag}(\omega_i)$, we can write:

$$\omega = \mathbf{R}^{-1}\mathbf{M}\mathbf{R} \tag{7}$$

$$z = \mathbf{R}Z \tag{8}$$

($\mathbf{R}$ is an orthogonal matrix; the components of Z are the normal coordinates).

Let us call $\mathscr{P}(z, z_0, t)$ the probability for the chain to go from z_0 at $t=0$ to z at time t. $\mathscr{P}$ satisfies a Fokker–Planck equation:[12]

$$\partial_t \mathscr{P} = ({}^t\partial_z k\mathbf{M} z + {}^t\partial_{\bar{z}} k\mathbf{M}\bar{z} + 2\,{}^t\partial_{\bar{z}}\partial_z)\,\mathscr{P} \tag{9}$$

where ∂_z (resp. $\partial_{\bar{z}}$) is a n-vector of components ∂_{z_i} (resp. $\partial_{\bar{z}_i}$) and ${}^t\partial_z$ (resp. ${}^t\partial_{\bar{z}}$) is the transpose of ∂_z (resp. $\partial_{\bar{z}}$). The solution can be written in terms of a path integral ($\mathscr{D}z\,\mathscr{D}\bar{z} = \prod_{i=1}^n \mathscr{D}z_i\,\mathscr{D}\bar{z}_i$):

$$\begin{aligned}\mathscr{P}(z, z_0, t) &= \det(e^{tk\mathbf{M}}) \int_{z(0)=z_0}^{z(t)=z} \mathscr{D}z\,\mathscr{D}\bar{z}\,\exp\left(-\tfrac{1}{2}\int_0^t {}^t(\dot{\bar{z}} + k\mathbf{M}\bar{z})(\dot{z} + k\mathbf{M}z)\,d\tau\right)\\ &\equiv F(z, z_0, t)\,.\,G(z, z_0, t)\end{aligned} \tag{10}$$

with

$$F(z, z_0, t) = \det(e^{tk\mathbf{M}})\,e^{-1/2({}^t\bar{z}k\mathbf{M}z - {}^t\bar{z}_0 k\mathbf{M}z_0)}$$

$$\begin{aligned}G(z, z_0, t) &= \int_{z(0)=z_0}^{z(t)=z} \mathscr{D}z\,\mathscr{D}\bar{z}\,\exp\left(-\frac{1}{2}\int_0^t ({}^t\dot{\bar{z}}\dot{z} + k^2\,{}^t\bar{z}\mathbf{M}^2 z)\,d\tau\right)\\ &= \langle z|\,e^{-tH_0}\,|z_0\rangle\end{aligned} \tag{11}$$

$$= \det\left(\frac{\mathbf{S}}{2\pi}\right)\exp\left(-\frac{1}{2}({}^t\bar{z}\mathbf{C}z + {}^t\bar{z}_0\mathbf{C}z_0 - {}^t\bar{z}\mathbf{S}z_0 - {}^t\bar{z}_0\mathbf{S}z)\right) \tag{12}$$

$$H_0 = -2\,{}^t\partial_{\bar{z}}\partial_z + \frac{1}{2}k^2\,{}^t\bar{z}\mathbf{M}^2 z \tag{13}$$

The matrices $\mathbf{S}$ and $\mathbf{C}$ appearing in (12) are defined as:

$$\mathbf{S} = k\mathbf{M}(\sinh(tk\mathbf{M}))^{-1}, \qquad \mathbf{C} = k\mathbf{M}\coth(tk\mathbf{M}) \tag{14}$$

Actually, $\mathscr{P}$, Eq. (10), can be easily deduced from the gaussian distribution of η (use (5); $\det(e^{tk\mathbf{M}})$ is simply the functional Jacobian for the change of variable $\eta \to z$[13]).

(12) is a generalization of the harmonic oscillator propagator.[14] It is obtained by using the normal coordinates. Furthermore, as can be easily checked, $\mathscr{P}$ is properly normalized: $\int dz\,d\bar{z}\,\mathscr{P}(z, z_0, t) = 1$.

Remark that an effective measure can be built for a distinguished monomer of the chain:[6] this can be done by integrating the Wiener measure (10) over all the paths of the other monomers. The result is a complicated expression that contains, in particular, a non local part (in time) exhibiting the non-Markovian character of the process for this monomer.

Nevertheless, we will show, in the sequel, that, despite this complication, we can compute some joint laws for several monomers (and *a fortiori* for one monomer).

So, let us turn to the computation of the area distribution $P(\{A_i\})$ for closed trajectories. Inserting the constraints

$$\prod_{j=1}^{n} \delta\left(A_j - \frac{1}{4i}\int_0^t (z_j\dot{\bar{z}}_j - \bar{z}_j\dot{z}_j)\, d\tau\right) \tag{15}$$

in the Wiener measure and using $\delta(x) = (1/2\pi)\int e^{iBx}\, dB$, we get the lagrangian for n particles subjected to uniform magnetic fields orthogonal to the motion plane (in addition to the harmonic interactions). Remark that, in principle, the magnetic fields are not the same for all the particles.

Introducing the $(n\times n)$ diagonal matrix $\mathbf{B}$ ($\mathbf{B}_{ij} = B_i\,\delta_{ij}$), we obtain

$$P(\{A_i\}) = \int\left(\prod_{j=1}^{n}\frac{dB_j}{2\pi}\, e^{iB_jA_j}\right)\left(\frac{Z_{\mathbf{B}}(t)}{Z_0(t)}\right) \tag{16}$$

$$Z_{\mathbf{B}}(t) = \mathrm{Tr}\, e^{-tH_{\mathbf{B}}}$$

$$H_{\mathbf{B}} = H_0 + V$$

$$V = \frac{1}{2}(-{}^t z\mathbf{B}\partial_z + {}^t\bar{z}\mathbf{B}\partial_{\bar{z}}) + \frac{1}{8}\,{}^t\bar{z}\mathbf{B}^2 z \tag{17}$$

In general, the matrices $\mathbf{B}$ and $\mathbf{M}$ do not commute and it is a difficult task to get the partition function $Z_{\mathbf{B}}(t)$. On the other hand, the distribution of the total area $A = \sum_{i=1}^{n} A_i$ is obtained by taking $B_j = B$ for all j. In this case, $\mathbf{B}$ and $\mathbf{M}$ commute. Using normal coordinates and known results about the partition function of the "2D harmonic oscillator + uniform magnetic field" problem,[15] we get the characteristic function of A ($\mathbf{I_n}$ is the $(n\times n)$ unit matrix):

$$\frac{Z_{\mathbf{B}}(t)}{Z_0(t)} = \prod_{j=1}^{n}\left(\frac{\cosh(tk\omega_j) - 1}{\cosh(t\sqrt{(k\omega_j)^2 + (B/2)^2}) - \cosh(t(B/2))}\right) \tag{18}$$

$$= \frac{\det(\cosh(tk\mathbf{M}) - \mathbf{I_n})}{\det(\cosh(t\sqrt{(k\mathbf{M})^2 + (\mathbf{B}/2)^2}) - \cosh(t(\mathbf{B}/2)))} \tag{19}$$

(the ω_j's are defined in (6)).

Each factor of (18) can be Fourier transformed in terms of modified Bessel functions and, finally, $P(A)$ is obtained by convolution. However, the result is not illuminating and we prefer to stick to the limit $t \to \infty$. A detailed study of (18) leads to:

$$\frac{Z_{\mathbf{B}}(t)}{Z_0(t)} \sim \exp\left(-\frac{tB^2}{8k}\sum_{i=1}^{n}\frac{1}{\omega_i}\right) = \exp\left(-\frac{tB^2 n(n+1)}{16k}\right) \tag{20}$$

Then, Fourier transformation shows that, in the large t limit, A is gaussian and scales like $\sqrt{t}$. Such a scaling is expected for all the areas A_i. This is what we will demonstrate by perturbation theory. When $t \to \infty$, we have

$$Z_{\mathbf{B}}(t) \sim e^{-tE_0(\mathbf{B})} \tag{21}$$

where $E_0(\mathbf{B})$ is the ground state energy. Moreover, due to the large oscillations of the factor $e^{iB_jA_j}$ in (16) when $A_j \to \infty$, only small values of B_j will contribute. So, it is enough to compute $E_0(\mathbf{B})$ at lowest order in $\mathbf{B}$. We will use the normal coordinates Z_i.

The eigenstates of H_0 are given by[16]

$$\Psi_{\{m_j\},\{n_j\}}(\{Z_j\}) = \prod_{j=1}^{n}\left(\sqrt{\frac{\omega_j n_j!}{\pi(n_j+|m_j|)!}}\, e^{im_j\theta_j}(\omega_j\,|Z_j|^2)^{|m_j|/2} \times L_{n_j}^{|m_j|}(\omega_j\,|Z_j|^2)\, e^{-1/2\,\omega_j\,|Z_j|^2}\right) \tag{22}$$

$$E_{\{m_j\},\{n_j\}} = \sum_{j=1}^{n}(2n_j+|m_j|+1)\,\omega_j \tag{23}$$

where $L_{n_j}^{|m_j|}$ is a Laguerre polynomial and the ground state is $\Psi_{\{0\},\{0\}}$. The perturbation V, (17), writes:

$$V = \tfrac{1}{2}(-{}^tZ\mathbf{R}^{-1}\mathbf{B}\mathbf{R}\partial_Z + {}^t\bar{Z}\mathbf{R}^{-1}\mathbf{B}\mathbf{R}\partial_{\bar{Z}}) + \tfrac{1}{8}\,{}^t\bar{Z}\mathbf{R}^{-1}\mathbf{B}^2\mathbf{R}Z \tag{24}$$

At first order in V, we get:

$$\Delta E_0^{(1)}(\mathbf{B}) = \int \Psi^*_{\{0\},\{0\}}\, V\Psi_{\{0\},\{0\}} = \frac{1}{8k}\operatorname{Tr}(\mathbf{B}^2\mathbf{M}^{-1}) = \frac{1}{8k}\sum_{m=1}^{n} mB_m^2 \tag{25}$$

Quadratic terms in $\mathbf{B}$ will also be produced at second order in V. The non-vanishing contributions will come out from the transitions from the ground-state to the states $\{m_j = \pm 1,\ m_l = \mp 1,\ m_i = 0 \text{ if } i \neq j, l\}$, $\{n_k = 0\}$. The computation gives:

$$\Delta E_0^{(2)}(\mathbf{B}) = -\frac{1}{16k} \sum_{m, m'=1}^{n} B_m B_{m'} \sum_{j \neq l} \mathbf{R}_{ml} \mathbf{R}_{mj} \mathbf{R}_{m'l} \mathbf{R}_{m'j} \left(\frac{(\omega_l/\omega_j) + (\omega_j/\omega_l)}{\omega_l + \omega_j} \right) \tag{26}$$

$$= -\frac{1}{8k} \sum_{m=1}^{n} m B_m^2 + \frac{1}{2k} \sum_{m, m'=1}^{n} B_m \mathbf{D}_{m, m'} B_{m'} \tag{27}$$

with:

$$\mathbf{D}_{m, m'} = \frac{1}{4} \int_0^\infty [(e^{-\tau \mathbf{M}})_{m, m'}]^2 \, d\tau + \frac{1}{8} \sum_{l=1}^{n} \frac{\mathbf{R}_{ml}^2 \mathbf{R}_{m'l}^2}{\omega_l} \tag{28}$$

So, to lowest order in **B**, we get:

$$\frac{Z_{\mathbf{B}}(t)}{Z_0(t)} \sim \exp\left(-\frac{t}{2k} \sum_{m, m'=1}^{n} B_m \mathbf{D}_{m, m'} B_{m'} \right) \tag{29}$$

As can be easily checked, (20) is recovered if we set $B_m = B$, $\forall m$.

With (16), we arrive at the probability distribution:

$$P(\{A_i\}) = \left(\frac{k}{2\pi t} \right)^{n/2} \frac{1}{\sqrt{\det \mathbf{D}}} \exp\left(-\frac{k}{2t} \sum_{m, m'=1}^{n} A_m (\mathbf{D}^{-1})_{m, m'} A_{m'} \right) \tag{30}$$

Thus, we observe that the areas A_i are correlated gaussian variables and that they scale like $t^{1/2}$ as expected. For the special case $n = 2$, we have:

$$P(A_1, A_2) = \sqrt{\frac{5}{3}} \frac{2k}{\pi t} \exp\left(-\frac{2k}{9t} (23 A_1^2 - 14 A_1 A_2 + 8 A_2^2) \right) \tag{31}$$

The width of A_2 is larger than the one of A_1: this is related to the fact that the second particle is, in average, farther from 0 than the first one. So, it sweeps larger areas.

Now, going to the winding angles $\{\theta_i\}$ around 0, we proceed as before and insert the constraint

$$\prod_{j=1}^{n} \delta \left(\theta_j - \frac{1}{2i} \int_0^t \left(\frac{z_j \dot{\bar{z}}_j - \bar{z}_j \dot{z}_j}{z_j \bar{z}_j} \right) d\tau \right) \tag{32}$$

in the Wiener measure. We are now faced to the problem of n harmonically bound particles submitted to the magnetic fields of point-like vortices located at the origin. The corresponding hamiltonian is:

$$H_\lambda = H_0 + W \tag{33}$$

$$W = \sum_{i=1}^{n} \lambda_i \left(\frac{1}{z_i} \partial_{\bar{z}_i} - \frac{1}{\bar{z}_i} \partial_{z_i} \right) + \sum_{i=1}^{n} \frac{\lambda_i^2}{2 z_i \bar{z}_i} \tag{34}$$

and the distribution $P(\{\theta_i\})$ is given by:

$$P(\{\theta_i\}) = \int \left(\prod_{j=1}^{n} \frac{d\lambda_j}{2\pi} e^{i\lambda_j \theta_j} \right) \left(\frac{Z_\lambda(t)}{Z_0(t)} \right) \tag{35}$$

Studying the limit $t \to \infty$, we cannot develop directly as before a perturbation theory with W: this is because of the last term in W that leads to a singular perturbation.[11] Due to this term, all the eigenfunctions of H_λ must vanish in 0 at least as $\prod_{i=1}^{n} (z_i \bar{z}_i)^{|\lambda_i|/2}$ ($\equiv U$). So we redefine those eigenfunctions:[11]

$$\Psi = U \tilde{\Psi} \tag{36}$$

The new hamiltonian acting on $\tilde{\Psi}$ is:

$$\tilde{H} = H_0 + \tilde{W} \tag{37}$$

$$\tilde{W} = \sum_{i=1}^{n} \left((\lambda_i - |\lambda_i|) \frac{1}{z_i} \partial_{\bar{z}_i} - (\lambda_i + |\lambda_i|) \frac{1}{\bar{z}_i} \partial_{z_i} \right) \tag{38}$$

That time, we can compute $\Delta E_0(\lambda)$ perturbatively and it will appear that only first order is necessary. Integrals of the form

$$\int e^{-1/2\, {}^t\bar{z} k \mathbf{M} z} \frac{1}{\bar{z}_i} \partial_{z_i} e^{-1/2\, {}^t\bar{z} k \mathbf{M} z} \, dz \, d\bar{z} \tag{39}$$

are involved. Integrating by parts and using $\partial_{z_i}(1/\bar{z}_i) = \pi\delta(z_i)$, we get, after some algebra:

$$\Delta E_0(\lambda) = k \sum_{j=1}^{n} \frac{|\lambda_j|}{(\mathbf{M}^{-1})_{jj}} = k \sum_{j=1}^{n} \frac{|\lambda_j|}{j} \equiv \sum_{j=1}^{n} \mu_j \, |\lambda_j| \tag{40}$$

So, for the winding angle distribution, we obtain:

$$P(\{\theta_i\}) = \int \left(\prod_{j=1}^{n} \frac{d\lambda_j}{2\pi} e^{i\lambda_j\theta_j} \right) e^{-t \sum_{j=1}^{n} \mu_j |\lambda_j|} \tag{41}$$

$$= \prod_{j=1}^{n} \left(\frac{1}{\pi\mu_j t} \frac{1}{1+(\theta_j/\mu_j t)^2} \right) \tag{42}$$

At large times, the winding angles are uncorrelated, they scale like t and are distributed according to Cauchy laws. The variance $\langle \theta_j^2 \rangle$ is infinite: this is, of course, due to the "small windings" occuring in the vicinity of the origin as will be seen explicitly at the end of this paper.

Moreover, we observe that θ_j scales like μ_j, i.e., like $1/j$. This is reasonable because, when j increases, the considered particle is, in average, farther from 0 and, consequently, its winding angle must decrease. What is somewhat unexpected is such a simple dependence of θ_j on j.

We also addressed the problem of winding angles around n different points of complex coordinates b_l, $l = 1,..., n$.

θ'_j being the angle wound by the particle j around the point b_j, we obtained for the set of variables $\{\theta'_j\}$ the same joint law as (42) except for the change of μ_j into μ'_j:

$$\mu'_j = \mu_j e^{-\mu_j |b_j|^2} \tag{43}$$

Owing to the rotationnal symmetry breaking when $b_j \neq 0$, the winding angles θ'_j are statistically reduced by the factor $e^{-\mu_j |b_j|^2}$. Nevertheless, even for large $|b_j|$'s, the variance $\langle (\theta'_j)^2 \rangle$ is infinite.

Setting all the b_j's to zero, we recover (42). This is what we will consider now and assume that we count the winding angles θ_j only when $|z_j| > r_0$ (i.e., the so-called "big windings"[9]). Still when $t \to \infty$, the perturbation W, Eq. (34), can now be used because $\lambda_j = 0$ when $|z_j| < r_0$. At first order in W, the linear contributions in λ_j will cancel. In the limit of a small, but finite r_0, we get, for the remaining contribution:

$$\Delta E_0^{(1)}(\lambda) \sim k \, |\ln r_0| \sum_{j=1}^{n} \frac{\lambda_j^2}{j} \tag{44}$$

The quadratic contributions in the λ_j's coming out from the second order in W will be finite (thus subleading) when $r_0 \to 0^+$. Finally, we get for the big winding angles asymptotic distribution:

$$P(\{\theta_j\}) = \prod_{j=1}^{n} \sqrt{\frac{j}{4\pi t k \, |\ln r_0|}} \exp\left(-\frac{j}{4tk \, |\ln r_0|} \theta_j^2 \right) \tag{45}$$

In this limit, the variables θ_j are uncorrelated (the correlations get smaller and smaller when r_0 decreases). They are now gaussian and scale like $\sqrt{t\,|\ln r_0|/j}$. Their variance grows to infinity when r_0 goes to 0, showing the increasing contribution of the small windings around 0.

To summarize, we have computed explicitly the asymptotic joint laws for the areas (that scale like $\sqrt{t}$) and for the winding angles (that scale like t when no critical region is excluded). The scaling variables we have got compare well with those involved in the Brownian motion on finite domains: this is not so surprising since the chain is bound to a fixed point.

Moreover, we have shown that physical interactions between particles (harmonic interactions here) can lead to statistical correlations (case of the areas) or not (case of the winding angles): it depends on the quantity we consider.

In a forthcoming paper,[17] we will study the statistical properties of the free Rouse chain. We will especially show that the areas and winding angles distributions are very different from those presented in this work. This is essentially due to the translation invariance that holds when the chain is free.

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