General mapping between random walks and thermal vibrations in elastic networks: Fractal networks as a case study

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We present an approach to mapping between random walks and vibrational dynamics on general networks. Random walk occupation probabilities, first passage time distributions and passage probabilities between nodes are expressed in terms of thermal vibrational correlation functions. Recurrence is demonstrated equivalent to the Landau-Peierls instability. Fractal networks are analyzed as a case study. In particular, we show that the spectral dimension governs whether or not the first passage time distribution is well represented by its mean. We discuss relevance to universal features arising in protein vibrational dynamics.

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I. INTRODUCTION

Mapping two different physical problems onto one another has been proven very useful in physics. Examples are the mapping between the Schrodinger and diffusion equations [1], the mapping between lattice-gas and Ising models [2], the mapping between quantum field theories and critical phenomena [2], and the mapping between random walks and electric networks [3]. Here we focus on another well-known mapping, the mapping between random walks and vibrations in the scalar elasticity model. This mapping has been recognized for a long time, and some of its consequences have been already exploited, mainly in the context of fractal and percolation networks [4]. Nevertheless, despite the large amount of work done in the field, vibrational analogs for several basic quantities arising in the theory of random walks remained unknown.

Vibrations on a network of masses and springs are one of the most well studied problems in physics. Scalar elasticity is a special case where the three components of the displacement vector of a network node are decoupled from each other. Recently we have shown that the scaling behavior of the mean first passage time (MFPT) on fractal networks can be derived by exploiting an analogy with elastic networks [5]. The MFPT is an extremely important quantity governing the rate of diffusion controlled chemical reactions [6,7]. Chennubhotla *et al.* expressed the MFPT in terms of thermal vibrational correlation functions [8]. However, the applied line of argumentation could not be carried on to higher moments of the first passage time, nor to its full distribution or other probabilistic quantities. In the present study, we consider networks of general topology and focus on the construction of a rigorous mapping between the problem of a scalar elastic network coupled to a thermal bath and the random walk problem. The roots of our approach can be traced back to the pioneering work of Hattori *et al.* who established a relation between random walks and thermal correlation functions of spin systems [9].

II. MAPPING RANDOM WALKS TO THERMAL VIBRATIONS

Consider an elastic network (EN) of N masses coupled by harmonic springs in the framework of the scalar elasticity model also known as the Gaussian network model (GNM) when applied to proteins [4,10,11]. The GNM is defined by the quadratic Hamiltonian equation,

$$H_{GNM} = \sum_i \frac{m(\dot{\vec{u}}_i)^2}{2} + \frac{\gamma}{2}\sum_{i,j} \Delta_{ij}(\vec{u}_i - \vec{u}_j)^2. \tag{1}$$

The first term represents the kinetic energy of the system, γ is the spring force constant which is assumed to be homogeneous, $\vec{u}_i \equiv (x_i, y_i, z_i)$ and $\vec{R}_i = \vec{R}_i^0 + \vec{u}_i$ are the displacement with respect to the equilibrium position $\vec{R}_i^0$ and the instantaneous position of the ith mass, respectively. Δ is the network connectivity matrix with the following entries: $\Delta_{ij}=1$ if $i \neq j$ and the pair i,j is connected by a spring, $\Delta_{ij}=0$ otherwise. Here we will assume that a path of masses and springs exists between any two masses on the network. The GNM is threefold degenerate so it is sufficient to consider one spatial direction. Denoting by $\vec{x}$ the vector whose entries are x_i, the equations of motion in the absence of friction are: $m\frac{d^2\vec{x}}{dt^2} = -\gamma\Gamma\vec{x}$. Here Γ is the network Kirchhoff matrix,

$$\Gamma_{ij} = \begin{cases} -\Delta_{ij} & if \ i \neq j \\ z_i & if \ i = j \end{cases}, \tag{2}$$

where $z_i \equiv \Sigma_k \Delta_{ik}$ is the coordination number of the ith mass. Since rigid translations are a solution of the equations of motion, one of the eigenvalues of the matrix Γ is zero and consequently this matrix has no inverse. As an alternative, we exploit the existence of a generalized inverse. Denoting the Moore-Penrose pseudoinverse of Γ by Γ^{-1} and coupling the elastic network to a thermal bath one can show that [10,12]

$$\Gamma_{ij}^{-1} = \frac{\gamma}{k_B T}\langle x_i x_j \rangle_T. \tag{3}$$

where $\langle x_i x_j \rangle_T$ is the thermal correlation function.

The EN we have described above can also be thought of as a network of nodes connected by links (which we will refer to as RWN), see Fig. 1. We now construct a continuous

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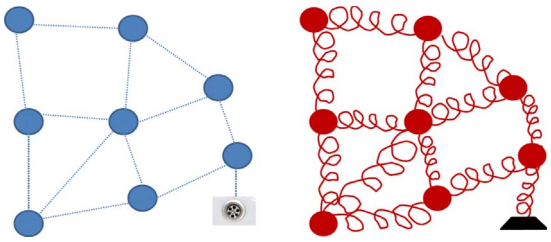

FIG. 1. (Color online) Left–a network of nodes connected by links (RWN), right—the corresponding elastic network of masses coupled by harmonic springs (EN). One can transform a RWN to an EN (and vice versa) by transforming nodes to masses and links to springs. Absorbing boundary conditions are tackled by introducing absorbing nodes and equivalent infinite mass walls. We study the relation between the random walk problem on the RWN and thermal vibrations of the EN.

time random walk on the RWN and show that under our construction the problem of vibrations on a network of masses and springs is practically equivalent to the random walk problem. In particular we express the *distribution of the first passage time* between two nodes (in Laplace plane) in terms of the vibrational correlation matrix defined in Eq. (3). Consider a random walk where the rate Γ_{jj} at which a random walker jumps out of node j is the coordination number of that node, $\Gamma_{jj}=z_j$, and that the random walker performs random jumps to its nearest neighbors without preference. In this scenario, $-\Gamma$ plays the role of a rate matrix whose elements $-\Gamma_{ij}$ are the transition rates from node j to node i. It follows that the change in occupation probabilities as a function of time follows the master equation: $\frac{d\vec{p}(t)}{dt}=-\Gamma\vec{p}(t)$, with the formal solution $\vec{p}(t)=\Sigma_{n=0}^{\infty}\frac{(-t\Gamma)^n}{n!}\vec{p}(0)\equiv\exp[-t\Gamma]\vec{p}(0)$.

When reflecting boundary conditions are introduced, probability is conserved. Indeed, from Eq. (2) it follows that: $\Sigma_{i\neq j}\Gamma_{ij}=-\Gamma_{jj}$, i.e., the rate of probability flow out of a site j is equal to the total rate at which probability flows into adjacent sites. The case of absorbing boundary conditions is tackled by introducing artificial absorbing nodes which are connected to some of the other network nodes. The EN analog is the introduction of equivalent infinite mass nodes which are connected by springs to the same masses as in the RWN. Mathematically this means that the network Kirchhoff matrix Γ would change such that $\Gamma_{ii}\rightarrow\Gamma_{ii}+n_i$, where n_i is the number of absorbing/infinite mass nodes connected to node i and we note that this would not change the form of the master equation. In addition, rigid translations are no longer a solution of the equations of motion. This implies that Γ has a true inverse Γ^{-1} for which Eq. (3) holds [12].

Denote by $P_{ij}(t)$ the probability that a random walker is found at site j at time t given that it was at site i at time $t=0$ and let $P(t)$ be the matrix whose entries are $P_{ij}(t)$. Denote by τ_{ij} the first passage time (FPT) (in the case of $i=j$ the first return time) of a random walker traveling from site i to site j and let $f_{ij}(t)$ be the probability density function of τ_{ij}. In the supplementary material accompanying this Letter we show that [12],

$$\begin{cases} \tilde{P}(s)=\dfrac{\hat{1}}{sN}\delta_{bc}+\dfrac{\Gamma^{-1}}{I+s\Gamma^{-1}} & \\ \tilde{f}_{ij}(s)=\dfrac{\dfrac{\delta_{bc}}{N}+\left(\dfrac{s\Gamma^{-1}}{I+s\Gamma^{-1}}\right)_{ij}}{\dfrac{\delta_{bc}}{N}+\left(\dfrac{s\Gamma^{-1}}{I+s\Gamma^{-1}}\right)_{jj}} & i\neq j \\ \tilde{f}_{ii}(s)=1-\dfrac{s}{(s+z_i)\left[\dfrac{\delta_{bc}}{N}+\left(\dfrac{s\Gamma^{-1}}{I+s\Gamma^{-1}}\right)_{ii}\right]} & i=j. \end{cases} \quad (4)$$

Here, $\tilde{P}(s)$, $\tilde{f}_{ij}(s)$, $\tilde{f}_{ii}(s)$ are the Laplace transforms of $P(t)$, $f_{ij}(t)$, $f_{ii}(t)$, respectively, $\hat{1}$ is a matrix whose *all* entries equal one and δ_{bc} equals either one or zero in the case of reflecting/absorbing boundary conditions, respectively. Equation (4) is the fundamental result that stands in the basis of this paper. It provides the connection between the statistical mechanics of an elastic network and the stochastic dynamics of a random walker on the same network. $P(t)$ and $f_{ij}(t)$ are uniquely determined by their Laplace transform and it follows that all the information about the stochastic dynamics of a random walker is contained in the pairwise correlation matrix Γ^{-1}. We proceed with the derivation of novel relations implied by Eq. (4).

III. ABSORBING BOUNDARY CONDITIONS

On a finite network with reflecting boundary conditions the probability of a random walker to eventually reach a target site j given it started at i is one. In general this probability is given by $F_{ij}=\int_0^\infty f_{ij}(t)dt=\lim_{s\rightarrow 0}\tilde{f}_{ij}(s)$ and our conclusion is revalidated by taking this limit in Eq. (4) ($\delta_{bc}=1$). The case of absorbing boundary conditions is more interesting since the random walker may be absorbed at the boundary before reaching its destiny. Taking the limit $s\rightarrow 0$ ($\delta_{bc}=0$) in Eq. (4) we obtain a vibrational interpretation for F_{ij} on a finite network equipped with absorbing boundary conditions,

$$\begin{cases} F_{ij}=\dfrac{\Gamma_{ij}^{-1}}{\Gamma_{jj}^{-1}}=\dfrac{\langle x_i x_j\rangle_T}{\langle x_j^2\rangle_T} & i\neq j \\ F_{ii}=1-\dfrac{1}{z_i\Gamma_{ii}^{-1}}=1-\dfrac{1}{\dfrac{\gamma z_i}{k_BT}\langle x_i^2\rangle_T} & i=j. \end{cases} \quad (5)$$

Doing the same for $\tilde{P}(s)$ yields a vibrational interpretation for the mean time spent in site j (given the walk started at site i) prior to absorption: $\int_0^\infty P_{ij}(t)dt=\frac{\gamma}{k_BT}\langle x_i x_j\rangle_T$. Interestingly, F_{ij} depends only upon the vibrational correlation between source and target and the vibrational mean square displacement (MSD) of the target. Similarly, the probability to eventually return to the origin F_{ii}, depends only upon the vibrational MSD of the origin and its coordination number z_i. We note that by definition $1\geq F_{ij}$, $F_{ii}\geq 0$ and hence in the case of absorbing boundary conditions: $\langle x_i^2\rangle_T\geq\frac{k_BT}{\gamma z_i}$ and $\langle x_j^2\rangle_T\geq\langle x_i x_j\rangle_T\geq 0$. In the scalar elasticity model the vector $\vec{x}$

is multivariate normal [10] and its equilibrium distribution is uniquely determined by the correlation matrix $\langle x_i x_j \rangle_T$. Since $\int_0^\infty P_{ij}(t)dt$ determines $\langle x_i x_j \rangle_T$, the stochastic dynamics of a random walker on the RWN uniquely determines the equilibrium distribution of the displacements vector on the EN.

Equation (5) allows us to examine the question of recurrence in infinite networks by taking the thermodynamic limit. In achieving this limit, we construct an infinite network as a limit of on growing finite subnetworks. Doing so, we keep all absorbing nodes at the periphery as subnetworks grow in size. We discriminate between two types of infinite elastic networks, those that are thermodynamically stable and those that are not. From Eq. (5) it is clear that $\lim_{N\to\infty} F_{ii}=1 \Leftrightarrow \lim_{N\to\infty}\langle x_i^2\rangle_T=\infty$ and that $\lim_{N\to\infty} F_{ii}<1 \Leftrightarrow \lim_{N\to\infty}\langle x_i^2\rangle_T<\infty$. A network is called recurrent if $F_{ii}=1$ for every site i [13,14]. By assumption there is path connecting any two nodes and one can show that in this case F_{ii} are either all equal to one or all smaller than one [14]. Correspondingly $\langle x_i^2\rangle_T$ are either all infinite or all finite and we conclude that an infinite network is recurrent if it is thermodynamically unstable and vice versa. Recurrence, in the context of random walks, is hence a term equivalent to the Landau-Peierls instability of elastic networks [15–17]. Note that no assumptions regarding network structure were made in arriving to this conclusion.

Since $\langle x_i^2\rangle_T$ either all diverge or all converge in the thermodynamic limit, considering the average vibrational MSD: $\langle x^2\rangle_T=\frac{1}{N}\Sigma_{n=1}^N\langle x_i^2\rangle_T$ is usually enough in order to determine if an infinite network is recurrent. On fractal elastic networks the evaluation of $\langle x^2\rangle_T$ is relatively simple since after going to normal modes and invoking the equipartition theorem one is left with a simple integral that gives [16]

$$\langle x^2\rangle_T \sim \begin{cases} \text{constant} & d_s > 2 \\ ln(N) & d_s = 2 \\ N^{(2/d_s)-1} & d_s < 2. \end{cases} \tag{6}$$

Here d_s is the network spectral dimension that governs the density of vibrational modes $g(\omega)$ at low frequencies via the scaling relation $g(\omega)\sim\omega^{d_s-1}$. For regular three-dimensional (3D), two-dimensional (2D), and one-dimensional (1D) networks the spectral dimension coincides with the regular dimension and one recovers the well known Debye density of states. Combing Eqs. (5) and (6) we revalidate that an infinite fractal network is recurrent if $d_s \leq 2$ (and vice versa) a result which was first obtained by Burioni *et al.* [16]. In addition we recover the average manner in which the limit $F_{ii}\to 1$ for $N\to\infty$ is achieved. Note that one must be more careful applying the above arguments when considering pathological cases for which, after taking the thermodynamic limit, $\langle x^2\rangle_T$ is infinite despite the fact that $\langle x_i^2\rangle_T$ are all finite and we refer the reader to a discussion on the related issue of "recurrence on the average" [14].

IV. REFLECTING BOUNDARY CONDITIONS

On a finite network with absorbing boundary conditions for every two sites characterized by $F_{ij}<1$, the probability that the random walker would never reach j ($\tau_{ij}=\infty$) is positive and the MFPT is therefore infinite. In contrast, in the case of reflecting boundary conditions, the random walker is sure to reach its destiny and the MFPT is then the single most important quantity describing τ_{ij}. The kth moment of τ_{ij} is given by: $E[\tau_{ij}^k]=(-1)^k\tilde{f}_{ij}(s)^{(k)}|_{s=0}$. Taking the first derivative and the limit $s\to 0$ ($\delta_{bc}=1$) in Eq. (4) we obtain

$$E[\tau_{ij}] = \begin{cases} \frac{N\gamma}{k_B T}[\langle x_j^2\rangle_T - \langle x_i x_j\rangle_T] & i \neq j \\ \frac{N}{z_i} & i = j. \end{cases} \tag{7}$$

Interestingly, the MFPT from site i to site $j\neq i$ depends only upon the vibrational correlation between source and target and the vibrational MSD of the target. The mean first return time is only affected by the number of nodes N and the coordination number z_i. Note that $E[\tau_{ij}]>0$ by definition and hence: $\langle x_j^2\rangle_T > \langle x_i x_j\rangle_T$, in contrast to the case of absorbing boundary conditions $\langle x_i x_j\rangle$ can be negative.

Equation (7) can be used in order to obtain a simple vibrational derivation of the scaling law for the global MFPT (GMFPT) on a finite fractal domain [18]. The global first passage time (GFPT) is defined as the random time it takes a random walker to reach a randomly selected target from a randomly selected origin. Here we assume that the source and target are different sites. The GMFPT is defined as the average GFPT: $\mathrm{GMFPT}(N)=\frac{1}{N(N-1)}\Sigma_{i,j,i\neq j}E[\tau_{ij}]$. Using Eq. (7) for $E[\tau_{ij}]$ and assuming that cross correlations add up (incoherently) to a negligible contribution, we obtain: $\mathrm{GMFPT}(N)\simeq\frac{N\gamma}{k_b T}\langle x^2\rangle_T$. The scaling of the GMFPT with the number of nodes follows from Eq. (6),

$$\mathrm{GMFPT}(N) \sim \begin{cases} N & d_s > 2 \\ N\, ln(N) & d_s = 2 \\ N^{2/d_s} & d_s < 2. \end{cases} \tag{8}$$

The MFPT does not provide a complete characterization of the FPT distribution. Another important, and much less studied quantity, is the variance. The variance gives a measure for the width of the FPT distribution around the mean. If the width is very large in comparison to the MFPT, the latter cannot be considered a reliable representative of the distribution. For $i=j$ we use Eq. (4) ($\delta_{bc}=1$) to obtain [12]: $E[\tau_{ii}^2]=\tilde{f}_{ij}(s)^{(2)}|_{s=0}=2(\frac{N}{z_i})^2[\frac{1}{N}+\frac{z_i\gamma}{k_b T}\langle x_i^2\rangle_T]$ and for the reduced variance [12]: $\sigma^2[\tau_{ii}]/E^2[\tau_{ii}]=\frac{2}{N}+\frac{2z_i\gamma}{k_B T}\langle x_i^2\rangle-1$. In fractal networks with a coordination number that is sharply distributed around a mean value z, we can replace z_i with z, average over all sites and use Eq. (6) to obtain

$$\frac{1}{N}\sum_{i=1}^{N}\frac{\sigma^2[\tau_{ii}]}{E^2[\tau_{ii}]} \sim \begin{cases} \text{const} & d_s > 2 \\ ln(N) & d_s = 2 \\ N^{2/d_s-1} & d_s < 2. \end{cases} \tag{9}$$

We conclude that in the case of $d_s>2$ the standard deviation is of the same order of the MFPT regardless of the number of nodes N. Conversely, when $d_s<2$ the reduced variance diverges as a power law of N and the MFPT cannot be considered a reliable representative of the FPT distribution. When

$i \neq j$ we average over all pairs distanced r apart and obtain the following approximation [12]:

$$\frac{1}{N_r}\sum_{i,j}\frac{\sigma^2[\tau_{ij}]}{E^2[\tau_{ij}]} \sim \begin{cases} [1-(r/a)^{d_f(2/d_s-1)}]^{-1} & d_s > 2 \\ ln(N)/ln(r/a) & d_s = 2 \\ N^{2/d_s-1}(r/a)^{-d_f(2/d_s-1)} & d_s < 2. \end{cases} \quad (10)$$

Here N_r is the number of pairs distanced r apart and the sum goes only over these pairs, a is the average distance between nearest neighbors and d_f is the network fractal dimension. We note that the joint domain in which the approximation is valid is given by: $\{N \gg 1, a \ll r \ll R_g\}$ where R_g is the radius of gyration. Equation (10) demonstrates that the reduced variance is a monotonically decreasing function of the distance between source and target. Keeping this distance fixed, the reduced variance is independent of the number of nodes in the case of $d_s > 2$ but when $d_s \leq 2$ it diverges with the number of nodes. Equation (10) stands in line with a similar result obtained independently by Bénichou *et al.* [7].

V. SUMMARY AND DISCUSSION

In this paper, we have rigorously mapped observables that appear in random walk theory to observables associated with the problem of an elastic network coupled to a thermal bath. Our mapping provides a systematic way for translating random walk problems to the realm of elasticity physics allowing for new theoretical, computational, and experimental approaches toward the random walk/elastic network problems. The mapping was demonstrated useful in the analysis of random walk problems on complex, scale invariant, media. It is important to emphasize that the MFPT heavily depends on network topology. The entire network topology is summarized in the network Kirchhoff matrix Γ and hence [using Eq. (7)] knowing the network topology amounts to knowing the MFPT. More surprising is the converse statement. Network topology can be reconstructed knowing the MFPT from any node to any other node and the second moment of the first return time [12].

Recently, we have utilized random walks on protein structures to study the vibrational dynamics of proteins [19]. In particular, we used a special case of Eq. (7) [8] relating the MFPT with the thermal variance in the instantaneous distance between amino acids. This equation has been used to unravel universal properties in the vibrational dynamics of proteins. In addition, we have shown that a sharp deviation from the mean universal behavior may result in the emergence of specific functionality. We suggest that exploiting the different relations between vibrations and random walks, as stated in this paper, can be beneficial in the research of functionality-dynamics interplay in proteins.

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