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Parametric ergodicity to measure roughness from a single sample

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ABSTRACT

We introduce a concrete random model system to study the concept of parametric ergodicity. It consists of a continuum mechanical cavity with an embedded random mass distribution, constrained by a parametrized boundary condition. The interest is twofold. On one hand, there is the practical interest of obtaining ensemble averages of physical quantities from a small number of experimentally available samples, in many cases only one. This is typically the case in studies on conductance fluctuations through disorder mesosocopic systems. On the other hand we want to develop more insight into the meaning of parametric ergodicity. For this, we focus on the statistical distribution of resonant frequency generated by the ensemble of random samples, and how to produce the same distribution from a single sample subject to changing a boundary condition — the external parameter. The paper shows how the changing of the boundary condition is equivalent to scanning the ensemble of equivalent samples.

1. Introduction

In this paper we develop a concrete model to study how to extract ensemble averages from a single sample by varying an external parameter. This problem is of paramount importance in experimental studies on disordered systems. There, it is typically the case that only a handful of samples (sometimes even only one) are available but one wishes to study statistical properties of similarly prepared ones. Even when many samples were available, it would be impractical to perform measurements on more than a few of them. In fact, this work was motivated by the need to obtain the distribution of resonant frequencies of nominally cylindrical nanosensors that had, by intrinsic limitations of the growth process, uncontrolled rough surfaces [1]. Indeed, the demand for nanosensors and nanoactuators capable of consistent sub-attonewton force and sub-attogram mass measurements, has prompted interest in nanoelectromechanical (NEMS) oscillators [2–4]. In addition, they are good candidates for the detection of single spin [5], and for the detection of motion approaching the quantum regime [6,7]. These nano-oscillators have already been used to detect attogram particles [4], of major interest for chemical and biological sensing. To improve the sensitivity and reliability of NEMS oscillator devices, knowledge of the material mechanical properties and mechanical response is essential. One of our goals is the understanding of the mechanical response of amorphous silica nanorods grown by electron irradiation. The experimental procedure allows one to grow one nanorod at a time and to monitor the process with the same transmission electron microscope (TEM) used for the irradiation. The control on the nanostructure with this technique is ideal for the study of the mechanical properties of silica at the nanoscale, and for the development of ultrasensitive nanoresonators. Close analysis of the silica nanorods shows surface roughness on the nominally cylindrical structures. In the field, these nanorods can be used as atomic force microscopy cantilevers and the contact force between the sensor and the sample under study can be used as an external parameter, a central topic of this paper.

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As a corollary of this study, and of no less importance, is the revision of the concept of parametric ergodicity. By means of a specific model we develop a better understanding of its meaning.

Generally speaking, when experimentally studying statistical properties of stationary systems it is useful to use, if possible, the property of ergodicity. In its common usage, ergodicity makes it possible to obtain time averages by considering instead ensemble averages [8]. This is helpful because under normal circumstances, the initial conditions of a system of many degrees of freedom are unknown and it would be impossible to take time averages. The ensemble average may describe our ignorance appropriately [9].

Another type of ergodicity that has been considered is parametric ergodicity [10]. There, an external parameter is varied quasistatically while a property of the system is monitored. A case in point is the study of frequency statistical fluctuations of wave modes in disordered systems [11]. While an external parameter (such as a magnetic field) is varied, the frequency spectrum is measured and it is assumed that the distribution of frequencies so obtained are those corresponding to an ensemble of similarly disordered systems. Tsyplyatyev et al. [12] proposed a rationale for the validity of parametric ergodicity when transport is measured through disordered samples—typically mesoscopic ones. It is known that ensemble fluctuations of conductance through mesoscopic samples are related to random dephasing at the substructural level. On the other hand, swapping of an external magnetic field on a single sample induces random dephasing at different locations, thus effectively generating the ensemble. Similar concepts have been shown [13] to be at play in universal conductance fluctuations, where the sample ensemble has been obtained by varying, on a single sample, not only the external magnetic field, but also the electron density. In this case they argue for parametric ergodicity on the basis of the idea that the external parameter induces sufficient transitions between microstates.

Parametric ergodicity is at the cornerstone of recent experiments. For example, experiments measuring the position-dependent elastic modulus of copper nanorods deposited on organosilicate glass [14]. For this purpose a resonator (an atomic force cantilever) is brought into contact with the sample. By varying the contact force (external parameter) they were able to perform a mapping of the elastic modulus by tracking a resonant frequency shift. Another example is the experimental measurement of conductance fluctuations in mesoscopic superconducting–normal-superconducting samples [15]. The fluctuations are the result of current channeling through subcomponent quantum dots: the total transmission is the result of the average transmission through the collection of quantum dots. They find empirically that this average over many samples can also be obtained by measuring on a single sample while varying the gate voltage (external parameter). In a third example [16], experiments of conductance through gold nanowires were done to clarify the microscopic mechanisms. For statistical analysis, instead of doing the experiments over a multitude of samples, they measure on only a few samples but vary an external magnetic field (external parameter) to produce random distributed dephasing.

The definition of parametric ergodicity given above deserves further consideration. Here, we study the problem through a concrete example of waves in disordered media that constructively develops the ensemble probability density function from parametric measurements on a single sample.

The paper is organized as follows. In Section 2, we introduce the model system. In Section 3, we introduce notation and the solution for the deterministic case, for a system with no disorder. In Section 4, we present the resonant frequency changes induced by disorder. This is the core section of the paper and also shows how, for a given sample, the frequency changes as a function of the boundary condition and why the frequencies so obtained correspond to those of an ensample of unconstrained samples. Section 5 is an example of application of the concepts to a small ensemble. This section has the purpose of clarifying the notation introduced in Section 4, and showing explicitly how the procedure of generating the ensemble by varying an external parameter works. Section 6 contains our conclusions.

2. The system

Consider an ensemble of random samples generated by an elastic string of length a, tension T, and random linear mass density. Let us suppose that we are interested in the statistical fluctuations of, for example, the lowest frequency in the string, generated by measuring the same quantity on many samples. As explained in the introduction, such an ensemble is not experimentally accessible. Moreover, let us further suppose that we only have a single string on which to make the measurements. The central goal of this paper is to obtain the statistical fluctuations of the frequency by measurements on a single sample. We proceed as follows. We introduce an external parameter that allows one of the fixed ends to become loose. More specifically, in Fig. 1 we represent this by a spring (of constant β) attached to the right end of the string. The $\beta \to \infty$ limit, corresponds to the fixed condition. The $\beta \to 0$ limit, corresponds to no vertical force on the right end of the string. We assume that there is always a horizontal force present to provide the required tension, as can be achieved by a small ring attached at the right end of the string and passing through a vertical fixed pole. Those two limits are conceptually relevant: while oscillating at its lowest natural frequency, in the former case large amplitudes occur in the center of the string, while in the latter, large amplitudes occur both at the center and more so at the right end. Thus, as we tune β from ∞ to 0, less kinetically active regions become more active. Therefore, the effect of the local mass on the global properties of the system (e.g. the resonant frequency) can be enhanced or reduced by varying β. From the point of view of ergodicity, we will show how to extract statistical properties of the ensemble from a single sample. This presupposes that the mass density is equally distributed along a single string or many—a standard situation which is the result of the specific sample manufacturing process, and that it is indeed the case in many relevant cases. For example, silica-based nanorods for sensor

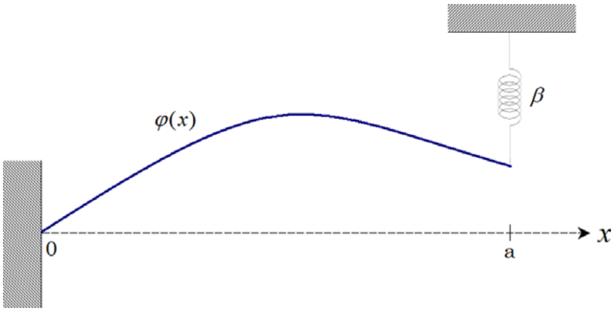

Fig. 1. A string with non-uniform mass along its length has an amplitude $\varphi(x)$ in the lowest frequency mode corresponding to the boundary conditions shown.

applications grown in Transmission Electron Microscope chambers present inherent surface roughness that is independent of the location along the rod [1]. Thus the ideas developed here can, mutatis mutandis, be applied to such a system.

3. Dynamics with no randomness

Let $\varphi(x)\cos\omega t$ be a normal mode of oscillation of the string with uniform density — the random density fluctuations will be introduced in the next section. To satisfy the wave equation and the fixed boundary condition at $x = 0$, we write

$$\varphi(x) = A\sin kx, \tag{1}$$

where k is the wavenumber, and ω the angular frequency.

At the right end, $x = a$, the total shear force on the cross section vanishes then,

$$0 = -T\left[\frac{\partial\varphi(x)}{\partial x}\right]_{x=a} - \beta\varphi(a). \tag{2}$$

Eq. (2) gives the correct limit when $\beta \to 0$, in this case corresponding to no spring, the small ring senses a zero vertical force when $\left[\frac{\partial\varphi(x)}{\partial x}\right]$. To check the correctness of (2) in the opposite limit, $\beta \to \infty$, we can rewrite it as $0 = -\frac{T}{\beta}\left[\frac{\partial\varphi(x)}{\partial x}\right]_{x=a} - \varphi(a)$, and we see that because the first term on the right hand side tends to zero then the boundary condition becomes $\varphi(a) = 0$, which is the correct result for an infinitely stiff spring.

Upon substitution of Eq. (1) in (2), we obtain an equation for k,

$$0 = Tk\cos ka + \beta\sin ka. \tag{3}$$

We introduce the dimensionless wavenumber $\xi = ka$, and the dimensionless spring constant $\gamma = \frac{\beta a}{T}$, and obtain,

$$\frac{1}{\gamma}\xi + \tan\xi = 0 \tag{4}$$

which gives an implicit dependence of the wavenumber on the spring constant, $\xi(\gamma)$. Fig. 2 shows a plot of the function obtained numerically. Also, as a guide, we found within less than 2% a good analytical approximation, as shown in Fig. 2, and given by

$$\xi = \frac{\pi}{2} + \arctan(\lambda_0\gamma) \tag{5}$$

with $\lambda_0 = 0.3788$.

For a linear medium such as the one we are considering here, Eq. (5) immediately renders the lowest frequencies $\omega(\gamma)$, recalling that $\omega = \sqrt{\frac{T}{\rho_0}}k$, where ρ_0 is the density of this uniform string.

4. Dynamics with randomness

To introduce the effect of random masses along the string, we will borrow ideas commonly used in electromagnetic cavities. Given a mode in an electromagnetic cavity, one is frequently interested in mapping the actual distribution of electric and magnetic fields. Alternatively, electromagnetic cavities are often used to measure the permittivity and conductivity of an unknown material placed inside. Although seemingly unconnected, the two applications can be understood from the same theory. Bethe and Schwinger [17] (BS) showed that when a small sample is introduced in the cavity, the mean frequency (and quality factor) of the resonant curve changes. This frequency change depends on the dielectric properties of the sample and

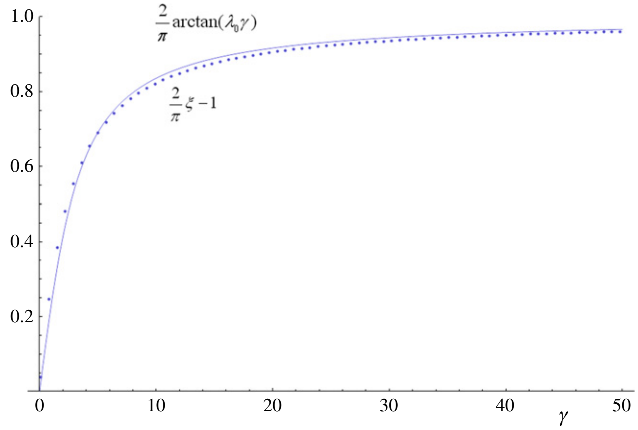

Fig. 2. Numerical solution to Eq. (4) for various values of γ (dotted line). Also shown, an analytical approximation (continuous line).

is proportional to the intensity of the field at the location of the sample. Thus frequency shifts while the sample is scanned encode information of the amplitude of the fields inside the cavity.

In fact, BS results are not restricted to the electromagnetic case, but apply to any wave phenomena. To clarify this point, it has been explicitly demonstrated [18] how to use the results on one-dimensional mechanical cavities, similar to those considered in this paper. For a small massive single inclusion in an otherwise homogeneous string, the frequency shift is given by

$$\frac{\omega - \omega_0}{\omega_0} = -\mu\varphi^2(x_n) \tag{6}$$

where ω_0 is the frequency of the homogeneous string, ω the frequency of the system including the inclusion, and μ the excess mass of the inclusion at position x_n. The negative sign means that a positive inclusion reduces the frequency, as expected on simple inertia grounds.

Eq. (6) can be extended to the case of multiple inclusions, thus

$$\Delta(\gamma) = \sum_{n=1}^{N} \left\{-\mu_n\varphi^2(x_n)\right\} \tag{7}$$

where μ_n is the excess mass at location x_n. N is the number of divisions of the string and is related to the excess mass correlation length ℓ, by $N\ell = a$. We also have introduced the definition of the fractional frequency change $\Delta(\gamma) = \frac{\omega(\gamma)-\omega_0(\gamma)}{\omega_0(\gamma)}$, where we emphasize the dependence with the external parameter, the spring stiffness γ.

Eq. (7) can be rewritten as

$$\Delta(\gamma) = -\frac{1}{2}\sum_{n=1}^{N}\mu_n + \frac{1}{2}\sum_{n=1}^{N}\mu_n \cos\left(\frac{2\xi(\gamma)x_n}{a}\right) \tag{8}$$

where we have used Eq. (1) for $\varphi(x)$ and set $A = 1$—the constant A can be set to any desired value before the experiments and depends only on the power of the external actuator.

The first sum on the right-hand side of the equation vanishes because μ_n has zero mean. For the second sum, we sample the external parameter γ in discrete steps m so that $\frac{\pi}{2} \le \xi \le \pi$ (see Fig. 2). This is done explicitly as follows: write $\xi_m = \frac{m-1}{N-1}\frac{\pi}{2} + \frac{\pi}{2}$ with $m = 1, 2, \ldots, N$, then, using Eq. (5) for the link $\xi_m = \xi(\gamma_m)$, we have $\gamma_m = \frac{\tan\left(\frac{\pi}{2}\frac{m-1}{N-1}\right)}{\lambda_0}$ which, for a given N, gives the sequence of γ_m, and is equivalent to giving a prescription how to sample the external parameter.

Defining

$$\Gamma_{mn} = \cos\left(\frac{2\xi_m x_n}{a}\right) \tag{9}$$

Eq. (8) for the fractional frequency shift can be rewritten as

$$\Delta_m = \frac{1}{2}\sum_{n=1}^{N}\Gamma_{mn}\mu_n \tag{10}$$

where, to emphasize, the index n labels the position along the string, and the index m labels the external parameter such that $m = 1$ corresponds to no spring and $m = N$ to a very stiff spring.

By taking N samples of m, Eq. (10) is a square linear problem in the μ_n. The external parameter could be sampled more finely and then μ_n could be found via a minimization algorithm, but that is a detail beyond the scope of the present article. In the conclusions we comment on this point for systems with correlated mass fluctuations.

We now recall our original interest in trying to extract the ensemble distribution of frequencies for the unloaded string (corresponding to $\gamma = 0$). To solve that problem, we proceed as follows. From Eq. (10),

$$\mu_n = 2\sum_{m=1}^{N}(\Gamma^{-1})_{nm}\Delta_m. \tag{11}$$

These mass fluctuations correspond to those of an element of an ensemble of statistically equivalent random strings. We generate other equivalent disordered strings as permutations of (11),

$$\mu_n^{(q)} = \sum_{n'=1}^{N} P_{nn'}^{(q)}\mu_{n'}, \tag{12}$$

where $\mu_{n'}$ is the mass fluctuation sequence given by (11) and $P_{nn'}^{(q)}$ is the matrix element of the qth permutation of order N.

We now return to Eq. (8) and set $\gamma = 0$ and, correspondingly, $\xi = \frac{\pi}{2}$,

$$\Delta^{(q)}(0) = \frac{1}{2}\sum_{n=1}^{N}\mu_n^{(q)}\cos\left(\frac{\pi x_n}{a}\right) \tag{13}$$

where we have again used that $\langle\mu\rangle = 0$ and have introduced all the elements of the ensemble through the index q.

Then from (11) and (12),

$$\Delta^{(q)}(0) = \sum_{n,n',m} P_{nn'}^{(q)}(\Gamma^{-1})_{n'm}\Delta_m\cos\left(\frac{\pi x_n}{a}\right) \tag{14}$$

we have again used that $\langle\mu\rangle = 0$ and have introduced all the elements of the ensemble through the index q.

Eq. (14) gives the explicit connection sought. By measuring the frequencies, Δ_m, of a given realization of a random system as a function of an externally controllable parameter (in this case the spring constant) one can through Eq. (14) obtain the statistical properties of the frequencies of the ensemble with a free boundary (or any other, by changing γ to a non zero value).

5. Example

To clarify the use of the method and the notation of the paper, we show explicitly how the method works for $N = 3$. This corresponds to considering a string made up of masses μ_1, μ_2, μ_3 on the left, middle and right sections respectively. N should be large in practical situations, but we work the $N = 3$ case (a small non-trivial case) to emphasize the construction of the method.

In this case, the matrix Γ defined in Eq. (9) is

$$\Gamma = \begin{pmatrix} 1/2 & -1/2 & -1 \\ 0 & -1 & -1 \\ -1/2 & 1 & 1 \end{pmatrix}. \tag{15}$$

This is because $\Gamma_{mn} = \cos\left(\frac{2\xi_m x_n}{a}\right)$, with $\xi_m = \frac{m-1}{N-1}\frac{\pi}{2} + \frac{\pi}{2}$ and $x_n = \frac{na}{N}$, that is $\Gamma_{mn} = \cos\left[\left(1 + \frac{m-1}{N-1}\right)\frac{n\pi}{N}\right]$ which is (15), with the columns labeling n (position along the string) and the rows labeling m (the various values of the external spring and the corresponding frequencies).

From (15),

$$\Gamma^{-1} = \begin{pmatrix} 0 & -2 & -2 \\ 2 & 0 & 2 \\ -2 & -1 & -2 \end{pmatrix}. \tag{16}$$

There are 3! permutation matrices for $N = 3$ namely

$$\begin{aligned} P^{(1)} &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad P^{(2)} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad P^{(3)} = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \\ P^{(4)} &= \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 1 \\ 1 & 0 & 0 \end{pmatrix}, \quad P^{(5)} = \begin{pmatrix} 0 & 0 & 1 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}, \quad P^{(6)} = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix}. \end{aligned} \tag{17}$$

Eq. (14) gives the frequencies of the free-end string for all the elements of the ensemble, $\Delta^{(q)}(0) = \sum_{n,n',m} P^{(q)}_{nn'}(\Gamma^{-1})_{n'm}$ $\Delta_m \cos\left(\frac{\pi x_n}{a}\right)$. Here the first matrix is given in (17), the second matrix in (16), the third vector $\begin{pmatrix}\Delta_1 & \Delta_2 & \Delta_3\end{pmatrix}$ has the three measured frequencies corresponding to the three spring constants on the actual single sample and the last vector is $\begin{pmatrix}\cos(\pi x_1/a) & \cos(\pi x_2/a) & \cos(\pi x_3/a)\end{pmatrix} = (1/2, -1/2, -1)$. Substituting,

$$\begin{cases} \Delta^{(1)}(0) = \Delta_1 \\ \Delta^{(2)}(0) = -\Delta_1 - \dfrac{1}{2}\Delta_2 - 2\Delta_3 \\ \Delta^{(3)}(0) = 3\Delta_1 + 2\Delta_2 + 4\Delta_3 \\ \Delta^{(4)}(0) = 2\Delta_1 + \dfrac{5}{2}\Delta_2 + 4\Delta_3 \\ \Delta^{(5)}(0) = -3\Delta_1 + \dfrac{1}{2}\Delta_2 - 2\Delta_3 \\ \Delta^{(6)}(0) = -2\Delta_1 + \dfrac{3}{2}\Delta_2. \end{cases} \tag{18}$$

Eq. (18) gives the 3! values of the frequencies that would be measured from 3! different strings with no springs (identified by the label 0) from the same ensemble. The frequencies Δ_1, Δ_2, Δ_3 are actual laboratory measurements on the single sample available, for three different springs. Notice that the first entry is just Δ_1, corresponding to the fact that $P^{(1)}$ is the identity matrix and it does not induce a real permutation. But, out of all the possible samples one does expect to measure the frequency Δ_1 once.

This example can be generalized to any N thus generating $N!$ elements of the ensemble and the corresponding frequencies for the no-spring strings.

6. Conclusions

In this paper we have introduced a concrete model random system, namely that of a string with mass density fluctuations, and shown how to obtain the statistical fluctuations of resonance frequencies from a single sample via parametric ergodicity. The central concept in this connection is the realization that the variation of an external parameter (in this case the stiffness of the connection of one end of the string to a rigid base) induces selective mechanical excitations of various parts of the string. In this sense, scanning the external parameter is equivalent to experimenting with different samples (prepared under similar conditions). This provides a better understanding to the meaning of parametric ergodicity. At the same time, the results are of practical interest since they show an explicit algorithm to use in experiments where only few samples are available. It is worth noticing that the number of divisions N in Eq. (12) is such that the mass fluctuations are uncorrelated. This allows us to introduce the permutation matrices to produce the ensemble. The method can be generalized to correlated fluctuations by substituting Eq. (12) to one that includes correlations. For example, in past work [1] we have considered self-affine roughness, r, correlations given explicitly by $C(r) = W^2 e^{-(r/\ell)^{2\alpha}}$, where the constants W, ℓ, α represent respectively the root mean square width, correlation length and roughness exponent. W is a measure of the roughness amplitude, ℓ is a gauge of the distance at which points on the surface are no longer correlated, and α quantifies the smoothness of the surface. If we used an N larger than that given by the coherence length, then we could vary the external parameter, solve for W, ℓ, α, and then generate other samples of the ensemble by using the self-affine correlation function.

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