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Dynamic Structure Factor of Vibrating Fractals

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Abstract

Motivated by novel experimental work and the lack of an adequate theory, we study the dynamic structure factor $S(k, t)$ of large vibrating fractal networks at large wavenumbers k . We show that the decay of $S(k, t)$ is dominated by the spatially averaged mean square displacement of a network node, which evolves subdiffusively in time, $\langle (\vec{u}_i(t) - \vec{u}_i(0))^2 \rangle \sim t^\nu$, where ν depends on the spectral dimension d_s and fractal dimension d_f . As a result, $S(k, t)$ decays as a stretched exponential $S(k, t) \approx S(k)e^{-(\Gamma_k t)^\nu}$ with $\Gamma_k \sim k^{2/\nu}$. Applications to a variety of fractal-like systems are elucidated.

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Naturally occurring fractals are ubiquitous [1]. Fractal models have been used to describe the dynamics of low temperature glasses and porous materials [2], proteins [3–8], sol-gel branched polymer clusters [9], and colloidal aggregates [10]. Scattering experiments, in which one is able to simultaneously probe correlations in space and time, allow the characterization of fractal structures. A key player in these experiments is the structure factor (SF) [11, 12]. While the static SF of fractals is well understood [2, 11, 12], dynamic structure factor (DSF) calculations are limited. In the context of solid fractals, the DSF has been extensively analyzed on the “single phonon” level [2], and in the absence of any source of friction. This provides a good description for the inelastic (Brillouin) scattering from solid fractals, but is not adequate for the quasi-elastic scattering from low dimensional fractals in solutions that have large fluctuations and friction dominated dynamics, such as branched polymers and colloidal aggregates [9, 10].

In this Letter we calculate the DSF $S(k, t)$ of vibrating fractal structures. A striking example of a biological fractal in solution is the spatial organization of chromatin in the nucleus. Recent experiments [13] stand in line with long standing theoretical predictions [14] suggesting a fractal (crumpled) globule structure. And yet, other fractal chromatin structures were also claimed consistent with experimental results [15]. Combined with the theory presented herein, DSF studies may aid resolving this conflict. Interestingly the DSF has been measured in a different biological system in which neutron spin-echo (NSE) studies were performed on horse heart myoglobin and bovine hemoglobin in solutions. In the large wavenumber k regime corresponding to $kR_g \gg 1$, where R_g is the gyration radius, and at low concentrations and times shorter than 1ns, the DSF decays as a stretched exponential [16]. As we demonstrate here this decay is distinctive of fractal structures and the fractal-like nature of proteins [4–6, 8] make them a natural case study for our theory. A stretched exponential decay of the DSF is also observed in dynamic light scattering experiments from suspensions of soft colloids that form glasses at large volume fractions. In these systems a universal stretching exponent that is independent of the volume fraction is found [17]. These findings can be explained by the theory advanced here assuming similarity of the structure (or, more precisely, of the force constant network) between these glasses and 3D percolation networks, as done for solid glasses [2].

Fractals are characterized by a few broken dimensions [18]: (i) the mass fractal dimension d_f , that governs the scaling $M(r) \sim r^{d_f}$ of the mass $M(r)$ enclosed in concentric spheres of ra-

dius r , (ii) the topological dimension d_l that governs the scaling $M(l) \sim l^{d_l}$ of the mass $M(l)$ enclosed in concentric “spheres” of radius l in the topological (or “manifold”/“chemical”) space, and (iii) the spectral dimension d_s that governs the scaling $g(\omega) \sim \omega^{d_s-1}$ of the vibrational density of states $g(\omega)$ with frequency ω [2]. The fractal dimension is experimentally measurable as it governs the power law behavior of the static SF. While known to be related, a general theory explaining the decay of the DSF in terms of d_f and d_s is still lacking and elucidation of d_s based on the DSF is usually not possible.

The main result of this Letter can be simply stated as follows. Assume a large bead-spring fractal network with $d_s < 2$ and arbitrary d_f , and consider a scattering experiment at large wavenumbers k such that both $kR_g \gg 1$ and $k\bar{u} \gg 1$, where $\bar{u} \equiv \sqrt{\langle u^2 \rangle}$ is the root mean square bead displacement. The latter obeys the generalized Landau-Peierls instability [5, 6], $\bar{u} \sim N^{1/d_s-1/2}$ where N is the number of beads. In this limit, and within a wide window of time, we find that $S(k, t)$ decays as a stretched exponential $S(k, t) \simeq S(k)e^{-(\Gamma_k t)^\nu}$, where the relaxation rate anomalously depends on k , $\Gamma_k \sim k^{2/\nu}$. The stretched exponential relaxation is a consequence of the anomalous diffusion of a network bead, with a mean square displacement (MSD) evolving as $\sim t^\nu$. The exponent ν depends on the fractal and spectral dimensions, $\nu = 1 - d_s/2$ in a Rouse model where the friction is local [3, 6, 19, 20], $\nu = (2 - d_s)/(2 - d_s + d_s/d_f)$ in a Zimm model where the friction is long range [7], and $\nu = 2 - d_s$ for vanishing friction [3]. The latter situation applies for solid fractals, yielding a result which goes much beyond the “single phonon/fracton” approach used previously and valid only for $k\bar{u} \ll 1$ [2]. Our result allows for an experimental evaluation of d_s .

We repeat briefly the definitions and assumptions of the scalar elasticity model [2]. The ground configurational state of the fractal is described by the set of coordinates $\vec{R}_{eq}(\vec{l})$, where \vec{l} is the coordinate of a bead in topological space, and deviations from the ground state are denoted by the displacements $\vec{u}(\vec{l}) = \vec{R}(\vec{l}) - \vec{R}_{eq}(\vec{l})$. The scalar elasticity “bead-spring” Hamiltonian is [2]

$$H \left[\{ \vec{u}(\vec{l}) \} \right] = \frac{1}{2} m \omega_o^2 \sum_{\langle \vec{l}\vec{l}' \rangle} \left(\vec{u}(\vec{l}) - \vec{u}(\vec{l}') \right)^2, \quad (1)$$

where $\langle \vec{l}\vec{l}' \rangle$ stands for pairs connected by springs, ω_o is the spring self-frequency, and m is the bead mass ($m\omega_o^2$ is the spring constant). The eigenstates (normal modes) of the Hamiltonian (1) form an orthonormal set and, on a fractal, are strongly localized in space, bearing the name “fractons” [2, 21].

In order to derive the DSF of vibrating fractals, we discuss first the relevant displacement pair correlation function $\langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}', 0))^2 \rangle$, where it is understood that spatial averaging has been performed, thus making this correlation function depend only on the relative separation $|\vec{\ell} - \vec{\ell}'|$ in topological space.

In Refs. [3, 6, 7] we derived the normal mode space Langevin equations for fractals in the high damping and vanishing damping limits. In the high damping limit, which is our main focus here due to its relevance to fractal dynamics in solutions, two models were considered [11, 20]: (i) a Rouse type model in which the hydrodynamic friction is local [3], and (ii) a Zimm type model where we accounted for the long range hydrodynamic interaction between different beads, that is transmitted through the velocity field of the solvent [7]. For both models, the Langevin equations of motion in the mode space can be written in the form

$$\frac{d\vec{u}_\alpha}{dt} = -\Gamma_\alpha \vec{u}_\alpha + \vec{\zeta}_\alpha(t) . \quad (2)$$

where $u_\alpha(t)$ is the amplitude of a normal mode α at time t , $\Gamma_\alpha = m\omega_\alpha^2 \Lambda_\alpha$ is the mode relaxation rate, $\vec{\zeta}_\alpha(t)$ is thermal white noise that obeys the fluctuation-dissipation theorem

$$\langle \vec{\zeta}_\alpha(t) \vec{\zeta}_\beta(t') \rangle = 2k_B T \Lambda_\alpha \delta_{\alpha,\beta} \delta(t - t') , \quad (3)$$

and Λ_α is the mode mobility coefficient. The dependence of Λ_α on ω_α is sensitive to the hydrodynamic model in question. To account for both models in a single formula, we shall write the relaxation rate as $\Gamma_\alpha \simeq \bar{A} \omega_\alpha^\theta$, where (i) in the Rouse model: $\theta = 2$; $\bar{A} = m/(3\pi\eta b)$ where η is the solvent viscosity and b is the bead diameter [3], and (ii) in the Zimm model: $\theta = 2 - d_s + d_s/d_f$; $\bar{A} = A m/(6\pi\eta b \omega_o^{d_s/d_f - d_s})$, where A is a numerical constant [7].

We use Eq. (2) to evaluate the two-point correlation function for the time regime $\tau_0 \ll t \ll \tau_N$, where $\tau_0 = \bar{A}^{-1} \omega_o^{-\theta}$ and $\tau_N \simeq \bar{A}^{-1} \omega_o^{-\theta} N^{\theta/d_s}$ are the shortest and longest vibrational relaxation times (respectively). We obtain the following scaling form

$$\langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}', 0))^2 \rangle = \frac{k_B T}{m\omega_o^{d_s}} (\bar{A}t)^\nu \Phi \left[|\vec{\ell} - \vec{\ell}'|/\ell(t) \right] , \quad (4)$$

where $\Phi[v]$ is the scaling function [22, 23] ($\Phi[0] = \text{const.}$) and $\ell(t) = \omega_o^{d_s/d_f} (\bar{A}t)^{\frac{d_s}{d_f\theta}}$ is the (dimensionless) length describing the propagation with distance, in topological space, of the bead-bead correlations or localized perturbations. In real space, this (dimensioned) propagation length is $\xi(t) \simeq b\ell(t)^{d_i/d_f} = b \omega_o^{d_s/d_f} \bar{A}^\zeta t^\zeta$ where $\zeta = d_s/(d_f\theta)$. Putting $\vec{\ell} = \vec{\ell}'$

in Eq. (4) it reduces simply to the (spatially averaged) MSD of a bead. Provided that $d_s < 2$, it shows the familiar *anomalous subdiffusion*

$$\langle \Delta \vec{u}(t)^2 \rangle \equiv \langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}, 0))^2 \rangle = B t^\nu . \quad (5)$$

where $\nu = (2 - d_s)/\theta$ and B is a constant.

The scaling form Eq. (4) implies a crossover behavior around a time $t^*(r)$ for pairs at a distance $r \equiv |\vec{R}_\ell - \vec{R}_{\ell'}|$ apart, $t^*(r) = \bar{A}^{-1} \omega_o^{-\theta} (r/b)^{d_f \theta / d_s}$ [24]. For $t \ll t^*(r)$, the correlation function is very close to its static value

$$\langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}', 0))^2 \rangle \approx \frac{k_B T}{m \omega_o^2} \left(\frac{r}{b} \right)^{d_w - d_f} . \quad (6)$$

where $d_w = 2d_f/d_s$. For longer times, $t^*(r) \ll t \ll \tau_N$, such that information has propagated much beyond the distance r , it approaches the MSD of a single bead, $\langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}', 0))^2 \rangle \simeq B t^\nu$, implying that the two beads are essentially moving together.

We note that the two-point correlation function Eq. (4) can be transformed from topological space to the real 3D Euclidean space,

$$\langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}', 0))^2 \rangle = \frac{k_B T}{m \omega_o^2} \left(\frac{r}{b} \right)^{d_w - d_f} \Xi \left[\frac{t}{t^*(r)} \right] . \quad (7)$$

$\Xi[u]$ has the following asymptotes: (i) $\Xi[u] \simeq \text{const.}$ for $u \ll 1$, and (ii) $\Xi[u] \sim u^\nu$ for $u \gg 1$. Eq. (7) is particularly useful for the numerical analysis that we perform next.

To test the above analytic expression we first evaluate numerically the pair correlation function on a vibrating Sierpinski gasket obeying the Rouse dynamics [1, 2, 18]. In Fig. 1(a) we plot, on a log-log scale, four averaged two-point correlation functions against the normalized time t/τ_0 [25]. Note the crossover from a constant value, that increases with increasing r as predicted by Eq. (6), to an anomalous subdiffusion time regime, identical to that of the single particle MSD (effectively equal to the correlation function for $r = b = 5A$). In the subdiffusion regime, the behavior is essentially independent of the distance r . Also note that the crossover time increases with increasing r as implied by the scaling with r of $t^*(r)$. A normalized version of Fig. 1(a), shown in Fig. 1(b), asserts the validity of Eq. (7).

We now turn to calculate the fractal DSF with rotational and translational motion arrested,

$$S(\vec{k}, t) \equiv \frac{1}{N} \left\langle \sum_{\vec{\ell}, \vec{\ell}'} e^{i\vec{k} \cdot (\vec{R}(\vec{\ell}, t) - \vec{R}(\vec{\ell}', 0))} \right\rangle , \quad (8)$$

where $\vec{R}(\vec{\ell})$ is the coordinate of a node $\vec{\ell}$ in the center of mass coordinate frame and the sums run over all network nodes. Changing to displacement variables, $\vec{R}(\vec{\ell}, t) = \vec{R}_{\text{eq}}(\vec{\ell}) + \vec{u}(\vec{\ell}, t)$, using the Gaussian property of the stochastic variable $\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}, 0)$ [11] and the isotropy of the scalar elasticity model, and performing angular averaging, we find, omitting from now on the subscript “eq” in $\vec{R}_{\text{eq}}(\vec{\ell})$,

$$S(k, t) = \frac{1}{N} \sum_{\vec{\ell}, \vec{\ell}'} \frac{\sin [k R_{\ell\ell'}]}{k R_{\ell\ell'}} e^{-\frac{k^2}{6} \langle (\vec{u}(\vec{\ell}, t) - \vec{u}(\vec{\ell}', 0))^2 \rangle}, \quad (9)$$

where $R_{\ell\ell'} = |\vec{R}(\vec{\ell}) - \vec{R}(\vec{\ell}')|$ is the Euclidean, real-space, distance between beads $\vec{\ell}$ and $\vec{\ell}'$.

At short times, $t \ll t^*(k^{-1}) \sim k^{-d_f \theta / d_s}$, information did not have time to negotiate a “blob” of linear size $\sim k^{-1}$ and we find that the DSF did not decay much, $S(k, t) \simeq S(k)$. At longer times, $t^*(k^{-1}) \ll t \ll \tau_N$, i.e. when $1 \ll k\xi(t) \ll kR_g$, information has propagated beyond the scattering wavelength $\sim 1/k$. Physically, this implies that the “blob” of size $\sim 1/k$, that is controlling the relaxation at wavevector \vec{k} , is now moving almost coherently as if it was a single bead. At this time regime we find a *stretched exponential* decay of the DSF,

$$S(k, t) \approx S(k) \exp[-(\Gamma_k t)^\nu] \quad (10)$$

where

$$\Gamma_k = (B/6)^{1/\nu} k^{2/\nu}. \quad (11)$$

Note that the stretching exponent is exactly the anomalous diffusion exponent ν . The stretched exponential decay, together with the dependence of the stretching exponent ν on the broken dimensions d_s and d_f , is thus a strong signature of the fractal structure. Corrections due to rotational and translational diffusion are considered elsewhere and are shown to vanish for large fractals [23]. In Fig. 2 we plot the DSF (and SF) for the Sierpinski gasket.

The anomalous wavenumber dependence of the relaxation rate, $\Gamma_k \sim k^{2/\nu}$, and the anomalous diffusion, $\langle \Delta \vec{u}(t)^2 \rangle \sim t^\nu$, can be explained using simple scaling hypotheses. For the relaxation rate we assume $\Gamma_k = D k^2 h(k\bar{u})$, where $h(x)$ is a scaling function and D is the center of mass diffusion coefficient of the fractal. We make use of the generalized Landau-Peierls instability, $\bar{u} \sim N^{1/d_s - 1/2}$, and take $D \sim N^{-1}$ and $D \sim R_g^{-1} \sim N^{-1/d_f}$ for the Rouse and Zimm type models of friction [11], respectively. Demanding that Γ_k is independent of N for $k\bar{u} \gg 1$, the scaling function for $x \gg 1$ must satisfy $h(x) \sim x^{2d_s/(2-d_s)}$ (for the

Rouse model) and $h(x) \sim x^{2d_s/[d_f(2-d_s)]}$ (for the Zimm model), leading to $\Gamma_k \sim k^{2/\nu}$ with $\nu = 1 - d_s/2$ (Rouse) and $\nu = (2 - d_s)/(2 - d_s + d_s/d_f)$ (Zimm) as stated. Similarly, for the MSD we assume $\langle \Delta \vec{u}(t)^2 \rangle = \bar{u}^2 y(t/\tau_N)$ where $y(x)$ is the scaling function and τ_N is the longest vibrational relaxation time. Assuming $\tau_N \simeq \bar{u}^2/D$, such that $\tau_N \sim N^{2/d_s}$ (Rouse) and $\tau_N \sim N^{2/d_s - 1 + 1/d_f}$ (Zimm), and demanding that for $t \ll \tau_N$ the MSD is independent of N , it follows that $y(x) \sim x^\nu$ for $x \ll 1$ (with ν taking the above stated values associated with the Rouse and Zimm models) leading to $\langle \Delta \vec{u}(t)^2 \rangle \sim t^\nu$.

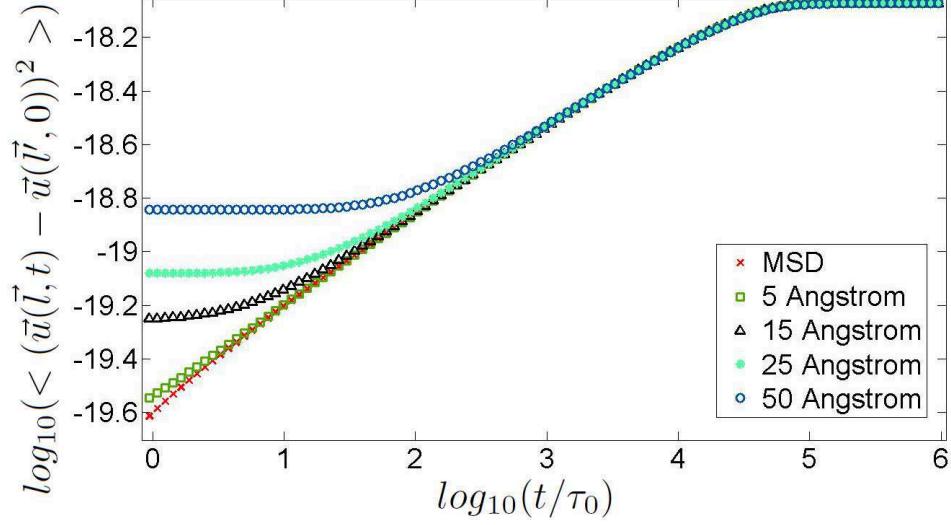
In this Letter we presented a through study of the DSF $S(k, t)$ of vibrating fractals in solutions. Our main result is that at large wavenumbers the decay of the DSF is strongly influenced by the anomalous diffusion of the spatially averaged MSD of a network bead. The result for large fractals is a stretched exponential decay of the DSF, which reduces to the known DSF of a linear Rouse polymer chain ($d_s = 1$), and a linear-Gaussian Zimm polymer chain ($d_s = 1, d_f = 2$) [11]. Among possible applications are: (i) Proteins, for which a stretched exponential decay has been recently measured by NSE [16], supporting their fractal-like structure. (ii) Glass forming colloidal suspensions [17]. Assuming analogy to 3D percolation network, that implies $d_f \simeq 2.48$ and $d_s \simeq 1.328$, and accounting for the hydrodynamic coupling, we suggest a stretched exponential decay with $\nu \simeq 0.556$, remarkably close to the observed value $\nu \simeq 0.6$. (iii) Colloidal gels [26], that show a clear fractal structure and for which a Zimm-type dynamics and bond-bending potential explains well the observed stretched exponential decay [7]. (iv) Chromatin [14, 15], for which it was recently shown that telomeres perform anomalous subdiffusion with $\nu \simeq 0.32$ [27]. This may be interpreted, within the Rouse model that yields $\nu = 1 - d_s/2$ (noting that in such a dense polymer system hydrodynamics is likely to be screened), by $d_s \simeq 1.36$. This value of d_s is remarkably close to that of percolation clusters in $2 < d < 5$ dimensions and suggests the presence of DNA crosslinks (e.g., via ligation). According to the present calculation, it is suggested that the DSF of chromatin will decay as a stretched exponential with stretching exponent $\simeq 0.32$, which can motivate experiments in this direction. We believe that our results can also be applied to a variety of other systems exhibiting fractal structure.

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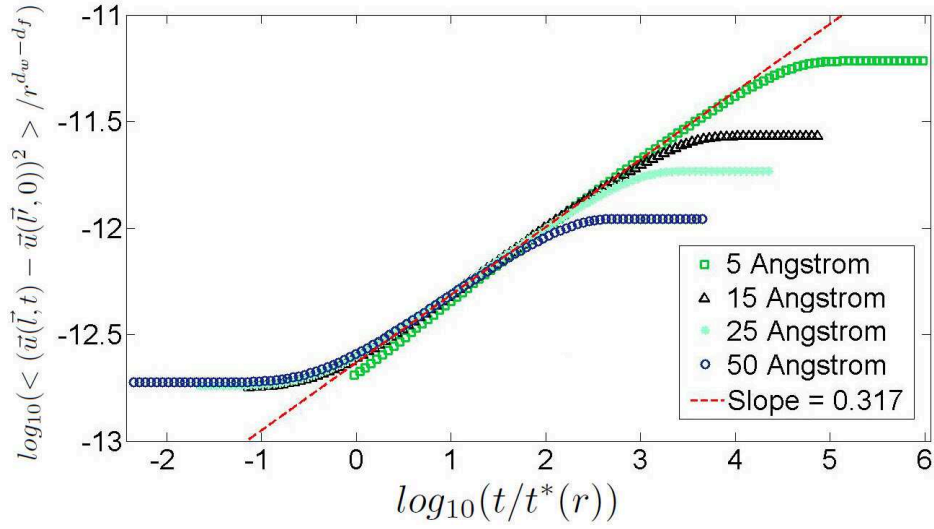
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actual models are quite different from their polymer analogs.

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- [24] Note that $t^*(R_g) \simeq \tau_N$.
- [25] We use $b = 5 \times 10^{-10}$ m, $m = 4 \times 10^{-25}$ kg, $\omega_o = 10^{12}$ s⁻¹, $\eta = 8.94 \times 10^{-4}$ Pa s, $T = 298$ K, yielding $\tau_0 \simeq 10.53$ ps. The size $N = 6561$ corresponds to $R_g \simeq 42.6$ nm and $\tau_N \simeq 4.11$ μ s. These parameter values are similar to those in proteins, except for N which is purposely exaggerated [23].
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(a)



(b)

FIG. 1: **(a)** The pair correlation function $\langle (\vec{u}(\vec{l}, t) - \vec{u}(\vec{l}, 0))^2 \rangle$ is evaluated numerically, using the Rouse-type model, for bead pairs located on a vibrating Sierpinski gasket with 6561 nodes [25]. For a *fixed* value of the inter-bead distance r , $\langle (\vec{u}(\vec{l}, t) - \vec{u}(\vec{l}, 0))^2 \rangle$ is calculated for all pairs distanced $r \pm \frac{1}{2}\text{\AA}$ apart where $r = 5, 15, 25, 50\text{\AA}$. For every point in time, the correlation functions, in each distance group, are averaged over all pairs in that group. **(b)** Following the predicted scaling behavior stated in Eq. (7), we normalize the correlation functions from (a) by $r^{d_w - d_f}$ and the time by $t^*(r) \sim r^{d_w}$. Data collapse to a single master curve is observed for $\tau_0 \ll t \ll \tau_N$. The slope ν in the subdiffusive time regime is found to be 0.317, in excellent agreement with the theoretical value of $\nu = 1 - d_s/2 \simeq 0.317$.

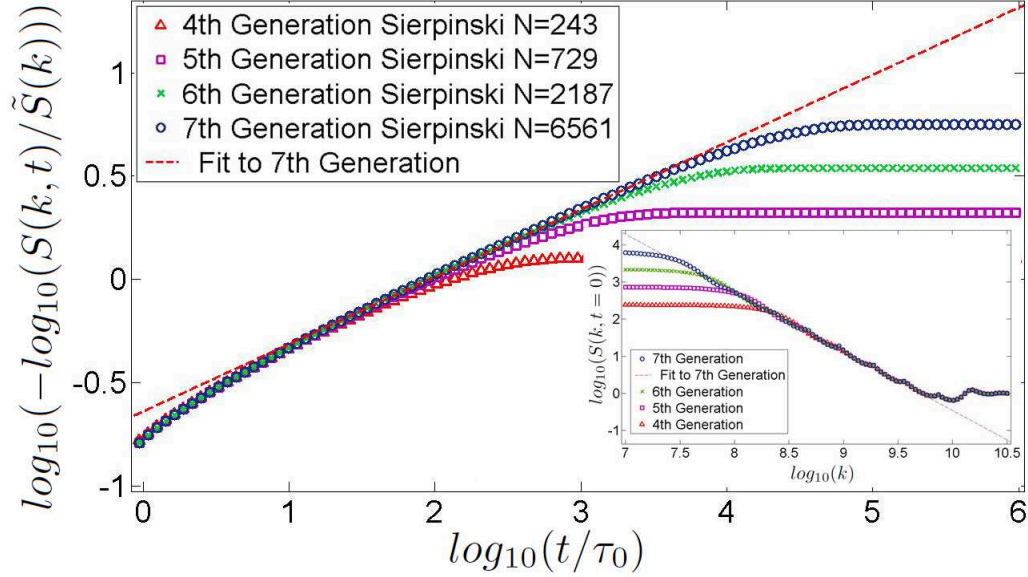


FIG. 2: $\log_{10}(-\log_{10}(S(k,t)/\tilde{S}(k)))$ for $k = 10^{10}\text{m}^{-1}$ is plotted *vs.* $\log_{10}(t/\tau_0)$ for vibrating Sierpinski gaskets of various sizes (Rouse model) [25]. Here $\tilde{S}(k)$ is the frozen SF (Eq. (9) with $e(\dots)$ set to unity). Note that a straight line, whose slope is positive, is formed on an intermediate time window that widens up as we move from smaller to larger gaskets, demonstrating the diminishing contribution of finite size effects. The stretching exponent obtained from the plot, 0.325, is in excellent agreement with the theoretical value $\nu \simeq 0.317$. **Inset:** The static SF, $S(k) \equiv S(k, t = 0)$, against k on a log-log scale. A clear power law decay, with an exponent of -1.583 , is visible for all gaskets, in excellent agreement with theory [2, 12] $\tilde{S}(k) \sim k^{-d_f}$, $d_f \simeq 1.585$. In contrast to $\tilde{S}(k)$, $S(k)$ does account for the contribution of thermal vibrations. However, as is evident from the plot, vibrations have a negligible effect on the static SF for the chosen parameter values [25] and $S(k) \simeq \tilde{S}(k)$.