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Vibrational spectra of a diffusion-limited aggregate

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Abstract. The vibrational densities of states for the diffusion-limited aggregate with varying elastic force constants are calculated by the recursion method of Haydock, Heine and Kelly. It is found that the spectrum with central forces is very different from that with isotropic forces which is characterized by a power-law behaviour in the fracton frequency regime. An interesting crossover is also observed when the ratio of the central to the non-central force constants of the Born model varies from $\alpha/\beta = 1$ to $\alpha/\beta = \infty$.

1. Introduction

Fractal structures are abundant in nature, and the development of fractal geometry (Mandelbrot 1982) led to remarkable advance in the description of many phenomena. Recently the elastic and vibrational properties of fractals have aroused much interest. In particular, much work has been carried out on the percolation system. For a period of time, the elasticity of random percolation network had been viewed as analogous to the problem of electrical conductivity of such a system. This analogy was first suggested by de Gennes (1976) in relation to the elasticity of gels and was later applied more generally (Hsu *et al* 1982). It can be understood within the framework of the Born model (Born and Huang 1954) with isotropic forces. In this model, the potential energy of the lattice is given by

$$V = \tfrac{1}{2}(\alpha - \beta) \sum_{\substack{\text{nn} \\ ij}} [(\boldsymbol{u}_i - \boldsymbol{u}_j) \cdot \hat{\boldsymbol{r}}_{ij}]^2 g_{ij} + \tfrac{1}{2}\beta \sum_{\substack{\text{nn} \\ ij}} |\boldsymbol{u}_i - \boldsymbol{u}_j|^2 g_{ij} \tag{1}$$

where $\boldsymbol{u}$ is the displacement, $\hat{\boldsymbol{r}}_{ij}$ is the unit vector from site i to site j; $g_{ij} = 1$ if sites i and j are both occupied and $g_{ij} = 0$ otherwise; the summation runs over all nearest neighbours. For the purely isotropic Born model, i.e. $\alpha = \beta$ in (1), the different Cartesian components of the displacements decouple:

$$V = \tfrac{1}{2}\beta \sum_{\mu} \sum_{\substack{\text{nn} \\ ij}} (u_{i\mu} - u_{j\mu})^2 g_{ij}. \tag{2}$$

It is obvious that each component maps exactly onto the scalar electrical conduction problem on such a network if one takes the displacement components to denote the voltage and β for the conductance of an occupied bond. Thus the isotropic Born model reduces to a scalar problem. On the basis of this scalar model, two results were obtained.

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(i) Young's modulus for the percolation system falls to zero as p approaches p_c as $Y \sim (p - p_c)^t$, where t is the percolating conductivity exponent, as noted by de Gennes (1976).

(ii) The concept of the fracton (Alexander and Orbach 1982, Rammal and Toulouse 1983) was introduced as the vibrational excitation of fractal structures. The vibrational density of states in the fracton frequency regime follows a power law $N(w) \sim w^{d_s-1}$ where d_s is the fracton dimensionality.

The above-presented results are elegant, but they fail to take into account the vector nature of the most realistic elastic systems. Recent developments (Feng and Sen 1984, Kantor and Webman 1984, Bergman 1985, Lemieux *et al* 1985, Deptuek *et al* 1985) have shown that percolation networks with central forces can have very different critical exponents for elastic moduli from those of the scalar model, i.e. $Y \sim (p - p_c)^f$ where f is different from the percolating conductivity exponent t. Thus it is important to investigate the vibrational density of states of fractal structures when the vector nature of the elastic forces is included. This paper presents the calculations of the vibrational spectra of a specific fractal: the diffusion-limited aggregate (DLA). In the solid state, fractals are commonly produced by aggregation processes. The DLA, first proposed by Witten and Sander (1981), provides a simple but non-trivial fractal model for a variety of aggregation and growth phenomena (Meakin 1988, Feder 1988). In section 2, we outline the method used in our work and the approximation involved. The calculations are presented and discussed in section 3.

2. Calculation method

We adopt the widely used recursion method of Haydock *et al* (1972, 1975) which offers fast computational speed and, above all, reliability. The vibrational density of states (VDOS) can be expressed as (Peng and Tian 1989)

$$N(w) = -(2\omega/\pi)\,\mathrm{Im}[\mathrm{Tr}(w_+^2 - \mathbf{W})^{-1}] = -(2\omega/\pi)\,\mathrm{Im}(\mathrm{Tr}\,\mathbf{G}) \tag{3}$$

where $\mathbf{G} = (w_+^2 - \mathbf{W})^{-1}$ is the Green function, $\mathbf{W}$ is the dynamical matrix, $w_+ = w + \mathrm{i}\mu$ ($\mu \to 0$). The trace of G can be written explicitly, and the VDOS then turns into a sum of local densities of states

$$N(\omega) = -\frac{2\omega}{\pi}\,\mathrm{Im}\left(\sum_i \langle u_i|\mathbf{G}|u_i\rangle\right) \tag{4}$$

where $|u_i\rangle$ is the displacement vector of site i. Unlike crystal and amorphous material, fractal structures have no characteristic length, i.e. the fractal is not uniform on any length scale, and so it is inconvenient to calculate the global VDOS from the superposition of local VDOSs. Instead, we choose an initial vector whose elements are random variables chosen from a distribution:

$$|0\rangle = \sum_i \xi_i |u_i\rangle \tag{5}$$

where ξ_i is the random variable. For a specific configuration $\{\xi_i\}$, one has

$$\langle 0|\mathbf{G}|0\rangle = \sum_i \xi_i^2 \langle u_i|\mathbf{G}|u_i\rangle + \sum_{i \neq j} \xi_i \xi_j \langle u_i|\mathbf{G}|u_j\rangle. \tag{6}$$

Averaging over the configurations yields

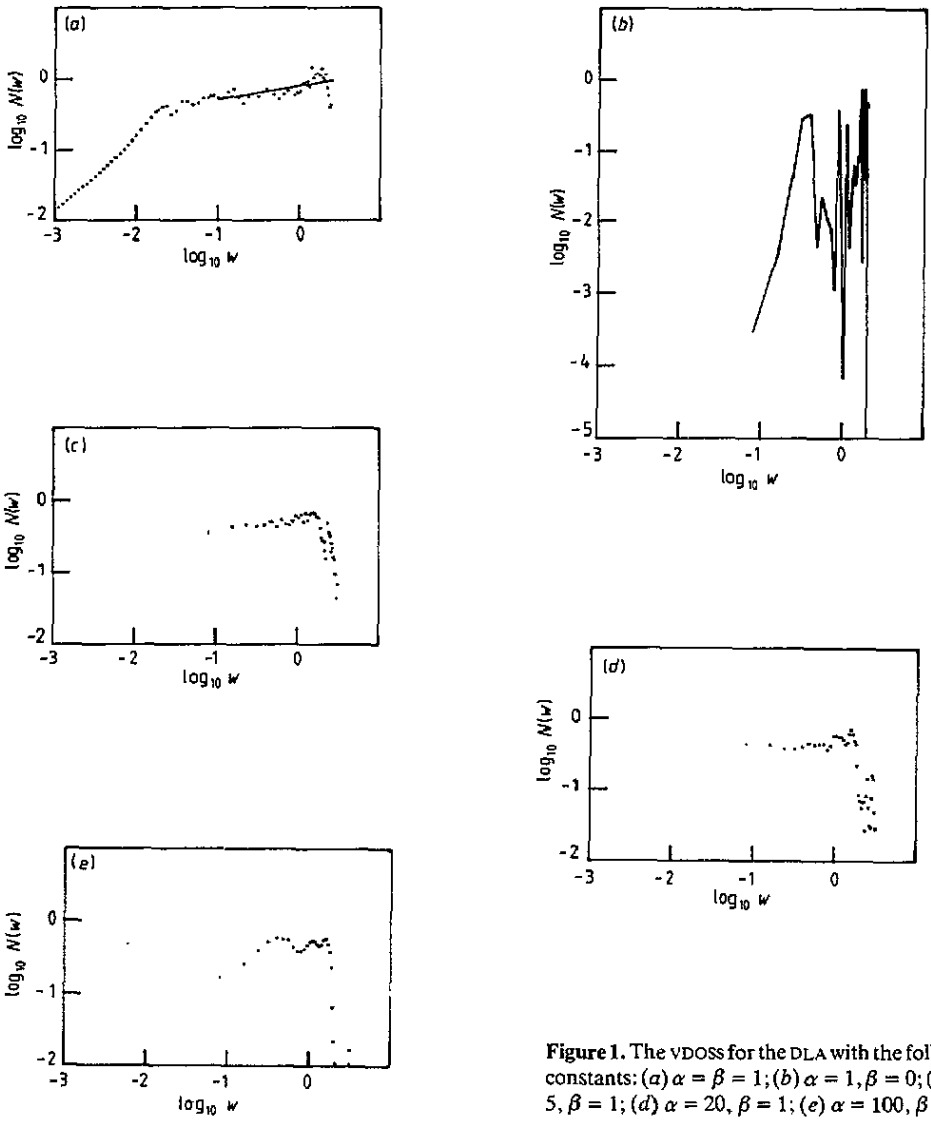

Figure 1. The VDOSs for the DLA with the following constants: (*a*) $\alpha = \beta = 1$; (*b*) $\alpha = 1, \beta = 0$; (*c*) $\alpha = 5, \beta = 1$; (*d*) $\alpha = 20, \beta = 1$; (*e*) $\alpha = 100, \beta = 1$.

$$\overline{\langle 0|\mathbf{G}|0\rangle} = \sum_i \overline{\xi_i^2}\langle u_i|\mathbf{G}|u_i\rangle + \sum_{i\neq j} \overline{\xi_i \xi_j}\langle u_i|\mathbf{G}|u_j\rangle. \tag{7}$$

We take the ξ_i-values as the uncorrelated random variables with mean zero and covariance 1, i.e.

$$\overline{\xi_i \xi_j} = \delta_{ij}. \tag{8}$$

So it is obvious that

$$N(\omega) = -(2\omega/\pi)\,\mathrm{Im}\,\overline{\langle 0|\mathbf{G}|0\rangle}. \tag{9}$$

In the calculation, random variables are chosen from a Gaussian distribution, and ten initial vector configurations have been averaged to get the global VDOS. The diagonal

element of the Green function $\langle 0|\mathbf{G}|0\rangle$ is obtained through the use of the recursion method which defines a new basis of orthonormal vectors $|n\}$ according to the recursive relation

$$b_{n+1}|n+1\} = (\mathbf{W} - a_n)|n\} - b_n^*|n-1\} \tag{10}$$

with initial conditions $|0\} = |0\rangle, |-1\} = 0, b_{-1} = 0, b_0 = 1$. As $\mathbf{W}$ is a tri-diagonal matrix in the new basis, it is easy to invert $w^2 - \mathbf{W}$ analytically to obtain the continued-fraction expansion of $\langle 0|\mathbf{G}|0\rangle$:

$$\langle 0|\mathbf{G}|0\rangle = 1/[\omega^2 - a_0 - |b_1|^2/(\omega^2 - a_1 - |b_2|^2/\ldots)] \tag{11}$$

where (a_n, b_n) are evaluated from the orthonormality property of the new basis.

3. Results and discussion

Our results are obtained in two steps. The first is the generation of the fractal; the second is the recursive calculation of the VDOS. The generation is after the algorithm of Witten and Sander (1981). One places a seed particle on a square lattice at time $t = 1$. At $t = 2$ a second particle is released from a random point on a circle surrounding and far away from the seed particle and allowed to undergo a random walk until it sticks irreversibly to the seed. At $t = 3$, a third particle is released, and this process continues until a large number of particles have been formed. The cluster size used in this work is 2314 particles. Using the density–density correlation function, we find the fractal dimension $D = 1.67 \pm 0.02$.

To calculate the dynamical matrix, we employ the Born model. For the isotropic case $\alpha = \beta$, the VDOS at low frequencies follows a power law $N(w) \sim w^{d_s - 1}$ down to the lowest frequency $w_{\min}$ determined by the finite size of the cluster. The power-law behaviour coincides with the fracton model (Alexander and Orbach 1982). Figure 1(*a*) illustrates the VDOS with free boundary condition when $\alpha = \beta = 1$. Since the system is not fractal on a length scale shorter than the wavelength $\lambda \sim 1$, the VDOS does not exhibit power-law behaviour above $w \sim 2\pi$. Fitting a least-squares line over the range from $\log_{10} w = -1.1$ to $\log_{10} w = 0.31$, we obtain the fracton dimensionality $d_s = 1.20 \pm 0.06$. This result is consistent with the diffusion studies of Meakin and Stanley (1983). Below $w_{\min}$ the VDOS fits another power law $N(w) \sim w^{d_1 - 1}$ with $d_1 = 2.09$, which is the result of the finite-size effects.

When the vector nature of the vibration is considered, i.e. $\alpha \neq \beta$, the VDOS may be expected to deviate from the prediction of the fracton model. In the case where $\alpha = 1$ and $\beta = 0$ in (1), the nearest-neighbour forces are central and the bond behaves as an ordinary spring. This purely central-force Born model is rotationally invariant. Figure 1(*b*) shows the spectrum in the frequency range above $w_{\min}$ when $\alpha = 1$ and $\beta = 0$. We see that the VDOS deviates from power-law behaviour drastically. An interesting crossover is observed when both central and isotropic forces are included. Figures 1(*c*)–1(*e*) show the spectra with the following α- and β-values, respectively: $\alpha = 5, \beta = 1$; $\alpha = 20, \beta = 1$; $\alpha = 100, \beta = 1$.

Feng (1985) has proposed a scaling argument on the low-frequency vibrational density of states for a percolation system with rotationally invariant elastic forces. He found that there exists a new crossover length scale l_c which depends on the relative strength of the microscopic bond-stretching and bond-bending elastic force constants such that, when the connectivity length ξ is much larger than l_c, the VDOS exhibits two different power-law behaviours in two different frequency ranges, with a crossover

between them. His results were based on the percolation model of bond-bending type. Although the Born model is not incorporated in the Feng model, the deviation of the VDOS from the power-law behaviour is observed by both Feng's scaling argument and our numerical calculation. This deviation is caused by the vector nature of the elastic forces. It is noted that the model discussed by Alexander (1984) corresponds to systems under internal stresses that are not explicitly rotationally invariant, whose elastic and vibrational properties fall into the same universality as the scalar model. Our model corresponds to systems which are not subjected to any significant internal stress.

4. Summary

We have calculated the VDOS for a specific fractal, the DLA, with central forces between the nearest neighbours and found the spectrum deviates drastically from that with isotropic forces which is characterized by a power-law behaviour in the fracton frequency regime. An interesting crossover is observed when both the central and the isotropic forces are included simultaneously.

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