Additivity of vibrational density of states in two-dimensional mesoscopic systems

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We investigate the vibrational density of states (DOS) in two-dimensional (2D) composite systems with nonhomogeneous geometry. The following three objects are selected as case studies: (i) the union between a 1D and a 2D crystallite; (ii) the union of a mass fractal with a 2D crystallite; and (iii) the union of a surface fractal with a 2D crystallite. In each case, it is found that the DOS of the composite system is, within a very good approximation, equal to the sum of the DOS of the components. This indicates the absence of a long-range contribution to the DOS of 2D macroscopic systems. This quantity can therefore be directly evaluated from the simple average of the DOS of its tessellated mesoscopic elements. The calculation of the vibrational DOS of a macroscopic solid can then be reduced to a feasible computational operation.

The knowledge of the density of states (DOS) of transverse phonons and quantum states is an essential tool to understand the properties of solids.[1] For large periodic systems of dimensionality d, the low-frequency DOS of the acoustic branches in the phonon spectra obeys the usual Debye law $\rho(\omega) \sim \omega^{d-1}$. When translational invariance is lost, however, the use of a simple geometrical description based on the global dimension d is not sufficient to characterize the vibrational behavior of the system. This is the case for disordered structures like amorphous and nonstoichiometric solids, polymers, polyatomic glasses, and fractals. In particular, it is known that the substitution of the dimensionality d by the fractal dimension in the Debye relation for phonons in fractal lattices gives erroneous predictions.[2–4] For systems in which the so-called "spectral partition" is possible, it has been theoretically predicted that a weighted average of the DOS can be used to characterize low-frequency vibrations.[5]

It is always possible to consider the macroscopic geometry of complex solids as simple unions of mesoscopic irregular cells. Being small, these objects are accessible to numerical simulation. If a physical quantity is shown to be additive at the mesoscopic scale, one can then reduce the complexity of the macroscopic system to a mesoscopic level. Note also that, in disordered solids, many interesting phenomena take place within the mesoscopic scale (e.g., the diffusion of phonons).

In the present communication, the aim is to compare the vibrational DOS of composite systems with the sum of the DOS of their isolated mesoscopic components. The result is that the DOS adds in a simple manner over the entire range of frequencies. Herein, we consider mesoscopic systems whose properties are those of irregular resonators, irregular drums or irregular quantum dots. These systems may exhibit anomalous localization and anomalous DOS behavior that have been theoretically investigated[4–6] and observed in recent experiments.[7]

In order to compute the eigenfrequencies of transverse vibrations in a two-dimensional (2D) system, we solve the following set of coupled differential equations:

$$m\frac{\partial^2 u_j}{\partial t^2} = \sum_\nu k(u_{j+\nu} - u_j) \text{ for } j = 1,2,\ldots,N, \qquad (1)$$

where m is the mass, N is the total number of particles, k is the spring stiffness, and the summation includes all nearest neighbors ν of particle j. The standard *Ansatz*, $u_{n,j} = u_j \exp(-i\omega_n t)$, leads to

$$m\omega_n^2 u_{n,j} = \sum_\nu k(u_{n,j} - u_{n,j+\nu}). \qquad (2)$$

This is an eigenvalue equation that can only be solved numerically for an irregular spatial distribution of mass. The different systems considered here are composites whose components may or may not be irregular in shape. Being of mesoscopic size, they contain between hundreds and thousands of atoms. As a consequence, the numerical computation of their eigenstates is accessible to modern numerical techniques.

The structures studied here have been selected in order to test the hypothesis of DOS additivity for the case of composite objects made with complex geometry. The first two structures are artificial [see Figs. 1(a) and 1(b)]. The third structure [Fig. 1(c)] is the union of a square lattice and the "infinite cluster" obtained in gradient percolation.[8] This is a well-known model geometry for the diffusion front of a soldering process. In each case, the total number of modes is identical since the number of particles is conserved through division into subsystems. Herein, we apply standard numerical methods to compute the whole set of eigenstates of the three systems shown in Fig. 1.

As depicted in Fig. 1(a), the "comb" structure consists of a 2D square lattice crystallite linked to one-dimensional chains containing approximately the same total number of particles. In Fig. 2 we show the DOS for this mixed system as a function of the reduced frequency $\omega = \omega_{\text{real}}(m/k)^{1/2}$. Note that the maximum frequency in 2D is given by $\omega_{\text{real}}^2 = 8k/m$. Also shown in Fig. 2 are the individual spectra obtained for the rectangular 2D system and the 1D chains.
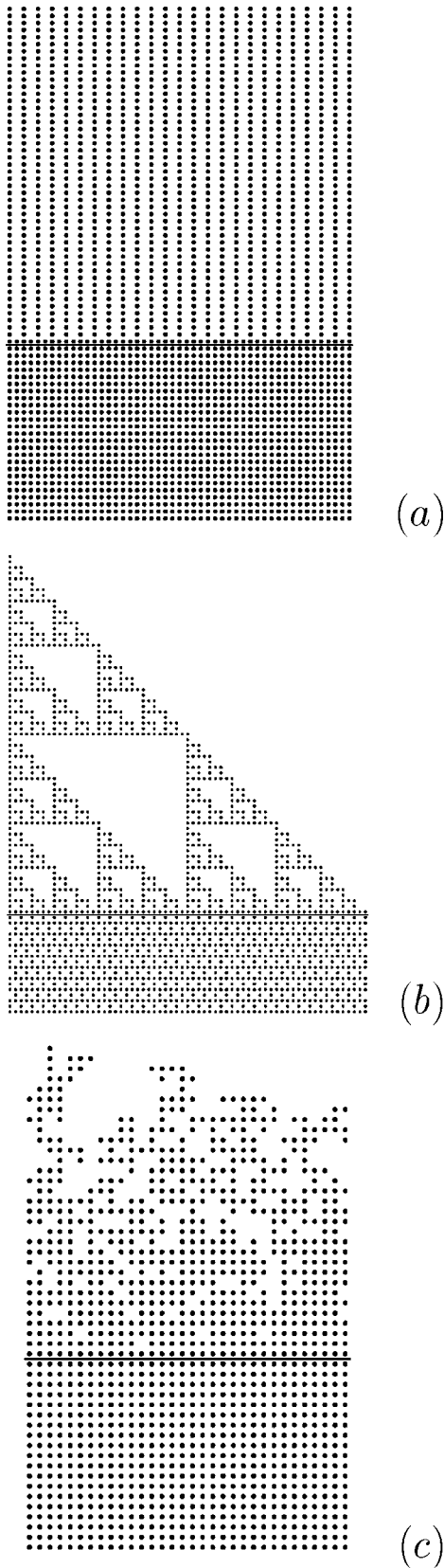
(a)

(b)

(c)

FIG. 1. The three composite systems under study: (a) the ''comb'' lattice is a combination of 1D chains linked to a 2D square lattice; (b) a standard 2D lattice interacting with a Sierpinski triangle; and (c) a square lattice terminated by the model geometry of a soldering. The external frontier is a fractal diffusion front.
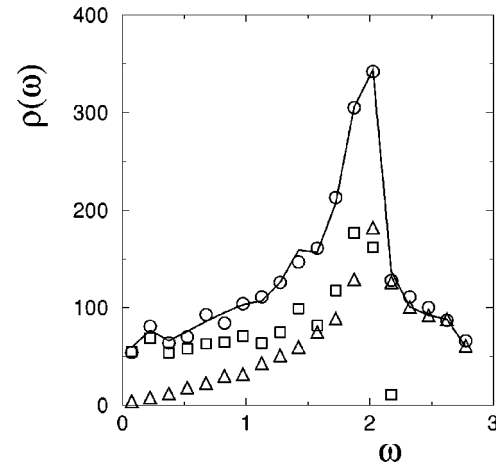

FIG. 2. Differential DOS $\rho(\omega)$ against the reduced frequency ω for the system shown in Fig. 1(a). The numerical values for the DOS are the number of states with reduced eigenfrequency between $\omega - \Delta\omega$ and $\omega + \Delta\omega$ with $\Delta\omega = 0.075$. The squares and triangles correspond to the DOS of the 1D chains and the 2D square lattice, respectively. Both spectra obey the corresponding 1D and 2D Debye laws at low frequencies. The circles represent the real spectrum of the interacting comb and the solid line corresponds to the sum of the components spectra. They are almost identical over the entire frequency range.

As expected, they are quite different; low frequency segments of both obeying the 1D and 2D Debye laws, respectively. The solid line in Fig. 2 represents the spectrum obtained by adding the two individual spectra. One clearly observes that the spectrum of the composite is nearly identical to the sum of the components spectra over the whole frequency range.

The second artificial system under consideration is a 2D crystal linked mechanically to a mass fractal [see Fig. 1(b)]. The vibrations in random or deterministic fractal lattices are known as *fractons* and are generally localized.[2–4] In addition, the low-frequency DOS for fractons is very different from the DOS of a regular lattice. The adequate exponent to be used in the Debye relation for fractons is called the spectral or fracton dimension whose value is often close to 4/3.[4] Due to the strong fluctuations observed in the differential DOS, we choose to express the numerical results for the system described in Fig. 1(b) in terms of its integrated DOS $I(\omega)$. This quantity measures the number of states with frequency smaller than ω. As shown in Fig. 3, the integrated DOS of the composite system is identical to the sum of the integrated DOS of the two noninteracting components.

The third structure under investigation is a geometrical paradigm often used to represent the stochastic process of diffusive penetration of a single crystal into an empty medium [see Fig. 1(c)]. The frontier of the irregular zone has been previously established to approximate a diffusion front with fractal dimension 7/4.[8] This system is then a surface fractal. The vibrations on surface fractals are called *fractinos*. For a given surface fractal, there exist localized and delocalized fractinos.[6] The differential DOS for both components and for the composite system displayed in Fig. 1(c) are shown in Fig. 4. The low-frequency spectrum for the square
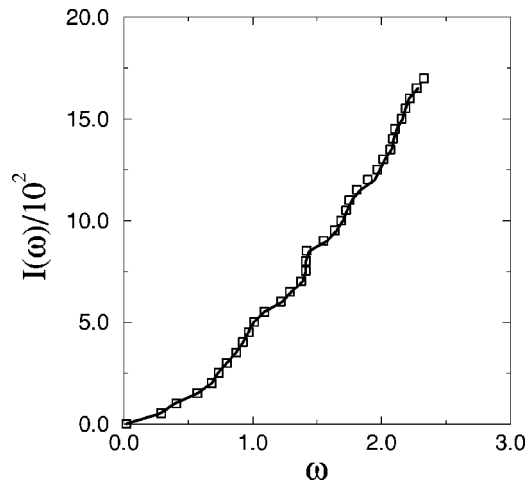
FIG. 3. The integrated DOS $I(\omega)=\int_0^\omega \rho(\omega')d\omega'$ against the frequency ω for the composite system presented in Fig. 1(b). The squares correspond to the spectrum of the global system and the solid line to the sum of the spectra of the two parts.

lattice obeys its corresponding Debye law. The spectrum of the irregular component is markedly different from that of the square lattice. Nevertheless, the additivity hypothesis is confirmed once again as shown in Fig. 4. In order to determine if the additivity of the DOS in this random system is generally valid, we performed additional simulations with 100 realizations of the composite geometry. Our results indicate that this property is self-averaging, i.e., the average DOS of this structure does not depend on the special set of configurations utilized and does not present relevant fluctuations from set to set.

Finally, nonstoichiometric lacunary systems of the type shown in Fig. 5 have also been investigated. In contrast with the previous systems presented in Fig. 1, such materials can be considered homogeneous above a certain characteristic
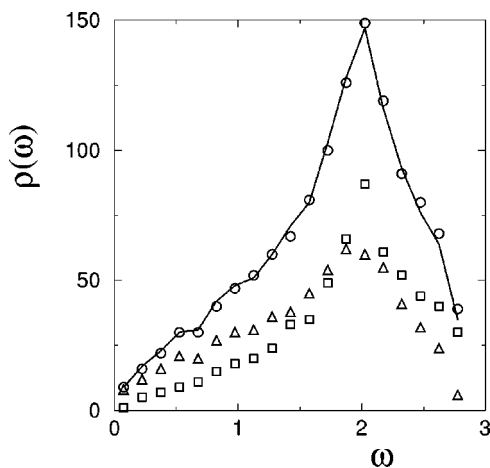

FIG. 4. DOS for the system shown in Fig. 1(c). The triangles correspond to the spectrum of the irregular part and the squares to the spectrum of the square lattice. The circles correspond to the spectrum of the whole system and the solid line represents the sum of the components spectra. The two spectra are identical over all frequency ranges.
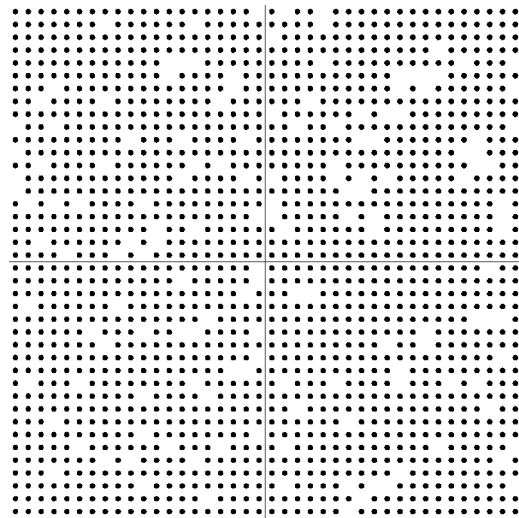

FIG. 5. Typical realization of a nonstoichiometric lattice with 10% of vacancies distributed randomly. The composite system is defined on a 40×40 square lattice. Vacancies are represented by empty sites. The system is divided into four components which are separated by the solid lines.

length ξ taken to be the average distance between defects. This length can be estimated as $\xi\approx aC^{-1/d}$, where C is the impurity (or vacancy) concentration and a is the lattice spacing. For example, ξ is of the order of tens of lattice spacing for a value of $C\approx1\%$. Qualitatively, one can then consider that a non-stoichiometric system is a pavement or juxaposition of cells of mesoscopic sizes with irregular geometry. The irregularity of the geometry arises from the compositional disorder itself.

The 2D nonstoichiometric solids studied here are generated by randomly allocating vacancies over a square lattice. Figure 5 shows a typical realization of the nonstoichiometric system generated for a concentration value of $C=0.1$. Eigenfrequencies are computed for the composite system and for the four separate components. The additivity property is also observed in this case (see Fig. 6). In order to provide a critical evaluation for the additivity hypothesis, we compute the more demanding quantity, $R=\rho(\omega)_{\text{total}}/\Sigma_{\text{parts}}\rho(\omega)_{\text{part}}$, where $\rho(\omega)_{\text{total}}$ is the differential DOS of the interacting system and $\Sigma_{\text{parts}}\rho(\omega)_{\text{part}}$ is the sum of the DOS of the components. The inset of Fig. 6 shows that, although the ratio R is not strictly constant over the entire range of frequencies ω, the value of $R\approx1$ still represents a fairly good approximation. The more pronounced discrepancy at low frequencies is due to the long wavelength states which are not present in the DOS of the components. In addition, we have performed simulations with 100 realizations of 40×40 nonstoichiometric lattices generated for two different values of the vacancy concentration, $C=0.1$ and 0.2. As expected, the results show that the sum of the separate spectra and the spectrum of the interacting ensemble are practically identical.

The additivity of the DOS in all composite systems studied here can be explained in terms of an effective weakness of the coupling among components. Although physically linked to each other, these parts interact at their frontier only. The additivity result, however, remains somewhat surprising
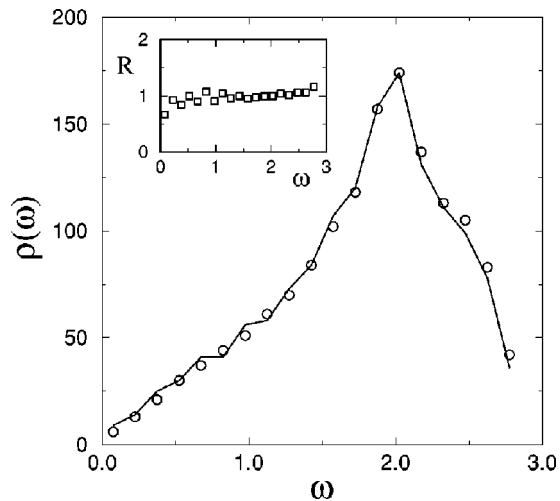
FIG. 6. DOS for the system shown in Fig. 5. The circles correspond to the spectrum of the whole system and the solid line represents the sum of the components spectra. The inset shows the dependence of the ratio R on the frequency ω. R is defined as the ratio between the differential DOS of the interacting system and the sum of the DOS of the components.

since mesoscopic tiles have a relatively large number of frontier sites. The additivity effect has a second cause: the eigenfrequencies in the different mesoscopic parts have a few, if any, resonant states. In the cases presented here the components interact only through their frontier and are out of resonance.

Another important contribution for additivity is the partial localization of the vibrational states. If present, such an effect obviously decreases the effective interaction between components. This is the case for vibrations of mass or surface fractals.[4,7] Also, vibrations in 2D disordered systems are always localized in the thermodynamic limit.[9–11] The localization of a state in the small scale structure of these solids is not affected by the occurrence of long wavelength modes when the different components are connected. Only the localization of the states which are at the boundary of the two objects will be significantly modified.

In summary, it has been shown that, within good approximation, the vibrational DOS of selected composite solids are equal to the sum of the calculated DOS of the different mesoscopic parts of the system. As these examples represent extreme cases in terms of geometry, this conclusion should be generally valid. For low frequencies, this observation confirms the recent theoretical prediction regarding spectral partition in infinite graphs[5]. This corresponds to the cases shown in Figs. 1(a) and 1(b). The additive property observed in the present study is not limited to low frequencies, but extends over the entire frequency range. Moreover, it also applies to systems where the concept of spectral dimension is meaningless [e.g., Figs. 1(c) and 4]. These results illustrate that the computational complexity of the DOS of a macroscopic solid can be dramatically reduced to the calculation of the DOS of mesoscopic systems. Finally, it should be recalled that, although not exact, some correspondence should exist between the vibrational DOS and the electronic DOS in a single orbital tight-binding approximation. We are currently investigating if the additivity hypothesis also applies for the tight-binding DOS of the same systems studied here as well as for the case of 3D nonstoichiometric structures.

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