Elastic behavior of materials with multifractal structures

Alexander S. Balankin*
Instituto Tecnologico y de Estudios Superiores de Monterrey, Campus Estado de México, 52926 Mexico
(Received 31 May 1995; revised manuscript received 14 August 1995)

A type of elasticity of random (multifractal) structures is suggested. Two phenomenological laws of reversible deformations of an elastic multifractal are postulated. The closed system of constitutive equations is obtained on the basis of these laws. Some classical formulas are derived as special cases of developed theory. The theoretical results are discussed with respect to available experimental data.

In the past ten years the theory of materials with random (fractal or multifractal) microstructure has become an attractive topic in mechanics and physics of solids.[1–6] Fractal geometry, developed by Mandelbrot,[7] allows the description of random structures which are more complex than Euclidean shapes. Statistical properties of a multifractal structure are characterized by the spectrum of generalized dimensions[6] $d_q=\lim_{r\to L_0}\{I_q(r)/\ln r\}$, $I_q=(q-1)^{-1}\ln\Sigma_{i=1}^{N}P_i^q(r)$, where I_q is the generalized entropy; $P_i(r)$ is the probability that a point of multifractal structure lies in box a i of covering network with boxes of size equal r; the index q ranges from $-\infty$ to ∞ and dimensions satisfy the general inequality $d_{q_1}\leqslant d_{q_2}$ for $q_2<q_1$; the equality being obtained in the case of monofractal structure.[6] The generalized dimension $d_{q=0}$ is equal to the metric (fractal) dimension evaluated by means of box-counting algorithm $d_0=d_F$, which for multifractal structure is greater than its topological dimension d_T, but smaller than, or equal to, the topological dimension d of the enveloping Euclidean space, i.e., $d_T<d_F\leqslant d$; the generalized dimension of order $q=1$ is equal to the information dimension, $d_I=d_1$, associated with Shannon entropy, and the dimension $d_{q=2}$ is equal to the correlation integral exponent $d_C=d_2$, also called correlation dimension. Aerogels, colloidal aggregates, polymers, some types of composite materials, porous media, etc. have a multifractal structure in a wide range of spatial scales $L_0<L<L_M$,[6–9] where L_0 is the microscopic cutoff (the minimal size of particles, components, pores, blobs, etc.) and L_M is the correlation length of self-similarity.

In practice or modeling elastic behavior of materials with multifractal microstructure it is broadly used three different types of elasticity:[1–5,10–13] (1) elasticity of energetic nature (elastic continuum, crystals, polycrystals, etc.); (2) entropy elasticity (elastomers); and (3) springlike elasticity (long polymer chains, foams, some structural composites, etc.). Theories of these types of elastic behavior are based on different phenomenological laws (experimental facts) and lead to different systems of constitutive equations.

The classical theory of elasticity is based on two experimentally established facts:[14] (1) the Hooke's law, according to which the strain ε_{ij} is proportional to the applied stress σ_{ij}, and (2) the Poisson's effect of lateral deformations (transverse strains) without corresponding stresses. In case of uniaxial extension (compression) of an elastically isotropic solid these facts may be represented in the form

$$(1)\ \ \sigma_{11}=E\varepsilon_{11};\quad (2)\ \ \varepsilon_{22}=\varepsilon_{33}=-\nu\varepsilon_{11},\quad \sigma_{22}=\sigma_{33}=0, \tag{1}$$

where E is the Young modulus, and ν is the Poisson's ratio. In practice, instead of these two postulates, the generalized form of Hooke's law is commonly used.[14] According to this law Poisson's effect is a consequence of the symmetry of the stress and strains tensors.

The basic postulates of the classical theory of rubber elasticity are[15] (1) the assumption that the chains of polymer network obey Gaussian statistics, and (2) the assumption of the incompressibility of elastomers. Under these assumptions it was derived dependence of stress F_1 (per unit cross-section area of sample in the initial state) versus relative strain λ_1 in direction of applied force:

$$(1)\ \ F_1=\frac{E}{3}(\lambda_1-\lambda_1^{-2});$$

$$(2)\ \ \lambda_2=\lambda_3=\lambda_1^{-0.5}\ \ (\lambda_1\lambda_2\lambda_3\equiv 1), \tag{2}$$

where $\lambda_i=L^i(F_1)/L_0^i$ (L_0^i and L^i are the sample dimensions in the i direction before and after application of the stress, respectively).

The springlike elasticity[8] is based on the empirical relation between relative strain λ_1 and applied force

$$F=E(\lambda_1-1), \tag{3}$$

which is valid for springs, long polymer chains, elastic foams, etc. The mechanism of lateral deformations for a material which exhibits springlike elasticity, for example an elastic foam,[16] differs from those for an elastic continuum (Poisson's effect) and for elastomers (incompressibility) and has a pure geometrical nature.[8]

In this article we suggest a more general type of reversible deformations of elastic random structures, that is the multifractal elasticity. It will be shown that two basic types of elasticity, e.g., the entropic elasticity (rubberlike and superelasticity) and the springlike elasticity may be interpreted as special cases of the multifractal elasticity. A possible generalization for developed theory is also briefly discussed.

In developing theory of multifractal elasticity we also start with two postulates (laws).

(1) When the external force F is applied to an elastic isotropic multifractal object, deformation occurs mostly on the length scale beyond certain characteristic length, which depends on F. Thus the presence of an external stress leads to the appearance of the unique characteristic scaling length L_F.

The initial morphology of multifractal can either be characterized by one or more length scale parameters L_i, or not to have them at all. In the first case the L_F may have physical significance such as the characteristic dimension of blobs in a polymer, characteristic size of cells, or mean distance between inclusions in a composite material, characteristic radius of correlations in a random network and aerogel, etc.[17] As it follows from the law postulated above, only one of the set of scale parameters L_i (or their invariant combination) is dependent on the external forces. If the initial multifractal structure has no scale parameters, then the physical meaning of L_F is the characteristic length above which deformations occur. It should be remarked that the idea that the presence of an external stress introduces a length scale to the problem of elastic deformations of random structures was invoked in[18] to study elastic macromolecules.

If this law is valid, from the second law of thermodynamics it follows that the force obeys relation

$$F=\left(\frac{\partial\Phi}{\partial L_F}\right)_T=\left(\frac{\partial U}{\partial L_F}\right)_T-T\left(\frac{\partial S}{\partial L_F}\right)_T. \tag{4}$$

The first term on the right of Eq. (4) is evidently the energy component of internal forces, and the second term is the entropic component.

It is well known, that in the zeroth long-wave approximation, at low frequencies, the difference between models of the media with microstructure disappears.[19] Therefore, to develop a static theory of elasticity of multifractals, which is considered below, it is possible (and sufficient) to formulate the first law in terms of relative deformations with scaling parameter $\lambda_F=L_{F_1}/L_{F_2}$, where L_{F_1} and L_{F_2} are the values of scaling length which correspond to $F=F_1$ and $F=F_2$, respectively. Notice, however, that instead of nonlocal theories of elasticity considered in Ref. 19, all of which in zeroth long-wave approximation are equivalent to classical model of elastic continuum, the zeroth long-wave approximation in our theory differs from the model of elastic continuum, because of distinctions in the basic laws.

(2) The information and correlation dimensions are not changed during reversible deformations of an elastic multifractal structure.

It is clear that elastic, i.e., homeomorphic (by definition[20]) deformations do not change the metric (fractal) dimension of deformed multifractal. Moreover, there are no changes in d_F, d_I, and d_C after an affine transformation (deformation) of multifractal structure,[21] while the limiting values $d_{q=-\infty}$ and $d_{q=\infty}$ of generalized dimension spectrum after an affine transformation generally differs from the initial ones. Hence the second postulate is valid at least in the case of affine elastic deformations. However, this postulate seems to be valid for any reversible (i.e., homeomorphic) deformation of a multifractal structure, and thus it is more general than the assumption[15,18] of affinity of elastic deformations.

The mass density ρ of a material with multifractal microstructure generally depends on the length scale L.[22] Dimensional analysis[23] implies the following general functional relation:

$$\rho=\rho_0\Psi\left(\frac{\xi_c}{L},\frac{L_F}{L}\right)=\rho_0\Psi(\lambda_c,\lambda_f), \tag{5}$$

where Ψ is a dimensionless function of its dimensionless arguments $\lambda_c=\xi_c/L$ and $\lambda_f=L_F/L$; ξ_c is the correlation length of multifractal (micro)structure. It is well known that within a bounded interval

$$L_F\ll L\ll\xi_c \tag{6}$$

the mass density of a multifractal structure $\rho(L)$ obeys a power law behavior.[22] According to (6) the second argument of dimensionless function $\Psi(\lambda_c,\lambda_F)$ is small ($\lambda_f\ll 1$), and at the asymptotic self-similar state we can apply to the function Ψ a scaling (incomplete self-similarity) representation,[23,24] so that

$$\rho=\rho_0\lambda_f^{-\alpha}\psi(\lambda_c),\quad \alpha=d-d_F, \tag{7}$$

where d_F is the fractal (metric, box-counting) dimension of the multifractal and ψ is a dimensionless function of λ_c (see also Ref. 25). Notice that ρ possess scaling behavior (7) in the initial state of multifractal structure as well as after any deformation of this structure, but λ_f, λ_c, and d_F, generally speaking, may be different before and after deformation.

However, according to the first postulated law, only the characteristic length L_F changes during the elastic (reversible) deformation of elastically isotropic multifractal; so that ξ_c must be constant. Furthermore, elastic deformations are homeomorphic by definition and thus do not affect the metric dimension d_F of deformed multifractal, so that the scaling exponent α is also constant.

By this means, the mass density of an elastic multifractal structure after its reversible (homeomorphic) deformations caused by different external forces F_1 and F_2 is equal to $\rho(F_1)=\rho_0\lambda_{f1}\psi(\lambda_c)$ and $\rho(F_2)=\rho_0\lambda_{f2}\psi(\lambda_c)$, respectively. Hence the ratio $\rho(F_1)/\rho(F_2)$ is not dependent on neither the variable length scale L nor the correlation length ξ_c and scales as

$$\frac{\rho(F_1)}{\rho(F_2)}=\left(\frac{\lambda_{f1}}{\lambda_{f2}}\right)^{-\alpha}=\left(\frac{L_{F_1}}{L_{F_2}}\right)^{-\alpha}_{T=\text{const}}=\lambda_F^{-\alpha},\quad \alpha=d-d_F, \tag{8}$$

i.e., the relation governing the change in the mass density ρ of elastically deformed multifractal[26] is similar to the relation that governs the change in the mass density because of the geometric changes in the dimensionalities of (multi)fractal structure [within the interval (6)]. We emphasize that this fact is a direct consequence of the first postulated law and the homeomorphism of reversible deformations.

It immediately follows from Eq. (8) that under uniaxial tension (compression) the change in the dimensionality of multifractal structure in the direction of external force F_x, which is $\lambda_x=L_x/l_x$, is accompanied by changes in the lateral dimensions of a deformed multifractal: $\lambda_i=L_i/l_i$, where $i=1,2,\ldots,d-1$. The lateral deformations λ_i are related to $\lambda_x=\lambda_F$ as

$$\lambda_i=\lambda_\perp=\lambda_x^{-\nu_F}=\lambda_F^{-\nu_F},\quad i=2,3,\ldots,d, \tag{9}$$

where ν_F is the transverse deformation exponent. Note that this exponent coincides with the Poisson's ratio $\nu=-\sqrt{(\lambda_i^2-1)/(\lambda_j^2-1)}$ only in the limiting case of infinitesimally small strains $\varepsilon=|\lambda_i^2-1|^{1/2}\ll 1$.

Substituting (9) into Eq. (8) we obtain $\alpha=1-(d-1)\nu_F$, so that

$$\nu_F=-\frac{\ln\lambda_\perp}{\ln\lambda_F}=\frac{d_F}{d-1}-1. \tag{10}$$

Therefore, if the first law is valid, the transverse deformation exponent ν_F of an elastically isotropic multifractal is determined by its metric dimension.

At the first glance, it is surprising that lateral deformations are independent of the detailed geometry of deformed multifractal structure, whose transverse deformation exponent is determined only by its metric dimension. Notice, however, that similar phenomena (power law distribution of stresses and strains with exponents, which are the functions of Poisson's ratio) are common within singular problems of the classical elasticity theory.[27]

Notice that Eqs. (9) and (10) are satisfied in the general case of n-dimensional deformation of a multifractal structure in d space. For example, for biaxial deformation in a three-dimensional space we have $\lambda_z=\lambda_F^{-\nu_F}$, $\lambda_F=(\lambda_x\lambda_y)^{1/(1-\nu_F)}$, where ν_F and d_F are still related by the Eq. (7), which is also valid for triaxial deformation, when $\lambda_F=(\lambda_1\lambda_2\lambda_3)^{1/\alpha}$ with $\alpha=3-d_F=1-2\nu_F$.

It must be emphasized that Eqs. (8) and (10) are not valid for materials obeying Poisson's effect (1) or, that is equivalent, the generalized Hooke's law.[20] For such materials the correlation length ξ_c is equal to the sample size in direction of applied external force, i.e., $\xi_c\equiv L_x$; so that ξ_c and

$$\lambda_C=\frac{\lambda_{c1}}{\lambda_{c2}}=\frac{\xi_{c1}}{\xi_{c2}} \tag{11}$$

must change after deformation. At the same time, for an elastic continuum $\lambda_f=0$ and $\lambda_F\equiv 1$, while for regular elastic lattice $\lambda_f=a/a_0$ (a and a_0 are the interatomic distance before and after deformation, respectively) and $\lambda_F=\lambda_C$. Hence for materials obeying Poisson's effect our first postulate is not valid.

Furthermore, the metric dimension of an elastic continuum as well as a regular elastic lattice [$d_F\equiv d_q$ for all q (Ref. 6)], which can be considered as a limiting case for multifractal structures, is equal to the topological dimension of structure d_T (Ref. 6); so that[28]

$$d_F=\begin{cases} d \\ d-1, \end{cases}\quad\text{and}\quad \alpha=\begin{cases} 0, & d_T=d, \\ 1, & d_T=d-1. \end{cases} \tag{12}$$

Now it is easy to understand that in the case of a Euclidean elastic structure obeying Eq. (1), the dimensionless function $\psi_P(\lambda_C)=\psi(\lambda_{c1})/\psi(\lambda_{c2})$ scales (in the limit of infinitely small strains) as

$$\psi_P=\left(\frac{\xi_{c1}}{\xi_{c2}}\right)^{-\beta}\equiv\left(\frac{L_x(F_1)}{L_x(F_2)}\right)^{-\beta}=\lambda_C^{-\beta}, \tag{13}$$

where $\beta=1-(d_T-1)\nu$. The second relation in (1) may be derived by substitution of Eqs. (13) and (9) with $d_T=d$ in the general relation (7).

Therefore, in the general case of an elastic multifractal structure, which also possesses a conventional Poisson's effect, the scaling relation (8) can be generalized into

$$\frac{\rho(F_1)}{\rho(F_2)}=\lambda_F^{-(d-d_F)}\lambda_C^{(d_T-1)\nu-1}. \tag{14}$$

It should be emphasized that relation (14) is valid only in the limit of infinitely small strains $\varepsilon_{ii}=\sqrt{|\lambda_i^2-1|}\ll 1$. Now, if multifractal structure possess Poisson's effect with $\lambda_C=\lambda_F$, the transverse deformation exponent is equal to

$$\nu_F=\nu-\frac{d-d_F}{d-1}. \tag{15}$$

Below we will consider only elastic multifractals reversible behavior which obeys two laws postulated above. The lateral deformations of such a structure have pure geometric nature ($\lambda_C\equiv 1$) and are governed by its metric dimension [see Eq. (10)].

Unfortunately, no case of accessible experimental data for an elastic behavior of multifractal structures has come to our notice. However, we can verify relation (10) by using available data for elastic monofractals.

For example, the fractal dimension of aerogel SiO_2 was measured by the small-angle neutron scattering method and by the molecular adsorption technique.[6,29] It was found that aerogel has the *monofractal* structure (i.e., $d_F=d_I=d_C$), which is characterized by the fractal dimension $d_F=2.3\pm 0.1$,[29] so that according to Eq. (10) we expect that $\nu_F=0.15\pm 0.05$. This value is in reasonable agreement with the experimental data $\nu=0.12\pm 0.08$,[27] which were obtained in the studies of longitudinal and transverse elastic waves propagation.

The metric dimension of monofractal strongly twisted polymer filament equals the fractal dimension of self-avoiding random walk[6] and thus is equal to $d_{s-a}=2$.[6,30] According to Eq. (10) we expect $\nu_F=0$, that agrees with experiments.[31] On the other hand, rubberlike polymers are characterized by $d_F=3$ (Refs. 32 and 33) and thus obey property of incompressibility,[15,31] i.e., $\nu_F=0.5$.

Looking back at the two proposed laws of reversible deformations for multifractal structures, we see that it is necessary to know the changes in entropy and internal energy as functions of deformations, before the closed system of constitutive equations can be obtained.

Using the definitions of information and correlation dimensions[6] and scaling properties of a multifractal,[34] with relations (4), (8) valid, it is readily shown that the changes in thermodynamic entropy $\Delta S(\lambda_i)$ and internal energy $U(\lambda_i)$ due to the reversible deformation of an elastic multifractal (in d-dimensional space) may be represented as

$$\Delta S = -C_2\left(\sum_{i=1}^{d} \lambda_i^{d_I} - d\right), \tag{16}$$

and

$$\Delta U = -C_1(\lambda_F^{\alpha_C} - 1), \quad \alpha_C = d - d_C, \tag{17}$$

respectively. Here C_1 and C_2 are constants (notice that parameters C_1 and C_2 also can be determined for any detailed model of structure[15,31]).

Substituting (16) and (17) into (4), and using relations (9) and (10) we can derive relationships between external force F_i and relative deformations λ_j for an elastic multifractal structure. For example, in the case of uniaxial deformation we obtain

$$F_1 = C_2\left\{ d_I\lambda_1^{d_I-1} - d_I[d_F-(d-1)]\lambda_1^{-d_I[d_F/(d-1)-1]-1} - \frac{C_1}{C_2}(d-d_C)\lambda_1^{d-d_C-1}\right\}. \tag{18}$$

According to the obvious condition $F(\lambda_i = 1) = 0$, it follows from Eq. (18) that

$$\frac{C_1}{C_2} = d_I\frac{d-d_F}{d-d_C} \leqslant d_F. \tag{19}$$

Hence the behavior patterns of elastic multifractals (for which two postulated laws are valid) can be determined by the metric, information, and correlation dimensions.

Moreover, it is easy to see that in the limit of infinitely small strains Eqs. (18) and (19) can be generalized by using Eqs. (14) and (15).

In the case of monofractal structure all generalized dimensions equal the metric (fractal) dimension, i.e., $d_q \equiv d_F = d_I = d_C$,[6] and Eq. (19) results in the relation

$$\frac{C_1}{C_2} = d_F, \tag{20}$$

so that Eq. (18) may be rewritten in the form

$$F_1 = C_1\{\lambda_1^{d_F-1} - [d_F-(d-1)]\lambda_1^{-\nu_F d_F-1} - (d-d_F)\lambda_1^{\alpha-1}\}. \tag{21}$$

The stress σ_{11} is related to the force $F_1(\lambda_1)$ by obvious equation $\sigma_{11} = F_1\lambda_1^{1-\alpha}$, which by using Eqs. (21) and (10) may be written in the form

$$\sigma_{11} = \frac{E}{1+6\nu_F+4\nu_F^3}[(\lambda_1^{1+4\nu_F}-1) - 2\nu_F(\lambda_1^{-1-2\nu_F^2}-1)], \tag{22}$$

where $E = (\partial\sigma_{11}/\partial\varepsilon_{11})_T$ is the Young modulus.

It can be clearly seen that within the limit of infinitesimally small strains, $|\varepsilon_{11}| = \sqrt{|\lambda_1^2 - 1|} \ll 1$, Eq. (22) leads to its classic counterpart Eq. (1). Thus, for monofractal structure we have $C_1 = 2(1+\nu_F)C_2 = (1+6\nu_F+4\nu_F^3)E$.

Similarly, we can derive relations $\sigma_{ij}(\lambda_k)$ for n-axial deformations of an elastically isotropic monofractal in d space. Pure shear is essentially a biaxial loading under the stresses σ_{11} and σ_{22} such that there is no change in the length along a second direction, i.e., $\lambda_2 = 1$.

The relationships between elastic moduli, i.e., Young's modulus E, shear modulus G, and bulk modulus B, and Lamé coefficients λ, μ of elastically isotropic monofractal, which are derived by analogy to the derivation of corresponding relationships in the classical elasticity theory,[14] are as follows:

$$G = \frac{E(d-1)}{2d_F}, \quad B = \frac{E}{d(d-d_F)}, \quad B = \lambda + \frac{2}{d}\mu. \tag{23}$$

Notice that these relations differ from those which were conjectured for elastic fractals by Bergman and Kantor.[1] At the same time, substituting Eq. (10) in the Eqs. (23) we obtain expressions that for $d = 2$ and $d = 3$ are identical to those for two- and three-dimensional elastically isotropic continuum.

Classical formulas for rubberlike elasticity (2) may be derived within a framework of Eqs. (8)–(10) and (18), (19) for multifractals obeying[35]

$$d_F = d = 3, \quad \text{and} \quad d_I = 2.$$

However, calculations based on Eq. (2) with the value of E adjusted by fitting are in reasonable agreement with the experiments only in the range of relatively small strains ($\lambda_i < 1.2$).[15] Traditionally, the refinement of the relation (2) is made by the phenomenological modifications of entropic theory, or by using empirical models for the elastic potential.[15,31] At the same time, elastomers are known to have fractal or multifractal microstructure.[10,13,36] Therefore, it is natural to describe the rubber elasticity of polymers by using the concept of multifractal elasticity.

Normally, the generalized dimensions of polymer networks swelled in a good solvent are within the range $2 < d_q \leqslant 3$.[32] Assuming in the first approximation that $d_F = d_I = d_C$ and substituting Eq. (10) into Eq. (21), we obtain a relationship between the nominal stress F_1 and the strain factor λ_1 in the following form:

$$F_1 = \frac{E}{1+6\nu_F+4\nu_F^3}\{\lambda_1^{1+2\nu_F} - 2\nu_F\lambda_1^{-1-2\nu_F(1+\nu_F)} - (1-2\nu_F)\lambda_1^{-2\nu_F}\}, \tag{24}$$

which was first derived in our work[4] by other means. Notice that behavior (24) differs from (2) even in the limit of incompressibly deformed material, when Eq. (24) reduces to the formula

$$F_2 = \frac{E}{4.5}(\lambda_1^2 - \lambda_1^{-2.5}), \tag{25}$$

obeying the experimental established asymptotic[15]

$$F_1 \propto \lambda_1^2, \quad \text{when} \quad \lambda_1 \gg 1.$$

It was shown in Refs. 4, 12, and 36 that calculations based on Eqs. (24) and (25) agree well with experimental data for rubbers without any adjustment of parameters (except E) right up to $\lambda_1 = 7$.

The nonlinear stress-elongation asymptotic for superelastic network[13]

$$\sigma_{11} \propto \lambda_1^{1/3} \tag{26}$$

is also a special case of constitutive equations (10), (18), and (19), which is valid for multifractals possessing $d_F + d_l - d = 1/3$, for example, $d_F = d = 2$, $d_l = 4/3$.

The basic relation for springlike elasticity (3) may be also derived from (18) and (19) in the case of multifractal structures for which $d_F = d_l = 2$.

Hence the proper regard for the real morphology allows an adequate description for the behavior of a reversible deformed material with fractal or multifractal microstructure. The examples considered in present work revealed that two laws of reversible deformations of a multifractal structure, which are postulated in the present work, are valid at least for some classes of materials with (multi)fractal (micro)-structure.

We hope this work will stimulate experimental research along this line. We expect that these investigations will support our concept of multifractal elasticity. If so, experimental data on elastic behavior can be used to estimate the metric, information, and correlation dimensions of the (multi)fractal microstructure.

The author is grateful to G. Cherepanov for useful discussions. The courtesy and support of the staff and faculty of Material Research Center at Monterrey University of Technology, and especially A. Bravo, are gratefully acknowledged.

*Formerly at Dzerzhinski Military Academy, Moscow, Russia. Electronic address: balankin@servdgi.cem.itesm.mx, FAX: (525) 326-57-89.

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