NEGATIVE POISSON'S RATIO
AND PERCOLATING STRUCTURES

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Abstract: Elastic properties of non-uniform, two-component systems are studied in frames of a model of percolation on a simple cubic lattice. It is shown that as the ratio of the bulk moduli of the components tends to zero, $\kappa = K_s/K_h \to 0$ (where s,h denote the softer and harder phase, respectively), the Poisson's ratio of the system tends to 0.2 at the percolation threshold of the harder phase, no matter what the values are of the Poisson's ratios of the components. A qualitatively new, collective mechanism leading to negative Poisson's ratio is suggested.

1. Introduction

The Poisson's ratio, being the negative ratio of the change of the transverse strain to the change of the longitudinal strain when the longitudinal stress is changed at fixed transverse stress, is one of the simplest characteristics of elastic properties of isotropic media. Typical materials exhibit positive Poisson's ratio, $\nu > 0$, *i.e.* expand (contract) transversely when stretched (pulled) longitudinally. For a long time is was believed that no isotropic material exists in nature for which ν can be less than zero [1]. Hence, various models exhibiting negative Poisson's ratio [2–4] were considered as mathematical curiosities rather than of any relation to reality and, hence, of no practical importance. After manufacturing foams of negative Poisson's ratio by Lakes [5], and finding negative Poisson's ratio of expanded polytetrafluoroethylene by Evans and coworkers [6] an increasing interest in studies of systems exhibiting such an unusual property has been observed [7–15]. Various potential applications of such systems [16, 17] encourage one to study new mechanisms which may lead to $\nu < 0$ [18, 19].

In this work we describe some results of theoretical studies of the Poisson's ratio of disordered structures composed of two phases of (strongly) different elastic properties. We

apply a renormalization group approach to a model of percolation on a hierarchical cubic lattice. (Although this approach was described in detail elsewhere [20], we present it briefly here, for completeness.) At the percolation threshold the Poisson's ratio, we obtain, is in perfect agreement with computer simulation results and a conjecture proposed by Arbabi and Sahimi [21]. Basing on this conjecture we postulate a new mechanism which should lead to negative Poisson's ratio for some materials, *e.g.* some polymers.

2. The model

Elastic properties of non-uniform systems of chaotic structure have been modelled in frames of percolating lattices [22–26]. In the simplest case one can consider lattices with two kinds of bonds, occupied and not occupied ones. At low densities of the occupied bonds they form clusters which are not connected to each other. When the concentration of the occupied bonds of the lattice increases, the clusters percolate at some critical concentration (the *percolation threshold*), p^*, and form a connected cluster of fractal structure [27]. Such systems can be studied theoretically by the renormalization group approach [28, 29] which consists in relating a set of characteristic properties of the system at a given length scale with the same set of the characteristic properties but corresponding to a larger length scale. Increasing the length scale in the following iterations one reaches the correlation length, above which the system can be thought of as a uniform one, and its properties do not change when the length scale is further increased.

Let us consider a simple cubic lattice whose elementary bonds of the length l are coloured with the probability distribution:

$$P^{(0)}(c) = p^{(0)}\delta(c - c_C^{(0)}) + (1 - p^{(0)})\delta(c - c_N^{(0)}),$$

where $0 \leq p^{(0)} \leq 1$ denotes the probability that a bond is connected (coloured by $c_C^{(0)}$) whereas $(1 - p^{(0)})$ is the probability that the bond is not connected (coloured by $c_N^{(0)}$). It is assumed that bonds of the same colour exhibit the same properties. The simplest iterational procedure consists in partitioning the lattice into super-cells of linear dimension bl, replacing them by effective elementary cells of the length bl, and rescaling the latter to the initial size l. Thus, the structure of the lattice remains unchanged after each iteration. The colours (*i.e.* the properties) of the elementary bonds do change, however, in general. To avoid long computations, necessary to determine complete characteristics of possible configurations of the super-cells, at the iteration n ($n = 0, 1, \ldots$), *i.e.* colours of the elementary bonds which replace them at the next iteration, $n + 1$, it is convenient to use an approximation which, for each direction α ($\alpha = x, y, x$), consists in dividing the set of all configurations of a super-cell into two disjoint subsets. The first subset, C_α, corresponds to these configurations of the bonds of the super-cell which connect a pair of these of the opposite boundaries (walls) of the super-cell which are perpendicular to the direction α. C_α The second subset, N_α, contains the remaining (not α-connected) configurations of the super-cell. Configurations corresponding to the subset C_α are replaced at the $n + 1$-th iteration by connected bonds, coloured as $c_C^{(n+1)}$ (assuming that x, y, z are equivalent we neglect here the index α), and those corresponding to N_α are replaced by disconnected bonds, coloured as $c_N^{(n+1)}$, in the directions specified by α, respectively. Thus, the probability distribution at the $n + 1$-th step reads:

$$P^{(n+1)}(c) = p^{(n+1)}\delta(c - c_C^{(n+1)}) + (1 - p^{(n+1)})\delta(c - c_N^{(n+1)}).$$

The probability that a configuration of a super-cell belongs to C_α is equal to the ratio of the number of the connected configurations of the super-cell to all configurations, $R[p^{(n)}]$, and this is just the probability, $p_\alpha^{(n+1)}$, that an effective bond at the $(n+1)$-th level is α-connected. By equivalence of x,y,z we neglect the index α and write:

$$p^{(n+1)} = R[p^{(n)}] \tag{1}$$

The above equation defines the transformation of the probability of a connected bond in the consecutive steps. This transformation has three fixed points $0, p^*, 1$, where 0 and 1 are stable and p^* is the unstable solution of (1).

If one knows the dependence of the effective properties of the system (colour of the bonds) in the subsequent step as a function of the effective properties (colour) in the preceding step:

$$c_I^{(n+1)} = f_I^{(n)}(c_C^{(n)}, c_N^{(n)}, p^{(n)}), \qquad I = C, N$$

then, taking into account the equation (1), the iterational procedure for finding the macroscopic properties of the system is complete. The macroscopic properties can be obtained as:

$$\lim_{n \to \infty} c_C^{(n)} = \lim_{n \to \infty} c_N^{(n)} = c_{macro}.$$

(In practice, the iterational procedure is finished when the properties of the lattice become independent of the number of the step n.)

In this work we used the cubic lattice and the function

$$R(p) = p^2(4 + 8p - 14p - 40p^3 + 16p^4 + 288p^5 - 655p^6 + 672p^7 - 376p^8 + 112p^9 - 14p^{10})$$

for calculating the probability of connected configurations. This function, implying the percolation threshold at $p_{crit} = .20846268\ldots$, was obtained in Ref. [30]. We should add here that this function has recently been applied by one of the authors to studies of conductance, dielectric properties, and elastic properties of composites, leading to good agreement with experimental results [31–33].

To calculate elastic properties of the percolating system we applied the well known Hashin-Shtrikman-Kröner formulae for the elastic moduli of perfectly disordered composite materials [34]. Within this approximation, the bulk modulus, K, and the shear modulus, μ, for the $(n+1)$-th iteration can be written as:

$$K_C^{(n+1)} = p^{(n)}K_C^{(n)} + (1 - p^{(n)})K_N^{(n)} - \frac{A_C^{(n)}D^{(n)}}{1 - p^{(n)}A_C^{(n)}(K_C^{(n)} - K_N^{(n)})}, \tag{2}$$

$$\mu_C^{(n+1)} = p^{(n)}\mu_C^{(n)} + (1 - p^{(n)})\mu_N^{(n)} - \frac{B_C^{(n)}M^{(n)}}{1 - p^{(n)}B_C^{(n)}(\mu_C^{(n)} - \mu_N^{(n)})}, \tag{3}$$

where

$$A_C^{(n)} = 3/(3K_C^{(n)} + 4\mu_C^{(n)}),$$
$$B_C^{(n)} = 6(K_C^{(n)} + 2\mu_C^{(n)})/(5\mu_C^{(n)}(3K_C^{(n)} + 4\mu_C^{(n)})),$$
$$D^{(n)} = p^{(n)}(1 - p^{(n)})(K_C^{(n)} - K_N^{(n)})^2,$$
$$M^{(n)} = p^{(n)}(1 - p^{(n)})(\mu_C^{(n)} - \mu_N^{(n)})^2.$$

The Poisson's ratio of the three-dimensional isotropic system is given by

$$\nu_C^{(n)} = \frac{3K_C^{(n)} - 2\mu_C^{(n)}}{6K_C^{(n)} + 2\mu_C^{(n)}}. \tag{4}$$

Equations for the non-connected configurations are obtained by replacing C by N, N by C, and p by $(1-p)$.

3. Results and discussion

In Figure 1 we show the Poisson's ratios obtained for very small ratios of the bulk moduli, $\kappa \equiv K_s/K_h$ (where the indices s, h denote the softer and the harder phase, respectively), and a few values of the Poisson's ratios of the composing phases as functions of the concentration of the harder phase. It can be seen that the concentration dependence of the Poisson's ratio can be non-monotonous in such a case (see, *e.g.*, the case when $\nu_s = 0$ and $\nu_h = -0.9$). It is also evident that the values of the Poisson's ratio of the system below the percolation threshold are determined mainly by the soft phase, and above the threshold – by the hard phase.

In Figure 2 we show the Poisson's ratios obtained for a few ratios of the bulk moduli and a few values of the Poisson's ratio of the composing phases as functions of the concentration of the harder phase. It can be seen that the extrema of the obtained curves become closer and closer when κ decreases. Extrapolating the positions of the extrema to $\kappa = 0$ one obtains $\nu_{crit} = 0.2$ at the percolation threshold, see Figure 2.
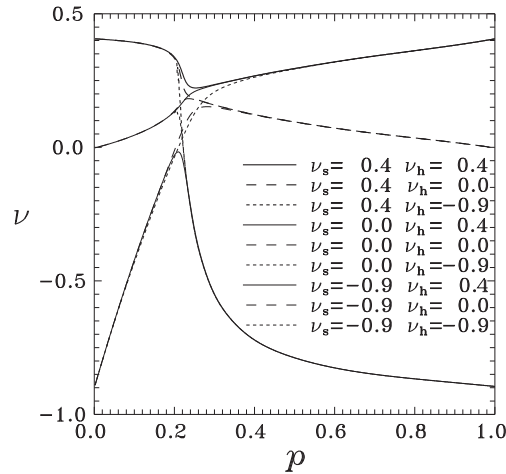

Figure 1. Poisson's ratios of the system composed of two percolating phases as functions of the concentration of the harder phase for various Poisson's ratios of the components for the ratio of the bulk moduli $\kappa = 10^{-5}$

The critical value $\nu_{crit} = 0.2$ was earlier obtained by direct numerical experiments [21, 25] for percolation of systems interacting by central and bending forces on simple cubic lattices. In the Ref. [21] a conjecture was proposed that at the percolation threshold one has

$$\mu/K = Z/8, \tag{5}$$

where Z is the coordination number of the lattice. The present calculations, which do not assume any specific form of the interactions, indicate that this is a general result for the cubic structure, *i.e.* does not depend on the elastic properties of the percolating phase. This suggests in turn that the Poisson's ratio at the percolation threshold is a universal characteristic of structure.
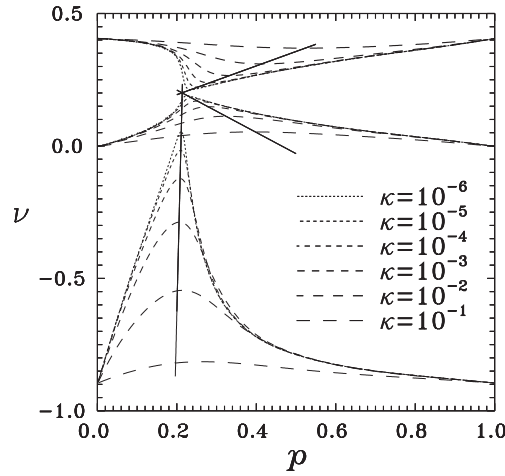
Figure 2. Poisson's ratios of the system composed of two percolating phases of various ratios of the bulk moduli, κ, as functions of the concentration of the harder phase for various Poisson's ratios of the components: $\nu_s = \nu_h = -0.9$ (thin lines), $\nu_s = \nu_h = 0.0$ (lines of the intermediate thickness), and $\nu_s = \nu_h = 0.4$ (thick lines). Extrapolating the extrema of the plotted curves one reaches ν_{crit} at the percolation threshold in each case

4. Cooperative mechanism which may lead to negative Poisson's ratios

Formula (5), which is consistent with our approximate theory, has an interesting and rather surprising consequence. Namely, it suggests that one can obtain negative Poisson's ratios near the percolation threshold for lattices of sufficiently high coordination number. The reasoning is as follows.

The Poisson's ratio for a d-dimensional isotropic body reads [35]:

$$\nu = \frac{dK - 2\mu}{(d-1)dK + 2\mu} = \frac{d - b}{(d-1)d + b} \tag{6}$$

where $b \equiv 2\mu/K$. The above equation implies that:

1. for $b > d$ one has $\nu < 0$, reaching its minimum value, $\nu_{min} = -1$, when $b/d \to \infty$,
2. at $b = d$ the Poisson's ratio equals zero, $\nu = 0$,
3. and when $b < d$ the Poisson's ratio is positive, reaching its maximum, $\nu_{min} = 1/(d-1)$, value when $b/d \to 0$.

Thus, if (5) holds true also when the number of bonds Z located at the vertices of the lattice is different from 6 (the case of the simple cubic lattice), the Poisson's ratio *at* the percolation threshold can be written as:

$$\nu = \frac{d - Z/4}{(d-1)d + Z/4}. \tag{7}$$

It follows from the above that ν should be positive when $Z < 4d$, zero when $Z = 4d$ (*e.g.* in the case of percolation on the f.c.c. lattice) and negative when $Z > 4d$.

It is worth to stress that, in contrast to some exactly soluble mechanical models of uniform structures which exhibit negative Poisson's ratios [2, 36] and which can be understood by considering few-particle clusters, the mechanism suggested above has its roots in a complex, collective behaviour of many particles forming non-uniform structures.

5. Conclusions

It has been shown that an approximate theoretical treatment of percolation on a cubic lattice exactly reproduces the Poisson's ratio obtained in computer simulations at the percolation threshold. This result may encourage one to use this approximation to describe various elastic properites of composites. It is worth noting that, recently, some critical indices have been calculated with high accuracy in frames of the present model [37].

Considerations presented in this paper suggest that at the percolation threshold the ratio of the shear modulus to the bulk modulus is a universal quantity which does not depend on the elastic properties of the percolating phase. It is well known, however, that this ratio depends on the coordination number of the lattice on which the percolation takes place [21]. Taking this into account, we conjecture that when the coordination number, Z, of the underlying lattice is more than four times larger than the dimensionality of the lattice, $Z > 4d$, the Poisson's ratio near the percolation threshold should be negative, no matter what the value is of the Poisson's ratio of the percolating phase.

Assuming that (5) is fulfilled also at the percolation threshold for random lattices with Z replaced by the *average* coordination number $\langle Z_i \rangle$ (where Z_i means the coordination number of the i-th lattice site), one can obtain negative Poisson's ratios by percolating systems of particles, *e.g.* polymer molecules, which can be bond to a sufficiently large number of neighbours, $\langle Z_i \rangle > 4d$.

Analytic studies and extensive computer simulations of various models in 2D and 3D are in progress to verify the above theoretical predictions.

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