

The elastic behavior of entropic “fisherman’s net”

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Abstract. A new formalism is used for a Monte Carlo determination of the elastic constants of a two-dimensional net of fixed connectivity. The net is composed of point-like atoms each of which is tethered to six neighbors by a bond limiting the distance between them to a certain maximal separation, but having zero energy at all smaller lengths. We measure the elastic constants for many values of the ratio γ between the maximal and actual extensions of the net. When the net is very stretched ($\gamma \sim 1$), a simple transformation maps the system into a triangular hard disks solid, and we show that the elastic properties of both systems coincide. We also show that the crossover to a Gaussian elastic behavior, expected for the non-stressed net, occurs when the net is looser ($\gamma \sim 3$).

PACS. 62.20.Dc Elasticity, elastic constants – 05.10.-a Computational methods in statistical physics and nonlinear dynamics – 65.50.+m Thermodynamic properties and entropies

Materials like rubber and gels are formed when polymers or monomers are cross-linked into macroscopically large networks. Due to the small energetic differences (of the order of kT) between the allowed microscopic configurations of these materials, their physics is primarily determined by entropy, rather than energy. This has been recognized long ago, and the peculiar physical properties of rubber and gels, in particular their great flexibility, are attributed to this microscopic feature. The classical theories of rubber elasticity, for instance, deal with Gaussian networks in which the internal elastic energy is completely ignored and the strands between cross-links are viewed as entropic springs [1]. These theories, however, do not explain well the elastic behavior of networks of certain types. Perhaps the most known unresolved problem in this field of research is the question of the critical elastic behavior of random systems near connectivity threshold. Most of the numerical works which aimed to investigate this issue during the last twenty years concerned the *energetic* elasticity [2]. Recent studies [3], however, suggested that close to the gel-point elasticity is dominated by its *entropic* component. A completely different aspect of entropic elasticity, which has been studied much less, is the behavior of highly connected networks, well above their connectivity threshold. The classical theories are inappropriate in this connectivity regime, since the strands between the junctions are very short and do not resemble Gaussian springs.

When the *boundary* of a thermodynamic system is *homogeneously* deformed, the distance between any two boundary points which prior to the deformation were sep-

arated by \vec{R} , becomes

$$r = [R_i R_j (\delta_{ij} + 2\eta_{ij})]^{1/2}, \quad (1)$$

where the subscripts denote Cartesian coordinates and summation over repeated indices is implied. The quantities η_{ij} are the components of the *Lagrangian strain tensor*, while δ_{ij} is the Kröner delta. The elastic behavior of the system is characterized by the *stress* tensor, σ_{ij} , and the tensor of *elastic constants*, C_{ijkl} , which are the coefficients of the free energy density expansion in the strain variables

$$f(\{\eta\}) = f(\{0\}) + \sigma_{ij}\eta_{ij} + \frac{1}{2}C_{ijkl}\eta_{ij}\eta_{kl} + \dots \quad (2)$$

Measuring the elastic constants is much more difficult in entropy-dominated systems than in energy-dominated ones. In the latter one needs to calculate energy variations around well-defined ground states. In the former, on the other hand, different microscopic configurations possess similar energies. Entropy in this case is essentially the (logarithm of the) number of allowed microscopic configurations. Measuring the variations of this quantity in response to external deformations applied on the system is usually very complicated. In order to simplify this task, and due to the fact that the exact energy details are quite irrelevant in entropy-dominated systems, the inter-atomic interactions in such systems are often modeled by “hard” potentials. Excluded-volume effects, for instance, can be modeled by the hard spheres repulsion, while chemical bonds can be replaced by inextensible (“tether”) bonds which limit the distance between the bonded monomers, but have zero energy at all permitted distances [4]. The energy of all the microscopic configurations in such models, which are

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called “athermal”, is the same, and their physics, therefore, is exclusively governed by entropy considerations. It is interesting to note that although athermal models have been investigated quite extensively in polymer and soft matter physics, the elastic properties of many of them are not well understood. Hard spheres systems, for instance, are studied for already more than 40 years [5]. They were, in fact, the first systems for which Metropolis *et al.* performed the first Monte Carlo (MC) simulations in 1953 [6]. The phase diagram of hard spheres, which is a function of a single parameter, their volume fraction, had been fully explored both in simulations and experiments [7]. Yet, despite the numerous works dedicated to *elasticity of hard spheres* [8], the accuracy of the values of their elastic constants still leaves much to be desired.

In the canonical ensemble, the elastic constants can be related to the mean-squared thermal fluctuations of the stress tensor components (just as the heat capacity is proportional to the mean-squared energy fluctuation). This relation, first expressed by Squire *et al.* [9], can be used for a Monte Carlo determination of the elastic constants. The method is known as the “fluctuation method”. The instantaneous stress, measured at a given microscopic configuration, is associated with the mean force (averaged over the entire volume) acting on the atoms [10,11]. The local forces originate from external potentials and interparticle interactions. In entropy-dominated systems, these forces are usually very small. They become extremely large only over very short time intervals when atoms come to the close vicinity of each other or when bonds are sufficiently stretched. Model with hard potentials can be regarded as the limiting case in which these time intervals vanish, while at the same time the instantaneous forces become infinitely large, keeping the rate of momentum exchange between atoms fixed. It is obvious that the stress in such systems must be related to the two-point probability densities of contact between spheres and occurrence of bond stretching, while the elastic constants (stress fluctuations) must be related to the corresponding four-point probability densities. Indeed, we have recently succeeded in formulating the exact relations. We obtained expressions enabling a direct measurement of the entropic contribution to the elastic constants, and demonstrated the accuracy and efficiency of the method using this formalism on three-dimensional hard spheres systems [12]. In this paper we apply this new formalism to measure, by means of MC simulations, the stress and elastic constants of topologically simple networks. We consider a “toy model” consisting of a two-dimensional (2D) network of atoms forming a triangular “fisherman’s net” (FN): atoms are point-like, *i.e.*, have no excluded volume, and each one of them is connected to six neighbors by a “tether” limiting the maximal distance between the atoms, but otherwise not exerting any force.

The FN is a highly connected network, whose physical behavior is entropy-dominated. Some of the elastic properties of the system have been studied by Boal *et al.* [13]. (Some extensions of that work can be found in Ref. [14].) In reference [13] fixed pressure ensemble has

been used, and the elastic moduli have been extracted from the fluctuations of the linear size of a rectangular cell. Our simulation applies to the fixed volume case, uses significantly larger systems and a wider range of stresses. We are also able to measure every elastic constant directly, without assuming any symmetry of the system. Whenever comparison is possible, our results are in agreement with reference [13].

The FN is six-fold symmetric when it is equally stretched along all the spatial directions. Its elastic properties in this reference state should be as of an isotropic system (see p. 35 (discussion on hexagonal systems) in Ref. [10]): Its stress tensor is diagonal with the elements $\sigma = \sigma_{xx} = \sigma_{yy} = -P$, where P is the *negative* external pressure (stretching) one needs to apply to the boundaries in order to balance the forces exerted by the net. Only four elastic constants of the net do not vanish: $C_{11} = C_{xxxx}$, $C_{22} = C_{yyyy}$, $C_{12} = C_{xxyy} = C_{yyxx}$ and $C_{44} = \frac{1}{4}(C_{xyxy} + C_{xyyx} + C_{yxyx} + C_{yxxy})$. Due to the isotropic nature of the system, only two of them are independent, and they satisfy the relations $C_{11} = C_{22}$, and, $2C_{44} = C_{11} - C_{12}$ [15]. It is quite common to describe the elastic properties of isotropic systems in terms of the *shear* modulus, μ , and the *bulk* modulus, κ , defined by $\mu = C_{44} - P$, and, $\kappa = \frac{1}{2}(C_{11} + C_{12})$. When these quantities are positive, the isotropic system is mechanically stable [16].

Our simulations were performed on systems consisting of 1600 atoms which were bonded to form a triangular 2D net. The topology of the net is such that the mean positions of the atoms form a regular triangular lattice with lattice spacing b_0 , while each pair of nearest-neighbor atoms is connected by a tether whose maximal extension is $b \geq b_0$. Periodic boundary conditions, which fixed the volume and prevented the net from collapsing, were applied. We denote by $\gamma \equiv b/b_0$ the ratio between the maximal and actual extensions of the net. Typical equilibrium configurations corresponding to two values of γ are depicted in Figure 1. We generated the MC configurations using a new updating scheme, recently proposed by Jaster [17], in which the conventional Metropolis step of a single particle is replaced by a collective step of chain of particles. At each MC time unit we made 1600 move attempts (with acceptance probability ~ 0.7), where at each attempt a new atom was selected randomly. (On the average, each atom was chosen once in a MC time unit.) Correlations between subsequent configurations were estimated from the autocorrelation function of the amplitude of the longest-wavelength phonon in the systems (both longitudinal and transverse phonons were checked). For all γ values, we found that after less than 1000 MC time units, the memory of the initial configuration was completely lost. We measured the stress and elastic constants for many values of γ . For each γ , we averaged the relevant quantities over a set of 1.5×10^7 configurations separated from each other by 3 MC time units. We also evaluated the standard deviations of the averages. The error bars appearing in the graphs which present our results correspond to one standard deviation. More technical details of the simulations,

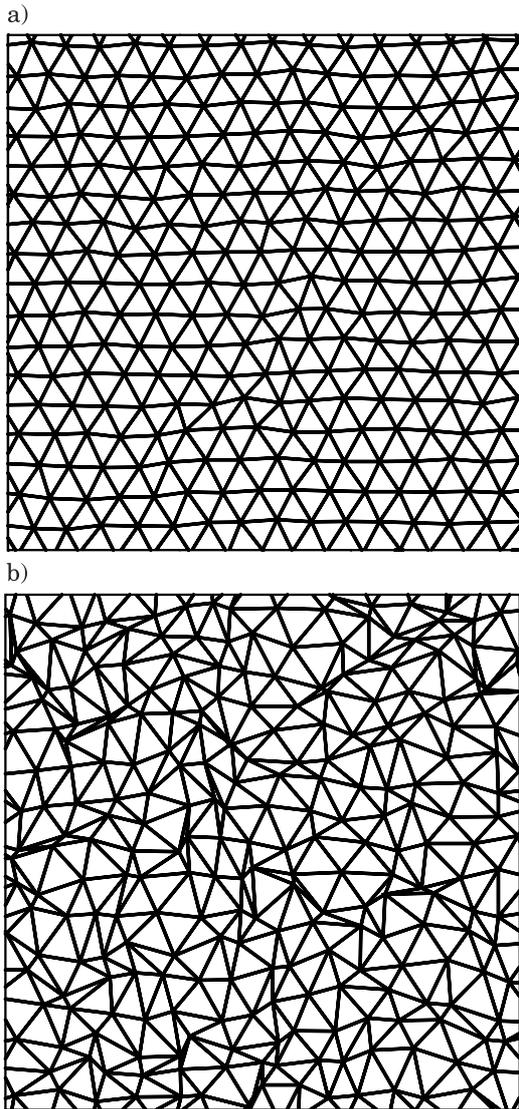


Fig. 1. Configurations corresponding to different values of the ratio γ between the maximal and actual extensions of the net: (a) $\gamma = 1.1$, (b) $\gamma = 1.5$. Only part of the net is shown in the figures.

as well as a detailed explanation on the formalism used in this work, were given in another publication [12].

When the net is fully extended ($\gamma = 1$), atoms cannot leave their mean lattice positions. Entropy, therefore, vanishes, while the stress and elastic constants diverge. For slightly larger values of γ , atoms are restricted to small thermal fluctuations around their lattice positions, as in Figure 1(a). A similar atomic-level picture appears in hard disks (2D “hard spheres”) solids for densities proximal to the close-packing density. In fact, the FN and the hard disks (HD) problems are closely related: In the latter (HD) the centers of the disks are not allowed to approach their neighbors a distance smaller than a , the diameter of the disks, while in the former (FN) atoms are not allowed to depart from their neighbors a distance larger the maximal extension of the bond, b . For HD solids, one can de-

fine the ratio $\delta = a/b_0 \leq 1$ between the diameter of the disks, a , and the mean lattice separation, b_0 . In the limits $\gamma \rightarrow 1$ and $\delta \rightarrow 1$ (corresponding to the FN and HD problems, respectively), the elastic constants of both systems coincide, as can be seen from the following argument: Let Π_{FN} and Π_{HD} be phase spaces of allowed configurations of a FN with a certain value of γ and of a HD solid with $\delta = 1/\gamma$, respectively. Each configuration in one of these phase spaces can be described by the set $\{\mathbf{u}_i\}$ of deviations of either the atoms of the net or the centers of the disks from their mean lattice positions. In the $\gamma, \delta \sim 1$ asymptotic regimes, we can assume that the size of all the deviations is much smaller than the lattice spacing, b_0 . One can easily check that if the set $\{\mathbf{u}_i\}$ represents an allowed microscopic configuration of the FN, then the set $\{-\mathbf{u}_i\}$ almost always corresponds to an allowed configuration of the HD system. Moreover, by this transformation we can generate almost all the configurations of Π_{HD} . The measure of the subgroup of configurations for which the mapping $\{\mathbf{u}_i\} \longleftrightarrow \{-\mathbf{u}_i\}$ between the two problems does not apply, diminishes proportionally to $\langle u_i^2 \rangle / b_0^2$. Thus, the mapping $\{\mathbf{u}_i\} \longleftrightarrow \{-\mathbf{u}_i\}$ is asymptotically a *one-to-one* transformation from Π_{FN} onto Π_{HD} . Since for both systems the Helmholtz free energy F is equal to $-kT \ln |\Pi|$, where $|\Pi|$ is the volume of the $2N$ -dimensional configuration phase space (N is the number of atoms), and since the Jacobian of the above transformation is unity, we readily find that the free energies F_{HD} and F_{FN} of the HD and FN systems, respectively, are related by

$$F_{\text{FN}}(N, \gamma) \simeq F_{\text{HD}}(N, \delta = 1/\gamma), \quad \text{for } \gamma \sim 1.$$

Suppose now that both systems are slightly deformed from their reference states. The displacements of the atoms from their original (undeformed) mean lattice positions can be divided into the set $\{\mathbf{u}_i\}$ of thermal fluctuations and the set $\{\mathbf{v}_i\}$ of small changes in mean lattice positions caused by the deformation. The transformation between Π_{FN} and Π_{HD} , in this case, maps both $\{\mathbf{u}_i\}$ to $\{-\mathbf{u}_i\}$ and $\{\mathbf{v}_i\}$ to $\{-\mathbf{v}_i\}$. The $\{\mathbf{v}_i\}$ mapping is equivalent to the reversal of the strain applied on the system. We therefore find that F_{HD} and F_{FN} will be equally modified, provided that opposite strains are applied on the FN and HD systems. The following asymptotic relations between the stress and elastic constants of these systems follow immediately: $\sigma_{\text{FN}}(\gamma) \simeq P_{\text{HD}}(1/\gamma)$, $\kappa_{\text{FN}}(\gamma) \simeq \kappa_{\text{HD}}(1/\gamma)$ and $C_{\text{FN}44}(\gamma) \simeq C_{\text{HD}44}(1/\gamma)$. These relations are very useful since the asymptotic expressions for P_{HD} , κ_{HD} and $C_{\text{HD}44}$ are available [18], and can be used to find the stress and bulk modulus of the FN. This gives us the *exact* expressions

$$\sigma_{\text{FN}}(\gamma) \simeq \frac{4/\sqrt{3}}{(\gamma^2 - 1)} \frac{kT}{b_0^2}, \quad (3)$$

$$\kappa_{\text{FN}}(\gamma) \simeq \frac{4/\sqrt{3}}{(\gamma^2 - 1)^2} \frac{kT}{b_0^2}. \quad (4)$$

For the elastic constant C_{44} , reference [18] finds only the asymptotic functional form, and therefore for our problem

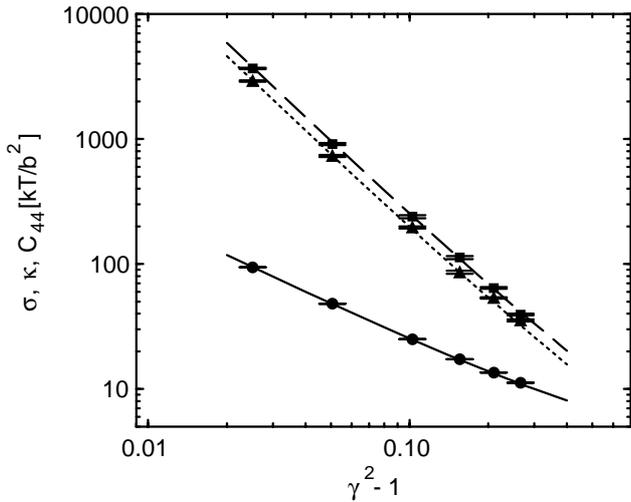


Fig. 2. Numerical results for the stress σ (circles), the bulk modulus κ (squares), and the elastic constant C_{44} (triangles), as a function of the ratio γ between the maximal and actual extensions of the net. Results are in kT/b^2 units. The solid, dashed and dotted curves depict the expressions on the right-hand sides of equations (3)–(5), respectively (with $A = 1.80$ in Eq. (5)).

we have

$$C_{\text{FN}44}(\gamma) \simeq \frac{A}{(\gamma^2 - 1)^2} \frac{kT}{b_0^2}, \quad (5)$$

with an unknown constant A . Our numerical results, presented in Figure 2, confirm these relations, which seem to be accurate over quite a large range of γ values. In equation (5) we use the value $A = 1.80 \pm 0.02$ obtained by fitting the asymptotic expression for C_{44} to the three data points corresponding to the smallest γ values. Note that while in equations (3)–(5) P_{HD} , κ_{HD} and $C_{\text{HD}44}$ are expressed in units of kT/b_0^2 , in Figure 2 they are given in units of kT/b^2 . In this representation, the stress and elastic constants of the FN are scaled to depend on the parameter γ alone.

Figure 3 shows the dependence of the stress and elastic constants on γ for weakly stretched nets. We observe a spectacular decay of elastic constants to almost zero for $\gamma \sim 3$, and at the same time we note that the stress becomes independent of γ . The same result (for the critical value of γ and the γ -independent pressure) has been obtained in reference [13]. The very fact of decrease of elastic constants with increasing γ should not be surprising, because it is intuitively clear that larger γ represent a “looser” and “weaker” solid. However, almost vanishing values already at $\gamma \sim 3$ are *not* direct consequences of the “weakness” of the solid, but of the fact that a “loose” network can be approximated by a network of *Gaussian springs*. Gaussian spring is a linear spring of vanishing unstressed length. The energy of such a spring, $E = \frac{1}{2}Kr^2$, is simply proportional to its squared end-to-end distance, r^2 . We will show that elastic solid formed by such springs has *exactly* vanishing elastic constants, independently of

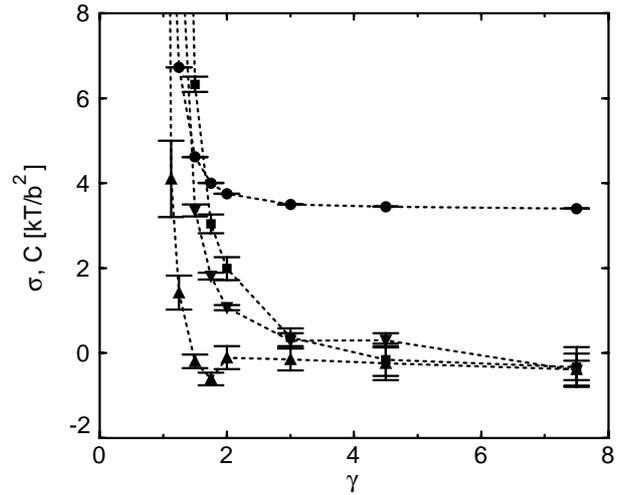


Fig. 3. Numerical results for the stress σ (circles), and the elastic constants C_{11} (squares), C_{12} (triangles pointing up) and C_{44} (triangles pointing down), as a function of the ratio γ between the maximal and actual extensions of the net. Results are in kT/b^2 units. The lines are guides to the eye.

the value of the spring constant K . Thus, the effect observed in Figure 3 is an indication of the Gaussian nature of the system.

It is easy to calculate the elastic properties of Gaussian networks at $T = 0$: The stresses of such networks depend on their topologies, namely on the details of the connectivity between the atoms and on the values of the springs constants between them. For 2D networks the stresses are not modified due to homogeneous changes in the size of the net, since the force exerted on the surface grows (diminishes) linearly with the length of the boundaries. Moreover, at $T = 0$, the free energy, F , coincides with the internal energy $E = \sum_{\text{bonds } \langle \alpha\beta \rangle} \frac{1}{2} K_{\alpha\beta} (r^{\alpha\beta})^2$, where $r^{\alpha\beta}$ is the length of the bond connecting atoms “ α ” and “ β ”, and $K_{\alpha\beta}$ is the spring constant assigned to this bond. From equation (1) it is obvious that the energy expansion in the strain variables includes only linear terms in η , and hence, by comparing with equation (2), $C_{ijkl}(T = 0) \equiv 0$ [19]. This identity, as well as the size independence of the stresses, holds at any other temperature since Gaussian networks have the interesting feature that their stress and elastic constants are temperature independent! For the stresses this feature is readily understood: The stresses can be expressed as the averages of quantities which are linear in the coordinates of the atoms. When the statistical weights of the distribution are Gaussian, *i.e.*, an exponent of a quadratic form of the coordinates, these averages coincide with the most probable values, namely their values at equilibrium. The temperature independence of the elastic constants then follows immediately, since the latter are just the derivatives of the stress components.

The similarity between non-stressed tethered and Gaussian one-dimensional (1D) nets, *i.e.*, linear polymers, is a consequence of central limit theorem [20]. For topologically two-dimensional regular (non-random) nets, such similarity was demonstrated by Kantor *et al.* [4]:

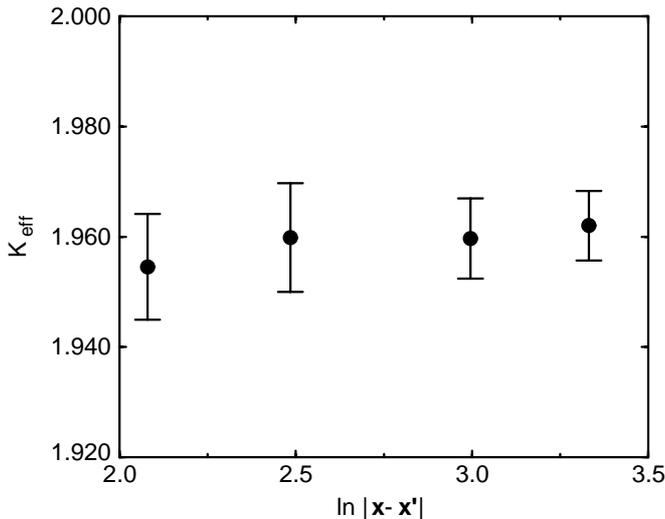


Fig. 4. The effective spring constant K_{eff} , extracted from MC measurements of $r_{\mathbf{x}\mathbf{x}'}^2 = \langle |\mathbf{r}(\mathbf{x}) - \mathbf{r}(\mathbf{x}')|^2 \rangle$ (see Eq. (6)). The error bars correspond to one standard deviation in the estimated value of $r_{\mathbf{x}\mathbf{x}'}^2$.

In both tethered and Gaussian two-dimensional nets, the mean-squared distance in the embedding space, $r_{\mathbf{x}\mathbf{x}'}^2 = \langle |\mathbf{r}(\mathbf{x}) - \mathbf{r}(\mathbf{x}')|^2 \rangle$, between two distant points whose internal positions in the net (measured in lattice constants) are \mathbf{x} and \mathbf{x}' , grows proportionally to $\ln |\mathbf{x} - \mathbf{x}'|$. One can define the effective spring constant, K_{eff} , as the value of K of a Gaussian network with the same connectivity and statistical properties as the tethered network. The value of K_{eff} is extracted from the ratio of the mean-squared distance, $r_{\mathbf{x}\mathbf{x}'}^2$, between two points \mathbf{x} and \mathbf{x}' on the FN, and the mean-squared distance $\tilde{r}_{\mathbf{x}\mathbf{x}'}^2$ between the same two points on a Gaussian network of unit spring constants:

$$K_{\text{eff}} = \tilde{r}_{\mathbf{x}\mathbf{x}'}^2 / r_{\mathbf{x}\mathbf{x}'}^2. \quad (6)$$

$\tilde{r}_{\mathbf{x}\mathbf{x}'}^2$ can be calculated exactly, while the value of the corresponding $r_{\mathbf{x}\mathbf{x}'}^2 = \langle |\mathbf{r}(\mathbf{x}) - \mathbf{r}(\mathbf{x}')|^2 \rangle$ can be extracted from MC simulations of the FN with free boundaries conditions (*i.e.*, in the absence of external pressure). We simulated a FN of $56^2 = 3136$ atoms and measured (using 10^7 different configurations) $r_{\mathbf{x}\mathbf{x}'}^2$ for several pairs of points \mathbf{x} and \mathbf{x}' at different lattice separations. With these measurements we evaluated the effective spring constants (using Eq. (6)), and found, as shown in Figure 4, that for the FN model $K_{\text{eff}} \simeq 1.96 kT/b^2$. In order to support our conclusion about the crossover into the Gaussian regime, we need to show that the constant value to which the stress drops in Figure 3 is just the stress applied by a Gaussian net with spring constants K_{eff} calculated for *non-stressed* FN. For a Gaussian net with $K \simeq 1.96$, one finds that $\sigma = \sqrt{3}K \sim 3.39 kT/b^2$ which indeed coincides with the value of $3.4 kT/b^2$, extracted from Figure 3.

The persistence of the Gaussian regime to intermediate values of γ ($\gamma \sim 3$) is not unique for 2D nets. Such behavior is also found, for instance, in 1D polymers. Let us consider, for a moment, a chain of $N \gg 1$ tethers of maximal length b , which is stretched by a force f , to an

end-to-end length $l = Nb_0$. It is a well known fact that this chain will be Gaussian, *i.e.*, f and l will be proportional to each other, provided that l does not exceed the order of magnitude of the root-mean-square size of the chain:

$$l = Nb_0 \lesssim \sqrt{N}b. \quad (7)$$

Yet, one must understand that in order to observe Gaussian elastic behavior, it is not essential to apply this criterion (7) to the whole chain, but only to small segments of it. If there exists a certain length scale at which the potential between the atoms becomes effectively quadratic, *i.e.*, can be replaced by a Gaussian spring, then the whole chain is like a chain of Gaussian springs, and therefore it is itself Gaussian. For a linear polymer chain, the effective potential between non-neighboring atoms is calculated by integrating out the spatial degrees of freedom of the atoms located between them. Such calculations are usually done iteratively, where on each “rescaling” step every second atom is integrated out. It appears that even elementary potentials which are very different from parabola, are brought into a parabolic form within a few “rescaling” steps. For the specific potential used in this work, three steps are sufficient, which means that a segment of $N \sim 10$ tethers may be justly considered as an effective Gaussian spring. Similarly to a macroscopically large chain, we expect that the Gaussian nature of this segment will persist as long as it is stretched to a length which does not exceed its root-mean-square size, namely, as long as $10b_0 \lesssim \sqrt{10}b$ (see criterion (7)). This relation gives the lower limit, $\gamma = b/b_0 \gtrsim \sqrt{10} \sim 3$, of the Gaussian regime of a 1D chain of tethers. For a 2D regular phantom net, the effective potential becomes approximately parabolic also for a distance of number of bonds, $N \sim 10$ [4]. The root-mean-square distance between two such points is $b\sqrt{\ln N}$. Thus, in order to observe Gaussian elastic behavior, we require that $10b_0 \lesssim b\sqrt{\ln 10}$, or, $\gamma = b/b_0 \gtrsim 10/\sqrt{\ln 10} \sim 4$, which is consistent with the value $\gamma \sim 3$, observed in Figure 3.

In summary, we have applied a new “fluctuation” formalism to MC determination of the stress and elastic constants of stretched tethered networks. These systems provide a convenient framework for studying the entropic contribution to elasticity in real polymeric systems. The Gaussian nature of entropic elasticity, observed for non-stressed phantom nets, was also found when stress was applied. It breaks only for highly extended networks, close to their full extension. This point has interesting implications to the problem of the critical elastic behavior of gels right above the gel-point. As already mentioned in the first paragraph of this paper, recently it was suggested by Plischke, Joós and co-workers that this behavior is dominated by entropy [3]. These authors studied numerically (using a different technique) the elastic behavior, at $T \neq 0$, of bond diluted (percolating) systems at which only a fraction p of the bonds was present. Their results in 2D for the critical exponent f characterizing the growth of the shear modulus above the percolation threshold p_c , $\mu \sim (p - p_c)^f$, match (within the range of error) the known result for the exponent t describing the conductivity of random

resistors networks $\Sigma \sim (p - p_c)^t$. The question is whether this result is universal. For Gaussian networks the identity, $f = t$, can be proven rigorously [21]. One can further argue that this result also applies to other types of interactions, provided that above a certain finite length scale, the network is *effectively* Gaussian. We have shown here that this property is not always insured. In a percolation problem, the elastic backbone is inhomogeneous and includes very tenuous parts where the tension applied to the network is distributed between very few strands. Such strands may deviate from Gaussian behavior when high stress is applied. Further complications can arise from excluded-volume effects which have not been discussed here at all.

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