Two-dimensional experimental simulation of polymers in annealed disordered media

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We model the motion of polymers in random media by chains of connected disks placed on an air stream table. The dynamic disorder is modeled by placing light disks on the table, which move in a random manner. We analyze the variation of the mean radius of gyration, $\langle R_g \rangle$, with the chain length N for different densities of mobile disks, finding that $\langle R_g \rangle \sim N^\nu$ where $\nu = 0.68 \pm 0.03$ for all densities. [S1063-651X(98)08803-5]

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The behavior of polymer chains in disordered media has been an active field of research for many years, since it is both a simple to state but far from trivial problem of statistical physics with external disorder, and a field with numerous practical applications [1–3]. Among these, we cite polymer transport through microporous membranes, gel permeation chromatography, viscoelasticity of polymer solutions, and so on. The main theoretical approach to the problem of polymers in random environments has been that of the self-avoiding walk (SAW) [4–23]. Despite the major effort that has gone into this work, both numerically and analytically, fundamental open questions still linger on. We concentrate in this paper on the radius of gyration of the polymers, $\langle R_g \rangle$. This is related to the number of monomers N or the length of the SAW by a power law

$$\langle R_g \rangle \sim N^\nu. \tag{1}$$

If the polymer (SAW) does not interact with its environment, the exponent $\nu = \nu_0$ is known to be 3/4 in two dimensions [1]. However, when there are interactions the behavior of ν is less clear. Kremer [4] argued that quenched disorder does not affect ν as long as the disorder is less than the percolation threshold. Baumgärtner and Muthukumar [24] studied numerically polymers in three dimensions without *a priori* assuming self-exclusion in an environment of fixed obstacles, i.e., quenched disorder. They concluded that there is a transition between a Gaussian phase, where $\langle R_g \rangle \sim N^{1/2}$ and a collapsed phase where $\langle R_g \rangle$ is independent of N when the number of obstacles is increased. This work was followed up by Edwards and Muthukumar [10] who studied the same system analytically in turn further refined by Cates and Ball [25] and Nattermann and Renz [26]. Cates and Ball also discuss the annealed-disorder case, and find through a Flory argument that $\nu = 1/4$ in the collapsed phase. Thirumalai [27] argued based on analytical arguments that in three dimensions *a priori* self-avoiding polymers do not have the same behavior as the free SAW when in the presence of weak annealed disorder. When the disorder is increased, the polymers undergo a transition to a collapsed state. Similar conclusions, valuable for two and three dimensions, but with quenched disorder, were drawn by Machta and Guyer [15] using a Flory approach. Honeycutt and Thirumalai [21] stud-

ied numerically the problem of anchored and nonanchored polymers in two-dimensional surroundings with annealed disorder, suggesting that $\nu = 2/3$ when anchored and smaller value, 1/6 or 1/8 depending on the disorder, if nonanchored. Le Doussal and Machta [22] on the other hand use arguments based on renormalization group theory to argue that $\nu < \nu_0$ for nonanchored polymers and $\nu > \nu_0$ for anchored polymers, both subject to annealed disorder. Cherayil [28] used renormalization group methods to study polymers without *a priori* self-exclusion in both annealed and quenched disorder. A transition between a collapsed phase and a phase dominated by self-avoidance was found, i.e., $\nu = \nu_0$. Both types of disorder—quenched or annealed—gave rise to the same asymptotic behavior. Obukhov [20] argued that self-avoiding polymers in two dimensions and quenched disorder are dominated by the self-avoidance, so that $\nu = \nu_0 = 3/4$. Wu *et al.* [29] studied numerically polymers without *a priori* self-exclusion in a two-dimensional environment with annealed disorder, but argue that the results are also valid for quenched disorder. They find a value for ν close to 1/4, which is the Flory value. Geroff *et al.* [30] studied numerically three-dimensional self-avoiding polymers moving among fixed obstacles, i.e., quenched randomness. The static
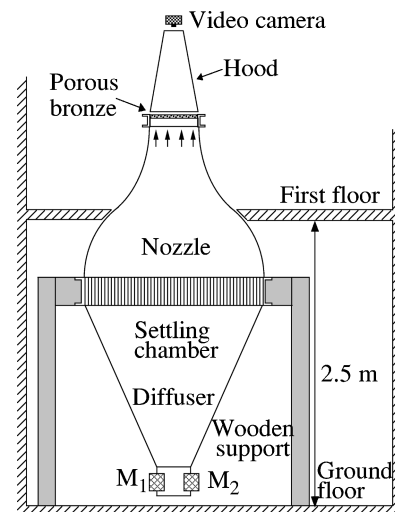

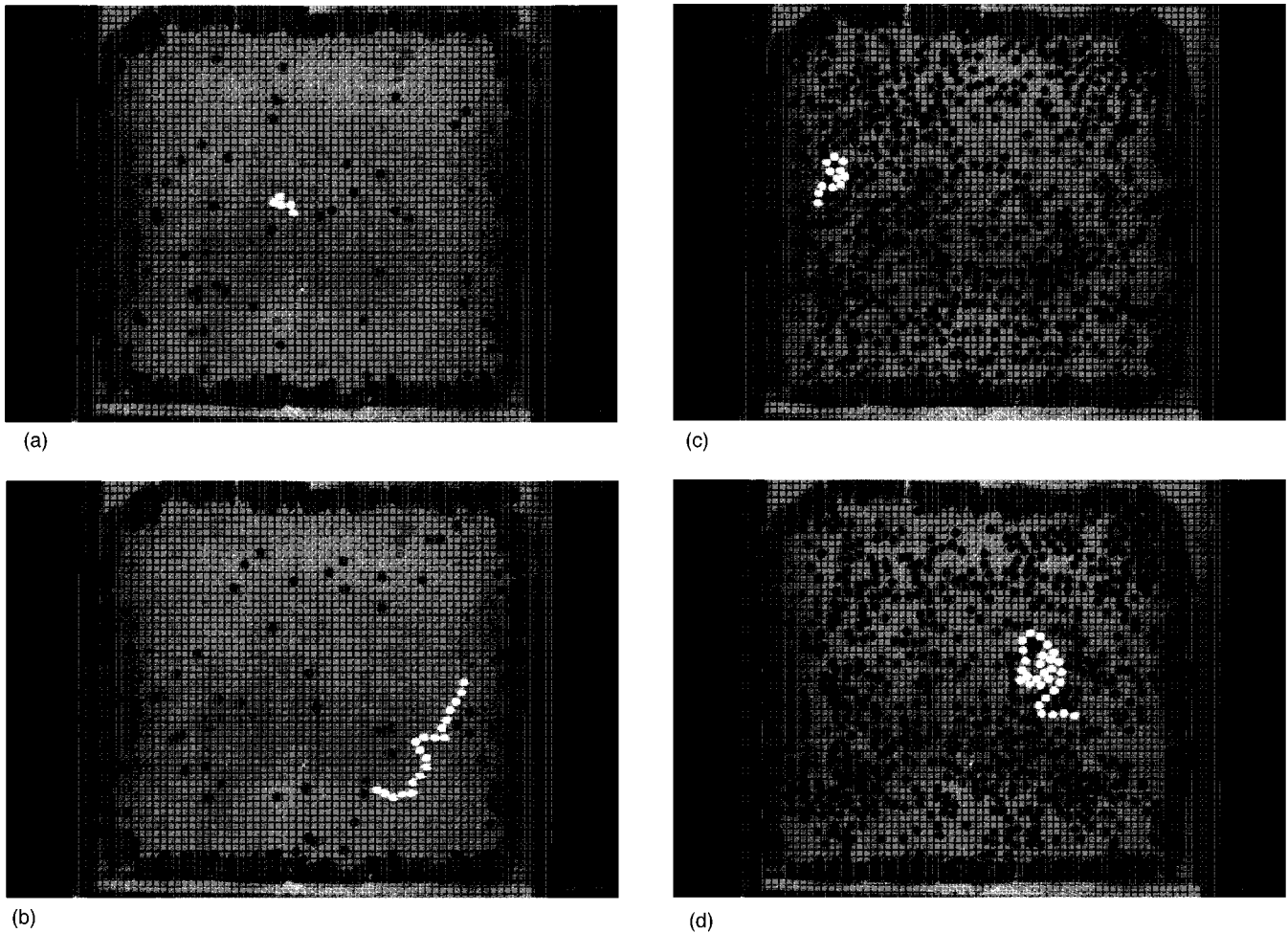FIG. 1. A schematic drawing of the air stream table.

FIG. 2. Images showing different configurations of the chain (white) for two densities of disks (black): (a) $N=4$ and (b) $N=19$ for $C=2.5\%$ and (c) $N=11$ and (d) $N=28$ for $C=20\%$.

properties of the polymers remained unaffected by the disorder. In an "experimental simulation," Tasserie *et al.* [31] have modeled nonanchored polymers interacting with a quenched disordered medium on an air table. They found $\nu=0.68\pm0.03$, seemingly independent of the densities of the objects forming the environment. Grassberger [32] simulated two-dimensional self-avoiding polymers in quenched random environments, finding $\nu=0.783\pm0.003$, in contradiction to the results of Tasserie *et al.* He also shows that there is a significant difference between the quenched and annealed disorder cases.

We report here an experimental study of the radius of gyration, $\langle R_g \rangle$, of a two-dimensional self-avoiding chain moving randomly in an environment with annealed disorder. The chain is placed on an air stream table built to study random two-dimensional systems [33]. It consists of a horizontal sintered bronze plate of size 50×50 cm^2 and thickness 5 mm. The plate is placed at the exit of a long vertical wind tunnel. Thus, the air flow (laminar and homogeneous) moves through the porous table, see Fig. 1. The air flow can be controlled by regulating a rheostat connected to two fans at the bottom of the wind tunnel. The fan power is linearly related to the air flow velocity and it was found that velocity profiles show a satisfactory overall uniformity at 150 V (air velocity of 30 cm/s). All experiments were performed under

the same conditions. At the other side of the plate, the flow is homogeneously turbulent on a small scale. This turbulence agitates both the chain and the disks that are placed on the table to simulate the random environment. These disks have a diameter of 8 mm and a thickness of 1 mm. They are
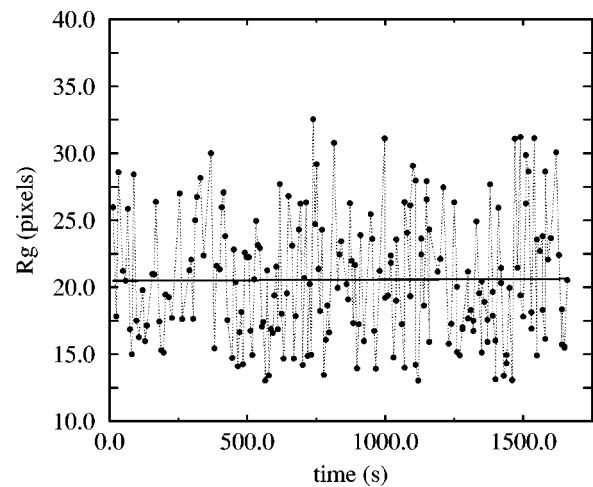

FIG. 3. Radius of gyration R_g (in pixels) as a function of time (in s) for $N=11$ in a disordered medium of density $C=2.5\%$.
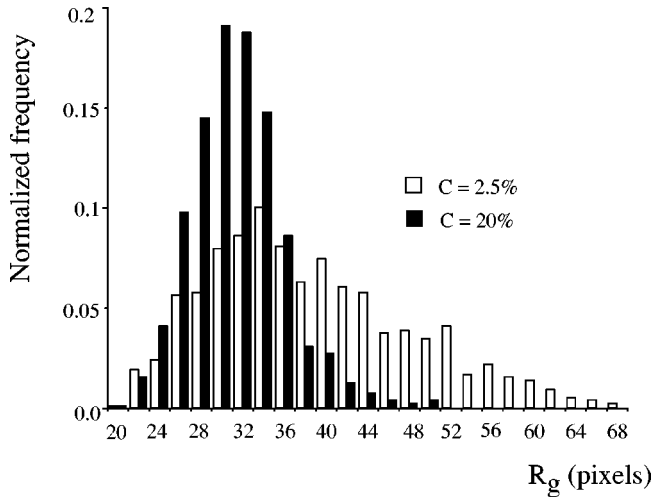
FIG. 4. Normalized distributions of the radius of gyration R_g for a polymer length of $N = 19$ for $C = 2.5\%$ and $C = 20\%$.

FIG. 5. Linear variation of the logarithm of $\langle R_g \rangle$ as a function of the logarithm of N for different densities C: From bottom to top the curves represent (a) $C = 2.5\%$, $\nu = 0.66$; (b) $C = 5.5\%$, $\nu = 0.66$; (c) $C = 10\%$, $\nu = 0.69$; (d) $C = 20\%$, $\nu = 0.68$.

suspended above the work surface due to the air stream, and move in a random fashion on the air table. This randomness is caused by the turbulent component of the flow field and the geometrical defects of the disks causing instabilities as do collisions between the disks. The disks, however, do not form a gas in a thermodynamic sense, but rather a driven dissipative system (kinetic energy is essentially dissipated during collisions). Energy is furnished by the air flow. Because of the lack of equipartition—clearly put in evidence when studying mixtures of disks with two different diameters—no temperature can be assigned to the gas. However, we may define a pseudotemperature T^*, which is a decreasing function of the number of disks [34]. The polymers are modeled by a chain of the disks described above. The disks are held together by a very thin silk thread glued at the center of each disk. Let us remark that the silk thread must be as thin as possible as the stiffness of the thread may easily dominate the individual agitations of the disks. This has been verified by Tasserie *et al.* [31]. In these conditions, the interactions between chain and gas are only due to collisions. In order to study the dynamical behavior of the chain, we record the images with a black and white videocamera placed 2 m above the porous bronze plate. This camera is connected to a work station. An image analyzer (VISILOG 4.0) allows us to record image sequences and to treat them automatically in order to obtain the different parameters. The disks forming the chains are white and disks constituting the disordered medium are black: each image acquisition is first binarized in order to insulate the chain from its environment. Figure 2 shows 4 images of some typical configurations taken by a chain of (a) $N = 4$ and (b) $N = 19$ into a gas particles of density $C = 2.5\%$ and (c) $N = 11$ and (d) $N = 28$ in a gas of density $C = 20\%$ (N is the number of bonds and C is the area occupied by disks divided by total area of the work surface). To obtain the radius of gyration R_g we compute coordinates corresponding to the mass center and then the second moments for a large number of decorrelated configurations on the time. 200 configurations were recorded for each chain with time intervals varying from 7 to 120 s (for the longest chains). These intervals were sufficiently long to ensure that correlations between consecutive configurations
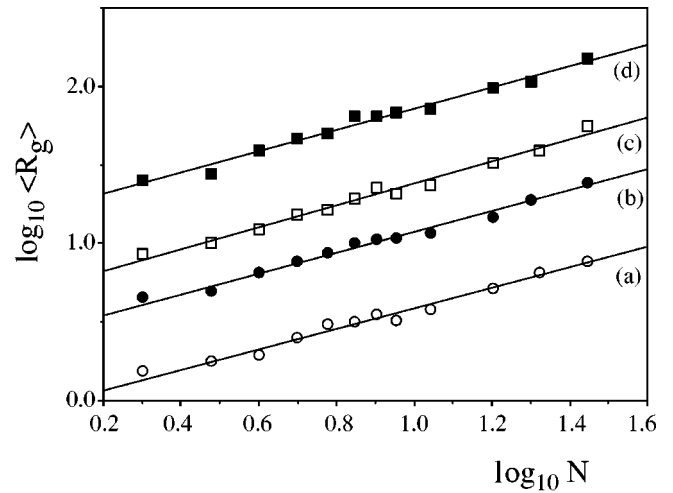
were negligible. Figure 3 shows a typical variation of the radius of gyration R_g as a function of time for $N = 11$ and for a density of objects $C = 2.5\%$. In this experiment, R_g has been computed roughly every 11 s. The mean radius of gyration $\langle R_g \rangle$ is then obtained by averaging R_g over all the configurations ($\langle R_g \rangle = 20.6$ in Fig. 3). In Fig. 4 we show the normalized distributions of the mean radius of gyration for a polymer of length $N = 19$ and for two densities of objects: $C = 2.5\%$ (in white) and 20% (in black). The histograms show that the distribution is largest at small density C of mobile obstacles. At highest density, the high number of objects keep the polymer from having stretched configurations. The mean value is more frequent at higher density than at the slower one. We have performed experiments for four densities C of objects of the gas particles: 2.5%, 5.5%, 10%, and 20% and for chain lengths of number of bonds varying from $N = 2$ to 28. Figure 5 shows the experimental results obtained for the four densities of gas particles. We have plotted the logarithm of the mean radius of gyration $\langle R_g \rangle$ as a function of the logarithm chain length N. We observe a linear variation corresponding to a power behavior between $\langle R_g \rangle$ and the length chain N, which is in good agreement with Eq. (1). The slopes of different curves are obtained by computing linear regressions over the experimental data for the four densities of disks: (1) $C = 2.5\%$, $\nu = 0.66$; (2) $C = 5.5\%$, $\nu = 0.66$; (3) $C = 10\%$, $\nu = 0.69$; (4) $C = 20\%$, $\nu = 0.68$. All the exponents are close to each other, and may be summarized as $\nu = 0.68 \pm 0.03$ (where the error is statistical). Within the quite limited range of chain lengths and densities C, the exponent seems universal. The exponent is smaller than $\nu_0 = 3/4$, the free SAW value, and indistinguishable from that obtained by Tasserie *et al.* who studied chains moving on the air flow table, but where the disorder was quenched in that the disks were fixed.

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