Density correlations in paper

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We present an analysis of areal mass density correlations in paper. Using β radiography, the local mass density of laboratory paper sheets has been measured. The real space density autocorrelation function calculated from the data reveals a nontrivial power law type of correlations with the decay exponent being roughly independent of the basis weight of the sheets. However, for low densities we find that correlations may extend at least an order of magnitude beyond the fiber length, whereas for heavier paper they quickly die out. [S1063-651X(96)51307-3]

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The sedimentation of particles from a suspension poses a formidable theoretical problem on a microscopic level [1]. There are challenging theoretical problems associated with the fundamentals of two-phase flow, and in particular with hydrodynamic interactions arising between constituent particles [2–4]. These interactions have been shown to strongly influence the structures formed by sedimentation [2,4], as compared to simple ballistic deposition models [5].

A striking example of a material formed by such a precipitation process is ordinary paper [6]. Its existence begins as a suspension of fibers in water, and it ends up forming a disordered fiber network. The structure of paper is an interesting problem with obvious commercial interest. The distribution of the local mass density in a paper sheet reflects the complicated physical processes during paper making and has been a long-standing subject of study [6]. Traditionally, its wavelength power spectrum has been used to characterize the size of ''flocs,'' or formation in paper [6–9].

In this paper, we address the fundamental question of *how random paper is* by studying real space correlations in the mass distribution of laboratory-made paper sheets. In particular, we investigate the existence and range of nontrivial *power law correlations*. This is motivated by two reasons. First, fibers in the suspension tend to *floc* together, giving rise to structures often clearly visible even to the eye [10,11] (see also Fig. 1). Flocculation in such suspensions may take place because of chemical or mechanical interactions between the fibers. A more complex element influencing flocculation is the hydrodynamics of the suspension. At present, there is no microscopic understanding of this since even for a static suspension of simple particles it is a formidable theoretical problem [4]. Second, power law correlations have important implications for recent experiments on front propagation (burning and imbibition), where paper has been assumed to be a uniformly random medium [12]. Our results indeed show the existence correlations well approximated by a power law type function that may persist up to about an order of magnitude beyond the fiber length. We also find that within the power law regime, the decay exponent is independent of areal mass density.

Our analysis is based on hand-made laboratory paper sheets from typical chemical pulp of industrial origin. The pulp is first diluted in a suspension of about 0.12% by weight of fibers. The paper making procedure is a standard one, where the suspension is poured into a sheet mold. At this stage, flocculation of fibers may take place, with the degree of it depending on how long the suspension is allowed to settle [13]. The liquid is then drained out by gravity through the bottom of the mold, where a wire mesh collects the deposited fibers. The actual paper sheet thus forms on the mesh. After formation, the sheets were wet pressed at 490 kPa and then dried in a drum. Three independent sets of paper sheets, A, B, and C were produced, and for sets A and C the pulp was fractionated [14]. Sheets of size of 16×16 cm^2 were prepared at several different final basis weights, ranging from 6.6 to 120 $\mathrm{g/m}^2$.

A map of the local mass density was obtained using β radiography [8,15]. Figure 1 shows an example of a radiograph from a sheet of basis weight of 80 $\mathrm{g/m}^2$. The radio-

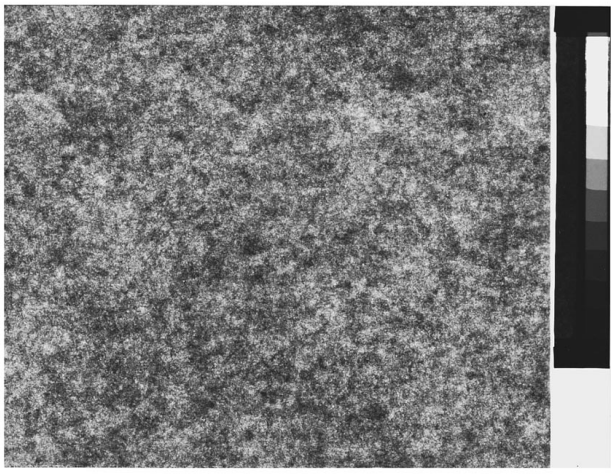

FIG. 1. A 14×17 cm^2 sample radiograph of a paper sheet with basis weight 80 $\mathrm{g/m}^2$. The gray scale shown on the side has ten shades, starting from zero basis weight and increasing with steps of 14.5 $\mathrm{g/cm}^2$.

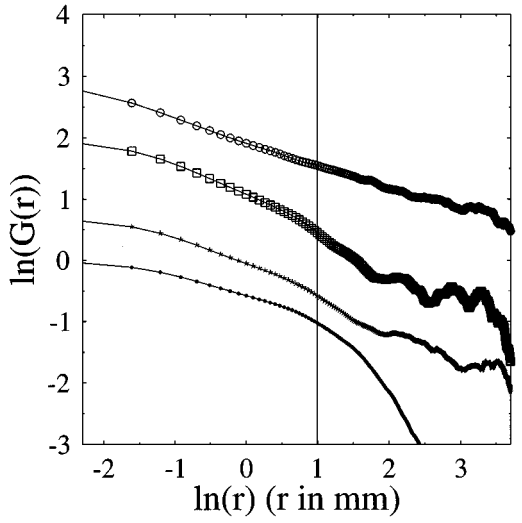

FIG. 2. A log-log plot of the correlation function $G(r)$ for four paper sheets of basis weights 7.2 g/m^2, 70.4 g/m^2, 86.3 g/m^2, and 118.9 g/m^2 (from top to bottom). The vertical line indicates the average fiber length $\lambda_A = 2.7$ mm. Data are from set A, and the curves have been shifted for clarity. For the lowest densities, the power law extends up to about 14 times the fiber length.

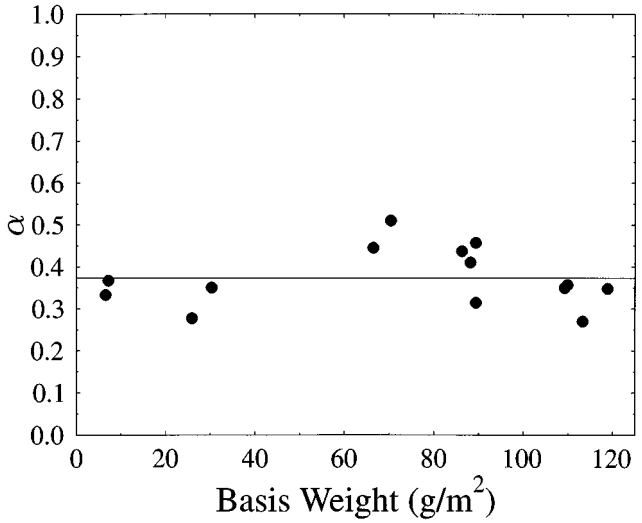

FIG. 3. The power law exponent α vs basis weight of the paper sheets for set A.

graphs typically have an imaging accuracy of about 0.2 mm (comparable to the thickness of the sheet [15]). Each set of digitized data is roughly 900×1600 pixels with a pixel size of about 0.09 mm. As a check, the global mass distribution was confirmed to be a Gaussian.

From the data, we calculated the two-point density correlation function as

$$G(\vec{r}) = \langle [m(\vec{x}) - \overline{m}][m(\vec{x}+\vec{r}) - \overline{m}] \rangle, \tag{1}$$

where $m(\vec{x})$ is the local mass density, $\overline{m}$ its average and $\langle\ \rangle$ an average over each sheet. As $m(\vec{x})$ can in this case be assumed to be isotropic, G is symmetric [16].

In Fig. 2 we show a log-log plot of Eq. (1) for four different paper sheets whose basis weights increase from top to bottom. For each basis weight studied, we tested three or four independent samples and found consistent results. In all cases studied, for $r<\lambda$, $G(r)$ follows to a good degree of approximation the form

$$G(r) \simeq r^{-\alpha}. \tag{2}$$

In Fig. 3 we plot α vs basis weight for set A, which spans a large range of thicknesses. We find that α is essentially *independent* of the basis weight, with an average value of $\alpha = 0.37 \pm 0.07$. Data sets B and C show the same result, with a slightly larger value of α. For comparison, we also examined two commercial paper grades of newsprint type finding similar results. The main difference was the anisotropy of commercial paper. Due to the manufacturing process there is a difference between the so called machine and cross directions in the final paper that leads to different power law exponents in the two directions [17].

For $r>\lambda$, we find that the *range* of $G(r)$ depends on the basis weight. In particular, the 7.2 g/m^2 sheets exhibit power law behavior for approximately 14 fiber lengths, whereas for the 118.9 g/m^2 sheets, no power law is seen beyond $r \approx \lambda$. Similar long range correlations are also observed for basis weights up to about 26–30 g/m^2 from set A. Analysis of the curves using, e.g., running exponents from $d\ln(G)/d\ln(r)$ shows that for low density cases, the power law behavior is rather consistent across and beyond the fiber length (see Fig. 2). For data sets B and C, all data obtained were of weight greater than 40 g/m^2 and thus showed no discernible power law correlations beyond $r \approx \lambda$. These results are qualitatively consistent with other experiments in the sense that the structure of paper is known to become more homogeneous with increasing basis weight [18].

For completeness, we also analyzed the *wavelength power spectrum* $k^2P(k)$ of our data, where $P(k)$ is the Fourier transform of $G(r)$. This measure is commonly used in paper physics to find the dominating wavelength component in the density distribution [8]. While $P(k)$ contains information equivalent to $G(r)$, the wavelength power spectrum for the present case does not reveal the initial power law behavior clearly. This is because of the additional k^2 factor in its definition, as well as cutoffs due to the finite size of the sample and the finite range of the power law in real space. Indeed, the wavelength power spectrum even for a two-dimensional (2D) uniformly random fiber network is just a smooth function that peaks at an uninteresting value of 0.05 times the fiber length [6].

The reason for nontrivial correlations must stem from the underlying physical processes in paper making, such as flocculation of fibers in the suspension and sedimentation, as explained in the Introduction. In particular, the persistence of a constant power law with $\alpha \approx 0.4$ for all densities studied here is quite interesting. This is because for a simple, uniformly random 2D fiber network, it can be shown that $\alpha = 1$ up to the fiber length, and no correlations exist beyond this [19]. The nontrivial experimental value of α may be due to enhanced flocculation in the suspension, or to enhanced hydrodynamic activity in the formation phase. However, trying to microscopically model the dynamics of fibers in a suspension even for hand-made paper sheets is a prohibitively difficult problem.

Recently, we have carried out an extensive study [19] of a simple 2D fiber deposition model with the rule that if a deposited fiber overlaps with another fiber it is always accepted. However, if it lands on empty space it is accepted only with a probability p, where $0<p<1$. We find that this model gives a constant nontrivial power law exponent $\alpha<1$ for all densities only if p is varied in such a way that clustering is enhanced for high densities. However, the model can at best only produce this power law up to about the length of a typical cluster in the fiber network. A plausible mechanism for obtaining long-range correlations at low areal densities is the introduction of effective cluster-cluster interactions. Such interactions could arise from hydrodynamics of the suspension. In Ref. [20] it has been shown that cluster-cluster interactions can indeed produce power law type density correlations well beyond the range of the cluster sizes. This is in agreement with the idea of enhanced flocculation within the suspension.

In conclusion, we have investigated the two-point density correlations in laboratory made paper sheets. Our results demonstrate the existence of nontrivial power law correlations. The power law exponent seems to be rather independent of paper density, but the range of the correlations is not. For basis weights greater than about 40 g/m^2, power law correlations only exist for distances of the order of the fiber length, while for smaller basis weights they can persist for distances more than ten times the fiber length.

The existence of nontrivial density correlations in paper sheets is of particular importance for experiments in which paper has been assumed to be uniformly random [12]. These include kinetic roughening of burning fronts [21] and wetting studies of directed percolation. Our results suggest that paper is *not* as random as assumed. Power law correlations extending to the centimeter range may induce undesirable effects such as crossover behavior.

The paper-making process is a complicated problem. We nevertheless hope to have demonstrated that the behavior of the correlation functions gives important information about paper structure and sedimentation in general. Many interesting questions still remain, such as the true microscopic origin of the power law correlations and the effect of the hydrodynamics of the suspension on the paper structure.

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