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Experimental Determinations of the Excluded-Volume Exponent in Different Environments

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The excluded-volume exponent ν is determined from a neutron scattering experiment by polystyrene chains, in the "intermediate" momentum-transfer range. The chains were successively dispersed in a good solvent, in a Θ solvent, and in the bulk material. The new result is the value of ν for the bulk material, which is discussed below.

The excluded-volume interaction introduces a characteristic exponent ν in the scaling law[1] of the end-to-end distribution function of chain segments separated by n steps,

$$P_n(r) = (1/n^{3\nu})f(r/n^\nu). \tag{1}$$

The theoretical value of ν is very close[2] to $\frac{3}{5}$ in the presence of an excluded-volume interaction, and $\frac{1}{2}$ in the case of a free chain. Two observables can be derived from Eq. (1):

(a) The mean squared end-to-end distance $\langle R^2 \rangle$, for a polymer chain of N segments,

$$\langle R^2 \rangle = KN^{2\nu}, \tag{2}$$

where K is a constant independent of N. Relation (2) provides an experimental test of the value of ν in the momentum-transfer range $q < (\langle R^2 \rangle)^{-1/2}$.

(b) The pair correlation function

$$g(r) = Be^{-r/Nl}r^{-(3\nu-1)/\nu} \tag{3}$$

in the range $l < r < (\langle R^2 \rangle)^{1/2}$, where l is the step length and B is a constant independent of N. Equation (3) suggests a scattering experiment in the intermediate momentum-transfer range

$$(\langle R^2 \rangle)^{-1/2} < q < l^{-1}. \tag{4}$$

The scattering law is

$$S(q) = \frac{12}{(ql)^{1/\nu} + O(1/N)}. \tag{5}$$

The value of ν is determined by the q dependence of the scattered intensity.

Relations (2) and (5) may be directly tested in a scattering experiment, provided that the single chain can be observed in its environment. This problem was discussed in earlier papers,[3,4] where it was shown that the neutron scattering technique is the only suitable one for the bulk material.

Real chains are nonintersecting and the measured value of ν is expected to be $\frac{3}{5}$, as in the case of a theoretical chain with excluded-volume interaction. This is observed in good solvents,[5] in the limit of zero polymer concentration. The environment of the chain may, however, produce situations in which the chain behaves as if it were free. A well-known case is the Θ solvent at the Flory[6] temperature $T = \Theta$. Another situation of interest is the bulk material, for which it is also thought[6] that the individual chains display no excluded-volume interaction. This was recently tested[7] by a neutron scattering experiment by deu-

TABLE I. Characteristics of the samples

Sample	Polymer	Molecular mass	Polydispersity M_w/M_n	Environment	Concentration (g cm^{-3})
A	PSD	1.1×10^6	1.17	CS$_2$, room temperature	2.5×10^{-3}
B	PSH	3.8×10^6	$\leqslant 1.2$	Deuterated cyclohexane, $T = \Theta = 40°C$	5×10^{-3}
C	PSD	1.1×10^6	1.17	Bulk of PSH chains of same molecular mass	2.05×10^{-2}

terated polystyrene dispersed in an undeuterated polystyrene matrix in the small-momentum range. This indicated that the size of polymer chains is identically the same in the bulk material and in a Θ solvent, over two decades of molecular masses. Thus for distances r comparable to the overall size of the chain, the scaling law (1) applies with $\nu = \frac{1}{2}$ in these two environments. The question is what happens at distance r smaller than $(\langle R^2 \rangle)^{1/2}$. The theory predicts the same value of ν for both Eqs. (2) and (3), for a chain in a good solvent. When the chain is in the bulk material there is no theoretical necessity for identical ν values in both momentum ranges. The question is open and can be answered by an experiment.

Here we report experimental values of ν obtained in the intermediate momentum range for polystyrene chains in three environments: carbon disulfide, cyclohexane at the Flory temperature, and bulk material. The scattering experiment was performed at the wave guide channel of reactor EL3 at Saclay. The wavelength of the incident beam was slected by a crystalline monochromator at $\lambda = 4.62 \pm 0.04$ Å. Collimation gave an angular divergence of less than 30 min of arc. The scattered intensity was recorded for angles θ so chosen that the momentum transfer $[q = (4\pi/\lambda)\sin\theta]$ had values between 3×10^{-2} and 10^{-1} Å^{-1}. This range satisfies the double inequality (4) as indicated by the characteristics of the samples in Table I. The intensity I scattered by the samples containing the labeled chains was decomposed into three parts, which are, respectively, the coherent signal of the labeled chains, the incoherent signal of the labeled chains, and the intensity scattered by the solvent (or matrix). The two last contributions were measured separately and subtracted from I, in order to determine the signal of interest. The values of ν were obtained by a best-fit method[8] between experimental data and Eq. (5). The plot of $\ln s^{-1}(q)$ as a function of $\ln q$ is shown in Fig. 1.

The experiment of the deuterated polystyrene (PSD) in CS$_2$ at room temperature (good solvent) gives a value for ν^{-1} equal to 1.68 ± 0.06. The nondeuterated polystyrene chain (PSH) in deuterated cyclohexane at $T = 40°C$ yields $\nu^{-1} = 2.06 \pm 0.08$. The same result is obtained for a deuterated polystyrene chain in an undeuterated polystyrene matrix, $\nu^{-1} = 2.01 \pm 0.08$.

The problem of multiple scattering effects was considered in the following way. After interpretation of the data, the experiment was carried out on a sample with thickness one half of the original sample. No modification of the exponents was observed. The fact that this experiment consists in comparing the effects of three environments also eliminates systematic errors such as those
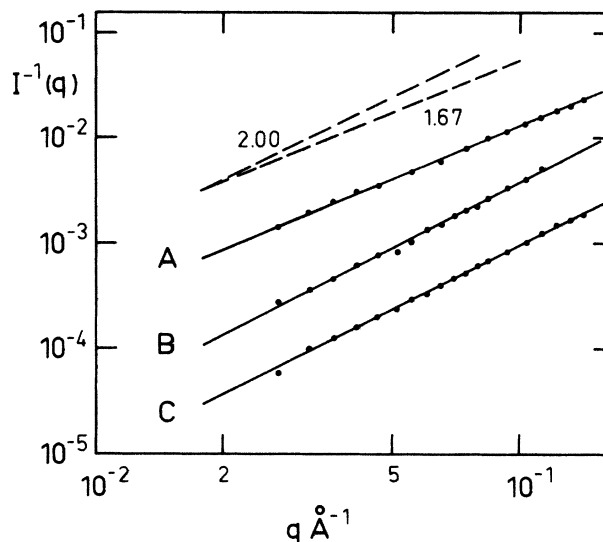

FIG. 1. Log-log plot of the inverse scattered intensity as a function of the momentum transfer q. The full points are experimental data for a polymer in a good solvent, curve a, a Θ solvent, curve b, and the bulk polymer, curve c. The full lines are the calculated curves. The dashed lines are reference straight lines with slopes of 2.00 and $\frac{5}{3}$.

introduced by multiple scattering.

The precision obtained in the intermediate-momentum range compares favorably with the one obtained in the small-momentum range. In an experiment based on Eq. (5) only one sample is needed and the result is independent of polydispersity. In contrast, the experimental determination of ν derived from relation (2) requires several samples, with well-defined molecular masses.

The value of ν obtained in good solvent in this experiment is consistent with results obtained using relation (2) and other experimental methods. The value of ν obtained in the bulk indicates the absence of an effective excluded-volume interaction over a large range of distances. A remarkable fact is that there is no deviation from the $1/q^2$ law for q values as high as $0.1 \ \text{Å}^{-1}$.

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Parametric Excitation of Alfvén and Ion Acoustic Waves

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The threshold is derived for parametric excitation of Alfvén waves in a uniform plasma in which the background magnetic field is modulated sinusoidally in time. Including the plasma pressure we show that ion acoustic waves can also be excited directly. Both cases are absolutely unstable for any interaction length. Finally, we analyze the subsequent decay of the excited Alfvén wave into an ion acoustic and another Alfvén wave.

The growth rate for the parametric excitation of Alfvén waves was calculated by Vahala and Montgomery.[1] The Alfvén waves were excited by oscillating the background magnetic field. This phenomenon was later observed experimentally by Lehane and Paoloni.[2] The purpose of this Letter is threefold; first, we derive the Alfvén wave instability threshold; second, we show that by taking the plasma pressure into account not only Alfvén waves may be excited directly but also ion acoustic waves; third, we obtain the threshold for the subsequent decay of the finite amplitude Alfvén wave so excited into an ion acoustic wave and another Alfvén wave.

We shall follow Vahala and Montgomery[1] and use simple magnetohydrodynamic equations with the addition of a plasma pressure term and resistivity. These equations are the following:

$$\rho \, \partial \vec{v}/\partial t + \rho (\vec{v} \cdot \nabla)\vec{v} = - \nabla p + \mu_0^{-1}(\nabla \times \vec{B}) \times \vec{B}; \quad (1)$$

$$\partial \vec{B}/\partial t = \nabla \times (\vec{v} \times \vec{B}) - (\eta/\mu_0)\nabla \times (\nabla \times \vec{B}); \quad (2)$$

$$\partial \rho/\partial t + \nabla \cdot (\rho \vec{v}) = 0; \quad (3)$$

we take the equation of state to be $p = c_s^2 \rho$.

The model considered by Vahala and Montgomery[1] was that of a uniform column of plasma of radius a with a uniform magnetic field pointing in the z direction. The source of energy for the