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SHADOWGRAPH STUDY OF GRADIENT DRIVEN FLUCTUATIONS

Gennady Nikolaenko and David S. Cannell
Department of Physics
UC Santa Barbara
Santa Barbara, CA 93106

ABSTRACT

A fluid or fluid mixture, subjected to a vertical temperature and/or concentration gradient in a gravitational field, exhibits greatly enhanced light scattering at small angles [1-3]. This effect is caused by coupling between the vertical velocity fluctuations due to thermal energy and the vertically varying refractive index. Physically, small upward or downward moving regions will be displaced into fluid having a refractive index different from that of the moving region, thus giving rise to the enhanced scattering [4]. The scattered intensity is predicted [5-7] to vary with scattering wave vector $\mathbf{q}$, as q^{-4}, for sufficiently large $\mathbf{q}$, but the divergence is quenched by gravity [8] at small $\mathbf{q}$. In the absence of gravity, the long wavelength fluctuations responsible for the enhanced scattering are predicted to grow until limited by the sample dimensions [9, 10]. It is thus of interest to measure the mean-squared amplitude of such fluctuations in the microgravity environment for comparison with existing theory and ground based measurements.

The relevant wave vectors are extremely small, making traditional low-angle light scattering difficult or impossible because of stray elastically scattered light generated by optical surfaces. An alternative technique is offered by the shadowgraph method, which is normally used to visualize fluid flows, but which can also serve as a quantitative tool to measure fluctuations [11, 12]. A somewhat novel shadowgraph apparatus and the necessary data analysis methods will be described. The apparatus uses a spatially coherent, but temporally incoherent, light source consisting of a super-luminescent diode coupled to a single-mode optical fiber in order to achieve extremely high spatial resolution, while avoiding effects caused by interference of light reflected from the various optical surfaces that are present when using laser sources.

Results obtained for a critical mixture of aniline and cyclohexane subjected to a vertical temperature gradient will be presented. The sample was confined between two horizontal parallel sapphire plates with a vertical spacing of 1 mm. The temperatures of the sapphire plates were controlled by independent circulating water loops that used Peltier devices to add or remove heat from the room air as required.

For a mixture with a temperature gradient, two effects are involved in generating the vertical refractive index gradient, namely thermal expansion and the Soret effect, which generates a concentration gradient in response to the applied temperature gradient. For the aniline/cyclohexane system, the denser component (aniline) migrates toward the colder surface. Consequently, when heating from above, both effects result in the sample density decreasing with altitude and are stabilizing in the sense that no convective motion occurs regardless of the magnitude of the applied temperature gradient. The Soret effect is strong near a binary liquid critical point, and thus the dominant effect is due to the induced concentration gradient. The results clearly show the divergence at low $\mathbf{q}$ and the predicted gravitational quenching. Results obtained for different applied temperature gradients at varying temperature differences from the critical temperature, clearly demonstrate the predicted divergence of the thermal diffusion ratio.

Thus, the more closely the critical point is approached, the smaller becomes the temperature gradient required to generate the same signal.

Two different methods have been used to generate pure concentration gradients. In the first, a sample cell was filled with a single fluid, ethylene glycol, and a denser miscible fluid, water, was added from below thus establishing a sharp interface to begin the experiment. As time went on the two fluids diffused into each other, and large amplitude fluctuations were clearly observed at low $\mathbf{q}$. The effects of gravitational quenching were also evident. In the second method, the aniline/cyclohexane sample was used, and after applying a vertical temperature gradient for several hours, the top and bottom temperatures were set equal and the thermal gradient died on a time scale of seconds, leaving the Soret induced concentration gradient in place. Again, large-scale fluctuations were observed and died away slowly in amplitude as diffusion destroyed the initial concentration gradient.

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Gradient

David Cannell and Gennady Nikolaenko

UC Santa Barbara

Marzio Giglio, Alberto Vailati and Fabrizio Croccolo

University of Milan

William Meyer

NASA Glenn Research Center

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Motivation

Basic Physics:

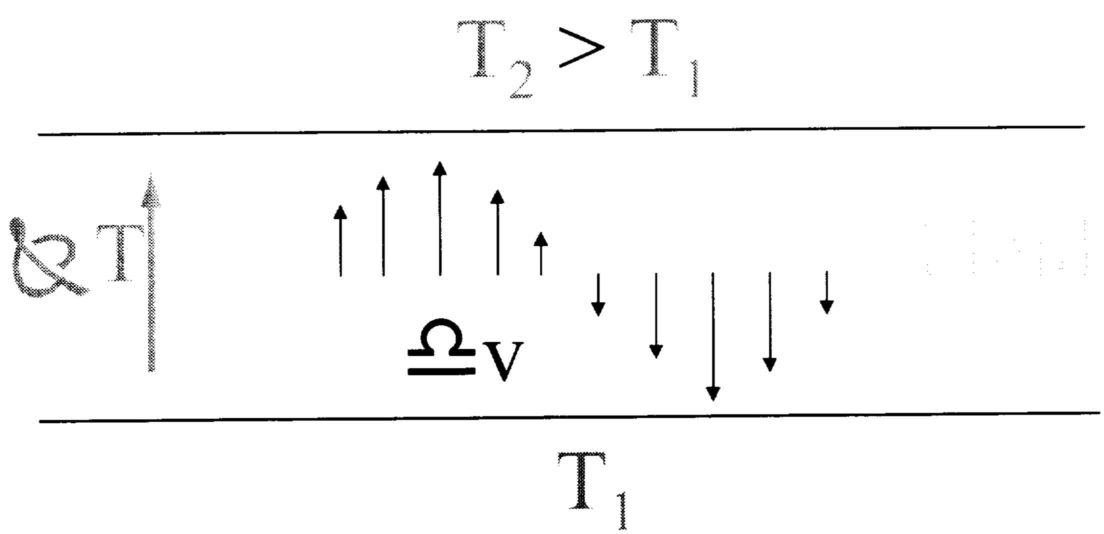
$$G(\mathbf{R},\tau) = < \delta\rho(\mathbf{R},\tau)\delta\rho(0,0) >$$

$$S(\mathbf{q},\omega) = FT[G(\mathbf{R},\tau)]$$

Normally short ranged and decays quickly, except near Critical Points

Becomes very long ranged when a fluid is subjected to a temperature gradient.

This effect is suppressed by gravity.



Thermally driven velocity fluctuations force colder more dense fluid into warmer less dense regions, and vice versa.

Result: Mode-mode coupling greatly enhances long-range density fluctuations.

Combined Physical Picture

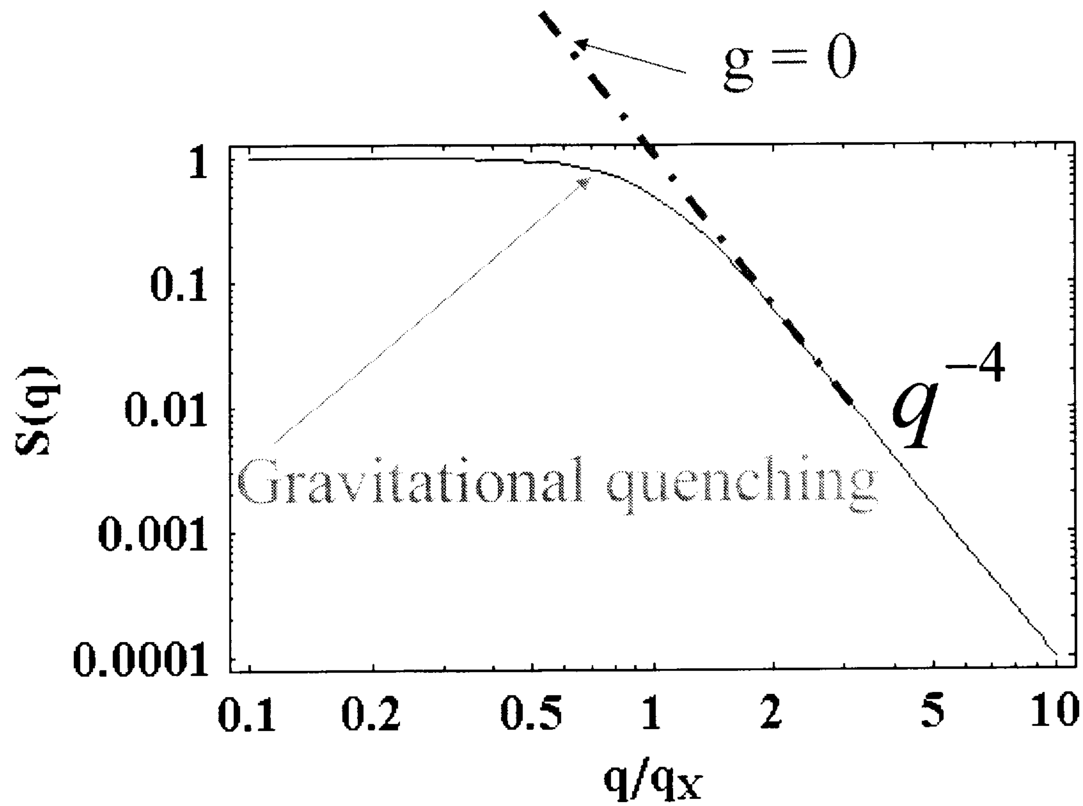
Short wavelength fluctuations are suppressed by viscosity and thermal diffusion.

$$\tau = 1/(D_T q^2),\ 1/(\nu q^2)$$

$$q = 2\pi / \Lambda$$

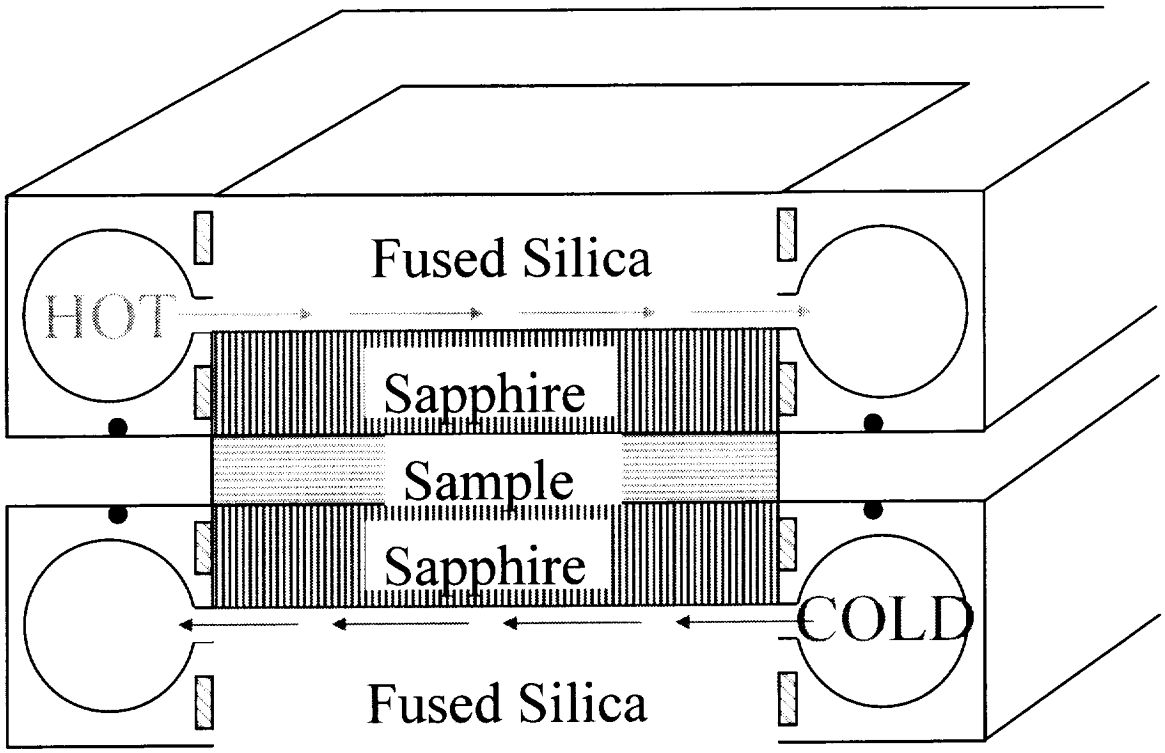
Long wavelength fluctuations last long enough for buoyancy to be important.

Quenched by gravity.



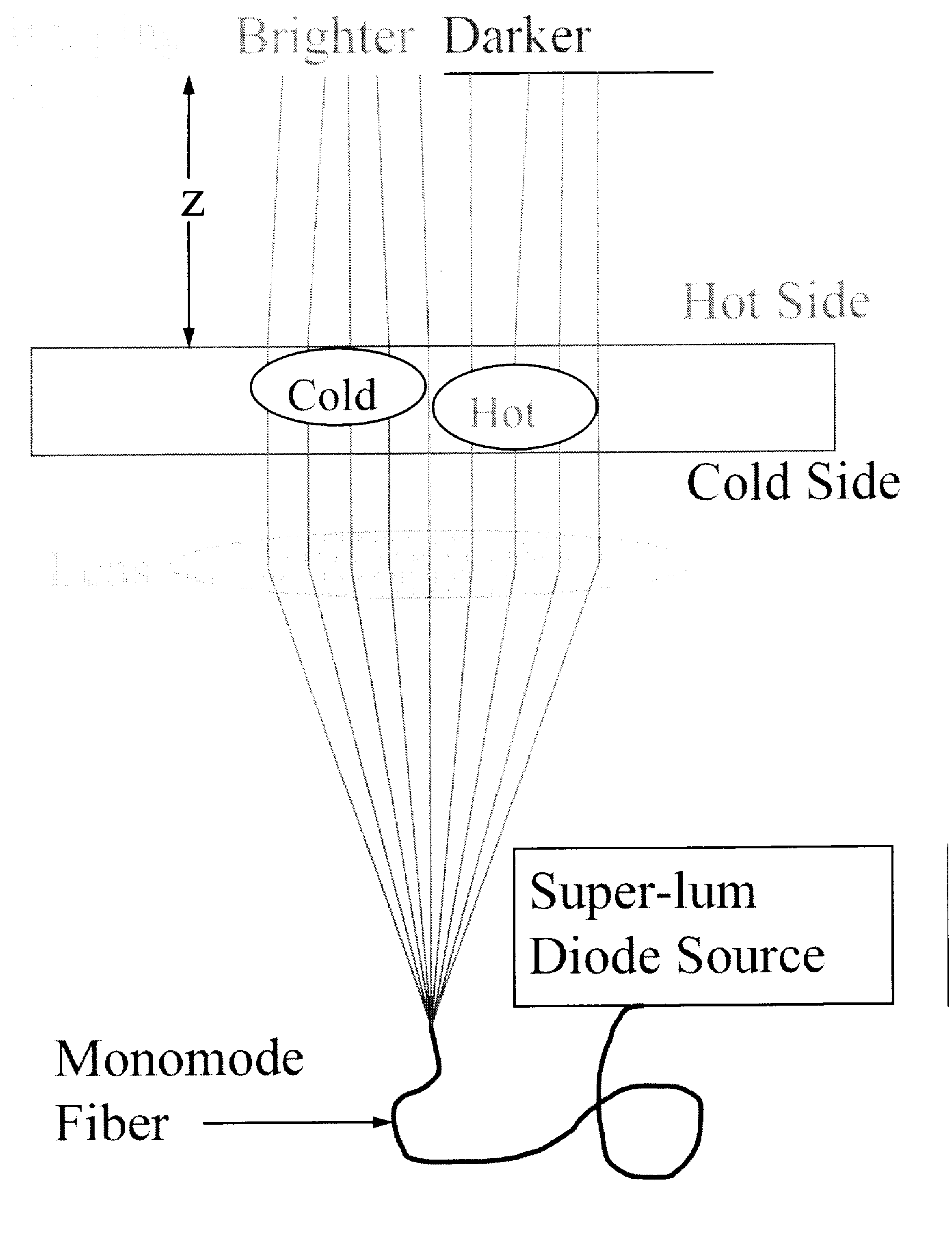
The structure factor S(q), predicted for a bulk fluid subjected to a thermal gradient.

Similar results hold for mixtures with a temperature and/or a concentration gradient.

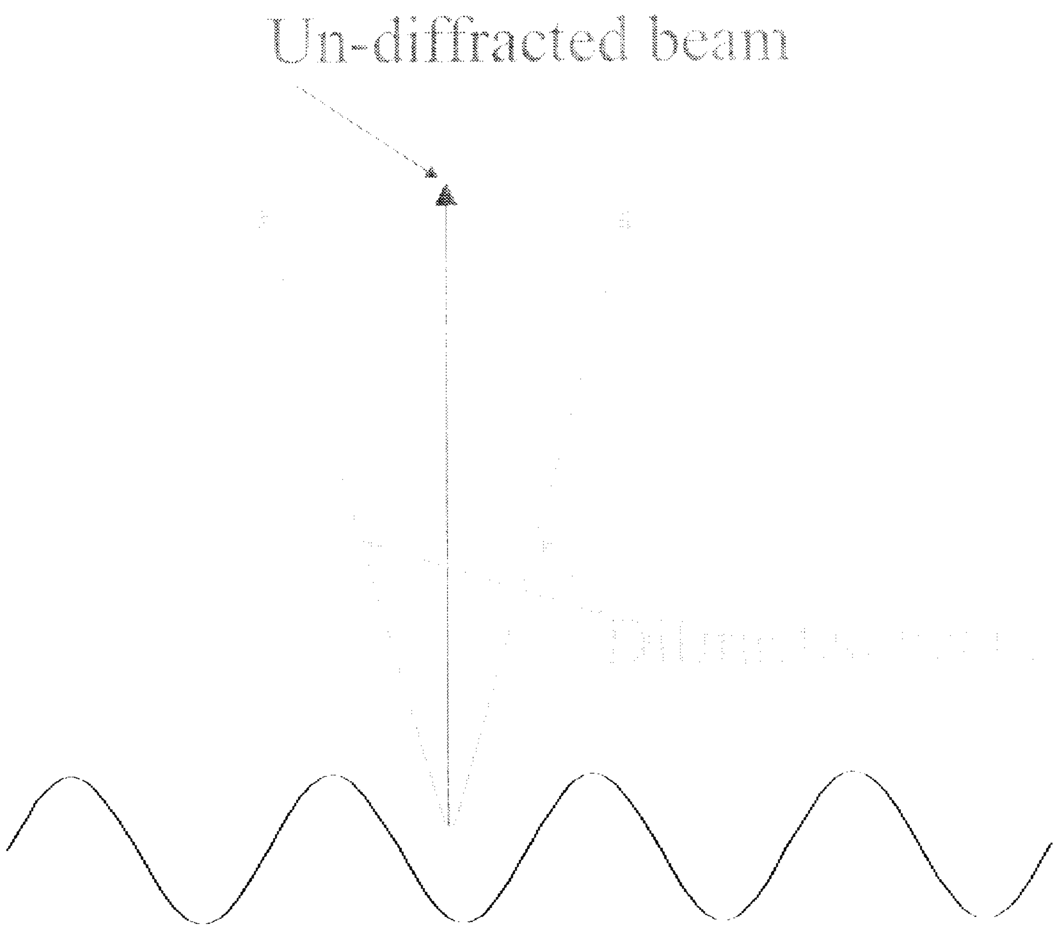


Cross section of the sample cell used to impose controlled thermal gradients on fluids.

Small angle scattering?
Interferometry?
Quantitative Shadowgraphy?


Brighter
Darker
Z
Hot Side
Cold
Hot
Cold Side
Lens
Super-lum
Diode Source
Monomode
Fiber

Physical Optics Treatment



Wave fronts emerging from the sample are phase modulated.

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2. Ratios of images quantify the fluctuations, $\delta I(x,y)$.

3. Fourier analysis of the ratio images reveals the spatial power spectrum of the fluctuations, $\delta I^2(\mathbf{q})$.

$$\delta I^2(\mathbf{q}) = A\ Sin^2(\mathbf{q}^2 z/(2k_o))\ S(\mathbf{q})$$

$$S(\mathbf{q}) = FT[G(\mathbf{R})]$$

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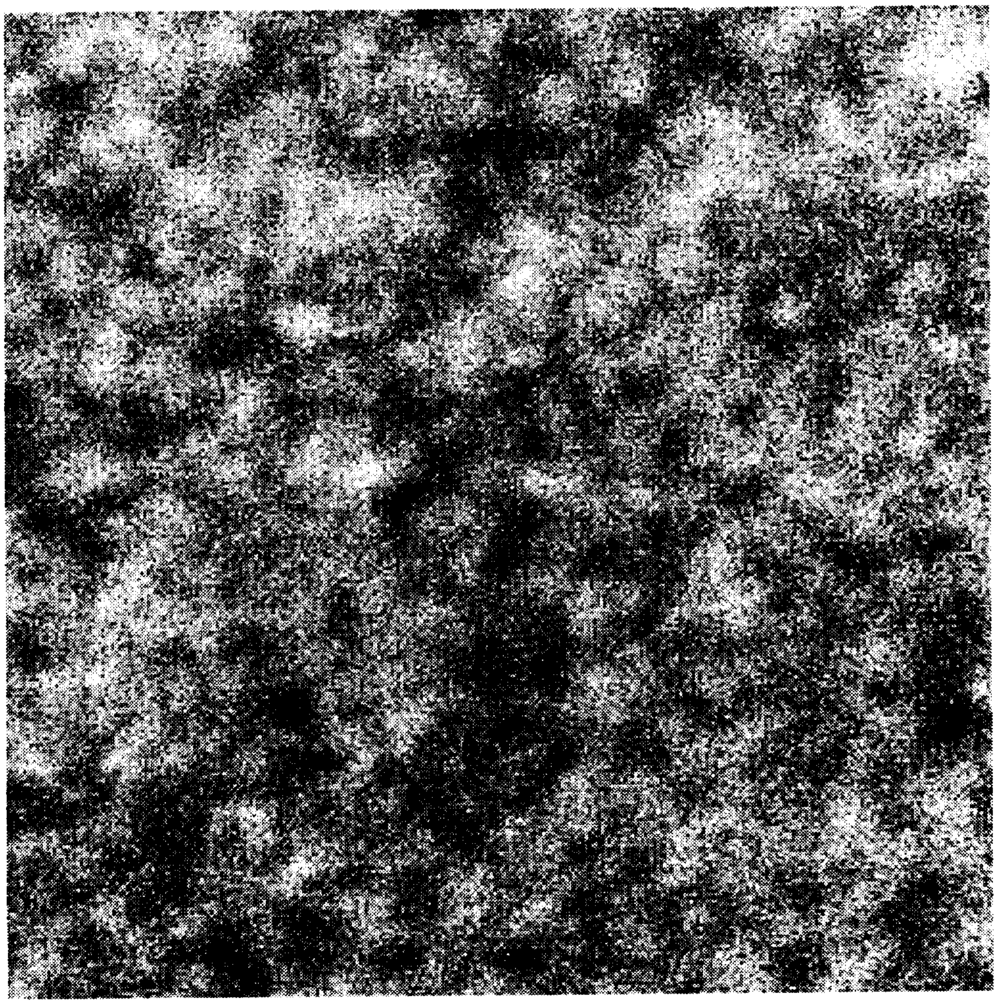
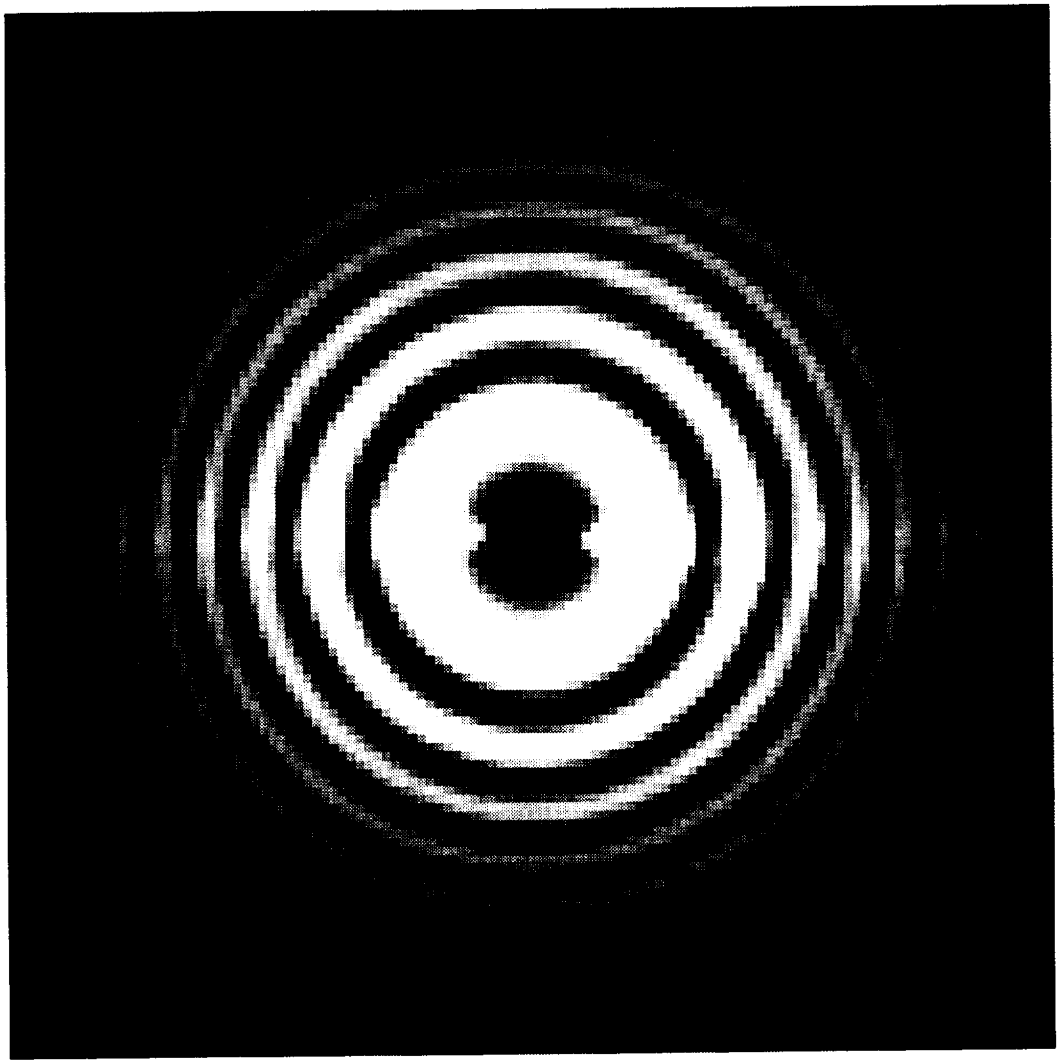
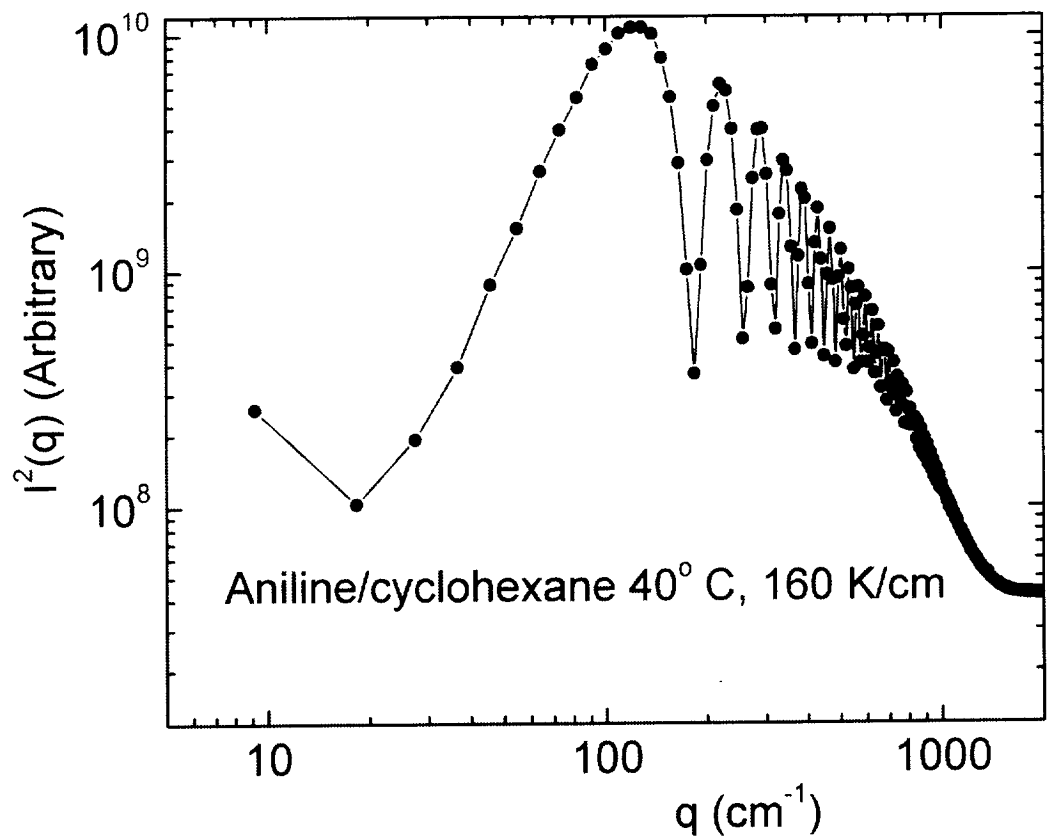


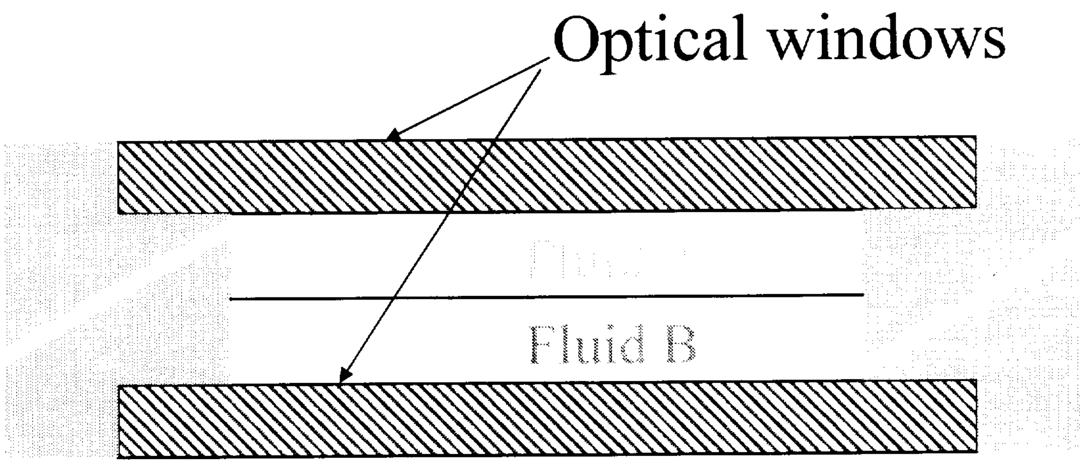
Ratio of two images taken 1 second Apart for aniline/cyclohexane at 40^{o} C with a gradient of 160 K/cm.



$I^2(\mathbf{q})$ for a 1 mm thick sample of 47 Wt.% aniline in cyclohexane at 40^o C with a gradient of 160 K/cm. Measured at z = 18 cm.

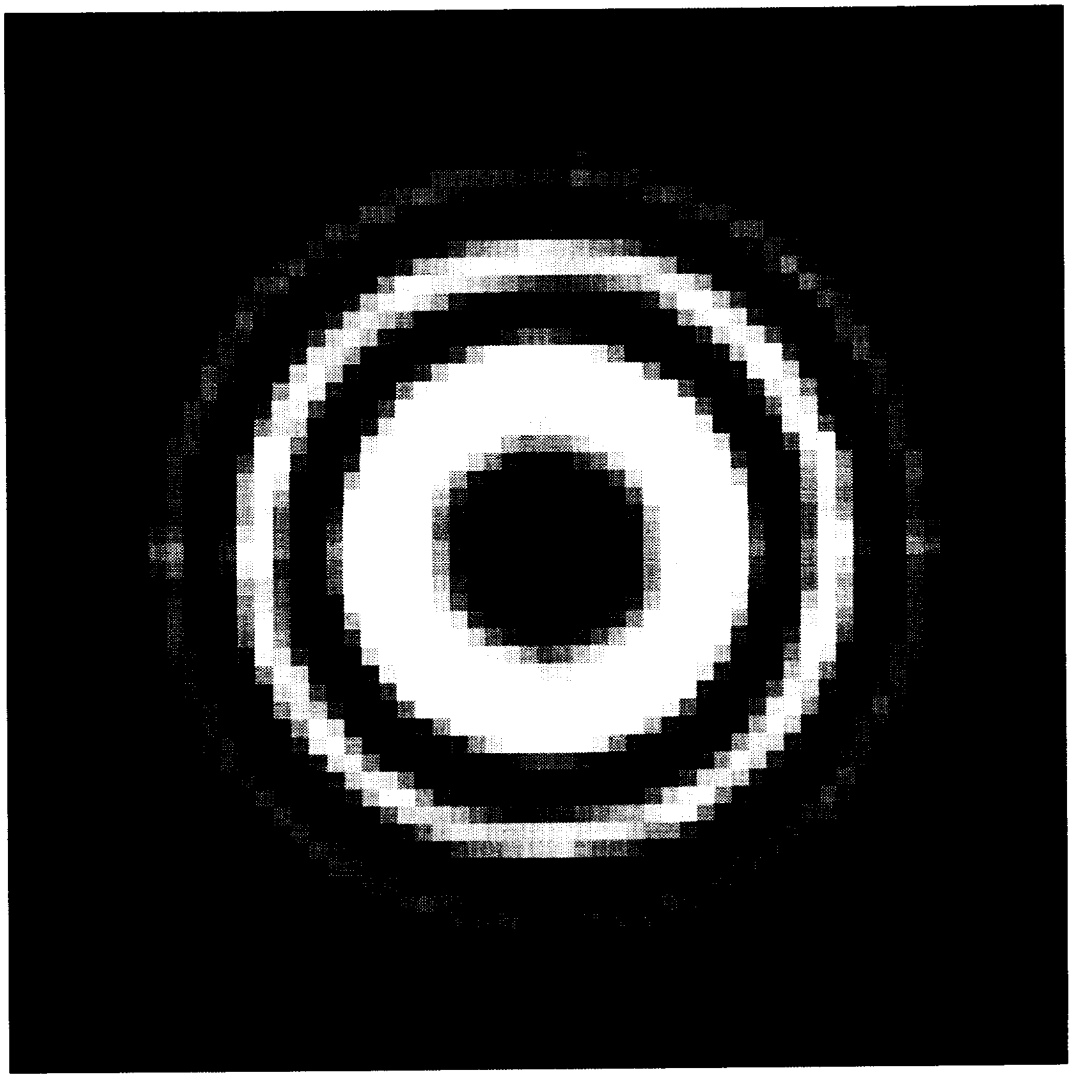


Log-Log plot of the spatial power spectrum of the fluctuations for the aniline/cyclohexane sample.

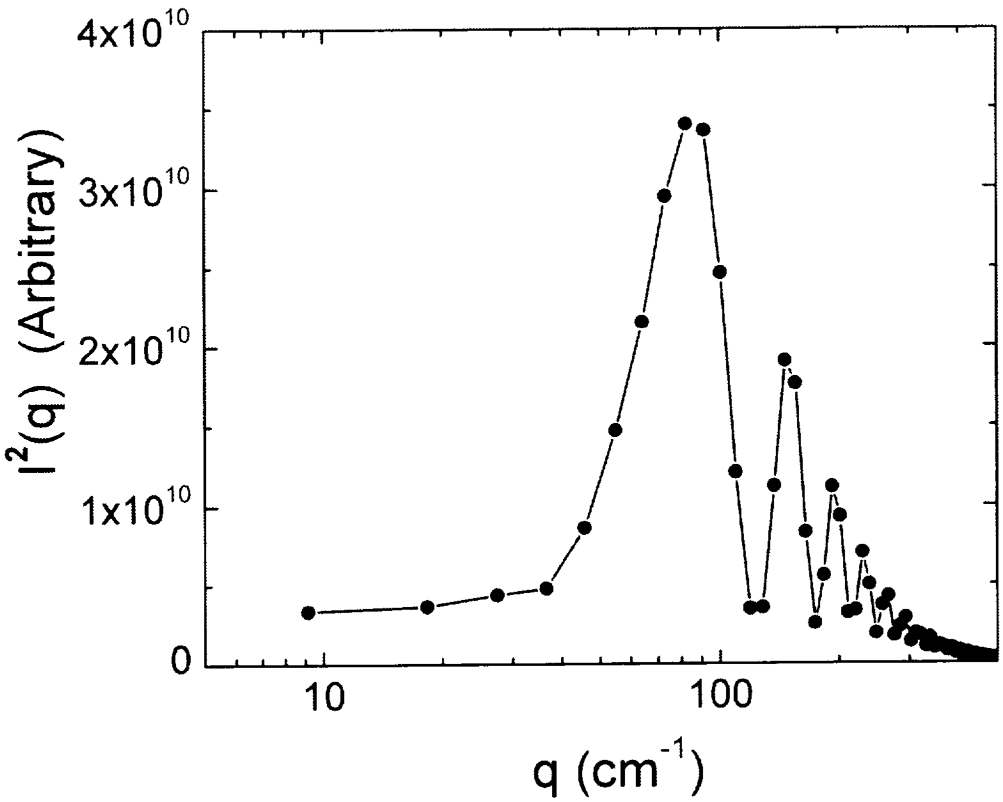


Cross section of the sample cell used to layer one fluid upon another, creating a flat interface

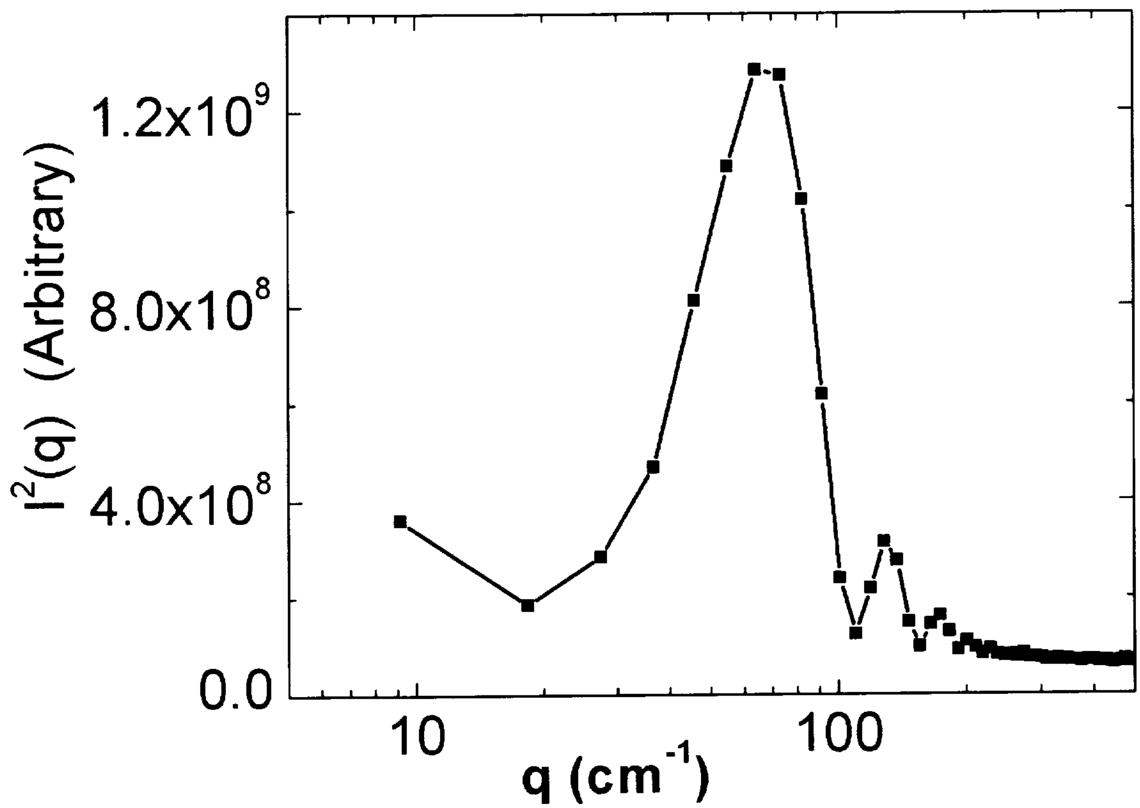
A large concentration gradient is present for many hours, during the process of free diffusion.



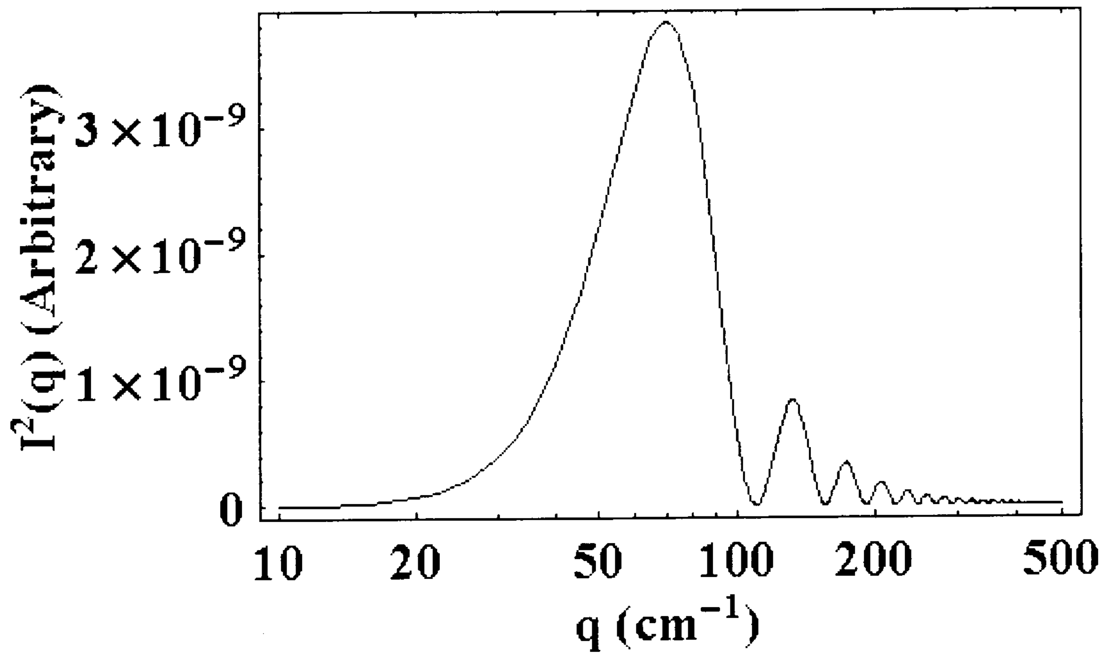
$I^2(\mathbf{q})$ for water diffusing into ethylene glycol.



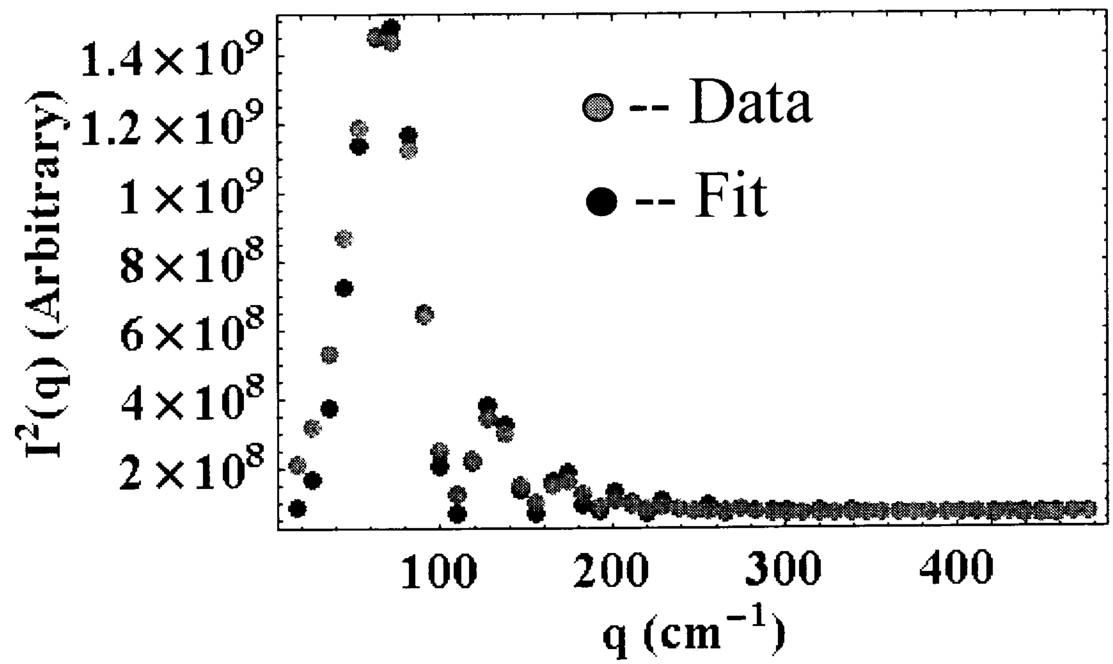
Log-Linear plot of the spatial power spectrum of the fluctuations for water diffusing into ethylene glycol.



Log-Linear plot of the spatial power spectrum of the fluctuations for a 1 mm thick toluene sample with a gradient of 200 K/cm.



Log-Linear plot of the shadowgraph signal expected for a 1mm thick toluene sample subjected to a gradient of 200 K/cm, and with an imaging distance of 50 cm.



Results of fitting existing theory to the data for toluene. The theory* includes both gravity and finite-size effects.

* José Ortiz de Zárate and Jan. V. Sengers, Preprint (2002).