

GRADFLEX, Fluctuations in Microgravity

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Abstract: We present the results of experimental investigations of gradient driven fluctuations induced in a liquid mixture with a concentration gradient and in a single-component fluid with a temperature gradient. We also describe the experimental apparatus being developed to carry out similar measurement under microgravity conditions.

The GRADient Driven FLuctuation EXperiment (GRADFLEX) involves the investigation of fluctuations induced in simple fluids and in binary mixtures by imposing a macroscopic temperature or concentration gradient under microgravity conditions. Recent experiments have shown that giant nonequilibrium fluctuations are present during diffusion processes in liquid mixtures and in the presence of a heat flux through a fluid. These fluctuations occur at all length scales between the microscopic and a macroscopic scale set by the sample dimensions. The fluctuations are due to corrugations in the diffusing front, whose fractal properties explain the presence of fluctuations involving all length scales. The fluctuations are generated by coupling between velocity fluctuations and the macroscopic gradient (concentration or temperature) which drives the flux. The amplitude of these fluctuations diverges as q^{-4} , where q is the wave vector of the fluctuation. Long wavelength fluctuations are stabilized by gravity, which quenches the q^{-4} divergence at the smallest wave vectors.

On Earth, gravity suppresses the long wavelength fluctuation below a typical cutoff wave vector. The aim of the GRADFLEX project is to investigate these fluctuations in the absence of gravity, where the long wavelength fluctuations are no longer predicted to be stabilized by gravity, and to compare the results with those obtained on Earth. Many materials science processes (for example, crystallization and growth of materials) are performed in microgravity because of advantages expected from the absence of convection. However, the presence of nonequilibrium fluctuations could lead to the unexpected presence of large scale inhomogeneities that could impair processing under microgravity conditions.

Two prototype systems to guide the engineering of flight hardware have been developed, one in the Optics and Microgravity Laboratory at the University of Milan by the Istituto Nazionale per la Fisica della Materia (INFM) and one in the Physics Department at the University of California at Santa Barbara (UCSB). Both systems use the shadowgraph method to measure the fluctuations. The system developed at INFM is devoted to the investigation of concentration fluctuations occurring during a Soret induced mass diffusion process, while that developed at UCSB is designed to investigate fluctuations induced by a thermal gradient in a single-component fluid. The project is scheduled for flight in 2006 onboard the Russian satellite capsule FOTON M3.

The current sensitivity of the shadowgraph method is now sufficiently developed to measure the scattering from the fluctuations, both on Earth and in microgravity. Samples are contained between parallel sapphire windows to provide the necessary thermal boundary conditions. The fluctuations give rise to phase perturbations in the wavefronts of a beam of light passing through the sample, resulting in measurable intensity modulation a sufficient propagation distance beyond the sample. This intensity modulation is time-dependent, and it can be analyzed to obtain both the mean squared amplitude of the fluctuations $S(q)$, and their power spectrum $S(q,\omega)$, for wave vectors as small as 20 cm^{-1} . Thus the method is useful well below the range where small angle light scattering is typically

impossible because of stray light and other effects. The resulting data are the product of $S(q)$ and the shadowgraph transfer function $T(q) = \text{Sin}^2(q^2 z / 2k_o)$.

We will present experimental results obtained on Earth for both $S(q)$ and $S(q, \omega)$ for polystyrene diffusing in toluene (INFM), and for thermal fluctuations in carbon disulfide (CS_2) with a temperature gradient (UCSB). We will also describe our current designs for the flight qualified measuring systems. Some examples of the data we have been able to obtain for $S(q)$ and $S(q, \omega)$ for the two systems on Earth are shown below.

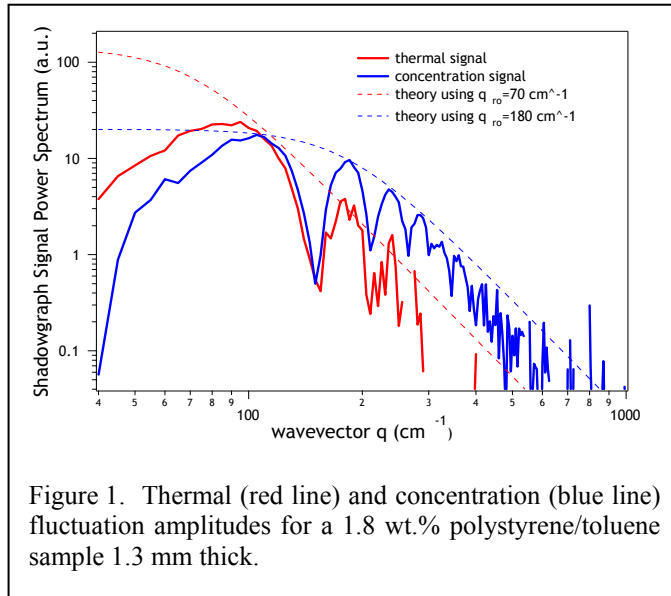


Figure 1. Thermal (red line) and concentration (blue line) fluctuation amplitudes for a 1.8 wt.% polystyrene/toluene sample 1.3 mm thick.

Figure 1 shows results for $T(q)S(q)$ for a 1.3 mm thick sample consisting of 1.8 wt.% of 9100 M_W polystyrene in toluene. The thermal and concentration fluctuations were measured separately in the following manner. A temperature gradient of 135 K/cm was applied during about 120 s, after which the thermal gradient was fully established, while the concentration gradient caused by the Soret effect had only begun to develop, because the relevant time constant is about 940 s. A set of 100 images taken at the rate of 1/s in the interval between 140 s and 240 s after beginning to establish the gradient were analyzed to obtain the results for thermal fluctuations. A similar set of images taken 6900 s after the start of the experiment were analyzed to obtain the results for the concentration fluctuations.

Theoretically, the thermal fluctuations should be quenched by gravity only below about 70 cm^{-1} ,

and the dashed curve for $S(q)$ shows that the data are quite consistent with this prediction. For concentration fluctuations the relevant wave vector is predicted to be about 226 cm^{-1} , while the data seem more consistent with a value of about 180 cm^{-1} as shown by the dotted curve for $S(q)$ for the concentration fluctuations.

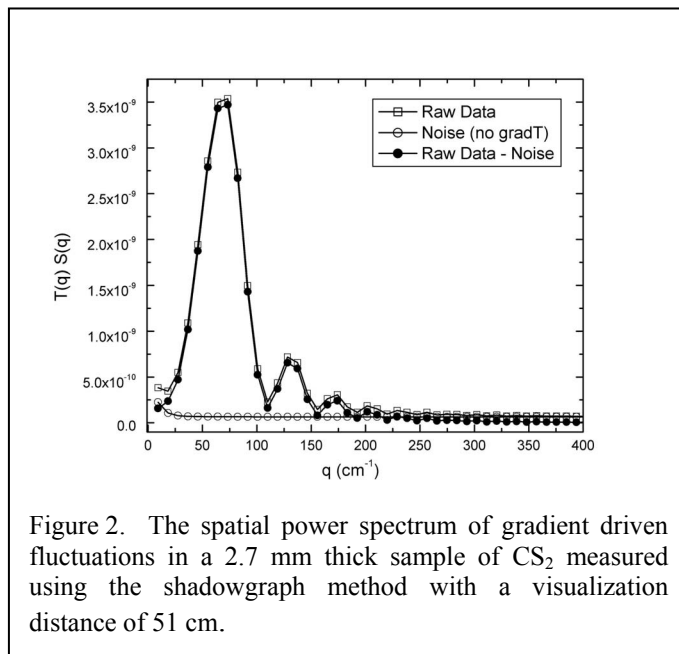


Figure 2. The spatial power spectrum of gradient driven fluctuations in a 2.7 mm thick sample of CS_2 measured using the shadowgraph method with a visualization distance of 51 cm.

Figure 2 shows results for $T(q)S(q)$ for a CS_2 sample. The data were obtained with a 23 K temperature difference applied across the 2.7 mm thick sample, which was held at a mean temperature of $32.5 \pm 0.5 \text{ }^\circ\text{C}$. The upper surface of the sample was hotter than the lower surface thus preventing any convective motion. Before applying the gradient we measured a background spectrum (open circles), primarily caused by camera noise, and this spectrum was subtracted from the spectrum measured with the gradient applied (open squares) to obtain the result shown by the solid circles. A total of 4000 images taken at 2 second intervals were analyzed to obtain the results shown. As can be seen, the amplitude of the fluctuations grows strongly with decreasing wave vector. Although the appearance of the data is complicated by the presence of the shadowgraph transfer function, this is not much of a problem in practice, because the data can be fit readily by the product $T(q)S(q)$.

The project is being carried out by INFM in collaboration with UCSB. The ground based research is being jointly supported by ASI and NASA. The flight hardware and flight expenses will be supported by ESA.