Grant Title: Gradient Driven Fluctuations

Final Technical Progress Report

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We have worked with our collaborators at the University of Milan (Professor Marzio Giglio and his group-supported by ASI) to define the science required to measure gradient driven fluctuations in the microgravity environment. Such a study would provide an accurate test of the extent to which the theory of fluctuating hydrodynamics can be used to predict the properties of fluids maintained in a stressed, non-equilibrium state. As mentioned above, the results should also provide direct visual insight into the behavior of a variety of fluid systems containing gradients or interfaces, when placed in the microgravity environment.

With support from the current grant, we have identified three key systems for detailed investigation. These three systems are:

1. A single-component fluid to be studied in the presence of a temperature gradient,
2. A mixture of two organic liquids to be studied both in the presence of a temperature gradient, which induces a steady–state concentration gradient, and with the temperature gradient removed, but while the concentration gradient is dying by means of diffusion, and
3. Various pairs of liquids undergoing free diffusion, including a protein/buffer solution and pairs of mixtures having different concentrations, to allow us to vary the differences in fluid properties in a controlled manner.

The sample choices are now very well defined for the first two categories, namely the single-component fluid should be carbon disulphide (CS_2) if possible, or toluene if this is preferable for flight, and the mixture should be an equimolar solution of n-hexane and toluene. For the third category, great flexibility in the choice of sample is possible, so the final choice can be made in terms of convenience/safety, interest to other microgravity efforts, etc.

In addition to identifying the specific samples, we have defined the spatial scale (hot and cold surface separation) of the sample cells that will be required to avoid significant boundary effects as being about 10 mm. In turn, this requirement fixes the required range of wave vector over which the fluctuations must be measured as extending from about 1 cm^{-1} up to several hundred cm^{-1}. Three possible experimental techniques have been considered and the two most promising ones, namely Small Angle Light Scattering, and Quantitative Shadowgraphy, have been assessed directly. The third possibility is Interferometry; however, the expected phase modulations are so small (of order $\lambda/500$) as to defy the use of direct interferometric methods. The wave vector range required extends to such small wave vectors that we have been able to eliminate Small Angle Light Scattering as a viable method, leaving Quantitative Shadowgraphy as the method of choice.

We have worked out a new theoretical treatment of the Shadowgraph [26], treating the method in terms of physical optics (previous theory was based on geometrical optics and could not provide predictions for fluctuations). The new theory enables the shadowgraph to be used as a quantitative measurement tool.

We have designed and constructed a sensitive shadowgraph instrument together with a thermal gradient cell, and have demonstrated by experiment that we can measure the shadowgraph signal from single-component fluids (toluene and CS_2) down to a wave vector of $10\ \mathrm{cm}^{-1}$, the limit set by the illuminated area of our current instrument, and up to over $1000\ \mathrm{cm}^{-1}$. To our knowledge, this is the first instance in which direct measurement of fluctuations in a single-component fluid heated from above have been made at such low wave vector. Previous measurements for single-component fluids heated from above have been made using dynamic light scattering, and they extend down to about 1500 cm^{-1}; thus the new method extends by over a factor of 100 the accessible wave-vector range. Furthermore, the newly available range is precisely that of most interest for the microgravity environment.

A two-dimensional image showing the shadowgraph results for a CS_2 sample 2.8 mm thick with an applied gradient of 100 K/cm is shown in Figure 1. In this representation, the brightness indicates the mean-squared amplitude of the fluctuations $S(\mathbf{q})$, multiplied by the shadowgraph transfer function $T(\mathbf{q})$. The image has been arranged so that $\mathbf{q}=0$ is located in the center, with $|\mathbf{q}|$ increasing with distance from the center. The transfer function $T(\mathbf{q}) = Sin^2(|\mathbf{q}|^2 z/2k_o)$, where z is the viewing distance and k_o is the wave vector of the light, and thus it has zeroes as a function of $|\mathbf{q}|$. These zeroes show up as dark rings in what would otherwise be a monotonically decreasing function of $|\mathbf{q}|$. This image is the result of processing over 600 individual shadowgraph images taken at intervals of one second. To

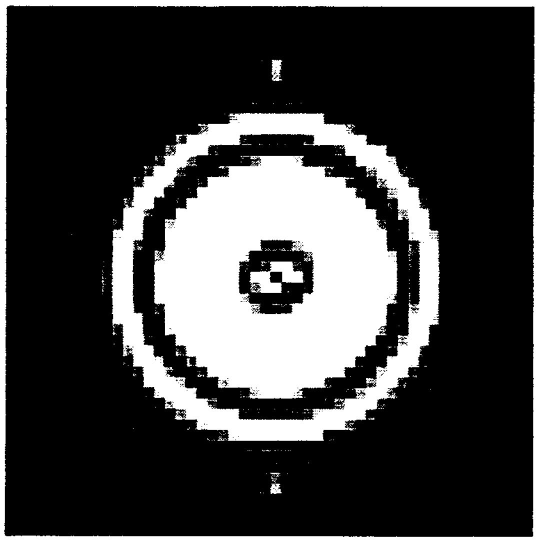

Fig. 1 Shadowgraph results for CS_2.

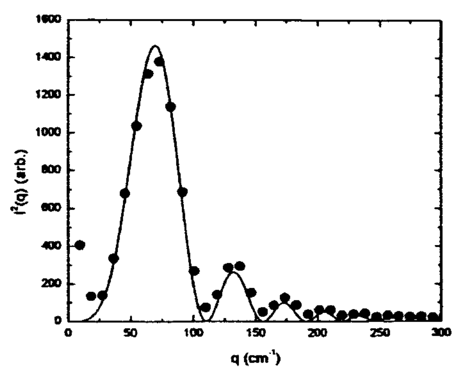

Fig. 2 Azimuthal average of the data shown in Fig. 1 together with theory.

our knowledge these are the first such measurements made.

As would be expected for an isotropic sample, the signal depends only on $|\mathbf{q}|$ and not on its direction, and thus it may be azimuthally averaged. The result of doing this is shown in Figure 2. The solid curve is the signal calculated using tabulated fluid properties for CS_2, multiplied by an arbitrary scale factor, **but with no other adjustable parameters**. Considering that we have not even subtracted the system noise floor, the data are quite well accounted for by current theory, which includes no-slip boundary conditions. The signal should decay smoothly to zero as $|\mathbf{q}|$ approaches zero, because of the transfer function, however there is a spurious rise at the 2 lowest $|\mathbf{q}|$ values measured, which we believe is due to imperfections in the sample cell. Considerable improvement can be made in this area. The work described above was carried out by the UCSB group with NASA support.

Professor Giglio and his group have focused their attention on the development of methods whereby a high-quality planar interface may be formed between two liquids, and/or two solutions in the microgravity environment. This problem is particularly difficult because any mechanical method, such as removal of a barrier, is certain to cause flow, thus introducing unwanted long wavelength fluctuations. The method chosen for development is that of introducing the two solutions continuously into a tubular apparatus, with each solution entering at one end of the cylindrical tube. At the center of the apparatus, there is a small circumferential crack perpendicular to the axis through which fluid is evacuated. The apparatus they have constructed enables the formation of a very smooth planar interface, **even when used with solutions in which the upper fluid is slightly denser than the lower one**, a situation that is intrinsically unstable in the Earth's gravitational field. We find this very encouraging and hope that this method will prove broadly useful in the microgravity environment.

In addition to the research described above, we have prepared a draft Science Requirements Document, spelling out in some detail the measurements of interest, the samples we think are most suitable for study, and the measurement technique we think is most likely to meet with success.

This grant allowed 4 undergraduates to participate in research. Their names are: Ian Bruce, Alexia Fu, Hunter McDaniel and Christopher Takacs.

There was one publication:

Steven P. Trainoff and David S. Cannell, "Physical Optics Treatment of the Shadowgraph," *Physics of Fluids*, **14**, 1340 (2002).

A post doc, Gennady Nikolaenko was employed for most of the grant period.