Non-equilibrium fluctuations on earth and in micro-gravity. 
The GRADFLEX experiment.

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We present the results of measurements of giant nonequilibrium fluctuations in a single component fluid (density fluctuations) and a mixture (concentration fluctuations) driven by applied temperature gradients, both on Earth and in space. Flight data were obtained during the September 2007 FOTON M3 mission. Spatial power spectra obtained using the shadowgraph method, during flight, confirm that the asymptotic $\nabla T^2 q^{-4}$ behaviour extends to such low wave vector $q$, as to be limited by the sample thickness. Quantitative comparison with theory is provided, and is generally quite good. Temporal sequences of shadowgraph images for the mixture, both on Earth and during flight will be presented to emphasize the dramatic differences. Fluctuation lifetimes of thousands of seconds were observed during flight. The rugged but sensitive shadowgraph scattering method (phase fluctuations below 10 milliradians measured to within a few percent absolute accuracy) will be described briefly.

1. Introduction
Equilibrium fluctuations are usually small and limited to molecular length scales. A notable exception occurs for fluids near second-order phase transitions where correlations readily extend to the micron scale as evidenced by the impressive phenomenon of critical opalescence [1]. While critical phenomena was one of the most studied areas in the sixties and seventies, a great amount of work was also carried out during that period in connection with two quite diverse physical systems, the laser, and the Rayleigh-Bénard (RB) convective instability (a liquid slab heated from below). These are examples of a general class of instabilities in open systems exhibiting a threshold value for the control parameter, above which macroscopic structures are formed. It was shown that the behaviour near the threshold of these instabilities closely mimics mean-field behaviour of critical systems, in the sense that system properties (order parameter, amplitude of fluctuations, relaxation times) present a power-law dependence on the control parameter much like that observed close to a classical critical point [2-4].

The literature on this topic is too vast to be quoted here. We single out however a UC Santa Barbara paper where large fluctuations below the onset of convection were first observed and
measured [5]. It was shown that fluctuations below threshold peaked around the wave vector of the convective pattern that eventually materialized above threshold, in keeping with critical fluctuations scaling arguments. The amplitude was also measured and agreed with theory to within experimental accuracy of about 50%. We quote Ref. 5 here because it is the first reported utilization of quantitative shadowgraph methods to study large-scale non-equilibrium fluctuations.

This paper presents results obtained using the shadowgraph method to measure long-range non-equilibrium fluctuations in both a fluid mixture and a single component fluid. The measurements were made both on Earth and during a flight experiment, the 13-day Foton-M3 mission. None of these measurements could have been performed without quantitative shadow methods.

The fluctuations we will discuss occur quite readily without the necessity of being close to a generalized critical point. Very much like the critical fluctuations, these are non-propagating fluctuations that scatter into the central Rayleigh component of the spectrum of the scattered light. The relevant theoretical work [6-9] was developed over a few years, without immediate reaction from experimentalists. It showed that subjecting a fluid or fluid mixture to a stabilizing temperature gradient by heating from above should result in non-equilibrium fluctuations with amplitudes and spatial ranges many orders of magnitude greater than those of equilibrium fluctuations. Incidentally, non-equilibrium propagating fluctuations under a temperature gradient were studied in Refs. 10 and 11 slightly earlier than references above, the effect of the gradient being of generating an asymmetry between the frequency shifted Brillouin components. For the non-propagating fluctuations we describe here, the hallmark of such gradient-driven fluctuations is the asymptotic behaviour for the structure factor of an unbounded sample in the absence of gravity. On Earth, gravity quenches this divergence at a well-defined wave vector determined by the fluid properties, while in the absence of gravity, the finite sample thickness was predicted to be the only limiting factor. Physically, the fluctuations are due to the coupling of thermal velocity fluctuations, with the quantity exhibiting a gradient (temperature or concentration, e.g.). While they have no effect on a homogeneous system, when they occur along a density or a concentration gradient, they can generate large fluctuations by displacing parcels of fluid having different density or concentration along the gradient. The limitation of such gradient-driven fluctuation on Earth occurs because very long wavelength fluctuations are suppressed by buoyancy which acts as a restoring mechanism for the displaced fluid parcels. In fact, such a restoring force can even result in oscillatory motion for sufficiently long wavelength fluctuations. Because of buoyant forces, $S(q)$ saturates to a constant below a critical wave vector $q_c$, which for a concentration gradient is given by $q_c = (\beta g \nabla c / (\nu D_c))^{1/4}$, where $\beta$ is the solutal expansion coefficient, $g$ is the acceleration of gravity, $\nu$ is the kinematic viscosity and $D_c$ is the concentration diffusion coefficient. For most common mixtures $q_c$ is of order few hundred cm$^{-1}$.

When the predictions were made they raised the issue of why not even a hint of the existence of these anomalously large fluctuations had ever been observed, despite the fact that solutions and mixtures subjected to gradients had been studied for many years. For example, using free diffusion methods to determine diffusion coefficients, or analytical centrifuges to obtain molecular weight through schlieren-sedimentation profiles [12], or while measuring Soret coefficients optically [13].

The reasons why they escaped observation are quite diverse and to some extent rather fortuitous. First, the common techniques use optical methods with the axis orthogonal to the applied gradient, while theory indicates that effects are maximized when the optical axis is aligned with the gradient, a geometry that is inappropriate (and very unpractical) for all the above methods. Second, because of the weak dependence of $q_c$ on density mismatch, it turns out that for any choice of single-component liquid and any mixture or suspension, gradient driven fluctuations on Earth remain confined to length-scales of the order of ten to a hundred microns, and this means that they were unlikely to be observed by visual inspection. Finally, the optical distortion introduced is rather modest in comparison with the
critical opalescence effects one would expect for a critical fluid with the same correlation length-scale. Critical opalescence implies that scattering is so strong, that it usually develops into strong multiple scattering, which in turns implies that a plane wave front will suffer random phase distortions large compared to $\pi$ in passing through the sample. Here, as we will show, wave front distortions remain a small fraction of $\pi$, and consequently instrumentation of high sensitivity is required to detect them. The stronger disturbance caused by critical opalescence is due to the fact that the structure function $S(q)$ decays approximately as $q^{-2}$ rather than as $q^{-4}$, and thus integration over wave vector results in large total scattering cross-sections.

The first observation of non-equilibrium fluctuations in a simple liquid subjected to a (stabilizing) temperature gradient is due to the Sengers group at the University of Maryland [14, 15]. The technique employed was a form of dynamic light scattering in which the scattering was optically mixed with stray light. The scattering due to the non-equilibrium fluctuations was very small compared to the enormous stray light contribution from optical imperfections and other sources. As dynamic measurements were performed, it was possible to cleanly separate the true signal from the stray light, and the $q^{-4}$ dependence was clearly observed, as well as the $|\nabla T|$ behaviour. The accessible scattering angles were much too large however, to permit the detection of the gravitational cutoff, and the Authors concluded that it was unlikely that it would be detected in the future. Ref. 14 and 15 are groundbreaking papers proving that experiments could be successfully executed in this area.

Subsequently, ultra low angle scattering and shadowgraph methods were employed by the Milan group to investigate non-equilibrium concentration fluctuations in Soret-driven and free diffusion processes [16, 17]. The cutoff at low $q$ due to the presence of gravity was observed, and found to be in excellent agreement with theory. Particularly important for the history of this project is a paper in Nature [17], describing an experiment in which no applied thermal gradient was involved, but instead the driving mechanism was the creation of a pure concentration gradient via the separation of two miscible solutions and subsequent free diffusion process. In the associated comment in the News and Views section, it was suggested that an experiment in microgravity conditions would be highly desirable, as this would give the possibility of observing fluctuations at length scales comparable with the physical size of the sample.

It was then decided to join forces between the Milan group and the UCSB group, proposals were submitted to ESA and NASA, and eventually the GRADFLEX (GRAdient Driven FLuctuation EXperiment) project was launched. It was decided that two experiments had to be flown, one on a single-component fluid (SINGLE), to be primarily the responsibility of the UCSB group, and one on a mixture (MIXTURE), for which the Milan group assumed primary responsibility. Both experiments employed a temperature gradient as the stimulus. It was applied to a 3-mm thick carbon disulphide sample in SINGLE to study density fluctuations and to 1-mm thick sample consisting of a 1.8 wt.% solution of $M_w = 9,100$ polystyrene dissolved in toluene for MIXTURE. The polymer solution, which has a very large Soret effect, developed a strong concentration gradient in response to the applied temperature gradient. Two publications have appeared recently describing the two experiments and the flight results, that are briefly reported here [18, 19]. As a general reference to the subject of non-equilibrium fluctuation we suggest the exhaustive and beautifully written book by Ortiz de Zarate and Sengers [20]. The present work is the first report containing results from both MIXTURE and SINGLE.

2. Experimental setups

Each setup consisted of a thin cylindrical sample cell with associated equipment to impose a stable temperature difference between the plates confining the sample. In both cases, the optical axis of a shadowgraph was oriented parallel to the gradient, and therefore optical windows that conduct heat much better than do the samples were required. Sapphire was chosen, as it has a thermal conductivity
hundreds of times larger than that of typical fluids. Because the cells are rather different, they will be described below separately. The **SINGLE** cell (and associated optics) was about 3 times larger than those of **MIXTURE** because the gravitationally induced crossover wave vector is about that much larger for the mixture than for CS2. For a single-component fluid, \( q_c = \left( \alpha g v T / \left( \nu D_r \right) \right)^{1/4} \), where \( \alpha \) is the thermal expansion coefficient, and \( D_r \) is the thermal diffusivity. Typically for single-component organic fluids, \( q_c \) is of the order of 70 cm\(^{-1}\) as compared to a few hundred cm\(^{-1}\) for mixtures. As the gravitational crossover is absent in microgravity, fluctuations are limited only by the finite sample thickness, and the cross over then occurs at a much lower value near \( q' = 2\pi / L \), where \( L \) is the sample thickness. Thus a dramatic change in the low \( q \) scattering was anticipated for **MIXTURE** even with a sample thickness of only 1 mm.

Both experiments utilized the quantitative shadowgraph technique. As this method proved to provide exceptional performance, but is very little known, we present below a brief description of the physical principles on which it is based and the origins of the rather complex instrumental transfer function \( T(q) \). The approach is unorthodox, no rigor will be claimed, but we hope it will be effective.

### 2.1. Scattering cells, choice of samples, stimuli and general layout

#### 2.1.1. **SINGLE**

**SINGLE** deals with large-scale gradient driven density fluctuations in a single component liquid, CS2, which was chosen to give a large signal. The apparatus was a flight version similar to the prototype developed to demonstrate the feasibility of measuring signals from a single-component fluid heated from above, something that had not been achieved previously. A 75-mm diameter 3 mm thick sample, at 1 atmosphere, was confined between the sapphire window and a silicon mirror. The window was coated with transparent conducting Indium Tin Oxide (ITO) on the surface in contact with the CS2, and the ITO was used to generate heat. Heat was removed from the mirror by means of 4 Peltier elements separated from the mirror by a thermal diffuser. The ITO and Peltier currents were regulated to impose gradients and were also used to maintain the mean temperature at 30\(^\circ\) C. Light of wavelength 680 nm from a super-luminous diode coupled into a mono-mode fibre, was used for illumination. The light diverged in a nearly Gaussian beam from the polished end of the fibre and passed through a beam splitter and then a lens mounted just upstream from the sample window. The lens collimated the light into a diffraction-limited beam, after which it passed through the sample and was reflected by the mirror. The beam splitter diverted 50% of the returning beam to a 1024 by 1024 CCD sensor, which recorded images at an effective distance \( z = 310 \) cm from the sample. By dividing images pixel by pixel, it was possible to detect the fractional intensity variation \( \delta I(x,y,t) \) caused by interference between the beam and light scattered by the fluctuations. Fourier transformation of the central 512 by 512 portion of such ratio images served to separate the fluctuations by wave vector. In the end the apparatus proved to be extremely sensitive and we were able to measure signals with applied temperature differences as small as 1 K. In taking data during the mission, nominal temperature differences of 1, 2, 5, 10, 15, 20 and 30 K were applied, and the same values were used while taking data on Earth after the mission.

#### 2.1.2. **MIXTURE**

**MIXTURE** deals with large-scale fluctuations of concentration, and to induce them, a concentration gradient must be established. A repeatable and controlled way to do so is to utilize the Soret effect [13]. Applying a temperature gradient generates a Soret mass flow, until the gradient reaches steady state where back diffusion balances the Soret flow. The resulting steady-state concentration profile is governed by \( \Delta c = -S_T c (1 - c) \Delta T \). Here \( S_T = 6.49 \times 10^{-2} \) K\(^{-1}\) is the Soret coefficient, and \( c = 1.8 \) wt.% is the concentration. In contrast with the sample choice for **SINGLE**, sample choice for **MIXTURE** offered a broad spectrum of choices, as \( S_T \) varies substantially with solute molecular weight and...
species. Unfortunately, the mass diffusion coefficient typically scales in the opposite way, and has to be chosen carefully in order to make time constants compatible even with a two-week mission. Ultimately, a low molecular weight polystyrene-toluene solution was used ($M_w = 9,100 \text{ g mol}^{-1}$), as it guaranteed a large Soret-driven concentration gradient and reasonably large mass diffusivity $D_c$ to keep time constants within reason. The magnitude of the concentration difference can be readily altered, and the experiment repeated as desired. The thermal-gradient cell was a flight-engineered version of a prototype used for ground-based tests [21], and was similar to designs developed previously [16, 17].

The sample was a 25-mm diameter by 1.00 mm thick layer of polymer solution confined between two parallel, 12 mm thick, sapphire windows that were temperature controlled to better than ± 0.01 K. The use of sapphire windows allowed the application of a relatively uniform temperature difference, while permitting optical access to the sample in a direction parallel to the imposed gradient. The sample was illuminated with collimated, 680 nm wavelength, light from a super-luminous LED coupled to a mono-mode optical fibre. Interference between the beam and the light scattered by the diffusing fronts resulted in small but measurable variations of the intensity in the images collected by a Charge-Coupled Device (CCD) with 1024x1024 square pixels. The central 512 by 512 portion of differences of succeeding images were Fourier decomposed to reveal the mean-squared amplitude of concentration fluctuations vs. the scattering wave vector $q$.

A measurement cycle involved an initial equilibration phase at a uniform temperature of 30.0° C for 260 minutes. Toward the end of this phase, a set of 539 reference images was acquired to characterize the optical background and the camera noise. This phase was followed by the rapid imposition of a temperature difference across the sample, which started the diffusion process. The temperature differences utilized were 4.35, 8.70 and 17.40 K. The time required for the formation of a linear temperature profile was about 100 s. In contrast, the time to create a steady-state concentration profile ($\tau_c = h^2/\left(\pi^2 D_c\right)$) was about 500 s; here $D_c = 1.97 \times 10^{-6} \text{ cm}^2/\text{s}$ is the diffusion coefficient, and $h = 1.00 \text{ mm}$ the sample thickness.

2.2. The shadowgraph method
The structure function of fluctuations $S(q)$ is usually determined by measuring the angular distribution of scattered light $I(q) \propto S(q)$, the scattering angle $\theta$ being related to the wave vector of the fluctuations by $q = 2k \sin(\theta/2)$, where $k$ is the wave vector of the light in the sample. In the simplest arrangement, sensors at various scattering angles are placed well clear of the transmitted beam. When fluctuations become spatially large, scattering angles must be reduced, and separating the scattered light from the beam becomes exceedingly difficult, as stray light scattered by imperfections in every optical surface encountered by the beam overwhelms the true scattering. It is in this regime that the power of light scattering via the shadowgraph is most valuable.

A shadowgraph instrument can be used as a scattering instrument [22], where one actually exploits the existence of the transmitted beam as a local oscillator that mixes with the weak scattered field. This mixing results in small amplitude time- and spatially-dependent fluctuations in the beam intensity which may be quantified using a digital camera. The sensor is normally so close that light scattered at any angle of interest falls onto the detector. A special scattering condition, the Raman Nath (R-N) condition (see below) is normally satisfied. Before proceeding further, one should stop for a second, and ponder the extreme simplicity of this method: a good quality plane wave, the scattering cell, and a sensor comprise the entire setup. Because of the interference between the beam and the scattered light, the low contrast intensity variations are proportional to the scattered field, rather than to the scattered intensity. Consequently, taking the Fourier transform of the intensity distribution gives the angular spectrum of the scattered radiation [22, 23]. So one could actually determine both the amplitude and
phase of the scattered plane waves in all directions. Indeed for a sample consisting of spheres, such as the calibration sample discussed below, one can generate the images of individual particle via a z scan [24]. However, as we are interested in the scattered intensity, measuring the power spectrum of the detected intensity distribution \( P(q) \) is more useful, rather than the overly-detailed Fourier transform. Of course phases are lost, and positions of particles are consequently also lost. From the above naive discussion, one might be inclined to think that \( P(q) \propto S(q) \). Unfortunately, the matter is more complicated, as a transfer function \( T(q) \) is generally involved, and \( P(q) = T(q)S(q) \). Actually, \( T(q) \) is the product of a number of transfer functions, each accounting for contributions due to various phenomena, such as temporal space coherence, averaging over pixels, optical resolution and windowing caused by taking finite-sized images, etc. Some of these effects are subtle, and will not be discussed here. Others, like finite time coherence will be briefly mentioned below.

The most evident feature of the power spectra of shadow images is the typical oscillatory, fringe-like behaviour, that is a consequence of the Raman Nath condition. It is rather simple to calculate \( T(q) \) due to R-N condition. By definition, it is the response to white noise signal, that is to light being scattered uniformly as a function of \( q \), as is the case for very small particles, that have \( S(q) = S(0) = 1 \). The interference at distance \( z \) for a small particle in the origin is given by

\[
I(x, y) = \frac{2}{k}S(0)\cos\left(\frac{k(x^2 + y^2)}{2z} + \phi\right)
\]

where \( z \) is the distance and \( \phi \) the scattered wave phase delay. The associated power spectrum is

\[
S(q_x, q_y) = \frac{4\pi^2}{k^2}|S(0)|^2\sin^2\left(\frac{q^2z}{2k} - \phi\right)
\]

and it coincides with the transfer function \( T(q) \). This implies that the random intensity distribution is made up by identical, infinite, constant amplitude interference fringes. The extra phase \( \phi \) accounts for size dependent phase delays, as discussed below [25]. Let us see when this holds true. As the fringe peak positions in \( q \) depend on distance, this implies that for all the \( q \) values of interest, the actual sample thickness must be so small that distance variations between \( z - L/2 \) and \( z + L/2 \) will not introduce appreciable changes for the positions of the outermost fringe like oscillations. It can be easily proven that this is the R-N condition \( q^2L/(2k) \leq \pi \). So, if \( S(q) \) decays over the wave vector range where R-N holds, it will depress accordingly the infinite sequence of fringe-like oscillations of \( T(q) \). So, in a sense, it is partially true that \( P(q) \) and \( S(q) \) are proportional, but in “average” over the \( T(q) \) oscillations, the case \( q = 0 \) being pathological because of the wide minimum, that requires special attention.

In concluding, thinking in terms of circular interference fringes as the source of the power spectra can be very profitable, because one learns how to get a gross idea about additional effects like limited time coherence of the superluminous diode that has been used in both setups. As \( \lambda/\Delta\lambda \) is typically of the order of 40, additional depression of the fringe like oscillations should be expected at the 40\(^{th}\) fringe, as limited coherence adds its share in depressing the oscillations, and it must be accounted for.

While one could figure out one by one the contributions to \( T(q) \), it was found much profitable to derive experimentally \( T(q) \) through a calibration procedure. As often done with scattering method, a colloidal suspension was chosen. Unfortunately very small particles, say 100 nm in diameter, otherwise ideal to guarantee uniform scattered intensity and zero scattered wave phase delay, do scatter too little, and finally 2 micron polystyrene spheres in isopropyl alcohol were chosen.

Below, the scheme used for the mixture experiment is reported, the one for single being almost identical. The transfer function can be quite generally written as

\[
T(q) = A(q)\sin(q^2z/2k_0) + C(q),
\]

where the envelopes of the minima and maxima are equal to \( C(q) \) and \( A(q) + C(q) \), respectively. Calibrating the optical system means basically to determine the functions \( A(q) \) and \( C(q) \).
To carry out the calibration, 320 images were taken with the cell filled with filtered isopropyl alcohol, to determine a background signal $S_b(q)$. The cell was then filled with the colloidal suspension, and 3500 images were taken. Between each image, the sample was stirred using an automatic piston, to ensure statistically independent images. After subtracting $S_b(q)$, the signal from the spheres is given by $S_{sp}(q) = A(q)\sin^2(q^2z/2k_0 + \phi_{sp}) + C(q)$, where $\phi_{sp}$ is an additional phase delay that accounts for the non-vanishing optical thickness of the 2 micron spheres in isopropyl alcohol [25]. We found the phase delay to be 1.78 radians, in very good agreement with Mie theory [26]. The functions $A(q)$ and $C(q)$ were determined by fitting the data for $S_{sp}(q)$, and this in turn allowed us to determine the instrumental transfer function $T(q)$.

3. Experimental results

3.1. Mixture
To show the striking comparison between ground and flight data, we show in Figure 1 a sequence of false colour two dimensional shadow images taken both on ground and in flight with the polymer solution and an applied temperature difference of 17.40 degrees. Images were taken 0, 500, 1000 and 2000 s (left to right) after the imposition of a 17.40 K temperature difference. The side of each image corresponds to 5 mm. Colours map the deviation of the intensity of shadowgraph images with respect to the time-averaged intensity.

The figure shows that in a gravity-free environment, fluctuations are boosted in amplitude and size. Images were acquired at a constant frame rate of 0.1 Hz while a temperature gradient was kept constant for 42 h. Estimates of the mean square amplitude of the fluctuations at various $q$ is performed by taking the azimuthal average of the two dimensional power spectrum.

![Figure 1](image)

**Figure 1.** Shadowgraph images of nonequilibrium fluctuations in microgravity (top panels) and on Earth (bottom panels). The sample is a 1.00-mm-thick solution of polystyrene in toluene. Images were taken 0, 500, 1000, and 2000 s (left to right) after the imposition of a 17.40 K temperature difference at $t=0$. The side of each image corresponds to 5 mm. The false-colours map the deviation of the intensity of shadowgraph images relative to the average intensity.

To isolate the signal of the concentration fluctuations from the contribution of nonequilibrium temperature fluctuations, as well as from other sources of noise (e.g. electronic noise from the CCD sensor), a dynamical analysis has been performed (see [18] for details). The power spectrum was
finally divided by the instrument transfer function $T(q)$ to compensate for the R-N oscillations, for finite time coherence and other effects not discussed here.

The resulting $S(q)$ obtained for temperature differences of 4.35, 8.70 and 17.40 K are shown in the top part of figure 2. The curves show the striking asymptotic behaviour $S(q) \propto q^{-4}$ for large values of $q$, while tend to saturate at low $q$ due to the finite-thickness of the sample (1 mm).

![Figure 2](image)

**Figure 2.** Top panel: experimental results obtained in microgravity in the presence of temperature differences of 4.35 K (black triangles), 8.70 K (blue squares) and 17.40 K (red circles). Bottom panel: experimental results divided by $|\Delta T|^2$.

From the actual values of the fractional intensity fluctuations (see false colour scale) one can derive an absolute estimate for the rms of the phase fluctuations. It amounts to less than $10^{-2}$ radians. So path length changes of few Angstrom are actually evaluated. Similar conclusions can be immediately reached by noticing that the scattering phase delay $\phi$ is very small. So the one millimetre size giant fluctuations have an optical thickness smaller than a 100 nm polystyrene colloidal particle in water [25]. Incidentally, this also implies that the main beam fractional power removal by the fluctuations is a small fraction of 0.1 %. The proportionality of the overall amplitude of non-equilibrium fluctuations to $|\Delta T|^2$ is confirmed in the bottom part of the figure 2.

Experimental data were compared with existing numerical predictions. In the absence of gravity, and with the radius of gyration of the polymer chains much larger than the size of solvent molecules, a theoretical model [20, 27] based on a single-mode Galerkin approximation predicts that the mean-squared amplitude of fluctuations should scale onto a universal curve independent of mixture properties or applied concentration difference which scales as at large wave vectors and saturates to a constant value at small wave vectors. Figure 3 shows the experimental data plotted with the theoretical prediction, in the absence of adjustable parameters. This result provides a straightforward confirmation of the scale invariance of the fluctuations up to wavelengths comparable to the sample thickness, and of the overall increase of amplitude of fluctuations when compared to data acquired on earth (dashed lines). Above a length scale comparable to the sample thickness, the results confirm that the scale invariance of the fronts of diffusion is frustrated by the finite size of the container, as predicted by theory [27] and by two-dimensional simulations [28]. Quite interestingly, very recent simulations
show that the presence of such fluctuations affects the mass transfer during a diffusion process [29, 30].

Figure 3. Comparison of the experimental results with the theoretical predictions. The solid line is the theoretical prediction for microgravity. The dashed lines are the theoretical predictions for fluctuations on Earth.

3.2. Single

We show in figure 4 a typical result for the azimuthal average of the two-dimensional power spectrum of the fluctuations for CS2 on ground, vs. the dimensionless wave vector $qL$, for an applied temperature gradient of 17.9 K/cm.

Figure 4. The product of the measured transfer function $T(q)$ and the calculated structure factor $S(q)$ vs. the dimensionless wave vector $qL$ for a 3 mm thick CS$_2$ sample subjected to a temperature gradient of 17.9 K/cm on Earth.

The data are shown as open circles, and the theoretical result is shown as the solid line. It should be emphasized that no parameters have been adjusted in making this comparison, including the amplitude. Thus the theory clearly does a remarkable job of describing the data, both in terms of absolute amplitude and wave-vector dependence. Of course, the theory actually predicts the structure
factor $S(q)$, while what is shown for direct comparison with the data is the product of the measured transfer function $T(q)$ and $S(q)$. The oscillatory behaviour associated with the transfer function is clearly evident. Similar results were observed for all applied gradients.

SINGLE exhibited some unexpected behaviour during flight, and to date we have no explanation for what was observed, however we have devised a reasonably robust method for extracting useful data despite the difficulties. Figure 5 shows shadowgraph images taken with no gradient applied (left) and shortly after application of a nominal 30 K temperature difference across the sample (right).

**Figure 5.** Shadowgraph image taken during flight with no temperature difference applied (left) and with a nominal 30 K difference applied (right). Note the presence of several white spots of various intensities in the image on the right, as well as the distortions present near the periphery. The speckled region at the bottom is of no consequence.

We observed that applying a temperature gradient during flight resulted in the sudden (within seconds) appearance of several white spots in the image, as though light were being focused in those regions. These bright regions appeared in exactly the same place every time a gradient was applied, and remained fixed in location as long as the gradient was maintained. Normal fluctuations were observed in those regions however. In addition to the white spots, it is evident that there are other perturbations present in the outer portions of the cell, and these perturbations evolved slowly over the 5-hour duration of a data run, suggesting that they may have been caused by weak lateral temperature gradients. The extent of this evolution was marked, and is shown by the two images of figure 6.

**Figure 6.** Shadowgraph images taken during flight with a nominal temperature difference of 30 K. The image on the left was taken immediately after the warm-up period, and the one on the right about 5.3 hours later, immediately after the end of the data-taking period.

The central region from which the data were obtained seemed little affected, however, the results obtained for $S(q)$ showed additional fluctuation power at low $q$, and this additional power varied with
what region was chosen for analysis and even from run to run. Time-domain spectral analysis of images taken in rapid succession (as rapidly as 30 Hz) revealed that presence of an additional noise source contributing very low temporal frequency power, but only for wave vectors lower than about 30 cm\(^{-1}\).

By fitting the higher frequency data using the predicted theoretical form and integrating the resulting spectrum it was possible to extract results for the product \(T(q)S(q)\). Examples of the time domain power spectrum \(S(q,\omega)\) for several very small wave vectors are shown in figure 7. As can be seen, the additional noise (visible in the data shown as x’s) is significant only for angular frequencies below about 0.2 rad/s (~0.03 Hz), and is thus a very low frequency phenomenon, allowing it to be separated from the data by fitting. Clearly, the form predicted by theory provides an excellent description of the dynamics of the fluctuations, as shown by the good agreement between the open circles and the solid lines. By automating the fitting process it was possible to analyze the flight data to obtain \(T(q)S(q)\) for many different q values, and the results are shown in figure 8, below.

**Figure 7.** Time-domain spectral power for several small wave vectors with an applied gradient of 101 K/cm. Data images were taken at 2 Hz for the upper panel and at 0.5 Hz for the lower two. The theoretical form (solid line) was fit to the data shown as open circles, while the data shown as x’s was excluded from the fit. The integral of the fitted function equals \(T(q)S(q)\).

**Figure 8.** Data for \(T(q)S(q)\) vs. the dimensionless wave vector \(qL\) taken in the absence of gravity with an applied gradient of 17.9 K/cm. The solid lines are the predicted results for \(S(q)\) after multiplication by the measured transfer function. As can be seen the theoretical prediction exceeds the data by about 15%, an effect we observed for all applied gradients.
Comparison of the vertical axis scale with that of figure 4 shows that removing gravity has enhanced the signal by about a factor of 8, and that the position of the lowest peak has shifted to lower wave vector. This occurs because removing gravity eliminates the gravitational suppression of the fluctuations, and their amplitude continues to diverge as \( q \to 0 \) until limited by the finite sample thickness of 3 mm.

To display the behaviour more generally we may divide the data by the measured transfer function (bearing in mind that near the minima this involves dividing by small numbers!). Doing so produces experimental results for the structure factor \( S(q) \) in absolute units, and the results are shown in figure 9 for several applied gradients, both on Earth and in the absence of gravity.

![Figure 9. Data for the dimensionless structure factor \( S(q) \) obtained by dividing the raw data by the \( q \)-dependent transfer function \( T(q) \), vs. the dimensionless wave vector \( qL \), for applied gradients of 17.9 K/cm (squares), 34.5 K/cm (triangles) and 101 K/cm (circles). The upper curves show the data obtained during flight and the lower ones show data obtained on Earth after the mission. The solid lines are the theoretical predictions without any adjustable parameters. From [19]

Note that the wide flat plateau region observed on Earth is replaced by a broad maximum in the absence of gravity. As predicted, this maximum occurs very near \( qL = \pi \), showing that in the absence of gravity, the only thing limiting the range and amplitude of the fluctuations is the finite sample thickness. Considering that these fluctuations are thermally driven, this is certainly a remarkable observation.

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