Non-Equilibrium Fluctuations in Microgravity: 
Initial Results of GRADFLEX Aboard FOTON-M3

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Abstract

Recent theory and experiments have shown that heat conduction in simple fluids and mass diffusion in binary mixtures are inevitably accompanied by the presence of large amplitude fluctuations in temperature or concentration, respectively. These fluctuations occur at all length scales, and their mean squared amplitude diverges as the fourth power of their size. However, on Earth, the divergence of large size fluctuations is prevented due to gravitational stabilization. The aim of the GRADFLEX experiment is to investigate the non-equilibrium fluctuations in microgravity, and to measure the divergence of large size fluctuations. GRADFLEX was flown on the FOTON M3 mission September 14-26, 2007. Some images of the fluctuations were downlinked to Earth during the mission by means of the Telesupport Unit. In this paper we present preliminary quantitative results based on the images downloaded during the mission. We show that under microgravity the amplitude of fluctuations diverges as anticipated by theory.

1. Introduction

Heat conduction (HC) and mass diffusion (MD) represent fundamental transport processes acting at the microscopic length scale. Many experiments performed under microgravity conditions take advantage of the absence of convection to exploit pure heat-conduction and diffusive-remixing processes.

Recently, however, both theory¹-³) and experiment⁴-¹⁰) have shown that these processes are inevitably accompanied by giant temperature and concentration fluctuations. The fluctuations are generated by the coupling between velocity fluctuations and the imposed temperature or concentration gradient. Velocity fluctuations are always present as they are generated by thermal energy. In the presence of a gradient, any motion of a small portion of fluid having a component in the direction of the gradient will result in the displacement of that portion into a layer with a different temperature or concentration.

The fluctuations occur at all length scales between the microscopic and macroscopic, their mean-square amplitude diverging as the fourth power of their size. On Earth, the divergence is limited by the stabilizing action of the gravitational force on fluctuations with wavelength larger than a certain cut-off wavelength \( \Lambda_{c}^{¹¹-¹³} \).

The aim of the GRADFLEX project¹⁴) is to investigate non-equilibrium fluctuations in HC and MD processes under microgravity conditions, and to measure the full divergence of fluctuations. Understanding such fluctuations may be particularly relevant for experiments performed in microgravity based on HC and MD processes, such as the crystallization of proteins which is a delicate process involving diffusion of macromolecules.

GRADFLEX was flown aboard FOTON-M3 September 14-26 2007. Some images of the fluctuations became available immediately thanks to the Telesupport Unit downlink to Earth. In this paper we present the first quantitative results for non-equilibrium fluctuations in microgravity obtained from some of the shadowgraph images downloaded to Earth during the flight.

2. Nonequilibrium fluctuations

During the early 80’s the presence of a macroscopic thermal gradient was predicted to enhance the amplitude of thermal fluctuations¹-³). This effect was initially reported for a single component fluid subjected to a temperature gradient⁴). It was then shown that it is also possible to enhance the concentration fluctuations in a binary mixture by applying a temperature gradient. In this case, the temperature gradient induces a concentration gradient by means of the Soret effect⁸,¹⁵). Similarly, a mixture of two fluids subjected to a pure concentration gradient under isothermal conditions was shown to exhibit greatly enhanced concentration fluctuations⁹). In all cases, the origin of the enhancement is the coupling of velocity fluctuations to the imposed gradient. For example, when a sugar solution is diffusing upward into a water layer, velocity fluctuations couple to the concentration gradient. Any vertical upward motion of a small portion of the solution results in the displacement of a concentrated fluid parcel into a layer where the concentration of
sugar is smaller\textsuperscript{16}. Similarly, a downward movement will drag a parcel of less concentrated fluid into a more concentrated layer. Displacements of fluid due to velocity fluctuations are always present due to thermal motion, but in the non-equilibrium state imposed by the presence of a concentration gradient, they become coupled with and enhance the concentration fluctuations\textsuperscript{9, 13).}
The mean-squared amplitude of the fluctuations driven by a temperature or concentration gradient has been observed experimentally\textsuperscript{5, 6, 8, 15), to diverge as \( q^{-4} \), where \( q \) is the wave vector of the fluctuation. This divergence is not predicted to continue indefinitely as \( q \) approaches zero. Instead, it is predicted to saturate when the fluctuation wavelength exceeds a value determined by fluid properties and the gravitational field\textsuperscript{12, 13). For the case of a fluid mixture subjected to a concentration gradient \( \nabla c \), the saturation phenomenon has been observed experimentally\textsuperscript{8, 9). The cut-off wave vector \( q_c \) is given by\textsuperscript{11-13):}

\[
q_c = \begin{cases} 
\left( \frac{\alpha g \nabla T}{\nu \kappa} \right)^{\frac{1}{2}} & \text{temperature NEF} \\
\left( \frac{\beta g \nabla c}{\nu D} \right)^{\frac{1}{4}} & \text{concentration NEF}
\end{cases}
\]

where \( \nabla T \) is the temperature gradient, \( g \) is the gravitational acceleration, \( \alpha \) the thermal expansion coefficient, \( \nu \) the kinematic viscosity, \( \kappa \) the thermal diffusivity, \( \beta \) the solutal expansion coefficient, and \( D \) is the mass diffusion coefficient.

For the case of a fluid mixture with an applied temperature gradient, the Soret effect determines the development of a concentration gradient in response to the applied temperature gradient. In general, both gradients couple to the velocity fluctuations and are effective in enhancing fluctuations\textsuperscript{12), but near a consolute point and for suspensions of macromolecules the effect of the induced concentration gradient dominates. In this case, the cut-off wave vector is still given by Eq. 1, but here the Soret effect on the concentration gradient is:\textsuperscript{7) \( \nabla c = -\left( k_T / T \right) \nabla T \), in which \( T \) is the absolute temperature, and \( k_T \) is the thermal diffusion ratio. In the absence of gravity, the cut-off wave vector is reduced to zero, and thus the mean-square amplitude of the fluctuations below the cut-off wave vector is expected to continue the \( q^{-4} \) divergence already observed above \( q_c \), the divergence being limited only by the confining dimensions of the sample\textsuperscript{18-21).}

3. The GRADFLEX experiment

In the previous section, we have suggested how macroscopic concentration gradients in a simple fluid or in a binary mixture can lead to anomalously large non-equilibrium fluctuations (NEF). The critical wave vector \( q_c \) defined by Eq. 1 distinguishes two completely different regimes. Above \( q_c \), the mean-squared amplitude of the fluctuations diverges with the explosive \( q^{-4} \) power law, and their dynamics is governed by purely diffusive processes. Below \( q_c \), the fluctuations are limited by gravity, and their decay is faster than what would be determined by diffusion alone. An estimate of \( q_c \) on Earth for typical mixtures parameters gives values of the order of 500 cm\(^{-1} \), while for simple fluids it is of the order of 100 cm\(^{-1} \). This implies that fluctuations having wavelengths larger than a fraction of a millimeter are limited in amplitude by gravity. In a microgravity environment, where the inhibiting effect of gravity is strongly reduced, the \( q^{-4} \) scale invariant behavior should be observable also for much larger wavelength fluctuations and their lifetime should be determined by heat conduction or mass diffusion alone. This may affect heat and mass transfer processes taking place in microgravity. The main practical reason is that the diffusive relaxation time scales with the square of the fluctuation size and this makes large-scale fluctuations very long-lived. A typical example where the long life of NEF could have dramatic consequences is the crystallization of a protein which typically involves diffusion in macromolecular solutions. It has been shown that even in microgravity conditions it is very difficult to grow high quality crystals. A possible explanation for this observation could lie in the existence of large-scale non-equilibrium concentration fluctuations whose lifetime could be long enough to result in uneven growth, causing thereby a defective crystal. It is also clear that the issue of spontaneous remixing in space may follow unexpected routes.

The aim of GRADFLEX (GRAdient Driven FLuctuation EXperiment), a joint project of the University of Milan and UC Santa Barbara, funded by ESA and NASA, is to investigate diffusion-driven fluctuations in the absence of gravity. The experiment was proposed to ESA in 1998 by UNIMI/CNR-INFM (Università degli Studi di Milano and Consiglio Nazionale delle Ricerche - Istituto Nazionale per la Fisica della Materia) and UCSB (University of California at Santa Barbara). A feasibility study was performed in the very early phases of the project. GRADFLEX was flown aboard the Russian capsule FOTON-M3, which was launched from the Baikonur Cosmodrome in Kazakhstan on September 14, 2007. Together with GRADFLEX, it carried a scientific payload of about 400kg made up of 43 experiments in fluid physics, biology, crystal growth, meteoritics, radiation dosimetry and exobiology. The mission spanned 12 days during which FOTON completed about 190 orbits around the Earth. FOTON-M3 eventually landed near the Russian border with Kazakhstan on September 26, 2007. The scientific payload was retrieved, and scientific data stored onboard will be retrieved, and scientific data stored onboard will be
available to scientists for analysis.

The GRADFLEX experiment involves two different samples. The first one, under the scientific lead of UCSB, investigates temperature fluctuations in a single-component fluid. The second one, under the lead of UNIMI-CNR-INFM investigates concentration fluctuations in a binary mixture. More details about the development of the GRADFLEX experiment can be found in a dedicated paper.

The first scope of feasibility studies was the selection of an appropriate mixing technique to characterize the non-equilibrium fluctuations. Many different optical methods have been considered including Low Angle Light Scattering (LALS), but it was found to suffer from increasing stray light problems as the scattering angle is reduced. It also requires fine and stable alignment, since the main beam must be disposed of without eliminating the light scattered at very small angles. 

Schlieren methods were also considered, and they do have some advantage, as the instrumental response function is not oscillatory like that of the shadowgraph. The simplicity of the Schlieren transfer function however is acquired at the expense of very good optical components and stable accurate blade positioning. A refined quantitative shadowgraph diagnostic technique was selected to obtain images of the fluctuations.

Sequences of images of the fluctuations are processed to recover their spatial power spectrum. The power spectrum provides the mean-square amplitude of the fluctuations \( S(\mathbf{q}) \) as a function of their wave vector. A shadowgraph is a rugged instrument which requires neither sophisticated optical parts, nor strict alignment of optical components. Therefore, it is well suited to operate under the harsh environment imposed on an experiment in microgravity.

The main drawback of the shadowgraph method is that the instrumental transfer function \( T(\mathbf{q}) \) is oscillatory, in the Raman-Nath regime identified by the condition \( q^2 d / 2 k_o \leq \pi \), where \( d \) is the sample thickness, and \( k_o \) is the vacuum wave vector of the light. For an ideal instrument, the transfer function in this regime is given by \( T(\mathbf{q}) = \sin^2 (q^2 z / 2 k_o) \), where \( z \) is the visualization distance. Physically, one may picture a fluctuation of a given wave vector as acting like a very weak diffraction grating, generating two weak beams traveling at small angle relative to the main beam, one to either side. While these diffracted beams (one positive and one negative order) still overlap the transmitted beam, they interfere with it and produce fringes having a wave vector equal to that of the original fluctuation. The amplitude of this interference pattern is periodic in space as the transmitted beam propagates beyond the sample (Talbot effect). For many simultaneous fluctuations, spatial Fourier transformation of the interference pattern can be used to decompose the fluctuations.

A second concern of our early efforts was that of establishing a macroscopic concentration gradient in a binary mixture, which is the main ingredient required to generate non-equilibrium concentration fluctuations. In the early phase of the GRADFLEX project, a free diffusion scheme was tested. By using this scheme, it is possible to generate large step-like concentration gradients, which in turn give rise to very large scattering signals. This can be achieved either by using a critical mixture with a huge Soret effect, or by using a flowing junction cell. However, it is very difficult to create a flat interface between two phases in the absence of gravity, where perturbations of the interface can relax only by diffusion. For this reason we preferred to create a concentration gradient through the Soret effect simply by applying a temperature gradient. This method appeared to be more suitable for a microgravity experiment. However, it must be pointed out that Soret induced concentration gradients are generally smaller than those created by free diffusion, with the exception of near-critical mixtures and complex fluids such as colloidal suspensions and polymer solutions. In near critical fluids, \( k_f \) diverges (while \( D \) goes to zero) at the critical consolute point. This divergence has been profitably used in the past for the generation of large amplitude NEF. However, a refined absolute temperature control and some handling/storage precautions to avoid phase separation are necessary and this poses severe limitations to the use in an unmanned space mission such as onboard a FOTON mission. A good alternative is represented by macromolecular and polymer solutions, which exhibit reasonably large Soret coefficients and do not require absolute temperature control and demanding handling/storage procedures. For a given linear polymer, the product \( k_f D \) is roughly independent of the size of the polymer chain and therefore, by suitably increasing the molar mass of the polymer, the thermal diffusion ratio can be increased at will. The optimal choice must obviously take into account that in microgravity conditions even the largest wavelength fluctuations decay by diffusion alone and therefore a small value of the mass diffusion coefficient \( D \) might give rise to a fluctuation lifetime so large to impair any study of the dynamics at low wave vector.

We selected as a solution of polystyrene (molecular weight 9100) in toluene. This combination has been extensively studied on Earth, and its thermo-physical properties are available in the literature. In the case of the single-component fluid, the sample of choice was CS2, based on expected signal levels. The thickness of this sample was 3 mm.

The GRADFLEX payload employs sophisticated thermal gradient cells to create a thermal gradient in a single-component fluid and a concentration gradient in a polymer suspension. During the flight, temperature and concentration fluctuations were induced by...
applying temperature differences in the range 1K-30K to the CS$_2$ sample and to the mixture. A preliminary qualitative check of predictions involved the comparison of shadowgraph images taken on the Earth and in space under the same experimental conditions. Fig. 1 shows a comparison for the single-component fluid sample with a 16.7 K/cm temperature gradient. The left panel shows the fluctuations on Earth, while the right panel those observed in space. One can immediately appreciate the enhancement, both in amplitude and in size, of the temperature fluctuations.

A similar comparison is shown in Fig. 2 for non-equilibrium fluctuations in the polymer solution. In this case, the enhancement in the amplitude of long wavelength fluctuations due to microgravity is much larger, being only slightly smaller than two orders of magnitude.

A further quantitative analysis involved the processing of series of images like those shown in Figs. 1 and 2 to obtain a quantitative statistical characterization of the properties of the fluctuations. This was accomplished by Fourier transforming such ratio images spatially and squaring to calculate the mean-squared amplitude of the fluctuations $S(q)$ from sequences of images. Of course the result is the product of $S(q)$ and the transfer function. Inhomogeneities in the illumination and noise due to the CCD camera were characterized by means of independent measurements and subtracted.

Fig. 3 shows the results for $S(q)$ for the single-component liquid obtained on Earth and in space when the sample was subjected to a temperature difference of 5K. The mean-squared amplitude of the fluctuations is enhanced roughly by a factor six upon removing gravity. There is also an observable shift toward lower wave vector.

![Fig. 1](image1.png)

**Fig. 1** Shadowgraph images of non-equilibrium fluctuations in a 3 mm thick layer of CS$_2$ with a 16.7 K/cm temperature difference. The left/right panels correspond to images taken on Earth and in micro-gravity conditions, respectively. The side length of the images is 37.5 mm in real space.

![Fig. 2](image2.png)

**Fig. 2** Shadowgraph images of non-equilibrium fluctuations in a 1 mm thick layer of a polymer solution with a 200 K/cm temperature gradient. The left/right panels correspond to images taken on Earth and in micro-gravity conditions, respectively. The side length of the images is 13 mm in real space.

![Fig. 3](image3.png)

**Fig. 3** Mean-squared amplitude $S(q)$ of non-equilibrium fluctuations in a 3 mm layer of CS$_2$ with a 5 K temperature difference. The solid circles correspond to data taken on Earth, and the open circles to data taken in space. The amplitude of the signal is modulated by the shadowgraph transfer function $T(q)$.

![Fig. 4](image4.png)

**Fig. 4** Mean-squared amplitude $S(q)$ of non-equilibrium fluctuations in a 1 mm thick layer of a polymer solution under the action of a 20 K temperature difference. The dotted line corresponds to data taken on Earth, while the solid line to data taken in space. The amplitude of the signal is modulated by the shadowgraph transfer function $T(q)$. 
In the absence of gravity, wavelength fluctuations is limited by gravity on Earth. Divergence is also apparent. The divergence of long-wavelength fluctuations predicted to occur under microgravity conditions. Here small $q$ corresponds to long-wavelength fluctuations and large $q$ to small-wavelength ones. Small wavelength fluctuations are not affected by gravity. This is apparent from the merging of the two curves at large $q$, where the $q^4$ divergence is also apparent. The divergence of long-wavelength fluctuations is limited by gravity on Earth.

In the absence of gravity, $S(q)$ increases about two orders of magnitude at small $q$. A more refined analysis of this result will involve the assessment of the contribution of temperature fluctuations to the signal measured on Earth.

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