

Thermally induced non-equilibrium fluctuations: gravity and finite-size effects

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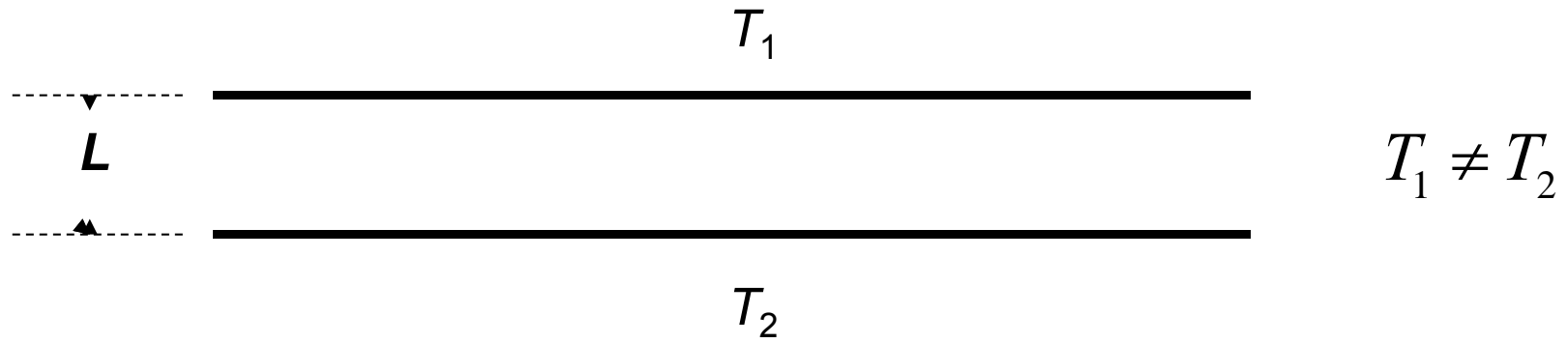


IWNET, Røros, Norway, August 19-24, 2012

Outline

- **1. Introduction: statement of the problem**
- **2. Non-equilibrium fluctuating hydrodynamics**
- **3. Light-scattering experiments**
- **4. Gravity effect on non-equilibrium fluctuations**
- **5. Gravity and finite-size effects near R-B instability**
- **6. Gravity and finite-size effects far away from R-B instability**

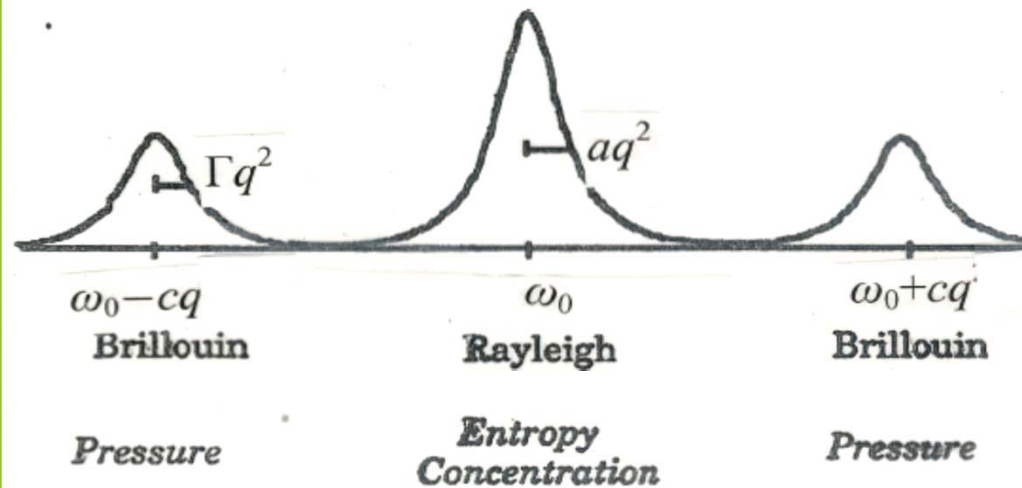
THERMAL FLUCTUATIONS IN FLUIDS



Light-Scattering Spectrum in Thermal Equilibrium

at constant pressure:

$$ds = \frac{c_p}{T} dT$$



$$I(q, \omega) \propto \left(\frac{\partial \rho}{\partial s} \right)_p^2 \langle \delta s^*(q, \omega) \delta s(q, \omega) \rangle + \left(\frac{\partial \rho}{\partial p} \right)_s^2 \langle \delta p^*(q, \omega) \delta p(q, \omega) \rangle$$

Rayleigh Line
Thermal Diffusion

+

Brillouin Lines
Sound Propagation

Fluctuating Hydrodynamics

Example: temperature evolution equation

(at constant pressure)

$$(\nabla \cdot \mathbf{v} = 0)$$

$$\rho c_p \left[\frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T \right] = -\nabla \cdot \mathbf{Q}$$

$$\mathbf{Q} = -\lambda \nabla T + \delta \mathbf{Q}$$

Linear phenomenological laws
are valid only “on average”:

$$\langle \delta \mathbf{Q} \rangle = 0$$

“**Fluctuating**” heat equation

$$\rho c_p \left[\frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T \right] = \lambda \nabla^2 T - \nabla \cdot \delta \mathbf{Q}$$

$$T = T_0 + \delta T(\mathbf{r}, t),$$

$$\mathbf{v} = \mathbf{0} + \delta \mathbf{v}(\mathbf{r}, t),$$

Thermal fluctuations in equilibrium

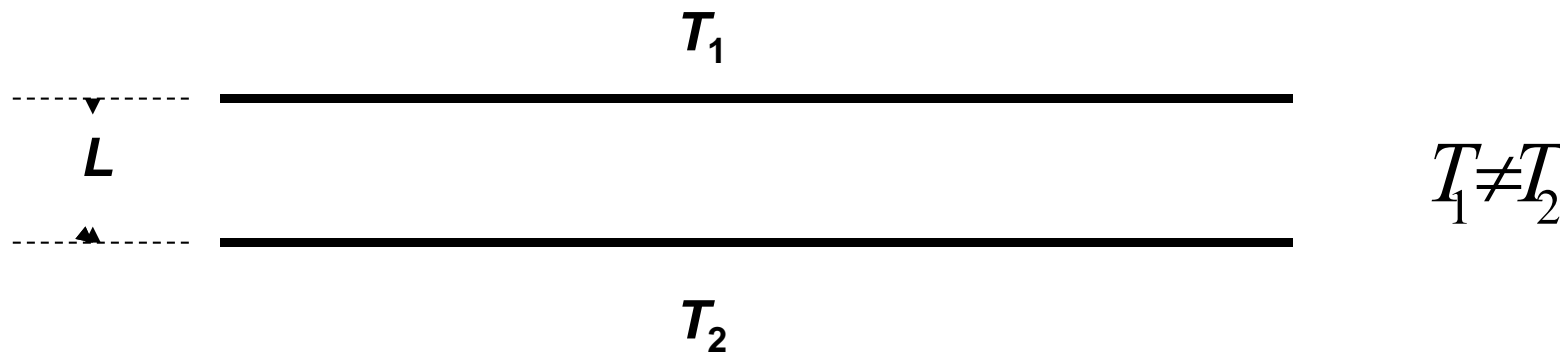
$$\rho c_p \frac{\partial \delta T}{\partial t} = \lambda \nabla^2 \delta T - \nabla \cdot \delta \mathbf{Q}$$

Fluctuation-dissipation theorem:

$$\langle \delta Q_i^*(\mathbf{r}, t) \cdot \delta Q_j(\mathbf{r}', t') \rangle = 2k_B \lambda T_0^2 \delta_{ij} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t')$$

$$\langle \delta T^*(q, t) \delta T(q, 0) \rangle = \frac{k_B T_0^2}{\rho c_p} \exp(-aq^2 t)$$

Thermal fluctuations in a temperature gradient



Rayleigh number:

$$R = \frac{\alpha L^4 \mathbf{g} \cdot \nabla T}{\nu a}$$

α is thermal expansion coefficient
 ν is kinematic viscosity
 $a = \lambda/\rho c_p$ is thermal diffusivity

Fluid in temperature gradient

$$\rho c_p \left[\frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T \right] = \lambda \nabla^2 T - \nabla \cdot \delta \mathbf{Q}$$

$$T = T_0 + \delta T(\mathbf{r}, t), \quad \mathbf{v} = \mathbf{0} + \delta \mathbf{v}(\mathbf{r}, t),$$

Fluctuating heat equation:

$$\rho c_p \left[\frac{\partial \delta T}{\partial t} + \delta \mathbf{v} \cdot \nabla T_0 \right] = \lambda \nabla^2 \delta T - \nabla \cdot \delta \mathbf{Q}$$

Fluctuating Navier-Stokes equation at constant pressure:

$$\frac{\partial \delta \mathbf{v}}{\partial t} = \nu \nabla^2 \delta \mathbf{v} + \frac{1}{\rho} \nabla \cdot \delta \mathbf{S}$$

Coupling between **heat** mode and **viscous** mode through ∇T_0

Assumption: **local equilibrium** for noise correlations

$$\langle \delta Q_i^*(\mathbf{r}, t) \cdot \delta Q_j(\mathbf{r}', t') \rangle = 2k_B \lambda T_0^2 \delta_{ij} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t')$$

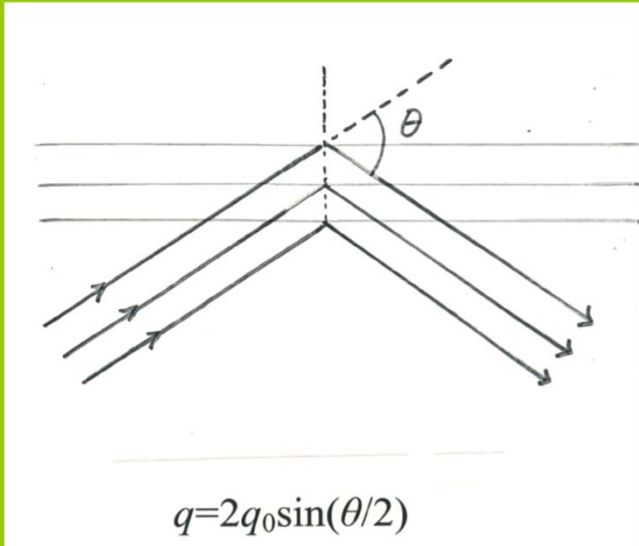
$$\begin{aligned} & \langle \delta S_{ij}^*(\mathbf{r}, t) \cdot \delta S_{kl}(\mathbf{r}', t') \rangle \\ &= 2k_B T_0 \rho v \left(\delta_{ij} \delta_{kl} + \delta_{il} \delta_{jk} \right) \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \end{aligned}$$

Fluids in a temperature gradient

$$C(t) = C_0 \left[(1 + A_T) \exp(-aq^2 t) - A_v \exp(-vq^2 t) \right]$$

$$A_T = \frac{c_p}{T_0(v^2 - a^2)} \frac{v}{a} \frac{(\nabla T_0)^2}{q^4} \quad A_v = \frac{c_p}{T_0(v^2 - a^2)} \frac{(\nabla T_0)^2}{q^4}$$

T.R. Kirkpatrick, J.R. Dorfman and E.G.D. Cohen, Phys. Rev. A 26, 995 (1982),
D. Ronis and I. Procaccia, Phys. Rev. A 26, 1812 (1982),
B.M. Law and J.V. Sengers, J. Stat. Phys. 57, 531 (1989).



Bragg-Williams condition

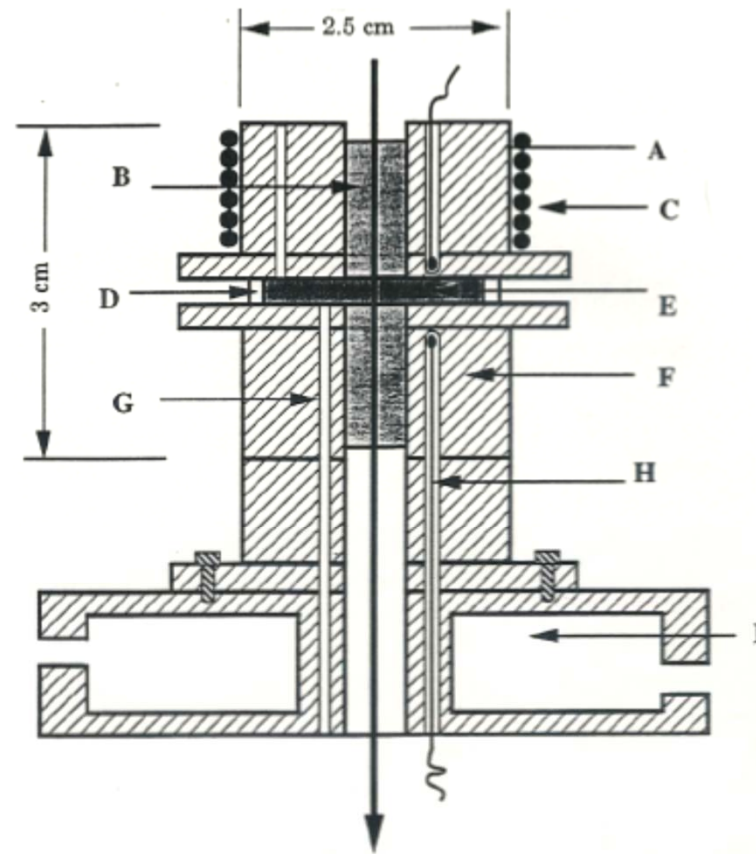
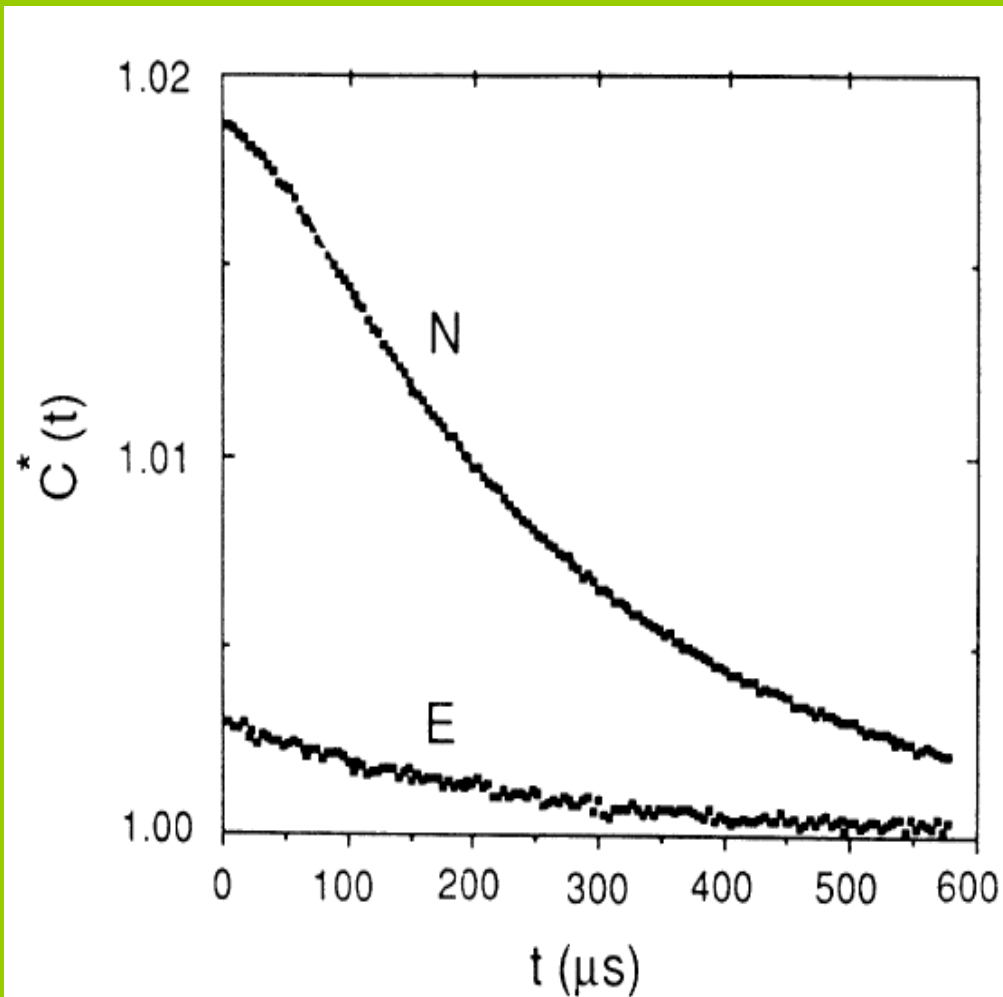


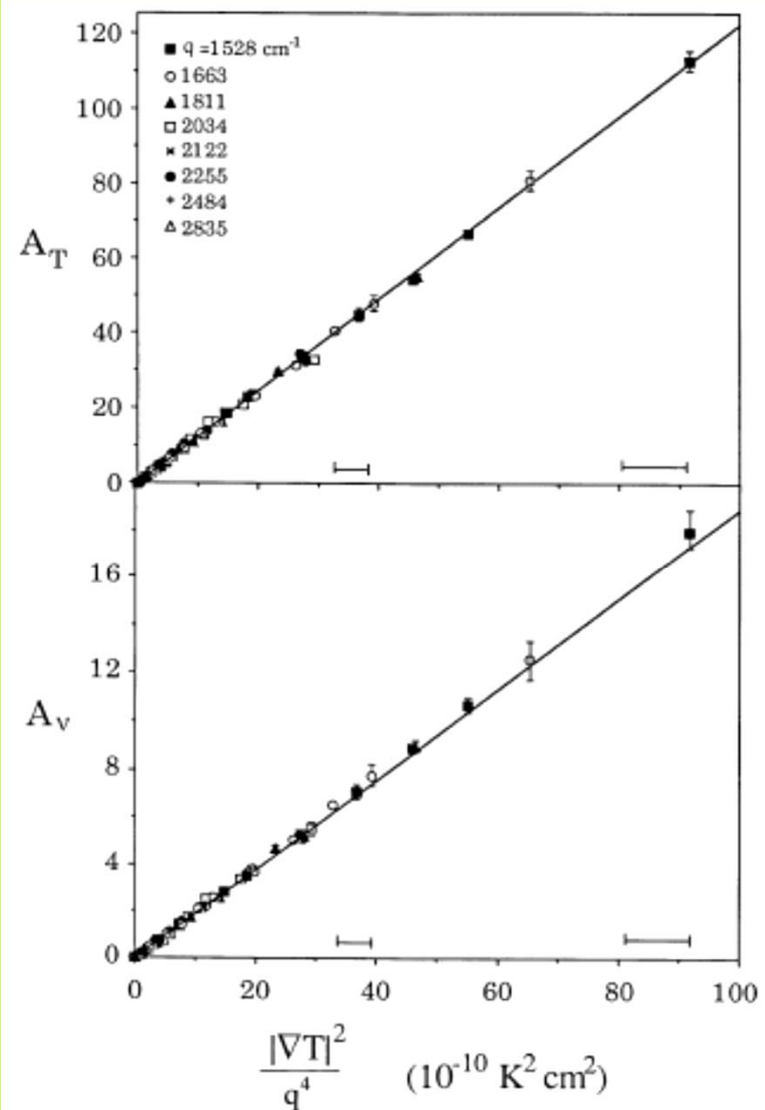
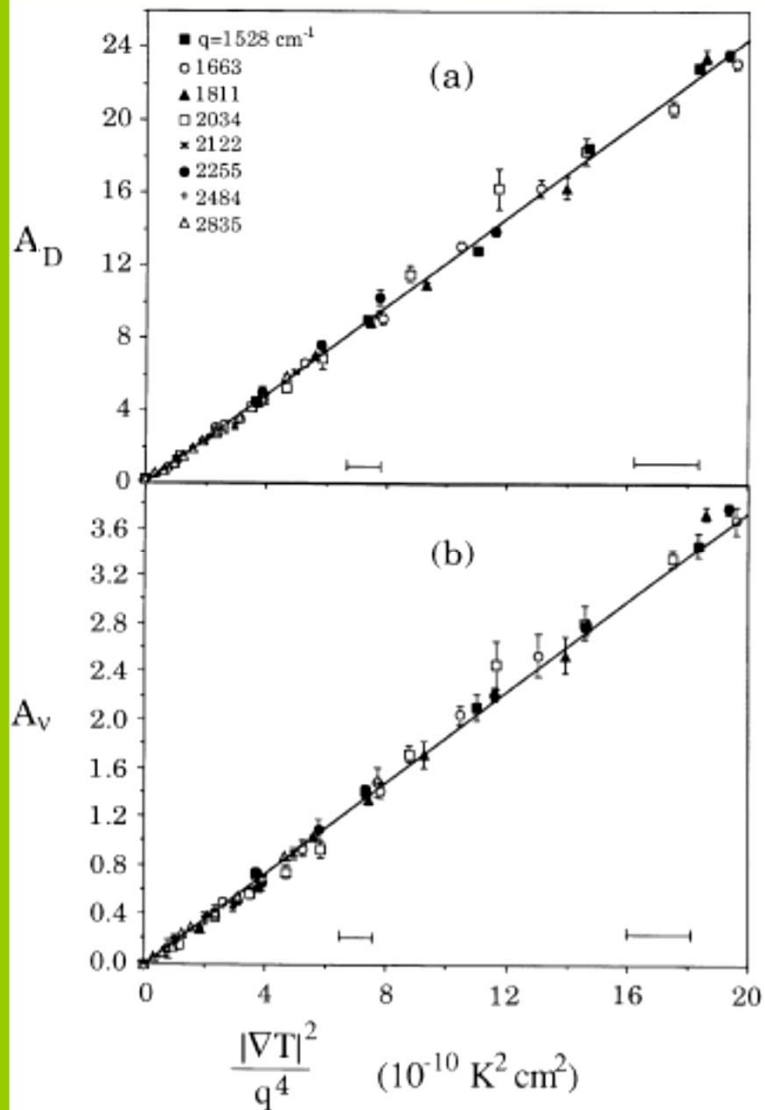
Figure 7.1 Optical cell for small-angle Rayleigh-scattering experiment. A: top plate. B: fused-silica window. C: Minco resistive heater. D: fused-quartz ring. E: sample liquid. F: bottom plate. G: liquid filling tube. H: thermistor. I: water circulating chamber. The laser passes through the liquid vertically.

$$C(t) = C_0 \left[(1 + A_T) \exp(-D_T q^2 t) - A_V \exp(-\nu q^2 t) \right]$$



Toluene
 $q=2255 \text{ cm}^{-1}$, $\nabla T=220 \text{ K/cm}$

Law, Segrè, Gammon, Sengers,
Phys. Rev. A **41**, 816 (1990)



$$A_T = \frac{c_p}{T(v^2 - D_T^2)} \frac{\nu}{D_T} \frac{(\nabla T)^2}{q^4}$$

$$A_V = \frac{c_p}{T(v^2 - D_T^2)} \frac{(\nabla T)^2}{q^4}$$

Segrè, Gammon, Sengers, Law, Phys. Rev. A **45**, 714 (1992)

Thermal fluctuations in a binary fluid

Decay rate of viscous fluctuations νq^2

Decay rate of thermal fluctuations $a q^2$

Decay rate of concentration fluctuations $D q^2$

In liquids: $\nu > a/D$

Lewis number

$$Le = a/D$$

Fluid mixtures in a concentration gradient

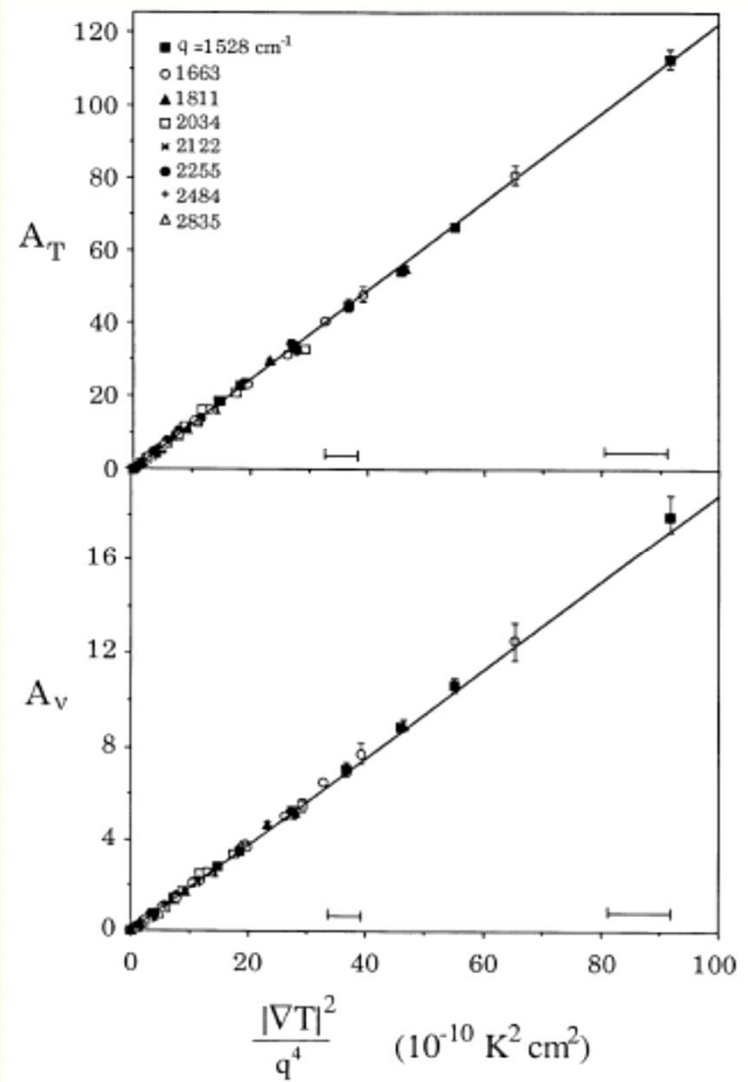
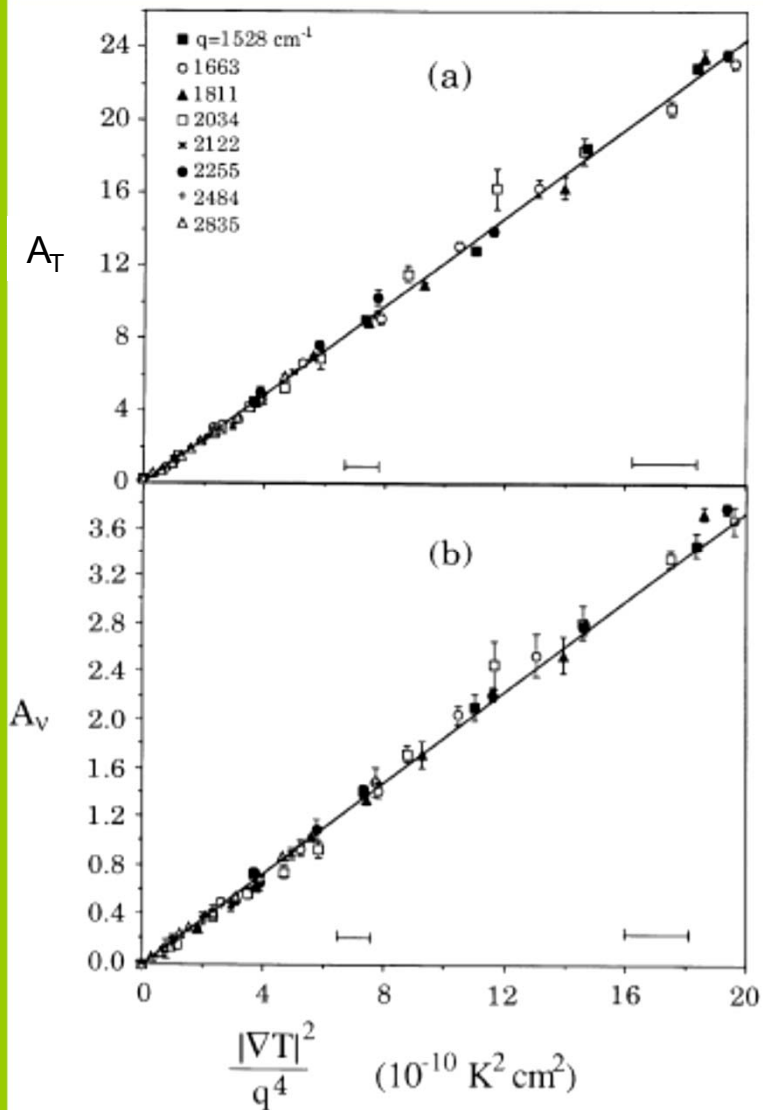
$$\frac{\partial}{\partial t} \delta c + \delta \mathbf{v} \cdot \nabla c_0 = D \nabla^2 \delta c - \frac{1}{\rho} \nabla \cdot \delta \mathbf{J}$$

$$\frac{\partial \delta \mathbf{v}}{\partial t} = \nu \nabla^2 \delta \mathbf{v} + \frac{1}{\rho} \nabla \cdot \delta \mathbf{S}$$

$\delta \mathbf{J}$ is fluctuating mass-diffusion flux

$$\langle \delta J_i^*(\mathbf{r}, t) \cdot \delta J_j(\mathbf{r}', t') \rangle = 2k_B T_0 \rho D \left(\frac{\partial c}{\partial \mu} \right)_{T,P} \delta_{ij} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t')$$

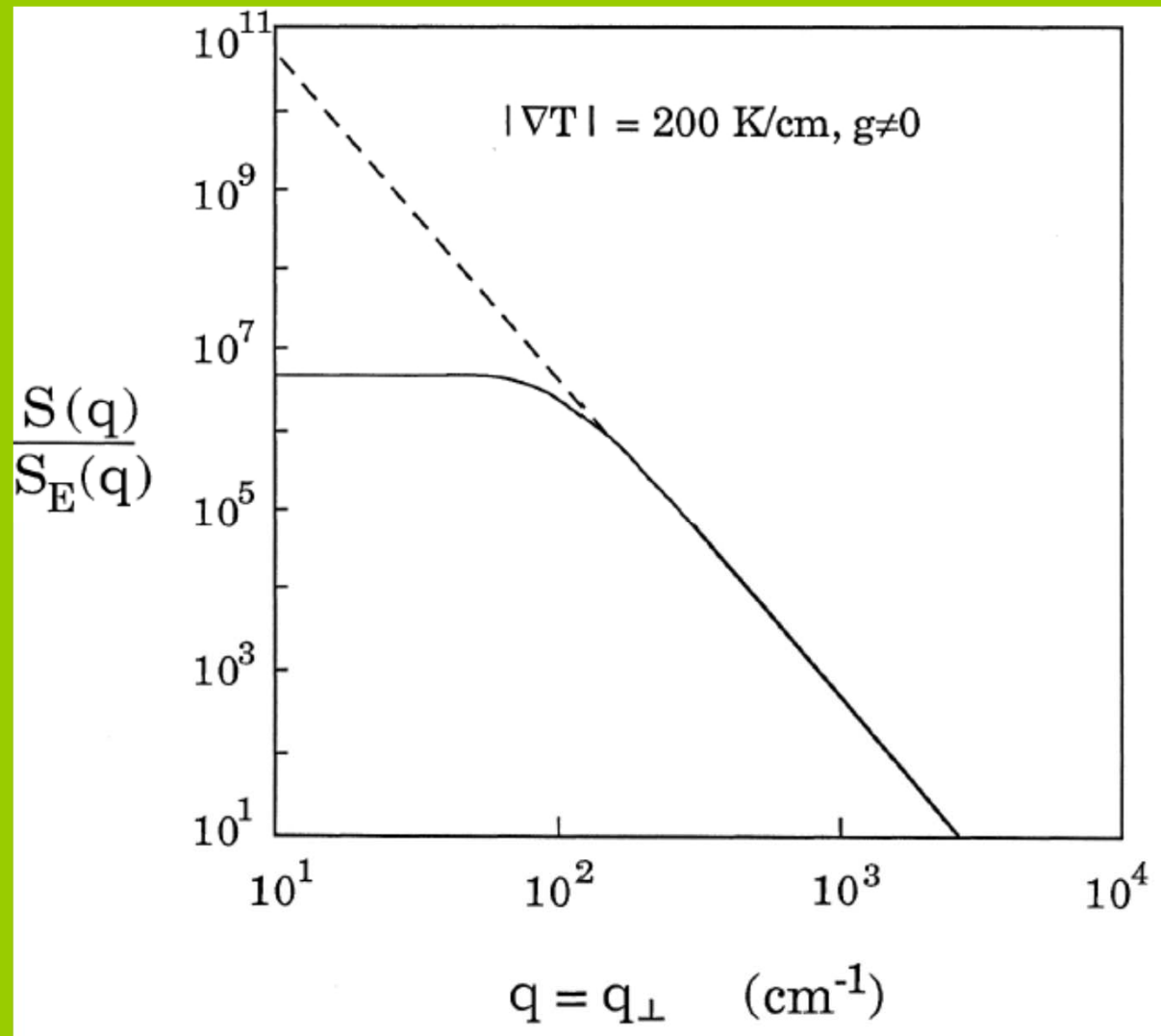
Coupling between **concentration** mode and **viscous** mode through ∇c_0



$$A_T = \frac{c_p}{T(v^2 - a^2)} \frac{v}{a} \frac{(\nabla T)^2}{q^4}$$

$$A_V = \frac{c_p}{T(v^2 - a^2)} \frac{(\nabla T)^2}{q^4}$$

Segrè, Gammon, Sengers, Law, Phys. Rev. A **45**, 714 (1992)



P.N. Segrè, R. Schmitz, J.V. Sengers, *Physica A* **195**, 31 (1993)

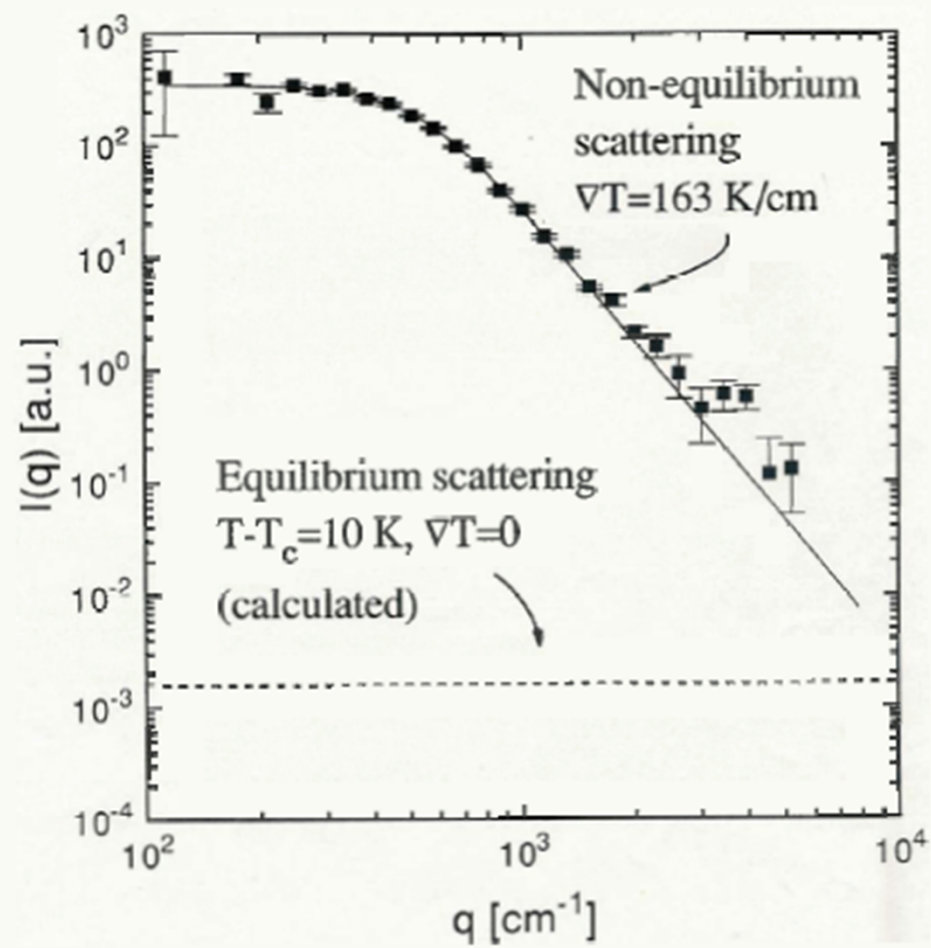
NONEQUILIBRIUM CONCENTRATION FLUCTUATIONS

EFFECT OF GRAVITY

$$S_{\text{NE}} = S_{\text{NE}}^0 \left[\frac{1}{1 + (q / q_{\text{RO}})^4} \right]$$

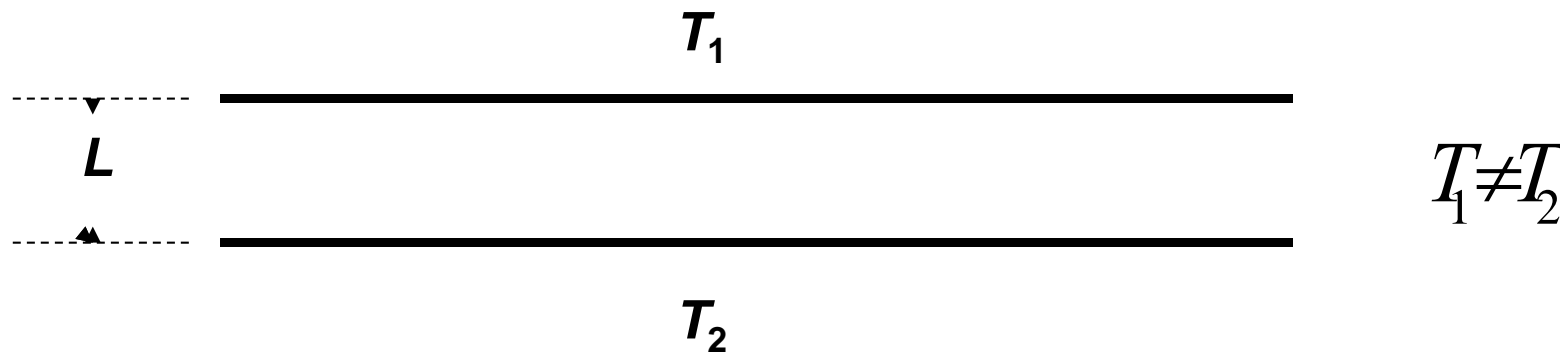
One-component: $q_{\text{RO}}^4 = \frac{1}{\nu \lambda \rho} \left(\frac{\partial \rho}{\partial T} \right)_P \mathbf{g} \cdot \nabla T_0$

Mixture: $q_{\text{RO}}^4 = \frac{1}{\nu D \rho} \left(\frac{\partial \rho}{\partial c} \right)_T \mathbf{g} \cdot \nabla c_0$



A. Vailati and M. Giglio, Phys. Rev. Lett. 77, 1484 (1996)

Thermal fluctuations in a temperature gradient



Rayleigh number:

$$R = \frac{\alpha L^4 \mathbf{g} \cdot \nabla T}{\nu a}$$

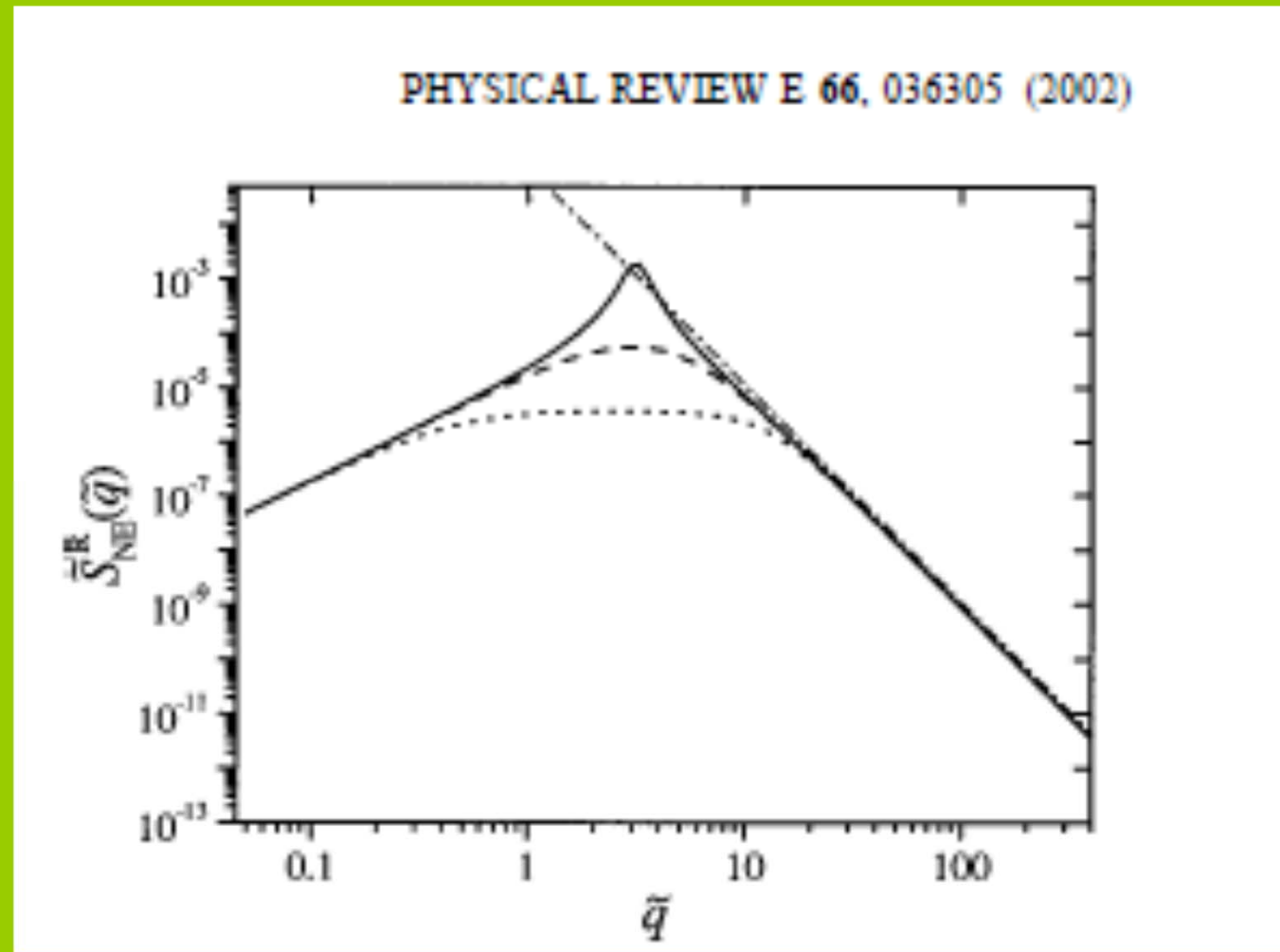
α is thermal expansion coefficient
 ν is kinematic viscosity
 $a = \lambda/\rho c_p$ is thermal diffusivity

J.M. Ortiz de Zaráte, J.V. Sengers,

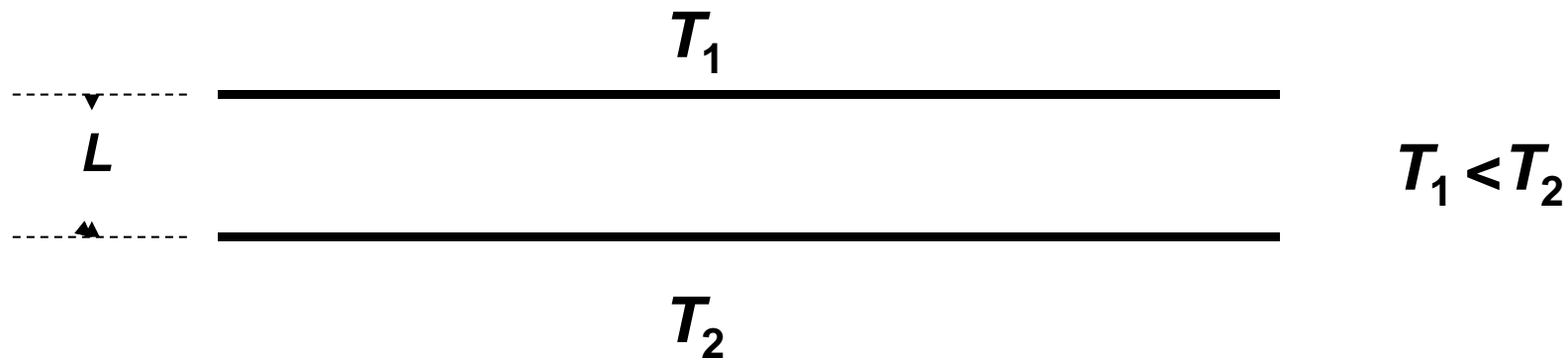
Solid curve:
 $R=1700$

Dashed curve:
 $R=0$

Dotted curve:
 $R=-25,000$



Thermal fluctuations in a temperature gradient: Heated from below



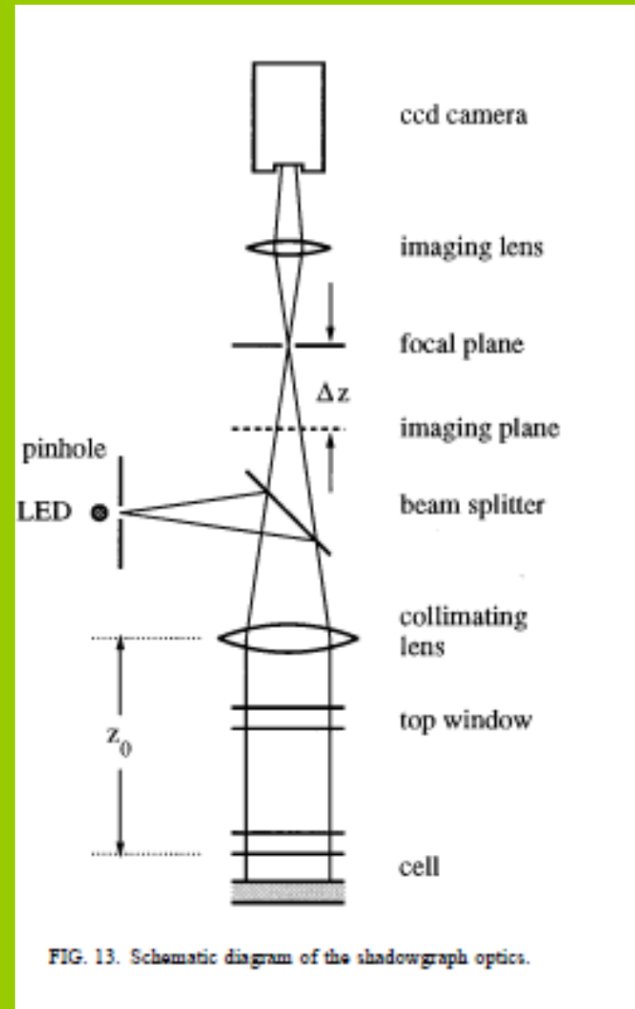
Rayleigh number:

$$R = \frac{\alpha L^4 \mathbf{g} \cdot \nabla T}{\nu a} \quad (\text{positive})$$

α is thermal expansion coefficient
 ν is kinematic viscosity
 $a = \lambda/\rho c_p$ is thermal diffusivity

Shadowgraphy

J.R. de Bruyn, E. Bodenschatz,
S.W. Morris, S.P. Trainoff, Y. Hu,
D.S. Cannell, G. Ahlers,
Rev. Sci. Instrum. **67**, 2043 (1996)



J.Oh, J.M.Ortiz de Zárate, J.V.Sengers, G.Ahlers
Phys. Rev. E **69**, 021106 (2004)

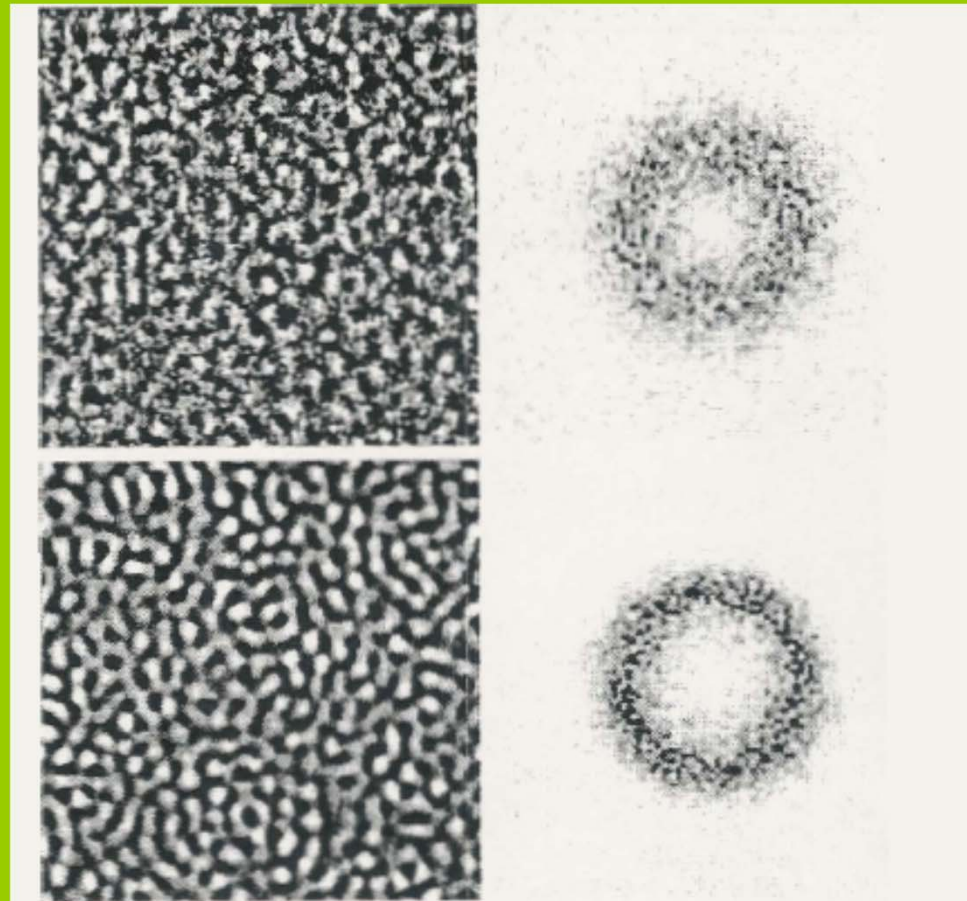
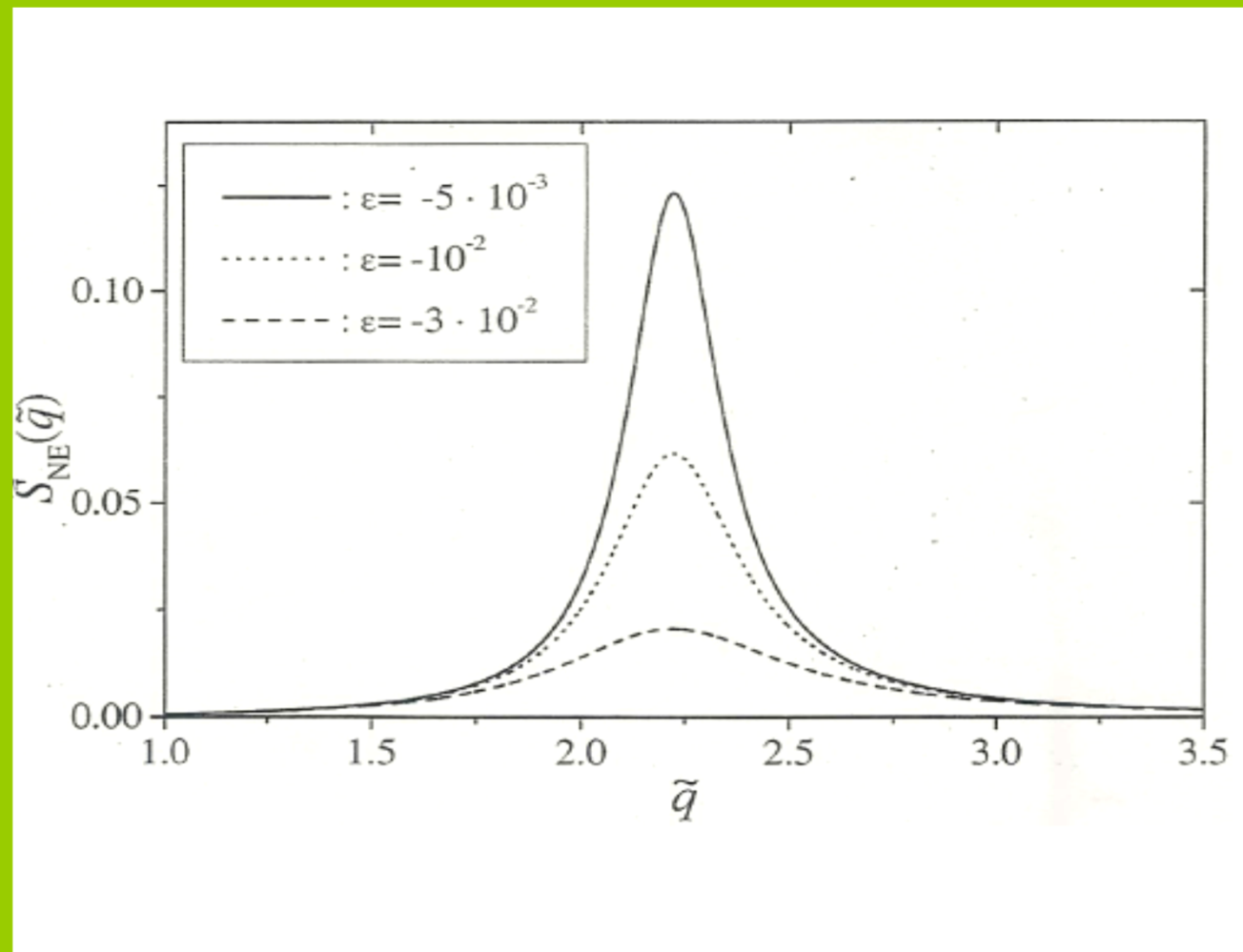


FIG. 5. Shadowgraph signals (left column, 1.3×1.3 mm²) and the moduli squared of their Fourier transforms (right column) for $\Delta T = 0.189$ (top row) and $\Delta T = 0.378$ K (bottom row). The exposure time was 500 ms.



$$\varepsilon = \frac{R - R_c}{R_c}$$

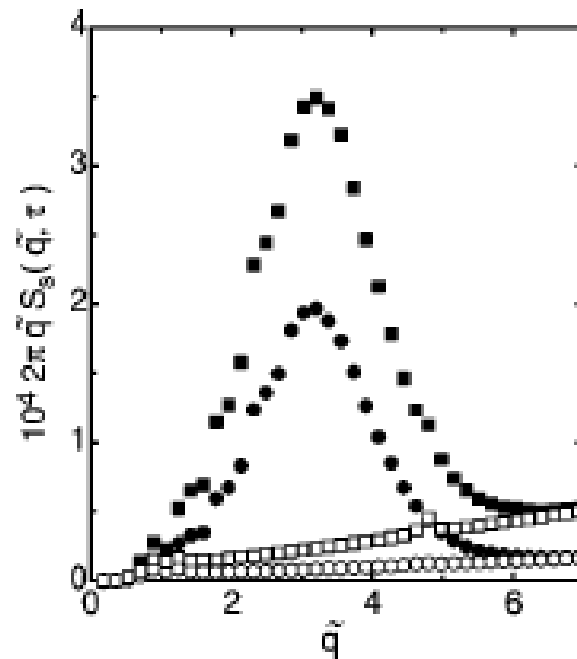


FIG. 6. Experimental shadowgraph structure factor $2\pi\tilde{q}S_s(\tilde{q},\tau)$ as a function of the dimensionless wave number \tilde{q} . These results are for $P=38.325$ bars, $\bar{T}=46.5$ °C, and $\Delta T=0.189$ °C. The solid squares (solid circles) are for $\tau=0.200$ s ($\tau=0.500$ s). The open symbols are the corresponding background measurements for $\Delta T=0$.

Oh, Ortiz de Zárate, Sengers, Ahlers, Phys. Rev. E **69**, 021106 (2004)

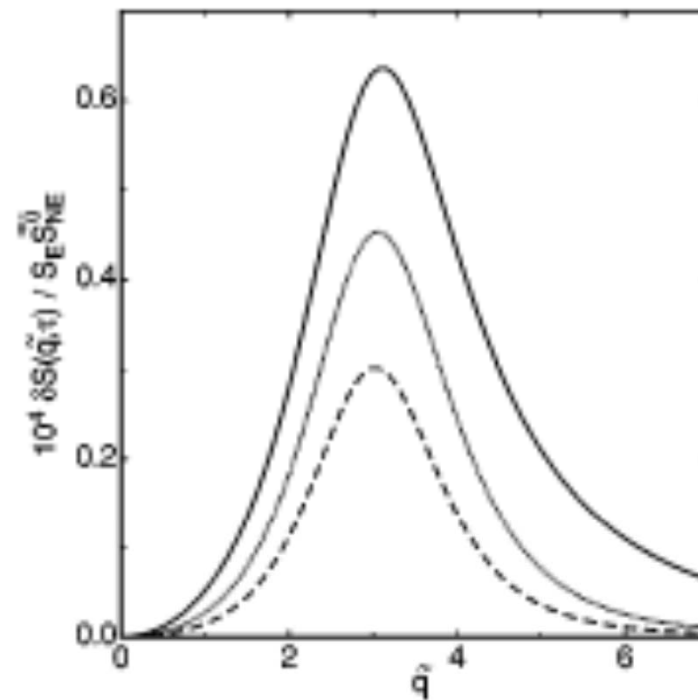


FIG. 1. Difference $\delta S(\bar{q}, \tau) / S_E S_{NE}^0$ [see Eq. (16) below] between the theoretical structure factors at $R=1371$ [Eq. (14)], and at $R=0$ [Eq. (15)] as a function of \bar{q} , for three different collecting times. The solid curve corresponds to $\tau=0$ ms, the dashed curve to $\tau=200$ ms, and the dotted curve to $\tau=500$ ms. The Prandtl number is $\sigma=34$ and $t_v=0.74$ s.

Oh, Ortiz de Zárate, Sengers, Ahlers, Phys. Rev. E **69**, 021106 (2004)

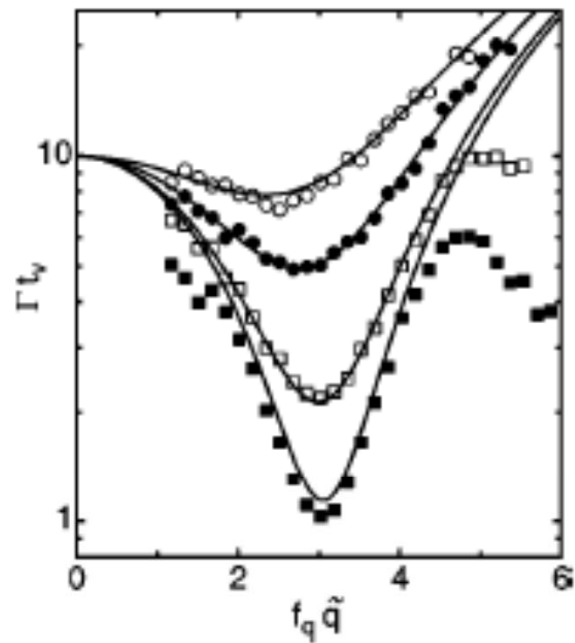
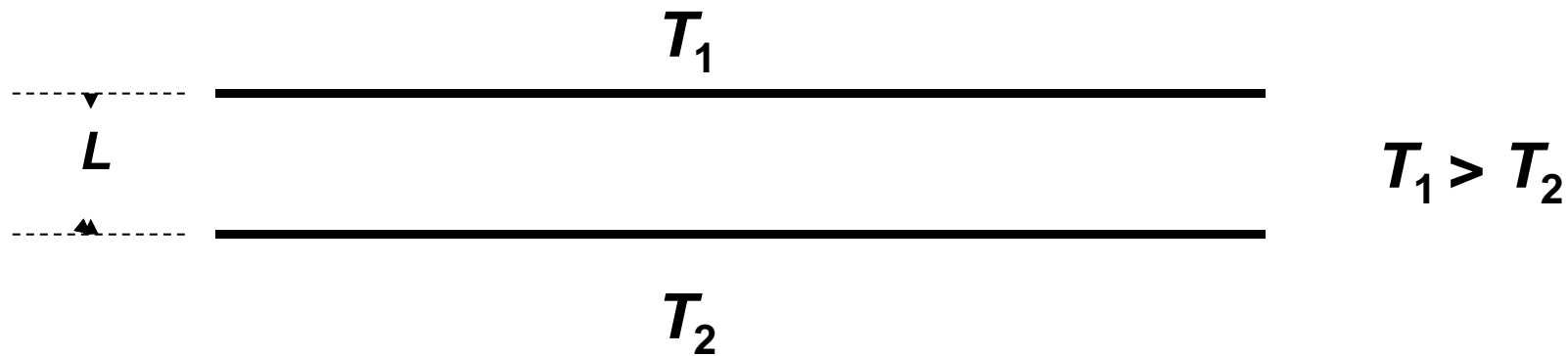


FIG. 9. Results for Γt_v as a function of $f_q \tilde{q}$, using the values $t_v = 0.551$ s and $f_q = 0.944$ from the least-squares fit described in the text. The data and symbols correspond to those in Fig. 8. The curves indicate the corresponding theoretical results obtained from Eq. (8) by using the scale factors of the Rayleigh number $f_{R,k}$ from the least-squares fit.

Oh, Ortiz de Zárate, Sengers, Ahlers, Phys. Rev. E **69**, 021106 (2004)

Thermal fluctuations in a temperature gradient: Heated from above



Rayleigh number:
$$R = \frac{\alpha L^4 \mathbf{g} \cdot \nabla T}{\nu a} \quad (\text{negative})$$

α is thermal expansion coefficient
 ν is kinematic viscosity
 $a = \lambda/\rho c_p$ is thermal diffusivity

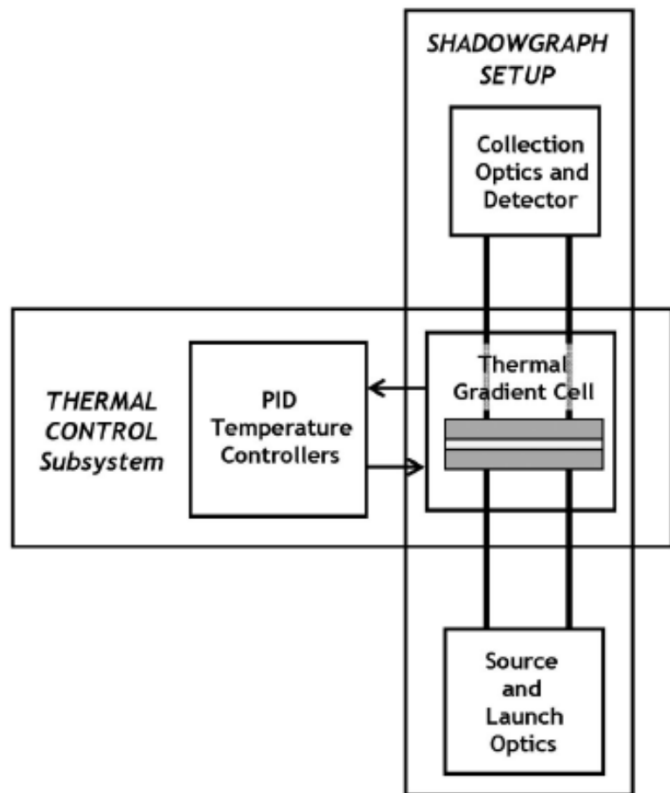


Fig. 2. Block diagram of the overall setup for the GRADFLEX flight experiment.

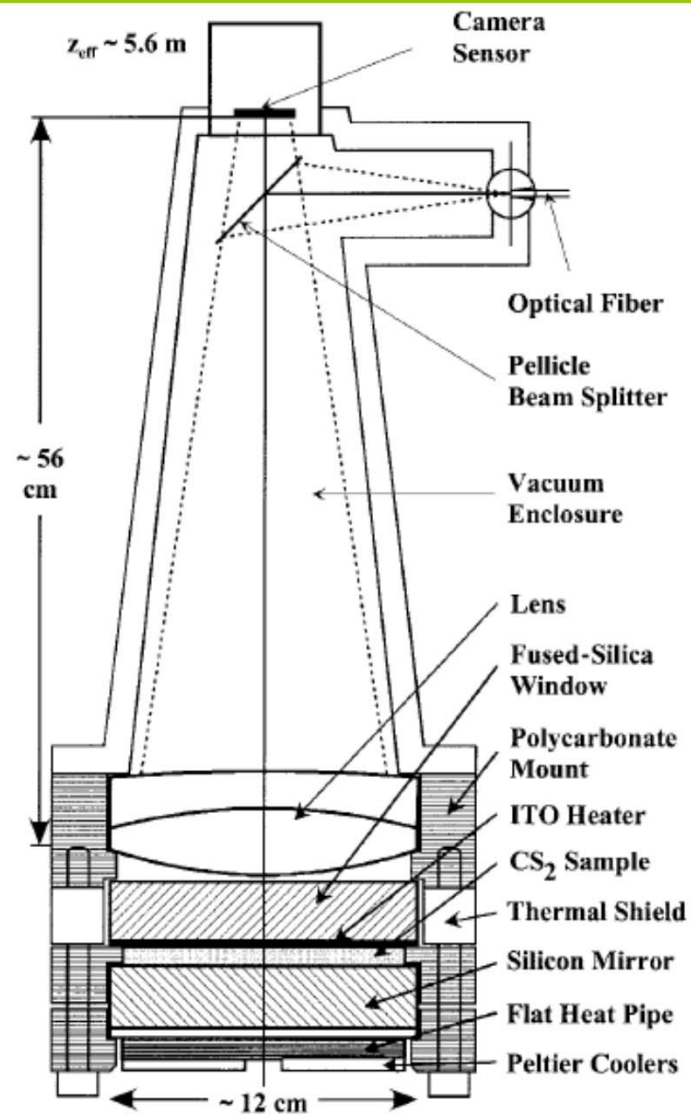
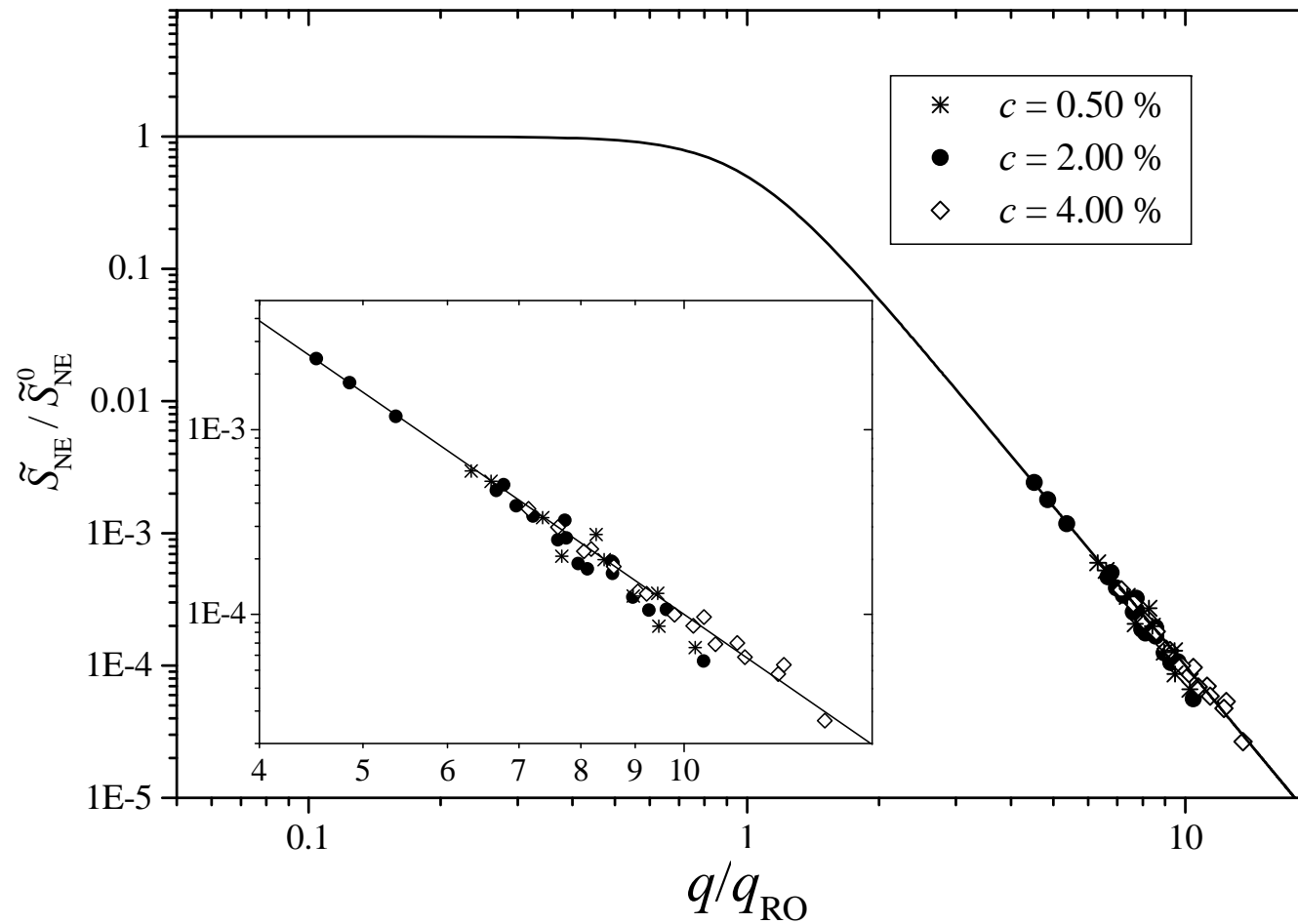


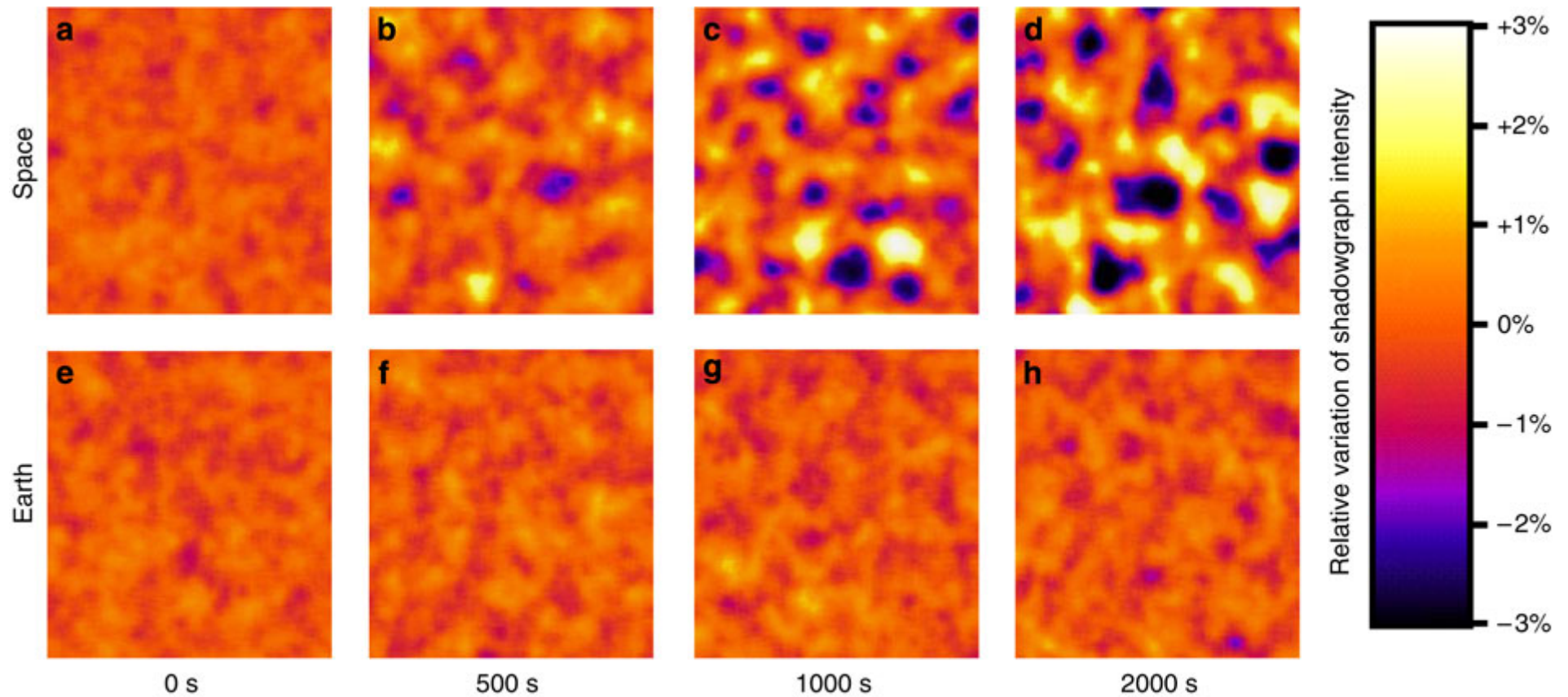
Fig. 3. Schematic diagram of the prototype apparatus used to measure fluctuations in a single-component fluid heated from above. The upper surface of the sample is heated via the transparent ITO film, and the heat is removed from the lower surface of the silicon mirror by means of four Peltier modules. The optical path is evacuated to eliminate optical disturbances from air.

Vailati, Cerbino, Mazzoni, Giglio, Nikolaenko, Takacs, Cannell, Meyer, Smart, Applied Optics 45, 2155 (2006)

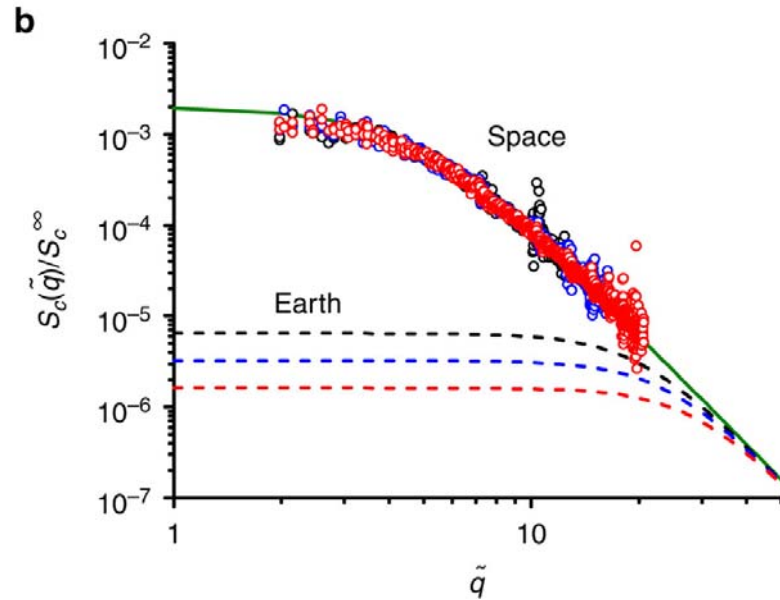
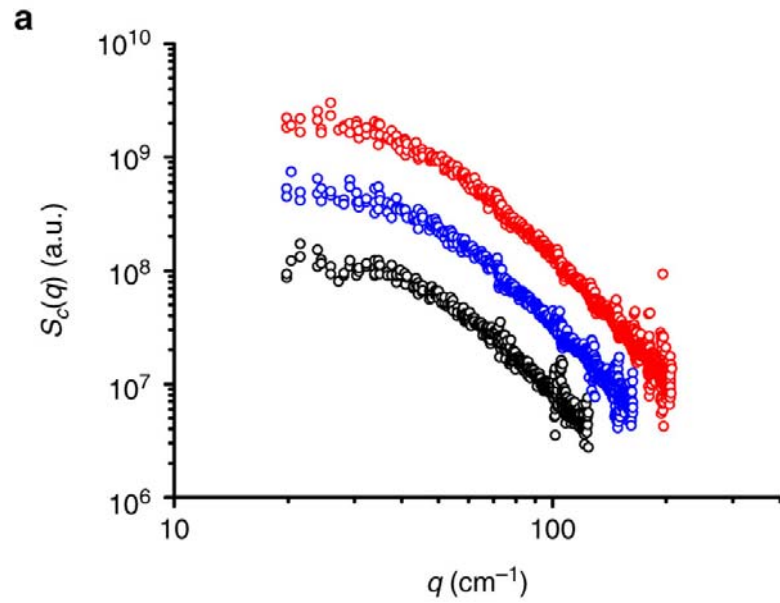


polystyrene-toluene solutions

polystyrene-toluene solution $\Delta T=17.40$ K



A. Vailati, R. Cerbino, S. Mazzoni, C.J. Takacs, D.S. Cannell, M. Giglio
Nature Communications **2**, article #290 (19 April, 2011)



A. Vailati, R. Cerbino, S. Mazzoni,
 C.J. Takacs, D.S. Cannell, M. Giglio
 Nature Communications **2**,
 article #290 (19 April, 2011)

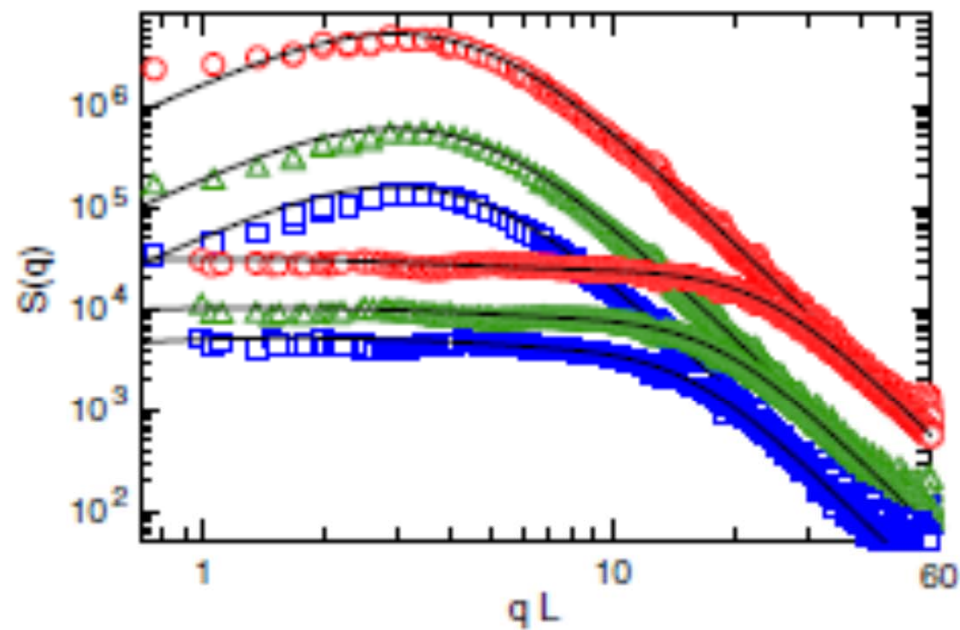


FIG. 4 (color online). Log-log plots of experimental results for $S(q)$ vs qL , with applied gradients of 17.9 (squares), 34.5 (triangles), and 101 (circles) K/cm, in microgravity (upper curves) and on Earth. The lines are the theoretical predictions.

CS_2

C.J. Takacs, A. Vailati, R. Cerbino, S. Mazzoni, M. Giglio, D.S. Cannell
PRL 106, 244502 (2011)

Conclusions

- **Validity of non-equilibrium fluctuating hydrodynamics has been confirmed experimentally by light scattering and shadowgraphy**
- **Thermal fluctuations exhibit always a strong non-equilibrium enhancement**
- **Non-equilibrium fluctuations are always long range encompassing the entire system**
- **Non-equilibrium fluctuations on earth are affected by gravity**
- **Non-equilibrium fluctuations are affected by the finite size of the system**