NEUF-DIX

Understanding Diffusion in Complex Fluids

TEAM MEMBERS			
Europe		**World**	**Industry**
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		ESA-Estec (INT) Marco Braibanti	

MASS TRANSFER IN LIQUIDS

Fundamental processes:

- **Diffusion**
- **Convection** (absent in μg)

- Growth and processing of materials (crystal growth)
- Trasport at the cellular level
- Rate of chemical reactions

Fickean diffusion: ∇c

Thermal diffusion: $\nabla T \Rightarrow \nabla c$

Mass diffusion

Adolf Fick

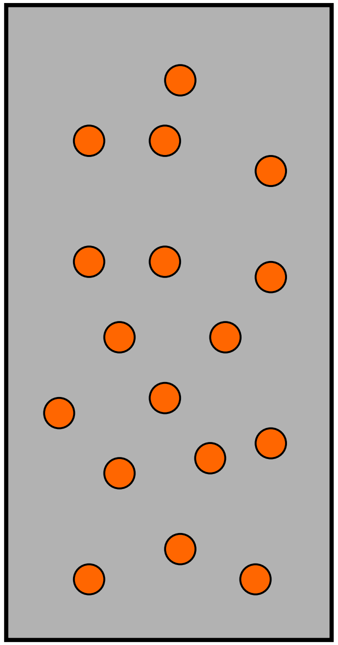
1855

$$j = -D\ \nabla c$$

NON-EQUILIBRIUM FLUCTUATIONS

Understanding diffusion at the mesoscopic scale

Microscopic scale



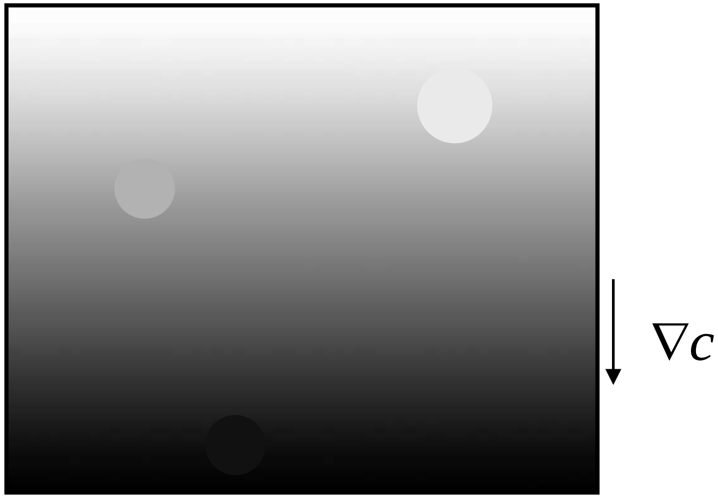
Robert Brown
1827

Albert Einstein
1905

Jean Perrin
1908

Thermal motion

Mesoscopic scale

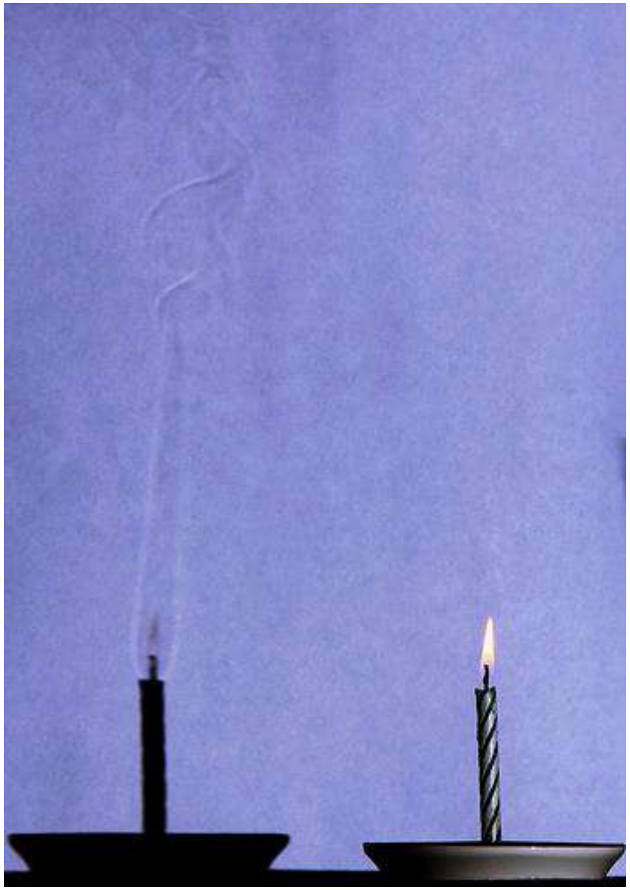


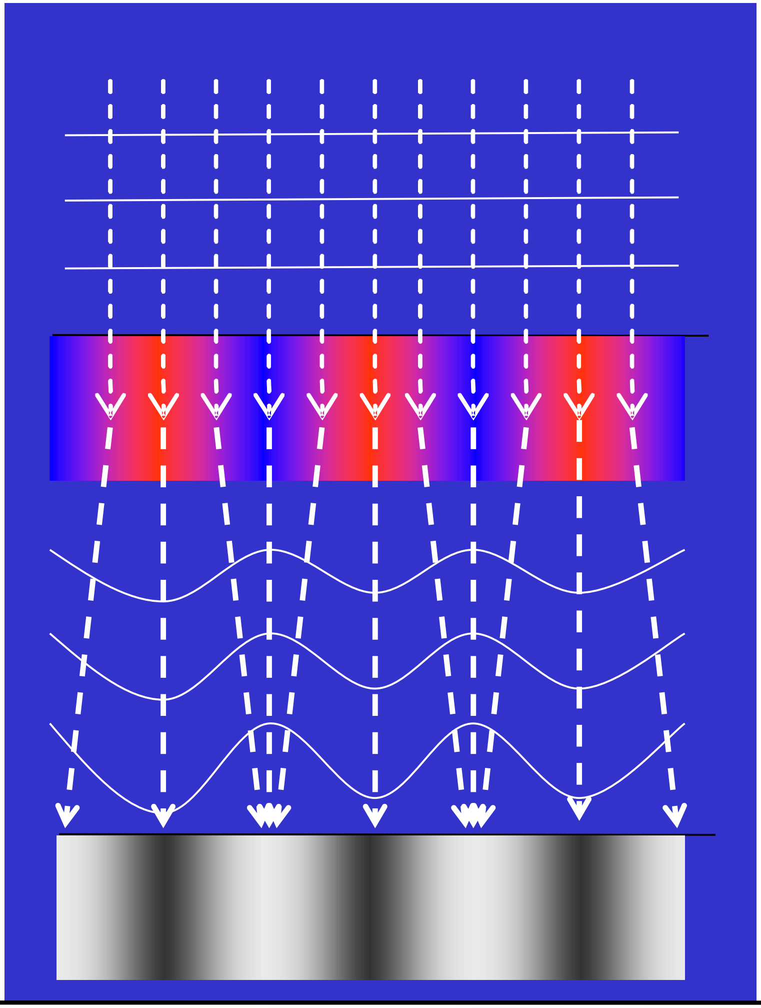
Velocity fluctuations
induce
concentration fluctuations

T. R. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, Phys. Rev A **26**, 1812 (1982)

OPTICAL DETECTION

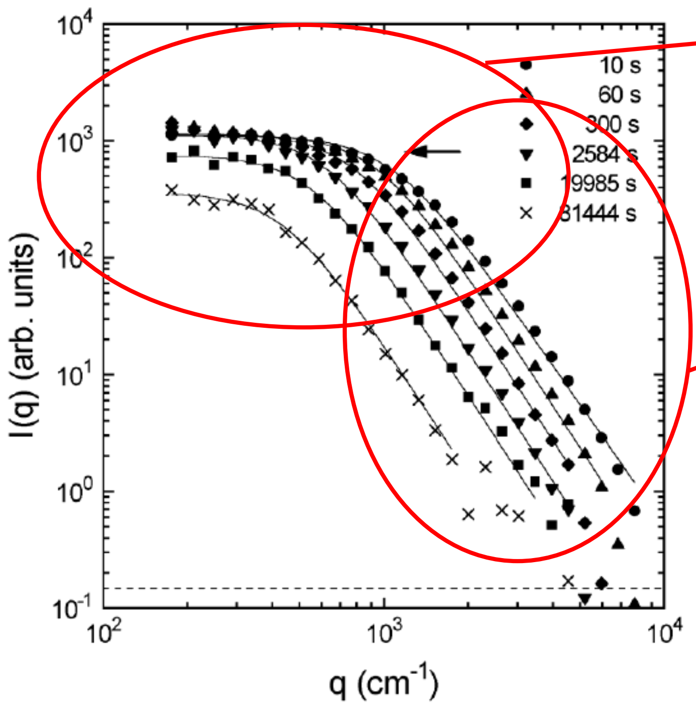
Shadowgraph





FLUCTUATIONS ON EARTH DURING DIFFUSION

Mean squared amplitude of fluctuations



A. Vailati and M. Giglio, "Giant fluctuations in a free diffusion process", Nature ***390****, 262 (1997)*

P. N. Segré and . V. Sengers, Physica A, 46, (1993)

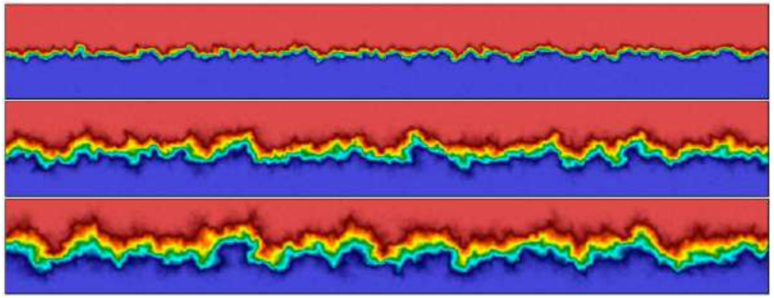
Gravitational stabilization

$I(q)=const$

Power law scaling

$I(q) \propto q^{-4}$ Self affine structure of the fronts of diffusion

2D diffusion in liquids with g=0

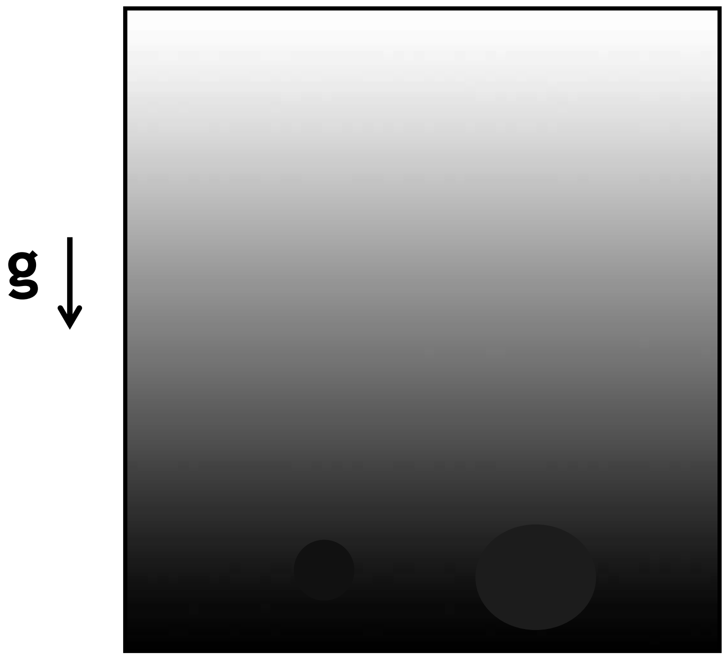


F. Balboa et al., SIAM J. Multiscale Modeling and Simulation, 10(4):1369-1408, (2012)

NEED FOR MICROGRAVITY

Gravitational Stabilization of Fluctuations

Mesoscopic scale



Dynamics of fluctuations dominated by buoyancy at small q:
quenching or amplification

Time scales

Diffusion $$\tau_{diff} = \frac{1}{Dq^2}$$

Buoyancy $$\tau_{grav} = \frac{\upsilon q^2}{\beta g \nabla c}$$

Rolloff wave vector

$$\tau_{diff} = \tau_{grav} \quad \Rightarrow \quad q_{RO} = \sqrt[4]{\frac{\beta g \nabla c}{\nu D}}$$

Università degli Studi di Milano · CNR - INFM · UCSB University of California Santa Barbara · esa · NASA · asi agenzia spaziale italiana · FOTON M3

RUAG Aerospace Defence Technology

OHB System An OHB Technology AG Company

VERHAERT

HIGHFTECH

PS in toluene

9100 MW, 1.8% w/w

1mm thickness

ΔT=20K

GRADFLEX

FOTON M3

14-26 September 2007

GRADFLEX

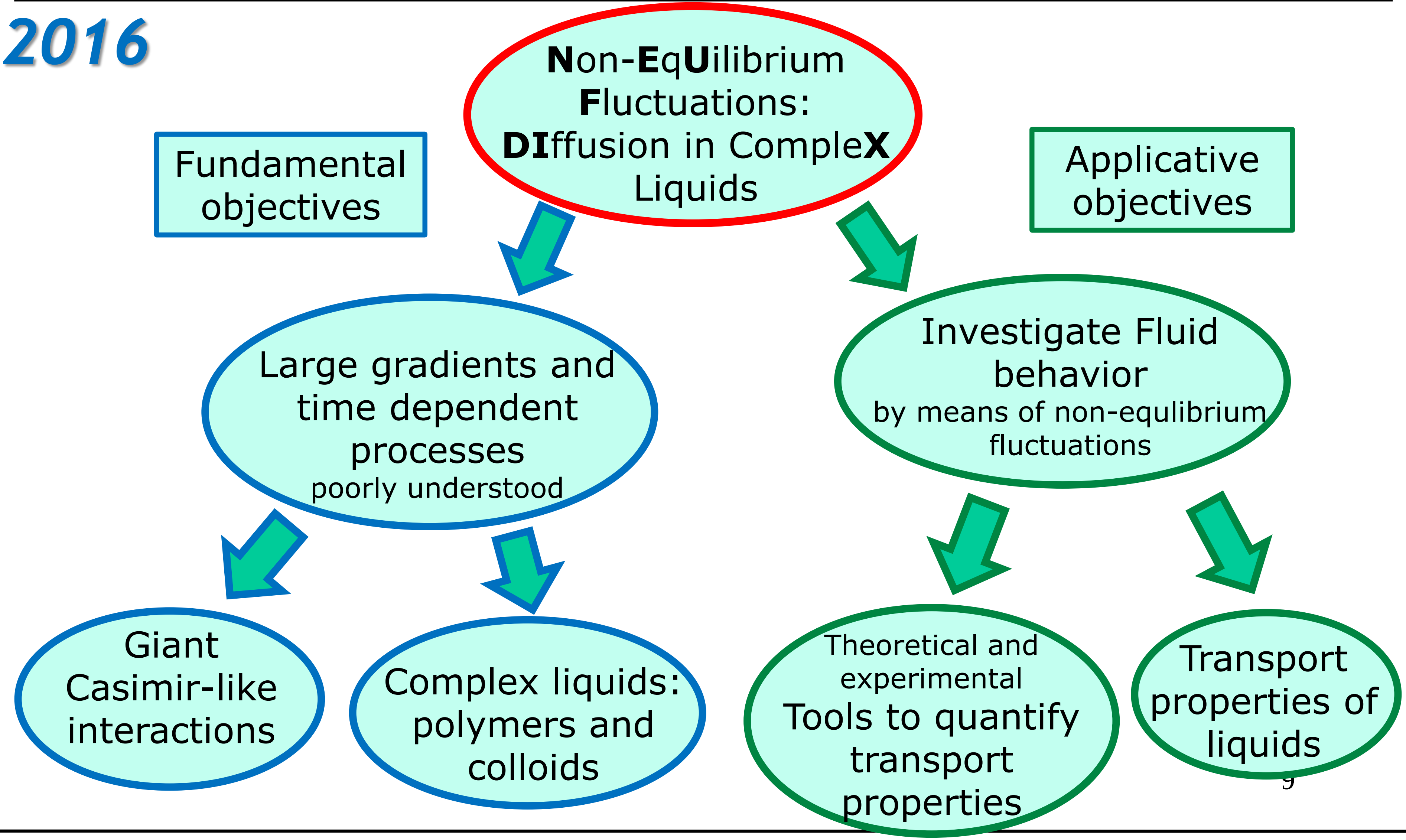
GRadient Driven FLuctuation EXperiment

SCIENTIFIC RETURN

- Linearized hydrodynamics: <u>small gradients, steady state</u>
- Diffusive dynamics of fluctuations
- Finite size effects

OVERALL OBJECTIVES

esa
European Space Agency



OUTSTANDING NOVEL EXPERIMENTS

1. Casimir forces out of equilibrium
2. Non-equilibrium fluctuations in a complex mixture including a polymer
3. Glass transition in a complex mixture including a polymer
4. Transient fluctuations: spinodal-like dynamics
5. Colloids: static and dynamic properties

Non-equilibrium Casimir forces

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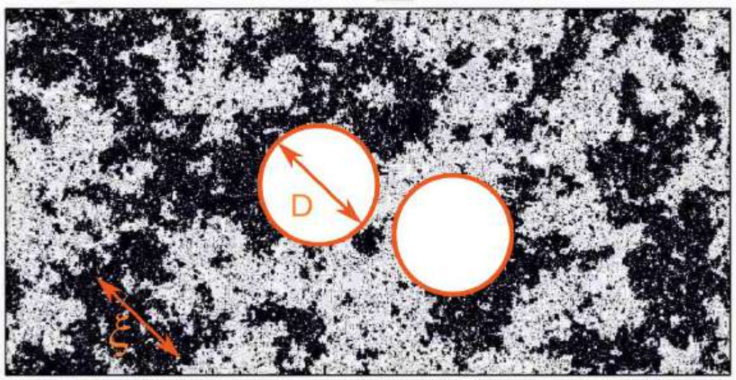
Classical critical Casimir effect

M. Fisher and P. G. De Gennes,
C. R. Acad. Sci. B 287, 207-209 (1978)

- correlation length ξ diverges near critical point
- long ranged fluctuations under confinement
- Scale invariant fluctuations at critical point



Credits: J. R. Nelson

TABLE I. Estimated Casimir pressures.

		$L = 10^{-6}$ m	$L = 10^{-4}$ m
p_{em}[a]		-1×10^{-3} Pa	-1×10^{-11} Pa
p_c[b]		-6×10^{-4} Pa	-6×10^{-10} Pa
p^w_{NE}	toluene + n-hexane[c]	$+2 \times 10^{-1}$ Pa	$+2 \times 10^{-3}$ Pa
p^w_{NE}	1-methylnaphtalene + n-heptane[c]	+9 Pa	$+0.9 \times 10^{-1}$
p^w_{NE}	aniline + methanol[c]	-3×10^{-1} Pa	-3×10^{-3} Pa

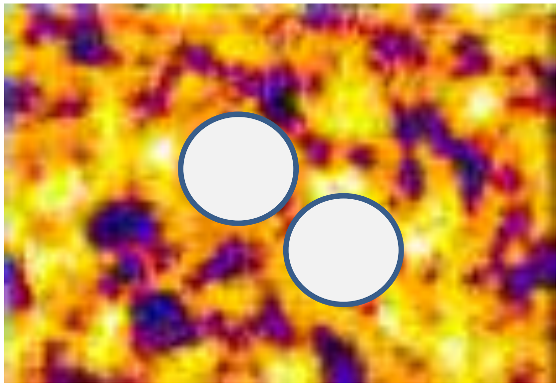
Non-equilibrium Casimir effect

Kirkpatrick, Ortiz de Zárate, Sengers,
Phys. Rev. Lett. 110, 235902, 2013,
Phys. Rev. Lett. 115, 035901, 2015.

Not yet observed experimentally

Long-ranged non-equilibrium fluctuations:

- generic scale invariance <u>far from a critical point</u>
- confinement: **fluctuation-induced (Casimir) forces**
- **Orders of magnitude larger than critical Casimir**



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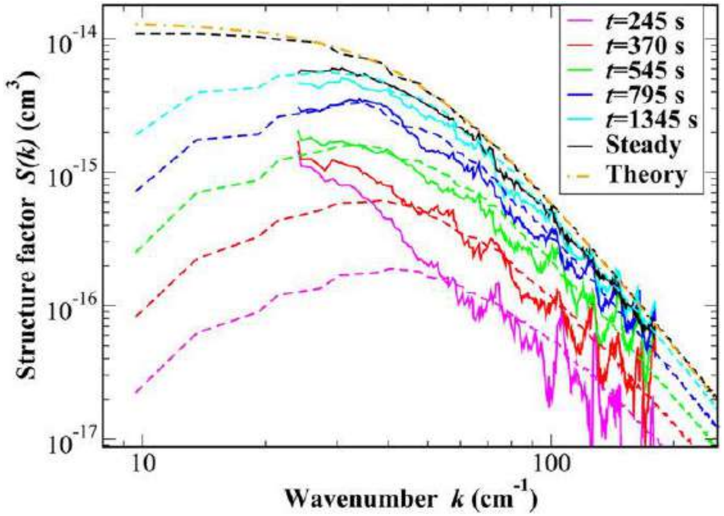
Spinodal-like scaling of NEF during transient diffusion

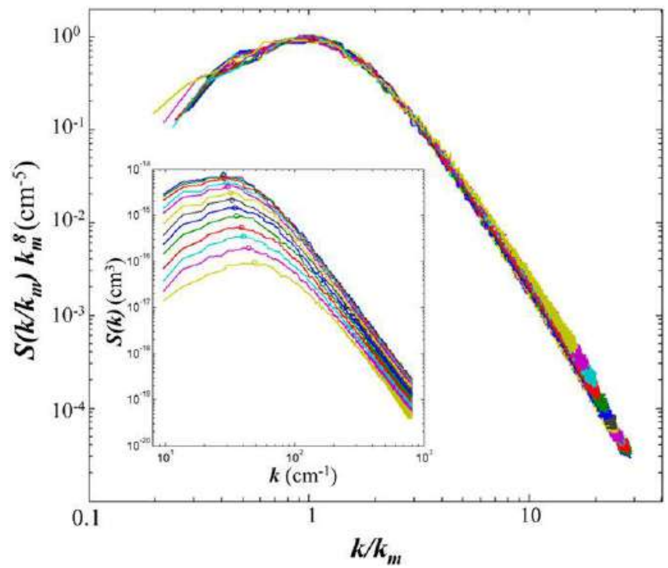


Scientific relevance

- time dependent process
- no available theoretical results
- birth of the fluctuations
- simulations show a spinodal like scaling of NEF

$$S(k/k_m,\ t) = k_m(t)^{-a}F(k/k_m)$$

- Gradflex results are partially compatible with simulations: need for a larger statistical sample



R. Cerbino, Y. Sun, A. Donev & A. Vailati,
Nature Scientific Reports 5, 14486 (2015)

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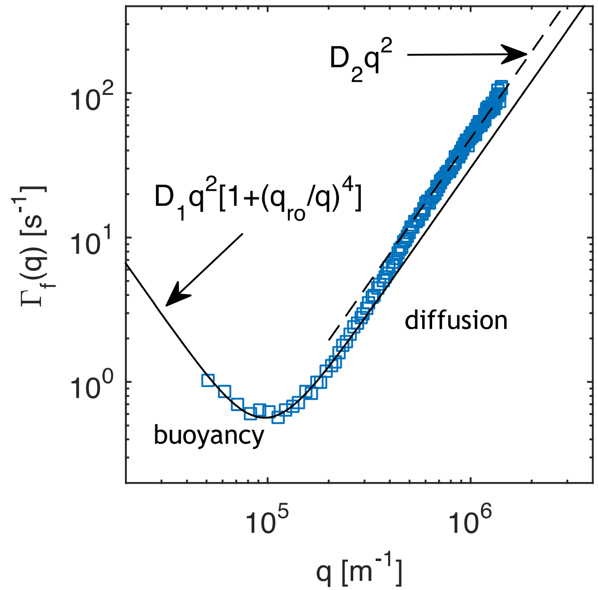
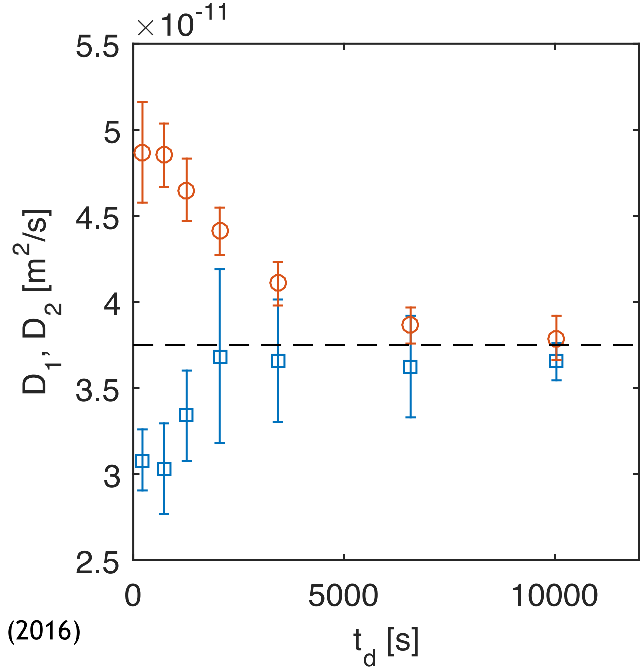
Non-equilibrium fluctuations in dense colloidal suspensions



Scientific relevance

- almost nothing is known about colloids and NEF
- only one theory for dense suspensions (Schmitz, 1994)
- Dense suspensions show a complex and richer dynamics
- non-linear theories needed?
- Ideal sample for probing Casimir forces induced by NEF



F. Giavazzi, G. Savorana, A. Vailati, R. Cerbino, Soft Matter 12, 6588 (2016)

Multiscale Simulations

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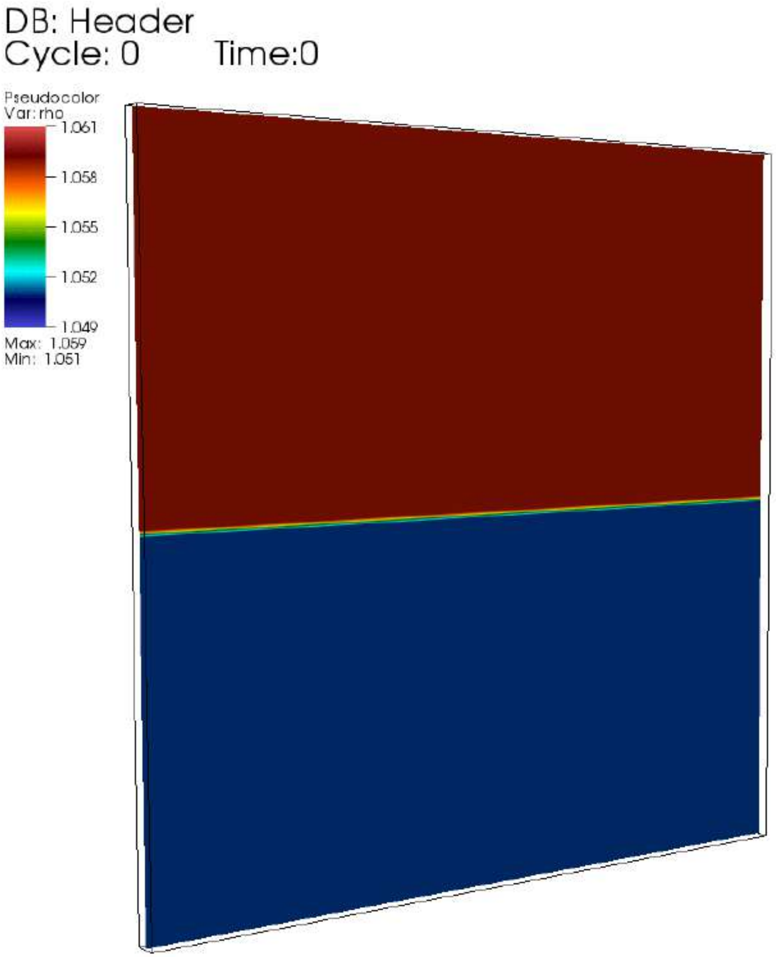

Development of an instability during diffusive mixing in a ternary mixture, triggered entirely by thermal fluctuations
A. Donev et al., Physics of Fluids, 27(3):037103, (2015)

- **Numerical methods to solve the equations of fluctuating hydrodynamics** in multispecies liquid mixtures developed by A. Donev and collaborators
- **simulation of fully time-dependent nonlinear equations**
- CHALLENGE: combine "big simulation" with "big data" generated by experiments under a Monte Carlo sampler

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THE EUROPEAN PHYSICAL JOURNAL E

Regular Article

The NEUF-DIX space project - Non-EquilibriUm Fluctuations during DIffusion in compleX liquids*

Philipp Baaske[1], Henri Bataller[2], Marco Braibanti[3], Marina Carpineti[4], Roberto Cerbino[5], Fabrizio Croccolo[2,6,a], Aleksandar Donev[7], Werner Köhler[8], José M. Ortiz de Zárate[9], and Alberto Vailati[4,b]