Perspectives on Non-Equilibrium Statistical Mechanics:
The 45th Anniversary Symposium of Yamada Science Foundation
Yukawa Institute for Theoretical Physics, Kyoto University, Japan

Memory effects in complex fluids

August 4, 2023

Francisco Vega Reyes

Departamento de Física and ICCAEx, Universidad de Extremadura, Spain

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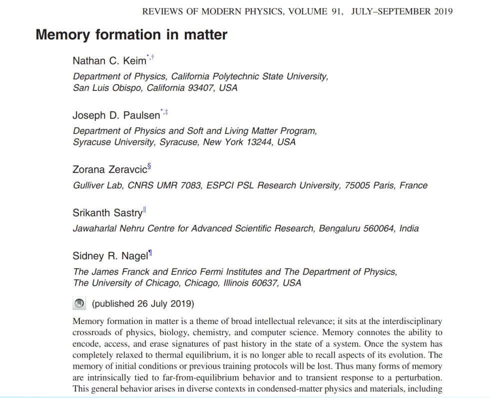
What is a memory effect?: Definition



REVIEWS OF MODERN PHYSICS, VOLUME 91, JULY–SEPTEMBER 2019

Memory formation in matter

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The James Franck and Enrico Fermi Institutes and The Department of Physics,
The University of Chicago, Chicago, Illinois 60637, USA

Memory formation in matter is a theme of broad intellectual relevance; it sits at the interdisciplinary crossroads of physics, biology, chemistry, and computer science. Memory connotes the ability to encode, access, and erase signatures of past history in the state of a system. Once the system has completely relaxed to thermal equilibrium, it is no longer able to recall aspects of its evolution. The memory of initial conditions or previous training protocols will be lost. Thus many forms of memory are intrinsically tied to far-from-equilibrium behavior and to transient response to a perturbation. This general behavior arises in diverse contexts in condensed-matter physics and materials, including

Keim *et al.* *Rev. Mod. Phys.* **91** (20019).

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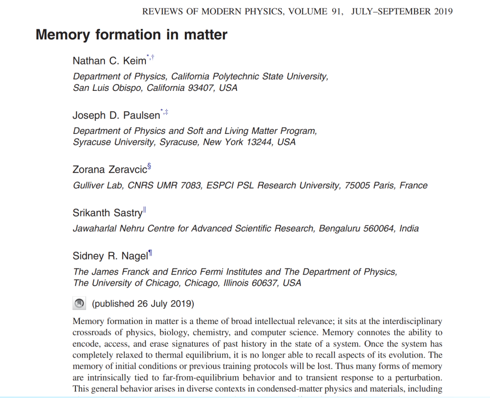

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(published 26 July 2019)

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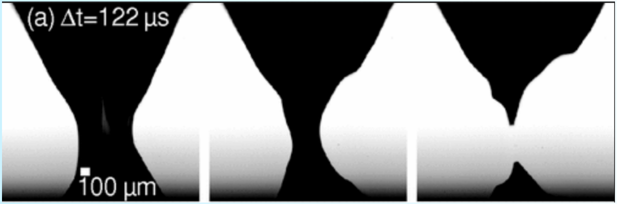
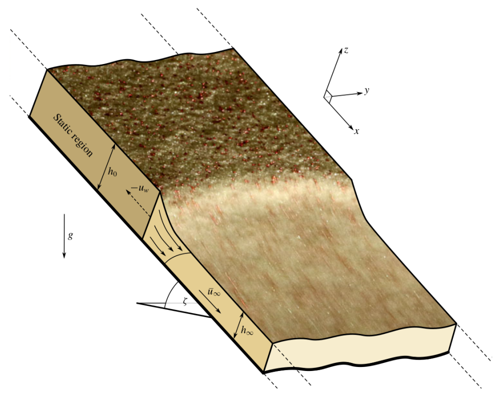
Examples of memory effects: Dripping faucet



In an underwater air bubble dripping from a faucet, the pinching neck of air is very sensitive to the geometry of the initial conditions. This occurs only if the dripping process is fast enough.

Keim *et al. Phys. Rev. Lett.* **97** 144503 (2006).

Examples of memory effects: Hysteresis in granular avalanches



Under certain circumstances, upwards avalanches occur in an inclined plane.

Russell, Johnson, Edwards & Viroulet *J. Fluid Mech.* **819** (2019).

Examples of memory effects: Shape memory

This wire is made of a material (nickel titanium) that under cooling of the initial cubic phase (paperclip-shaped), undergoes a transition to a martensite phase which can be deformed with no modification of microscopic bond topology. Thus, the material gets back to paperclip-shaped if the cubic phase is recuperated after heating back. This has applicattions such a as aerospace industry
Boeing Frontiers Online. `www.boeing.com`

Examples of memory effects: Shape memory in red blood cells

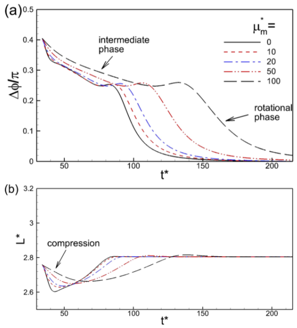
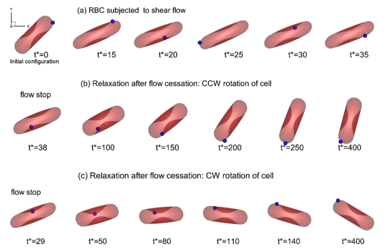


FIG. 15. The effect of membrane viscosity on the RBC shape memory response: (a) Return of a marker point to its original location is shown using the phase angle $\Delta\phi$ for different values of μ_m^*. (b) End-to-end cell length. Only the recovery part is shown. BCSF cells, $\Delta\phi_S/\pi \approx 0.4$, $\varepsilon = 0.3$, $\lambda = 0.1$.

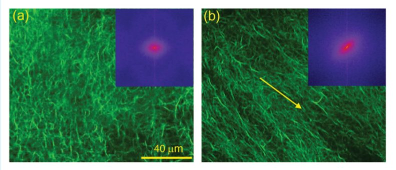


Red blood cells are biconcave-discoid-shaped at rest.

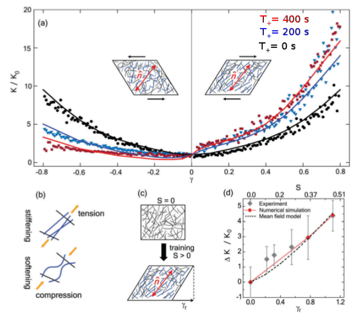
Red blood cells return to their biconcave discoid resting shape as the external forces are withdrawn.

Cordasco & Bagchi *Phys. Fluids* **29** 041901 (2017).

Examples of memory effects: Mechanical memory in cells' actin networks

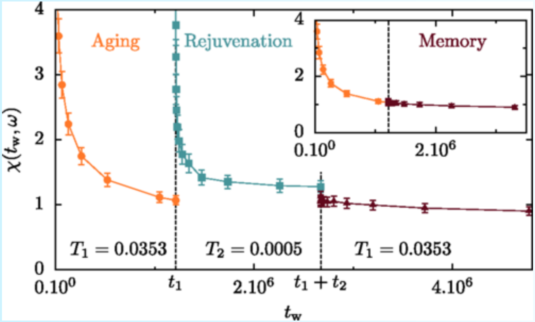


Confocal image of actin sample;
Majumdar, Foucard, Levine, Gardel, *Soft Matter* **14** 2052 (2018).



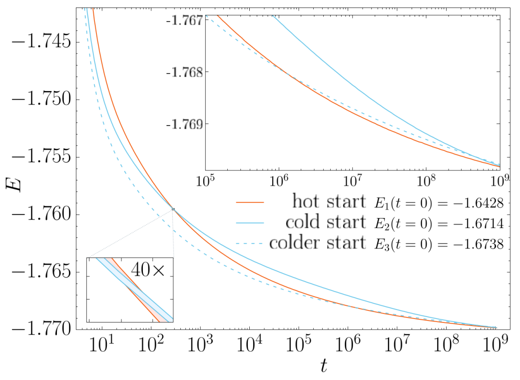
History dependent shear response in actin networks. T_+: training time; γ: shear strain; K/K_0: stiffening ($K \equiv \frac{\mathrm{d}\sigma}{\mathrm{d}\gamma}$);
Majumdar, Foucard, Levine, Gardel, *Soft Matter* **14** 2052 (2018).

Examples of memory effects: Memory in glasses



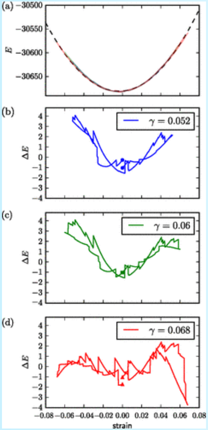
Aging, rejuvenation and perfect memory in glasses.
Scalliet & Berthier, *Phys. Rev. Lett.* **122** 255502 (2019).

Examples of memory effects: Memory in spin glasses



Hot spin glass that cools faster.
Baity-Jesi, Lasanta et al., *Proc. Nat. Acad. Sci* **116** 15350 (2019).

Examples of memory effects: Shear memory in amorphous solids

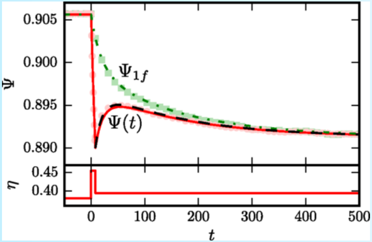


The three lines initially follow the same path (for positive strains) and separate as the respective amplitudes are reached.
Fiocco, Foffi & Sastry, *Phys Rev. Lett* **112** 025702 (2014).

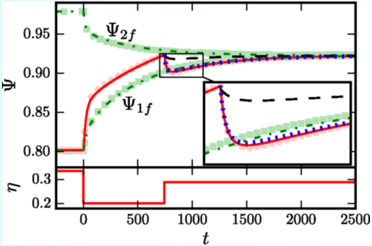
In a series of simulations, a binary mixture of spheres interacting through a Lennard-Jones pontential undergoes cyclic shear training at strain $\pm\gamma$.

The system response is different depending of the strain amplitude.

Examples of memory effects: Thermal memory in active particles



Here, the system is kindled.

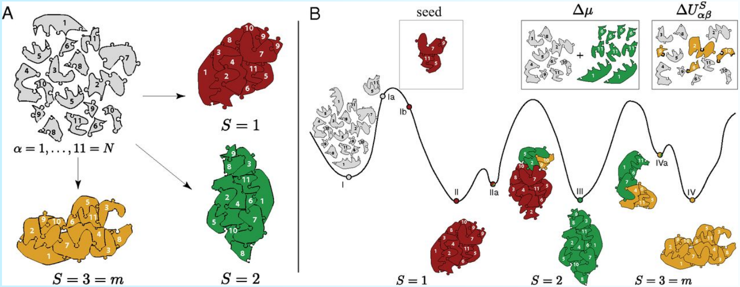


Here, the system is quenched.

The polar order parameter experiences a hump if subject to a thermal protocol (here, η is the thermal noise intensity).

Kürsten, Sushkov & Ihle, *Phys. Rev. Lett.* **119** 188001 (2017).

Examples of memory effects: Associative memory



Certain square building blocks bonds are stronger, which results in a set of more permanent structures, or *associative memories.*

Murugan, Zeravcic, Brenner & Leibler, *Proc. Natl. Acad. Sci* **112** 54 (2015).

Examples of memory effects: Water has thermal memory

. . . The fact that the water has previously been warmed contributes to its freezing quickly: for so it cools sooner. Hence many people, when they want to cool hot water quickly, begin by putting it in the sun. So the inhabitants of Pontus when they encamp on the ice to fish (they cut a hole in the ice and then fish) pour warm water round their reeds that it may freeze the quicker. . .

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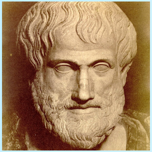

Aristotle, in *Meteorologica*, part 12, Book I, (350 BC)

Examples of memory effects: Water has thermal memory!

Making Snow with Boiling Water

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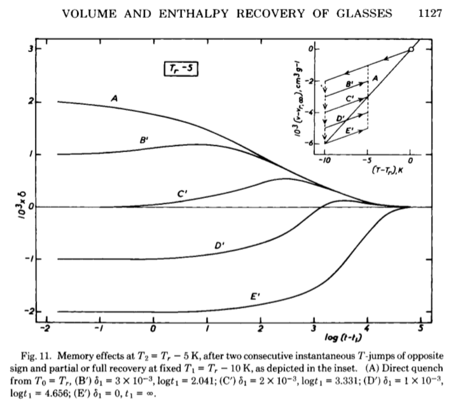
Making Snow with Boiling Water

The Mpemba effect can be interpreted as initial state fast deletion, due to a very different environment.

Examples of memory effects: Volume memory in polymers



VOLUME AND ENTHALPY RECOVERY OF GLASSES 1127

Fig. 11. Memory effects at $T_2 = T_r - 5$ K, after two consecutive instantaneous T-jumps of opposite sign and partial or full recovery at fixed $T_1 = T_r - 10$ K, as depicted in the inset. (A) Direct quench from $T_0 = T_r$, (B′) $\delta_1 = 3 \times 10^{-3}$, $\log t_1 = 2.041$; (C′) $\delta_1 = 2 \times 10^{-3}$, $\log t_1 = 3.331$; (D′) $\delta_1 = 1 \times 10^{-3}$, $\log t_1 = 4.656$; (E′) $\delta_1 = 0$, $t_1 = \infty$.

The Kovacs memory effect: a piece of polymer undergoes a volume jump as a consequence of a temperature protocol, before relaxation to the equilibrium volume.
Kovacs *et al.*, *J. Polym. Sci. Pt. B-Polym. Phys.* 17 1907 (1979).

The granular gas in nature

- A granular gas is a system composed of many macroscopic particles that collide with each other, loosing a fraction of kinetic energy upon collision.

The granular gas in nature

- The characteristic particle collision time is finite.

The granular gas in nature

- Granular gases may be considered as *thermalized* granular materials.

The granular gas in nature

- Granular materials are ubiquitous in nature. In particular, granular fluids are common in low gravity environments.

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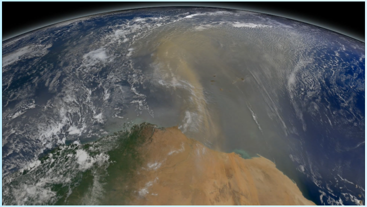
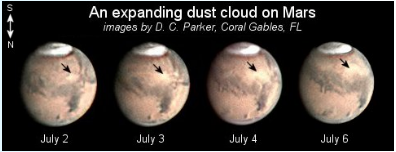


Results derived from NASA's CALIPSO satellite
H. Yu et al., *J. Geophys. Lett.* **42** 1984 (2015).



Mars dust cloud.
Source: NASA.

Memory effects in the granular gas

Kovacs effect

(1) *Simple Kovacs effect:* A single temperature rebound (hump) is observed during relaxation towards the stationary value.

Memory effects in the granular gas

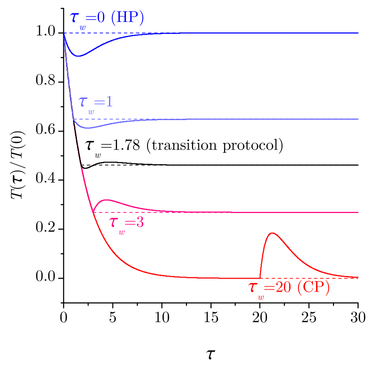
Kovacs effect

(1) *Simple Kovacs effect:* A single temperature rebound (hump) is observed during relaxation towards the stationary value.

(2) *Complex Kovacs effect:* Multiple temperature rebound (in the form of damped oscillations) are observed during thermal relaxation.

Kovacs effect

The granular fluid is subject to a thermal impulse and left to cooling a time τ_w before fixing the heat source back again.



Lasanta, VR, Prados & Santos, *New J. Phys.* **21** 033042 (2019).

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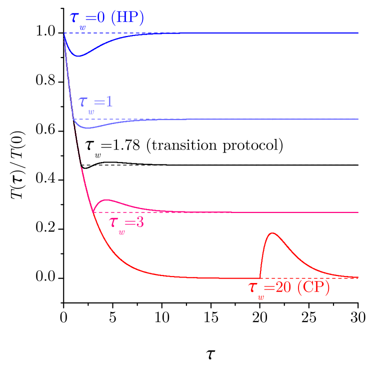

- The granular gas can react, as molecular gases, by *remembering its initial temperature* (hump up).

Lasanta, VR, Prados & Santos, *New J. Phys.* **21** 033042 (2019).

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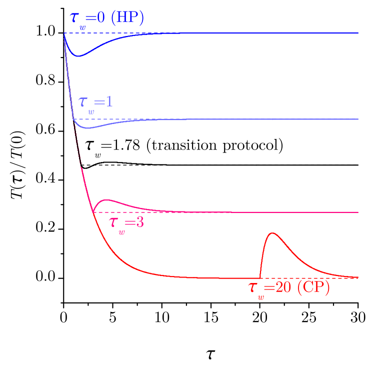


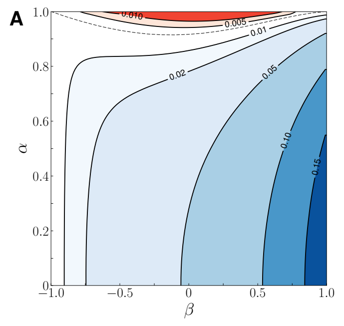
- The granular gas can react, as molecular gases, by *remembering its initial temperature* (hump up).
- BUT, it can also react *remembering by its inherent cooling rate* (hump down).

Lasanta, VR, Prados & Santos, *New J. Phys.* **21** 033042 (2019).

Kovacs effect

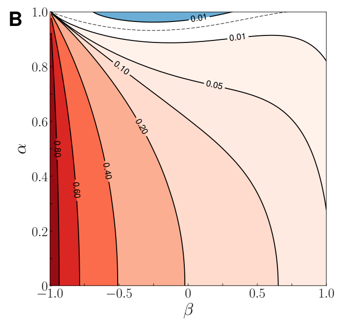
Temperature fixed

right after heat impulse



Temperature fixed

long after heat impulse



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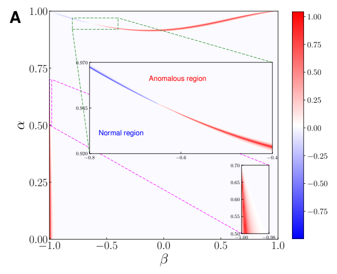

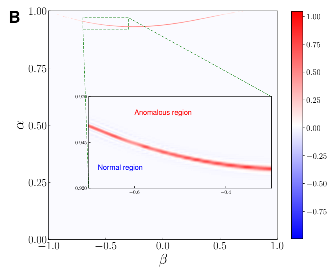

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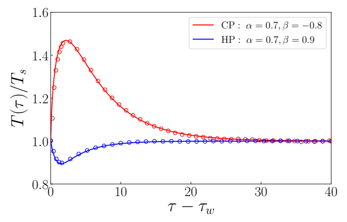
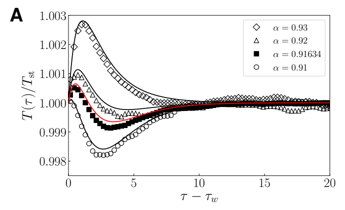
Kovacs effect

Giant temperature humps



Multiple temperature humps



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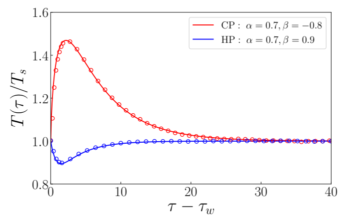

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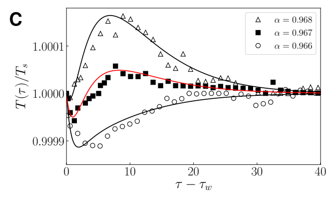

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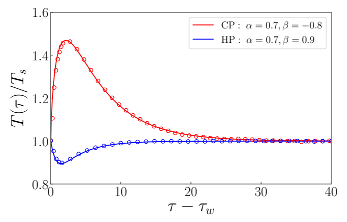

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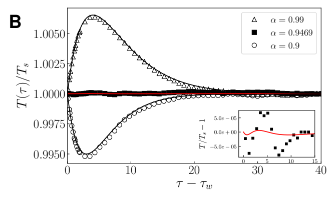

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Memory effects in the granular gas

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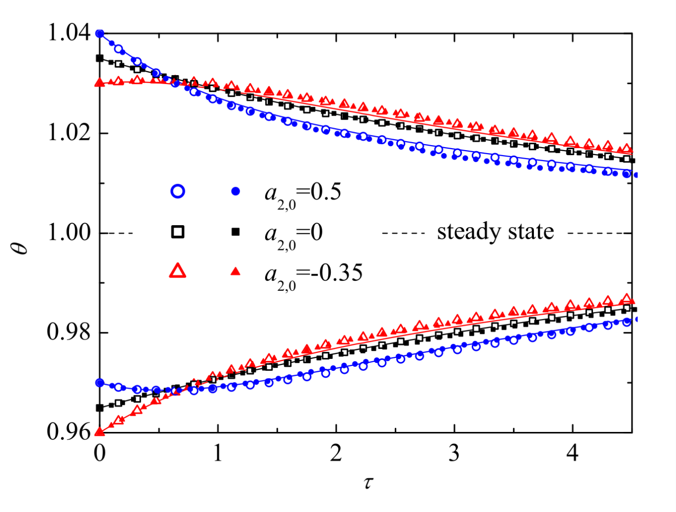
Mpemba effect

(1) *Direct Mpemba effect*: Among two systems at different initial temperatures, it is the hottest one the first to achieve the (shared) stationary value.

(2) *Reverse Mpemba effect*: Conversely, an initially colder system may heat up faster than a hotter one to their shared stationary temperature.

Memory effects in the granular gas



Phys. Rev. Lett. **119** 148001 (2017)

Theoretical basis: The $T - a_2$ coupling: Mpemba effect

$$\frac{dT}{dt} = -\frac{2K}{3}\left(\mu_2 T^{3/2} - \chi\right), \tag{1}$$

$$\frac{d\ln(1+a_2)}{dt} = \frac{4\kappa}{3T}\left(\mu_2 T^{3/2} - \chi - \frac{\frac{1}{5}\mu_4 T^{3/2} - \chi}{1+a_2}\right), \tag{2}$$

where $\kappa \equiv 2ng(n)\sigma^2\sqrt{\pi/m}$ and $\chi \equiv (3m/2\kappa)\xi^2$.

Theoretical basis: The effects of additional cumulants: complex Kovacs effect

$$\partial_\tau \ln\left(1+a_{20}^{(0)}\right)=\frac{4}{15}\left[5\left(\mu_{20}^{(0)}-\gamma\right)-\frac{\mu_{40}^{(0)}-5\gamma}{1+a_{20}^{(0)}}\right], \tag{3}$$

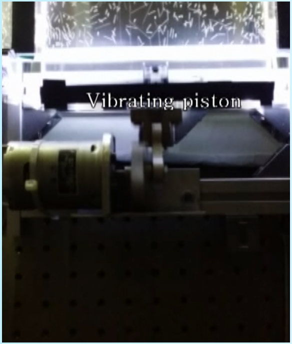
$$\partial_\tau \ln\left(1+a_{02}^{(0)}\right)=\frac{4}{15}\left(5\mu_{02}^{(0)}-\frac{\mu_{04}^{(0)}}{1+a_{02}^{(0)}}\right), \tag{4}$$

$$\partial_\tau \ln\left(1+a_{11}^{(0)}\right)=\frac{4}{9}\left[\frac{3}{2}\left(\mu_{20}^{(0)}+\mu_{02}^{(0)}-\gamma\right)-\frac{\mu_{22}^{(0)}-\frac{3}{2}\gamma}{1+a_{11}^{(0)}}\right], \tag{5}$$

$$\partial_\tau \ln\left(1+a_{11}^{(0)}+\frac{5}{2}a_{00}^{(1)}\right)=\frac{4}{3}\left[\frac{\mu_{20}^{(0)}+\mu_{02}^{(0)}-\gamma}{2}-\frac{\mu_{00}^{(2)}-\frac{1}{2}\gamma}{1+a_{11}^{(0)}+\frac{5}{2}a_{00}^{(1)}}\right]. \tag{6}$$

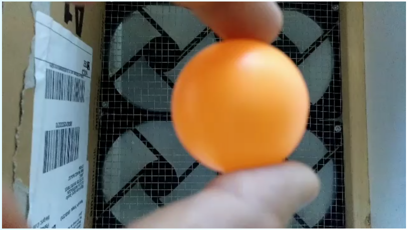
Experimental realizations in the lab of a granular gas

Vibration



Experiment performed at Sapienza, Roma
Pontuale, Gnoli, VR & Puglisi, *Phys. Rev. Lett.* **117** 098006 (2016).

Air Fluidization:
Spheres on a plane



Experiments performed at *Granular Dynamics Imaging Lab*, ICCAEx, Universidad de Extremadura, Badajoz, Spain
VR, López-Castaño, Saavedra, Rodríguez-Rivas, Yuste & Abad, *work in progress*, 2019.

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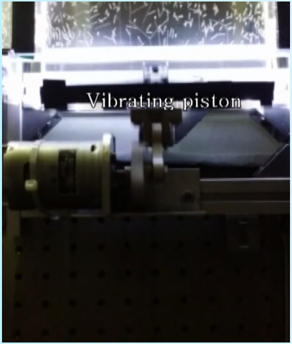

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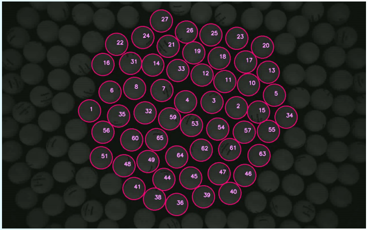

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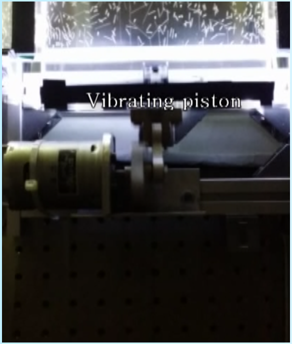
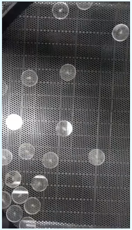


Experiment performed at Sapienza, Roma
Pontuale, Gnoli, VR & Puglisi, *Phys. Rev. Lett.* **117** 098006 (2016).

Air Fluidization:
Disks on a plane



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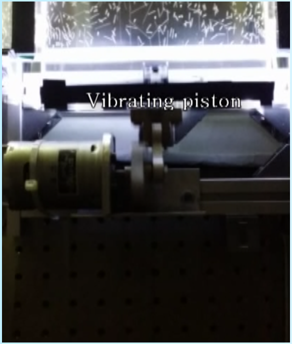

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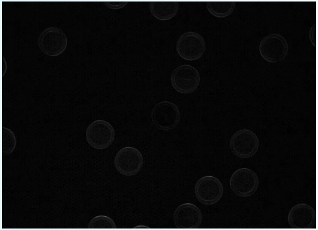

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Experimental evidence of memory effect in the granular gas: Kovacs Effect

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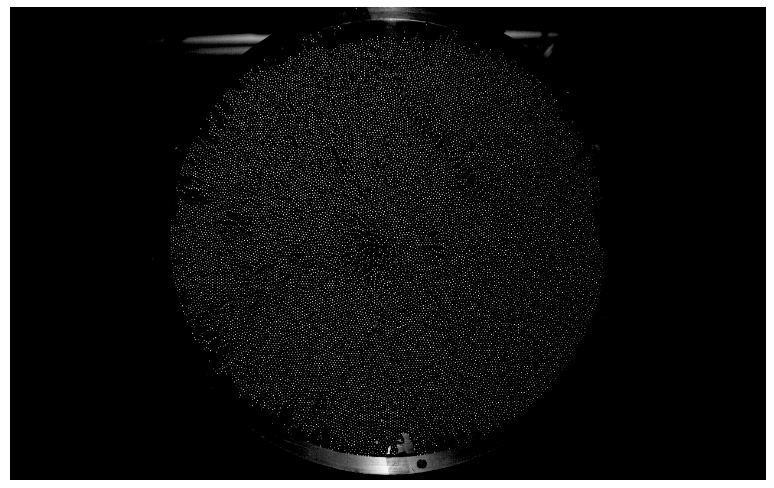

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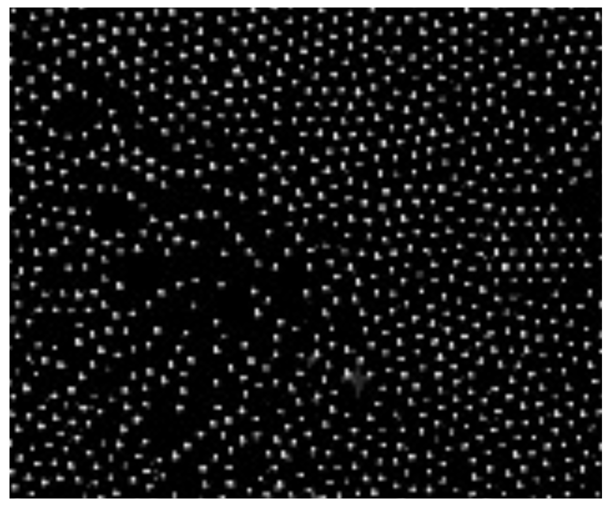

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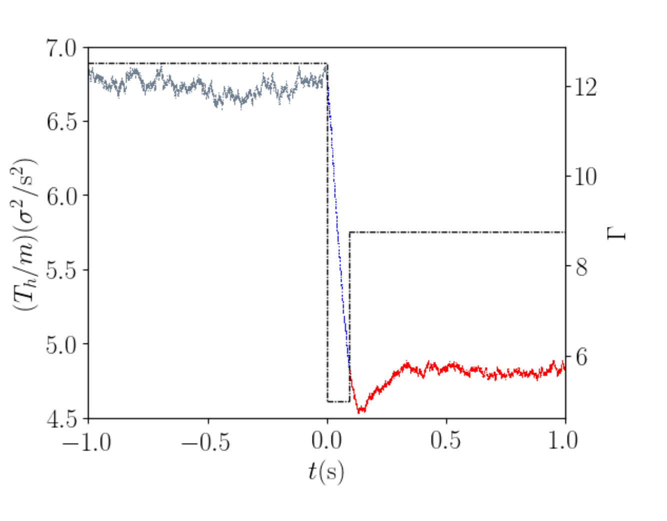


Then, why memory effects?

Memory effects should appear always during transients for which the particle distribution has not undergone a contraction to average fields; i.e., the system is not yet under a *normal state*

$$\{a_i, n, T, \mathbf{u}\} \rightarrow \{n, T, \mathbf{u}\} \tag{7}$$

Then, why memory effects?

According to D. Hilbert's theory, normal states are always attainable if the system has the ability to evolve towards an equilibrium state.

Existence of normal states in intrinsically non-equilibrium states is however, not guaranteed.

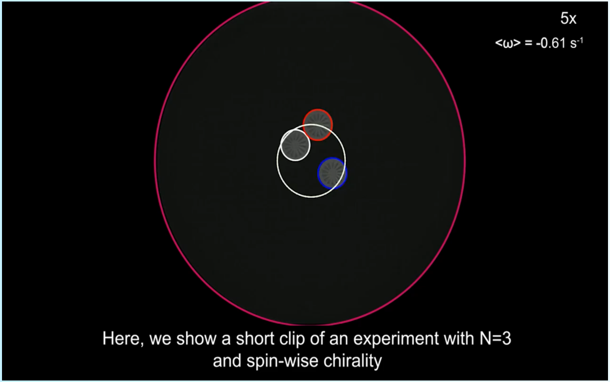
Therefore, non-equilibrum systems should be more prone to display persistentmemory effects.

In particular, active matter is a good candidate for lack of scale separation and thus, for long transients where memory effects can occur.

Normal (hydrodynamic) states vs. kinetic states

Begründung der kinetischen Gastheorie, David Hilbert, *Mathematische Annalen* **72**, 562577 (1912).

Long memory in a quiral active particle: Self-restrained diffusion



Here, we show a short clip of an experiment with N=3
and spin-wise chirality

Flow chirality inversion

Chirality transitions in a system of active flat spinners
M. A. López-Castaño, A. Márquez Seco, A. Márquez Seco, A. Rodríguez-Rivas, and F. Vega Reyes
Phys. Rev. Research **4**, 033230 (2022).

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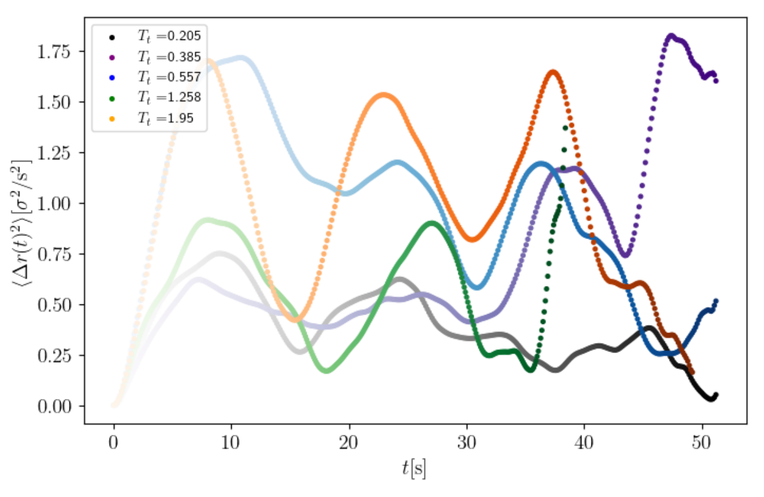

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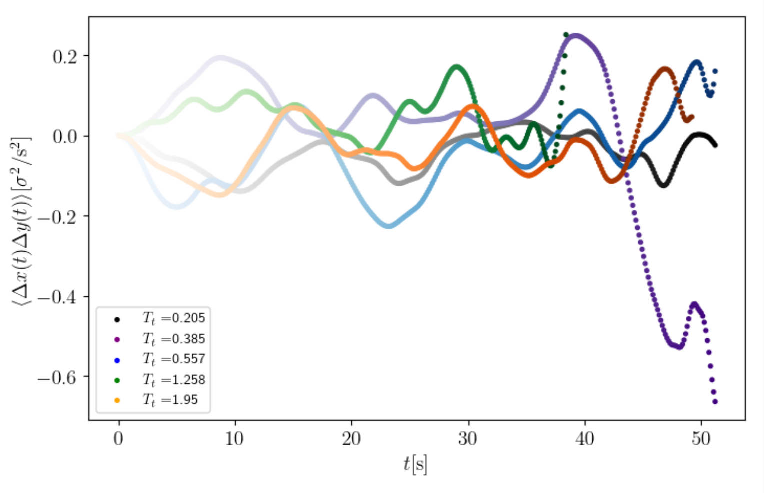
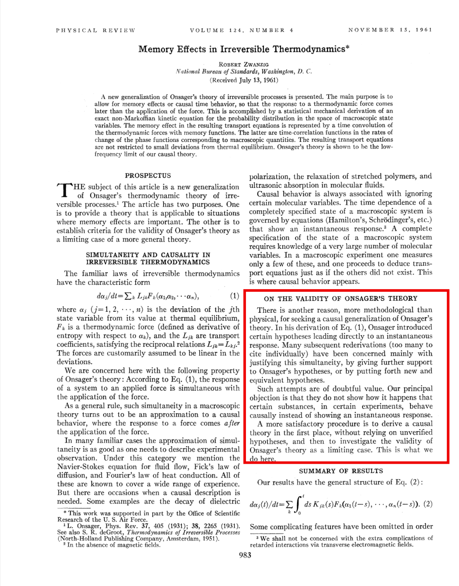


Mori-Zwanzig formalism



PHYSICAL REVIEW VOLUME 124, NUMBER 4 NOVEMBER 15, 1961

Memory Effects in Irreversible Thermodynamics*

ROBERT ZWANZIG
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(Received July 13, 1961)

A new generalization of Onsager's theory of irreversible processes is presented. The main purpose is to allow for memory effects or causal time behavior, so that the response to a thermodynamic force comes later than the application of the force. This is accomplished by a statistical mechanical derivation of an exact non-Markoffian kinetic equation for the probability distribution in the space of macroscopic state variables. The memory effect in the resulting transport equations is represented by a time convolution of the thermodynamic forces with memory functions. The latter are time-correlation functions in the rates of change of the phase functions corresponding to macroscopic quantities. The resulting transport equations are not restricted to small deviations from thermal equilibrium. Onsager's theory is shown to be the low-frequency limit of our causal theory.

PROSPECTUS

THE subject of this article is a new generalization of Onsager's thermodynamic theory of irreversible processes.[1] The article has two purposes. One is to provide a theory that is applicable to situations where memory effects are important. The other is to establish criteria for the validity of Onsager's theory as a limiting case of a more general theory.

SIMULTANEITY AND CAUSALITY IN IRREVERSIBLE THERMODYNAMICS

The familiar laws of irreversible thermodynamics have the characteristic form

$$d\alpha_j/dt=\textstyle\sum_k L_{jk}F_k(\alpha_1,\alpha_2,\cdots\alpha_n), \qquad (1)$$

where α_j $(j=1, 2, \cdots, n)$ is the deviation of the jth state variable from its value at thermal equilibrium, F_k is a thermodynamic force (defined as derivative of entropy with respect to α_k), and the L_{jk} are transport coefficients, satisfying the reciprocal relations $L_{jk}=L_{kj}$.[2] The forces are customarily assumed to be linear in the deviations.

We are concerned here with the following property of Onsager's theory: According to Eq. (1), the response of a system to an applied force is simultaneous with the application of the force.

As a general rule, such simultaneity in a macroscopic theory turns out to be an approximation to a causal behavior, where the response to a force comes *after* the application of the force.

In many familiar cases the approximation of simultaneity is as good as one needs to describe experimental observation. Under this category we mention the Navier-Stokes equation for fluid flow, Fick's law of diffusion, and Fourier's law of heat conduction. All of these are known to cover a wide range of experience. But there are occasions when a causal description is needed. Some examples are the decay of dielectric polarization, the relaxation of stretched polymers, and ultrasonic absorption in molecular fluids.

Causal behavior is always associated with ignoring certain molecular variables. The time dependence of a completely specified state of a macroscopic system is governed by equations (Hamilton's, Schrödinger's, etc.) that show an instantaneous response.[3] A complete specification of the state of a macroscopic system requires knowledge of a very large number of molecular variables. In a macroscopic experiment one measures only a few of these, and one proceeds to deduce transport equations just as if the others did not exist. This is where causal behavior appears.

ON THE VALIDITY OF ONSAGER'S THEORY

There is another reason, more methodological than physical, for seeking a causal generalization of Onsager's theory. In his derivation of Eq. (1), Onsager introduced certain hypotheses leading directly to an instantaneous response. Many subsequent rederivations (too many to cite individually) have been concerned mainly with justifying this simultaneity, by giving further support to Onsager's hypotheses, or by putting forth new and equivalent hypotheses.

Such attempts are of doubtful value. Our principal objection is that they do not show how it happens that certain substances, in certain experiments, behave causally instead of showing an instantaneous response.

A more satisfactory procedure is to derive a causal theory in the first place, without relying on unverified hypotheses, and then to investigate the validity of Onsager's theory as a limiting case. This is what we do here.

SUMMARY OF RESULTS

Our results have the general structure of Eq. (2):

$$d\alpha_j(t)/dt=\sum_k\int_0^t ds\, K_{jk}(s)F_k(\alpha_1(t-s),\cdots,\alpha_n(t-s)). \qquad (2)$$

Some complicating features have been omitted in order

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* This work was supported in part by the Office of Scientific Research of the U. S. Air Force.

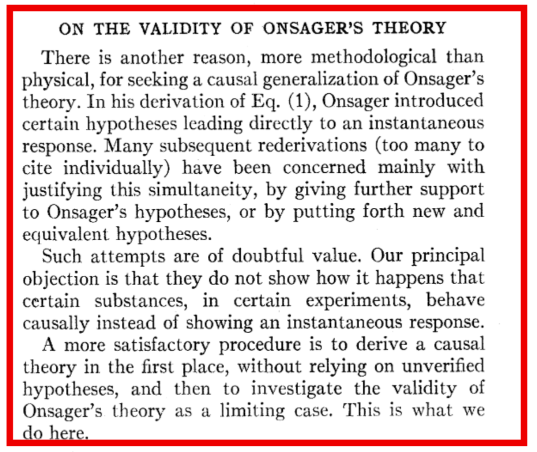
[1] L. Onsager, Phys. Rev. **37**, 405 (1931); **38**, 2265 (1931). See also S. R. deGroot, *Thermodynamics of Irreversible Processes* (North-Holland Publishing Company, Amsterdam, 1951).

[2] In the absence of magnetic fields.

[3] We shall not be concerned with the extra complications of retarded interactions via transverse electromagnetic fields.

"Memory Effects in Irreversible Thermodynamics", Robert Zwanzig, *Phys. Rev.* **124** 983 (1965).

Mori-Zwanzig formalism



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Conclusions

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2. *On the emergence of large and complex memory effects in nonequilibrium fluids*
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3. *Diffusive regimes in a two-dimensional chiral fluid*
   F. Vega Reyes, M. A. López-Castaño, A. Rodríguez-Rivas, *Commun. Phys* **5** 256 (2022).

GRACIAS!! :)

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