Molecular Dynamics simulations to study dissipation in amorphous materials: starting with Ta_2O_5

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INFN-Pisa

10th Einstein Telescope Symposium
Orosei, Italy
12th April 2019

Overview

In current interferometric gravitational wave detectors, **thermal noise** from the mirrors limits the detector sensitivity in most of the detection band.

Current mirror coating are amorphous (= non-crystalline).

Intensive experimental activity led to significant improvements

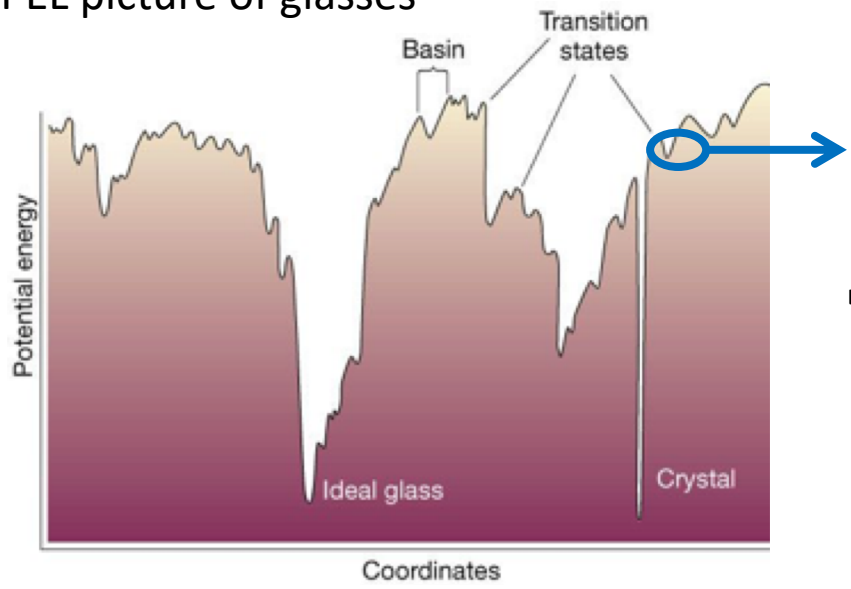
but a **fundamental understanding** of dissipation mechanisms is still missing.

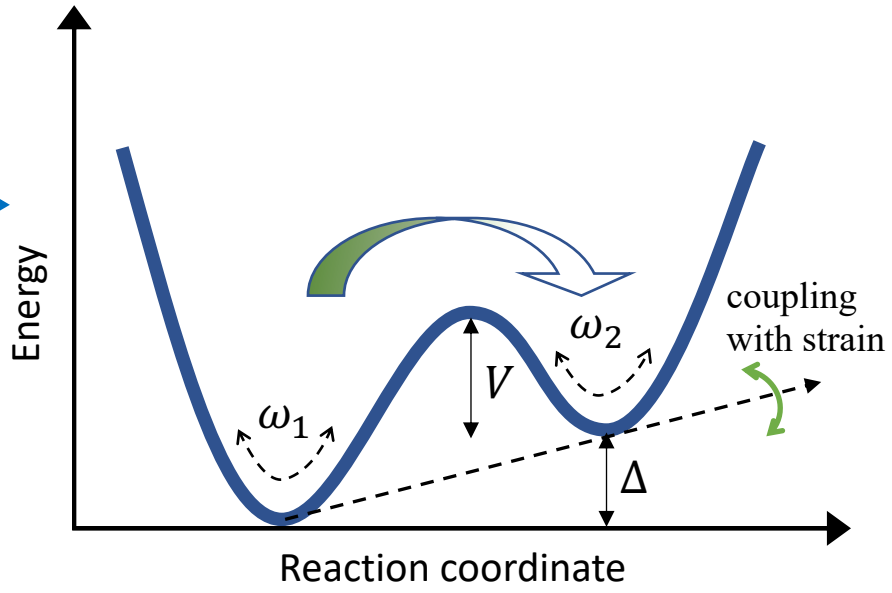
Modeling and simulations could provide insights into the microscopic processes controlling dissipation.

The common approach to this problem builds on the framework of the so-called two-level systems (TLS) model.

TLS model and internal friction

PEL picture of glasses





Potential Energy Landscape (PEL) of a glass = collections of two-level systems.

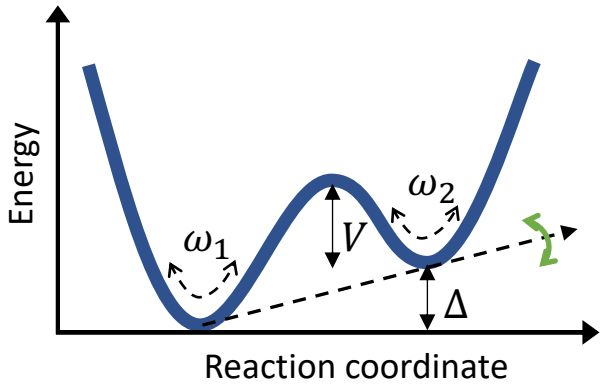
Transitions from one well to another are possible thanks to **coupling between external strain and thermal motion**.

Dissipation dues to atomic motion during transitions from one well to another.

TLS model and internal friction

"Simplified" expressions for the **quality factor**

(assumptions: independent TLS, harmonic transition state theory, factorizable distributions…)



strain coupling constants

$$Q_{l/t}^{-1}(\omega,T) = \frac{\gamma_{l/t}^2}{E_{l/t}k_BT}\int_0^\infty\int_0^\infty \frac{\omega\tau}{1+\omega^2\tau^2}\operatorname{sech}^2\left(\frac{\Delta}{2k_BT}\right) \times f(\Delta)g(V)d\Delta\,dV,$$

elastic modulus

asymmetry distribution

barrier distribution

$$\tau^{-1} = \tau_0^{-1}\cosh\left[\frac{\Delta}{2k_BT}\right]\exp^{-V/k_BT},$$

attempt frequency

In the last decades, the TLS model has been used to predict dissipation via atomic scale simulations with rather satisfying results.

Our big question

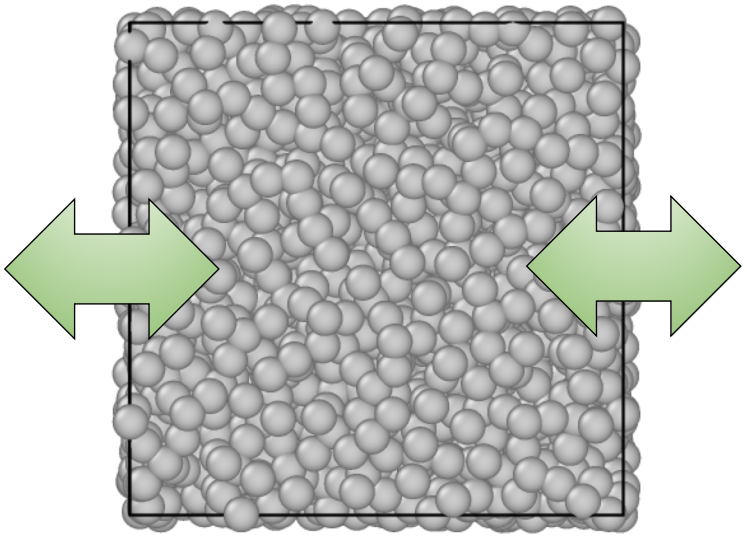
Can we develop an alternative and **theory-independent** approach to provide reliable prediction of the behavior of losses in coating materials?

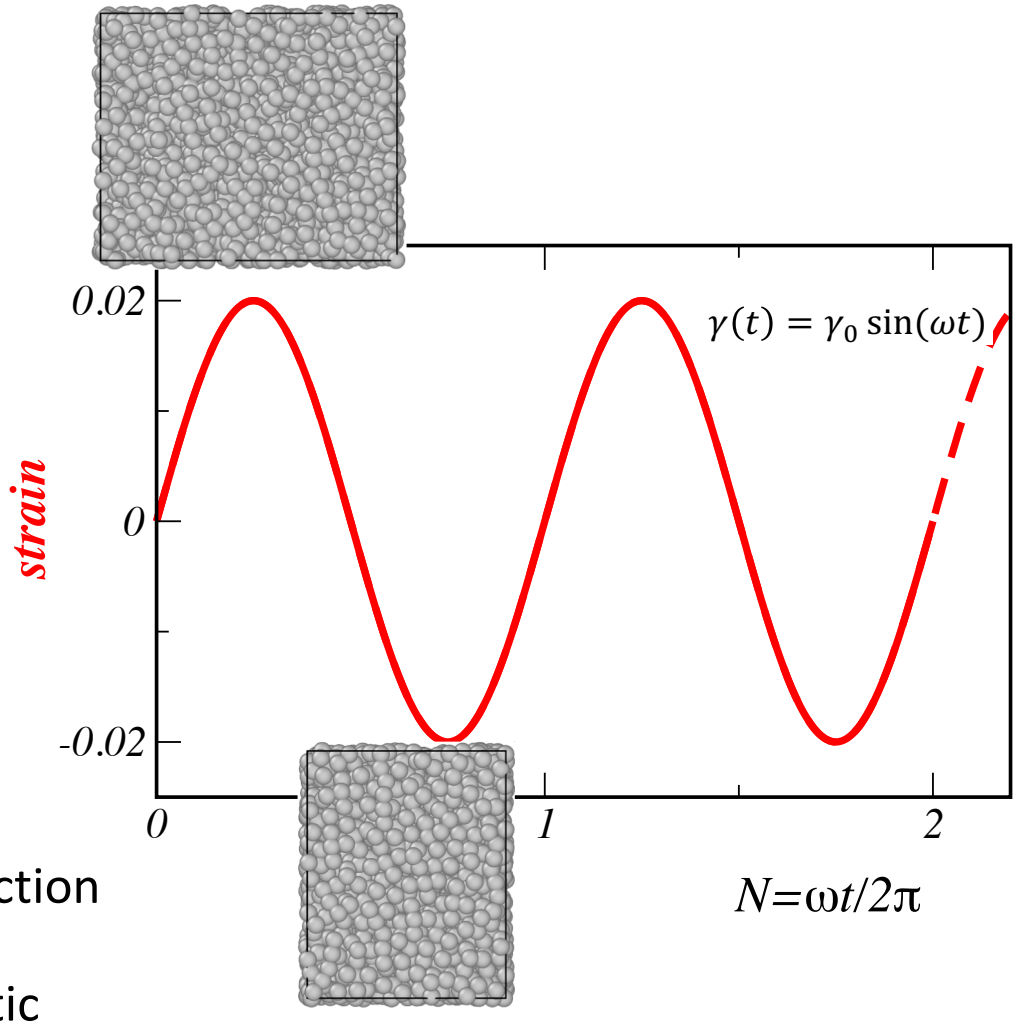
Plan of the talk

- Molecular Dynamics simulations of Dynamical Mechanical Spectroscopy (MD-DMS).
- Ta_2O_5 model and details of the simulations.
- Glassy samples from melt quenching.
- Simulation results and comparison with experimental data.

Dynamical Mechanical Spectroscopy

Unilateral tensile oscillatory deformation



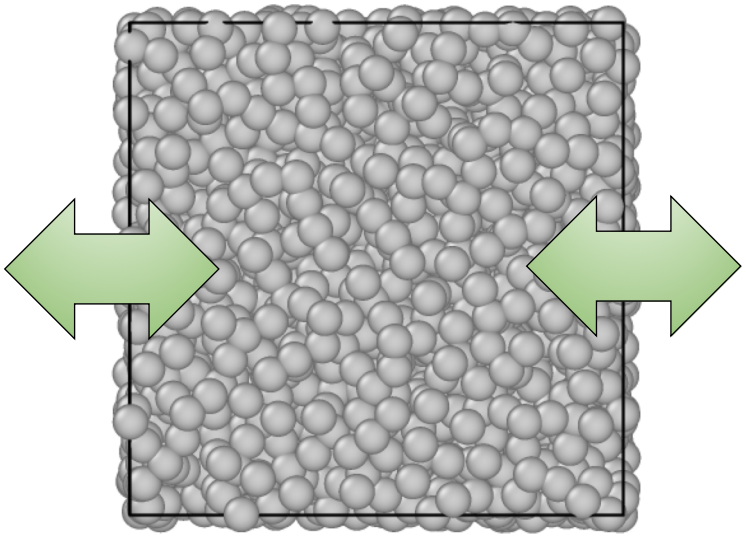


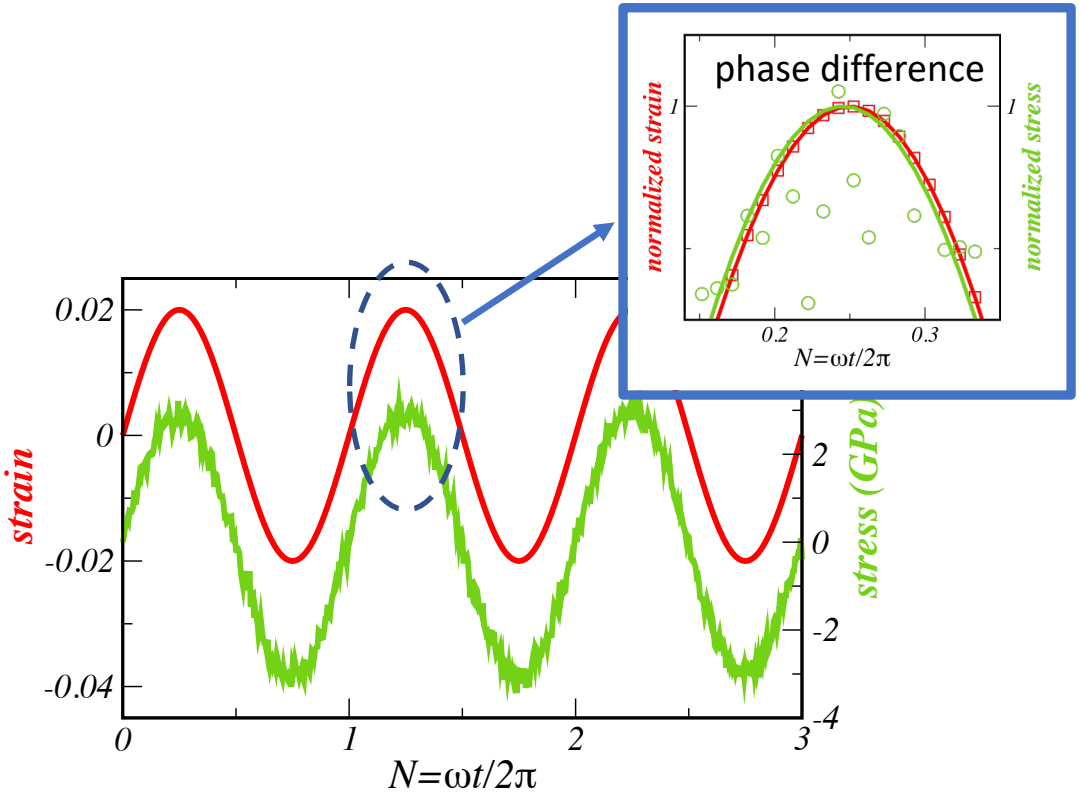
Apply a sinusoidal strain $\gamma(t)$ with selected frequency along one direction

Strain amplitude in the linear elastic regime

Dynamical Mechanical Spectroscopy

Unilateral tensile oscillatory deformation





Storage and loss moduli

$$\mathrm{E}' = \frac{\omega}{N\pi}\int_0^{N2\pi/\omega} \sin(\omega t)\,\frac{\Sigma(t)}{\gamma_0}\,dt \qquad \mathrm{E}'' = \frac{\omega}{N\pi}\int_0^{N2\pi/\omega} \cos(\omega t)\,\frac{\Sigma(t)}{\gamma_0}\,dt$$

Dissipation $$Q^{-1} = \tan\delta = \frac{E''}{E'}$$

Ta_2O_5 model and simulation details

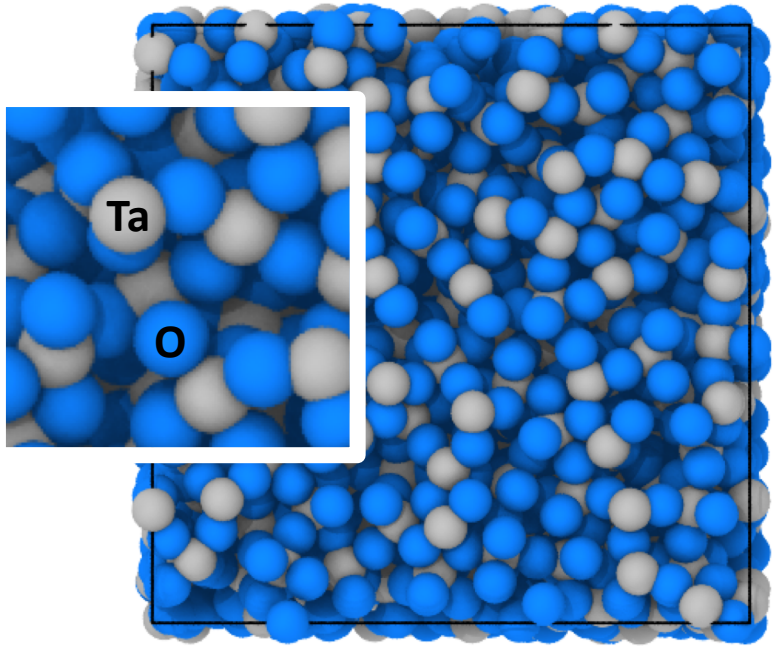
Benchmark system: **Ta_2O_5** for which we have:

- reliable interatomic potential
- experimental data to compare with

Classical MD simulations

Interatomic potential: BKS potential (Coulomb + exponential repulsion + VdW attraction) + covalent bonding (Morse potential)
[Trinastic et al., JCP 2013]

Optimization: Wolf truncation with cut-off
[Damart et al. JAP 2016]



Interatomic potential is the <u>only</u> ingredient we put by hand.

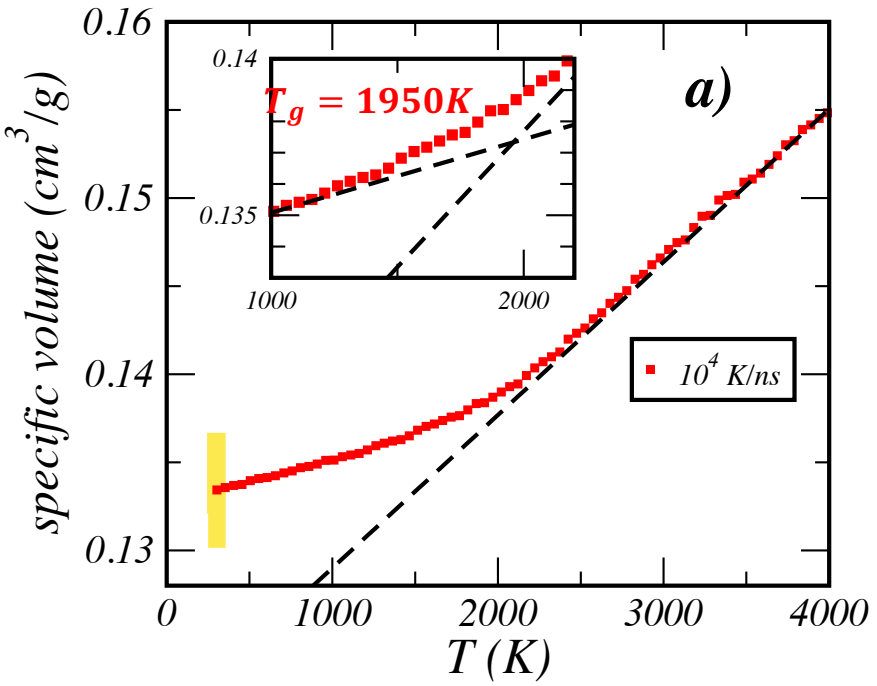
Glasses via melt cooling

Liquid samples: start from crystal at 300 K, fast heating to 5000 K, equilibrate at 5000 K for 50 ns.

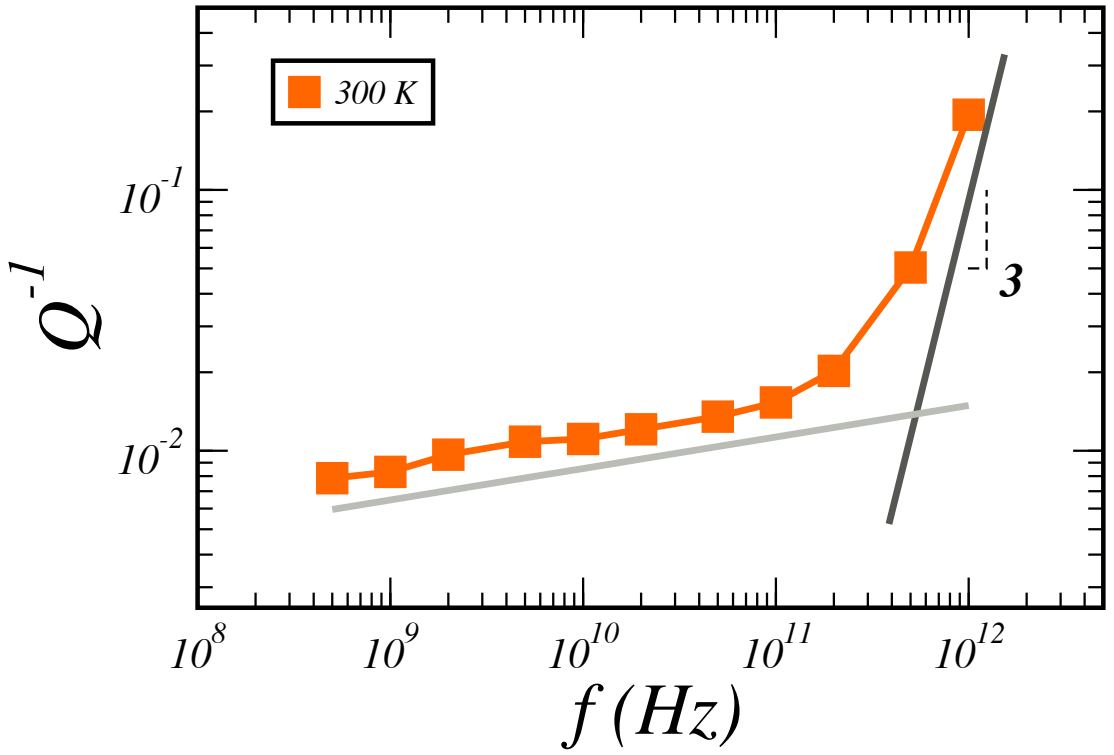
Glasses by cooling the liquid at constant rate.

Density of glassy samples matches the experimental value range of **thin films** [Alderman et al. PRMaterials 2018]

Young's modulus is $148 \pm 4\ GPa$ (from quasistatic simulations), in agreement with the experimental value $140 \pm 15\ GPa$ by nanoindentation on thin films [Alaca et al. Nanotechnology 2002]



MD-DMS in Ta_2O_5: first results

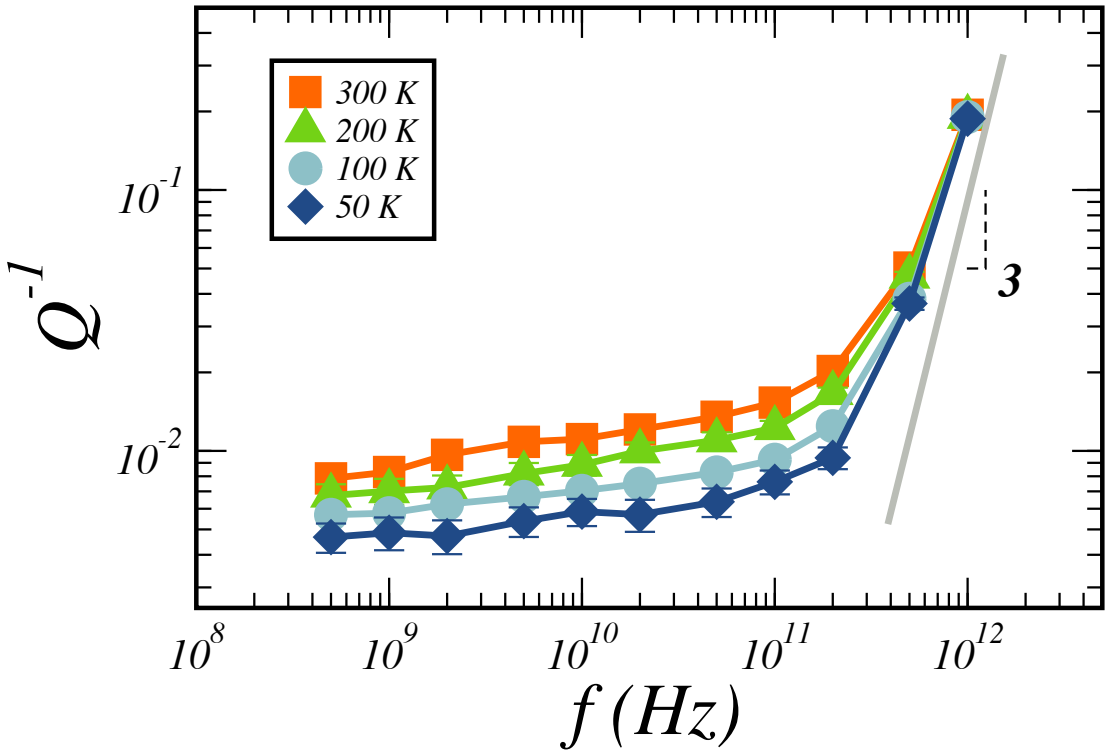


Average over 30 samples, 3 deformation directions and 20 oscillation periods.

In THz range , harmonic damping controlled by structural disorder: $Q^{-1} \sim f^{3}$.

Around 10-100 GHz, sharp crossover to an almost-flat but not completely flat frequency dependence.

MD-DMS in Ta_2O_5: first results



The high-frequency regime is virtually T independent.

Lowering the temperature reduces the dissipation in GHz range but seems not to affect the weak frequency dependence.

The crossover slightly shifts to lower frequencies upon cooling.

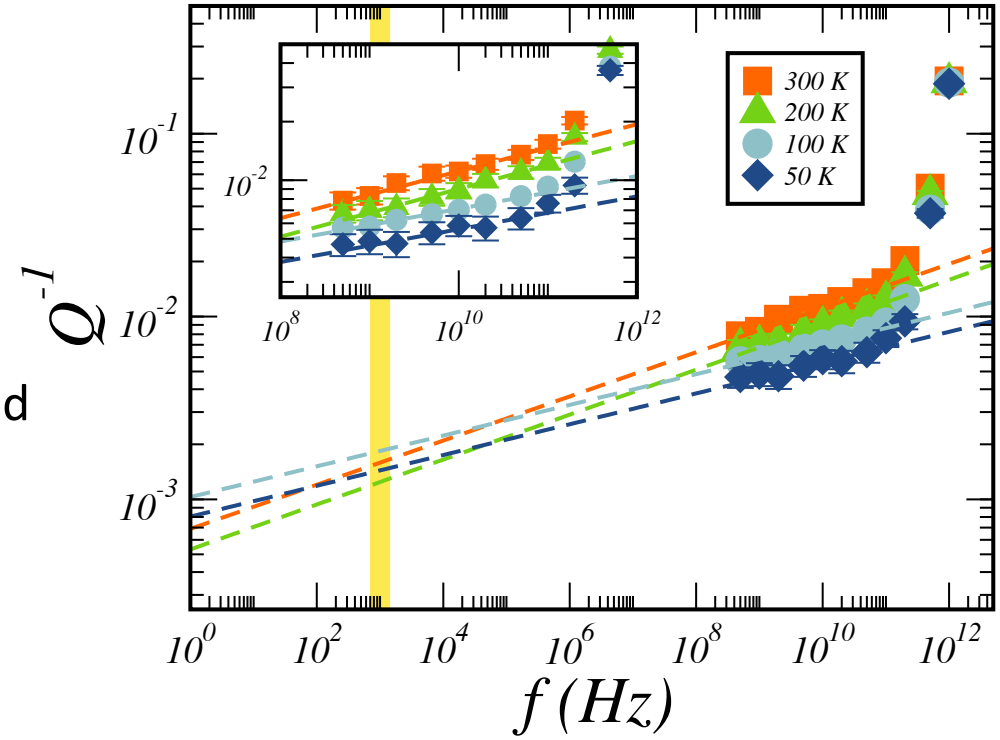
From simulation to experimental frequencies

In the GHz range we observe a power-law dependence on frequency over about 2 decades.

Power-law behavior reported in experimental works (see Huynh et al. PRMaterials 2017) and predicted by theory and models (TLS).

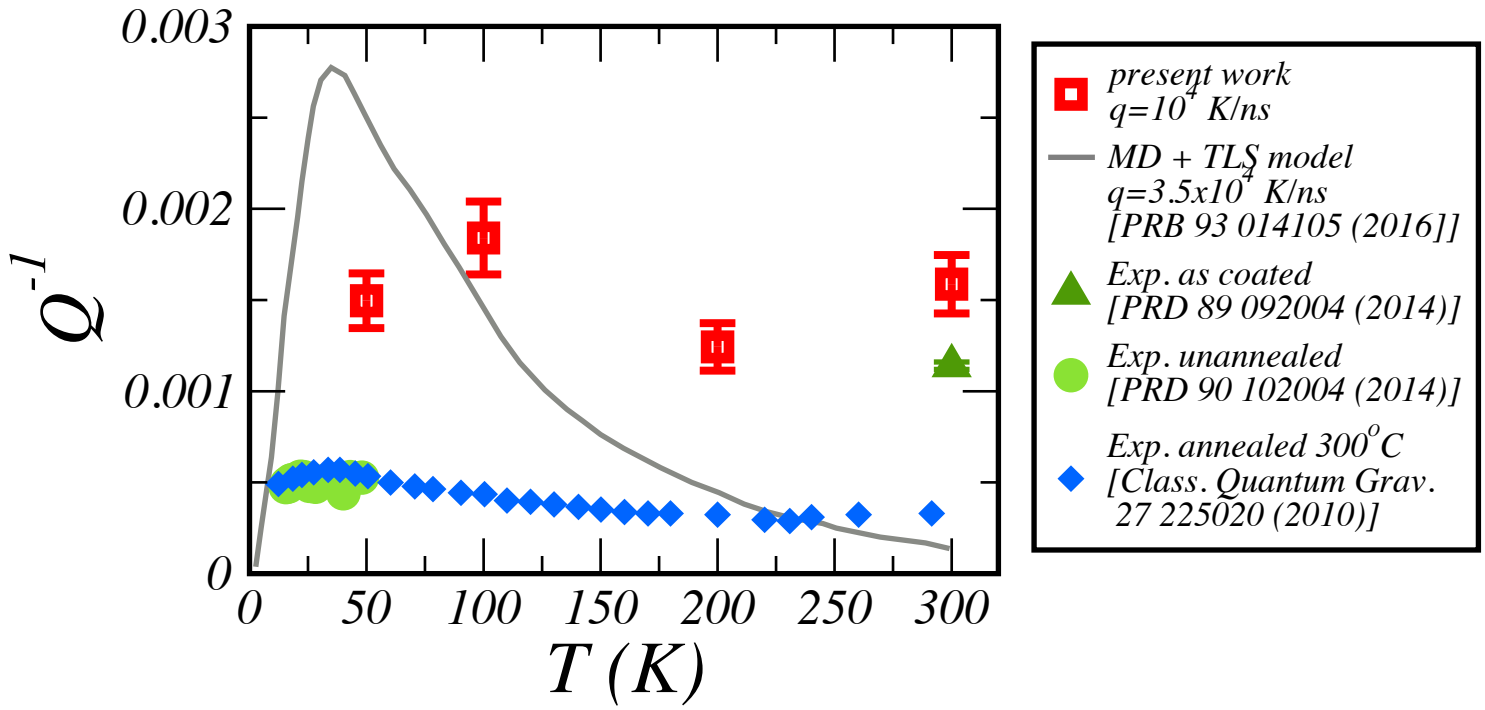
Fitted exponent:

$$n(300K) = 0.121,$$
$$n(200K) = 0.123$$
$$n(50K) = 0.084$$



We extrapolate the power-law from GHz to the acoustic frequencies

Comparing with experiments and TLS modeling



With respect to experimental data on as-coated/as-deposited Ta_2O_5:

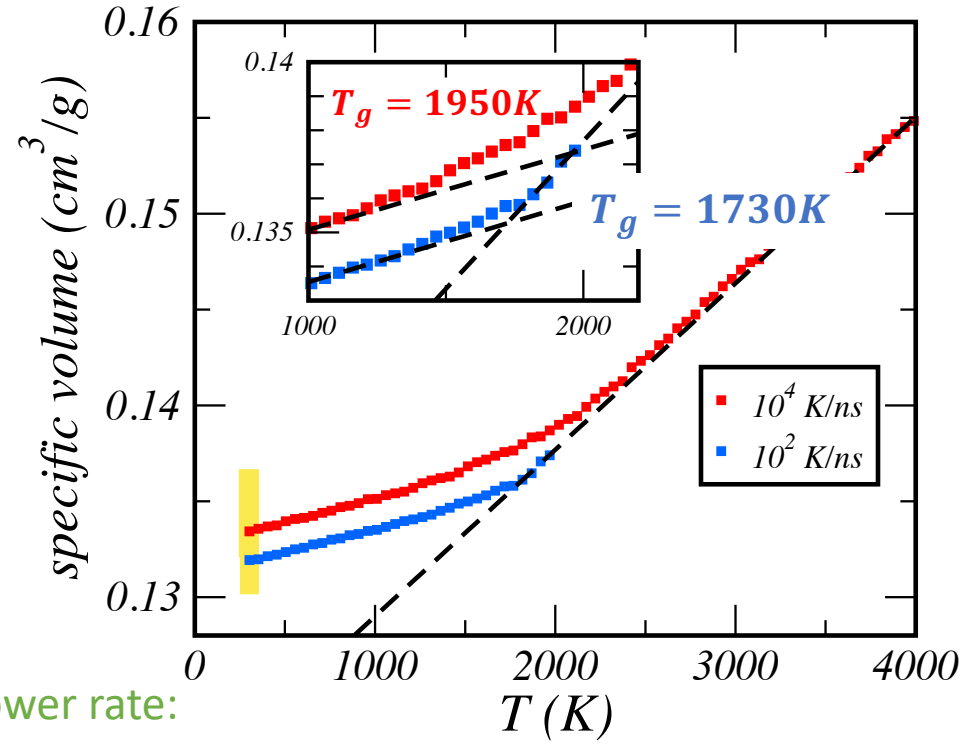
- MD-DMS overestimates dissipation by a factor 1.5 – 3
(but exp data strongly depends on the preparation method)
- **captures the temperature dependence**

Note: our preparation protocol is only qualitatively similar to experimental ones.

Annealing vs slow quenching

24h annealing at higher temperature is out of reach for simulations

The best we can do is to slow-down the cooling rate



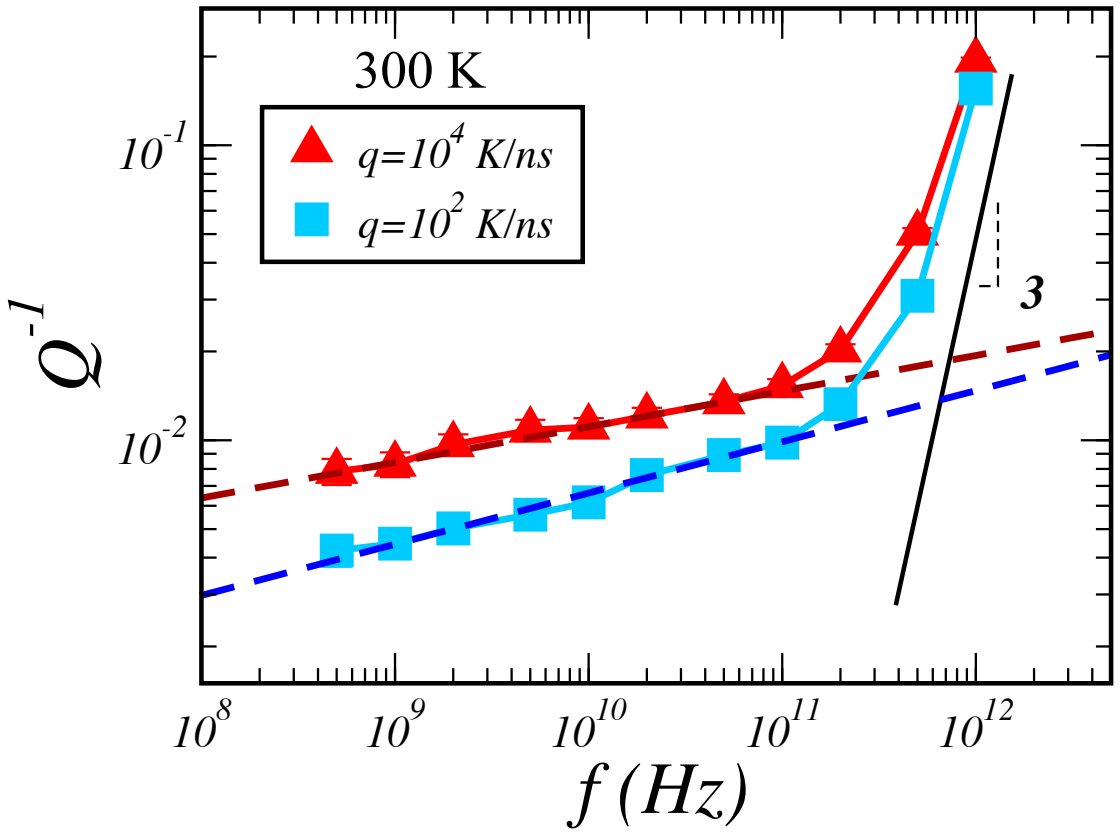
If cooling with a x100 slower rate:

Simulated glass transition shifts of about $200\ K$

Density increases from $7.52 \pm 0.04\ g/cm^3$ to $7.62 \pm 0.04\ g/cm^3$ (+1.3 %)

IS energy decreases from $-15.3 \pm 0.1\ eV/atom$ to $-15.5 \pm 0.1\ eV/atom$ (-1.25 %)

Mechanical losses via MD-DMS



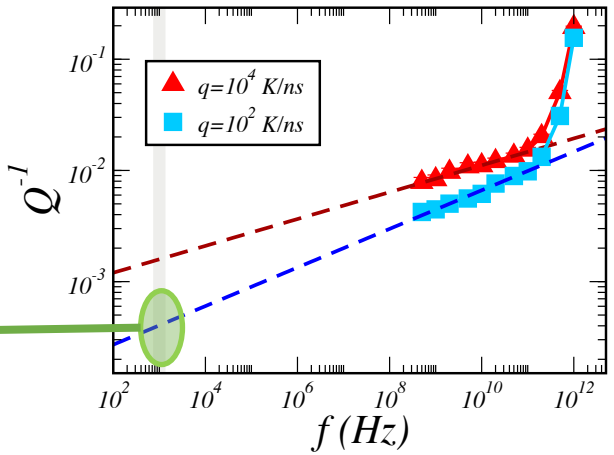
The slowly quenched glasses have lower mechanical losses in the accessible frequency range.

The **power-law dependence on f** (confirmed!) gets stronger:
the exponent changes from ~0.12 to ~0.19

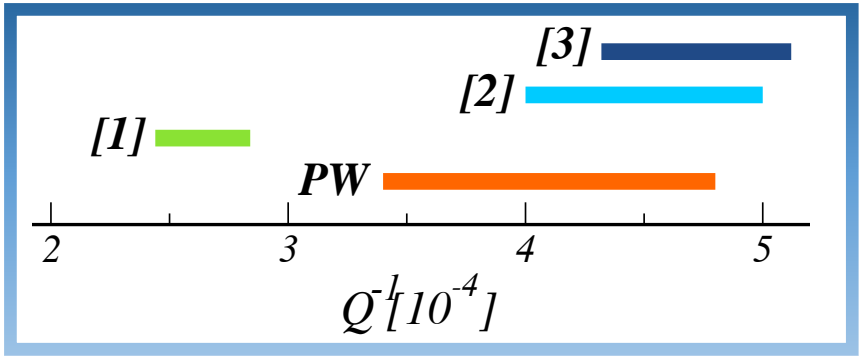
Extrapolating and comparing with experiments

Our estimate at 1 KHz

$Q[10^{-4}]$: 4.1 ± 0.7



	Exp. annealed [1]	Exp. annealed [2]	Exp. annealed [3]
$Q[10^{-4}]$	2.64 ± 0.2	4.5 ± 0.5	4.72 ± 0.4
our estimate	$+55\%$	-9%	-13%



[1]: Martin *et al.* Class. Quantum Grav. **26** 155012 (2009)

[2]: Vajente *et al.* Class. Quantum Grav. **35** 075001 (2018)

[3]: Principe *et al.* Class. Physical Review D **91** 022005 (2015)

Summary

We propose to apply **DMS** to measure mechanical losses in numerical simulations of glasses

We test the method on a model of **glassy Ta_2O_5**, obtained via melt quench, and whose properties match those of **experimental thin films**.
Two cooling rate, fast and slow, are considered.

Q^{-1} shows a robust **power-law behavior** in the range of frequency accessible to simulations (GHz), which allow to estimate the mechanical losses at acoustic frequencies.

If compared with relevant systems (not-annealed for fast quench or annealed for slow quench), **simulation results are in very good agreement with experimental data**.

In the whole procedure, **the only external input is the interatomic potential**.

Next steps and perspectives

Complete the characterization of Ta2O5 (temperature, further slow-down the cooling, …)

Apply the MD-DMS method to other systems:

- doped materials (Ta_2O_5 + ?)
- high coordination number glasses

Go beyond the “experimental” approach:

microscopic investigation of the dissipation mechanism(s)

<u>Our goal:</u>

A **reliable** (compare well with exps) and **efficient** (for demanded time and resources) method to calculate losses (Q) in any material of potential interest for the GW community.

We are only at the beginning but the road looks promising…

Thanks to the IT Center (University of Pisa) for providing valuable computational resources.