Mechanical loss calculation: In silico broadband spectroscopy of amorphous Ta$_2$O$_5$

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Overview

The goal of the coating collaboration: reduction of mechanical loss in mirror coatings

“Fundamental” (vs pragmatic) approach: understand the microscopic processes controlling dissipation

Current mirror coatings are amorphous

Standard modeling approach based on Two—Level System (TLS) model: dissipation due to atomic motion during transitions from one well to another, which are possible via coupling between external strain and thermal motion.

TLS model encloses some delicate points:
  i) TLS are independent? ii) coupling between TLS and strain described with macroscopic elasticity, despite disorder iii) sampling of barrier distribution $p(V)$, the key quantity...

Our work: Develop an alternative and theory-independent approach to provide reliable prediction of the experimental behavior of losses (Q factor) in coating materials.
Plan of the talk

- Molecular Dynamics simulations of Mechanical Spectroscopy
- Ta$_2$O$_5$ model and details of the simulations
- Glassy samples via melt quenching
- Simulation results and comparison with experimental data
Molecular Dynamics – Mechanical Spectroscopy (MD-MS)

Unilateral tensile oscillatory deformation

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

Strain amplitude in the linear elastic regime

$$E' = \frac{\omega}{N\pi} \int_{0}^{N2\pi/\omega} \sin(\omega t) \frac{\Sigma(t)}{\gamma_0} dt$$

$$E'' = \frac{\omega}{N\pi} \int_{0}^{N2\pi/\omega} \cos(\omega t) \frac{\Sigma(t)}{\gamma_0} dt$$

Storage and loss moduli

Dissipation

$$Q^{-1} = \tan \delta = \frac{E''}{E'}$$
Benchmark system: \( \text{Ta}_2\text{O}_5 \) 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 only 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]
“Annealing” with slower cooling

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 %)
Structural characterization: Pair Distribution Function

![Diagram showing pair distribution function with peaks at different temperatures: 300 K and 50 K. Peaks labeled Ta-O, Ta-Ta, O-O.]

- **300 K**
  - Peak at 2 Å for Ta-O
  - Peak at 4 Å for Ta-Ta
  - Peak at 3 Å for O-O

- **50 K**
  - Peak at 1.5 Å for Ta-O
  - Peak at 3.5 Å for Ta-Ta
  - Peak at 2.5 Å for O-O

Legend:
- □ 10⁴ k/ns
- ▲ 10² k/ns

$r$ (Å) vs. $g(r)$
Structural characterization: Bond Angle Distribution

$O$-$Ta$-$O$

$Ta$-$O$-$Ta$

![Graphs showing bond angle distribution at 300 K and 50 K](graphs.png)
Structural characterization: Voronoi analysis

Volume of Voronoi cells

Credits:
Voro++ project
Structural characterization: polyhedra connectivity

corner sharing (CS)

edge sharing (ES)

face sharing (FS)

Credits for representation K. Prasai et al.
Structural characterization: take-home message

Small but statistically significant structural differences between fast and slow quenched glasses.

Question:
how do they impact on losses?
**MD-MS in Ta$_2$O$_5$**

Average over 30 samples, 3 deformation directions and 20 oscillation periods. **The slowly quenched glasses have lower mechanical losses** (factor 2-3) in the accessible frequency range.
In THz range, harmonic damping controlled by structural disorder: \( Q^{-1} \sim f^3 \). High-\( f \) regime is virtually T-independent.

Around 10-100 GHz, sharp crossover to an almost-flat but not completely flat frequency dependence. The position of the crossover is slightly \( f \)-dependent.
In GHz range we observe a well-defined power law regime $Q^{-1} \propto f^{\alpha}$ with $\alpha \sim 0.1 - 0.2$ depending on $T$ and cooling rate.

Power-law behavior:

- supported by large body of evidence (sediment, soil and rocks, biological tissues, amorphous SiO$_2$, unstressed Si$_3$N$_4$ and other anelastic solids)
- compatible with exponential barrier distribution (narrower for slow quench)
We extrapolate the power-law from GHz to the acoustic frequencies.

We compare with TLS model predictions on glasses prepared by melt cooling at a comparable rate.
Comparing with experiments: fast quenched and as-deposited films

The spread of the exp data results from the influence of manufacturing and blurs the comparison.

Yet, our results capture the weak role of T, observed in experiments

\[ Q^{-1} \times 10^4 \]

\[ f (\text{Hz}) \]

\[ T (\text{K}) \]

\[ PW q=10^4 \text{ K/ns} \]

\[ \text{exp before annealing (d1)} \]

\[ \text{MD + TLS model} \]

\[ \text{exp as coated (d2)} \]

\[ \text{exp as deposited (d3)} \]

\[ d1: \text{M. Granata et al., in preparation} \]

\[ d2: \text{M. Granata et al., PRD 93, 012007 (2016)} \]

Comparing with experiments: slow quenched and annealed films

Remarkable qualitative and quantitative agreement between MD results and experimental data

*d4:* M. Granata et al., PRD 93, 012007 (2016).
*d5:* I.W. Martin et al. Class. Quantum Grav. 27 225020 (2010).
*d6:* G. Vajente et al. Class. Quantum Grav. 35 075001 (2018).
*d7:* M. Principe et al., PRD 91, 022005 (2015).
Summary

We propose to apply **Mechanical Spectroscopy** to calculate losses in numerical simulations of glasses.

We test the method on a model of **glassy Ta$_2$O$_5$**, 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 in the losses at acoustic range.

If compared with relevant systems (not-annealed for fast quenched / annealed for slow quenched), **simulation results are in impressive agreement with exp. data**.

**In silico** mechanical spectroscopy has an interesting predictive potential: it could provide quick and reliable estimates of the behavior of materials.
“Fundamental” (vs pragmatic) approach: understand the microscopic processes controlling dissipation (slide 2)

Fundamental AND pragmatic?

Thanks for your attention
What about other materials: the SiO$_2$ case

The low-frequency (TLS?) contribution become apparent already at $f \sim 10$ GHz, within the range accessible for the MD-DS method.