New insights on the glass transition through optical manipulation of chromophores

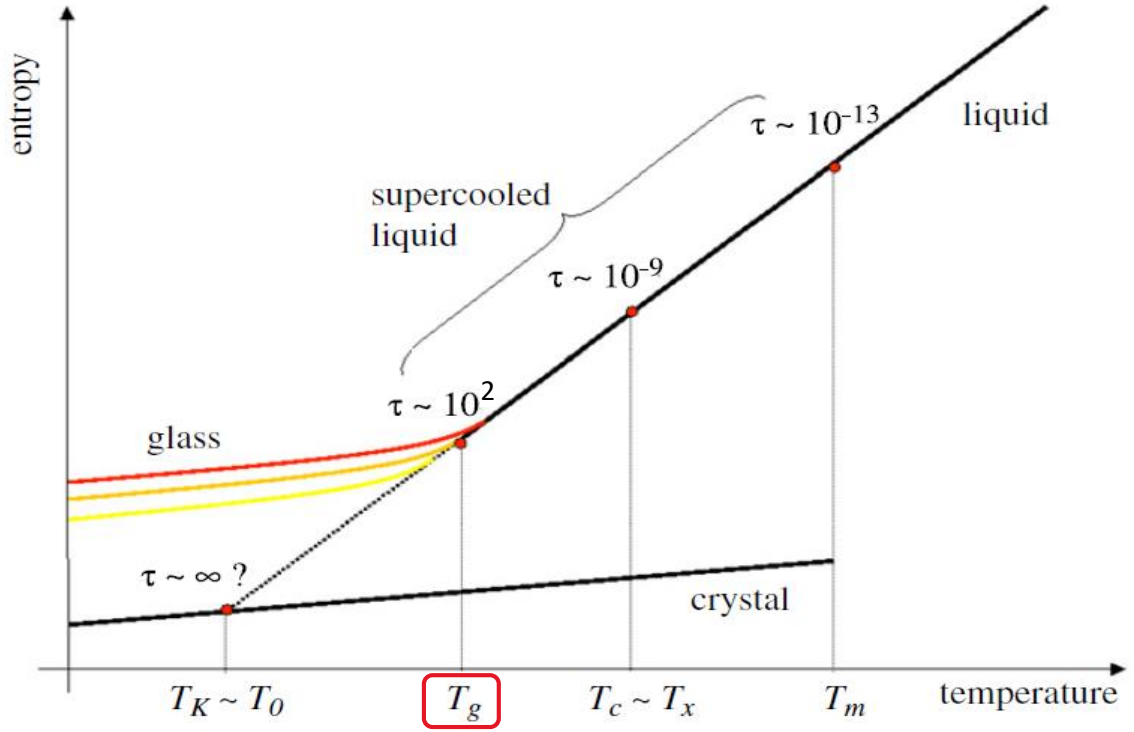
Eden DZIK

3rd year PhD student

Supervisors: François LADIEU (SPHYNX), David CARRIERE (LIONS)

The glass transition may be thermodynamic (or not)

At $T < T_g$, no time to wait for equilibrium



A. Cavagna, Physics Reports (2009)

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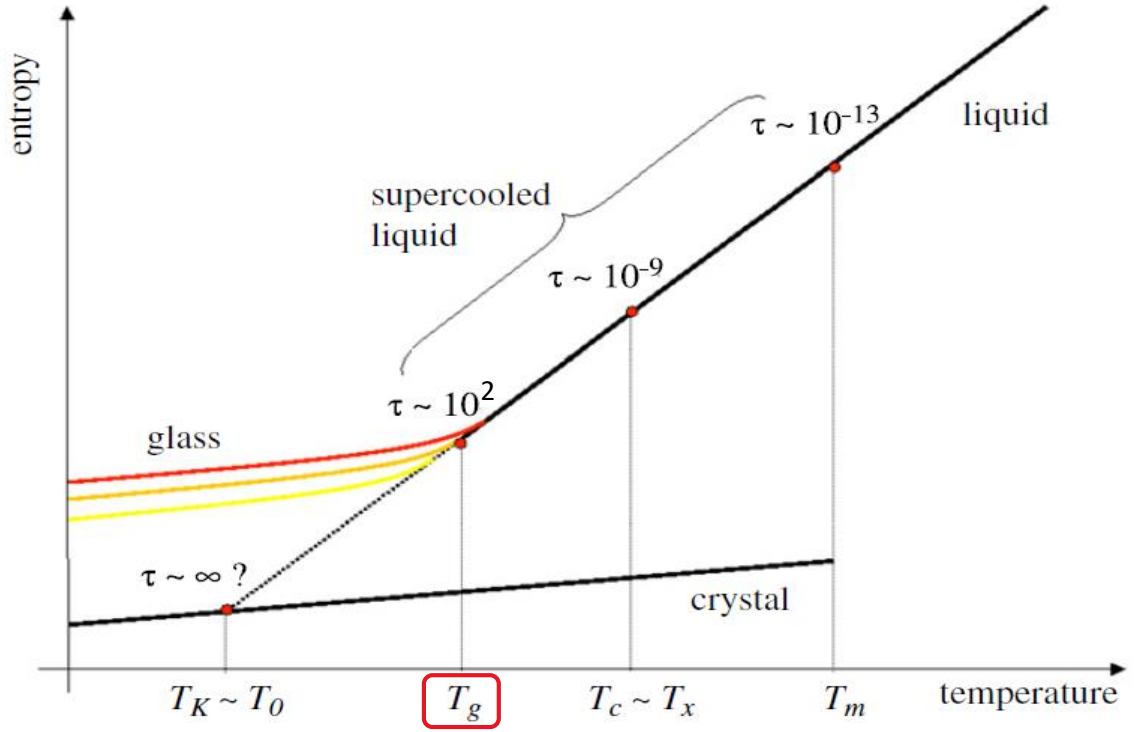

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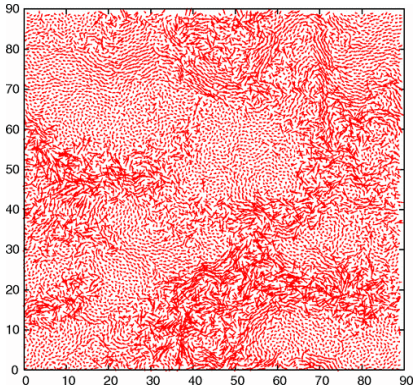


L.Berthier & G. Biroli, Rev. Mod. Phys (2011)

Dynamic correlations
➡ … static correlations?

Is the transition thermo/dynamic ?

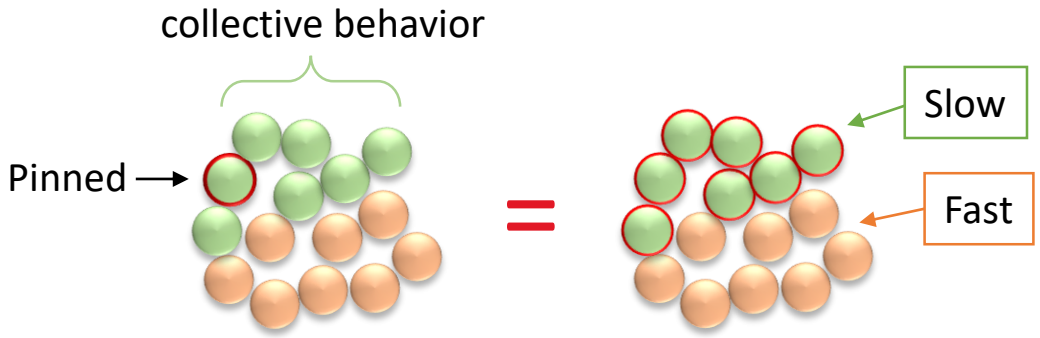
Amorphous order (RFOT)

Dynamic facilitation (KCM)

Amorphous order implies random pinning and photofluidization

Random pinning

Equilibrium glass obtained
by trapping 1 particle/glassite



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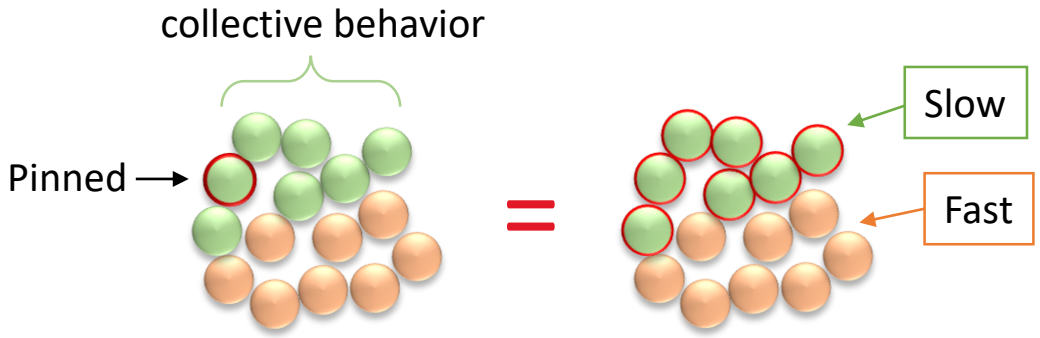
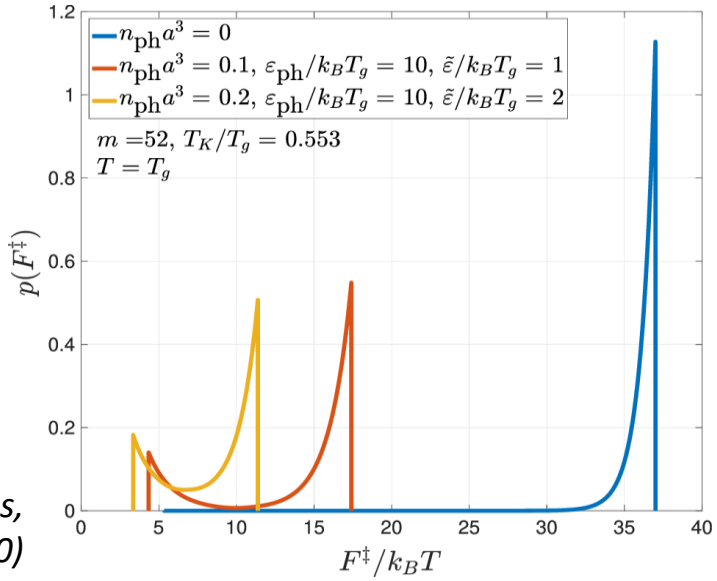


Photofluidization

Agitating local probe
lowers the free energy barriers



V. Lubchenko & P.G. Wolynes,
J. Phys. Chem. B (2020)

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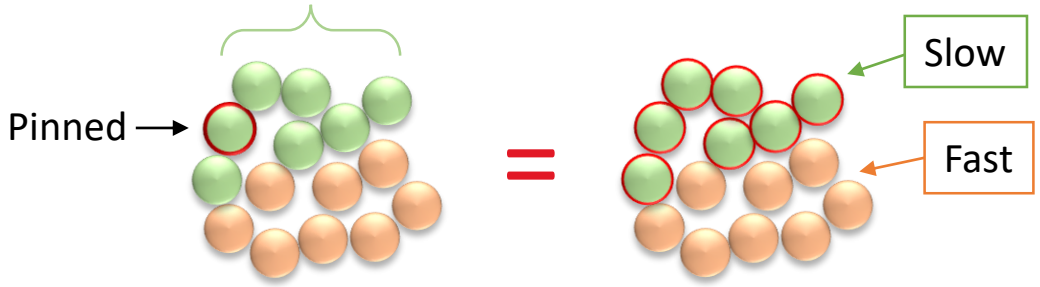
collective behavior



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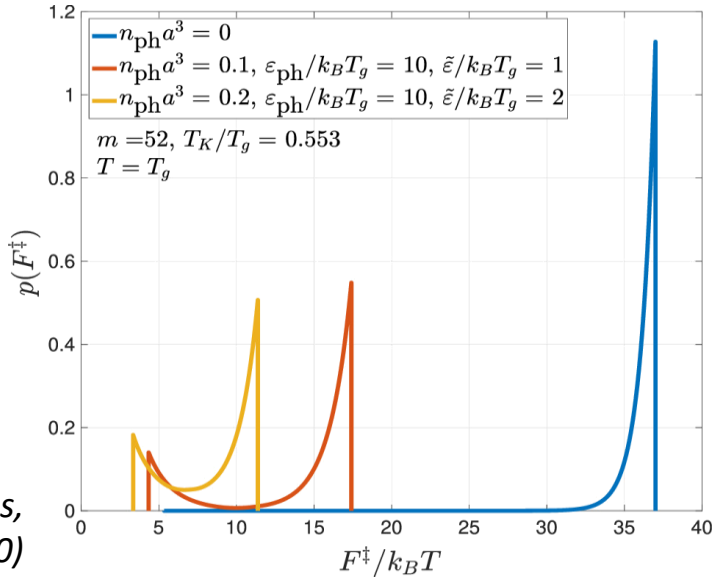


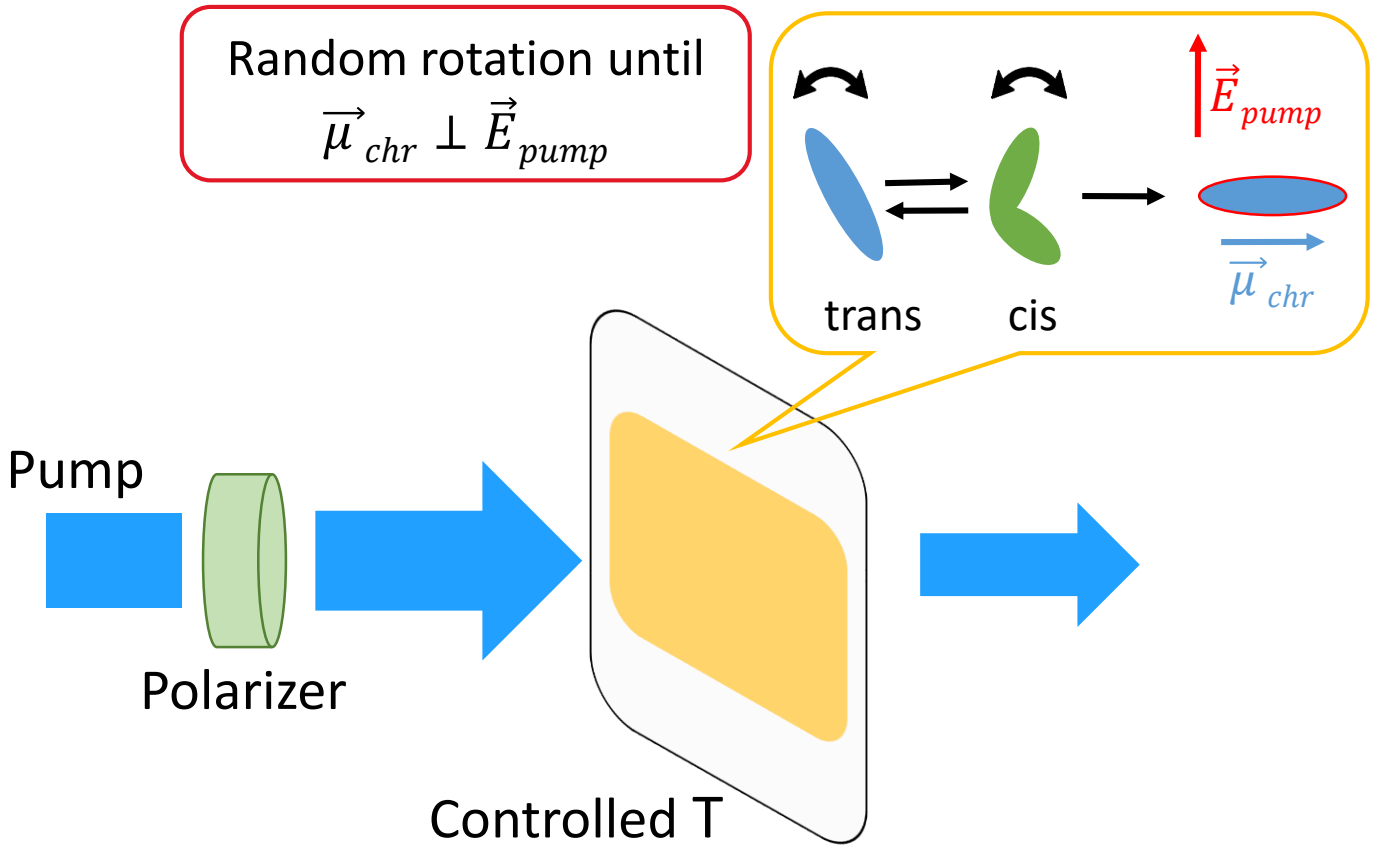
V. Lubchenko & P.G. Wolynes,
J. Phys. Chem. B (2020)

Dielectric spectroscopy
All molecules align with
an alternate field

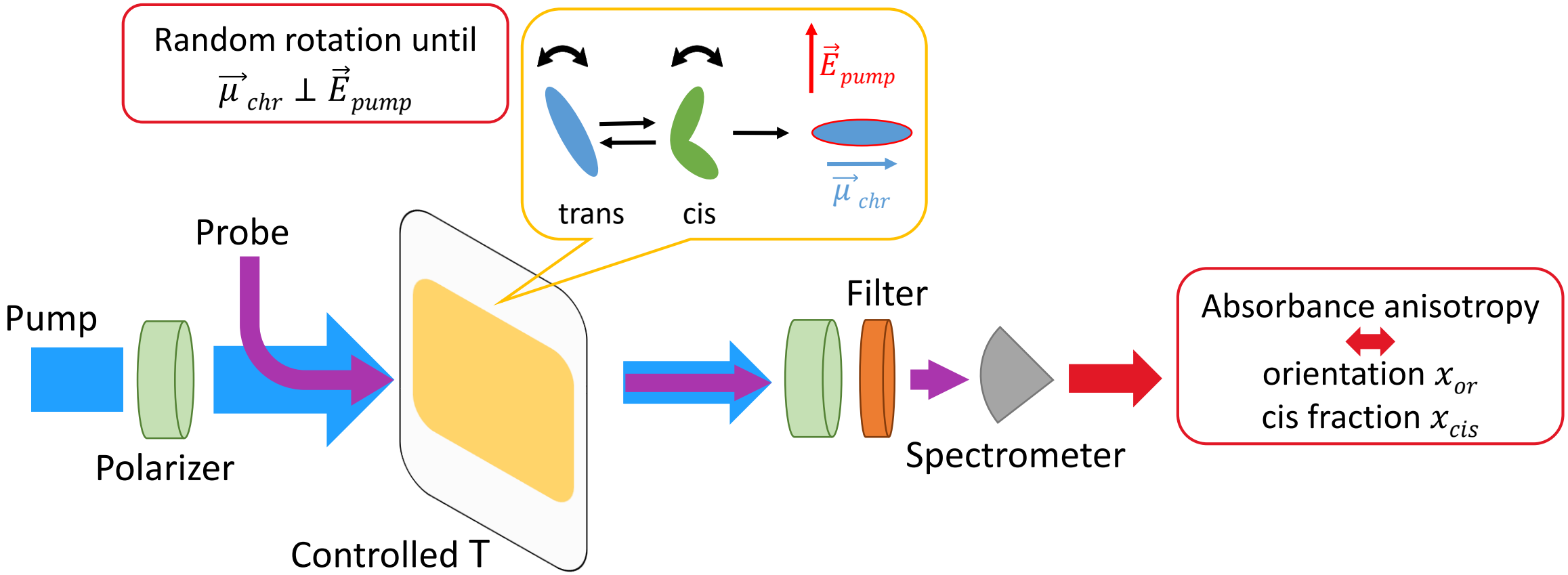
Goal:
Measure the glass global response
to a local perturbation

Photoactive probes
A few azo-grafted molecules
isomerize with light

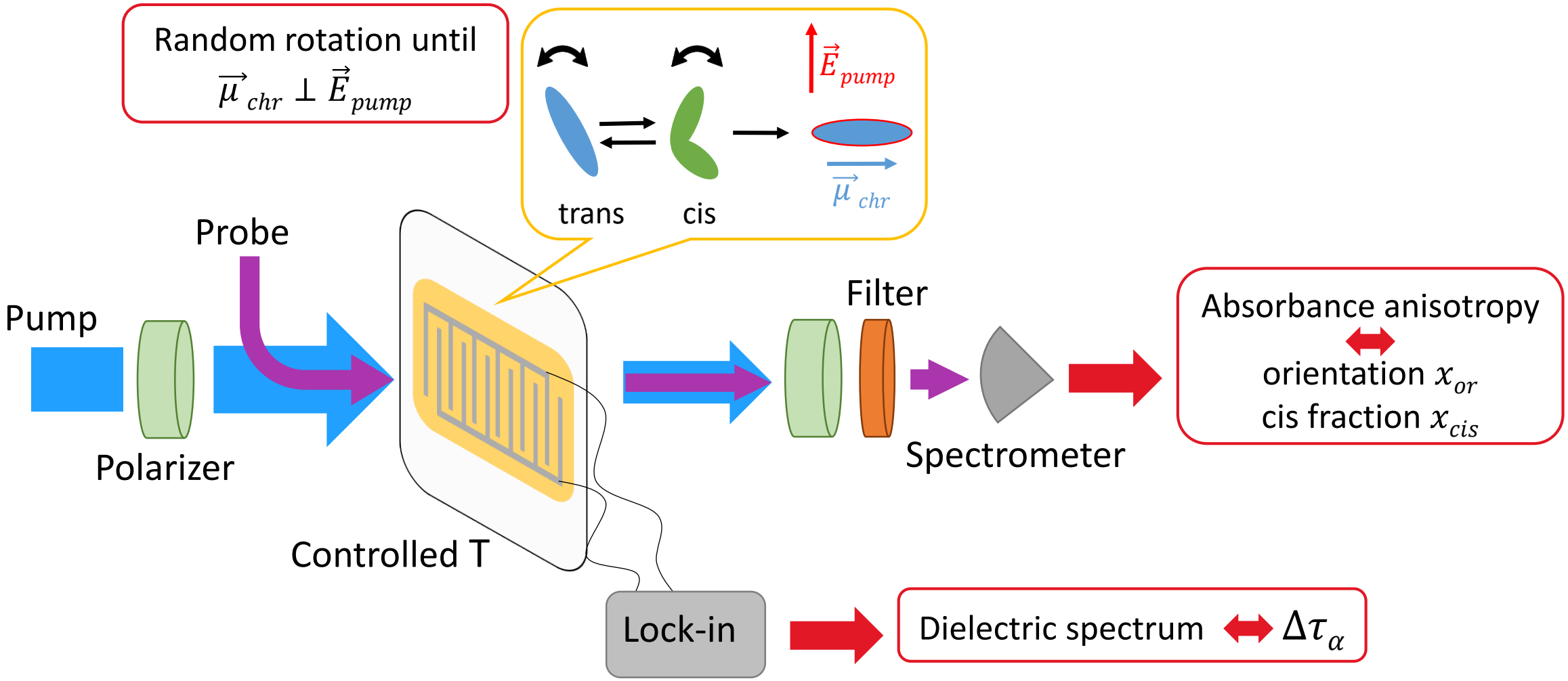
Photactivated azo-grafted molecules perturb the glass locally



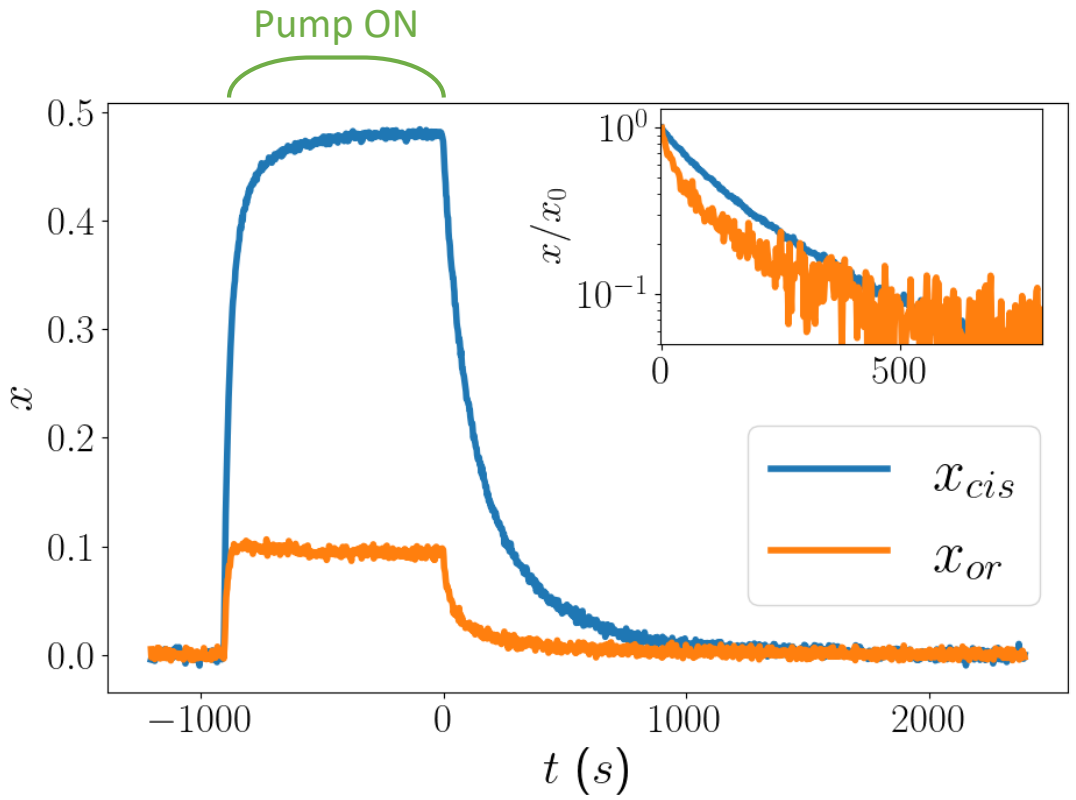
The chromophores activity creates a measurable dichroism



Dielectric spectroscopy measures the glass viscosity



Chromophores are coupled to the glass matrix



Heterogeneous relaxation
➡ stretched exponentials

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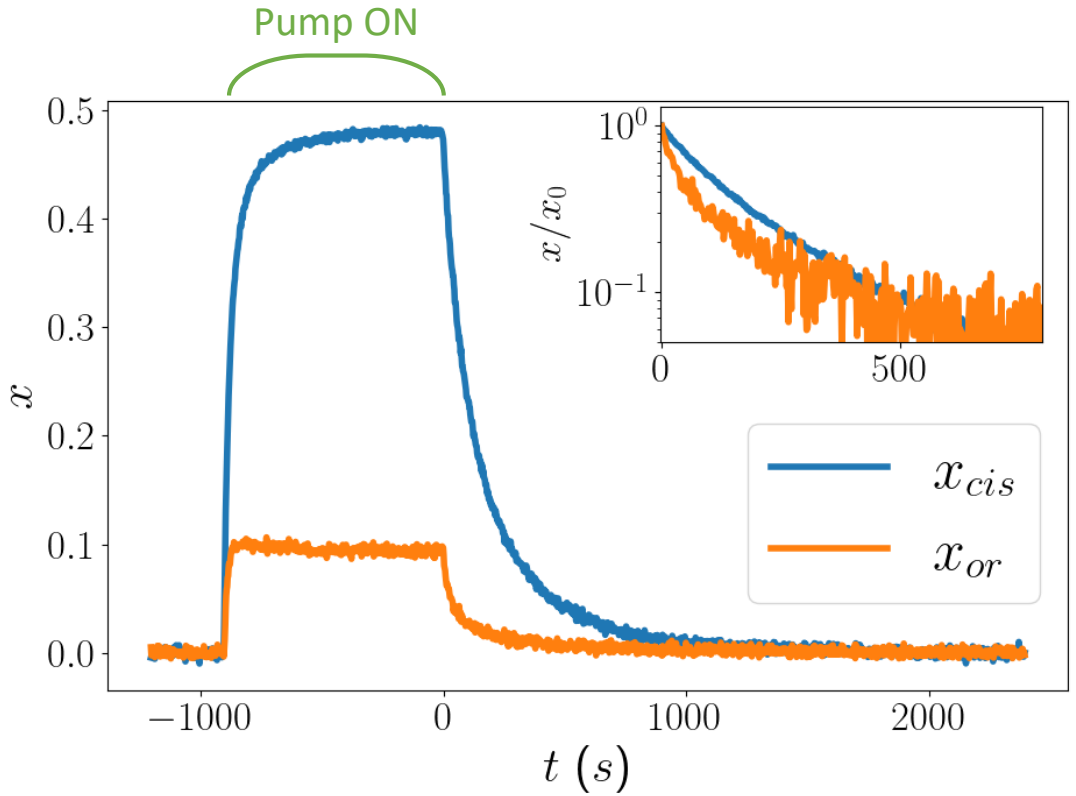
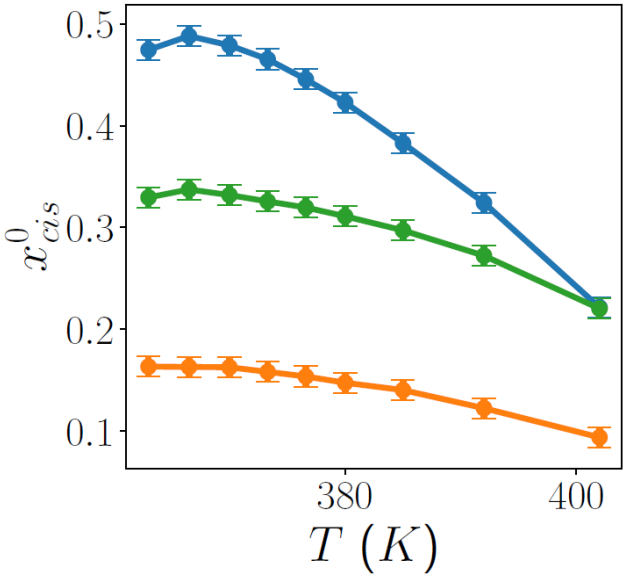
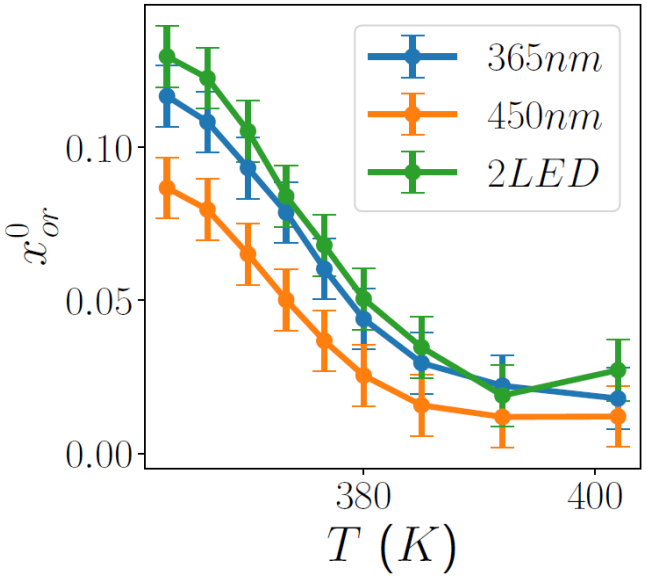


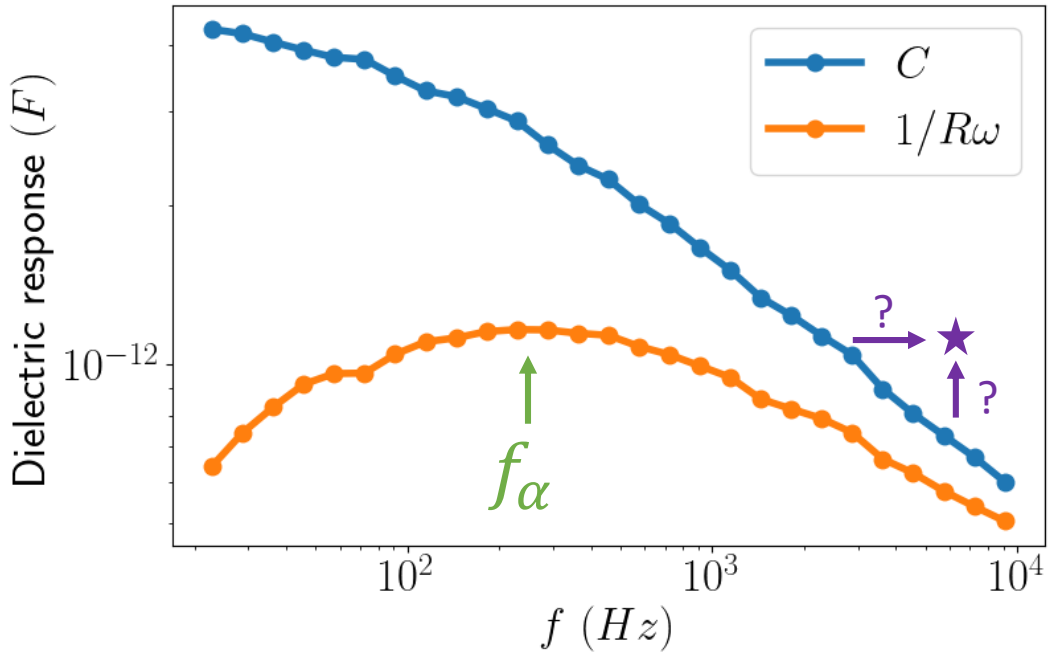
Heterogeneous relaxation
➡ stretched exponentials

x_{or} <<1 at high T $(\tau_\alpha < \tau_{isomerisations})$





The whole glass is affected by local perturbations



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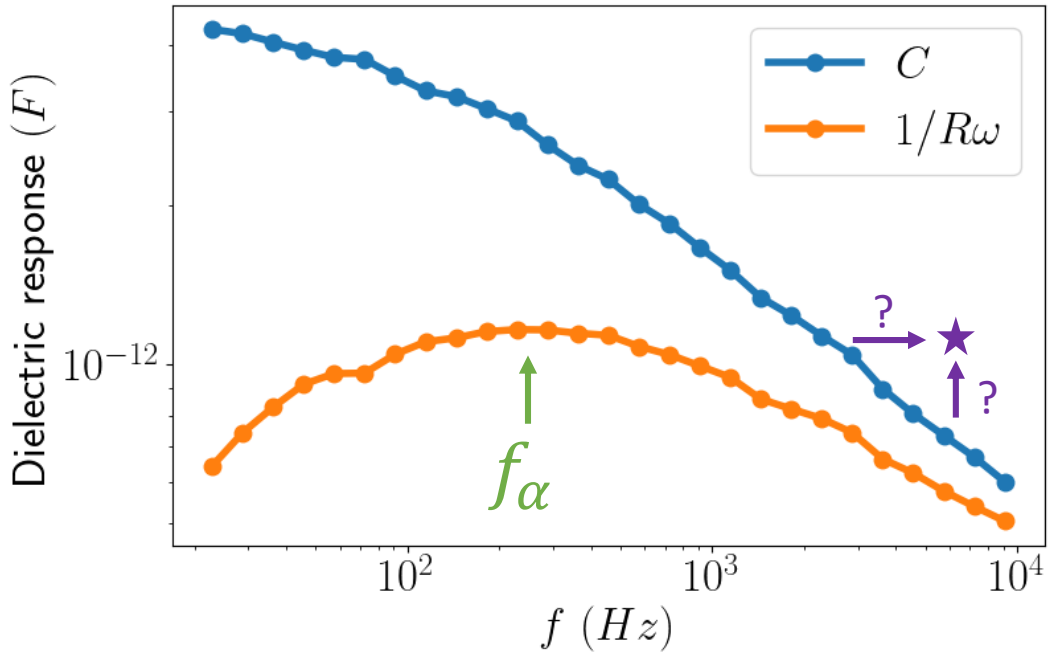

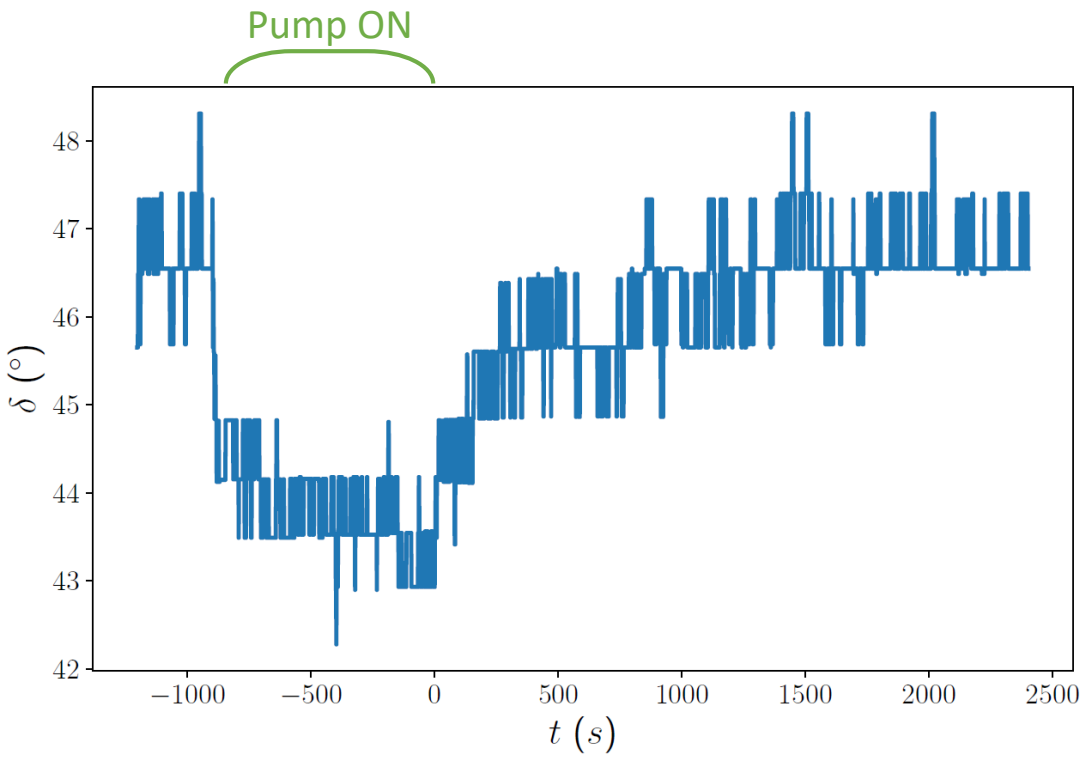

With only 1% azo,
the glass relaxes significantly faster

Conclusion

- Chromophores are coupled to the glass matrix
- The whole glass is affected by local perturbations

Our setup is ready to test RFOT predictions

Thank you for your attention!