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New insights on the glass transition through optical manipulation of chromophores

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Why do supercooled liquids dynamics slow down so abruptly upon cooling towards the glass transition temperature T_g ? This difficult question can be explored, among other techniques, with local probes, such as azobenzene-grafted molecules. They isomerize and orient themselves under polarized illumination, and their dynamics is affected by the glass matrix [1]. They are also proven to modify glass properties, as used for photo-induced mass transport in glassy polymers [2]. This strong coupling should allow to use these optically active molecules to change the dynamics of glasses at the scale of dynamical heterogeneities. Here, by using a pump-probe optical set-up on an azobenzene-doped molecular glass, we build up and measure the orientation parameter and cis-isomers fraction. At the same time, we follow the dynamics of the material by dielectric spectroscopy. With only 1% of azobenzene-grafted molecules, we observe a jump in the dielectric phase δ when the pump light is turned on with a maximum at $T = 370K$ (close to $T_g = 363K$). This effect is robust, i.e. it is similar when exciting only at 365nm (trans absorption peak) and also when adding a 450nm excitation (cis peak). Because δ is only sensitive to the relaxation time τ_α , we thus conclude that the relaxation time of the glass matrix varies significantly under illumination. Our results will be compared to a recent RFOT prediction [3] about the photo-activation of glassy dynamics around T_g . We will show that our experiment unveils new fundamental aspects of the glass transition.

REFERENCES

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