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^{8}Li Spin Relaxation as a Probe of the Modification of Molecular Dynamics by Inelastic Deformation of Glassy Polystyrene

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Glasses occupy more volume than required for molecular close packing. The distribution of this "free volume" is related to other key properties such as dynamic heterogeneity (stretched exponential relaxation). As a glass ages, it equilibrates by thermally activated structural relaxation producing permanent densification with slowed relaxation times. Mechanical deformation can significantly alter glassy structure and relaxation, leading to apparent over-aging or rejuvenation via irreversible plastic shear flow that explores microscopic configurations that are otherwise inaccessible[1].

Nanoimprint[2] is a technique that deforms thin polymer films by indentation of a patterned die for lithographic patterning and measuring mechanical properties. Few techniques are capable of studying local properties of polymer films, however the spin-lattice relaxation of implanted $^{8}Li^{+}$ is sensitive to the molecular dynamics in the glassy state, including modification by processing parameters[3]

We report initial results on a 300 nm thick atactic polystyrene film plastically modified by nanoimprint stamping[4] using a 1 mm ultra-smooth spherical die. While the \elip\ beam can easily be stopped in the film, the beamspot is $\sim$ 2 mm in diameter, so a large array of imprints was produced over an area $\sim$ 3 mm^2, leaving an inelastic strain of a few tenths of a percent over an areal fraction $\sim$ 20%.

To ensure the beam overlapped the imprinted area, a new method was developed. Using scintillation from an Al_2O_3 crystal, the beamspot image was fit with a Gaussian profile. Partially automation allowed the overlap to be maximized in real time. We find a small but significant change in the bulk of the film (away from the surface), compared to an unimprinted control, the relaxation is slower and more inhomogeneous (lower stretching exponent).

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[2] Traub, Ann.Rev.Chem.Bio.Eng. **7**, 583 (2016).
[3] McKenzie, SoftMatter**14**, 7324 (2018).
[4] Cross, Rev.Sci.Inst.**79**, 013904 (2008).

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