Induced cooperative motions in a medium driven at the nanoscale: Searching for an optimum excitation period

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Résumé en anglais	*Recent results have shown the appearance of induced cooperative motions called dynamic heterogeneity during the isomerization of diluted azobenzene molecules in a host glass-former. In this paper, we raise the issue of the coupling between these "artificial" heterogeneities and the isomerization period. How do these induced heterogeneities differ in the saturation regime and in the linear response regime? Is there a maximum of the heterogeneous motion versus the isomerization rate, and why? Is the heterogeneity evolution with the isomerization rate connected with the diffusion or relaxation time evolution? We use out-of-equilibrium molecular dynamics simulations to answer these questions. We find that the heterogeneity increases in the linear response regime for large isomerization periods and small perturbations. In contrast, the heterogeneity decreases in the saturation regime, i.e., when the isomerization half-period ($\tau p/2$) is smaller than the relaxation time of the material ($\tau\alpha$). This result enables a test of the effect of cooperative motions on the dynamics using the chromophores as Maxwell demons that destroy or stimulate the cooperative motions. Because the heterogeneities increase in the linear regime and then decrease in the saturation regime, we find a maximum for $\tau p/2 \approx \tau\alpha$. The induced excitation concentration follows a power-law evolution versus the isomerization rate and then saturates. As a consequence, the α relaxation time is related to the excitation concentration with a power law, a result in qualitative agreement with recent findings in constrained models. This result supports a common origin for the heterogeneities with constrained models and a similar relation to the excitation concentration.*
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