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Direct Nanoscale Characterization of Submolecular Mobility in Complex Organic Non-linear Optical Systems DANIEL KNORR, TOMOKO GRAY, TAE-DONG KIM, JINGDONG LUO, ALEX JEN, RENE OVERNEY — For organic non-linear optical (NLO) materials composed of intricate molecular building blocks, the challenge is to deduce meaningful molecular scale mobility information to understand complex relaxation and phase behavior. This is crucial, as the process of achieving a robust acentric alignment strongly depends on the availability of inter- and intra-molecular mobilities outside the temperature range of the device operation window. Here, we introduce a nanoscale methodology based on scanning probe microscopy that provides direct insight into structural relaxations and shows great potential to direct material design of sophisticated macromolecules. It also offers a means by which mesoscale dynamics and cooperativity involved in relaxation processes can be quantified in terms of dynamic entropy and enthalpy. This study demonstrates this methodology to describe the mesoscale dynamics of two systems (1) organic networking dendronized NLO molecular glasses that self-assemble into physically linked polymers due to quadrupolar phenyl-perfluorophenyl interactions and (2) dendronized side-chain electro-optic (EO) polymers. For the self assembling glasses, the degree of intermolecular cooperativity can be deduced using this methodology, while for the dendronized side-chain polymers, specific side chain mobilities are exploited to improve EO properties.

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