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| Title: | Fluorescence Depolarization Kinetics Captures Short-Range Backbone Dihedral Rotations and Long-Range Correlated Dynamics of an Intrinsically Disordered Protein |
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| Abstract: | Intrinsically disordered proteins (IDPs) do not autonomously fold into well-defined three-dimensional structures and are best described as a heterogeneous ensemble of rapidly interconverting conformers. It is challenging to elucidate their complex dynamic signatures using a single technique. In this study, we employed sensitive fluorescence depolarization kinetics by following picosecond time-resolved fluorescence anisotropy decays to directly capture the essential dynamical features of intrinsically disordered α-synuclein (α-syn) site-specifically labeled with thiol-active fluorophores. By utilizing a long-lifetime (≥10 ns) anisotropic label, we were able to discern three distinct rotational components of α-syn. The subnanosecond component represents the local wobbling-in-cone motion of the fluorophore, whereas the slower (∼1.4 ns) component corresponds to the short-range backbone dynamics governed by collective torsional fluctuations in the Ramachandran Φ–Ψ dihedral space. This backbone dihedral rotational time scale is sensitive to the local chain stiffness and slows down in the presence of an adjacent proline residue. We also observed a small-amplitude (≤10%) slower rotational correlation time (6–10 ns) that represents the long-range correlated dynamics involving a much longer segment of the polypeptide chain. These intrinsic dynamic signatures of IDPs will provide critical mechanistic underpinnings in a mosaic of biophysical phenomena involving internal friction, allosteric interactions, and phase separation. |
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