Assessment of molecular dynamics force fields for conjugated polymers using neutron and X-ray scattering

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Conjugated polymers (CPs) have been utilized in efficient photovoltaics, batteries, light emitting diodes, chemosensors, and many other new technologies. These materials are highly tunable, and so with the flexibility of organic synthesis techniques, this enables the application of molecular design principles to produce materials with superior performance. However, the use of computational methods for CPs is still limited by the lack of properly validated molecular dynamics (MD) modeling parameters (force fields) that can be used to probe structure and dynamics at length and time scales relevant to charge transport mechanisms. In this work, we outline our use of neutron and x-ray scattering techniques for an evaluation of molecular simulation force fields specifically produced for poly(3-hexyl thiophene) (P3HT), a model CP. Quasi-elastic neutron scattering (QENS) experiments are used along with MD simulations to quantitatively compare proposed force fields to an extensive set of experimental data. Wide-angle x-ray scattering is also used to compare experimental and computational polymer structures. QENS validation of MD force fields presents a unique opportunity to increase the accuracy of highly uncertain parameters that are used in the simulation of conjugated polymers, including backbone torsion potentials, atomic partial charges and non-bonded Lennard-Jones parameters. Many of these parameters are determined from *ab-initio* calculations, but unlike parameters for small molecules, they are not often parameterized to experimental data. Moreover, there is high variability in these parameters for the few force fields developed for CPs in the literature. Here, we provide an honest assessment of these force fields, discussing both strengths and limitations. Finally, we provide a vision for future work to accelerate the development of accurate force fields for these types of materials.