Efficient Single-run Implementation of Generalized Einstein Relation to Compute Transport Coefficients: A Binary-based Time Sampling

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A robust and simple implementation of the generalized Einstein (GE) formulation using a single equilibrium molecular dynamics (MD) simulation is introduced to compute diffusion and shear viscosity. The unique features underlying this framework are: (1) The use of a simple binary-based method to sample time-dependent transport coefficients results in a uniform distribution of data on a logarithmic time scale. Although we sample ``on-the-fly", the algorithm is readily applicable for post-processing analysis. Overlapping same-length segments are not sampled as they indicate strong correlations. (2) Transport coefficients are estimated using a power law fitting function, a generalization of the standard linear relation, that accurately describes the long-time plateau. (3) The use of generalized least squares (GLS) fitting estimator to explicitly consider correlations between fitted data points results in a reliable estimate of the statistical uncertainties in a single run. (4) The covariance matrix for the GLS method is estimated analytically using the Wiener process statistics and measured variances. (5) We provide a Python script to perform the fits and automate the procedure to determine the optimal fitting domain.The framework is applied to two fluids, binary hard sphere and a Lennard-Jones near the triple point, and the validity of the single-run estimates is verified against multiple independent runs. The approach should be applicable to other transport coefficients, since the diffusive limit is universal to all of them. Given its rigor and simplicity, this methodology can be readily incorporated into standard MD packages, using on-the-fly or post-processing analysis.