# Using accurate and precise determination of diffusion properties to complement quasi-elastic neutron scattering measurements

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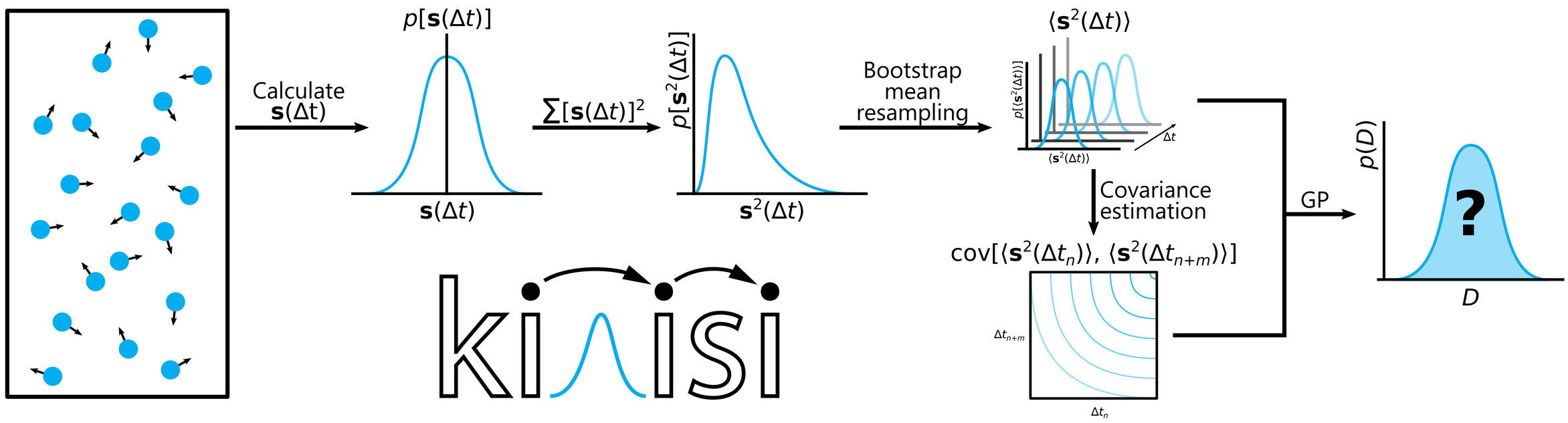
## Can we accurately estimate the variance in diffusion coefficients from molecular dynamics simulations?

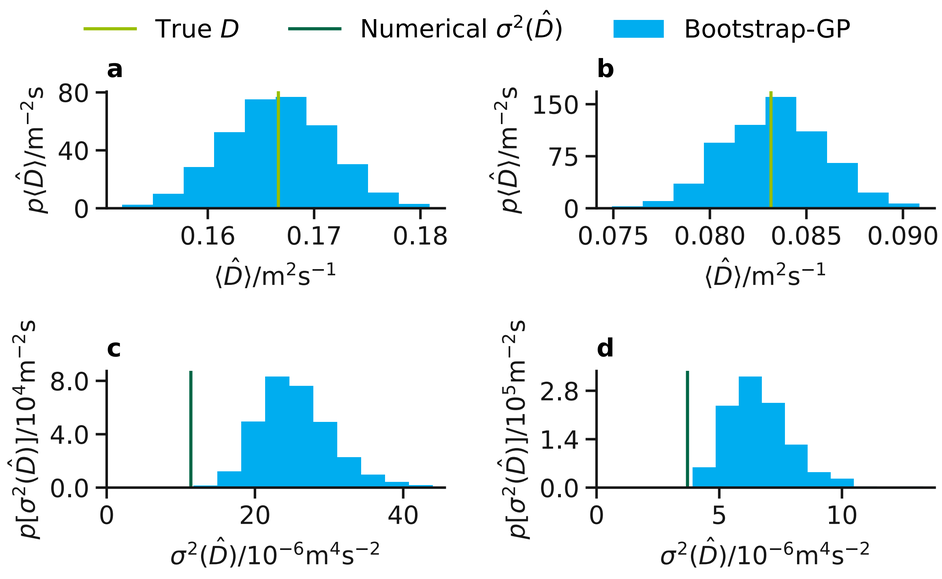
Atomic scale diffusion is a fundamental process, critical to the application of a range of functional materials and biological systems. Experimentally, this can be probed with quasi-elastic neutron scattering (QENS) measurements, which are often combined with molecular dynamics (MD) simulations. The stochastic nature and limitations in system size (number of atoms and simulation length) mean that a series of equivalent simulations will give a range of estimates of the diffusion coefficient (*D*). It is possible to describe the atomic mean-squared displacements with a Gaussian process, parameterised from a single MD simulation, to generate diffusive relationships equivalent to running repeated simulations, from which we can probe the distribution of *D*. This methodology is available as an open-source Python package, `kinisi`.




A schematic of the process `kinisi` uses for the estimation of the diffusion coefficient, with accurate variance quantification.

## Accurate distributions of *D* from a single simulation

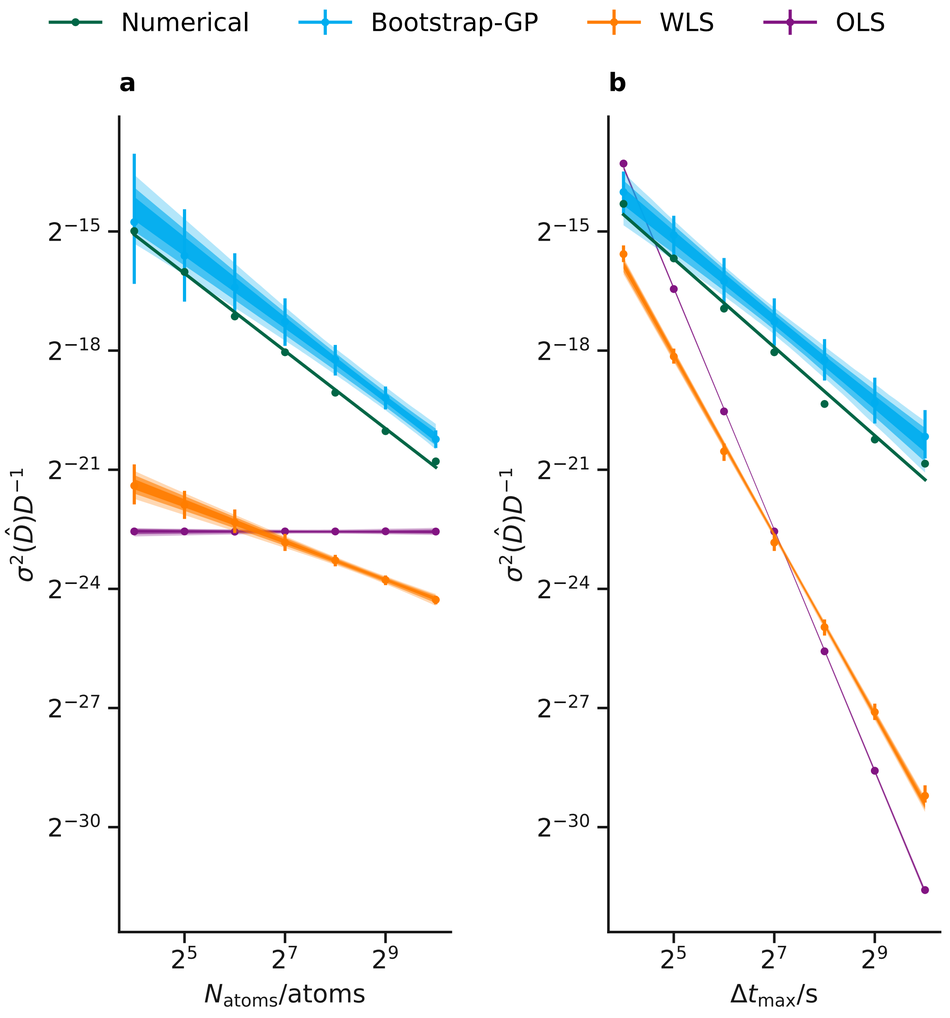
From a single simulation, this approach can accurately estimate the diffusion coefficient with high precision. Additionally, the variance, that would be observed from repeated simulations, is also accurately estimated, and slightly overestimated.




Accuracy of mean (**a** and **b**) and variance (**c** and **d**) estimation using the bootstrap-GP approach for a truly random (**a** and **c**) and temporally anti-correlated random walks (**b** and **d**). Determined for 128 atoms over 128 s and a step size of 1 ms$^{-1}$.

## Good accuracy across a range of system sizes

The bootstrap-GP variance estimation has good accuracy across a range of system sizes. Analogous for ordinary- and weighted-least squares approaches fail to accurately quantify the variance. Accuracy at low numbers of atoms is promising for investigations of collective motion.




Influence of system size on variance; changing number of atoms (**a**) or simulation length (**b**). Lines indicate modelled power-law relationships, where the thickness is indicative of the credible intervals. In (**a**) the simulation length is fixed to 128 s and in (**b**) the number of atoms is fixed to 128, both use a step size of 1 ms$^{-1}$.

## Can this be used to improve QENS analysis?

QENS is an important experimental technique to probe atomic and molecular diffusion. The analysis may be coupled with atomistic simulations, which may help in interpretation and/or extend the investigated range into regions inaccessible experimentally. The bootstrap-GP gives an unbiased estimation of the true diffusion coefficient. Furthermore, accurate uncertainty quantification enables better comparison with experimental results and allows for the application of Bayesian inference.

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