Exploring the Influence of Three-Body Interactions on the Self-Diffusivity of Krypton

Isabel Nitzke[1, S, C] and Jadran Vrabec[1]

[1]*TU Berlin, Berlin, Germany*
nitzke@tu-berlin.de

Historically, effective pair-potentials have dominated molecular modeling and simulation. However, recent advances in computing capabilities and the availability of trustworthy *ab initio* force fields allow for the exploration of more complex potentials that explicitly consider the three-body interactions. For decades, molecular simulations typically employed the Lennard-Jones potential as a basis to calculate the self-diffusivity of noble gases, such as krypton. Such simulations provide accurate results across a broad range of conditions, with the exception of high-density scenarios.

To address this limitation, quantum-based *ab initio* interaction potentials for two-body and nonadditive three-body interactions proposed by Jäger et al. [DOI: 10.1063/1.4943959] are implemented to investigate their influence on the self-diffusivity of krypton under gas-like and liquid-like supercritical conditions. This research aims to answer the critical question: How do three-body interactions impact the self-diffusivity of krypton, particularly at higher densities? By employing the force field by Jäger et al., a molecular dynamics study using the Green-Kubo formalism is conducted. A comparison of the present results with traditional two-body interaction simulations and experimental data sheds light on this issue. The scarcity of experimental data for transport properties, particularly in dense gases, has amplified the importance of such computational investigations. A thorough analysis not only enhances our grasp of noble gas behavior but also has wider implications for the development of more accurate force fields and the improvement of predictive models in materials science.