A NEW POTENTIAL ENERGY SURFACE FOR N_2O–He, AND PIMC SIMULATIONS PROBING INFRARED SPECTRA AND SUPERFLUIDITY

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High resolution infrared spectra of an N_2O molecule seeded in ultra-cold helium custers have been reported by two groups.[a,b] Early simulations of N_2O–$(He)_n$ clusters[c,d] were performed using potential energy surfaces (PESs) for which the dopant N_2O molecule was frozen at its equilibrium geometry. Since the evolution of the shift of the ν_3 band-origin of N_2O provides a key link to bridge the gap between micro and macro world, a new 3D PES was generated which incorporated the asymmetric-stretch Q_3 vibrational motion of the N_2O.[e] Bosonic PIMC simulations[f] based on this surface were then used to study rotational dynamics, and energetic and superfluid properties of N_2O–$(He)_n$ clusters. The evolution of the calculated shifts agree reasonably with the experimental results, but some quantitative discrepancies remain. To address this problem, a new four-dimensional N_2O–He PES has now been obtained which also takes account of the change in the average value of the Q_1 symmetric-stretch coordinate on excitation of ν_3. It has been fitted to a generalized MLR functional form[g] which imposes better long-range behaviour. Results obtained using this new surface will be presented.

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