

Rovibrational Excitation and Relaxation of Molecules in ${}^4\text{He}$ Nanodroplets

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We have performed quantum Monte Carlo simulations of N_2^+ ions in ${}^4\text{He}$ clusters and of Rb_2 in the triplet ground state, ${}^3\Sigma_u^+$, adsorbed on a ${}^4\text{He}$ surface. For that purpose we have obtained the potential energy surface for the N_2^+ -He interaction and for the Rb_2 -He interaction by ab initio coupled cluster calculations. Due to the weak attraction between Rb_2 and He, the ground state is an Andreev state, with the Rb_2 axis oriented parallel to the surface. The anisotropic dimple formed in the ${}^4\text{He}$ surface by Rb_2 is shallow, which explains why we found almost free Rb_2 rotation parallel to the surface. Out-of-plane rotation is hindered by the surface. We furthermore used correlated basis function theory to calculate damping of Rb_2 vibrations due to coupling to the ${}^4\text{He}$ surface. N_2^+ is solvated inside ${}^4\text{He}$ due to the strong attraction between the ionic molecule and ${}^4\text{He}$. We calculated binding energies, the structure of the ${}^4\text{He}$ “shell” around N_2^+ , and the rotational dynamics of $\text{N}_2^+ \cdot {}^4\text{He}_N$ clusters. We discuss the effect of the additional He-He repulsion due to the dipole moments induced by N_2^+ on the chemical potential and on the ${}^4\text{He}$ density around N_2^+ . Regarding rotational dynamics, N_2^+ is an interesting case of a light rotor (favoring weak rotational coupling with ${}^4\text{He}$) with a strong interaction with ${}^4\text{He}$ (favoring strong coupling). We present our results of rotational excitations of $\text{N}_2^+ \cdot {}^4\text{He}_N$.