

Microscopic manifestation of superfluidity in doped helium clusters: Spin solvent effects

Doing a parallelism of nuclei/electrons with dopant/helium atoms, a quantum chemistry approach is used to perform spectral simulations of molecular species embedded in helium clusters of variable size. To account for the spin characteristics of the solvent, Hartree or Hartree-Fock methodology is applied to obtain energies and structural properties of the aggregates. In this frame, one considers the distortion induced on the molecular dopant by the surrounding helium atoms. As a consequence of the different spin multiplicity, inherent to the boson/fermion nature of the helium environment, selection rules for Raman[1] or infrared (IR)[2] spectra depending on the polar/non polar character of the dopant, predict completely different profiles in these diverse scenarios. This finding agrees with, *e.g.* the experimental infrared spectra of oxygen carbon sulfide, OCS, in helium nanodroplets[3]. Below the critical transition temperature of ${}^4\text{He}$, ~ 2.12 K, the boson profile is close to the gas phase spectrum of the dopant. So, the **impurity seems to be freely rotating inside the droplet, a microscopic manifestation of superfluidity**. In turn, even at lower temperatures (but above ~ 0.003 K, the superfluid transition temperature of ${}^3\text{He}$), the corresponding fermion spectrum shows an unstructured broad shape resembling the case of heavy molecules immersed in liquids. **This peculiar behavior is illustrated in Fig. 1** through simulated IR spectra of the iodine chlorine (ICl) molecule, a species which mimics to the quasi-linear OCS molecule.



Figure 1: Simulated IR spectra of $\text{ICl}(X)$ in helium nanodroplets: left panel, ${}^4\text{He}$ environment; right panel, ${}^3\text{He}$ environment.

References

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