Nonadiabatic Alignment of Molecules in Helium-4 and Superfluidity around Clusters

Robert E. Zillich

Institut fuer Theoretische Physik, Johannes Kepler Univ., Altenbergerstr. 69, Linz, Austria

Pump-Probe spectroscopy has been recently used for alignment of molecules in superfluid helium-4 droplets. In gas phase, a short non-adiabatic pulse leads to a coherent revival pattern of the molecule orientation. In 4He, the dynamics is profoundly altered (e.g. the revivals are missing), hence the response to the helium environment to this highly nonlinear kick of the molecule cannot be understood in the familiar terms of superfluidity. We present our approach to solve the coupled dynamics of molecule and helium and show some early results. In a second part, we take the step from molecules to clusters in helium-4 and discuss the solvation of Mg and Na clusters. Using path integral Monte Carlo simulations, we demonstrate that the superfluid response around a larger dopant like a Mg11 cluster differs from the response around a small dopant: the decoupling of dopant rotation and helium motion can set in before the dopant is fully solvated. We calculate the helium solvation density and energy of various Mg and Na clusters, based on simple models for the interaction. As expected from experiments, small Na clusters form dimples on the He droplet surface rather than being surrounded by He, although the limit of large He droplets as encountered in experiments is out of reach in our simulations.