Laser-induced alignment of molecules in helium nanodroplets: Environment-limited rotation

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We use 10 ps long moderately intense laser pulses to initiate rotation of molecules embedded in helium nanodroplets and measure the time-dependent molecular alignment by femtosecond-laser-pulse-induced Coulomb explosion imaging. Our studies carried out on I_2, OCS and CS_2 and C_2HI molecules show pronounced yet decreasing oscillations in the degree of alignment for the first 200 ps after the alignment pulse. These oscillations have a period of 40-50 ps – depending on the molecule. For each molecule the period decreases as the intensity of the alignment pulse is increased but, strikingly, saturates at a certain value that does not change even when the alignment pulse intensity is more than doubled. The observations stand in stark contrast to results for gas phase molecules measured under identical laser conditions where no oscillations are observed.

Our preliminary interpretation of the observations is that the surrounding helium droplet environment imposes a maximum value on the angular speed that the molecule and its local solvation shell of He atoms can attain from the interaction with the alignment pulse. This maximum is related to the Landau velocity, i.e. the maximum velocity at which objects in superfluid helium can move without causing elementary excitations. As such the experimental findings may provide a direct measurement of superfluidity in finite-size helium systems.

In addition, by lowering the energy of the alignment pulse we observe rotational revivals in several molecules inside helium nanodroplets. Previously, revivals have only been observed for I_2 molecules [1]. These new experiments show that rotational revivals is a general phenomenon and will help to understand alignment of molecules inside helium droplets better. Further analysis is ongoing and being developed based on the angulon quasiparticle theory [2].

References:
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