Three dimensional molecular alignment inside helium nanodroplets

Adam S. Chatterley[*1], **Benjamin Shepperson**[*2], **Henrik Stapelfeldt**[*3],

[*] Institute for Kemi, Aarhus University, 8000 Aarhus C, Denmark

Synopsis Three-dimensional laser-induced molecular alignment has been demonstrated at a high repetition rate for molecules doped into helium nanodroplets. The cold environment leads to better alignment than can be obtained in an isolated environment.

We demonstrate that three-dimensional spatial alignment of molecules embedded in helium nanodroplets can be achieved using nonresonant elliptically polarized 160 ps laser pulses at a 1 kHz repetition rate. Using Coulomb explosion imaging, the 3D confinement of 3,5-dichloroiodobenzene in helium droplets is compared to that of isolated molecules. Ion-ion covariance mapping reveals that the primary differences in the observed Coulomb explosion ion images are due to scattering off of helium atoms inside the droplet. When this non-axial recoil is taken into account, we find that a higher degree of 3D alignment can be obtained inside the droplets than in the isolated environment.

The 3D alignment follows the alignment laser pulse adiabatically in the gaseous environment, however in the helium environment a delayed response is observed, opening the possibility of field-free 3D alignment. This technique should pave the way for next-generation molecular dynamics and diffraction experiments, performed within a cold helium solvent.

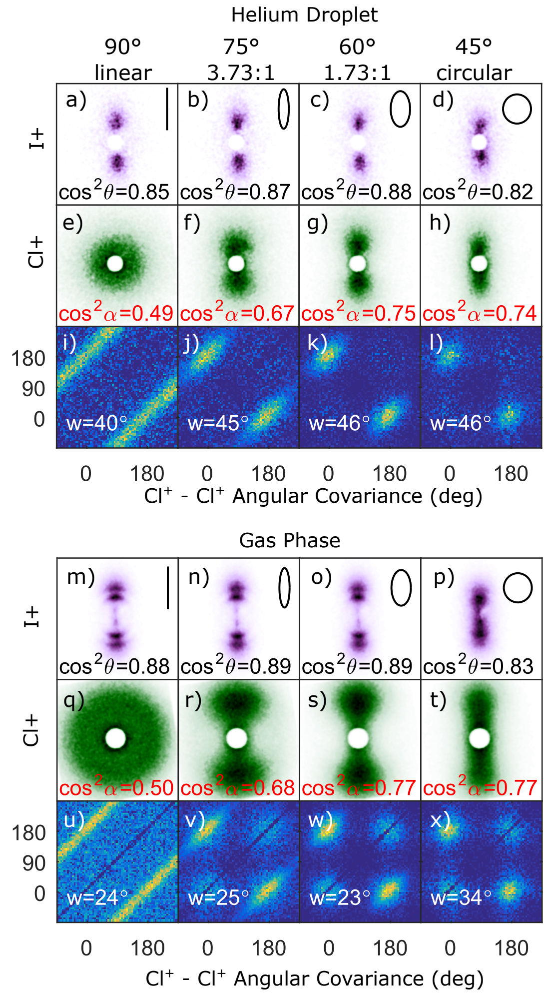

Figure 1. Ion images and covariance maps for 3D aligned 3,5-difluoroidobenzene with differing alignment ellipticities. Top: helium droplet, Bottom: gas phase. The top rows show I^+ images, the middle row Cl^+ images, and the bottom row Cl^+ - Cl^+ covariance maps.

[1]E-mail: aschatterley@chem.au.dk
[2]E-mail: shepp@chem.au.dk
[3]E-mail: henriks@chem.au.dk