Spatio-temporal dynamics of molecular alignment in intense femtosecond laser fields

Francesco Mazza[1, 2], Leonardo Castellanos[2], Meena Raveesh[3], Joakim Bood[3], Michele Marrocco[4], Isaac Boxx[1] and Ali Hosseinnia[1]

1. *Chair of Optical Diagnostics in Energy, Process and Chemical Engineering, RWTH Aachen University, 52062 Aachen, Germany*
2. *Faculty of Aerospace Engineering, Delft University of Technology, Kluyverweg 1, 2629 HS Delft, The Netherlands*
3. *Division of Combustion Physics, Department of Physics, Lund University, S-221 00 Lund, Sweden*
4. *Dipartimento di Fisica, Università di Roma "La Sapienza", I-00185 Roma, Italy*

The interaction of femtosecond (fs) laser pulses with molecular rotors in gas-phase media results in the excitation of Raman coherence between rotational eigenstates of the molecules. The same interaction affects the statistical populations of the eigenstates, leading to "permanent" alignment of the molecules along the polarization plane of the fs laser field [1]. This effect, though often ignored in fs coherent Raman scattering (CRS) diagnostics, has been extensively reported in molecular alignment experiments. Less attention has been paid to the spatial distribution of permanent alignment over the cross-section of the probe volume and its temporal evolution in the collisional environment of the gas. As the permanent alignment depends quadratically on the intensity of the fs laser pulse, the degree of alignment over the cross-section of the probe volume reflects the irradiance profile of the fs laser beam.

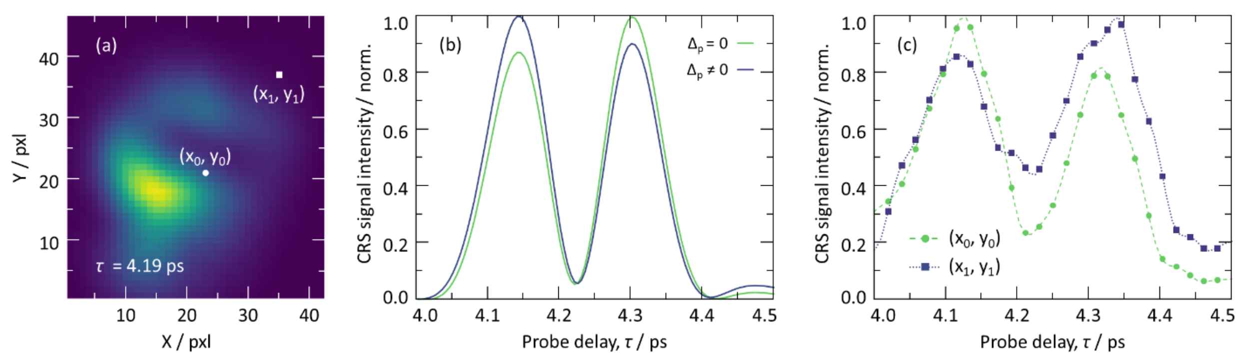

Figure (a) Shot-averaged fs CRS image at half-revival of the N_2 wave packet (4.19 ps) [2]. (b) Theoretical half-revival trace with (blue) and without (green) permanent alignment effects. (c) Experimental half-revival traces, measured at the two locations of the probe volume cross-section indicated in panel (a).

Here we show how the evolution of the rotational wave packet at each location in the probe volume arises from the non-linear interaction of quantum revivals and permanent alignment. The spatial phase [2] and amplitude profiles of the laser fields both contribute to the local alignment dynamics, as shown in the figure. We furthermore perform pump-probe experiments to quantify the lifetime of the permanent alignment. Our experimental results indicate a lifetime of a few tens of ps, much shorter than the collisional dephasing of the wave packet and comparable to its rephasing period. These results have important implications for fs CRS diagnostics and its measurement accuracy.

1. Md. Z. Hoque *et al.*, *Phys. Rev. A* **84** (2011) 013409.
2. A. Hosseinnia *et al.*, *in review*.