Intrinsic Calibration of Molecular Alignment Using Rotational Echoes

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Coherent control of molecular rotations has been thoroughly studied and vastly utilized in the last three decades. Motivated by obtaining spectroscopic signatures from the molecular frame, researchers have invented a large and sophisticated toolbox for controlling the angular distribution of gas phase molecules via their interaction with strong laser pulses. In fact, the field of rotational control has evolved in parallel to and from rotational coherence spectroscopy that aims to obtain accurate measurements of molecular rotational coefficients, from which the molecular structure can be deduced. © 2021 The Author(s)

The basic physics of laser-induced rotational dynamics is well understood: an ultrashort laser pulse interacts with the molecules via their anisotropic polarizability and applies an effective torque that rotates them toward the polarization axis of the pulse. Shortly after the interaction the rotating molecules become aligned, i.e. with their molecular axes preferentially lying along the polarization axis of the excitation pulse. However, the quantification of the degree of this alignment (or, how effective the interaction between the pulse and the molecules is) is quite challenging and requires accurate assessment of various experimental parameters, that are not necessarily stable or accessible. In my talk I will discuss a method we have developed of utilization of rotational echoes for intrinsic calibration of this alignment factor.

Echo spectroscopy is a central technique in various ranges of spectroscopy (NMR, vibrational, electronic and rotational), enabling researchers to distinguish dynamical dephasing from decoherence phenomena. In recent works we have published, we studied the unique response of the multi-level rotational system to two, time-delayed pulses that generate the echo signal (inset a in figure) \cite{1,2}. When observing the echo dependence on the pulses’ intensities and specifically on second pulse intensity, $I_2$, we have realized that it can be utilized for calibration of the degree of alignment. Since the echo signal has an oscillatory trend with the 2\textsuperscript{nd} pulse: $S_{echo} \propto \sin^2(I_2)$, there is a maximum point that can be found (for a specific delay between the pulses) both in simulation and in experiment, as shown in insets b,c in the following figure.

Afterwards, a simple simulation of alignment with the optimal pulse intensity (inset d) generates the exact value of the alignment factor. In this figure’s example, a 10.5\textmu J pulse is equivalent in simulation to a pulse of 4.7 arbitrary unit intensity, and the alignment factor achieved by it is of the value $\Delta \cos^2 \theta = 0.0272$.

This suggested calibration method allows bypass of complicated (and sometimes impossible) quantifications of experimental parameters and other obstacles that are inevitable in the experimental system such as collisional decoherence of the molecules and convolution of the pump and probe pulses. A manuscript regarding this intrinsic calibration method is now under review.
References:

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[2] D. Rosenberg, R. Damari, and S. Fleischer, Phys. Rev. Lett. 121, 234101 (2018).