***Laser Pulse Shaping and Coherent Control of Molecules**, Brijuni, Croatia, August 26th - 31st, 2007.*

Spinning molecules at will

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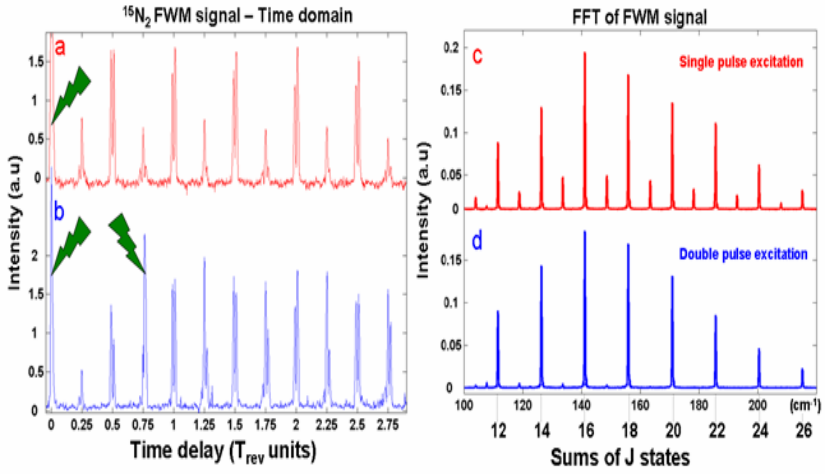
We experimentally demonstrate a new approach to selective excitation of close molecular species in mixtures[1,2]. Following excitation by an ultrashort, strong laser pulse, the molecules are repetitively aligned (depending on their rotational constants). In our double-pulse scheme, the first pulse excites both components in a binary mixture, and the second pulse de-excites one, while enhancing the excitation degree of the other. This enables the application of another strong, linearly polarized pulse for preferential dissociation/ionization of the selected molecular component; thereby enrichment of the sample becomes feasible. Since this process is nonresonant and does not require any special conditions like temperature etc. this approach is general and can be applied to most linear molecules. In our work we implemented this approach to molecular isotopes and spin isomers.

The case of molecular isotopes is based on slight difference in the masses of the molecular components, and supported by the periodic process of repetitive alignment one can distinguish between the isotopic components and selectively affect them. The case of spin isomers[3] is more complicated since there are no differences in the mechanical or electrical properties of the spin isomers to be selectively controlled. Here we utilize the symmetry and statistics of the specific molecular wavefunction and demonstrate selective excitation of Ortho/Para nitrogen using non resonant laser pulse at room temperature. Typical data is shown in the figure where we compare the molecular response to single (red, top) and double (blue, bottom) pulse in time (left) and frequency (right) domains. The absence of odd frequencies in the double pulse case, serve as a signature of selective excitation of a single isomeric component.



Time domain FWM signal from $^{15}N_2$ following **a)** a single pulse. **b)** 2 pulses delayed by ~¾Trev. **c)** and **d)** Fourier transforms of **a)** and **b)** respectively.

References:
[1] Sharly Fleischer, Ilya Sh. Averbukh and Yehiam Prior, Phys. Rev. A. **74**, 041403 (2006).
[2] Sharly Fleischer, Ilya Sh. Averbukh and Yehiam Prior, J. Mod. Opt. in press.
[3] Sharly Fleischer, Ilya Sh. Averbukh and Yehiam Prior, PRL, in press.