Dynamic field-free orientation of polar molecules by intense two-color femtosecond laser pulses

Sankar De[1*], Dipanwita Ray[1], Irina Znakovskaya[2], Fatima Anis[1], Nora G. Johnson[1], Irina Bocharova[1], Maia Magrakvelidze[1], B. D. Esry[1], C. L. Cocke[1], Matthias F. Kling[2] and Igor V. Litvinyuk[1†]

[1] J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506-2604, USA
[2] Max-Planck Institute of Quantum Optics, Hans-Kopfermann Strasse 1, D-85748 Garching, Germany

Synopsis: We present the first experimental observation of dynamic field-free orientation of a heteronuclear molecule (CO) induced by intense two color (800 and 400 nm) femtosecond laser pulses. We have used the two color pulse as pump to orient the molecules and a more intense 800 nm pulse as probe to measure the angular distributions. In addition to dynamic alignment seen in time dependence of $\langle\cos^2\theta\rangle$, we observe clear orientation in $\langle\cos\theta\rangle$ traces, which revives with the rotational period and can be reversed by changing the relative phase of the two colors. We studied the dependence of degree of orientation on the pump pulse intensity, and compared the results with theoretical calculations.

Combining a fundamental frequency of a laser and its second harmonic with a definite relative phase results in an asymmetric electric field and broken inversion symmetry. Such field-asymmetric laser pulses can generate macroscopic orientation in polar molecules. Here we present the first experimental implementation of this technique.

In our experiments we combined 50 fs 800 nm pulses from a Ti:Sapphire laser with their second harmonic field of 400 nm wavelength. The resulting field-asymmetric two-color pulses were focused onto a supersonic jet of CO molecules inside a velocity-map imaging (VMI) spectrometer. Rotationally excited CO molecules were interrogated at a varying time delay by a more intense single-color (800 nm) laser pulse. The probe pulse Coulomb exploded the molecules and the resulting C^{2+} fragments were momentum analyzed using VMI.

Figure 1 shows the experimental time traces of $\langle\cos^2\theta\rangle$ and $\langle\cos\theta\rangle$, where θ is the angle between the CO molecular axis (represented by the momentum direction of C^{2+}) and the laser polarization. $\langle\cos^2\theta\rangle$ represents the alignment of CO and the values of $\langle\cos\theta\rangle$ indicate a net macroscopic orientation of the molecular ensemble. The observed orientation exhibits rotational revivals after each rotational period. The results demonstrate field-free orientation of CO, which might be used to study angle-differential properties of this heteronuclear molecule, such as various ionization and scattering cross-sections.

We studied the dependence of the degree of orientation on the pump pulse peak intensity and identified an optimal pulse intensity for the maximum degree of orientation. We also compare our experimental results to a series of theoretical calculations and confirm that the orientation of CO is dominated by an asymmetry in the molecular hyper-polarizabilities rather than the molecules' permanent dipole moment.

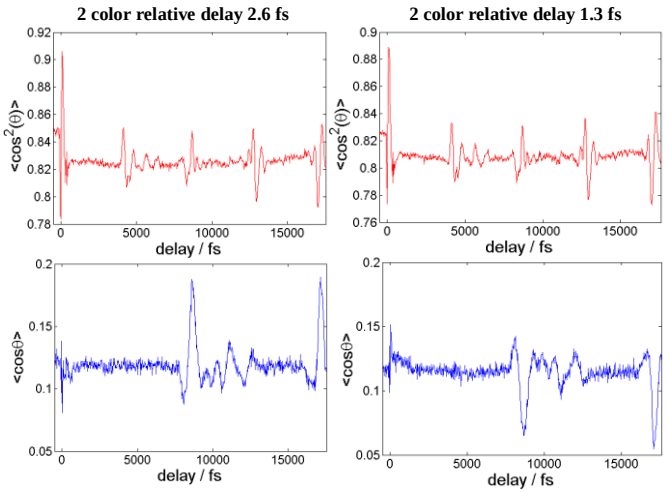

Fig. 1. Time dependence of molecular alignment ($\langle\cos^2\theta\rangle$ - upper row) and orientation ($\langle\cos\theta\rangle$ - lower row) for two different relative phases of 800 nm and 400 nm beams corresponding to opposite asymmetric fields.

This work was supported by Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

* E-mail: sankar@phys.ksu.edu

† E-mail: ivl@phys.ksu.edu