Controlling the orientation of the molecular angular momentum by shaping the polarization of a fs laser pulse

G. Karras[1], E. Hertz[1], M. Ndong[1], F. Billard[1], D. Sugny[1], B. Lavorel[1], and O. Faucher[1]

[1] *Laboratoire Interdisciplinaire Carnot de Bourgogne (ICB), UMR 6303 CNRS-Université de Bourgogne, 9 Av. A. Savary, BP 47 870, F-21078 DIJON Cedex, France*

Presenting Author: gabriel.karras@u-bourgogne.fr

Over the last twenty years, lasers have been offer great potentials for controlling the rotational dynamics of molecules. The wealth of the information accessed and the number of potential applications is far richer as a molecular ensemble looses its isotropic behavior. Laser induced field-free molecular alignment is a representative example for the above [1]. In this case, the molecules are aligned along and perpendicular to the laser polarization axis at well spaced fractions of the molecular rotational period even after the pulse has been switched off. In the current work, we present an original way for enriching the aforementioned control of molecular rotation by orienting the angular momentum or equivalently by inducing unidirectional molecular rotation. The investigation is conducted in N_2 at atmospheric pressure. The desired unidirectional rotation was embedded to the molecular targets after they were irradiated with a pulse of rotating linear polarization. The effect is quantified using the rotational Doppler effect [2]. In translational Doppler effect, a photon with momentum $p=h/\lambda$ reflected by a moving object with momentum $p=mu$ is frequency shifted by $\Delta v=2u/\lambda$. The fact that electromagnetic waves posses also spin angular momentum gives rise to the angular analog of the aforementioned effect. Thus, when a circularly polarized photon with spin angular momentum $\hbar$ interacts with a rotating body with angular velocity Ω, its frequency is shifted by $\Delta\omega=2\Omega$. The experimental results are reproduced quite well by numerical simulations and comparison between the different schemes proposed for inducing angular momentum orientation is discussed. The dependence of the rotational frequency shift on different experimental parameters, such as laser intensity, polarization state, and pulse duration is also presented and compared with theoretical findings.

References

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