Field-free unidirectional molecular rotation

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Abstract: By varying the polarization and delay between two ultrashort laser pulses, we control the plane, speed, and sense of molecular rotation. This control may be implemented to individual components within a molecular mixture.

The interest in laser induced field-free alignment and control of molecular rotation led to several applications which are based on the time-dependent alignment[1,2,3], where the ensemble averaged alignment factor $\langle \cos^2 \theta \rangle$ was the main observable. Preparation of molecules in a state of fast, unidirectional spinning is a much more complicated task that has been so far achieved via a sophisticated control over time-dependent field polarization[4]. Here we show that field-free, unidirectional molecular rotations can be induced by two properly polarized and time delayed ultrashort laser pulses.

An ultrashort, linearly polarized strong laser pulse, induces in linear molecules coherent molecular rotations which, due to quantum revivals, are manifested in a series of aligned ("cigar") and antialigned ("disk") angular distributions. (figure 1).

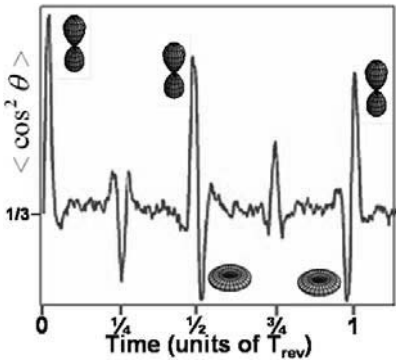

Figure 1: Alignment factor as a function of time measured in N_2 gas at room temperature. The two angular distributions ("Cigar" and "Disk" discussed above) are depicted near their corresponding peaks.

In order to induce unidirectional rotation we apply the first pulse polarized in z-direction, and let the molecular angular distribution evolve under field-free conditions towards an aligned "cigar" (or an anti-aligned "disk") state. At the time of alignment, the molecules are confined in a narrow cone around the polarization direction of the first pulse. At this moment, a second pulse, linearly polarized at a different angle, is applied, and thereby induces molecular rotation in the plane defined by the two polarization vectors.

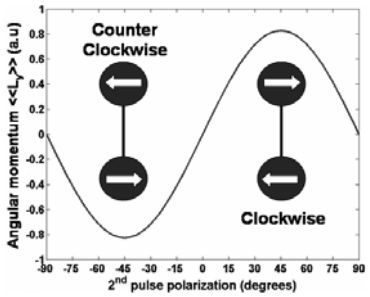

Figure 2: Mean angular momentum along the y-axis as a function of the polarization angle of the second pulse applied to the pre-aligned molecules ("Cigar" distribution) around the half rotational revival time (see fig. 1). The induced sense of rotation is depicted in the figure.

For a relative angle of +45 degrees between the two polarizations in the xz plane, the molecular ensemble rotates mostly clockwise (positive angular momentum), and the opposite is seen for the -45 degrees case. For a second pulse polarized parallel to the first one, no angular momentum along the y -axis is induced. In all cases, the mean angular momentum along the x -axis remains zero.

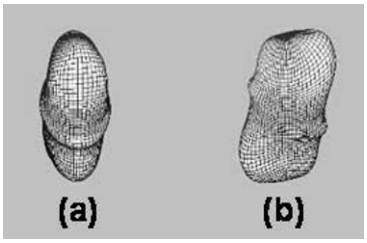

Figure 3: Calculation of $|\psi_{rot}|^2$ averaged of 1 T_{rev} of propagation. The anisotropic distribution is clearly seen from the comparison between the two view angles (90 degrees apart) (a) and (b).

As was mentioned above, molecules in this state are constrained to rotate in a plane, leading to highly anisotropic angular distribution. In figure 3 we observe the calculated $|\psi_{rot}|^2$ subjected to 2 pulses polarized 45 degrees at the right delay to induce unidirectional clockwise rotation, averaged over 1 revival time of propagation. This anisotropy in angular distribution is expected to yield anisotropic collisional cross section and corresponding anisotropic diffusion in space.

Up to this point we have considered the case where the second pulse, polarized at 45 degrees to the first pulse, is applied at the point of maximal alignment. If the second pulse is applied at the point of antialignment ("disk") its effect on the angular momentum gained by the molecular ensemble is opposite. This can be seen in figure 4, where the second pulse, polarized at $+45^0$ to the first one is applied at different delays. Since around half revival time (and full revival time) the molecules alternate between two dramatically different angular distributions, the effect of the second pulse is expected to depend on the exact delay.

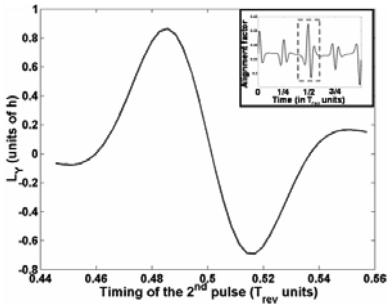

Figure 4: Calculated ensemble averaged angular momentum $<< L_y >>$ for different time delays around the half T_{rev} (shown in the inset). The second pulse is polarized 45^0 to the first one.

As can be seen in figure 4, by varying the time delay between the two pulses around the half revival time, one can induce clockwise or counterclockwise (positive or negative $<< L_y >>$ respectively) unidirectional rotation, depending on the angular distribution attained by the molecules at the time of application of the second cross polarized pulse. Based on this observation and on our previous works[3] we can induce selective unidirectional rotation in mixtures of molecular isotopes and nuclear spin isomers. Due to the rational ratio of the revival times of molecular isotopes, there is a specific time where one isotope completes an integer number of revival periods while the other completes and integer and a half of its own periods. At this time we find the two isotopic species at opposite angular distributions ("disk versus "cigar"), making them amenable for opposite induction of the sense of rotation (clockwise versus counterclockwise). The same can be done for nuclear spin isomers, based on their different behavior around ¼ and ¾ T_{rev}. The case of nuclear spin isomers is shown in figure 5.

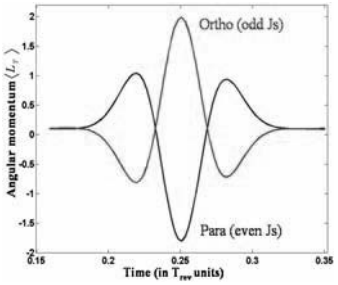

Figure 5: The mean angular momentum along the y-axis of two spin isomers of $^{15}N_2$ is plotted against the delay between the pulses for a second pulse polarized at +45 degrees to the first and applied at $\frac{1}{4}T_{rev}$.

When the second pulse is applied at specific times around $\frac{1}{4}$ and $\frac{3}{4}T_{rev}$ but is polarized at an angle to the first one, i.e. at +45 degrees, different senses of unidirectional rotation are clearly seen (Figure 5) for the individual nuclear spin species (Ortho, Para).

In conclusion, we show that two, time delayed, properly polarized ultrashort laser pulses can induce unidirectional rotational molecular motion in a gas of linear molecules. Based on the different rotational constants of molecular isotopes and the different symmetry properties of nuclear spin isomers and corresponding transient angular distributions, we can induce selective senses of rotation to the different species. Such opposite sense of rotation can be used for physical separation of the different species in the sample through scattering from a surface. Moreover, unidirectional rotation is expected to yield interesting optical features such as induced optical activity.

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