

Unidirectional Rotation of Molecules Measured by the Rotational Doppler Effect

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Abstract. A pair of linearly polarized pump pulses induce field-free unidirectional molecular rotation, which is detected by a delayed circularly polarized probe. The polarization and spectrum of the probe are modified by the interaction with the molecules, in accordance with the Rotational Doppler Effect.

Introduction

The interaction of light with rotating bodies leads to a frequency shift of the reemitted light known as the Rotational Doppler Shift, RDS [1]. In 2005 the optical RDS due to molecular rotation driven by a microwave field was observed [2]. In Ref. [3], a method of inducing field-free unidirectional molecular rotation (UDR) was suggested, which was then demonstrated in [4], and further extended in [5]. In the present paper, we use the RDS phenomenon to detect and quantify the field-free molecular UDR.

In its simplest form, RDS arises when a birefringent object rotating at frequency Ω interacts with circularly polarized (CP) light [1]. The transmitted light contains an admixture of a CP field of opposite handedness shifted in frequency by 2Ω . The shift can be red or blue, depending on the sense of the object's rotation.

Experimental setup

A schematic drawing of the experimental setup is shown in Figure 1. UDR of gas phase molecules contained in a cell is induced by two linearly polarized pump pulses, with the second one arriving at the time of maximal alignment of the molecules and polarized at 45° to the first pulse [3]. A CP probe is introduced at a variable time delay after the second pump pulse. A set of appropriately aligned polarizing analysers is used to transmit the generated light polarized oppositely to the incident probe. The measured observable is the Doppler-shifted spectrum of the transmitted beam.

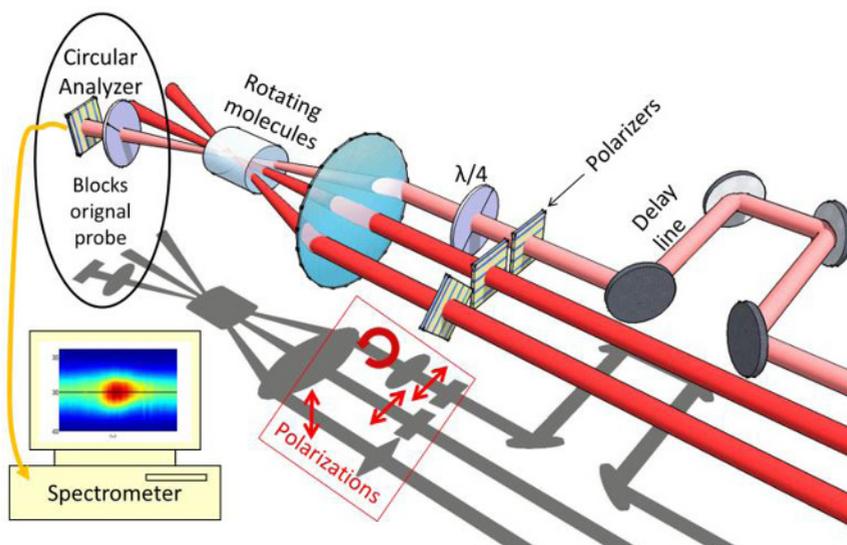


Fig. 1. Experimental setup. The probe pulse trajectory begins at the delay line and ends at the circular analyzer. The other two trajectories correspond to the linearly polarized pump pulses, which induce the unidirectional rotation in the gas cell. The Doppler-shifted wave passes through the circular analyzer and reaches the spectrometer.

When slowly rotating molecules are used, the RDS ($\sim 2\Omega$) is much smaller than the probe pulse bandwidth and it is hard to resolve the frequency shift. To overcome this difficulty, we introduced an interference filter to create a narrow spectral notch at a known position in the probe pulse spectrum [6]. This spectral notch enables the apparatus to resolve shifts of the order of the notch width, which is much smaller than the full spectral bandwidth of the probe.

Experimental Results

The results shown in Figure 2 were obtained for D_2 molecules at ambient conditions. The peak irradiance of the pump pulses was set to 100 TW/cm^2 , just below the ionization threshold. The FWHM of all three pulses was 40 fs. The delay between the two pumps was set equal to this value. Spectrograms of the probe beam were recorded in 350 fs time slots measured in the vicinity of the first molecular alignment. In the left panel of Figure 2, the probe polarization rotates in the opposite direction to that of the molecules, while on the right it rotates in the same direction, leading, as expected, to blue and red shifts, respectively. If the probe arrives when the molecules are not aligned, the molecular angular distribution is essentially isotropic. In this case there is no net birefringence, and no light is transmitted through the analyser.

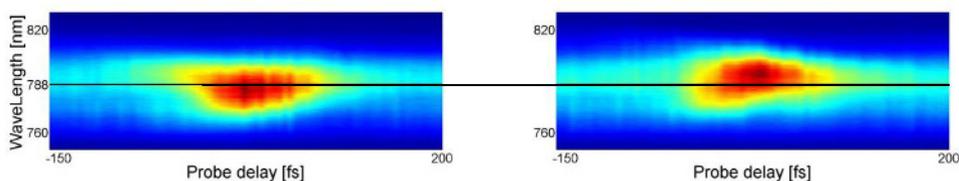


Fig. 2. D_2 spectrograms recorded at probe delays in the vicinity of the first alignment. The line at 788 nm indicates the maximum of the probe spectrum in absence of the pump pulses. In the left panel the UDR was in opposite sense to the original probe polarization, resulting in a blue shift, while in the right panel the UDR was in the same sense of the original probe polarization, resulting in a red shift.

A second set of experiments was performed using CO₂ molecules, under similar conditions. In this case a notch was introduced at the center of the probe's spectrum. The delay between the pumps was set to 200 fs, and the probe delay was scanned in the vicinity of the first half revival of the molecular alignment. The resulting spectrum of the transmitted light is shown in Figure 3. As expected, the Doppler shift of the transmitted light is manifested by a spectral dip below the notch wavelength line on the left panel (UDR in the opposite sense to the probe's CP) and above it on the right panel (UDR and CP in the same sense).

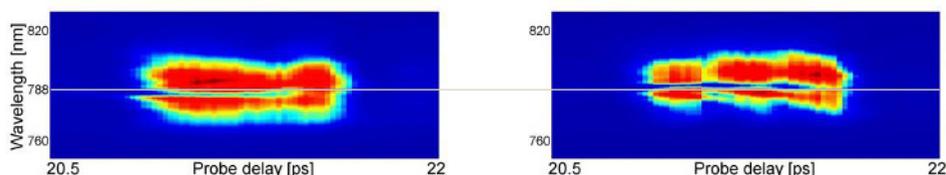


Fig 3: CO₂ Spectrograms around half revival time. The horizontal 788 nm line across the figure designates the original notch wavelength. In the left panel the UDR was in opposite sense to the original probe polarization, resulting in a blue shift, while in the right panel the UDR was in the same sense of the original probe polarization, resulting in a red shift.

Summary

Unidirectional rotation of gas phase molecules was produced by a pair of delayed laser pulses, linearly polarized at 45° with respect to each other, and detected with a circularly polarized probe. The UDR is manifested by an asymmetric spectral shift of the transmitted light produced by the Rotational Doppler Effect.

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References

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