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Chapter 1

Pulse shaping control of spatially aligned rotational wavepackets of N_2 and O_2

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1. Abstract

Control on the structure of the laser-induced field-free alignment revivals in diatomic molecules at room temperature is demonstrated. The control is achieved by temporally shaping the laser field, which modifies the excitation of the rotational Raman wave packets and their subsequent evolution.

2. Introduction

Laser-induced molecular alignment is a powerful tool to create spatially aligned distributions in an otherwise randomly oriented, gaseous sample of molecules [1]. In the regime called “impulsive alignment”, it is possible to obtain field-free molecular alignment at the times when the “revivals” occur. In those, diatomic molecules oscillate between the situation of alignment and *anti*alignment in tens to hundreds of femtoseconds - faster than their rotational period. Alignment refers to a situation where the internuclear axis is preferentially aligned along the direction of the polarization vector of the electric field of the laser. In *anti*alignment the internuclear axis is preferentially aligned on the plane perpendicular to the polarization vector.

The phenomenon of laser-induced molecular alignment is currently receiving a great deal of attention, with most recent progress concentrating on the effort to control the dynamics of the rotational wave packets by use of tailored laser fields or sequences of pulses. Despite a considerable body of theoretical work on the subject [2-4], experimental data are scarce. Two recent papers [5,6] have reported the enhancement of the degree of alignment by applying pairs of pulses. Renard et al. [7] applied a pulse shaping technique that allowed them to control the symmetry of the wave packet and thus the rephasing period.

In this contribution, we will show that it is possible to control the structure of the revivals using pulses with a modified temporal structure. We show control on the structure of the revivals in N_2 and O_2 at room temperature; for instance, enhancement of alignment at the expense of antialignment or vice versa. The results are discussed with reference to theoretical simulations that imply solving the time-dependent Schrödinger equation for a quantum rotor.

3. Simulations

In the non-resonant case for linearly polarized light interacting with a linear molecule, the effective Hamiltonian is given by [1,8,9]

$$H(t)=BJ^2-1/2\ \varepsilon^2(t)\ [(\alpha_{||}-\alpha_{\perp})\cos^2\theta+\alpha_{\perp}] \qquad (1)$$

where ε is the electric field, θ is the angle between the molecular axis and the field polarization vector and $\alpha_{||}$, $\alpha_{\perp}$ are the components of the polarizability tensor parallel and perpendicular to the molecular axis.

The signal stems from the birefringence of the aligned molecules, which induces some ellipticity on the initially linearly polarized probe pulse, and is proportional to

$$I_{signal} \propto [\ \langle\cos^2\theta(t)\rangle - 1/3 + C\ ]^2 \qquad (2)$$

where C is due to a residual elliptical polarization of the probe pulse. If C=0, the sign of $\langle\cos^2\theta(t)\rangle - 1/3$ is lost (homodyne detection). For $C >> \langle\cos^2\theta(t)\rangle - 1/3$, the sign can be retrieved from the data (heterodyne detection) [10].

The calculation is performed in the basis of field-free stationary rotational states $|\ J, M\ \rangle$ of the molecule. Only the matrix elements $\langle\ J, M\ |\cos^2\theta|\ J, M\ \rangle$ and $\langle\ J, M\ |\cos^2\theta|\ J \pm 2, M\ \rangle$ are non-zero. For a given start wave function $\psi_0 = |\ J, M\ \rangle$, M remains fixed and the final wave function after the interaction consists of a superposition of either only odd or even J states. With increasing laser intensity, more J states become significantly populated. Finite temperature is considered by a thermal ensemble of initial wave functions with a weight given by the Boltzmann factor and the multiplicity due to nuclear spin statistics.

4. Experimental details

The experimental approach follows the scheme of [10]. The output beam from an amplified 1 kHz Ti:sapphire laser system (785 nm center wavelength, 30 fs FWHM duration, 0.8 mJ per pulse) is split in two branches that act as a strong alignment beam and a weak probe beam. The temporal shape of the alignment beam is controlled with a home built pulse shaper [11]. Its polarization is then rotated by 45^o and the beam is focused onto the air sample with a 100 mm focal length lens. The alignment is probed by a weak-field polarization technique that is sensitive to the instantaneous birefringence that appears in the gas sample at a revival time. To that end the frequency-doubled probe beam is delayed with respect to the alignment beam using a motorized linear delay stage.

5. Results and discussion

By the application of the pulse shaping technique, control on the revival structures of N_2 and O_2 has been achieved. In this report, we will concentrate on the influence of third order dispersion (TOD) on the (half-) revival structure, where molecules switch from a situation of alignment to *anti*alignment. We can show, both in simulations and experimentally, that it is possible to enhance alignment at the expense of *anti*alignment or vice versa at the half revival by choosing the appropriate pulse shapes.

The results can be seen in Figure 1. A phase retardation in the shape of a cubic polynomial (TOD) was applied in the Fourier plane of the pulse shaper (left panels). The variation of the sign and magnitude of this cubic phase affected the temporal shape of the pulses (middle panels). Fig. 1(c) shows the structure of the first half revival in N_2 when a positive chirp of +20000 fs^3 was applied. This, in the time domain, corresponds to a pulse train with the highest intensity at the leading edge. Fig. 1(f) shows the result obtained for a pulse where a cubic phase of the same magnitude but opposite sign (reversed temporal structure) was applied. In both (c) and (f) the first peak corresponds to alignment and the second, to *anti*alignment.

A theoretical simulation was run for a room temperature sample of N_2 molecules for an electric field shaped as in the experiment. In the simulation, $(\langle\cos^2\theta(t)\rangle - 1/3)^2$ is evaluated and scaled to the signal. As can be seen in the Figure, agreement with the experimental results was obtained.

In conclusion, the control of one parameter (TOD) allowed us to promote either alignment or *anti*alignment at will in a half revival of a diatomic molecule. Even though the inverse problem is harder to tackle (obtaining a pulse shape that produces a given desired structure for a revival), the versatility of pulse shapers, allowing to finely control the wavepacket rephasing dynamics, was demonstrated and shows our increasing ability to manipulate the external degrees of freedom of molecules.

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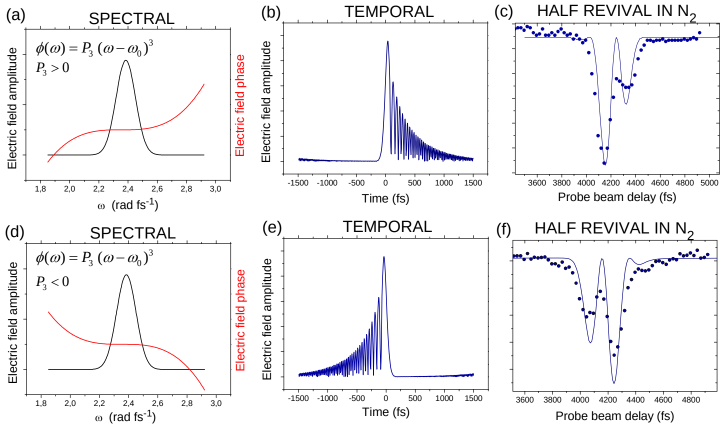

Figure 1. Control of the magnitude of alignment (*anti*alignment) in the first half revival of N_2 by applying pulses with a positive (negative) TOD. (a), (d) Amplitude and phase of the electric field in the spectral domain. (b), (e) Envelope of electric field in the time domain. (c), (f) The dots correspond to the experimental results: total homodyned signal detected around the time of a half revival in N_2. The lines correspond to the theoretical simulation.

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