J. At. Mol. Sci.
doi: 10.4208/jams.070510.073010a

Vol. **2**, No. 2, pp. 143-154
May 2011

# Controlling the molecular rotational wave packet accurately by femtosecond laser pulses

Zeng-Qiang Yang[a,b,*], Jian-Jun Zhang[a], and Yan-Wen Zhang[a]

[a] *Key Laboratory of Ecophysics and Department of Physics, Normal College, Shihezi University, Shihezi 832003, China*
[b] *Xinjiang Laboratory of Phase Transitions and Microstructures of Condensed Matters, Yili Normal University, Yining 835000, China*

Received 5 July 2010; Accepted (in revised version) 30 July 2010
Published online 28 February 2011

**Abstract.** We investigate the rotational wave packet modified by femtosecond laser pulses. The calculations are performed by solving numerically the full time-dependent Schrödinger equation for the $N_2$ molecule at finite rotational temperature. It is demonstrated that the rotational wave packet induced by the first laser pulse can be controlled exactly just by selecting the optimal time at which the second laser pulse is introduced. Whether the pulse duration of the two lasers is equal or not, the molecular alignment induced by the first laser pulse can be enhanced or degraded by precisely inserting the peak of the second laser pulse at the maximum or minimum position of the slope curve for the alignment parameter by the first laser. Furthermore, the already enhanced alignment by the two lasers can be enhanced or degraded by precisely inserting the peak of the third laser at the maximum or minimum position of the slope curve for the alignment by the two lasers. The already degraded alignment by the two lasers can be increased again from the isotropic distributed ensemble by precisely inserting the peak of the third laser at the peak position of the slope curve by the two lasers.

**PACS**: 33.80.Rv, 42.50.Hz, 33.90.+h

**Key words**: rotational wave packets, femtosecond laser pulses, time delay, the slope curve for the alignment

## 1 Introduction

When a laser pulse with both ultra short pulse width and ultrahigh laser intensity illuminates a molecule, the induced dipole moment will impart a torque. The molecule can then rotate to the laser polarization direction. As a result, molecules are partially aligned or oriented along the laser polarization direction [1–3], the field-free alignment and orientation of gas

*Corresponding author. *Email address:* zqyphys@yahoo.com.cn (Z. Q. Yang)

 

molecules have recently found many imperative applications. Molecular properties such as refractive index [4,5], ionization rate [6], high harmonic generation efficiency [7,8], diffraction [9,10], and molecular imaging [11] depend strongly on molecular alignment. Whatąŕs more, molecular alignment and orientation also play an important part in molecular electronic stereo dynamics [12–14].

The molecular alignment can be achieved adiabatically and nonadiabatically [2], adiabatic alignment is induced from field-free rotational states to pendular statesčň but it is lost at the end of the laser pulse. Nonadiabatic alignment of the molecules attracts much attention for being able to study the molecules in a field-free alignment condition. It is demonstrated that the excellent aligned molecules can be obtained by selecting the proper parameter of the aligning laser pulse [15–17] and reducing the rotational temperature [18,19]. However, the improvement is often limited by the saturation of molecular alignment and the maximum intensity that can be applied to the molecules without ionizing it. Spectral phase shaping method [20] provides a passive control schemes for improving the alignment degree beyond the limit. The alignment degree can be maximized by the accurate optimized laser pulse by evolutionary algorithm. However, this strategy is often time consuming and very hard to apply in experiment.

An intriguing and more efficient way for achieving an enhanced alignment degree is to employ a train of laser pulses [21]. Since the alignment is resulted from the in-phase overlapping of spherical harmonics in the time evolution of rotational wave packet, the odd and the even rotational states evolve out of phase around quarter revival times and in phase around full revival times. The alignment degree can be enhanced by applying the second laser pulse at the rising edge of the alignment parameter induced by the first laser pulse [22–24]. On the other hand, the rotational wave packet induced by the first laser pulse can be annihilated by the second laser pulse introduced at the half revival time [25,26]. What's more, a coherent molecular rotational wave packet can be manipulated selectively by strong femtosecond laser pulse [27,28].

Recently, a general method that relates the two-pulse alignment degree to their delay times has been developed, a new strategy to actively determine the optimized time delay between the two pulses with the same pulse duration is proposed according to the so called 'slope rule' [29]. In fact, the control for the rotational wave packet depends strongly on the pulse duration of two lasers. In this paper, the relation between the slope of alignment and the optimum time delay between the two pulses with both same pulse duration and different pulse duration is investigated, the enhancement and the annihilation of the molecular alignment are controlled exactly using two and three lasers with different peak intensity and pulse duration.

## 2 Theoretical method

We calculate the nonadiabatic alignment of linear molecules using the method presented in references [19,22]. For a molecule in a linearly polarized laser field, the Schrödinger equation