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Pulse train induced rotational excitation and orientation of a polar molecule

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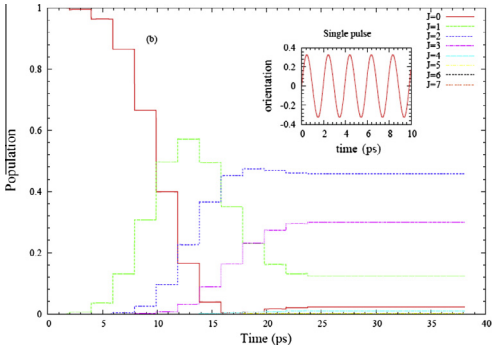
HIGHLIGHTS

- Rotational excitation and orientation of a polar molecule is studied in presence of pulse train of different shape envelopes.
- Both rotational excitation and orientation show strong dependence on shape of the pulse train.
- Rotational dynamics and orientation show strong dependence on the shape of the pulse train.

GRAPHICAL ABSTRACT

The figure shows the effect of thirteen pulse train (Gaussian shape) on the population dynamics of ground and excited rotational states of HBr molecule. Inset shows the orientation of the molecule due to single pulse.



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ABSTRACT

We investigate theoretically the rotational excitation and field free molecular orientation of polar HBr molecule, interacting with train of ultrashort laser pulses. By adjusting the number of pulses, pulse period and the intensity of the pulse, one can suppress a population while simultaneously enhancing the desired population in particular rotational state. We have used train of laser pulses of different shaped pulse envelopes. The dynamics and orientation of molecules in the presence of pulse train of different shapes is studied and explained.

Introduction

Population transfer to a desired coherent superposition of atomic and molecular states (i.e., a wave packet) has been a major goal during the past three decades and continues to be a challenge, for instance, for implementation of chemical and biological processes [1,2], for fast quantum information processing [3], and for nonlinear optics [4]. Since the invention of the mode-locked lasers, pulse trains have been used in the measurement of spectroscopy [5]. The generated single pulse in the train can be of attosecond duration [6] or of few-cycle oscillations [7]; the repetition frequency of the pulse train can be in the THz regime [8] and it can

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be tuned in the experiments [9]. In molecular systems, Warren and Zewail [10] showed that a sequence of nonresonant strong pulses could dramatically increase population inversions and multiphoton pumping. Diels and Besnainou [11] showed that matching the frequency peaks, associated with the train spectrum, with the successive transition frequencies of an anharmonic ladder of vibrational levels resulted in a nearly complete population transfer to the upper state. Vitanov and Knight [12] showed that the excitation of a nondecaying two-level atom by a train of weak pulses leads to the coherent accumulation of population in the excited state. Bouchene [13] has shown that the phase relation between pulses in a train is paramount in controlling dispersion in the propagation of trains with terahertz repetition rates. Cryan et al. [14] observed experimentally a high degree of alignment in nitrogen at STP resulting from impulsive-Raman excitation with a train of eight linearly polarized laser pulses. Gogyan et al. [15] have reported theoretically an efficient method for population transfer to desired superposition in multilevel systems using a train of pulses combined with weak controlled lasers. de Araujo [16] has reported a pump–dump scheme based on femtosecond pulse trains to transfer population between ground vibrational levels of the molecule via an excited stationary state. As far as the orientation of molecule [17] is concerned, system needs an asymmetry field that distinguishes up and down. As already known, a variety of methods have been proposed to break the field symmetry, including introductions of weak electrostatic or magnetic or both fields, in addition to an off-resonant pulsed laser field [18] and the conventional schemes of the coherent excitation with the laser fields of frequencies ω and 2ω [19] . Sugny et al. [20] and Dion et al. [21] has reported molecular orientation by using sequence of half cycle pulses.

Controlling interactions between light and matter has been a long standing goal in atomic and molecular physics. The work presented in this paper investigates the interaction between a train of laser pulses and matter. The aim of this paper is to build up a general picture of how a train of pulses interacts with an atomic ladder system in cold molecules. In a ladder system, an excitation is transferred to the higher energy levels like climbing a ladder. The interaction with a small number of pulses helps us to understand the physics of a ladder system's interaction with an arbitrary number of pulses in a train. Significant achievements in ultrafast laser technology have produced a new generation of optical short pulses whose duration (10 fs) is shorter than the atomic relaxation time and, indeed, the time scale of nuclear motion [22]. Shaping the spectral phase and amplitude of ultrafast laser pulses is already a commercially available technology for many research endeavors [23]. Generating a train of femtosecond laser pulses offers a new set of useful properties and has great potential for further development in many applications. For example, the interaction time of ultrafast lasers provides the capability to study the interaction with an atomic and molecular ladder system because these short pulses coherently interact with the system before radiative relaxation occurs. Now, it is possible to obtain shaped laser pulses by tailoring the ultrashort laser pulses in the frequency domain [24]. Since these laser pulses can be experimentally realized by the ultrafast pulse shaping method, we believe that the present technique is feasible and can be applied to various polar molecules. In this paper, we have investigated the effects of pulse propagation on the rotational population and orientation by train of ultrashort pulses of different shaped envelope which has not been studied yet.

Theory

The laser induced rotational excitation dynamics is studied for HBr molecule due to train of pulses of different pulse envelopes. The HBr molecule has small permanent dipole moment as compared to the traditional polar molecules (such as LiCl and KCl), molecule is treated within the rigid-rotor approximation (frozen internal vibrational motion), interacting with the fields. The Hamiltonian for this model is

$$H(t) = BJ^2 + V_E(t) \tag{1}$$

where B, is the rotational constant and J^2 is the squared angular momentum operator. The interaction potentials $V_E(t)$ between the molecule and the pulse train is defined as:

$$V_E(t) = \sum_{k=0}^{N} \Delta p_k \delta(t - t_k) \tag{2}$$

Here t_k, is the time at which the kth pulse is applied and Δp_k is the area under the kth pulse. Physically, Δp_k is the momentum transferred by the kth pulse to the molecule.

The laser field is defined as

$$E(t) = E_0 f(t) \sin(\omega t) \tag{3}$$

where E_0, is the electric field amplitude; and $f(t)$ is the sine square envelope defined as

$$f(t) = \sin^2(\pi t/t_p); \qquad 0 < t < t_p \tag{4}$$

$$f(t) = 1; \qquad \textit{for rectangular pulses} \tag{5}$$

In Eq. (3), ω, is the frequency of the applied laser field, which fits the main features of the experimental shape peak amplitude E_0 and duration t_p (full width at half maximum FWHM) of laser pulse. In order to ensure that the observed rotational excitation is induced by non-adiabatic rotational excitation (NAREX) process, a quantum dynamical calculation is performed to calculate the population by solving the time dependent Schrödinger equation (TDSE) in the presence of laser pulse [25]

$$\imath\hbar \frac{|\partial \psi_{rot}(t)\rangle}{\partial t} = (H_{rot} - \mu \cos\theta E(t))|\psi_{rot}(t)\rangle \tag{6}$$

The time dependent rotational wavepacket $|\psi_{rot}(t)\rangle$ is expanded in the terms of the rotational eigenstates $|JM\rangle$ of H_{rot} as

$$|\psi_{rot}^{J_iM_i}(t)\rangle = \sum_{J=0}^{J_{max}} \sum_{M=-J}^{J} C_{JM}^{J_iM_i}(t)|JM\rangle e^{-\imath E^J t/\hbar} \tag{7}$$

with the initial condition

$$|\psi_{rot}^{J_iM_i}(t=0)\rangle = |J_iM_i\rangle \tag{8}$$

inserting Eqs. (7) and (8) into Eq. (6), one obtains a set of coupled equations for the time-dependent expansion coefficients,

$$\imath\hbar \frac{dC_{JM}^{J_iM_i}(t)}{dt} = -\mu E(t) \sum_{J'=0}^{J'_{max}} \sum_{M'=-J'}^{J'} \langle JM|\cos(\theta)|J'M'\rangle \times e^{-\imath(E^{J'}-E^J)t/\hbar} C_{J'M'}^{J_iM_i}(t) \tag{9}$$

$$C_{JM}^{J_iM_i}(t=0) = \delta_{JJ_i}\delta_{MM_i} \tag{10}$$

The selection rules $J' = J \pm 1$ and $M' = M$ simplify the differential Eq. (9) as

$$\imath\hbar \frac{dC_{JM_i}^{J_iM_i}(t)}{dt} = -\mu E(t) \sum_{J'=0}^{J'_{max}} \langle JM_i|\cos(\theta)|J'M_i\rangle \times e^{-\imath(E^{J'}-E^J)t/\hbar} C_{J'M_i}^{J_iM_i}(t) \tag{11}$$

and

$$C_{JM}^{J_iM_i}(t) = 0 \qquad \textit{for} \quad M \neq M_i \tag{12}$$

The time-dependent rotational wavepacket (7) is thus