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Towards Using Molecular States as Qubits

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Abstract

Molecular systems are presented as possible qubit systems by exploring non-resonant molecular fragmentation of n-propyl benzene with femtosecond laser pulses as a model case. We show that such laser fragmentation process is dependent on the phase and polarization characteristics of the laser. The effect of the chirp and polarization of the femtosecond pulse when applied simultaneously is mutually independent of each other, which makes chirp and polarization as useful ‘logic’ implementing parameters for such molecular qubits.

Keywords

Molecular qubits; Multiphoton Ionization; frequency chirp; polarization

INTRODUCTION

Molecular states are fundamentally useful for exploring quantum computing due to their unique scalability advantages. Still, difficulties in possible manipulation of the molecular states and the fragility in their coherence lifetimes pose serious restrictions on their use as qubits. An extension in the coherence lifetime of molecular states can be achieved under laser field induced conditions when the laser pulses are themselves modified through pulse shaping techniques [1]. Isolation of molecular states can also be achieved experimentally in gas phase by the use of the supersonic molecular beam expansion techniques. Under such conditions, precise laser molecular interactions can generate molecular fragments that are not subject to immediate inter-molecular collision decoherence. Controllability of such quantum systems can be useful in defining their role as qubits in the rapidly growing field of molecular information processing [2]. Optical techniques are definite favorites owing to their promise of scalability and universality [3]. In fact, the use of optimally shaped pulses to guide the temporal evolution of a system and controlling its future has been an active field of research. Decades of research has resulted in the important non-trivial realization that all quantum mechanical processes are wave phenomena that are subject to constructive and destructive interferences. As such, an active manipulation of such quantum interferences is an essential step towards quantum control [4]. Since the vibrational processes responsible for such interferences are in several femtoseconds (10^{-15} s), femtosecond pulse shaping plays a significant role in the development of temporal control. Thus, coherent control uses optimally shaped pulses to guide the temporal evolution of a system wherein the resulting optimally shaped pulses are due to multi-parameter optimization, where each parameter is a ‘control knob’ for the experimentalist.

Recently, towards this direction, we demonstrated selective photo-fragmentation of n-propyl benzene under supersonic molecular beam conditions using chirp as a ‘control knob’ [5].

Similarly, other useful and widely used experimental control parameters are: pulse energy, amplitude, polarization, pulse width, inter-pulse separation, etc. Many such parameters are, however, interdependent. More importantly, some determine the feasibility of the concerned experiments. Thus, they are essential enabling conditions rather than control parameters and cannot be modulated independently nor can they be explored simultaneously. There is a continued effort in the search of independent ‘control knobs’ that can be simultaneously used in experiments. Our experimental results show that the control knobs, ‘polarization’ and ‘chirp’ of a femtosecond laser pulse are mutually exclusive and can act independent of each other, in the particular case of controlling multiphoton fragmentation of n-propyl benzene. Such mutually exclusive control knobs would be of immense interest in the implementation of quantum logic gates as mutually independent operations possible with the independent control knobs can be combined irrespectively to attain specific logic operations.

EXPERIMENTAL

Our experimental setup consists of a Ti:Sapphire multipass amplified laser system (Odin, Quantronix Inc.), which operates at 800 nm with 50 fs FWHM pulses at 1 kHz with energy of ~1 mJ. This is seeded by a home-made Ti:Sapphire oscillator (pumped by Verdi-5, Coherent Inc.) generating 27 fs transform-limited pulses centered at ~800 nm with 450 mW at a repetition rate of 94 MHz. The compressor of the Odin is suitably modified to produce chirped ultrafast laser pulses. When frequency components are arranged linearly within the laser pulse bandwidth, it results in linear chirping. As the spacing between the compressor gratings was increased relative to the optimum position for minimum pulse duration of 50 fs, we generate a negatively chirped pulse. Conversely, decreasing the inter-grating distance resulted in the positively chirped pulse. We measured the pulse durations for all such pulses using a home-made autocorrelator. The instantaneous phase variation in such femtosecond laser pulses at its center frequency (ω_0) can be expressed as:

$\varphi(\omega) = \varphi(\omega_0) + \frac{1}{1!} \frac{\partial \varphi}{\partial \omega} \big|_{\omega=\omega_0} (\omega - \omega_0) + \frac{1}{2!} \frac{\partial^2 \varphi}{\partial \omega^2} \big|_{\omega=\omega_0} (\omega - \omega_0)^2$, where the second derivative of spectral phase at the center frequency is defined as the linear chirp coefficient introduced by the compressor (*i.e.*, chirp parameter in the frequency domain) and can be calculated as:

$\beta = \tau_0 \sqrt{(\tau^2 - \tau_0^2)/4 \ln 2}$, where τ is the pulse duration of the chirped laser pulse and τ_0 is the chirp-free pulse (transform limited pulse) [5].

The laser polarization is varied from linear to circular by using a quarter-wave plate (QWPO-800-15-4-AS20) and from parallel to perpendicular by using a half-wave plate (QWPO-800-15-2-AS20). The amplified laser pulses are then focused with a lens (focal length = 50 cm) on the supersonically expanded molecular beam of n-propyl benzene that has been selected by the skimmer at the centre of a time-of-flight (TOF) chamber. Design of our molecular beam setup has been discussed in detail earlier [6].

The n-propyl benzene sample at room temperature was used without further purification (98%; Sigma Aldrich) and was seeded in Helium at 2 atm backing pressure. The mass spectra were recorded with a Wiley-McLaren type linear time-of-flight mass spectrometer (TOFMS). The fragment ions are detected using an 18 mm dual micro-channel plate (MCP) detector coupled to a 1 GHz digital oscilloscope (Lecroy 6100A). When the mass resolution of n-propyl benzene cation calculated from the time-axis data available from the TOFMS, it was found to be $t/2\Delta t \sim 1100$, where t is TOFMS flight data position in time and Δt is the FWHM of the highest peak, at the mass to charge ratio, $m/z=120$. We typically averaged our mass spectra over 500 laser shots.

RESULTS AND DISCUSSIONS

One of the simplest possible shaped pulses that have been effective in controlling molecular processes has been the use of simple linear chirped pulses. Recently, by using the same experimental setup as described here, we have shown that the phase characteristics of the laser field play a very important role in the fragmentation of n-propyl alcohol [6]. We now have extended these experiments to include polarization of the laser pulses as well. In Figure 1, the results of the individual versus the simultaneous effect of chirp and polarization on fragmentation of n-propyl alcohol are shown.

We find that though a change in polarization affects the overall fragmentation efficiency, the fragmentation pattern of n-propyl benzene molecule remains unaffected in contrast to the chirped case. Thus, for the coherently enhanced fragments ($C_5H_5^+$, $C_3H_3^+$) with negatively chirped pulses and C_6H_5 with positively chirped pulses, polarization effect is the same as compared to that in case of transform limited pulses, making these control parameters mutually exclusive.

The fragmentation channels are suppressed when we alter the polarization from linear to circular for both the unchirped and chirped cases (Figure 1b, Figure 1c). We record the mass spectra of n-propyl benzene using linearly chirped and unchirped laser pulses with varying angle of the quarter-wave plate. Average energy (~200 mW) of the laser pulses was kept constant using a variable ND-filter. We then compared the mass spectra by calculating their respective integrals under the peak. All the fragmentation channels which were enhanced with chirped pulses were suppressed to roughly the same extent as compared to the unchirped pulse case (Figure 2).

The fragmentation pattern is unchanged if we alter the polarization at any particular chirp of the laser pulse. The relative ratios of the fragment ions are essentially similar. We find that when we change the polarization from linearly polarized light (LP) to circularly polarized light (CP), molecular ion yield is suppressed by a factor of 0.52, as $\alpha = [(M+CP)/(M+LP)] = 0.52$. Up to a certain amount of chirp, this ratio, α , is almost constant. However, α increases when the laser pulses are highly chirped. This can be due to the lowering of laser peak intensity at high chirps. The lowering of the fragment ion yields for circularly polarized light can be attributed to the lowered probability of the rescattered electrons inducing dissociative ionization [7].

Such a mutually exclusive nature of the two control parameters make the polarization and chirp as attractive logic defining operations in quantum computing applications involving molecular ions. Molecular ions or the vibrational states of isolated molecules can effectively act as multiqubit [8] quantum building blocks, which can effectively manipulated selectively with independent operators for quantum computation. As of now, one of the foremost applications of such mutually exclusive control operations would be to implement a model quantum logic operation using the molecular ions under molecular beam environment. Possible scalability of such model quantum systems would be the most attractive feature of such quantum computing goals.

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FIGURE 1.

Effect of the chirp and polarization on fragmentation: (a) effect of only laser pulse chirp on the variation of different fragment ion yields, (b) effect of only laser polarization angle on the variation of different fragment ion yields (for reference, the corresponding linear component of laser intensity is also plotted), and (c) simultaneous effect of polarization angle at different amounts of chirp for laser pulse on variation of a specific ion yield fragment, $C_3H_3^+$ ion.

FIGURE 2.

Polarization dependence of different fragment ion yield at (a) Transform-limited condition (b) Negatively chirped pulse ($\beta=-8064 \text{ fs}^2$), and (c) Positively chirped pulse ($\beta=+6246 \text{ fs}^2$).