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Towards using molecular ions as qubits: Femtosecond control of molecular fragmentation with multiple knobs

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Abstract. Non-resonant molecular fragmentation of n-propyl benzene with femtosecond laser pulses is dependent on the phase and polarization characteristics of the laser. We find that 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 knobs.

Keywords. Multiphoton ionization; frequency chirp; polarization.

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1. Introduction

In the rapidly growing field of information processing, controllability of quantum systems is the key [1]. In the recent past, optical techniques have been often favoured because they are scalable and universal [2]. Using optimally shaped pulses to guide the temporal evolution of the system thereby controlling its future, has been an active field of research. Though non-trivial, decades of research has resulted in the realization that all quantum mechanical processes are wave phenomena that are subjected to constructive and destructive interferences, and as such their active manipulation is an essential step towards quantum control [3]. Femtosecond pulse shaping plays a significant role in developing temporal control because the vibrational processes responsible for such interferences are in several femtoseconds (10^{-15} s). Coherent control often uses optimally shaped pulses to guide the temporal evolution of a system. Such optimally shaped pulses are the result of multiparameter optimization where each parameter is a 'control knob' for the experimentalist.

Thus, using chirp as a 'control knob', we recently demonstrated selective photofragmentation of n-propyl benzene under supersonic molecular beam conditions [4]. Other useful and widely used experimental parameters are: pulse energy, amplitude, polarization, pulse width, interpulse separation, etc. Many of these parameters however are interdependent and determine the viability of the concerned

experiments. So they are essential enabling conditions rather than control parameters and can neither be modulated independently nor be explored simultaneously. Thus, 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 implementing quantum logic gates as mutually independent operations possible with the independent control knobs can be combined to attain specific logic operations.

2. Experimental set-up

Our experimental set-up consists of a home-made low power (∼500 mW) high repetition rate (94 MHz) Ti:sapphire oscillator laser (pumped by Verdi5, Coherent Inc.) producing 27 fs transform-limited pulses centred at ∼800 nm. These 20 fs laser pulses from the oscillator are amplified with a Ti:sapphire multipass amplifier (Odin, Quantronix Inc.), which operates at 800 nm with 50 fs pulse width at 1 kHz with an energy of ∼1 mJ. Chirped ultrafast laser pulses are generated from our suitably modified compressor for the amplified laser system. As the spacing between the compressor gratings is increased relative to the optimum position for a minimum pulse duration of 50 fs, we generate a negatively chirped pulse. Conversely, decreasing the intergrating distance results in the positively chirped pulse. 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 focussed with a lens (focal length = 50 cm) on a supersonically expanded molecular beam of n-propyl benzene at the centre of a time-of-flight chamber. The design of our molecular beam set-up is discussed in detail earlier [4].

The n-propyl benzene sample at room temperature is used without further purification (98%; Sigma Aldrich) and is seeded in helium at 2 atm backing pressure. The mass spectra are recorded with a Wiley-McLaren type linear time-of-flight mass spectrometer. The fragment ions are detected using an 18 mm dual microchannel plate (MCP) detector coupled to a 1 GHz digital oscilloscope (Lecroy 6100A). The mass resolution of n-propyl benzene cation is calculated and is found to be $t/2\Delta t$ ∼1100 at $m/z = 120$. Mass spectra are typically averaged over 500 laser shots.

3. Results and discussions

One of the simplest possible shaped pulses that have been effective in controlling molecular processes is the simple linear chirped pulse, which, for example, selectively alters the fragmentation pattern of n-propyl benzene molecule. On the other hand, with a change in polarization of the laser pulse, the fragmentation pattern of n-propyl benzene molecule remains unaffected though the overall fragmentation efficiency is affected. Thus, for the coherently enhanced fragments ($C_5H_5^+$, $C_3H_3^+$)

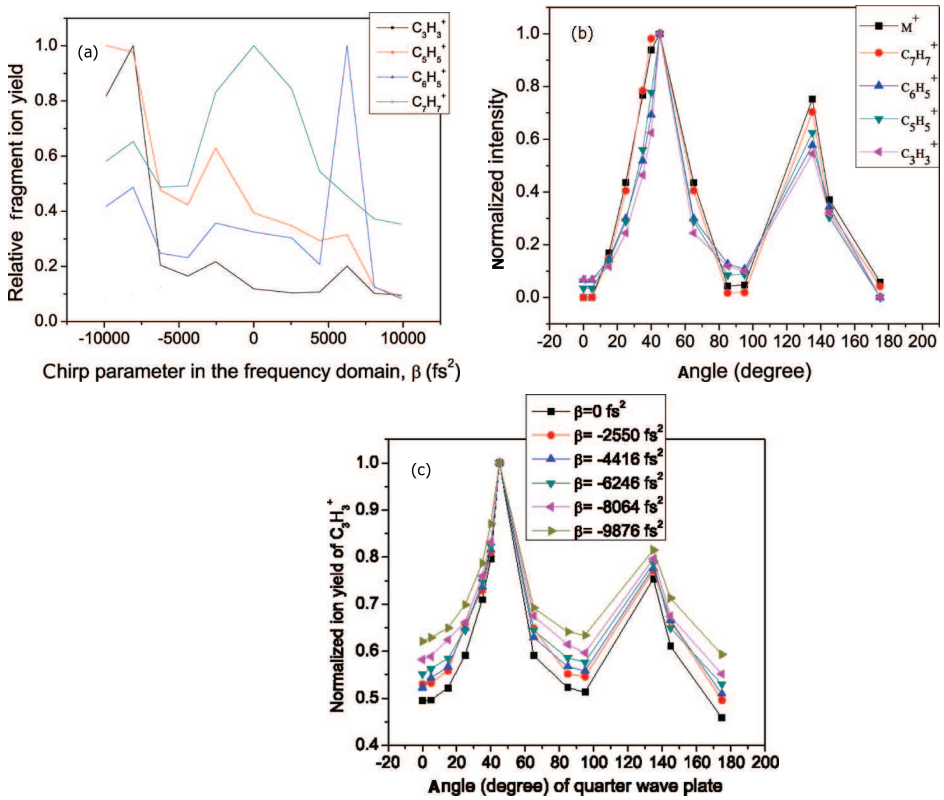

Figure 1. Individual vs. simultaneous affect of the chirp and polarization: (**a**) Variation of different fragment ion yields with chirped laser pulses, (**b**) variation of different fragment ion yields with polarization angle and (**c**) variation of $C_3H_3^+$ ion yield with polarization angle at different amounts of chirp for laser pulse.

with negatively chirped pulses and $C_6H_5^+$ with positively chirped pulses, polarization effect is the same compared to that in the transform-limited pulses, making these control parameters mutually exclusive.

For both unchirped and chirped pulses, the fragmentation channels are suppressed when we alter the polarization from linear to circular (figures 1b and 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 is kept constant using a variable ND-filter. We then compare the mass spectra by calculating their respective integrals under the peak. All the fragmentation channels which are enhanced with chirped pulses are suppressed to roughly the same extent when compared with the unchirped pulse (figure 2).

So the fragmentation pattern is unchanged if we alter the polarization at any particular chirp of the laser pulse. Relative ratios of the fragment ions are essentially similar. When we change the polarization from linearly polarized light (LP) to

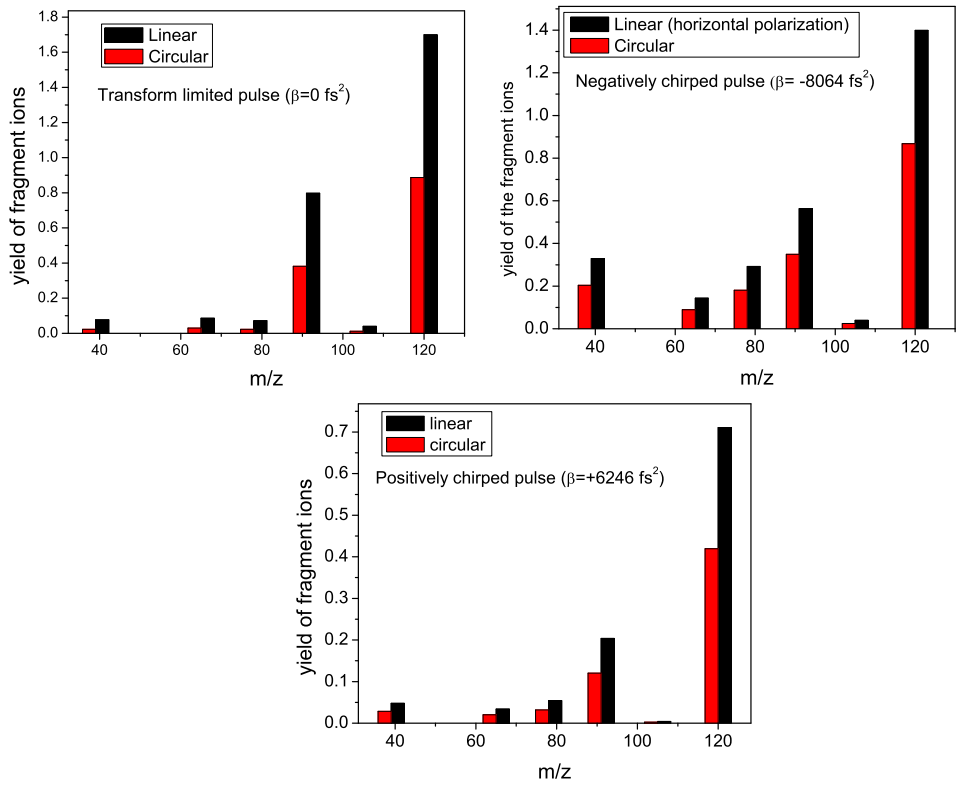

Figure 2. Polarization dependence of different fragment ion yields at different chirps.

circularly polarized light (CP), molecular ion yield is suppressed by a factor of 0.52, as $\alpha = [(M^+)_{\mathrm{CP}}/(M^+)_{\mathrm{LP}}] = 0.52$. Up to a certain amount of chirp, this ratio, α, is almost constant. But, when the laser pulses are highly chirped, α increases. 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 re-scattered electrons inducing dissociative ionization [5].

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 multiqudit [6] quantum building blocks, which can be effectively manipulated selectively with independent operators for quantum computation.

4. Future perspective

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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