Phase-Matched Generation of High Order Harmonic for Study of Molecular dynamics

L.V. Dao, K.B. Dinh, H. L. Vu and P. Hannaford
Centre for Quantum and Optical Science, Swinburne University of Technology, Melbourne, Australia
Author e-mail address: dvlap@swin.edu.au

Abstract: We present a pump-probe experiment based on the use of a second field to modulate the intensity and the spatial profile of the phase-matched high-order harmonics radiation for study of the dynamics of molecular gases.
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1. Introduction

The wave-packet created by the laser field plays a fundamental role in understanding the quantum picture and provides a bridge between the quantum picture and the classical concept of the trajectory of a particle. The motion of the wave-packet reflects the time evolution of a coherent superposition of the system. Useful information regarding the discrimination and visualization of the wave-packet dynamics can be obtained from time-resolved photoelectron imaging because of the sensitivity of the photoelectron angular distribution to the electronic symmetry. The high order harmonic generation (HHG) process is a coherent recombination of wave-packets and can be used to study wave-packet properties [1]. In conventional pump-probe spectroscopy a strong laser pulse is used to excite a molecule and the variation of the optical properties following the recovery or excitation of the molecules is probed by a second delayed pulse. In a HHG experiment the propagation and phase of the fundamental and harmonic fields depend on the optical properties of the medium and are reflected in the observed harmonic intensity through the so-called phase mismatch factor. A second off-axis delayed pulse can be used to perturb the propagation process or to change the harmonic phase which leads to a variation of the total harmonic intensity and the harmonic beam profile [2].

We report the use of a second, off-axis, long-pulse, laser beam to control the HHG process and show that the dynamics of the excited wave-packet can be studied through the modulation of the HHG intensity and the variation of the beam profile. The experimental approach proposed here shows that the properties and dynamics of the quasi-bound electron wave-packet can be studied and characterized.

2. Experimental Results and discussions

A 1 kHz multi-stage, multi-pass, chirped-pulse amplifier system producing up to 10 mJ pulses with a duration of 35 fs and centered at 810 nm is used in our experiment. The laser beam is split into two beams with an intensity ratio of first beam to second beam of about 4:1. A dispersion medium is added in the path of the second beam to give a pulse duration of ~ 80fs. The phase mismatch between harmonic and fundamental field $\Delta k = k_q - qk_0$ along the axis of the fundamental laser beam, which is reflected in the output of harmonic intensity, can be expressed as

$$\Delta k(z) = \textit{dispersion of the neutral medium} + \textit{the plasma dispersion} + (\textit{geometric term}) + (\textit{dipole phase term}) + (\textit{dispersion of highly excited medium}) \quad (1)$$

The dispersion of the neutral medium is negative, and the plasma dispersion is positive. The geometric term is dependent on the configuration of the HHG setup. Electrons which have tunneled under the interaction of the laser field at the maximum electric field of a laser cycle can return to the atom after one cycle with zero kinetic energy and oscillate with the laser frequency in the next cycle. These electrons can be captured into high excited states after a few optical cycles of the pulses. We include the fifth term due to the dispersion of the highly excited medium to take account of excitation effects.

When the second beam is absent the HHG radiation is obtained along the optical-axis of the first beam. A good beam profile and high HHG intensity indicate that the phase mismatch is small for a short quantum pathway [3]. When the second laser field is applied the harmonic intensity varies with the delay time between the two beams because the phase-mismatch Δk is altered. The modulation of the harmonic intensity is very strong for parallel polarizations for delay times up to -300 fs. When an 80 fs hyperbolic secant second pulse is applied the ration

between the first and second fields strength is ~ 0.05 and 0.01 at delay time of -200 fs and -300 fs, respectively. The second field is not change the field strength of the first field but it is enough to modify the trajectory of free electron. A high modulation frequency, which is the same as the laser frequency, and a low modulation frequency (20-25 THz) can be seen in Figs. 1. The electrons are most likely produced at the peak of the electric field but they return to the core with zero kinetic energy and trap into quasi-bound state. They are not contributed to HHG emission but their contributions to phase-matching cannot be neglect. The second (weak) pulse excites the quasi-bound and high excited wave-packet, which is likely a high excited Rydberg wave-packet, into the ionized continuum. The free electron wave-packets created by the first laser field, when they interact with the field before or after maximum, and the other free electron wave-packets created by the second laser field through the ionization of high –excited wave packet can interfere with each other. The ionization of the quasi-bound wave-packet and the interference of the two wave-packets lead to a change of the plasma dispersion. The interaction of the second laser field with the free electrons modifies the trajectory and re-scattering of the electron causes a change in the dipole phase. In the longitudinal direction or in the total harmonic intensity the contribution of the dipole phase to the phase-mismatch is smaller than that of the plasma dispersion. The Fig. 2 show the beam profile of H21 and H17 for oxygen at around -100 fs where the phase-mismatch is large and the harmonic intensity is low. Within one optical cycle we observe a shifting of the maximum with delay time away from the axis (for delay ~ - 108 fs) to an asymmetrical annular beam (around the optical axis of driving field) (for delay ~ - 109 fs) and then an abrupt change back to on-axis (for delay ~ - 110 fs). We can expect that the variation of the spatial profile depends on the atomic and molecular structures.

In conclusion, using a long off-axis pulse to modify the phase-matched harmonic intensity and the spatial distribution, the contribution of different electron trajectories can be revealed. We demonstrate that information on the quasi-bound state and the Rydberg state can be obtained from the high-order harmonic generation process. This experimental technique offers the possibility of studying the dynamics of the phase matching of an atom and the molecular structural dynamics with high time and spatial resolution.

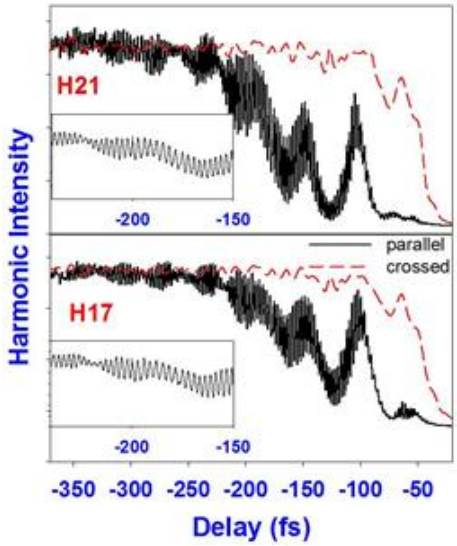

Fig. 1: Modulation of the intensity of harmonics H17 and H21. The inset shows a zoom of the region near the time delay of - 200 fs.

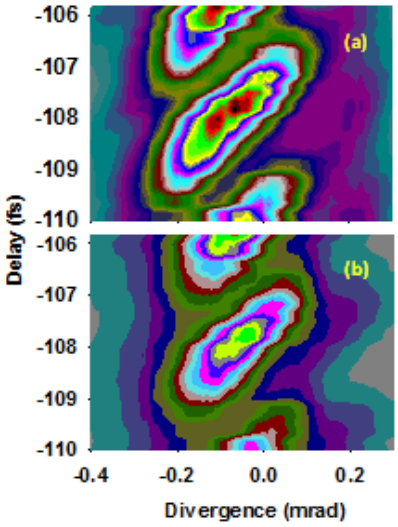

Fig. 2: the variation of beam profiles of H17 (a) and H21 (b) for oxygen at around -110 fs.

4. References

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