Correlated Terahertz and High Harmonic Generation from Aligned Molecules

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***Abstract*—We report the joint measurements of terahertz wave and high harmonic spectra generated from aligned nitrogen molecules. Our results show the yields of terahertz wave and high harmonics are sensitive to the alignment angle. This observation clarifies that electronic structure play an important role in the generation of terahertz and high harmonics from molecules.**

I. INTRODUCTION

WHEN laser beams are focused on atoms and molecules, wide spectral range of photons can be radiated from the source. In the region of high energy, high harmonic generation (HHG), covering tens to hundreds electron volts, emit within the attosecond timescale. In the low energy region, terahertz (THz) wave can also be generated. It's still an open question on the correlations between these two types of radiations in generating mechanism. In this letter, we report the joint measurements on pre-aligned molecules. We trace the rotation of the transiently aligned nitrogen and map the angular distribution of THz and high harmonics. The results suggest that the electronic dynamics play an important role in radiating photons from molecules in strong field.

II. RESULTS

The experimental setup consists of a 785nm, 25fs, 1.5mJ, 1kHz titanium: sapphire laser system, vacuum chambers for generation and detection HHG and THz. The output beam is split into three arms. The first arm is to impulsively align the molecules. The second arm is sent through a 30μm β-BBO to generate its second harmonic, whose group velocity dispersion is compensated by calcite and polarization is rotated into the same plane as the fundamental pulse. The high-harmonic spectra and the THz wave are recorded at different time delays around the rotational half-revival moment (~4.1ps of nitrogen), as shown in Fig.1. The modulation of THz is similar to the former results from the experiments conducted in air. A rotatable half wave plate is mounted on the first arm to adjust the alignment angle. These two pulses are focused ~0.2mm below and 2mm before the orifice of a continuous nozzle (0.2mm in diameter) generating a supersonic expansion. The third arm is used to detect the THz waveform by electron optics sampling (EOS) technology.

Then we keep the polarization of generation pulse fixed, and rotate the half wave plate to trace the angular THz and high harmonic signals. In Fig.2, we present the angular distribution of THz and harmonic order 21st from experiment and simulation at different time delays. The signal measured in the laboratory frame is a convolution of the single molecule response with the alignment distribution. By following the deconvolution process, we get the single molecule radiation. We also show the calculated ionization rates based on MO-ADK theory. The similar shapes imply that the angular ionization rate, which is closely associated to the electronic structure, determine the angular THz and high harmonic distribution.

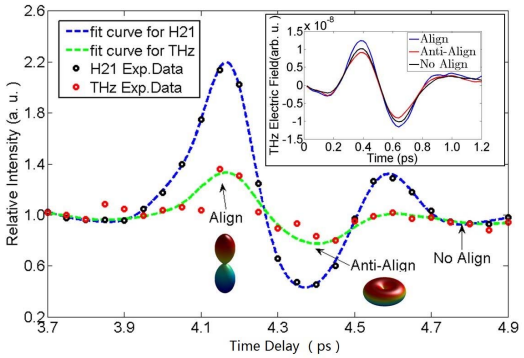

Fig. 1. The relative intensity of harmonic 21st (black circles) and THz (red circles) as a function of the delay between alignment and generation pulses, the blue and green dotted-curves are the simulation results. The moments of alignment, anti-alignment and no significant alignment have been pointed out on the figure, and the calculated wave packets are shown under the arrows. The inset presents the THz waveform detected at the three moments.

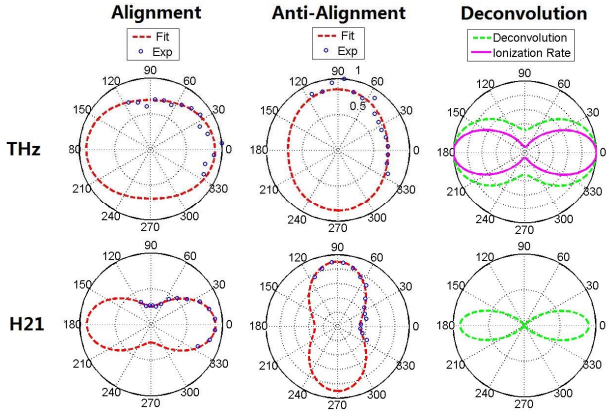

Fig. 2. Left and middle column show the experiments (blue circles) and calculations (red dotted curves) of the angular THz (up) and HHG (down) at alignment and anti-alignment moment. In the right column we demonstrate the deconvolution results (green dotted curves) of single molecular angular radiation and the calculated ionization rate (pink solid curves).

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