Harmonic enhancement in high-order harmonic generation from laser-ablated plume

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Abstract: In this study, we present some of our recent observations with high-order harmonic generation from Sn^+ using ~20 fsec mid-infrared driving laser pulses to understand the mechanism of single-order harmonic enhancement.

1. Introduction

High-order harmonics (HOHs) generated using low-density laser-ablated plume (LAP) have proven to be an excellent source of intense coherent extreme-ultraviolet (XUV) and soft X-ray radiation. The mechanism of high-order harmonic generation (HHG) can be explained by the three-step model, involving the tunnel ionization of the valence electron of an atom/ion upon interaction with a high-intensity laser pulse, acceleration away from the parent ion towards the continuum, and finally the emission of HOHs after recombination into the ground state upon reversal of the laser pulse electric field [1]. LAP-based HOH sources are capable of generating μJ of pulse energies in each harmonic order over a relatively broad spectral range, hence the possibility of intense attosecond XUV pulse generation [2, 3]. It has also been demonstrated that LAPs can emit intense harmonics over a relatively narrow spectral range through the phenomenon of resonant harmonic (RH) generation, which could be useful for applications in spectroscopy and imaging [4,5]. The RH generation mechanism is slightly different from the three-step model, but rather follows the four-step model [6]. In the four-step model, instead of direct recombination of the electron from the continuum into the ground state, the electron is scattered into an autoionizing state (AIS) present in the continuum, which is subsequently followed by radiative transition to the ground state emitting RH. Recently, the scope of LAPs as an XUV source has been further broadened by the demonstration of intense XUV vortices using the vortex driving laser beam, along with the 50% RH suppression observed under vortex laser beam irradiation, which motivates further efforts in understanding the laser-matter interaction during RH generation using the vortex driving laser [7].

More recently, using mid-infrared driving laser pulses, we have observed significant harmonic enhancement close to the ionization potential I_p = 18.87 eV of In^+, along with the previously well-known intense RH near 19.92 eV [8]. To explain this phenomenon, we hypothesized that the presence of several Rydberg states just below I_p could be responsible for the harmonic enhancement. In this paper, we show that harmonic enhancement below I_p depends on the LAP material. We study HHG from Sn^+ by using broadband ~20 fsec mid-infrared driving laser pulses, focusing on the behaviour of the harmonic close to I_p=14.6 eV of Sn^+ to study the response of Rydberg states present just below I_p.

2. Experimental Setup

We perform our experiments using the 100 Hz laser line of the Advanced Laser Light Source (ALLS). Fig. 1 shows a schematic diagram of the experimental setup. The uncompressed output of an amplified Ti:sapphire laser (800 nm central wavelength, ~210 psec pulse duration) is split into two beams using a 30:70 beamsplitter. To create the LAP, we focus the 30% beam portion onto a solid target at an intensity of ~10^{10} W cm^{-2}. The 70% beam portion is compressed down to 45 fsec and is used as an input to the optical parametric amplifier (OPA, TOPAS-800) for the frequency conversion.

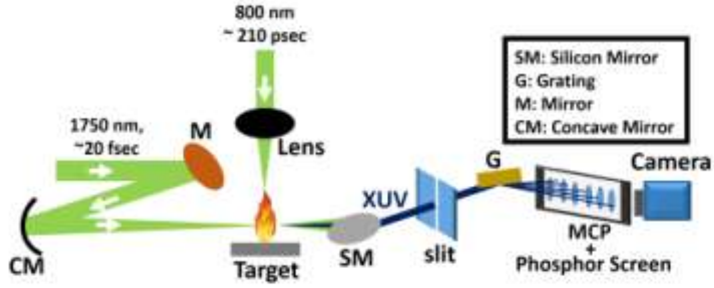

Fig.1. Schematic diagram of the setup for the generation of high-order harmonics from laser-ablated plumes

The tuneable output of the OPA is amplified parametrically and is sent to a gas-filled hollow-core fibre setup for further pulse compression. The details of the setup are given elsewhere [9]. For HHG, the laser pulses with adjusted pulse energy are focussed onto the LAP creating an intensity ~10^{14} W cm^{-2}. The generated XUV co-propagating with the laser beam is filtered by a silicon mirror installed at a Brewster angle, reflecting only the XUV. This XUV signal is frequency dispersed by an XUV spectrometer containing a fixed vertical slit, and a Hitachi 1200 lines/mm XUV grating. The XUV spectrum is then detected using a microchannel plate and a phosphor screen, which is recorded with a 16-bit CMOS camera (model PCO-edge, PCO AG, Germany).

3. Results

We use driving laser pulses with a central wavelength of 1750 nm and ~20 fsec pulse duration to generate the harmonics from Sn^+. The harmonic spectrum obtained is shown in Fig.2. The maximum photon energy generated is ~27.5 eV. Following the four-step model, a strong RH near 26.3 eV, which is 37-photon resonant with the 1750 nm central wavelength of the driving laser pulses is generated.

In our previous investigations using In^+ as a source of HHG, we had observed harmonic enhancement close to I_p, and we hypothesized the role of various Rydberg states just below I_p in harmonic enhancement. The 20 fsec laser pulses used in our experiment with Sn^+ generate broadband harmonics resulting in a continuum of harmonics up to 21 eV. This continuum covers the range of energies near I_p=14.6 eV of Sn^+. However, as can be seen in Fig.2, we do not observe any harmonic enhancement in the energy range close to I_p. We also performed similar experiments using tuneable driving laser wavelengths of OPA (data not shown in this paper), and no harmonic enhancement near I_p was observed. In the presentation, we will describe the mechanism by which some materials show below-I_p harmonic enhancement, while others do not, along with the detailed experimental results.

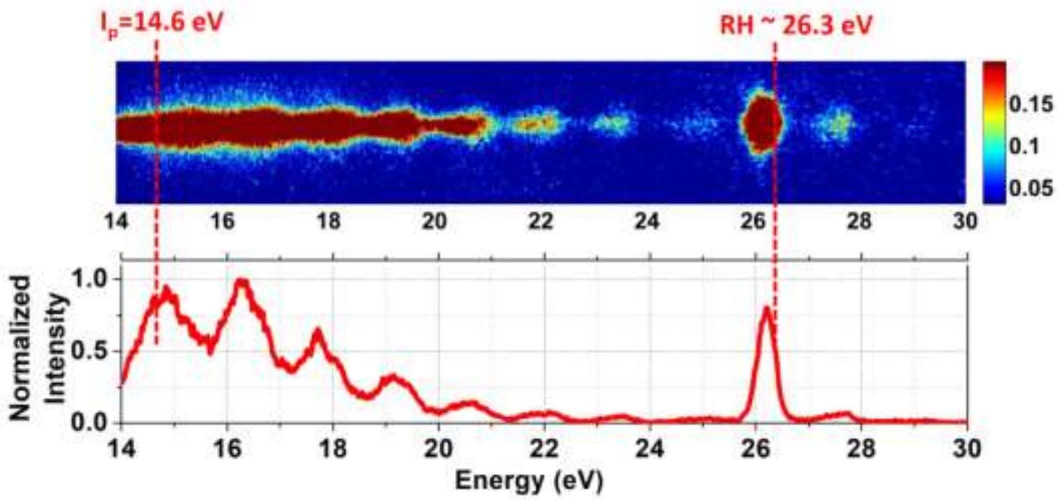

Fig.2. High-order harmonic spectrum from Sn^+

4. Conclusions

We studied HHG from Sn^+ using mid-infrared ~ 20 fsec driving laser pulses. A continuum of harmonics up to 21 eV and a maximum of 27.5 eV harmonic is generated. While the well-known RH near 26.3 eV is generated, no harmonic enhancement is observed near I_p=14.6 eV, demonstrating that only the presence of Rydberg states just below I_p does not result in the harmonic enhancement.

5. References

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