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High-order harmonic generation below the ionization potential using laser-ablated Indium plume

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***Abstract*—We study high-order harmonic generation from laser ablated Indium plumes below the ionization potential at various driving laser wavelengths. Apart from intense harmonic from the autoionizing state, we also observe enhanced harmonic generation below the ionization potential.**

***Keywords*—high-order harmonics, resonant harmonics, below-threshold harmonics.**

I. Introduction

High-order harmonic generation (HHG) is a highly nonlinear process involving the interaction of extremely intense ultra-short femtosecond laser pulses with gases or low-density plasmas [1, 2]. HHG has been found to be a very efficient and excellent table-top source of highly coherent extreme ultraviolet and soft X-ray radiation. The HHG process even has the potential to generate attosecond optical pulses by using femtosecond laser pulses as the driving laser [3]. The HHG process is very well explained by the three-step model [4]. In this model, an electron is tunnel ionized upon interaction with an ultrashort laser pulse **(step-1)**, then accelerated in the continuum under the influence of the laser electric field **(step-2)**, and finally recombines into the initial ground state upon reversal of the laser field to emit high-order harmonics **(step-3)**.

Interestingly, intensity enhancement of a single harmonic by one or two orders of magnitude has been observed in many laser ablated plasma plumes [2]. This happens when the energy of a harmonic order matches with a strong radiative transition with very high oscillator strength (*gf* value) and the enhanced harmonic is called resonant harmonic (RH). This has led to the modification of the three-step model, resulting in the introduction of the four-step model involving trapping of tunnel ionized electron by autoionizing state (AIS) of an atom, and finally radiative transition to the ground state [5].

In this paper, we study HHG from indium plasma plumes at various driving laser wavelengths. We demonstrate that apart from the enhanced RH, the harmonic intensity is significantly high close to the ionization potential (I_p). This enhancement is observed for a range of driving laser wavelengths and appears even when the RH disappears completely at off-resonant driving laser wavelengths.

II. Experimental Scheme

We first used Ti-Sapphire laser operating at 800 nm wavelength. The uncompressed 210 psec pulse is focused to create the plasma at the target surface. The compressed 50 fsec laser pulse is then focused inside the plasma plume to generate the XUV radiation. The silicon mirror installed at a brewster angle eliminates the 800 nm radiation, while reflecting the XUV. The XUV signal is spectrally dispersed by a flat-field grating (Hitachi, 1200 lines/mm). This signal is detected by an MCP and phosphor screen and is recorded by a camera (16 bit CMOS, PCO-edge). See Fig. 1.

The I_p of In^+ is 18.87 eV, which is 12-photon resonant with the 800 nm driving field. With the 800 nm driving field, a typical high harmonic spectrum will contain a series of odd harmonics. Therefore, to explore harmonics close to I_p, we need to generate even harmonics. For this, the second

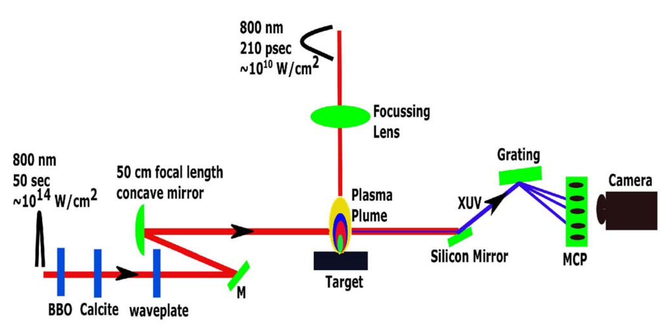

Fig. 1: Schematic diagram for the two-color HHG setup

harmonic field (400 nm) is added to the 800 nm using a BBO crystal. Calcite crystal rotation compensates the temporal delay, and the polarization of the 800 nm and 400 nm field is made parallel by using a half wave-plate. This wave-plate rotates only the 800 nm polarization by 90 degrees and makes it parallel to the 400 nm field. This setup is called two-color HHG setup. We also study the generation of high harmonics with an Optical parametric amplifier (OPA: TOPAS-800) with single-color HHG setup. Details of this laser source are given elsewhere [6]. The OPA central wavelength can be tuned from 1.2 μm to 2.2 μm.

III. Results and Discussion

Fig. 2 shows the harmonic spectrum recorded with Indium plasma plume by using two-color HHG setup at 800 nm driving field. This spectrum contains even as well as odd harmonics. In laser-ablated Indium plume, In+ is the active species responsible for HHG. The AIS ($4d^9 5s^2 5p$) exists at 19.92 eV, which is above the ground state ($4d^{10} 5s^2$) of In+. This is a very strong transition with a high *gf* value of 1.11, and is 13-photon resonant with 800 nm driving field [7]. As can be seen clearly in Fig. 2, there is an intensity enhancement at 13H. However, surprisingly, the 12H, which is close to I_p and is one photon below the AIS is also seen to be very intense as compared to the neighboring harmonics. The 12H is 24 times more intense than 8H, 10 times more intense than 11H and 7 times more intense than 14H and 15H.

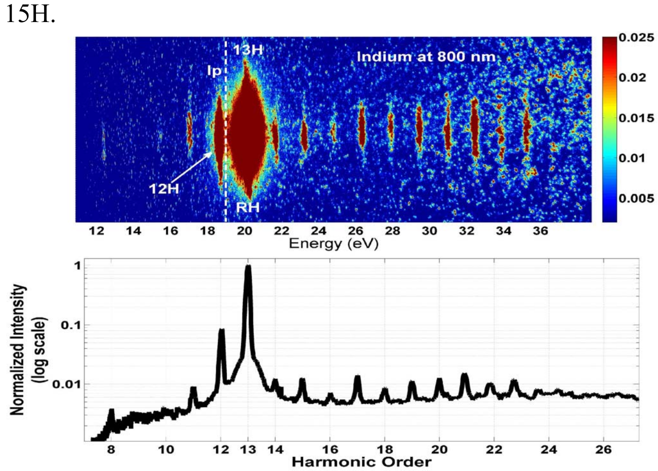

Fig. 2: Harmonic spectrum recorded with Indium plasma plume with two-color driving field

To further explore this enhancement behavior, we study the generation of high harmonics with an OPA. This gives us the ability to tune the driving laser wavelength. Fig. 3 shows various spectra recorded for different driving laser wavelengths. At 1.78 μm driving wavelength, the RH is close to 29 photon resonance and the I_p is exactly 27-photon resonant. We clearly see the enhanced 27H with 1.78 μm driving field. An interesting behavior is seen when we tune the driving laser wavelength around the 1.78 μm. Consider for example the 1.745 μm driving wavelength. At this wavelength, RH energy is 28-photon resonant (which is absent with our single-color setup) and the I_p is also off-resonant. As expected, we don't see harmonic generated around RH. However, we still see harmonic signal near I_p. Similar effect is seen at 1.735 μm and 1.88 μm. At both of these wavelengths, the resonant harmonic is missing but strong harmonic close to I_p is still clearly seen.

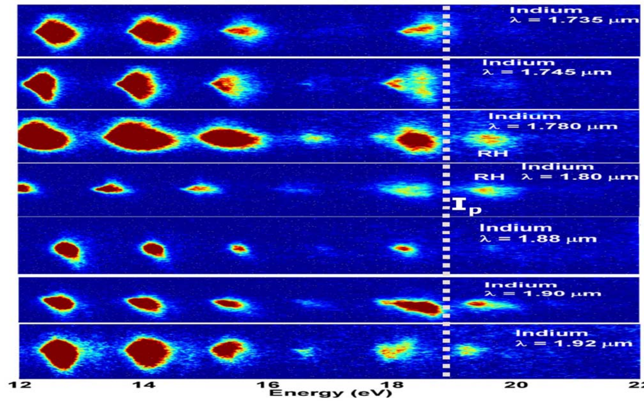

Fig. 3: Harmonic spectrum recorded with Indium plasma plume at various driving wavelengths obtained using an OPA.

The presence of several Rydberg states could be responsible for the generation of enhanced harmonics below I_p [8].

IV. Conclusion

In conclusion, we have demonstrated the intensity enhancement of harmonic below I_p in Indium. This enhancement has been seen both at 800 nm driving field obtained using Ti-Sapphire laser as well as with a wide range of driving wavelengths obtained using an OPA. The below I_p enhancement is seen even when RH completely disappears at certain driving wavelengths due to off-resonance behavior. We attribute this enhancement behavior to the involvement of several Rydberg states present below I_p in singly charged Indium ion. Further study and comparison of these findings with the simulation results obtained with different models will be useful for us in extending our understanding of physics and the electronic trajectories involved in the generation of harmonics below I_p.

V. Acknowledgement

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