High-order harmonics generation and attosecond dynamics in laser-produced plasma

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High-order harmonic generation (HHG) has emerged as an excellent tabletop source of coherent extreme ultraviolet (XUV) and soft X-ray radiation, providing a compact means to access high photon energies in the laboratory. Since high-order harmonics are intrinsically generated in attosecond bursts due to the nonlinear interaction between intense laser fields and matter, HHG is also opening a new domain of attosecond science, enabling the generation of ultrashort pulses for probing ultrafast electron dynamics and fundamental processes in matter.

The HHG process in most conventional nonlinear media, including atomic gases, is well explained by the semi-classical three-step model, which involves tunnel ionization of an electron, acceleration of the free electron in the laser field, and its recollision with the parent ion, resulting in high-energy photon emission. However, for certain media such as laser ablation plumes (LAP), the intensity of a specific harmonic order can be enhanced by one to two orders of magnitude relative to neighbouring harmonics produced by the conventional three-step process. This enhancement is observed when the harmonic photon energy is near an autoionizing resonance of the nonlinear medium, leading to resonance-enhanced harmonic generation.

In this presentation, I will first review prior studies on HHG from laser-ablation plumes, summarizing key experimental findings and the mechanisms of resonance enhancement. I will then present our recent work on intense, highly monochromatic high-order harmonics generated from gallium plasma, demonstrating its capability as a narrowband XUV source for ultrafast studies.

Our experiments were performed at the Advanced Laser Light Source (ALLS) facility of the Institut National de la Recherche Scientifique (INRS), which provides high-repetition-rate ultrafast laser systems. We used the second harmonic of a Ti:sapphire laser operating at 400 nm to pump a gallium laser ablation plume, creating conditions for efficient HHG. The measured harmonic spectrum from gallium shows strong resonance enhancement at specific harmonic orders. Gallium ions (Ga^+) with an ionization potential of 20.52 eV exhibit a strong resonance in the photoionization cross-section, peaking at 21.9 eV. This resonance corresponds to the transition from the ground state $3d^{10}4s^2\ ^1S_0$ to the excited autoionizing state $3d^94s^24p\ ^3P_1$. The transition from this autoionizing state back to the ground state is resonant with the absorption of seven photons from the 400 nm laser, corresponding to the 7th harmonic (7H).

In our measurements, we observed a highly intense resonant harmonic (RH) at this 7th harmonic order. We define the enhancement ratio (ER) as $ER = 2I_q/(I_{q-2} + I_{q+2})$, where I_q is the integrated intensity at the q^{th} harmonic order. Using the 400 nm pump laser, we observed an ER of 714, which is significantly higher than the previously reported ER of around 100 from indium plasma using similar LAP-HHG techniques. This makes the ER of 714 the highest value reported so far using the LAP technique, establishing gallium plasma as a source for generating intense, narrowband resonant harmonics in HHG.
The full-width at half-maximum (FWHM) bandwidth of the generated 21.9 eV resonant harmonic is 0.28 eV, narrow enough to support a Fourier-transform-limited pulse duration of around 6.5 femtoseconds, which is advantageous for time-resolved studies requiring spectral purity.

Recent studies show that resonance harmonics reveal various phenomena occurring on sub-femtosecond and attosecond timescales, providing insights into transient electron dynamics and autoionization processes. In this talk, I will discuss several such phenomena that have been investigated through our experimental and theoretical studies, demonstrating how resonance-enhanced HHG serves as both a bright, narrowband XUV source and a sensitive probe for exploring ultrafast processes in complex media.

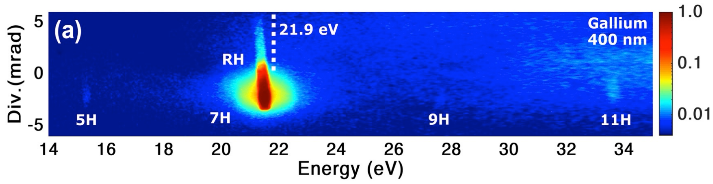

Fig. 1. HHG spectra generated from Ga^+, using 400 nm laser wavelength. The laser intensity used to generate harmonic spectrum at 400 nm is 1.6×10^{14} W cm^{-2}. [1]

References
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[2] R. A. Ganeev *et al.*, Opt Lett **31**, 1699 (2006).