High order harmonic generation in plasma plume of *in-situ* laser produced silver nanoparticles

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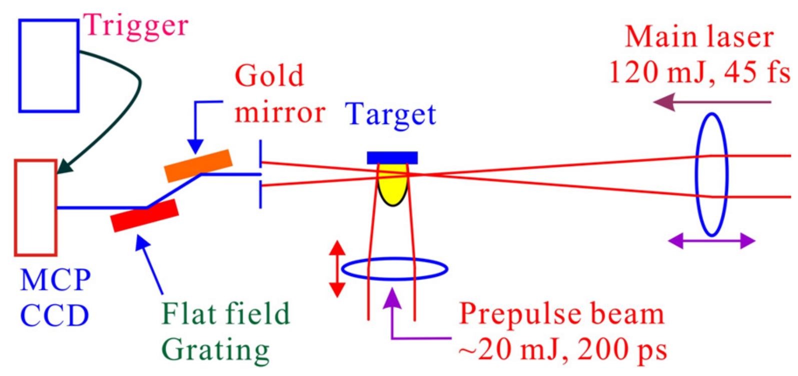
Abstract

- The results of the experimental study of high order harmonic generation (HHG) from the interaction of 45 fs Ti:sapphire laser pulses with plasma plumes of *in-situ* produced Ag nanoparticles are presented.
- The nanoparticles were generated by the interaction of 200 ps, 20 mJ laser pulses with bulk silver target at an intensity of ~1×10^{13} W/cm^2.
- AFM analysis was carried out to confirm the presence of nanoparticles in plasma plume.
- The spectral characteristics of the HHG from *in-situ* produced nanoparticles are compared with the HHG from mono-particle plasma plumes and with the HHG from preformed nanoparticle containing plasma plumes.
- The cutoff harmonic order generated using the *in-situ* silver nanoparticles is at the 21st harmonic order.

Motivation

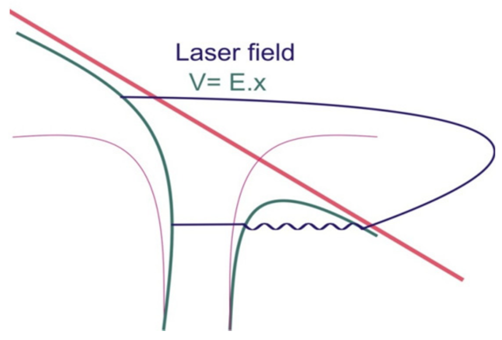
- ➢ High order harmonic generation (HHG) : A good source of ultra-short, tunable, coherent, XUV radiation.
- ➢ Can be used in various pump-probe, spectroscopic applications.
- ➢ Femtosecond laser interaction with gases/under-dense plasmas → odd harmonics of laser in the XUV region.
- ➢ For practical applications : highly efficient and stable harmonics
- ➢ HHG from nanoparticle targets : high efficiency, less stability
- ➢ HHG from mono-particle plasma : less efficiency, high stability
- ➢ HHG from in-situ produced plasma : may contain best of two

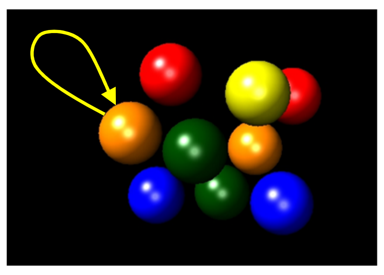
Experimental setup



- ❑ HHG : Interaction of fs laser pulse with preformed plume
- ❑ XUV spectrograph: Grazing incidence mirror to focus harmonics vertically, Flat field grating to disperse the spectrum, MCP+CCD assembly to detects the harmonics.
- ❑ Plasma plumes : generated by focussing a 200 ps laser at I ~ 10^{13} W/cm^2, contains Ag nanoparticles predominantly.
- ❑ Delay required for expansion of plume ~ 60 ns
- ❑ Femtosecond laser parameters : Energy – 120 mJ, Pulse duration – 45 fs, intensity : 10^{14-15} W/cm^2
- ❑ Intensity of laser can be changed by adjusting the positions of the lens.
- ❑ At this prepulse intensity: higher excitation of plasma plume → triggered MCP (15 ns) to cut stray light

Mechanism of HHG



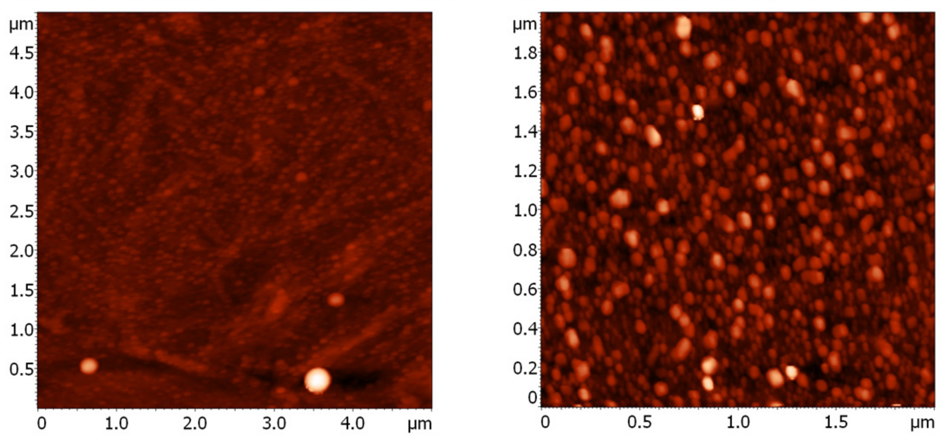


- Laser field → distorted atomic potential → tunneling of bound electron → acceleration in laser field → may return to parent atom/ion → recombine → HHG
- Propagation of harmonics inside plume → odd harmonic generation due to constructive interference
- Photon energy (max) : E_{max}= IP+3.17U_P(el. quiver energy)
- U_P ↑ as I_L ↑, but for HHG $I_L \leq I_{Sat}$ where ionization effects destroy phase-relationships
- Nanoparticles : mean free path << electron excursion length → HHG from surface only,
- Nanoparticles : separation between constituents << electron excursion length → electron can recombine with neighbouring ion → increased recombination cross-section → increased HHG efficiency
- Nanoparticles : field of neighbouring atoms distort atomic potential & increased collision heating → reduction ionization threshold → decrease in saturation intensity → decrease in HHG cutoff

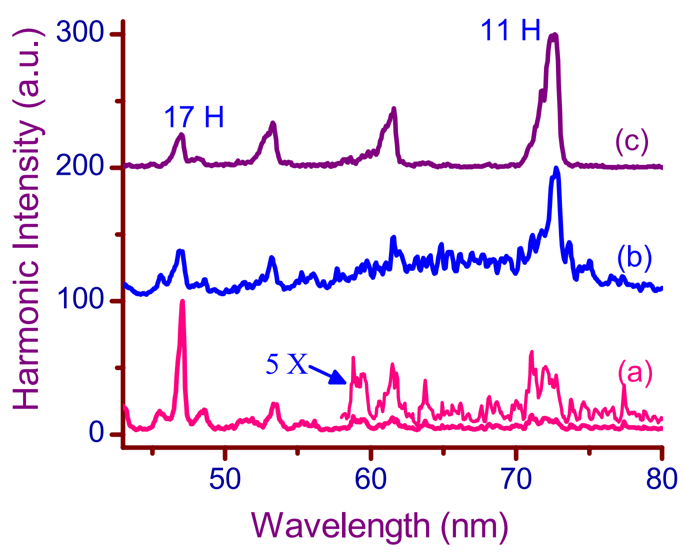
Nanoparticles : Increased efficiency, Decreased cutoff

Results and discussion

Formation of nanoparticles



- ❑ Left figure : AFM image of the deposited sample at intensity 10^{10} W/cm^2, **no nanoparticles**
- ❑ Right figure : AFM image of the deposited sample at intensity 10^{13} W/cm^2 : Nanoparticles with Average particle size 30 nm.



- ❖ Plot (a) shows HHG from silver monomer, (c) shows HHG from Ag nanoparticles (preformed)
- ❖ Plot (b) shows the HHG at prepulse intensity 10^{13} W/cm^2. Comparison of Plot (b) with (a) and (c) indicates that HHG is from insitu produced nanoparticles.

Discussion

- At prepulse intensity 10^9-10^{10} W/cm^2 : Plume predominantly consists of neutral and singly charged particles & Free electron density inside plume is low → good phase matching conditions, HHG from mono-particle plasma
- At prepulse intensity 10^{13} W/cm^2 : Plume predominantly consists of nanoparticles and highly exited plasma → Free electron density inside plume is high → bad phase matching conditions → no HHG from mono-particle plasma but higher nonlinear response of nanoparticle overcomes the phase-mismatch conditions → harmonics from nanoparticles produced *in-situ*.

Conclusions

- ➢ First results on HHG from *in-situ* produced nanoparticles are presented.
- ➢ Although the intensity of the HHG is less but it could be increased by optimizing the nanoparticle generation conditions.
- ➢ This will be helpful in building an efficient and stable source of coherent XUV radiation.

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