L.5: On high harmonic generation from nanoparticle containing plasmas

Generation of coherent XUV radiation through the high order harmonic generation (HHG) process during the interaction of ultra-short laser pulses with gaseous media and laser ablated plasma plumes, is an area of active research. The Laser Plasma Division of RRCAT had earlier demonstrated the use of nanoparticle (NP) containing plasma plumes as a medium of efficient HHG. It was shown that the HHG yield in the lower harmonic orders (9^{th} - 29^{th}) is enhanced by a factor of 10-200 in NP plasma plumes compared to the corresponding mono-atomic plasma plumes. It was also observed that HHG from NPs is relatively insensitive to the material and size of the NP. Till now there was no plausible mechanism in literature to explain the HHG process from NPs. One such mechanism has now been proposed by us.

As per our proposed mechanism, due to the small size and high density of the NPs, the electronic structure inside a metal NP gets distorted and become quasi-continuous. The separation (δ) between two successive levels (the Kubo gap) can be written as $\delta=4E_f/3N$, where E_f is Fermi energy and N is total number of atoms in the NP. Usually $\delta < 1$ meV. In this case, the electrons inside the NP can move freely as electron gas. This effect is weaker for the atoms at the surface of the NP and one can assume that the valence electrons in these atoms are still attached to parent nuclei. This results in a disappearance of the material specific properties in HHG from NPs.

The usual process of HHG is understood in terms of the so-called three step model in which harmonics are generated when 1) the valence electron gets tunnelled out from an atom placed in laser field, 2) it gets accelerated in laser field and 3) it then radiatively recombines with the parent nuclei to emit radiation. Due to cyclic nature of this process at laser frequency, the emitted radiation is harmonic of the incident laser radiation. Now, due to the overlap of the wave functions of the constituent atoms in a NP, the whole NP acts as a single coherent body and the electron from one atom in the NP, which recombines with a neighboring atom can still emit the radiation without losing the coherent properties, which increases the effective recombination cross-section. As a NP starts getting ionized, its temperature increases, thereby resulting in a reduction in the de-Broglie wavelength of the electrons inside the NP. This in turn, results in a separation of the atomic wave-functions. The distance (Δx) between two adjacent atoms for our test case (10 nm, silver) NPs can be calculated to be ~0.3 nm, which remains comparable to the de-Broglie wavelength for the temperatures up to 20 eV. Hence the assumption of the NP as single coherent entity will remain valid if the temperature remains under a certain limit.

A numerical code has also been developed to simulate the evolution of the ionization state and the temperature inside the NP and the results are shown in Fig.L.5.1 and Fig.L.5.2.

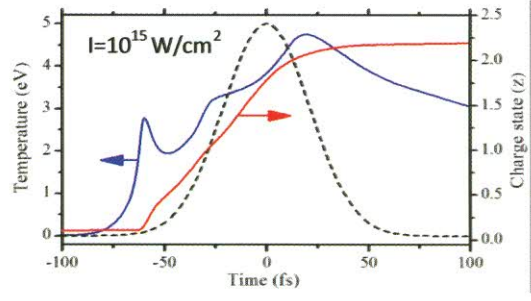

Fig. L.5.1: The temporal evolution of the nanoparticle temperature and average charge state

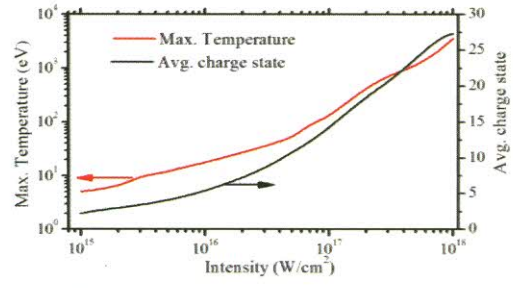

Fig. L.5.2: The variation of the nanoparticle temperature and average charge state with laser intensity

It has been shown that only the electrons which are ionized from the surface of NP can contribute towards the HHG. The enhancement in HHG efficiency due to NP effect has been estimated and the results are shown in Fig. L.5.3 for various harmonic orders. For higher orders, the electron should recombine with higher kinetic energy, which reduces its de-Broglie wavelength. Hence, the enhancement factor decreases rapidly with increasing harmonic order. The same trend was also observed experimentally and reported earlier. For more details of the present work, please refer to H. Singhal et al., J. App. Phys. 115, 033104 (2014).

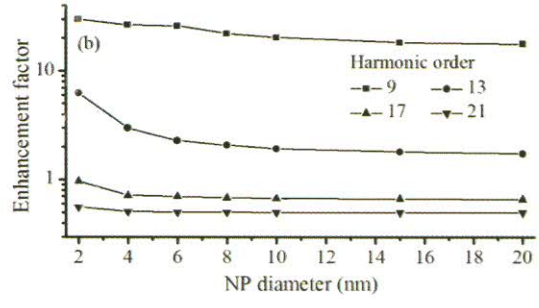

Fig. L.5.3: The variation of the enhancement factor with nanoparticle diameter for various harmonic orders

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