Intense Femtosecond Laser-induced X-ray/Ultrasound Emission From Aqueous Solutions in Air

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1. Introduction

Intense femtosecond laser interaction with solutions induces various interesting nonlinear phenomena such as laser ablation, plasma formation, and so on. Such phenomena are sensitively dependent on experimental conditions and accompanied with X-ray/ultrasound emission, which can be useful sources for various analytical studies in laboratories. Experimental conditions on the laser excitations and the sample preparations are key issues not only for such practical developments of X-ray/ultrasound sources but also for detailed mechanism discussions on the intense laser-matter interactions, which will be introduced in this talk.

2. Experiments

Pulse shape/width-controlled femtosecond laser pulses (800 nm, 35 fs at shortest, 1 kHz, linearly-pol., Mantis, Legend HE USP, Coherent) were tightly focused by an objective lens (10x) into circulated solution flow in a glass tube for ultrasound emission [1-3] or by an off-axis parabolic mirror ($f \sim 50$ mm) in air onto a film-like solution flow set in an automatic position controller for X-ray emission [4-7]. Ultrasound emission intensity was measured by transducers at the frequency of 5, 10, and 25 MHz and X-ray emission intensity was measured in air by a Geiger counter. A solution sample was distilled water or gold nano-colloidal suspensions with different particle sizes and shapes. Double pulse excitations were carried out with optical delay lines for the time range 0-15 ns.

3. Results and Discussions

3-1. Ultrasound

Photoacoustic signal intensity usually increases as the sound frequency decreases from 5 to 10 and 25 MHz and the intensity with gold nano-rods (12x35 nm) is higher than that with nano-spheres (20 nm ϕ) [1]. This may be because of the effective absorption at the wavelength of 800 nm due to surface plasmon absorption. Chirped-pulse irradiations up to 800 fs induces higher photoacoustic signal intensity though white light emission is most effective with the shortest pulse irradiation [2]. Further enhancements of the photoacoustic signal intensities were observed with double-pulse irradiations and the intensity ratio for the main and the pre-pulses at 80:20 shows the highest enhancement with the time delay at 15 ns [3]. This enhancement is ascribed to laser ablation phenomena induced by the pre-pulse irradiation.

3-2. X-ray

Appropriate positions of the solution surface relative to the laser focus for the highest X-ray intensity, which was automatically measured by the position controller with a LabView code, shift upstream of the laser incidence as the laser power increases [4]. This indicates that such X-ray emission in air is inevitably related to the air plasma formation. Size dependence of gold nano-spheres on X-ray intensity shows that the highest intensity was obtained with 40 nm in diameter, which FDTD calculations reproduce the efficiency of scattering and absorption [5]. Similarly to the case of the ultrasound emission, chirped-pulse irradiation induces higher X-ray emission, which indicates the complex mechanisms in the single laser pulse from the ionization and the electron acceleration to X-ray emission though inner-shell excitation and bremsstrahlung [5]. Double-pulse irradiations with the time delay up to 15 ns show another X-ray intensity peak in the upstream or the downstream positions when the time delay is in the picosecond range or in the nanosecond range, respectively [6, 7]. These peaks are also ascribed to the phenomena induced by the pre-pulse irradiation such as pre-plasma formation and laser ablation associated with shockwave expansion and droplet formation.

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