Time Resolved, Phase-Matched Harmonic Generation from Exploding Noble Gas Clusters

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Abstract. Third-harmonic generation from noble-gas clusters by ultrafast probe pulses is more sharply-enhanced than linear absorption following heating by an ultrafast pump pulse, in good agreement with simulations of cluster expansion and collective electron dynamics with phase matching consideration.

INTRODUCTION

Recent experiments [1,2] have shown that the linear absorption and refractive index of van der Waals bonded clusters are resonantly enhanced at time delays $\Delta t_{crit} \sim 1$ ps following excitation by a short pump pulse, as illustrated by our time-resolved linear absorption data for Ar clusters in Fig. 1a. This resonance occurs as the natural plasma frequency ω_p of the expanding clusters drops through the probe laser frequency ω, and thus occurs earlier for smaller clusters and/or higher pump intensity. The resonance is quite broad, spreading over ~ 1 ps, prompting Milchberg et al. [3] to propose that a moving surface critical density layer, rather than a bulk collective resonance [4], dominates the linear optics of the expanding clusters.

EXPERIMENT

Here we report additional pump-probe experiments that measure the third-order nonlinear susceptibility of the plasma inside the gas jet medium which is composed of expanding clusters. Argon clusters were heated by an intense 400nm, 200fs pump pulse, then allowed to freely expand. An 800nm, 80 fs probe beam with parallel polarization then probed the micro-plasma at a small angle ($\sim 1^0$) with respect to the pump propagation direction and generated a $3\,\omega_{probe}$ signal ($\lambda = 266$ nm). When pump and probe overlap ($\Delta t \sim 0$), $3\,\omega_{probe}$ signals can be generated by sum-frequency generation (SFG) or 4-wave mixing (4WM) involving both pump and probe, in directions determined by phase-matching (Fig. 1b). In addition, the probe alone generates 3rd-harmonics (THG) along its propagation direction at any Δt. Figure 1c shows results in which the detector collected both the SFG at $\Delta t = 0$ and resonantly-enhanced THG at time delay $\Delta t_{crit} \sim 200$-300 fs. For given pump intensity, the THG

resonance occurs earlier than the linear absorption maximum (Fig. 1a), as expected for ω_p resonance with the higher frequency $3\,\omega$. In addition, and quite interestingly, it is much narrower than the linear absorption resonance, suggesting the dominance of collective electron dynamics with phase matching consideration.

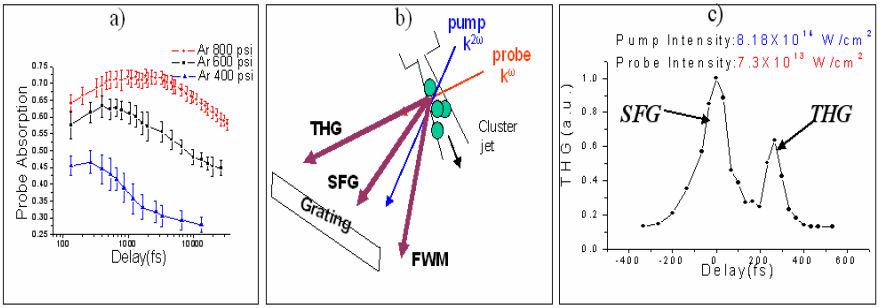

FIGURE 1. Pump-probe experiments and results from argon clusters. **a)** Time-resolved linear absorption of probe pulse (800nm, 80 fs) following excitation of Ar clusters by pump pulse (200 fs, 400nm). Higher backing pressure corresponds to larger clusters (800 psi: R ~ 21 nm; 600 psi: R ~ 17nm; 400 psi: R ~ 13 nm). **b)** Non-collinear pump-probe scheme used for results in c), showing signals at $\lambda_{probe}/3$ resulting from Third Harmonic Generation (THG) by the probe, Sum Frequency Generation (SFG) and Four Wave Mixing (FWM) by the pump and the probe at zero delay, which are separated spatially by momentum conservation. **c)** Delayed third harmonic resonant enhancement from Argon clusters (800 psi., average cluster radius~ 21nm), and SFG peak at zero delay. THG resonance occurs earlier, and lasts a much shorter time, than corresponding linear absorption resonance shown in a).

The simple model calculation based on references [4, 5] shows that the coherence length (L_{coh}) for THG, normally very small (<μm), peaks sharply as clusters expand and the 800nm and 266nm light become phase matched. During this peak, which has a very short time duration (~ several fs), L_{coh} becomes comparable to the collinear interaction length (~1mm), inducing a greatly enhanced conversion efficiency of the THG (Fig. 2). In order to verify the phase matching theory, we made the overlap length ($L_{overlap}$) small (~30μm) by using a large angle (~20^0) pump (800nm)-probe (800nm) geometry (Fig. 3a).Compared with the nearly collinear case(~1^0), the THG resonance enhancement for the large angle case is greatly suppressed because the interaction length is quite small (Fig. 3b). The delayed resonance is also quite sensitive to the position of the pump-probe focus within the cluster jet, which is also consistent with phase-matching sensitivity.

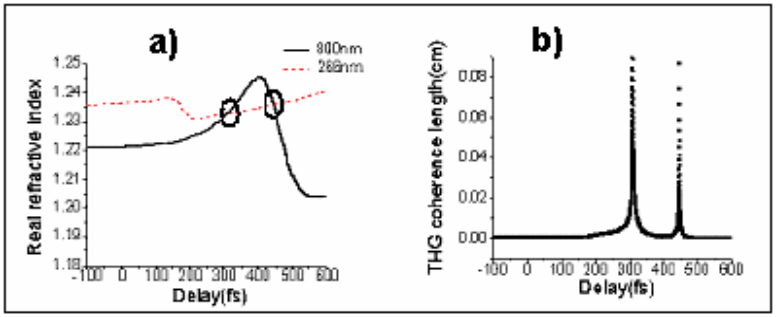

FIGURE 2. Model calculation for phase matching when the Argon clustered jet (30nm radius) is pumped by the 200fs, 400nm beam with the peak intensity $5x10^{14}$ W/cm^2. **a)** The phase matching resonance between 800nm and THG takes place when the real refractive indices are same. **b)** If phased matched, the THG coherence length grows enormously during a short time period.

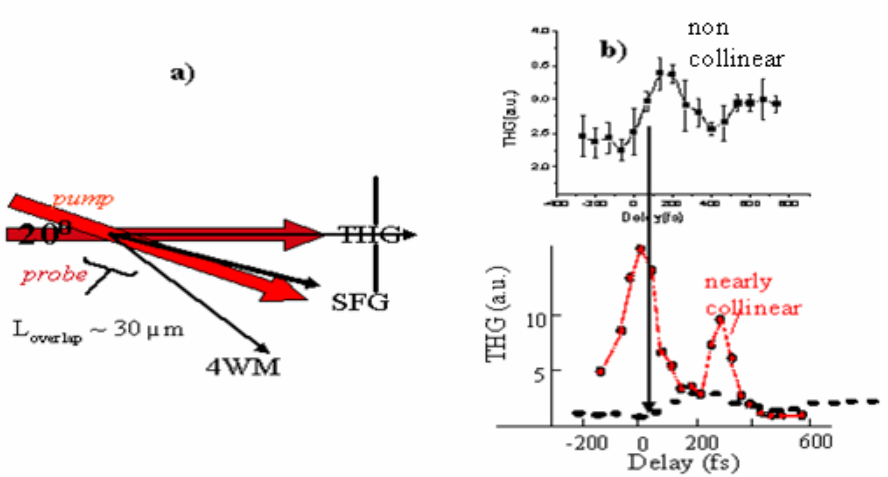

FIGURE 3. Large angle (20^0) Pump(800nm)-probe(800nm) experiments and results from argon clusters. **a)** The overlap length (~30μm) is much smaller than the nearly collinear (~1^0) case (~1mm). **b)** Comparison of THG resonance enhancement. The enhancement of phase matched resonance for the large angle case is suppressed because of small interaction length compared with the nearly collinear case. There is no SFG and 4WM peak near zero delay because of 800-800 pump-probe interaction. The pump intensity was $9x10^{13}$ W/cm^2 and the probe intensity was $2x10^{13}$W/cm^2. The average cluster radius is 21nm for the nearly collinear case and is 13nm for the large angle case.

The fact that only the phase matched third harmonic signal is enhanced even though the THG resonance itself has a long time duration explains the short time duration of the THG peak.

We were also able to see the THG resonance in the collinear pump(400nm)-probe(800nm) geometry and performed pump and probe intensity dependence scans

(Fig. 4). The pump intensity scan with fixed probe intensity (Fig. 4a) shows that at lower pump intensities, the SFG and 4WM processes peak at zero delay, whereas the THG resonance peaks at a later delay and is well separated from the first peak. At higher pump intensities, the clusters expand more rapidly, so the two peaks merge. For a given pump intensity, we found that the THG resonance grows more nonlinearly with probe intensity than the first peak (Fig. 4b, 4c), which is a rough agreement with the theory. The SFG and 4WM should depend linearly on the probe intensity and the THG should have a cubic dependence. The fit analysis of the second peak shows more deviation from cubic (from 2.46 to 1.43) if the third data point with 1.5×10^{14} W/cm^2 probe intensity is included. This intensity is close to the first ionization threshold of Argon gas, therefore the phase matching condition is changed because of additional ionization at this intensity, which makes the dependence deviate more from cubic. Also the probe intensity dependence can be complicated by the mixture of three signals. More thorough experiments are on-going.

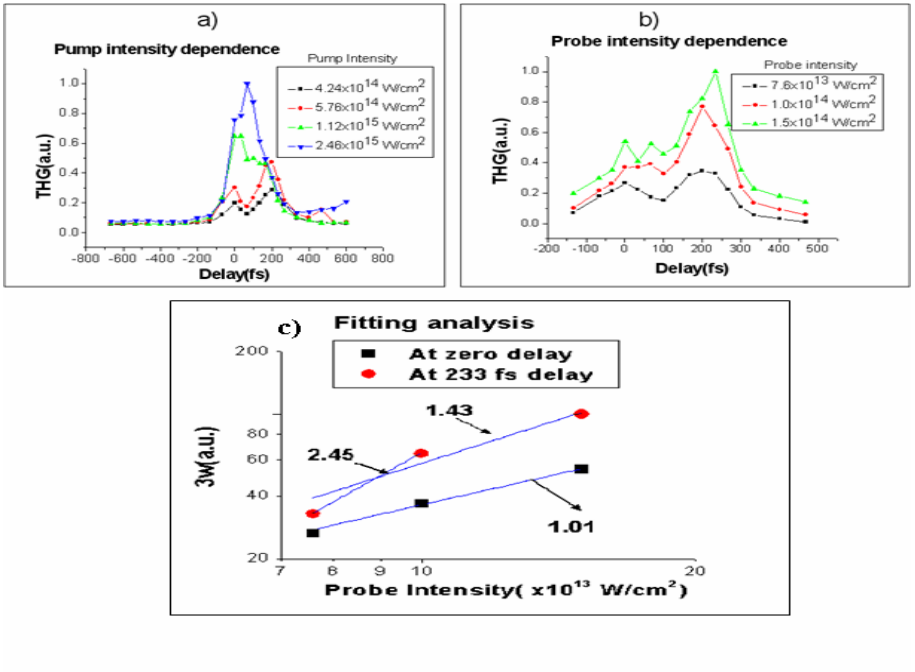

FIGURE 4. Pump and probe intensity dependence from Argon clusters in the collinear geometry. **a)** Pump intensity dependence. By increasing the pump intensity, the clusters expand more rapidly and reach the THG resonance earlier. Therefore the first peak (SFM+4WM) at zero delay and the second peak (THG resonance) at later delay merge together. The probe intensity was fixed at 4×10^{14} W/cm^2. **b)** Probe intensity dependence. The second peak shows more nonlinear growth than the first peak, which verifies that the second peak is the THG resonance peak. The pump intensity was 5×10^{13} W/cm^2. **c)** Fit analysis at each peak. The first peak shows the linear dependence and the second peak is more nonlinear (ideally cubic dependence).

PIC SIMULATION

Our analytical theory describes an individual fully ionized cluster [7].Third harmonic generation results from nonlinear response of the cold electron core [6] in the potential of the positively charged ion background. The model shows that cluster non-uniformity, resulting from cluster expansion, is essential for harmonic generation. The resonant enhancement of harmonic generation occurs when the eigen-frequency of the electron core $\omega_p(t)/\sqrt{3}$ matches the third harmonic of the laser pulse. A PIC simulation of such enhancement is shown in Fig. 5. A more comprehensive model, describing the macroscopic nonlinear optical properties of the clustered gas medium, is being developed. This model will address the phase matching issue in a non-collinear geometry, and the dependence of the third harmonic signal on the relative polarization and angle of the pump and the probe.

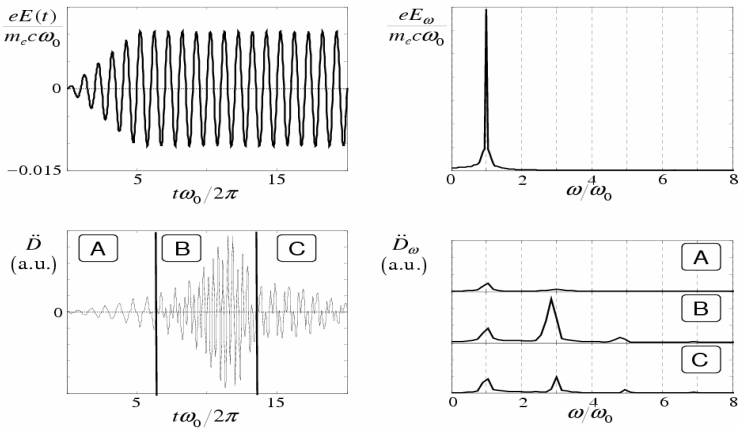

FIGURE 5. Resonance enhancement of third harmonic generation in an isolated cluster ionized by the pump laser pulse. The simulation was done by axisymmetric electrostatic PIC code [7]. The graph on the left shows the second time derivative of the cluster dipole moment *D* in response to the probe pulse. The figures on the right show the spectra of the second time derivative at three different periods of time: A). At an early time the cluster is uniform and has solid density. B). As the cluster expands, the density becomes non-uniform and at the same time the eigen-frequency of the collective electron mode in the cluster tunes to the third harmonic of the probe, leading to resonant enhancement of the nonlinear response at 3ω. C). The response decays because of the detuning from the resonance and leakage of the cluster electrons.

FUTURE WORK

We are now directly measuring the time evolution of $n(\omega)$ and $n(3\omega)$ using frequency-domain interferometry with probe pulses at both ω and 3ω. We expect the refractive indices for ω and 3ω to cross at $\Delta t \sim$ 200-300 fs, which is coincident with the measured THG enhancement.

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