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Title: Coherent Control of Harmonic Generations from exploding clusters

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Recent experiments [1] showed that linear absorption of van der Waals bonded clusters is resonantly enhanced at time delays $\Delta t_{\text{crit}} \sim 1$ ps following excitation by a short pump pulse. 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 higher pump intensity. An analogous enhancement in harmonic generation from exploding clusters was predicted theoretically [2], but has proven elusive to observe in the laboratory.

Here we report pump-probe experiments in which the predicted enhancement in Third Harmonic Generation (THG) from a gas jet of expanding clusters is observed definitively and reproducibly. Equally important, our experiments demonstrate 3 reasons why this result has been elusive: (1) Cluster $\chi^{(3)}$, THG coherence length and probe absorption evolve independently, and compete with each other. Each must be independently measured, modeled and controlled to achieve the desired optimization of THG efficiency. (2) The pump-probe overlap region must coincide very closely with the gas jet to avoid degradation of probe beam quality when propagating through un-pumped clustered gas. (3) THG efficiency from the un-ionized gas jet medium (un-ionized gas + un-ionized clusters) is much stronger than was realized in theoretical work [2]; optimizing THG to a higher level in the ionized medium was therefore correspondingly more difficult than predicted. This comprehensive understanding of the problem is important because it points the way toward coherent controlled enhancement of high-order harmonic generation extending into the soft x-ray regime.

Experiments were performed with the University of Texas Tri-color Terawatt (UT³) laser system with a supersonic Argon gas jet (~1mm length). The gas jet, composed of 80~90% un-clustered gas and 10-20% clusters (20nm average radius), was heated by an intense 400nm, 100 fs pump pulse, then allowed to freely expand. An 800nm, 100 fs beam then probed the micro-plasma at a small angle ($\sim 2^\circ$) and generated a $3\omega_{\text{probe}}$ signal ($\lambda = 266$ nm). We used the non-collinear geometry in order to avoid $3\omega_{\text{probe}}$ signals generated by sum-frequency generation (SFG) or 4-wave mixing (4WM) involving both pump and probe, in directions determined by phase-matching when pump and probe overlap ($\Delta t \sim 0$). We modified the uniformly expanding cluster model [3] including the phase matching effect to explain the experimental data. When the probe is weak (2×10^{13} W/cm²), we observe a sharp enhancement of THG at $\Delta t \sim 0$ caused by the cluster ionization; the delayed enhancement caused by cluster expansion, however, is masked by increased probe absorption, as predicted by our self-consistent model. When we increased the probe intensity to $\sim 10^{15}$ W/cm², on the other hand, the $\Delta t \sim 0$ enhancement is no longer observed because the probe ionizes clusters and gas, thus quenching the nonlinearity caused by pump ionization. However, we observe the predicted enhanced THG from cluster expansion at time delay $\Delta t \sim 200$ -300 fs. The model predicts that the enhancement is a combination of $\chi^{(3)}$ resonance and local phase-matching. It also predicts further THG enhancements at higher probe intensity, if pre-pulses in the probe line can be suppressed, and at higher harmonics, if a vacuum UV detection system can be acquired.

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