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Enhanced Third Harmonic Generation from Anisotropically Expanding Clusters.[1] BONGGU SHIM, GREG HAYS, RAFAL ZGADZAJ, TODD DITMIRE, MICHAEL DOWNER, FOCUS Center, Department of Physics, University of Texas at Austin — We report controlled enhancement of optical third harmonic generation (THG) from hydrodynamically expanding Ar clusters (6 x 10^5 atoms per cluster) several hundred femtoseconds following ionization and heating by ultrashort pump pulses. A 400 nm, 100 fs pump beam ionized a gas jet composed of Ar clusters and residual gas. An 800 nm, 100 fs probe then generated third harmonic radiation from expanding clusters at controlled delays. Simulations show that the nonlinear third order susceptibility of individual clusters and the THG coherence length of the clustered plasma medium were optimized nearly simultaneously as the pre-heated clusters expanded, and both contributed to the observed THG enhancement. When resonantly enhanced, THG developed temporary polarization anisotropy, showing that the clusters expanded faster along the pump polarization. By contrast, the linear optical response was isotropic. The physical mechanisms contributing to enhanced THG are scalable to relativistic probe intensity and to high-order harmonics extending to the soft x-ray regime.

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