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2015 J. Phys.: Conf. Ser. 635 122002

(http://iopscience.iop.org/1742-6596/635/12/122002)

Low-order harmonic generation in a ZnS laser ablation plasma

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Synopsis Low-order (3rd to 9th) harmonic generation of a near-infrared driving laser (1064 nm) is reported in a laser ablation plasma of ZnS. Temporal analysis shows two distinct components with respect to the ablation event. The late temporal component exhibits high conversion efficiency for the highest harmonic orders observed. This is attributed to a dramatic modification of the plasma medium with the driving laser.

Harmonic generation has become a standard technique for the synthesis of coherent radiation in the high frequency regions of the electromagnetic spectrum, from the UV through VUV and XUV, to X-ray. Low-order harmonic generation is possible with moderately intense lasers of nanosecond duration, and is used as a tool for VUV spectroscopy [1]. In contrast, high-order harmonic generation is only possible in the high laser intensity regime (typically above 10^{14} W/cm^2) only easily attained in systems that deliver ultrashort laser pulses of femtosecond duration.

Atomic and molecular gas jets have been the most common nonlinear media employed for harmonic generation, but the universality of the phenomenon makes alternative sources viable. Laser ablation plasmas offer interesting avenues for research, and have been explored as nonlinear media for harmonic generation in recent years in a number of works [2,3] both in the low and high intensity regimes. Particularly attractive is the possibility to tailor the characteristics of the medium through the control of the ablation laser characteristics. In particular, control of the density, composition, spatial extent or degree of ionization are possible with a wise choice of ablation laser parameters like wavelength, intensity, pulse duration or spatial properties of the laser beam on the surface of the target.

In this work, we report on the generation of low-order harmonics (from the 3rd to the 9th) of a fundamental NIR laser source of nanosecond duration at 1064 nm in a NIR laser ablation plasma of the wide bandgap semiconductor ZnS. The ablation laser irradiated the target at normal incidence, and the driving laser propagated parallel to the target at a controllable distance (typically 1 mm). The delay between the ablation event and the harmonic generation event was fully controllable electronically, and the focal position of the driving laser could be placed on-axis with respect to the ablation plasma propagation direction, or out-of-axis. This permitted full freedom of spatiotemporal exploration.

Harmonic generation was found to maximize at two distinct temporal delays with respect to the ablation event, as shown in Figure 1. The first peak was found around 250 ns, and the second, around 10 μs. Interestingly, the early component dies off quickly with increasing harmonic order (becoming absent for the 9th harmonic), whereas the late component is extremely intense for the 7th harmonic and is still clearly visible with the 9th harmonic. This evidence points to the idea that the driving laser induces a secondary plasma in a precursor population composed of ZnS aggregates and nanoparticles, and this secondary plasma is characterized by a particularly high conversion efficiency towards higher harmonic orders. The control of the properties of the secondary plasma using this method opens new routes for finding efficient nonlinear media for nonlinear wavelength conversion.

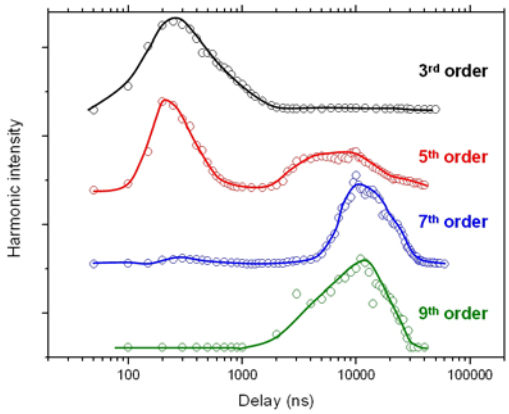

Figure 1. Normalized harmonic emission at orders 3, 5, 7 and 9 of the fundamental NIR driving laser in a ZnS laser ablation plasma as a function of delay with respect to the ablation event.

References

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