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Plasma structures for quasiphase matched high harmonic generation

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A scheme for creation of periodic plasma structures by ablating a lithographic pattern is demonstrated. A proof of principle experiment was conducted, and plasma parameters were measured as a function of time with spatial resolution <10 and 100 μm periodicity. Several possible applications, in particular, quasiphase matching for high harmonic generation in plasma are considered. © *2011 American Institute of Physics*. [doi:10.1063/1.3578407]

High harmonics generation (HHG) is a standard technique for generating coherent radiation in the EUV and even x-ray regimes, with various applications from attosecond physics[1,2] to biology.[3] In the standard scheme of (nonrelativistic) HHG an ultrafast laser pump beam at intensities around 10^{14} W/cm^2 is focused into gas jet, generating high harmonics.[4] The yield of such schemes is inevitably limited by dispersion in the medium.[5] Across a distance equal to the coherence length a phase mismatch of π grows and causes destructive interference between the pump and high harmonic beams. This process is a major limitation on the conversion efficiency of HHG.

Quasiphase matching (QPM) is a well known "fix" for the phase mismatch problem.[6] In QPM the medium is modulated with a coherence length period so that pump phase or harmonic emission is changed to prevent the destructive interference caused by the phase mismatch.[3] For HHG in the EUV regime and beyond, dispersion in the gaseous medium can be mostly attributed to free electrons generated by laser ionization of the medium. Under this assumption the Coherence length (at ~ 0.8 μm wavelength) is given (in meters) by:[2] $L_c \propto 10^{15}/qN_e$ where N_e is the free electron density (in per cubic centimeter) and q is the harmonic number. Recently QPM was realized by using multiple gas jets whose pressure and separation were properly controlled.[7] However, the realization of this technique is limited by geometrical constraints on the number and minimal separation of the jets.

In this paper we are proposing a simple method for fabricating numerous plasma jets, tailored for HHG, reliving technical restraints on the dimensions of the jets and their periodicity. In our scheme the jets are produced by ablation of a microlithographic periodic stripe pattern (Fig. 1). Cylindrical plasma jets formed by ablation extend the lithographic pattern into the space above the target, creating a row of narrow plasma jets of different material composition. The efficiency of HHG in plasma has been demonstrated to vary considerably with the atomic composition,[8] and the periodic change in this efficiency enables QPM-HHG. The electron density and plasma structure that the HHG pump laser is incident upon can be adjusted (much like in using standard gas jets) by a multitude of control parameters. These parameters are: structure and material composition of the ablated target, the delay between the ablation and the pump incidence, the distance and angle at which the HHG pump is incident and the ablating laser's parameters.

While we focus our attention to applications in HHG, this scheme can be considered with some variations for other high-field applications such as transient plasma gratings for high intensity optical systems,[9] or tunable millimeter wave gratings.[10]

A proof of principle experiment was conducted using an Nd:YAG laser, that delivered 1064 nm, 15 ns, 140 mJ pulses at a 0.2 Hz rep rate as the ablator. The ablating beam was focused by cylindrical leans to a ~ 1 mm$\times 30$ μm [full width at half maximum (FWHM)] spot on the target's surface, in vacuum (<200 mTorr). The target was $50\times 50 \times 1$ mm^3 plastic covered with 40 μm of copper, etched to produce a stripe pattern with a ~ 100 μm (see Fig. 1) by a relatively cheap photoelectromorphing process. At laser intensities required for jet generation, only a small layer of material was ablated with each shot, and roughly 100 shots were made at the same location on the target before moving to a new (unexposed) area.

In order to find optimal conditions suitable for HHG we measured the plasma parameters (temperature and density) as a function of the delay after the ablation onset. Plasma parameters were measured by means of optical emission spectroscopy. The radiation emitted by the plasma was collected at a small angle with respect to the target (see Fig. 1) by UV-transparent optics, and imaged onto spectrometer entrance slit (0.3 m optical length, 1200 g/mm grating, 300 nm blaze) coupled to a gated Andor ICCD. The gating was var-

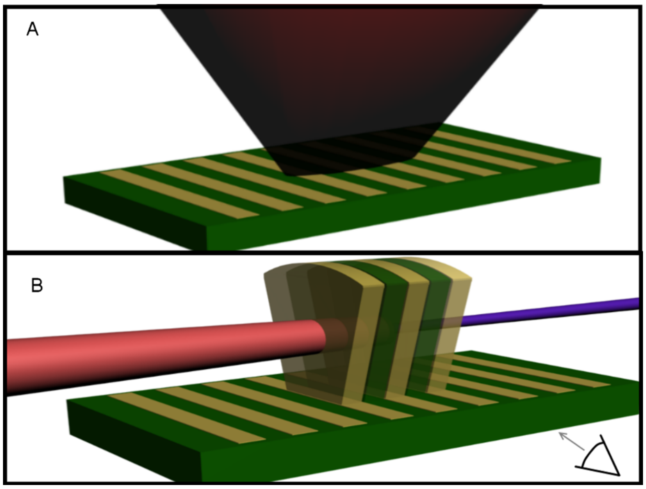

FIG. 1. (Color online) System schematics: the lithographic pattern hit by a relatively low intensity laser beam (a), and the formed plasma jets in which the high intensity laser pump facilitates HHG (b).

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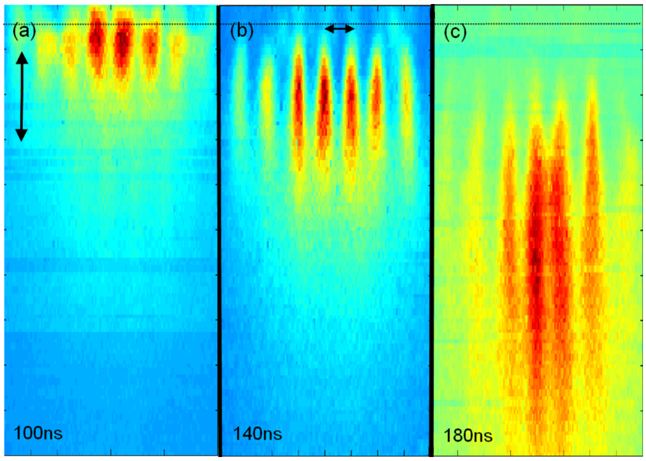

FIG. 2. (Color online) Monochromatic imaging of the plasma jets at different times using a 30 ns gate. The dashed line marks the target's surface, the double arrows are 200 μm scale.

ied between 20 and 200 ns to within a few ns jitter with respect to the ablating laser. The spectral resolution was better than 5 Å. Two-dimensional (2D) spatial resolution was achieved by imaging the plasma on the ICCD through a narrow-band interference filter (326 ± 10 nm FWHM), which transmits the 324.8 and 327.3 nm characteristic lines of neutral copper atoms. While the plasma is not entirely optically sparse at these wavelengths, these pictures give a good *qualitative* 2D picture of the plasma and it's spatial modulation.

The plasma electron temperature was derived from intensity ratios[10] of the lines emitted by neutral copper atoms at 330.8, 510.6, and 521.8 nm, assuming local thermal equilibrium and negligible Doppler line broadening.[11] The temperature of ~ 0.8 eV was measured at 140 ns with respect to the ablation onset and at 100 to 300 μm away from the target surface. The plasma electron density was derived from line broadening caused by the linear Stark effect in hydrogen[12] (from both plastic and impurities in the copper), with Stark coefficients determined by the temperature values we previously measured.[8] By imaging the jet across the entrance slit the spatial dependence of the temperature and density across the z-axis (toward the ablating laser) was obtained. A one-dimensional hydrodynamic code HYADES (Ref. 13) was used to simulate the plasma conditions obtained in the experiment and compared to the electron density measured by Stark broadening.

These results demonstrate a simple method for generation of periodic plasma structures by ablating a previously made lithographic pattern. By passing a high intensity laser pulse through such plasma patterns suitable conditions for QPM required for HHG can be created. Our measurements suggest such conditions exist around 140 to 180 ns after initiation of plasma by the ablating laser pulse. Within this temporal window the plasma jets are several hundred microns wide and have relatively uniform temperature and relatively low electron density of $\sim 10^{17}$ cm^{-3}, whereas at later times the plasma structure begins to fade.

Examining Fig. 2 in depth shows that the modulation of the plasma density is significant, and suggests that much finer periodicities (suitable for generation of higher harmonics) could be obtained by using finer lithography in preparation of the target. Plugging the measured temperature and density into the Saha equation results shows the degree of ionization for this plasma region above 1. This implies that even at high pump intensities the QPM inducing plasma structure will not be destroyed considerably by subsequent ionization induced by the HHG pump laser as it goes through the plasma. Moreover, the proper choice of target materials can allow for increased laser intensities that may lead to higher HHG yield (Fig. 3).

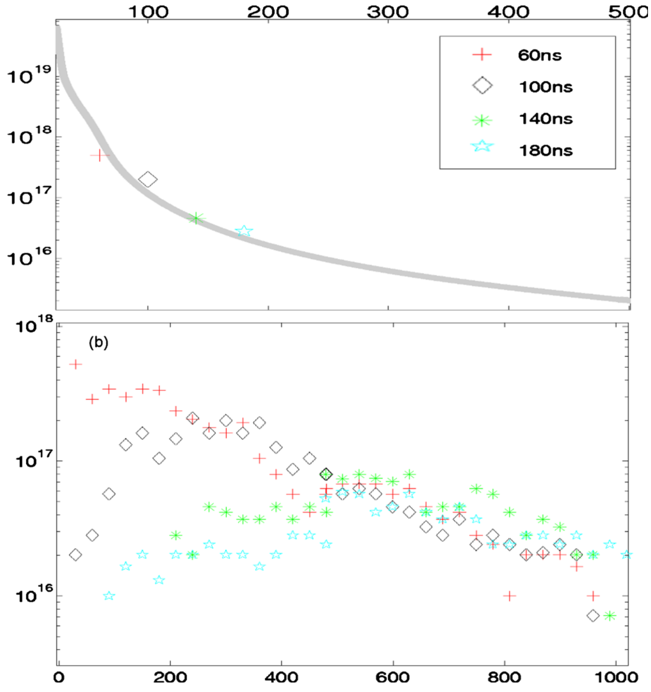

FIG. 3. (Color online) Typical electron density in per cubic centimeter vs time (in nanosecond) comparison of the experimental results (points) to simulation (continuous line) (a), and the electron density derived from the stark broadening of the hydrogen using a 20 ns gate (b).

In conclusion we have demonstrated the feasibility of a robust scheme for tailoring plasma structures with control over material composition, temperature, and density (both of free electrons and neutrals), through ablation of specifically prepared lithographic targets. We have examined our ability to control these plasma conditions for a specific case of a periodic plasma grating which we believe can support quasiphase matched HHG.

While we have demonstrated a periodicity of 100 μm and only ablated ~ 6 jets per shot, the scalability of the proposed solution to more numerous jets and other periodicities is obvious. Based on our findings, we estimate that 10 μm periodicity is easily achievable with a finer target. While we have focused our efforts on variations to the spatial and temporal delays between ablation and HHG, farther degrees of control can be explored by modifying the ablating laser's intensity, the angle of the HHG laser beam with respect to the plasma structure, and the periodicity, depth and material composition of the lithographic pattern.

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