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Citation: Phys. Plasmas **19**, 030703 (2012); doi: 10.1063/1.3693388
View online: http://dx.doi.org/10.1063/1.3693388
View Table of Contents: http://pop.aip.org/resource/1/PHPAEN/v19/i3
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# Surface-plasmon-enhanced MeV ions from femtosecond laser irradiated, periodically modulated surfaces

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(Received 23 November 2011; accepted 17 February 2012; published online 16 March 2012)

Enhanced emission of high energy ions is measured from sub-wavelength metallic grating targets under irradiation by intense ($10^{15}-10^{16}$ W cm$^{-2}$), p-polarized, 50 fs, 800 nm laser pulses. The maximum ion energy is 55% higher and the ion flux is 60% higher for the modulated surface in comparison with polished surfaces of the same atomic composition. The ion emission, a result of enhanced light coupling and hot electron production in the grating targets, is correlated with enhancement in hard x-ray bremsstrahlung emission. The results are well reproduced by particle-in-cell simulations. The study reveals that the enhanced laser coupling by surface plasmon excitation on metallic gratings is directly responsible for the enhancement of ion energies. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3693388]

One of the exciting spin offs of intense, femtosecond laser interaction with solids is the generation of short bunches of energetic charged particles with low emittance.[1–6] Such charged particle as well as electromagnetic emissions are caused by hot electrons, which arise from preferential absorption of laser light by specific collisionless absorption processes like resonance absorption (RA)[7] and vacuum heating (VH)[8] in the dense plasma at non-relativistic laser intensities. Although several efforts have been made to enhance this coupling using the conditioning of laser pulses and targets,[9–11] the most promising way of efficient laser energy coupling appears to be the micro- and nano-structuring of the target surface. Metal nanoparticles,[12,13] nanowires,[14,15] and micrometer scale polystyrene spheres[16] have all been investigated for enhancing such coupling to produce higher x-ray emission. Recent results from our group have shown that the sub-wavelength structural modification[9,12] of the target surface couples laser energy more efficiently (up to 93%) compared to conventionally used polished surfaces. The enhanced laser coupling to the nano-structured surface produced plasmas is basically attributed to (a) the "lightning rod" effect, a purely geometrical effect depending on the shape of the structures leading to localized enhanced electric field amplitude near the sharp edges and (b) the excitation of surface plasmon waves (SPWs), in moderate laser intensity regime. Experimental studies[17] and reports based on numerical simulations[18] at relativistic laser intensities suggest near complete absorption of incident laser energy. The enhanced laser absorption yields enhanced energetic x-ray emission primarily attributed to localized enhancement of laser electric field in the vicinity of sub-wavelength structures. But there appears to be very little work on using structured surfaces for enhancing ion emission. Recently, we presented a counterintuitive observation[13,15] that enhanced laser absorption leading to higher production of hotter electrons from nano-structured targets does not necessarily lead to the enhancement of the ion energy. The evolution of hot electrons in the plasma eventually decides the ion emission characteristics. Therefore, controlling the plasma evolution with an aim to improve ion emission characteristics still remains a challenge.

In this paper, we demonstrate that the extra laser energy absorbed by metallic sub-wavelength surface modulation can indeed be guided for enhanced energetic ion emission from the freely expanding laser produced plasma. We report that for non-relativistic laser intensities sub-wavelength surface modulations lead to more than 50% increase in the highest ion energy and 60% more ion flux with intensity dependent divergence in comparison to a polished target of same atomic composition paving the way for multi-kilohertz tabletop operation of such ion sources. We also performed two-dimensional (2D) particle-in-cell (PIC) simulation, which faithfully reproduces the basic experimental observations.

The targets used in the present experiment are (a) gold coated triangular blazed (17.45°) grating (AuGR) on a glass substrate having a periodicity of 555 nm and groove depth of 158 nm and (b) gold surface (Au) with optical surface polish ($\lambda/10$), used as a reference for comparison. The gold coating is much thicker than the optical skin depth of laser. P-polarized, 50 fs, 800 nm, 10 Hz Ti:Sapphire laser pulses (nano-second pre-pulse contrast better than $10^{-6}$) are focused

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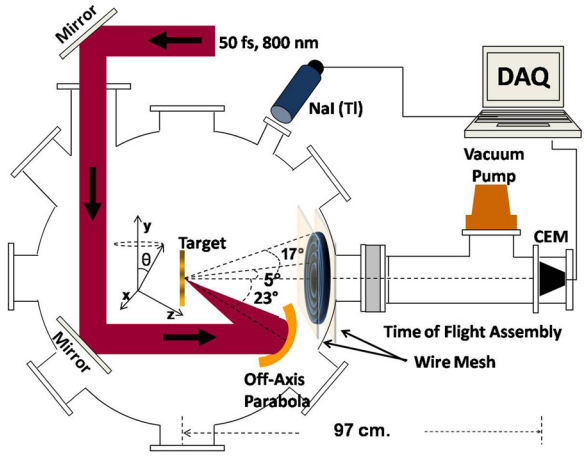


FIG. 1. (Color online) Schematic experimental arrangement. P-polarized, 50 fs, 800 nm laser is focused on to the sub-wavelength grating (AuGR) and polished gold (Au) targets, placed adjacently for comparison under identical experimental conditions. The bremsstrahlung emission is recorded by time-gated NaI (Tl) detector and the charged particle emission is characterized by a CEM and four concentric AnFCs.

by a f/4 off-axis gold coated parabolic mirror to a focal spot size of 10 $\mu$m at an incident angle of 23°, which is the phase matching angle,[17,19] for this grating to excite surface plasmon induced absorption. The polished and grating targets are mounted adjacently in a vacuum chamber (base pressure $10^{-6}$ Torr) and are moved by a motorized precision stage to ensure a fresh target portion for each laser irradiation. A schematic of the experimental apparatus is shown in Fig. 1.

Hard x-ray emission in the 20–300 keV range is measured with calibrated NaI (Tl) detectors, (a) gated in time with respect to the incident laser pulse to ensure almost background free data acquisition and (b) covered by thick lead walls and lead apertures to reduce "pile up" events. The signal from the detector is collected by a multichannel analyzer (MCA) connected to a computer. The energy of the ions emitted from the plasma, along the target normal direction, is measured by a channel electron multiplier (CEM) used in proportional mode, exploiting conventional ion arrival time measurement technique. Four large annular faraday cups (AnFCs) (Ref. 20) are placed at different angles ($\theta$) of 5° (FC1), 8° (FC2), 12° (FC3), and 17° (FC4) with respect to target normal direction to measure the total flux as well as the angular divergence of the ions emitted from the plasma. A biased nickel mesh of large transmission was installed in front of the AnFC to repel the low energy electrons.

The bremsstrahlung spectra from both samples obtained at an intensity of $2 \times 10^{16}$ W cm$^{-2}$ yield hot electron temperature of $11 \pm 0.6$ keV and $26 \pm 0.7$ keV for Au and gold grating target (AuGR), respectively. This clearly signifies hotter and denser plasma formation[12,17] in the latter. The bremsstrahlung x-ray yield recorded over 128 consecutive laser shots at different laser intensities reveals that the x-ray yield from the grating surface is consistently higher than the polished gold surface over the entire intensity range considered in the experiment. The surface plasmon resonance (SPR) on a sub-wavelength grating surface occurs for a certain angle, which is highly polarization-dependent and leads to nearly complete absorption at the resonance angle.[17] The AuGRs are kept at the particular phase matching angle during the experiment to excite SPR at the grating surface. S-polarization under the same conditions does not enhance the emissions. Angle and polarization dependencies clearly prove the role of SPR. The yield ratio, however, decreases with increase in laser intensity because of detrimental modification of surface topography by the rising edge of the laser pulse similar to other observations.[13,15,20] Note that, based on a simple electrostatic calculation, the maximum enhancement of the local electric field on the grating surface can be estimated[21] as $\eta'_{max} = 2|\epsilon'_m|^2 \cos\theta_{in}/\epsilon''_m\,(|\epsilon'_m| - 1)^{1/2}$, where $\epsilon_m = \epsilon'_m + i\epsilon''_m = -33.26 + i14.52$ is dielectric permittivity[22] of gold at 800 nm and $\theta_{in}$ is the angle of incidence of laser. Note that the above expression is derived under the condition that $\epsilon''_m$ is much smaller than $\epsilon'_m$—that is, for weak damping of the plasmon. Under actual conditions, the amplification is large but will be suitably limited by the damping present in the system. In this case, the local electric field enhancement comes out to be 24 times higher than the polished surface. The "hot" electron temperature scaling law[23] gives an increase of 2.9 times the "hot" electron temperature obtained from polished surface. This estimate is reasonably close to the experimentally observed "hot" electron temperature ratio of 2.36 from both surfaces. Now we move on to address the question whether the enhanced laser energy absorption leads to enhanced ion emission. We have measured the energy distribution and the angular divergence of the ions emitted from freely expanding plasma as described below.

From CEM ion energy measurement, it is evident that more energetic ions have been measured in case of AuGR as compared to Au as shown in Fig. 2(b). The maximum ion energy from AuGR surface is consistently higher that of the Au surface by more than 50% as shown in Fig. 2(a). The straight lines show the result of fitting $(I\lambda^2)^{\alpha}$ to the experimental data. Both data follow reasonably this scaling of $\alpha = 0.4$ as several experiments have previously reported.[24,25] Note that the maximum ion energy emitted from freely expanding laser produced plasmas is proportional[26] to the "hot" electron temperature $T_{hot}$ of the plasma. In the present context, the expected increase in the maximum ion energy $(\sqrt{T_{hot}^{AuGR}/T_{hot}^{Au}} = \sqrt{26/11} = 1.54)$ matches very well with experimentally obtained enhancement factor of 1.57. This clearly shows the direct channeling of the coupled laser energy into the ion emission.

The maximum ion energy recorded Fig. 2(b) shows the typical ion energy spectra recorded during the experiment at an intensity of $2 \times 10^{16}$ W cm$^{-2}$. Note that, at higher laser intensity, a low energy component of the AuGR ion distribution (<20 keV) overpasses the Au ion distribution (see the dotted circle on Fig. 2(b)). Considering the total ion flux emitted from the plasma, it is seen that the AuGR yields 60% more ions compared to Au validating that AuGR surface couples more laser energy to the plasma compared to the polished Au surfaces.

To estimate the angular divergence of the ion beam emitted from both samples (Au and AuGR), four concentric AnFCs are employed. The AnFCs, placed at different angles (5°, 8°, 12°, and 17°) with respect to the target normal

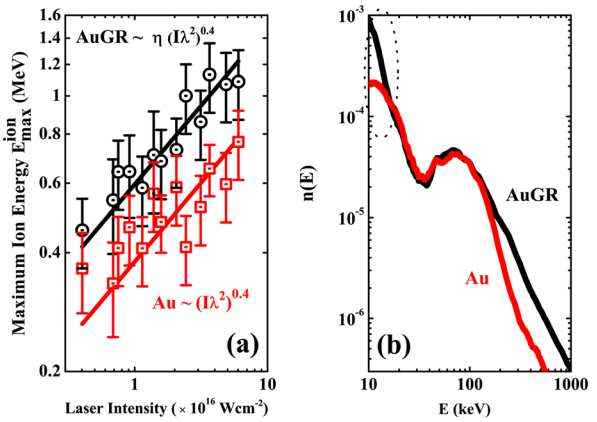


FIG. 2. (Color online) The maximum energy of ions emitted from both the samples under identical laser focusing conditions is shown in (a). The straight lines indicate $(I\lambda^2)^{\alpha}$ scaling behavior. The average maximum ion energy recorded from AuGR is consistently higher by a factor of 1.57 which matches well with the theoretically expected enhancement factor of 1.54 (see text for details). Plot (b) shows typical ion energy spectra obtained at an intensity of $2 \times 10^{16}$ W cm$^{-2}$.

direction, provide angular divergence measurements for every laser shot incident on the samples. The angular divergence of the ion beam from Au target follows the well-known bell shaped ion flux distribution with the peak lying along the target normal direction. This trend remains invariant with increase in laser intensity. On the other hand, the ion emission from AuGR shows a different behavior. First, the ion emission becomes more divergent in nature which increases with laser intensity. The ion flux emitted from AuGR seems to have to major components. The first one, similar to Au, lies mostly along target normal direction (CEM $\equiv 0^{\circ}$). While the other bunch, highly divergent in nature, is mostly detected by the outermost AnFC 4 placed at angle of 17° with respect to the target normal direction. This distribution is similar to what has been reported earlier.[17,18] In between these two distributions, a hollow region with a relatively low ion flux appears. The width of this hollow region appears to increase further with increasing laser intensity. To represent the relative variation of ion flux distribution from the planar and corrugated surfaces, the normalized ratio of ion flux emitted by AuGR to that of Au as recorded by each AnFC is plotted as a function of its angular position (Fig. 3(a)). The relatively larger divergence from AuGR might be indicative of a small amount of non-planarity in plasma expansion.[24,25,27]

It is to be noted for the entire intensity range of laser intensities considered in this report; the total ion flux emitted from the AuGR is consistently higher than the ion flux from polished Au. To elucidate this point, in Fig. 3(b), a comparison of the ion flux recorded by AnFC 3 (placed at an angle of 12° with respect to the target normal direction) for both the samples are presented over the entire intensity range. The same trend is observed for the other AnFCs too. This reconfirms the enhanced laser absorption because of surface plasmon excitation at the grating surface. While it may appear that this contradicts the trend shown by x-ray yields, mentioned earlier, note that at higher intensities the detrimental effect of the rising edge of the incident laser pulse on surface topology results in strong ablation of low energy (<20 keV) debris (dotted circle, Fig. 2(b)) from the modulated surface. This is responsible for the consistent increase of ion flux from modulated surface with increasing laser intensity. We have, indeed, varied the angle of incidence and the polarization of the incident 50 fs, 800 nm laser beam using a $\lambda/2$ plate and recorded the bremsstrahlung spectra and ion energy spectra from both the surfaces. We find that the grating surface does not yield anything different from a gold surface for s-polarization.

To understand the above experimental observations, 2D-PIC simulations are performed for both the polished gold and the gold grating targets at a laser intensity of $5 \times 10^{16}$ W cm$^{-2}$ with identical laser pulse parameters used in the experiment. The initial plasma density was taken to be 10 $n_{cr}$ ($n_{cr} = 1.72 \times 10^{21}$ cm$^{-3}$ for 800 nm) with ion charge states taken as +4. The simulation box is $8240 \times 3200$ ($206\lambda_0 \times 80\lambda_0$), and there are 196 macro-electrons and 49 macro-ions in a grid cell. Figure 4(a) shows the fraction of incident energy absorbed in the target. In the PIC simulations, the effect of the SPR is manifested in terms of increase in localized electrostatic field at the grating surface.[18,19] In the present case, the grating surface absorbs 54% more laser energy compared to polished surface, which is contributed by the enhancement effect of the local electrostatic field at the grating surface.[18] The laser energy absorbed is converted to hot electrons first, as shown in Fig. 4(b). The hot electron temperatures for the modulated surface and polished surface are 44 keV and 28 keV, respectively. Later, the hot electron energy is partially transferred to heavy ions slowly. Therefore, the ion temperature of the gold grating should be much higher due to the larger laser absorption, which is confirmed by the PIC simulation results in Fig. 4(c) showing ion energy distribution in logarithmic scale. The ion temperature for the gold grating target (17 keV) is significantly higher than that for the polished target (8 keV). Note that the simulation reproduces the basic qualitative trends in the observations, though the ion energy found in the simulation is still much smaller than that observed in the experiments. A possible explanation is that some gold atoms are ionized to much higher charge states than $Au^{4+}$ adopted in our simulation due to the local electric field enhancement associated with SPR on a sub-wavelength grating surface. Also in the three-dimensional geometry, the effect of local field enhancement can be even significant, which shall be pursued in future.

The strongest fields for the gold grating target are at the vacuum-target boundary (within the grooves), but the strongest fields for the polished gold target case are far away from the boundary, which comes from the addition of the incident and reflected laser fields.[18] This enhancement effect of electrostatic fields at the boundary is responsible for the high laser energy absorption in the grating. Initially, the electric field density is found to be quite high at the grooves, an effect which has been noticed even in the low intensity linear field calculations employing finite difference schemes.[28,29] The enhanced electric field density initiates plasma formation at the groves, and the electrons generated are influenced by the time dependent interference fringes of the laser electric field resulting in a stochastic[30,31] motion. The electrons

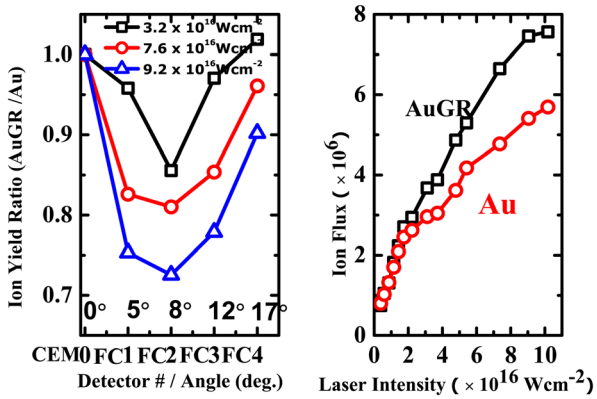


FIG. 3. (Color online) (a) The ratio of ion flux measured by AnFCs from AuGR and Au surface with increasing laser intensity pointing towards the intensity dependent divergence of the emitted ion flux. Note that the AuGR/Au yield ratio is set to 1 at CEM (0°) to make a relative comparison of the yields. This figure indicates that ions from AuGR have a larger divergence. Plot (b) shows the total ion flux captured by FC3, placed at an angle of 12° with respect to the target normal with increasing laser intensity.

collide with the surrounding groove walls and, therefore, produce hotter and denser plasma as compared to the polished surface as recorded in the bremsstrahlung spectra.

As the electric field density initially maximizes at the shallow grooves, the expanding plasma carries an imprint of the modulated surface and is expected to induce modulation[32] in sheath profile. As the ion expansion occurs mostly normal[33,34] to the local surface, a diverging ion distribution is expected. This is evident in the relative divergence measurement (Fig. 3). We believe the intensity increase at the surface of the grating is compromised by surface damage at high intensities resulting in ion expansion pattern similar to polished surface. The experimental observation of increase in maximum ion energy is very well validated in the experiments (Fig. 2(a), an average 55% increment in maximum ion energy is observed and in PIC simulations, Fig. 4(a)). The striking similarity of the 54% extra absorption of incident laser energy

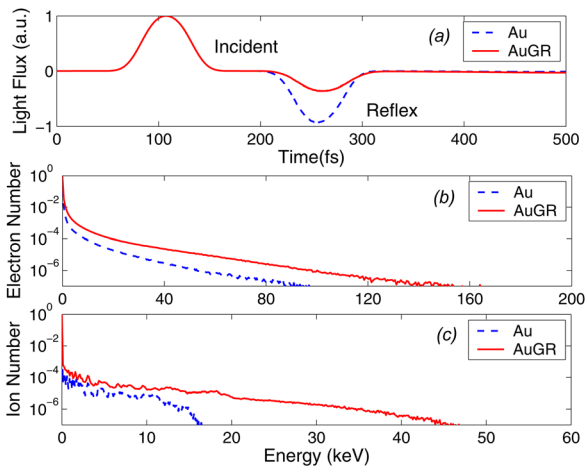


FIG. 4. (Color online) 2D-PIC simulation results displaying the response of modulated and plane surfaces irradiated with an ultrashort laser pulse at intensity of $5 \times 10^{16}$ W cm$^{-2}$. (a) The laser energy absorption by the AuGR and the polished gold target (Au). The grating surface absorbs 61% and the polished surface absorbs only 7% of the incident laser energy. The electron energy distributions from both surfaces at 150 fs are shown in plot (b). Plot (c) shows the ion energy distribution at 500 fs.

by the grating surface with the enhancement of ion accelerating electric field clearly establishes the fact that this extra absorbed laser energy by the surface modulations is directly responsible for accelerating ions to higher energies.

In summary, we have presented a study on the enhancement of MeV ion emission from periodic metallic surfaces from intense femtosecond laser produced solid plasmas in comparison with polished metallic target of the same atomic composition. The study clearly points out that the modulated surface couples more laser energy into the plasma via surface plasmon absorption producing hotter plasma. The extra energy absorbed because of surface modulations is directly responsible for producing higher energy ions from modulated surfaces. We believe that further investigations are necessary to understand the transfer of energy from the electrons to the ions for a particular local geometry. It is also necessary to carry out more measurements under different laser and target conditions. It may then be possible to further enhance the ion fluxes and maximum ion energies. It is hoped that this study opens up the possibility of designing efficient targets towards practical multi kilohertz, tabletop femtosecond ion accelerators.

M.K. acknowledges a Swarnajayanthi Fellowship and G.R.K. a J.C. Bose Fellowship from DST (GOI).

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