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Efficient hard X-ray source using femtosecond plasma at solid targets with a modified surface

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Abstract

Recent results on constructing of an efficient hard X-ray source using solid targets irradiated by high-contrast 200-fs laser pulses with an intensity above 10^{16} W/cm^2 are presented. We used different solid targets with a laser- and electrochemically modified surface layer: craters, pyramidal cavities, porous silicon, gratings. Experimental data obtained confirms that using solid targets with a corrugated surface one can achieve a prominent increase both in the efficiency of hard X-ray generation (in the quanta range 2–30 keV) and in the hot electron temperature of plasma.

Keywords: Femtosecond laser plasma; Modified surface; X-ray source

1. INTRODUCTION

The interaction of intense femtosecond laser pulses (above 10^{16} W/cm^2) with solid targets is of current interest both for the study of high-density plasmas and for the construction of short-pulse X-ray sources. It is well known that high-temperature, solid density, nonequilibrium plasma formed under such an interaction is the bright source of prompt X-ray flashes in the soft and hard X-ray regions (Murnane *et al.*, 1989; Gulietti & Gizzi, 1998). One of the attractive approaches to the enhancement of X-ray yield from a femtosecond laser–plasma interaction consists of using targets with a modified surface (Wulker *et al.*, 1996; Volkov *et al.*, 1997). This approach allows us to get an increase in hard and soft X-ray yield, hot electron temperature, second harmonic generation efficiency, and so forth, without changing the intensity, energy, or duration of a laser pulse. Surface modification can be obtained in different ways: grating burning or crater formation by laser radiation (Golishnikov *et al.*, 2001; Volkov *et al.*, 2003), black metal layer sputtering (Gordon *et al.*, 1994; Wulker *et al.*, 1996), porous layer formation by electrochemical etching (Nishikawa *et al.*, 1997; Volkov *et al.*, 1997, 1998), and production of an array of tightly packed nanowires or holes by electrochemical surface processing (Gordienko & Savel'ev, 1999; Kulcsar *et al.*, 2000; Nishikawa *et al.*, 2001; Gordienko *et al.*, 2003).

In this article we report on our recent experimental advances in investigation of hot electron generation and efficient hard X-ray production with various types of modified targets—laser induced gratings, electrochemically etched pyramidal cavities on Si wafer, and laser-induced craters. For the latter issue we obtained experimental data for a very broad class of laser-modified targets (LM targets) including crystal and amorphous targets, conductors and dielectrics: Si, Ge, crystalline and fused quartz, KDP, Fe, Ti, TiD, W, Zr, Pd.

2. EXPERIMENTAL SETUP AND TARGET SURFACE PROCESSING

A dye-laser system (616 nm, 200 fs, 0.3 mJ) was used in the experiments (Volkov *et al.*, 1997). The laser pulse was focused to the spot with a diameter of 3.5 ± 0.5 μm yielding intensity at the surface of a target above 10^{16} W/cm^2 with intensity contrast better than 10^5 (*p*-polarized unless otherwise specified). The target was placed inside the vacuum chamber with residual pressure less than 10^{-2} Torr (Fig. 1). Two NaI(Tl) detectors equipped with changeable X-ray filters made of Be, Al, Ta, and Cu foils were used for X-ray yield measurements. This double channel hard X-ray detection scheme allowed us to estimate plasma hot electron temperature in every laser shot (Gordienko *et al.*, 2002). All

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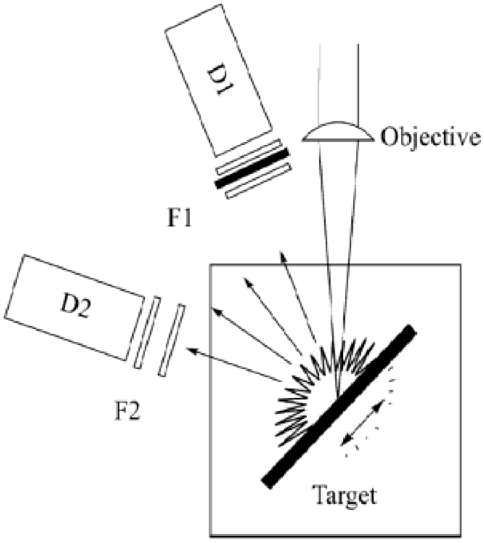

Fig. 1. Experimental setup: D1, D2: detectors with X-ray filters (F1 and F2).

the results obtained with modified targets were compared with data obtained using the flat surface of the same target.

We used three different approaches to the target surface processing: (1) electrochemical etching providing us with pyramidal cavities on Si wafer (Fig. 2a), (2) surface modification by a tightly focused laser pulse with subsequent crater formation (Fig. 2b,c), and (3) quasi-periodic grating generation by a series of femtosecond laser pulses with an intensity slightly above the surface melting threshold (Fig. 2d).

Pyramidal cavities on Si wafer were prepared by chemical etching of *n*-type (100) Si wafer with a resistivity of 5 $\Omega \cdot$cm through rectangular windows formed in an SiO_2 mask, using the well-known technique of isotropical etching (Sangwal, 1987). The etching was performed in 40% KOH solution at 60°C for 5 min. Chemical etching of uncovered areas resulted in formation of inverse pyramidal cavities bounded with (111) crystallographic planes. The measured base size was $8 \times 8\ \mu m^2$ with the base angle of the pyramids of 57.8° (Fig. 2a). The resulting cavity depth was 6.4 μm. After cavity formation the oxide mask was removed by HF acid.

During this experiment angles of incidence of the femtosecond laser radiation with the mean target plane were 22.5°. Only a single laser shot was made for each cavity. Special care was taken to ensure precise adjustment of the femtosecond laser beam waste and a single cavity. We placed an optical pin-diode to monitor the reflection from the target 45° from the direction of the specular reflection. We found that under irradiation of a cavity with femtosecond laser pulse, reflection was diffusive, whereas specular literally unperturbed reflection appeared if the laser pulse interacted with the flat part of the target surface. In the first case, the pin-diode signal increased dramatically while being negligible in the latter one.

The other method of surface modification consists of crater formation by laser pulse. This is much easier to

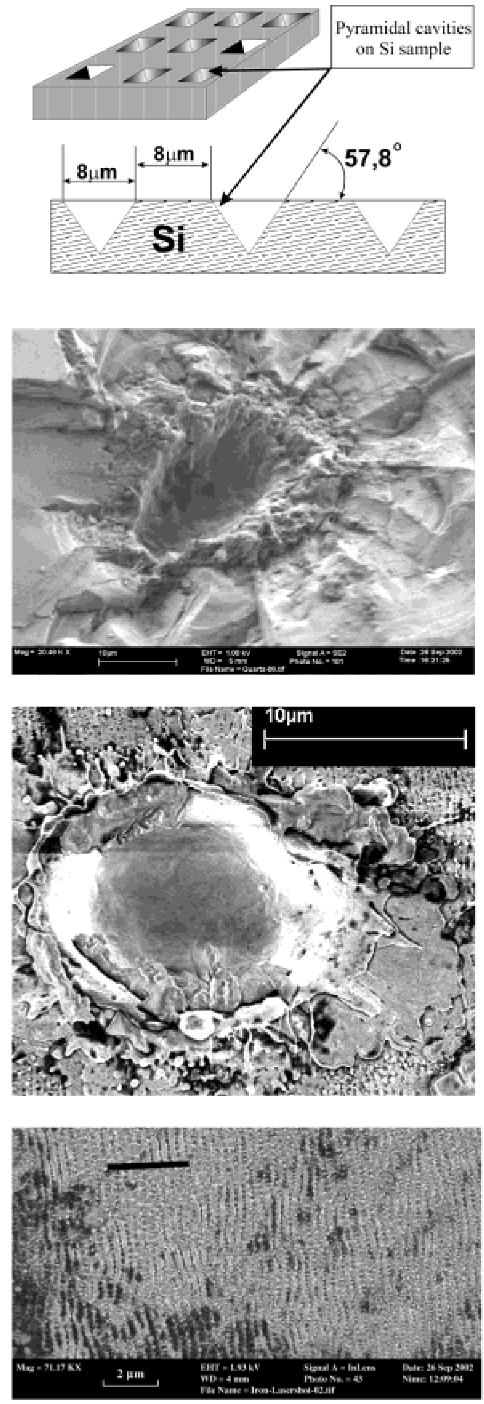

Fig. 2. Targets with modified surface. a: Schematic view of pyramidal cavities at silicon substrate. b: SEM image of the laser-induced crater at crystalline quartz. c: SEM image of the laser-induced crater at iron surface. d: SEM image of laser-induced grating at iron surface.

realize directly during a laser–plasma experiment, because no special target processing is necessary in advance. In the commonly used experimental scheme, the target is moved, providing a fresh flat surface for each laser shot. In contrast, we did not move the target from shot to shot. Even a single laser shot formed a crater with a well-developed surface (see

Fig. 2b). In our experiments, the first laser shot was used to prepare a crater with structured walls and bottom. The next shot created plasma from these structures and formed new structures also (both laser pulses had a 45° angle of incidence to the mean surface). It was checked (measuring hard X-ray yield from the plasma) that up to 4–5 shots can be applied without target displacement. Figure 2b shows a SEM image of the crater produced at the flat surface of crystalline quartz at an intensity of about 20 PW/cm^2. The crater diameter is slightly larger than the plasma spot size. A rough surface is formed on the periphery and bottom of the crater. It should be noticed here that a well-structured surface was formed in this way provided crystalline targets were used (see experimental data in the next section). We found that with amorphous and polycrystalline (metal) targets no crater surface structuring appeared. This can be seen from Figure 2c presenting a SEM image of a laser-induced crater at the iron surface. We think that structuring occurs only at crystalline targets, probably due to cracking of a target material.

For metallic targets we developed the other method of surface structuring. It is well known that generation of periodic structures at the target surface can be obtained by exposing a surface to laser radiation with an intensity slightly exceeding the melting threshold. This happens due to interference between the surface electromagnetic wave (SEW) exited by vacuum pump wave and the pump wave itself (Volkov *et al.*, 2003; Gauthier *et al.*, 1995). In our experiments *p*-polarized femtosecond laser radiation was used to generate periodic structures at the surface of an iron target. The intensity of the femtosecond laser pulse was reduced to $\sim 10^{12}$ W/cm^2 by shifting the focusing objective by 300 μm from the tight focusing position toward a target. This intensity was slightly higher than the threshold of target melting. The angle of incidence at the target was in this case 45°. The result of 15 successive shots is shown in Figure 2d. After this procedure the focusing lens was shifted to the precise focusing position and single laser shot was made to create laser plasma.

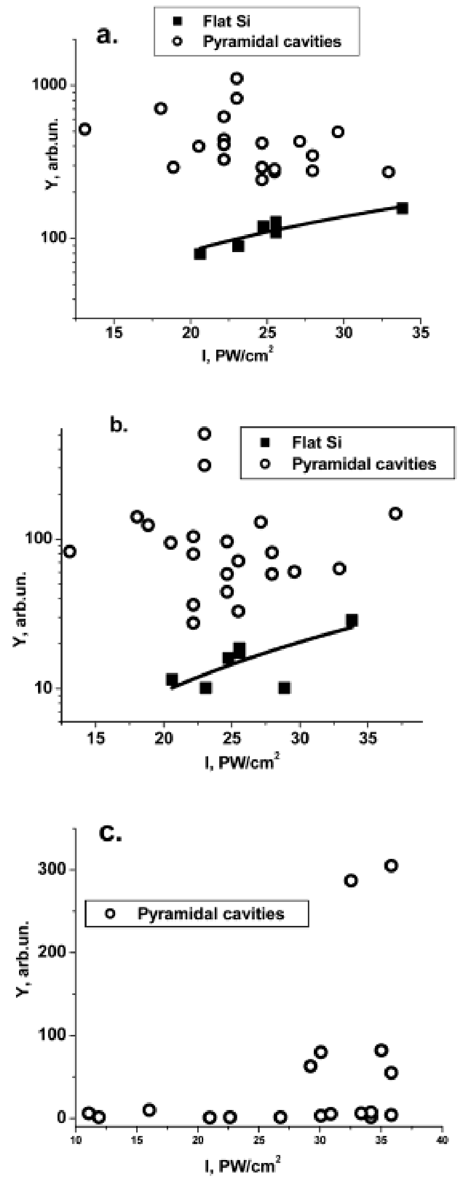

Fig. 3. The dependence of X-ray output in different energy ranges from laser pulse intensity for flat and electrochemically modified Si target (pyramidal cavities on Si substrate). Energy ranges: a: above 2 keV, b: above 9 keV, c: above 36 keV (on flat target no quanta above 36 keV were detected).

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Pyramidal cavities

Experimental data on X-ray yield in different spectral ranges is presented in Figure 3. For this target we observed a prominent increase of the hard X-ray yield above 2 keV compared with that from flat silicon. For example, above 9 keV we estimated that X-ray yield grow up 7 ± 3 times in comparison with the flat Si target. Moreover, X-ray quanta with energies exceeding 36 keV were detected only for the target with pyramidal cavities. Note that X-ray yield looks nearly independent on the laser pulse energy (at least in the laser pulse energy diapason used in our experiments), whereas raw data is very disperse. We attributed such a behavior to the imperfect adjustment of the center of the pyramidal cavity (8×8 μm base) with laser focal spot (3 μm). In fact quality of this adjustment depends not only on the quality of mechanical movement and positioning of the target but also on the laser beam stochastic angle deviations from shot to shot causing 3–5 μm jumps of the focal point.

Noticeable changes were also detected in the value of the hot electron temperature for the target with pyramidal cavities. Figure 4 presents the distribution of the hot electron temperature deduced from our X-ray data for the flat silicon and the silicon specimen with pyramidal cavities. The average intensity was $I = 20 \pm 10$ PW/cm^2. For the electrochemically modified sample, the mean hot electron temperature

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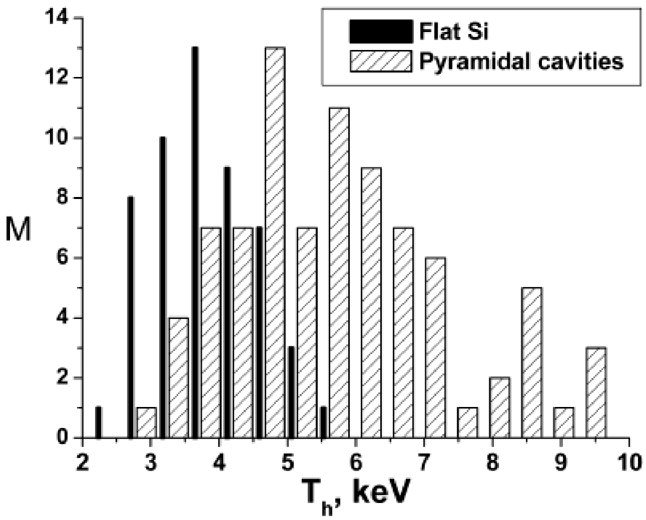

Fig. 4. Distribution of the number of realizations *M* with given temperature T_h for the flat silicon target and pyramidal cavities.

was 5.8 ± 1.5 keV, compared with the 3.7 ± 0.7 keV value measured for the flat Si target. In the case of the flat silicon target, width of the distribution in Figure 4 is due to fluctuations of such laser pulse parameters as pulse duration and energy. In the case of a target with pyramidal cavities the distribution is much wider. This might be attributed to the same obstacle mentioned in the previous paragraph: In each realization, the focal spot falls into different parts of a pyramid (vertex, edge, or rib). It should be noted that in the case of a pyramidal cavity, both X-ray yield and hot electron temperature do not depend on the polarization state of incident beam, as the same values were obtained both for *s*- and *p*-polarized radiation.

A prominent increase in hard X-ray yield and hot electron temperature for targets with pyramidal cavities might be attributed to an increase of the effective absorption and the local light intensity due to multiple reflections from cavity walls. To get at least a qualitative explanation of our results numerical modeling should be done by a two-dimensional (2D) PIC code. Such calculations are underway now. Anyway, because the contrast of our pulses is high, major mechanisms responsible for the hot electron production in our experiments are vacuum heating (Brunel, 1987) and the anomalous skin effect (Andreev *et al.*, 1992; Gibbon *et al.*, 1992). Experimental evidence comes from the two findings: (1) the hot electron temperature does not depend on the polarization state of the light for targets with pyramidal cavities, being strongly dependent on this quantity for the flat one, and (2) the scaling of the hot electron temperature for flat Si targets deduced in our experiments was $T \sim (I\lambda)^{0.6-0.8}$ (Golishnikov *et al.*, 2001), which is close to the theoretical scaling $T \sim (I\lambda)^{2/3}$ for these mechanisms of hot electron generation and does not agree with $T \sim (I\lambda)^{1/3}$ scaling that is a distinctive feature of resonant absorption (Gibbon & Forster, 1996).

3.2. Crater formation by laser pulse

In Golishnikov *et al.* (2001), we described experiments with laser-induced craters on a silicon surface. Here we systematized our study for other LM materials (see Table 1). From this table one can see that an increase in hot electron temperature is the characteristic feature only of crystalline targets. To check this we made a comparative study of fused (amorphous) and crystalline quartz as a target material (see Fig. 5), because the only difference between the two is in their crystalline structure. For the LM crystalline quartz, prominent growth of the mean hot electron temperature in

Table 1. *Hot electron temperature for different target materials: $I \sim (1.5–3.5) * 10^{16}$ W/cm^2*

Flat target		Target with modified surface	
Target material	T_{hot} (keV)	Type of surface modification	T_{hot} (keV)
Crystalline targets			
Si	3.7 ± 0.8	Electrochemical, pyramidal cavities	5.8 ± 1.6
		Electrochemical, porous Si (Volkov *et al.*, 1998)	5.1 ± 1.7
		Laser-induced crater	8 ± 3
KDP	3.7 ± 0.7	Laser-induced crater	7 ± 3
Ge	4.9 ± 1.4	Laser-induced crater	6.3 ± 2.1
SiO_2 (crystalline)	4.8 ± 1.0	Laser-induced crater	7.5 ± 2.2
Amorphous and polycrystalline targets			
SiO_2 (fused)	4.1 ± 1.7	Laser-induced crater	4.4 ± 1.6
TiD	6.4 ± 2.2	Laser-induced crater	10 ± 4
Zr	5.2 ± 1.8	Laser-induced crater	5.7 ± 1.4
Pd	7 ± 2	Laser-induced crater	10 ± 3
W	7 ± 2	Laser-induced crater	10 ± 4
Fe	4.7 ± 1.2	Laser-induced crater	5.3 ± 1.2
		Laser-induced gratings	8 ± 2

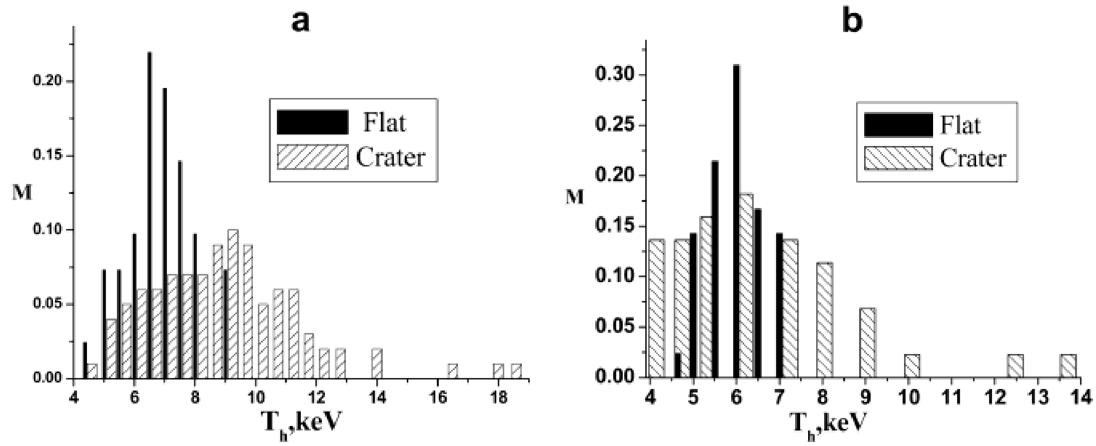

Fig. 5. Distribution of the fraction M of realizations with given temperature T_h for the quartz target. a: Crystalline quartz; b: fused quartz.

comparison with the flat one is clearly observed (Fig. 5a), whereas for the LM fused quartz, target distribution of the hot electron temperature from shot to shot is almost the same as for the flat fused quartz (Fig. 5b). At the same time the distribution of the hot electron temperature for LM quartz is wider independently of the initial structure of a flat target, and this is a characteristic feature for all LM targets tested. The mean value of the hot electron temperature was about 4.5 keV (see Table 1) for both the flat samples and the LM fused quartz, whereas this quantity reaches 7.5 ± 2.2 keV for LM crystalline quartz.

Although for crystalline LM targets the X-ray spectra are shifted toward higher quanta energies due to the increase in the hot electron temperature, the absolute value of the hard X-ray yield above 2 keV, 9 keV, or 13 keV does not increase, and sometimes even falls. The only spectral range where an increase always takes place was for quanta energies above 36 keV. In this spectral range detectors working in a single quantum detection regime registered no signal for any flat target, but did for all the crystalline LM targets. Note that a huge enhancement in hard X-ray yield observed by Hironaka *et al.* (1999) using a Cu target is to be attributed to the unsuitable *s*-polarization state of the first laser shot producing the crater.

Hence our findings confirm that structuring occurs only with crystalline targets probably due to cracking of the crater walls (see also Fig. 2b). This structuring may lead to efficient absorption of the laser energy, thermal flux suppression and, possible local field enhancement in the well-developed surface of the crater walls (Gordienko *et al.*, 2003).

3.3. Laser-induced gratings

Experiments with laser-induced gratings at the iron surface were described in detail by Volkov *et al.* (2003). In brief, here we observed a two- to threefold increase of the X-ray yield above 7 keV in comparison with the flat iron target. In the spectral range above 20 keV the resonant grating provides an even higher, fourfold increase in the X-ray output. This indicates that the temperature of the hot electrons was also higher for the target with laser-induced grating. Our measurements confirmed that the hot electron temperature at laser-induced gratings was 1.4 ± 0.2 times higher than the one at the flat iron surface (see Table 1). This phenomenon was explained by the local field enhancement under interaction of the femtosecond laser pulse with resonant gratings (Volkov *et al.*, 2003).

4. CONCLUSIONS

Modification of the structure of the surface allows one to control the process of hot electron generation under femtosecond laser plasma interaction at moderate intensities of 10^{16} W/cm^2. Namely, we observed an increase in the temperature of hot electrons and a shifting of the X-ray spectra toward shorter wavelengths for targets with a structured surface layer. The efficiency of hard X-ray production also can be enhanced with our approach, but this feature strongly depends on the surface treatment procedures.

We observed a significant hard X-ray yield increase from plasma created at the surface of an electrochemically modified solid-state target (pyramidal cavities). Hot electron temperature enhancement was also found for these targets. Even more interesting results might be obtained using the same structures but with a shorter structure period of the order of 1 μm. This also should reduce instability due to misalignment between the laser focal spot and cavity.

Creating craters with a single femtosecond laser shot at the crystalline materials surface we also achieved an increase in the hot electron temperature. Laser modification and structuring of the target surface (craters on crystalline targets and gratings on metal targets) looks very attractive because all the processing procedures take place *in situ*.

The obtained increase in hot electron temperature can be used to enhance excitation efficiency for low-lying nuclear levels. It should be noted that such an increase in hot electron temperature should also provide for higher energies

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of fast ions emitted by plasma (Volkov *et al.*, 2002). This allowed us to observe at moderate intensities thermonuclear neutrons from a D-enriched Ti target with laser-induced craters (Volkov *et al.*, 2000; Chutko *et al.*, 2001).

Recent studies (Salzmann *et al.*, 2002; Reich *et al.*, 2001) found that for fixed laser energy and intensity, there is an optimum electron temperature to generate Kα photons in material of a given atomic number Z. The modification of the target surface allows us to control hot electron temperature and thus may lead to more efficient Kα production.

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