Intensity clamping and controlled efficiency of X-ray generation under femtosecond laser interaction with nanostructured target in air and helium.

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Abstract. We have performed theoretical calculations aimed at modeling the propagation dynamics of a focused femtosecond laser beam in a nonlinear gas medium. It was obtained that when helium is blown into the focal area, the intensity clamping level is about 2.5 times higher than when air is present. The conditions for optimal placement of the target relative to the position of the vacuum focus and, accordingly, allowing to achieve maximum intensity are found. It was found that under interaction in air femtosecond Cr:forsterite laser (\( \lambda = 1.24\ \mu \text{m}, E < 1\ \text{mJ}, 100\ \text{fs} \)) with nanostructured iron containing tape the intensity clamping effect is observed. Replacing the air medium with helium leads to essentially increase in line (6.4keV) X-ray yield. Increasing the pulse duration to 300fs led to an increase in the X-ray output twice and reached \( 3 \times 10^5 \) photons/pulse in solid angle.

1. Introduction

Nowadays, laser based sources of ultrashort X-rays have found very important applications in tasks of X-ray ultrafast time-resolved spectroscopy and X-ray diffraction [1-3]. For this applications source of short x-ray pulse with a large photon number of \( 10^8 \) ph/pulse in solid angle is required. Simplest way to get X-rays is to focus high intensity laser on solid target. Femtosecond laser generation of X-rays arising as a result of the stimulated production of high-energy electrons is largely associated with the use of schemes for interacting with a target under vacuum conditions at high laser intensities [4,5]. The femtosecond laser-solids ultrafast X-ray source operating at atmospheric pressure is a convenient device for ultrafast time-resolved measurements and biomedical application. On the one hand the X-ray source scheme can be reduced by eliminating the vacuum system that is usually used. On the other hand vacuum-free X-ray system prevents the known debris problem. In gas environment debris cannot reach the focusing lens or mirror, which is placed not far from the solid target. The presence of near-surface gas serves as a filter that does...
not allow contamination and ultimately damage to the surrounding optics. Besides, in a vacuum, there are also problems associated with the operation of the target.

So there is a need for an efficient X-ray source capable of operating in air with a tabletop femtosecond laser. In that conditions ionization of air molecules in the near-surface area of the target is accompanied by depletion of laser energy, as well as self-induced defocusing of the laser beam [6]. These processes with increasing laser energy cause a restriction (clamping) of the laser beam intensity level (compared to the “vacuum” level). The conversion efficiency to X-ray under the conditions of laser-target interaction in a gas environment is limited at a lower level than under vacuum conditions. Known, that it is possible to reduce energy loses and increase X-ray yield using local blowing gas with high ionization potential (helium) into the interaction area [7,8]. In this connection the wavelength influence and its effect on the nonlinear propagation regime and energy delivery of focused femtosecond laser radiation to the target in air and helium has not been previously considered.

It is known, that using structured targets such as porous materials or different periodically structured targets leads to an increase in the absorption of laser energy by the target and correspondingly enhancing X-ray yield from near-surface plasma [9,10]. Considerable local field enhancement and increasing energy coupling may appear due to laser generation of surface electromagnetic waves (SEWs), which results in more efficient X-ray production. Note that SEW generation depends on period of structure of the target, laser wavelength, polarization [10-13]. Besides for applications of laser-plasma x-ray sources, it is necessary to have a renewable state of the target surface for each new interaction act. One of the ways to solve the problem is the use of such targets as rotating metal disc [14], moving metal wire or tape [15-17]. Well-known structured target in which the appearance of a surface electromagnetic wave can be laser stimulated and the surface of which can be simply updated to each act of laser action is a commercially available magnetic tape. [14].

The aim of this work is to theoretically study the features of nonlinear propagation of focused intense femtosecond laser beam in air and helium as a function of the wavelength and laser pulse duration, as well as to experimentally investigate the conditions for effective conversion to x-ray that occurs when a focused beam of a femtosecond mJ-level Cr:forsterite laser impact to a nanostructured iron containing tape in air and helium.

2. Theoretical modeling

We have performed theoretical calculations aimed at modeling the nonlinear propagation dynamics of a focused laser beam in a nonlinear gas medium. The propagation of a femtosecond laser pulse in gas mixture was numerically solved using a system of recognized equations. The equation for the electromagnetic field takes into account diffraction, dispersion, Kerr and plasma nonlinearities, the self-steepening of the pulse, and the loss of energy for ionization.

Clamping is a well-known phenomenon that manifests itself in limiting the intensity during the production and propagation of the filament in gas. An increase in the initial laser energy and/or intensity leads to a change in such filament parameters as its beginning of formation and the diameter, but the intensity of the filament changes slightly. Can we expect to limit the intensity under the conditions of the plasma existence, but the absence of a filament, i.e. the absence of an extended plasma channel with a length several times greater than the Rayleigh length? We consider the following experimental scheme. A laser beam with a diameter (at the level of 1/e²) of 2.5 mm is focused by a lens (F= 6 cm) on the target situated in a helium (or air) medium at a pressure of 1 bar. It was theoretically obtained (see Figure.1) dependence of intensity on distance z for laser wavelength of 0.8μm and 1.24 μm in air and He. We see that intensity clamping level is about 2.5 times higher for helium compare with air. This is due to the high ionization potential in He (24.6
3 eV) compared to air (14.6 eV). Thus, in He, a much higher intensity is needed to create the plasma
concentration necessary to limit the intensity, i.e., to defocus the laser beam.

![Figure 1](image1.png)

**Figure 1.** Dependence of intensity on distance $z$ for different laser wavelengths in helium. Pulse duration 100 fs, energy 1 mJ. $z=6$ cm corresponds to the focus position.

![Figure 2](image2.png)

**Figure 2.** Dependence of parameter $I \lambda^2$ from distance $z$ for different laser wavelengths in helium. Pulse duration 100 fs, energy 1 mJ. $z=6$ cm corresponds to the focus position.

The conditions for optimal placement of the target relative to the focus position in a vacuum and, consequently, achieving maximum intensity can also be seen in Figure 1. It was theoretically shown that an increase in the wavelength from 800 nm to 4000 nm with comparable laser parameters and focusing mode provides an increase in the product $I \lambda^2$ by almost an order of the magnitude (see Figure 2). As a result of the calculation, it was shown that with increasing the pulse duration while maintaining the input energy the maximum intensity as a result of clamping reduced slightly (see Figure 3). The increase in pulse duration leads to a significant increase in energy density (see Figure 4) on the surface of the target and, as a consequence, should lead to an increase in the output of X-ray photons.

![Figure 3](image3.png)

**Figure 3.** Dependence of maximum intensity on pulse duration in He

![Figure 4](image4.png)

**Figure 4.** Dependence of energy density on distance $z$ at different pulse durations in He. $z=6$ cm corresponds to focus position.
(1) $\tau_p=500\text{fs}$, (2) $\tau_p=300\text{fs}$, (3) $\tau_p=200\text{fs}$, and (4) $\tau_p=100\text{fs}$. Laser wavelength 1.24 $\mu$m

Thus, the made theoretical analysis showed that:

a - the intensity restriction occurs even in the absence of an evident filament and the clamping intensity strongly depends on the ionization potential of the medium;

b - the conditions are revealed under which it is possible to obtain high intensity values (He blowing) and at such high intensity values to increase the fluence (number of photons) at the target by increasing the pulse duration.

3. Experimental setup and results

The experiments are conducted with Cr:forsterite laser system at 10 Hz repetition rate. The setup supplies near-IR-pump pulses of 100 fs at 1240 nm with pulse energy of 1 mJ. The laser beam was focused on the moving tape target surface by a lens with a focal length $F = 6\text{cm}$ to the spot with the diameter of 10 $\mu$m (NA=0.1). So called vacuum intensity was about $10^{16}\text{W/cm}^2$. The moving tape target was placed in air. A commercially available cassette player is used as a moving tape target. The tape speed of 4.76 cm/c can provide renewing surface up to 1 kHz regime of laser interaction. The tape contains a thin top magnetic layer supported by a thicker base film. Top layer of the tape consists of iron oxides $\text{Fe}_2\text{O}_3$ in the form of nanocylinders (10-20 mm diameter and 100-1000 mm height). The thickness of the magnetic layer of 3 microns, the thickness of the substrate 16 microns. The target porosity is about 20%. Nanocylinders are oriented mainly in the longitudinal direction.

Therefore, it was possible to expect the manifestation of polarization dependence in the process of interaction of laser radiation with the tape target. The polarization state was changed with a $\lambda/2$ plate. The schematic layout of the experimental setup is shown in Figure 5. We can perform experiment also in helium gas. To do this, was made a small chamber in the form of a piece of silicone hose with maylar window of 20 $\mu$m to monitor X-rays from the nearsurface laser induced plasma. The chamber can be filled with helium by using small nozzles with a diameter of 0.4 mm. The consumption of helium should be less than 0.25 l/min. To detect the X-rays, we used PMT 9107FLB equipped with a NaI scintillator and beryllium filter and Amptek spectrometer, working in the single-photon mode, which was placed at the distance of 8-11 cm from the target.
**Figure 5.** Table top experimental system for repetition rate X-ray production from moving nanostructured tape 1 is the Cr-forsterite femtosecond laser, 2 is the λ/2 plate, 3 is the lens, 4 is the small chamber, 5 is the reducer, 6 is the tape, 7 is the image of the tape, 8 is the PMT, 9 is the X-ray spectrometer Amptek.

To determine the conditions under which the maximum output of X-ray from the laser induced microplasma on the surface of the structured tape is achieved, the X-ray yield depending on the state of polarization of the laser radiation was studied. It was obtained that X-ray yield from the nanostructured tape is 2 times higher when using P-polarized pulses. Therefore, P-polarized laser radiation was used in subsequent experiments.

At the next stage, experimental measurements of the temperature of hot electrons in air and helium were carried out depending on the laser energy (see Figure 6). The electron temperature was measured as an approximation of the X-ray bremsstrahlung spectrum (an example of the spectrum in helium and approximation is shown in Figure 7) As can be seen from the figure, the electron temperature in air stabilizes at a level of 2.5 ± 0.5 keV at an energy of 0.3 μJ and remains unchanged with an increase in energy to 1 mJ, which indicates the stabilization of the intensity. At the same time, the temperature of hot electrons in helium was the same as in air at 0.3 μJ and was 1.5-2 times higher at 1 mJ, and reach the value of 6 ± 1 keV. This indicates the fact that in He the intensity stabilize at higher level than in air as was predicted theoretically.

![Figure 6. Dependence of hot electron temperature in air and He from laser energy.](image)

At the next stage, we measured the X-ray yield. Figure 8 shows the dependence of the number of K-α X-ray photons (6.4 keV) in air and helium from the laser energy. The behavior of the X-ray yield correlates with the electron temperature: X-ray yield in air really does not change with increasing laser energy and at a maximum energy of 1 mJ it achieve 2.5*10⁴ photons/pulse in solid angle. With helium blowing, the X-ray yield turns out to be 6 times greater and amounts to 1.3*10⁵ photons/pulse in solid angle. Considering the dependence of X-ray yield from laser intensity as

\[ Y_{X\text{-ray}} \sim A \exp(-E/T_{\text{hot}}) \]

\[ T_{\text{hot}} \sim 5 \text{ keV} \]

![Figure 7. Fe K-α X-ray spectra in He.](image)

\[ Y_{X\text{-ray}} \sim I^2 \] [17], it indicates growing the intensity level 2.5 times as predicted theoretically. A similar dependence was obtained for the integral energy of X-ray radiation in the range of more than 3 keV. In this case, X-ray yield in helium was 20 times higher than in helium. This value was higher than increasing in K-α X-ray yield. The explanation of this may be the following: with an increase in intensity in He the number of high-energy quanta increases, which make the main contribution to the integral signal output.
At the next stage we performed experiments with chirped pulses of 300 fs. As shown in Figure 8, Figure 9 this result in 2-fold increase in the K-α X-ray yield (as predicted by the theory). Since an increase in the duration should not lead to a significant change in the intensity, the temperature of the hot electrons remained unchanged, and, accordingly, the proportion of high-energy quanta also remained in this case. Therefore, the increase in X-ray yield turned out to be the same both in the case of characteristic photons and for an integral energy of more than 3 keV. It should be noted that in the performed experiments optimal placement of the target relative to the position of the vacuum focus and, accordingly in order to achieve the maximum intensity was found. Besides the enhancing the fluence in the conditions of the clamping effect with an increase in the pulse duration and focusing conditions is obtained. Thus, the developed X-ray source with the number of photons more than $10^6$ photons/pulse can be used for time-resolved experiments using polycapillary optics. Further prospects for increasing the number of photons are associated with an increase in the laser energy and a simultaneous increase in the duration of the laser pulse.

4. Conclusions

1. It is theoretically shown and experimentally confirmed that the propagation of a focused femtosecond laser beam in a gas medium (air, helium) is accompanied by saturation of the intensity (clamping manifestation), the value of which depends on the ionization potential of atoms or molecules of the medium.
2. It was found that when a focused (NA = 0.1) polarized radiation of a femtosecond laser on Cr: forsterite (energy 1 mJ, pulse duration 100 fs) is exposed to iron-containing nanostructured tape in air, the laser beam energy density is stabilized at the level of 16 J/cm², temperature of hot electrons is limited at a level of 2.4 keV. In this case, the yield of characteristic X-ray (6.4 keV) is limited to about $2.5 \times 10^4$ photons / pulse in solid angle.
3. A non-vacuum approach has been proposed, which makes it possible to optimize the obtaining of a high yield of characteristic X-ray radiation under interaction of intense repetition rate femtosecond laser with nanostructured tape.
4. It was found that when a femtosecond Cr:forsterite laser (energy up to 1 mJ, 100 fs, NA = 0.1) is exposed nanostructured tape in He leads to an increase in X-ray yield with $1.3 \times 10^5$ photons/pulse (in solid angle), electron temperature grows 1.5-2.0 times and achieved 6±2 keV.
5. It was observed that under chirped femtosecond Cr:forsterite laser (energy up to 1 mJ, 300fs, NA = 0.1) interaction with nanostructured iron containing tape in He line X-ray yield grows two times and achieve 2.8×10⁵ photons/pulse (in solid angle).

6. It was theoretically predicted that using mid-IR laser due to product (Iλ²) one can obtained essential enhancing laser beam energy density and as a result increasing in X-ray yield.

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References

[1] Fetisov G.V, 2020 UFN 190 2-36 (in Russian)
[2] O’Neil, G C, Miaja-Avila L, Joe Y I, Alpert B K, Balasubramanian M, Sagar D M, Dorisew W, Fowler J W, Fullagar W K, Chen N, Hilton G C, Jimenez R, Ravel B, Reintsema C. D., Schmidt D R, Silverman K L, Swetz D S, Uhlig J and Ullom J N 2017 J of Phys. Chem. Letters 8 1099-04
[3] Chergui M and Collet R 2017 J Chem. Rev 117 11025-65
[4] Li M, Huang K, Chen L, Yan W, Tao M, Zhao J, Ma Y, Li Y and Zhang J. 2017 J Rad. Phys. and Chem. 137 78-82.
[5] Martin L, Benlliure J, Cortina D, Gonzales D, Llerena J J, Peña J and Ruiz C 2019 Commissioning of a laser-plasma x-ray micro-focus source for phase contrast imaging. In Relativistic Plasma Waves and Particle Beams as Coherent and Incoherent Radiation Sources III 11036 110360J
[6] Bukin V V, Vorob’ev N S, Garnov S V, Konov V I, Lozovoi V I, Malyutin A A, Shchelev M Ya and Yatskovskii I S 2006 J Quantum Electronics, 36 638
[7] Hou B, Easter J, Krushelnick A M and Nees, J. A. 2008. J Optics Express, 16 17695-705
[8] Hada M and Matsuo J 2010 J App. Phys B 99 173-9.
[9] Murnane M M, Kapteyn H C, Gordon S P, Bokor J, Glytsis E N and Falcone R W 1993 J App. Phys. Let. 62 1068-70.
[10] Wang H, Li Z and Chen Z 2018 J App Phys B, 124 172
[11] Gordienko V M and Savel’ev A B 1999 UFN 169 78-80
[12] Gordienko V M, Mikheev P M, Savelev A B, 2001 J Laser Phys 11 600-5
[13] Samsonova Z, Höfer S, Hollinger R, Kämpfer T, Uschmann I, Röder T, Trefflieh L, Rosmej O, Förster E, Ronning C, Kartashov D and Spielmann C 2018. J Applied Sciences, 8 1728.
[14] Rathore R, Arora V, Singhal H, Mandal T, Chakera J A and Naik P A 2017 J of Laser and Particle Beams, 35 442-9.
[15] Kutzner J, Silies M, Witting T, Tsilimis G and Zacharias H 2004 J Appl. Phys. B 78 949-55.
[16] Son J G, Hwang, B J, Seo O, Kim J M and Ko D K 2018 J of the Korean Phys. Society 73 1834-9.
[17] Arora V, Naik P A, Chakera J A, Bagchi S, Tayyab M and Gupta P D 2014 J AIP Advances 4 047106.