Kα X-ray Emission from Nanowire Cu Targets Driven by Femtosecond Laser Pulses for X-ray Conversion and Backlight Imaging

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ABSTRACT: A high-quality X-ray source was proposed by modifying the target material structure characteristics driven by ultrahigh laser energy. The experiments were performed on the Ti:sapphire femtosecond laser beam device (4.3–6 J, 30 fs), one of the three XG-III lasers in Laser Fusion Research Center of China Academy of Engineering Physics. The femtosecond laser beam drove the nanowire copper material with an average length of 18–50 μm and a diameter of about 260 nm. A single-photon counting charge-coupled device was employed to measure the copper Kα X-ray emission of the nanowire and foil targets. A clear maximum photon yield of the nanowire target was calculated to be 3.6 × 10⁸ photons sr⁻¹ s⁻¹, the conversion efficiency was up to 0.0087%, and the average yield was 2.5 times that of the copper foil targets. In addition, by using a pinhole imaging method of φ10 μm, the minimum full width at half maximum spot size of the X-ray source was calculated in the range of 85–240 μm, which was similar to that of the copper foil material with a long radius of 170 μm and a short radius of 63 μm. The experimental data illustrate that the nanowire has the potential to enhance the energy absorption of femtosecond laser for X-ray conversion and backlight imaging.

1. INTRODUCTION

Ultrashort and intense laser pulses are focused on a solid target on a time scale of hundreds of femtoseconds to tens of picoseconds, and the laser can produce plasma to generate X-rays via bremsstrahlung, K-shell ionization, and hot-electron.1,2 There are many characteristics like that of the ultrashort femtosecond laser pulse.3 Ewald et al.4 analyzed that these unique multi-keV X-rays were originated from high-temperature plasmas with anomalous properties such as being very bright (up to 10¹² of Ti Kα), of extremely short duration at the range of a 100–5000 fs, and even having a small focal spot size of only 8 μm.5 Because of the design of many different materials, the tremendous potential of ultrashort X-ray techniques has initiated the development of ultrashort X-ray sources. Therefore, the sources, with a high spatial coherence, may be of interest for many applications, such as the diagnostic tools used to study the structure and performance of materials and hot compressed matter,6 as well as served as a pump source for X-ray lasers7 to study ultrafast dynamics.8 Compared with synchrotron sources, ultrashort laser-generated plasma X-ray sources, with very high instantaneous peak brightness, small size, and low cost, are a more economical and compact alternative to ultrashort hard X-ray sources. Workman et al.9 reported a point-backlighter X-ray source with a spatial resolution of 10 μm. Chen et al.10 proposed a conversion efficiency (CE) up to 0.01% and an X-ray photonic yield of 1.3 × 10⁷ photons sr⁻¹ s⁻¹ on the 20 TW laser system of the Extreme Light (XL-II). Chen et al.11 analyzed the transformation of a CE laser into fast electron between 20 and 40%. Changing the structure of the material is a potential and more economical method to improve CE, which can increase the absorption of laser energy. Serbanescu et al.12 proposed that the Kα X-ray CE produced by laser-driven Cu target was 0.0032% and the minimum spot size was up to 8 μm. The experiments on copper surface modification such as Cu nanowire, nanoparticle,13 nanoparticle, and nanorod arrays14 showed that nanostructure material could improve the CE of the hot electron to X-ray source.15

In this paper, we reported an experimental improvement of the X-ray yield, as well as the spot, which was related to the
Above the laser intensity of $1.9 \times 10^{18}$ W/cm$^2$ on XG-III facility, the improvement of the material surface had a potential significant effect on the K$_\alpha$ X-ray yield and CE. The results showed that copper K$_\alpha$ X-ray could be efficiently obtained from nanostructured material driven by ultrashort laser pulses on intensity laser systems. X-ray K$_\alpha$ yield of $3.6 \times 10^8$ photons sr$^{-1}$ s$^{-1}$ and CE of 0.0087% were produced at ultrashort femtosecond laser energy ($4.3-6$ J, 30 fs). The experimental data showed that copper X-ray K$_\alpha$ of the surface-modified nanowire targets could produce higher CE, and the focal spot of copper K$_\alpha$ X-ray source ranging from 85 to 240 μm was similar to that of the copper foil target of 10 μm, which would hopefully be diagnosed by X-ray microscopy$^{18}$ and electron distribution$^{19}$ with higher time-resolved diffraction.

### 2. EXPERIMENTAL DETAILS AND NANOWIRE MATERIAL

#### 2.1. Experimental System

The experiments were performed on an XG-III laser device located at Laser Fusion Research Center (LFRC) of China Academy of Engineering...
Physics (CAEP). The device had three different wavelength laser beams, which could output nanosecond, picosecond, and femtosecond simultaneously, with wavelengths of 527, 1053, and 800 nm, respectively. The experiment only used the horizontal femtosecond laser beamline Ti:sapphire laser system that mainly consisted of four parts: the front-end system, two amplifiers (50 TW/500 TW), and a vacuum compressor. The installation and diagnostic layout of nanowire and foil targets are shown in Figure 1. The femtosecond laser beam with a pulse of \( t_0 = 30 \) fs was focused through an \( f/10 \) off-axis parabolic Au mirror, with the inclined incident on the foil targets and nanowire targets at 35 and 45°, respectively. In case it was necessary to generate plasma, the main pulse of this device was delivered with a maximum intensity of \( 1 \times 10^{23} \) W/cm² and the laser intensity contrast was \( 10^5 \). This experimental intensity of the femtosecond laser is shown in Table 1. A 40 \( \mu \)m diameter laser focal spot already contained approximately 20% of the energy in the pulse. The pulse energy range of this experiment was 4.3–6 J.

Single-photon counting (PI LCX CCD) method was employed to measure bremsstrahlung and Kr X-ray radiation yield on each shot ranging from 2 to 30 keV. The charge-coupled device (CCD) vertical direction was 23.7° to the reference surface of the vacuum target chamber and 64.1° to the laser incident direction. A 0.6 T magnet was set between the LCX and target to change the electronic trail. A filter consisting of 120 \( \mu \)m Cu and 500 \( \mu \)m Be was used in front of the LCX CCD. For Cu Kr X-rays, the photon detection efficiency of the LCX CCD was only 45%.

To diagnose the Cu Kr focal spot, a PIXIS-XB CCD system was used with a 10 \( \mu \)m pinhole. In this experiment, the pinhole camera system was composed of 20 \( \mu \)m thick tantalum with a 5 × 6 matrix pinhole and PIXIS-XB CCD, the size of which was 13 \( \mu \)m/pix. The filter used was a 5 \( \mu \)m thick aluminum film to shield energy X-rays below 4 keV.

The backlight imaging experiment was also performed with a resolution plate and 13.5 \( \mu \)m/pixel PI MTE CCD. Steel 45 # was the designed flange adapter for MTE CCD. In front of the MTE CCD, a filter consisting of 20 \( \mu \)m Al and 400 \( \mu \)m Be was employed with the transmittance rate of 0.69. Where X-ray beam passes through a homogenous material, the ballistically transmitted intensity follows the well-known Beer–Lambert law \( I/I_0 = e^{-\mu x} \), where \( I \) is the intensity of the X-rays passing through the sample and \( I_0 \) is the intensity without the sample in vacuum. The parameter \( \mu \) is the absorption rate of films, and \( x \) is the thickness of the sample.

2.2. Nanowire Cu Target Material. Among the advanced ion or laser-wake field acceleration schemes, the target coated with a low-density, such as a near-critical, nanostructured layer is emerging as a promising strategy. The interaction of ultra-intense laser and plasma with near-critical density has the characteristics of strong coupling and effective energy absorption. The low-density layer can be a controlled preplasma or a solid nanostructured material with ultrahigh porosity, which allows for a high absorption efficiency of the laser energy by the hot electron population for a flat foil simple. Passoni et al. mentioned insights on the role played by a nanostructure upon the interaction with a superintense laser pulse.

Cu nanowire, with a thickness of 30 \( \mu \)m, was fabricated by the dealloying process as the target of laser irradiation for X-ray generation and was characterized by a simple template preparation process, cheap raw materials, highly ordered hole array, and adjustable hole diameter within a certain range. As shown in Figure 2, the results of the nanowire target based on scanning electron microscopy (SEM) indicated that the structure with more surface area would perform better in laser energy absorption. The SEM micrograph was a commercial anodic aluminum oxide (AAO) template with a nominal pore size of 200 nm. The holes in the template were not in a regular hexagonal arrangement. The pore size was unevenly distributed, and the shape of the holes was irregular but they were arranged in parallel. The internal dimensions were comparable to the orifices. After dissolving the template, statistical analysis showed that the pore size distribution ranged from 120 to 260 nm, the size of 64% of the pores ranged from 180 to 220 nm, with an average pore size of 198 nm, an average pore center distance of 280 nm, and the nanowire length of about 20 \( \mu \)m. As copper nanowires were deposited and grown in the pores of the hard template, the pore size and distribution of the AAO template had an important influence on the microstructure of the copper nanowires.

3. RESULTS AND DISCUSSION

Because the laser spot satisfied the Gaussian distribution, the 20% energy of the laser was concentrated within the radius of 20–30 \( \mu \)m to transfer high-energy electrons. This spot size was relatively small in the low-contrast and ultra-intensity laser system above 2 J @30 fs and 800 nm.

3.1. Yield and CE of X-rays. To calculate the CE, it was necessary to record the number of photons according to the single-photon mode of the LCX CCD channel, so the probability of two photons competing at one pixel of CCD was very small. Moreover, the background difference method was often used to calculate the number of X-ray photons, which referred to the background data collected before the experiment, and then the experimental spectrum was distinguished from the background. The CE of the femtosecond laser energy to Kr X-ray photons was calculated, including the intermediate process of converting electrons based on the number of photons recorded by LCX CCD. It is acknowledged that there is a small deviation in the manufacturing accuracy of the detection instrument and the experimental measurement methods, so every experiment has a certain degree of cumulative error.

Figure 3 showed the Cu target X-ray spectra of the laser-driven two-shot foil and the four-shot nanowire target recorded by LCX CCD. All these spectra included two copper foil backgrounds. After dissolving the template, statistical analysis showed that the pore size distribution ranged from 120 to 260 nm, the size of 64% of the pores ranged from 180 to 220 nm, with an average pore size of 198 nm, an average pore center distance of 280 nm, and the nanowire length of about 20 \( \mu \)m. As copper nanowires were deposited and grown in the pores of the hard template, the pore size and distribution of the AAO template had an important influence on the microstructure of the copper nanowires.

![Figure 2](https://dx.doi.org/10.1021/acsomega.0c01135) SEM photographs of AAO at 15,000 times (left) and Cu nanowire material at 40,000 times (right) magnification. The average of pore center distance is 280 nm.
the Cu characteristic line, corresponding to the Kα and Kβ peaks of copper X-rays. Obviously, no. 2022 revealed that the X-ray CE was the highest emission intensity with nanowire Cu target @6 J laser energy. However, the broadband spectrum included bremsstrahlung emission and characteristic spectrum. Nonetheless, its full width at half maximum (fwhm) was obviously fatter than other shots. It might also be that the laser energy was the largest and the number of X-ray photons from the nanowire was the largest. Serbanescu et al. analyzed X-ray source based on various focal spot conditions and varied degrees of preplasma at kilohertz repetition rate, which clearly showed that the level of preplasma was an important parameter affecting the efficiency of Kα X-ray.2

The number of Cu Kα photons produced by the two Cu foil targets was \(1.2 \times 10^8\) and \(6.0 \times 10^7\) photons sr\(^{-1}\) s\(^{-1}\) at about 2 J laser energy, respectively. The energy of the two driving lasers was basically the same, but the number of photons was half different, which might be caused by experimental errors. The number of Kα X-ray photons of nanowires increased with the driving femtosecond laser energy, from a minimum of \(1.1 \times 10^8\) to \(3.6 \times 10^8\) photons sr\(^{-1}\) s\(^{-1}\), among which the maximum laser intensity was \(4.6 \times 10^{18}\) W/cm\(^2\) @6 J in Table 1, which was consistent with the results of Dobosz’s.26 The exponential relationship between the average number \(N_p\) of photons and peak laser intensity \(I_{peak}\) was satisfied, which was expressed as

\[ N_p \propto I_{peak}^{3/2} \]  

(1)

Of course, the number of photons obtained by the CCD was also affected by the detection angle. The photon distribution was in the shape of a lotus flower, which was simulated by Chen through 1D fully electromagnetic LPIC++ at the incidence of 45°.7 The average Kα photo yield of the nanowire target was \(2.2 \times 10^8\) photons sr\(^{-1}\) s\(^{-1}\), which was about 2.5 times that of the foil target. It may be concluded that due to high surface to volume ratio or laser intensity increase, the absorption ability of the nanowire structure target to laser energy is stronger than that of the foil target.

When calculating the energy conversion of Cu Kα X-ray, the 120 μm copper and 500 μm beryllium thick windows were taken into account, and the X-ray photon transmission distance in the air was ignored instead of the distance from the vacuum chamber window to the detector. Therefore, the transmission factor \(T = 0.0034\) of Cu Kα X-ray was calculated from the radiation source to the detector yield. Assuming that the isotropic X-ray emission is \(2\pi\) sr, the CE \(\eta_{K\alpha}\) is also calculated as a \(2\pi\) sr sold angle. The CE for Cu Kα X-ray photons produced is defined as

\[ \eta_{K\alpha} = \frac{NE_{K\alpha}}{E_{laser}} \]  

(2)

where \(N\) is the number of Cu Kα photons, \(E_{K\alpha}\) is the Cu Kα photon energy, and \(E_{laser}\) is the laser pulse energy incident on
the target. As the fs laser intensity increased, the CEs $\eta_{\text{Kr}}$ of the copper nanowire target was also improved in the range of 0.0036–0.0087%, as shown in Table 1. The Cu Kr average CE per pulse was 0.006%, which was 2 times that reported by Serbanescu (3.2 ± 0.4) × 10−5 for 1 kHz repetition on the order of magnitude, compared with the Cu foil of 0.0045%. In general, the improved nanostructure target could absorb more laser energy and generate more photons, which also improved the conversion from a femtosecond laser to X-rays.

Although our laser intensity was higher than others', the experimental result also showed that enhancements in local electromagnetic fields result in excess absorption and hotter electrons at about 1018 W cm−2. Rajeev et al. quantitatively explained the enhancement to the design of structured surfaces via surface plasmon and “lightning rod” effects, which irradiated with 100 fs, 806 nm laser pulses, focused to intensities in the range of 1014 to 1015 W cm−2. Sudipta et al. reported that the Cu nanorod array samples produced about 20–40 times higher X-ray yield than the optically polished Cu targets irradiated at 1016 W cm−2, assuming isotropic emission of X-ray photons over the range of 150–300 keV.

3.2. Spot Imaging of X-rays Source. Generally, as one of the high-quality backlight sources, the femtosecond laser energy concentration can be inferred from the focal spot size of Cu Kr X-ray source. The smaller the spot, the higher the need for resolution applications, such as biological high-resolution X-ray imaging systems. In order to verify the good laser energy absorption ability of the nanostructure, the X-ray source size was measured by pinhole and PIXIS-XB CCD.

The filter used was a 5 μm thick aluminum film with a transmittance of 50% at 8 keV, which could effectively absorb soft X-rays below 4 keV. The quantum efficiency of PIXIS-XB CCD was about 20%. The resolution of a pinhole camera was evaluated by the mean square of geometric resolution $\Delta_n$ and the physical resolution $\Delta_\alpha$. The geometric resolution can be expressed as

$$\Delta_n = d(1 + 1/M)$$

Here, $d$ is the pinhole diameter of 10 μm and $M$ is the magnification. The physical resolution from the angle of the pinhole diffraction effect is

$$\Delta_\alpha = k\lambda d^{-1}$$

Here, coefficient $k$ is 2.44, $\lambda$ is the X-ray wavelength of 1.54 nm for Cu Kr, and $d$ is the object distance of 175 mm, the actual magnification $M$ is 4.7. Under this experimental condition, the design resolution meets $\Delta \approx \Delta_\alpha \approx \Delta_n$.

The spot sizes for X-ray of two copper foil targets and four nanowire targets are shown in Table 1. Apparently, the average spot was 85–240 μm from the nanowire Cu targets, which was similar to the 63–170 μm of the copper foil target. The ratio of the short radius to the long radius was about 0.6 for the Cu foil and 0.5 for the nanowire target. It could also be seen in Figures 4 and 5 that the focal spot image of the four nanowire targets was relatively round, while that of the two foil targets were elliptical. In addition, the spot measured by the pinhole imaging method was slightly larger than the knife-edge method, while the spot measured with the knife-edge was 47 and 59 μm from laser-creased Cu foil, respectively. In addition to the different incidence angles, the fs laser energy interaction with the nanowire material was also stronger, with more output and more focus. It might also be obliquely incident on the target surface, but the spot image looked elliptical.

Based on the experimental geometry, the fs laser focal spot was about 30 μm at the fwhm incident on the targets. According to the report of Fourmaux et al., the X-ray spot was only 2–8 times that of the femtosecond laser, whether it was copper foil target or nanowire targets material. For example, the X-ray focal spot size of no. 2010 was 90/170 μm (short/long radius) and that of no. 2018 was 112/240 μm with X-ray hole image (see Figures 4 and 5), the aspect ratio of which was about 1.5–2.5. The spot roundness of the nanowire was larger than that of a flat target creased by fs laser. According to the equilibrium stage separation process, including the material balance equation (M equation), the

Figure 5. Focus spot size with pinhole image of nanowire Cu in no. 2018 and 2020–2022. Obviously, the roundness of nanowire is better than those of Cu foil targets.
normalization equation (S equation) of phase equilibrium equation and each component of each equilibrium stage, and the thermal equilibrium equation (H equation), a new position of the computational mesh was chosen.29 In Figures 6 and 7, two-dimensional Figures 4 and 5 were transformed into the three-dimensional mesh, and the results showed that the X-ray focal spots of the nanowire targets were more uniform than those of foil targets driven by fs laser via pinhole imaging. In

Figure 6. Mesh of X-ray focal spot from two targets of no. 2010 and 2011 from foil targets driven by fs laser.

Figure 7. Mesh of X-ray focal spot from four targets of no. 2018 and 2020–2022 from nanowire driven by fs laser. Every shot included a 3 × 3 matrix pinhole.

Figure 8. X-ray image of resolution plate. From left to right, the resolution image of per shot reflects the features sizes varying from 6 to 9 lp/mm in no. 2018 and 2020–2022.
addition, Figure 7 also showed that the focal image was $3 \times 3$ instead of $5 \times 6$ as designed, which was incomplete for the magnification ratio.

The nanowires encouraged faster electron collimation, which would reduce divergence and reduce the X-ray spot size. However, the X-ray resource spots of the planar target were approximately smaller than that of the nanowire. From the comparison between Figures 4 and 5, the X-ray emission spot of the nanowire was more round. However, the X-ray emission spot projection intensity was a few factors lower in the vertical ($y$-direction) than in the transverse ($x$-direction) direction in counts. The fwhm in the transverse direction was narrower than the half-height in the $y$ direction, indicating that the vertical direction was more concentrated, as shown in Figure 5 and Table 1. The X-ray spot of the nanowire was larger than that of plain foil.

3.3. Resolution Plate Image of Backlight. In the experiment, a resolution plate was also used to measure the imaging resolution. A 10 line pair/mm (lp/mm) resolution plate was placed between the target and a 0.6 T magnet, 120 mm from the target. Imaging experiment was also performed to distinguish the spatial resolution with 9-line pair/mm array scanner. As shown in Figure 8, the resolution image could clearly reconstruct the structural features of the resolution plate. The X-ray source resolution was analyzed from the X-ray point emission. From left to right, the feature size of each shot resolution image varied from 6 to 9 lp/mm. Although all lines were distinguishable, the resolution plate was significantly reduced to 50% at 9 lp/mm and was no longer visible at 10 lp/mm. According to the principle of backlight imaging, the spatial resolution of $1000/9/2/M = 42.8 \mu m$ of the X-ray source was distinguished, where parameter $M = 1.3$ was the magnification coefficient in the experiments. The single-shot radiation imaging was clear, indicating that the number of photons was high enough. In any case, the stripes of $42.8 \mu m$ were clear and the source size was much smaller than $42.8 \mu m$ shown as Figure 8.

It was key for a small X-ray point source to be widely used in the backlight imaging for dense plasma, material, physics science, and other fields. The resolution plate results showed that the X-ray point source was smaller than $42.8 \mu m$. However, spot size was larger than $63 \mu m$ with the pinhole image method, which was also larger than the result of $40 \mu m$ measured by knife-edge.35 Hollinger et al. reported that the increase in X-ray flux, the small source size of $\sim 5 \mu m$ deduced from the penumbra in a knife-edge test,30 and the picosecond pulse duration makes these plasma an excellent X-ray point source for time-resolved flash radiography, which was 10 times that of our experimental laser intensity with 10 times CE. Zhou et al. opened new paths to investigate the amplification of electron density and quasistatic magnetic fields simulated by PIC.31

4. CONCLUSIONS

To improve the surface of materials to absorb more laser energy, multiple X-ray photons are one of the research methods at present. The nanowire target was designed by the AAO method, which increased the surface area of the target. Furthermore, it also provided a channel for super-hot electrons, which was the key to increase the X-ray yield. Two different types of targets, nanowire target and copper foil target, were tested on XG-III laser equipment. Compared with the Cu Kα X-ray photon yield of the foil target produced by femtosecond laser, that of the nanowire target was as high as $3.6 \times 10^8$ photons sr$^{-1}$ s$^{-1}$. The average yield of the nanowire target was $2.2 \times 10^8$ photons sr$^{-1}$ s$^{-1}$, which was 2.5 times that of the foil target. Compared with the average CE of 0.0045% of the foil target, that of the nanowire target was 0.006%. Among them, the maximum valve was 0.0087%. The focal spot size of the nanowire was $85 \sim 240 \mu m$, which was similar to or slightly larger than that of Cu foil of $63 \sim 170 \mu m$ measured by $910 \mu m$ pinhole imaging. Finally, the resolution plate was used to test the imaging capability of the X-ray point source. The experimental data showed that the modified nanostructure wire material could significantly improve the output of an X-ray photon driven by ultrashort laser, which also provided a potential economic reference method for the application of high-resolution X-ray imaging in the future.

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