解説**REVIEW ARTICLE****高性能新型軟X線レーザーの開発と分子生物・医学**黒田 寛人¹, Rashid Ganeev¹, 鈴木 将之¹, 馬場 基芳¹, J. Zhang¹, 尾崎 恒之²¹東京大学物性研究所・先端分光部門²カナダ・国立科学研究所

(平成18年1月20日受理, 平成18年1月31日掲載決定)

STUDIES OF HIGHLY ADVANCED SOFT X-RAY LASERS FOR BIOMEDICAL AND NANO-LASER MEDICINE APPLICATIONSH. Kuroda¹, R.A. Ganeev¹, M. Suzuki¹, M. Baba¹, J. Zhang¹, and T. Ozaki²¹*Institute for Solid State Physics, University of Tokyo, Tokyo Japan*²*Institut national de la recherche scientifique (Quebec) Canada*

(Received January 20, 2006, Accepted January 31, 2006)

Abstracts

Current activities on our research of soft x-ray lasers and higher harmonic generations using intense, subpicosecond lasers are reviewed. Especially for soft x-ray lasers we experimentally demonstrate that by longitudinally pumping 2-mm-long molybdenum preformed plasma with high-intensity 475 fs duration laser pulse, a highly directive soft x-ray laser at 18.9 nm wavelength is generated. The divergence of the beam is evaluated to be of the submillirad order, and only requires pump laser energy of 150 mJ. Simulations show that the pedestal in the main pump pulse can generate electron density and gain profiles with large spatial gradients, which result in the selective amplification of low-order transverse modes. The present result is the demonstration of an efficient and alternative method of improving the spatial coherence of x-ray lasers with amplified spontaneous emission medium, with possibilities of becoming an excellent tool to explore various application experiments. In higher harmonic generation, topics related to observation of blue shift due to collisionless absorption process is described. We demonstrate the generation of high harmonics (up to the 65th order, $\lambda=12.24$ nm) of a Ti:sapphire laser radiation after the propagation of femtosecond laser pulses through the low-excited boron plasma produced by a prepulse radiation on the surface of different targets. High-order harmonics generated from the surface plasma of most targets showed a plateau pattern. The harmonic generation in these conditions assumed to occur due to the interaction of femtosecond pulses with ions. The conversion efficiencies in the plateau region were varied between 10^{-7} to 8×10^{-5} depending on the target. The main contribution to the limitation of harmonic generation efficiency and cutoff energy was attributed to the free-electrons-induced self-defocusing of main pulse.

Key Words: Soft x-ray laser, high harmonic generation, XUV spectroscopy, ultra-short pulses, nano laser medicine.¹〒277-8581 千葉県柏市柏の葉5-1-5 (5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan)²1650 boul. Lionel-Boulet Varennes (Quebec) Canada J3X 1S2

1. Introduction

Since the time when first ruby laser was demonstrated, future development of x-ray laser remains as one of a dream of laser physicists and almost all researchers engaged in laser applications involving biomedical applications and basic medicine. In this papers, present status of highly advanced soft x-ray laser based on longitudinally Ni-Mo 18.9 nm laser which might possess the capability of opening a new phase in x-ray laser application is described. This is the first clear demonstration that we have now obtained very efficient, smart table top highly qualified soft x-ray laser. This 18.9 nm Ni-Mo x-ray laser which are developed by us recently has and extraordinary small divergence, and also operates at pump laser energies of several 100 mJ. These are over ten times good divergence and 100 times smaller energy compared to other results. The present scheme is based on the transient-collisional- excitation (TCE) scheme extended to the Ni-like ion. This method uses a sub-picosecond or femtosecond, high-intensity laser pulse to generate a short-lived, high gain laser medium of only 2 mm length within preformed plasma. The experimental results reveal an additional favorable characteristic of longitudinal-pumped TCE x-ray laser, that is ultra-small divergence of less than one milliradian (0.3 mrad). These unique features will have a large impact on x-ray laser application. An example is the precise measurement of time-resolved emission spectroscopy in live tissue, which is tremendously difficult even with advanced synchrotron radiation sources. Nonlinear frequency conversion sill allow tunability of the x-ray laser wavelength, allowing us the measure real atomic structure via photoelectron holography. Such methods will allow the precise measurement and observation of various nano-structures, with which we hope to renew our understandings of new fields such as nanotechnology, nanobiology and laser medicine. Efforts to develop shorter wavelength around water window region (2.2 nm-4.4 nm) based on Ni-W 4.3 nm laser will be continued.

A new approach towards more advanced scheme based on a transient collisional scheme equipped with high beam qualities pumped with by far small energy (table top soft x-ray laser) was proposed¹⁾, and it has been successfully demonstrated producing 18.9 nm highly directive Mo soft x-ray laser in Ni-Mo target.^{2,3)} In the present paper, aspect of this important observation is described.

On the other hand, we are also devoting extensive studies on higher harmonic generation from solid surface plasma, which is also recognized as an alternative way to pursue coherent light source in soft x-ray regions. In recent studies, one interesting observation is blue shift of generated higher harmonics without broadening due to self phase modulation. These shifts are brought about by collisionless absorption process, which shows an onset of new interaction process predicted for a mechanism responsible for ultra short and intense, laser-matter interaction. In this paper, aspects of these new topics associated with soft x-ray laser (SOX) and higher harmonic generation (HHG) are discribed.

The laser plasma has long been used as a medium for harmonic generation. The high-order harmonic generation (HHG) in visible and extreme ultraviolet (XUV) ranges has been investigated in the laser plasma formed by optical breakdown in gases and at the sur-

face of solid targets¹⁻³⁾. The application of various ions provided the important information on their roles for the HHG. However, the maximum observed order of harmonics reported in those studies was limited to the 27th one due to some concurred effects in high-excited plasma. No plateau pattern for high harmonics was reported in those studies.

In this paper we demonstrate the efficient generation of coherent XUV radiation after the propagation of femtosecond pulse through the low-excited plasma produced on the surface of different solid targets. The harmonics up to the 65th (12.24 nm) order and the conversion efficiencies up to 8×10^{-5} were achieved in these studies. We show our results on the spectral modification of harmonic distribution for some plumes. Strong resonance enhancement of single harmonic will also be discussed.

2. Highly directive 18.9-nm nickellike molybdenum x-ray laser operating at 150 mJ pump energy

2.1 Experimental Results

In the present paper, we show experimental results that might change our preconceptions on x-ray lasers. The 18.9 nm Ni-like molybdenum x-ray laser described in this paper has an extraordinary small divergence, and also operates with only 150 mJ pump laser energy.^{2,3)}

A 2-mm-long slab molybdenum target (T) is placed within a vacuum chamber, which is pumped by two laser beams from a tabletop 1.06 μm -wavelength Nd:glass / Ti:sappire hybrid chirped pulse amplification laser system. First, the 300-ps duration prepulse is line-focused onto the target surface using a cylindrical lens, which produces the highly ionized molybdenum preplasma. After a typical time of several nanoseconds, an intense 475-fs main pulse is point-focused and pumps the preplasma from a longitudinal direction, producing a high transient gain at 18.9 nm while propagating through the preplasma. The dimensions of the long pulse at focus are 100 μm wide and 2 mm long, and the spot diameter and confocal parameter of the short pulse is 30 μm and 10 mm, respectively. Typical intensities of the long and short pulses are $1.5 \times 10^{11} \text{ Wcm}^{-2}$ and $3 \times 10^{16} \text{ Wcm}^{-2}$, respectively.

In Fig.1 we show the intensity trace of a typical on-axis soft-x-ray spectrum between 15 and 25 nm observed using the above experimental setup. The background originating from the noise of the detector was sampled from a portion of the image without the spectrum, and subtracted to obtain the actual spectral intensity trace. The pump laser energy in the long and short pulse is 30 mJ and 120 mJ, respectively, and the peak-to-peak delay time between the two pulses is 4 ns. The wavelength of this spectrum is calibrated by replacing the molybdenum target with a boron target, and using the high-order diffraction of the resonance lines of He-like and H-like boron. We can see from this figure that the on-axis emission from the molybdenum plasma is completely dominated by a single line at 18.9 nm, which is assigned to the $3d^9 4d \ ^1S_0 - 3d^9 4p \ ^1P_1$ line of Ni-like molybdenum.

The most interesting point about the observed spectrum is the narrow spectral width of the 18.9 nm line, in spite of the fact that the spectrograph slit is widely opened to a width of 1.6 mm. In fact, the size of the 18.9 nm line at the detector plane is not only

Fig.1 Intensity trace of the on-axis soft-X-ray spectrum between 15 and 25nm. The background was subtracted from the trace to obtain the actual intensity profile.

Fig.2 Spectrum image of the on-axis emission for plasma length of (a) 2.0mm and (b) 0.5mm. The sensitivity of the photocathode camera was increased by about eight times for (b)

small in the direction of spectral dispersion, but also in the direction vertical to it. This can be seen in Fig.2(a), which is the raw image of the on-axis spectrum between 16 and 22 nm wavelengths for 2-mm-long molybdenum plasma. By fitting the observed intensity profile of the 18.9 nm line assuming a Gaussian transverse mode, the full-width at $1/e^2$ maximum of this spot at the detector plane is 93 μm in the direction of spectral dispersion and 85 μm in the vertical direction.

A big question arises as to why such highly directive x-ray lasers can be generated from short 2-mm-long plasma. We attribute this unique phenomenon to the presence of a pedestal in the longitudinal pump beam. We have experimentally measured the pedestal-to-peak contrast ratio of our 475 fs pump laser to be about 1×10^{-5} for the present pump conditions, using photomultiplier tube and calibrated neutral density filters. There are two outcomes of this pedestal that can greatly affect the divergence of the present x-ray laser. First is the localization of the Ni-like ion abundant region to the position of the longitudinal beam.

Simulations show that the $1.5 \times 10^{11} \text{ Wcm}^{-2}$ intensity prepulse is only capable of producing preplasma with an average ionization of 8, much less than 14 required for Ni-like molybdenum ions. However, by introducing a 300 ps duration, 1×10^{-5} contrast ratio pedestal to the main pulse, the temperature and density is increased to 90 eV and $1 \times 10^{20} \text{ cm}^{-3}$, respectively. As a result, narrow Ni-like molybdenum abundant region is produced within the preplasma, which can generate high gain with the irradiation of the high intensity main pump.

Fig.3 Effect of pedestal in pumping pulse

2.2 Pre-plasma conditions

A big question arises as to why such highly directive x-ray lasers can be generated from short 2-mm-long plasma. We attribute this unique phenomenon to the presence of a pedestal in the longitudinal pump beam. We have experimentally measured the pedestal-to-peak contrast ratio of our 475 fs pump laser to be about 1×10^{-5} for the present pump conditions, using photomultiplier tube and calibrated neutral density filters. There are two outcomes of this pedestal that can greatly affect the divergence of the present x-ray laser. First is the localization of the Ni-like ion abundant region to the position of the longitudinal beam. Simulations show that the $1.5 \times 10^{11} \text{ Wcm}^{-2}$ intensity pre-pulse is only capable of producing pre-plasma with an average ionization of 8, much less than 14 required for Ni-like molybdenum ions. However, by introducing a 300 ps duration, 1×10^{-5} contrast ratio pedestal to the main pulse, the temperature and density is increased to 90 eV and $1 \times 10^{20} \text{ cm}^{-3}$, respectively. As a result, narrow Ni-like molybdenum abundant region is produced within the pre-plasma, which can generate high gain with the irradiation of the high intensity main pump. To illustrate this effect, the result of gain calculation with and without the above pedestal condition is shown in Fig.3, respectively. Here the pedestal produces a density profile within the pre-pulse-produced plasma in the x-direction normal to the target surface, which is assumed to be uniform in the z-direction parallel to the x-ray laser axis. We find that gain coefficients exceeding 70 cm^{-1} are generated at the entrance of the pre-plasma, over a length of 500 μm . A long and narrow positive gain region extends over the full length of the 2-mm-long pre-plasma, which corresponds to the region that is pumped by the pedestal. Note that gain outside of this region is strongly negative, resulting in strong absorption of the x-ray laser. In case of a longitudinal pump without pedestal, the x-ray beam is bent by a relatively large electron density gradient and no gain is observed both in the experiments and in the simulation. Fig.3 shows the simulation result.

2.3 Pedestal conditions ^{4,5)}

Another important outcome of the presence of a pedestal in the main pulse is the generation of electron density and gain distributions that have large spatial gradients. The parabolic elec-

Fig.4 The parabolic electron density and gain profile that are produced by the pedestal is found to limit the number of transverse modes that are amplified in the gain medium.

tron density and gain profile that are produced by the pedestal is found to limit the number of transverse modes that are amplified in the gain medium. In the work by London *et al.*⁶⁾, it was numerically shown that for gain medium with such unique profiles, the efficiently guided modes that receive large amplification could be restricted to the lower order modes (See Fig.4). Using formulations from this paper, we can calculate the number of transverse modes that receive large amplification in our experiment. Assuming that the density and gain profile is homogeneous along the x-ray laser axis, we take the lower limit of $0.18 \times gL_{\max}$ for counting the number of modes, where $gL_{\max}$ is the maximum gain-length product experienced by a single transverse mode. If the density profile vertical to the target surface was flat, the number of effectively amplified one-dimensional Hermite-Gaussian modes is 47, and in this case the geometrical dimensions of the gain region determines the divergence of the x-ray laser. However, this number is reduced to 5 for the parabolic profile predicted in our experiment, with the maximum gain obtained for the diffraction-limited transverse mode. This shows that for our experimental condition, amplification prefers transverse modes with high spatial coherence, which explains the observed small divergence.

3. Tunable coherent high order harmonics generations due to collisionless absorption

The availability of compact, high-intensity sub-picosecond lasers with chirped pulse amplification (CPA) has opened the new field of study of laser-matter interactions with solid targets. At the high laser intensities above 10^{16} Wcm^{-2} , the electron temperature is more than one keV and the main laser absorption is related to collisionless absorption. Collisionless absorption mechanisms include vacuum heating, $\mathbf{J} \times \mathbf{B}$ heating, sheath inverse Bremsstrahlung, and the anomalous skin effect. Hansen *et al.* reported for the first time that the second harmonic is blue shifted when the laser has intensity exceeds $5 \times 10^{16} \text{ Wcm}^{-2}$ and the blue shift is due to the absorption by the anomalous skin effect⁷⁾. However, no detailed study of the blue shifts in the other harmonic orders, nor the dependence on the laser intensity at the incidence angle of 45 degrees has yet been reported. It was commonly observed that efficiency of harmonic generation for the p-polarized laser pump was much larger than that for the s-polarized laser beam. Only a few experimental studies have been reported on the effects of polarization on the harmonic radiation. However, the

cause of the disagreement between some recent theoretical and experimental results is not yet clear.

In this paper, we present blue shifts of up to about 2 nm in the second harmonic and for the first time about 5 nm in the fifth. We carry out more detailed model-calculations of the electron temperature in our experimental condition using a 1D hydrodynamic code HYADES. We find that the blue shift in our experi-

ment can be well explained from the absorption mechanism due to the anomalous skin effect. We also show that the harmonic dependence on the pump laser polarization decreases when laser intensity exceeds 10^{17} Wcm^{-2} . Fig.5 shows the dependence of the wavelength shift of the second harmonics on the incidence laser intensity. The peak frequency of the second harmonic emission is blue shifted for intensities greater than $2 \times 10^{16} \text{ Wcm}^{-2}$. It is found that the increase of this shift energy with an increase in the laser intensity is small in comparison with the experimental results by Hansen *et al.*⁷⁾.

Unfortunately, we cannot detect spectral shift below $1 \times 10^{16} \text{ Wcm}^{-2}$ that the fifth harmonic light is blue shifted by 5.1 nm for p-polarization at the intensity of $1 \times 10^{17} \text{ Wcm}^{-2}$. This shift is much larger in magnitude than the Doppler shift seen in the spectrum of the fundamental. Therefore, these results cannot be explained by the hydrodynamic motion of plasma. The calculation results by Rozmus *et al.*⁸⁾ show that at low temperature ($T_e < 300 \text{ eV}$), the collision absorption mechanism is dominant, and at higher temperature ($T_e > 800 \text{ eV}$), the collisionless absorption mechanism due to the anomalous skin effect becomes dominant. Fig.6 shows schematic model of anomalous collisionless absorption within a one sheath.

The electronic temperature profile in our experiment condition was calculated using a 1D hydrodynamic code HYADES. From the results, it is found that the electron temperature in the skin depth from the critical density surface is much higher than 800 eV. Therefore

Fig.5 Location of the second harmonic peak as a function of incident intensity in our experiment.

Fig.6 Schematic of collisionless absorption image on the solid surface plasma irradiated by an ultra-intense laser pulse

from our experimental and calculation results, we attribute the harmonic frequency shift to the collisionless absorption mechanism, and the difference in the wavelength shift between the orders of harmonics in our experimental results is attributed to the phase shift accumulation or phase shift rotation at a certain cycle. A more detailed analysis of the experimental data has been reported elsewhere⁹⁾.

4. Brilliant highly directive higher harmonics and soft x-ray lasers from solid target plasma pumped by tabletop Ti:S laser

4.1 Experimental arrangements

Fig.7 shows the experimental setup for the high-order harmonic generation. The pump laser used in this research was consisted of chirped-pulse amplification Ti:sapphire laser (Spectra-Physics, Tsunami + TSA10F) operated at a 10-Hz pulse repetition rate, whose output was further amplified using a three-pass amplifier. A portion of uncompressed radiation (pulse energy $E=15$ mJ, pulse duration $t=210$ ps, central wavelength $\lambda=796$ nm) was split from the main beam by a beam splitter and used as a prepulse. This prepulse was focused by a spherical lens on a solid target located in the vacuum chamber and produced a plume predominantly consisting on neutrals and singly charged ions¹⁰⁾. The composition of plasma was analyzed using the time-integrated spectral measurements of plume in XUV, UV, and visible ranges. The focal spot diameter of prepulse beam on the target surface was adjusted to be approximately $600\ \mu\text{m}$. The intensity of picosecond prepulse, I_{pp} , on the target surface was varied from $7 \times 10^9\ \text{Wcm}^{-2}$ to $8 \times 10^{10}\ \text{Wcm}^{-2}$. After some delay, the femtosecond main pulse ($E=8$ mJ, $t=150$ fs, $\lambda=796$ nm) was focused on the target plasma from the orthogonal direction using 200-mm or 100-mm focal length lenses. Our experiments were carried out up the maximum intensity of the femtosecond main pulse of $I_{fp}=1 \times 10^{15}\ \text{Wcm}^{-2}$.

The high-order harmonics were analyzed by a flat-field grazing-incidence XUV spectrometer with a Hitachi 1200-groove/mm grating¹¹⁾. An additional gold-coated grazing-incidence cylindrical mirror was used for the image translation from the plasma area to the detector. The XUV spectrum was detected by a microchannel plate with phosphor screen and recorded by a CCD camera¹²⁾.

Fig.7 Experimental setup. First, the prepulse was focused on the target surface. The preplasma was produced by the prepulse. Then the main pulse was focused in the preplasma. Delay between the prepulse and the main pulse was from 3 ns to 94 ns.

4.2 Results and discussion

We used different solid targets (Ag, B, In, C, W, Se, Mo, Nb, Si, Zr, Mg, Cd, Pd, Al, Cr, Ta) for the preparation of suitable laser plumes for our experiments. The choice of targets was stipulated by our aim to analyze the influence of atomic number, ionization potentials, and other parameters of samples on the HHG from different plumes. Our efforts were concentrated on determination of the optimal conditions for achieving the maximum harmonic order (i.e., cutoff energy) as well as maximum conversion efficiency for high harmonics using low-excited laser plasma.

The plateau pattern of high-order harmonic distribution was appeared in these experiments. In particular, high harmonics up to the 63rd order at a wavelength of 12.6 nm were observed in experiments with boron plasma at loosely focused conditions ($b=6$ mm, $L_p=0.6$ mm, b is confocal parameter, L_p is the plasma sizes). The HHG from the B plume appeared to be similar to those observed in gas-jet experiments, with characteristic shape of plateau for the harmonics exceeding 17th order (Fig.8, curve 1). The conversion efficiencies were in the range of 10^{-4} (for the 3rd harmonic) to 10^{-7} (for the harmonics in plateau region). The plateau disappeared when the prepulse energy was increased above 10 mJ. In that case the highest harmonic observed was the 19th one. When the prepulse energy was further increased, the strong spectral lines from multiply ionized B appeared in the spectrum, and we did not observe any harmonics below 65 nm¹³⁾. Higher harmonics (up to the 65th order, $\lambda=12.24$ nm) were observed at the conditions when the confocal parameter of focused radiation was changed ($b=1.2$ mm) to be close to the plasma sizes. However, in that case we observed the instability of HHG in the terms of the plasma conditions. Most of our further presented studies were carried out at $b=6$ mm. Analogous pattern was appeared in the case of longitudinal scheme, when femtosecond beam propagated through the laser plasma and hole drilled in the target¹⁰⁾. High harmonics up to the 57th order ($\lambda=13.96$ nm) were routinely observed in these experiments (Fig.8, curve 2). The conversion efficiency within the plateau range in this scheme was measured to be 10^{-7} , analogous to the one in the case of orthogonal pump scheme. The plateau and high harmon-

Fig.8 Harmonic output as a function of the wavelength of generated XUV radiation in the cases of (1) orthogonal and (2) longitudinal pump schemes.

ics in the case of longitudinal scheme disappeared at high intensities of prepulse radiation ($I_{pp} > 5 \times 10^{10} \text{ W cm}^{-2}$) causing the excitation of strong B III and B IV lines.

Higher harmonics (up to the 65th order, $\lambda = 12.24 \text{ nm}$) were observed at the conditions when the confocal parameter of focused radiation was changed ($b = 1.2 \text{ mm}$) to be close to the plasma sizes. However, in that case we observed the instability of HHG in the terms of the plasma conditions. Most of our further presented studies were carried out at $b = 6 \text{ mm}$. Analogous pattern was appeared in the case of longitudinal scheme, when femtosecond beam propagated through the laser plasma and hole drilled in the target¹⁰⁾. High harmonics up to the 57th order ($\lambda = 13.96 \text{ nm}$) were routinely observed in these experiments (Fig.8, curve 2). The conversion efficiency within the plateau range in this scheme was measured to be 10^{-7} , analogous to the one in the case of orthogonal pump scheme. The plateau and high harmonics in the case of longitudinal scheme disappeared at high intensities of prepulse radiation ($I_{pp} > 5 \times 10^{10} \text{ W cm}^{-2}$) causing the excitation of strong B III and B IV lines.

The plateau pattern was appeared in the case of the HHG from most of plasma samples used in these experiments, while in some cases (W, C, Mg, Se) we observed a steady or even steep decrease of conversion efficiency for each next harmonic. The origin of this phenomenon is not clear and further studies are required for the clarification of the difference between harmonic distributions and cutoff energies observed from various targets. Nevertheless, we observed a direct relation between the cutoff energy of the harmonics generated from different targets and the second ionization potential of atoms. Fig.9 shows a linear dependence between these two parameters in the case of the targets where we observed a plateau pattern. These observations underline a decisive role of free electrons appearing during further ionization of singly charged particles that led to the restriction of cutoff energy. The conversion efficiencies in the plateau region were varied from 10^{-7} to 8×10^{-6} depending on the target.

Fig.9 Dependence between the cutoff harmonic and ionization potential of ions (second ionization potential) for different targets. The plateau pattern was not observed in the cases of C, Se, W, and Mg plumes.

Fig.10 Harmonic output from magnesium plasma in the case of (1) 2 ns and (2) 13 ns delays. Inset: the dependence between the 25th harmonic intensity and time delay in the case of palladium plasma.

We investigated the influence of the delay between prepulse and main pulse on the harmonic yield. The harmonic efficiency considerably increased when the time delay was increased above 2 ns (Fig.10). The harmonic yield remained approximately constant up to 30 ns delay and then steadily decreased for a fixed distance between the main beam and target surface (see the inset in Fig.10 showing the dependence between the 25th harmonic intensity and time delay in the case of palladium plasma). However, we were able optimizing the conversion efficiency at longer delays by moving a focal spot farther from the target.

The optimal conditions for harmonic generation at 18 ns delay were found to be at the distances of 100 - 150 μm from the target surface. The high harmonics were observed up to the maximum delays of 100 ns used in these experiments. Most of our experiments were carried out at 13 ns to 18 ns delays.

While most of targets showed a prolonged plateau-like distribu-

Fig.11 Harmonic spectrum from chromium plasma in XUV range. 27th order harmonic is disappeared using Cr laser plasma

Fig.12 High-order harmonic spectra from (1) indium and (2) silver plumes

Fig.13 Spectrum of the high order harmonic generation was obtained using the indium plasma. The conversion efficiency of 13th harmonic is 8×10^{-5} .

tion of high harmonics, some of them demonstrated very interesting features that were not observed previously in gas-jet experiments. In particular, in the case of chromium plasma we registered a considerable suppression of 27th harmonic with comparing to the neighbor harmonics (Fig.11). Such a feature was observed in the cases of different focusing geometries using both 200-mm and 100-mm focal length lenses. The absence of 27th harmonic was

registered in a broad range of main pulse intensities.

The investigation of the conditions of enhanced conversion efficiency for some group of harmonics in the plateau region was carried out in various studies. However, in our studies we observed another process, a considerable restriction of a certain harmonic in the plateau region, while neighbor harmonics' yield was relatively equal. A possible mechanism explaining this effect could be related with an influence of free electrons and depletion of neutrals and singly charged ions that led to a decrease of coherence length for some specific harmonics. At the same time, a propagation effects could not completely suppress a single harmonic. Probably, the target properties play crucial role on a considerable suppression of 27th harmonic intensity in the case of chromium plasma.

Most intriguing pattern was appeared in the case of indium plasma. We observed anomalously strong 13th harmonic radiation ($\lambda = 61.2$ nm), which almost two orders of magnitude exceeded the neighbor ones (Fig.12 and 13). These studies were repeated using different focusing conditions and plasma geometries. The conversion efficiency of 13th harmonic was measured to be 8×10^{-5} .

For the confirmation that this strong line in the range of 61.2 nm was originated from the nonlinear process rather than from a simple re-excitation of plasma line we investigated the influence of 13th harmonic output on polarization characteristics of the main pulse. The 25 degree rotation of quarter-wave plate led to the complete disappearance of 61.2 nm emission, as it should be assuming the origin of HHG, while the excited lines of plasma spectrum observed at different polarizations of main beam remained unchanged. Another test of this process was carried out by the variation of the wavelength of main beam to analyze whether the excited transitions from indium plasma influence on a plateau pattern of harmonic distribution. We were able to change the central wavelength of the output radiation of our laser from 770 to 820 nm. The 13th harmonic output was considerably decreased with displacement of the fundamental wavelength from 796 nm to the short-wavelength region. At the same time we observed a strong enhancement of the 15th harmonic at the wavelength of main pulse of 775 nm, while other harmonics remained relatively constant. All these observations show the influence of atomic and/or ionic transitions on the nonlinear susceptibilities of individual harmonics.¹⁴⁾

The question arises, why did we obtain a strong enhancement of single harmonic in the plateau range in the case of In plasma? Is it due to propagation effects, i.e., phase matching, or is it reflects a single-ion response? The propagation effects caused by macroscopic medium response in gaseous media started to be explored in early stages of HHG studies. Efficiency optimization for high harmonics was recently thought to be achieved, as several groups reported phase-matched generation, and then reached the so-called "absorption limit", for which re-absorp-

tion limits the effective medium length.

An enhancement of HHG efficiency should include the optimization of both macroscopic and microscopic responses. From the macroscopic point of view, such an optimization dominantly means an achievement of ideal phase matching conditions. Microscopic optimization includes the methods of the growth of polarization of harmonics that depends on nonlinear susceptibility of a separate harmonic. The last parameter considerably depends on the relation between the excited states of nonlinear medium in the vicinity of harmonic wavelength. To our opinion, the strong 13th harmonic was a result of resonance-induced enhancement of nonlinear susceptibility in the vicinity of 61 nm.

5. Conclusion

As topics in soft x-ray Laser research, an ultra-small divergence x-ray laser has been demonstrated at 18.9 nm, based on longitudinally pumped Ni-like molybdenum scheme. The divergence estimated from the far-field pattern of the x-ray laser beam is submrad, which is the smallest ever demonstrated for a TCE x-ray laser. The output energy of this x-ray laser is measured to be 27 nJ. Furthermore, the pump energy that is required is only 150 mJ, making it possible to operate this high performance x-ray laser over at ten-hertz multi-hertz repetition rates using present-day laser technology.

Together with soft-x-ray laser research, investigations have been performed on the harmonic generation from solid surface plasmas using the subpicosecond Nd:Glass laser and several interesting phenomena have been observed. The blue shift of the harmonics by collisionless process is one of important topics. These results are important demonstrations of the large harmonic blue shift that can provide a tool to realize a tunable coherent subpicosecond light source.

In order to develop the high repetition rate x-ray laser, the longitudinal-pumped TCE x-ray laser is one of the most effective pumping schemes. We have described the results of longitudinally pumping TCE x-ray laser and high order harmonic generation from the solid surface plasma. The most important factor of the longitudinally pumping x-ray generations is concave density profile of pre-plasma and pedestal of the pumping laser pulse. To pursue the high repetition rate of the laser-driven TCE x-ray laser, we have designed new 20 TW Ti:Sapphire laser system (600 mJ, 30 fs, 10 Hz). Special attention was paid to improve the contrast ratio and control of pulse shape and phase by adaptive AOPDF and 1 kHz preamplifier. At present we have already obtained the laser energy of 1.2 J at 10 Hz repetition rate before the pulse compressor. Precise measurements of characteristics and soft x-ray laser experiment by this system will soon be started.

In conclusion, we demonstrated the generation of high harmonics (up to 65th order, $\lambda=12.24$ nm) after the propagation of femtosecond laser pulses through the low-excited surface plasma created by a prepulse radiation. High-order harmonics generated from most plumes showed a plateau pattern. The harmonic generation in these conditions assumed to occur due to the interaction of femtosecond pulses with ions. The conversion efficiency for the harmonics generated in the plateau region was varied in

the range of 10^{-7} to 8×10^{-6} depending on target. The main contribution to the limitation of harmonic generation efficiency was caused by free-electrons-induced self-defocusing of main beam.

Our observation of the considerable resonance-induced enhancement of a single harmonic ($\lambda=61.2$ nm) in the plateau region with the efficiency of 8×10^{-5} in the case of In plume can offer some expectation that analogous processes can be realized in other plasma samples even in the shorter wavelength range where the highest harmonics were achieved. It is difficult to realize this approach in gas puffs due to the necessity of the preparation of specific conditions for the excitation of appropriate levels of nonlinear medium prior to laser-matter interaction.

Acknowledgements

This work was partly supported by the Grant-in-Aid for Creative Scientific Research (14GS0206) of Japan Society for the Promotion of Science (JSPS).

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