

Ultra-fast switching of light by absorption saturation in vacuum ultra-violet region

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Abstract: Advances in free electron lasers producing high energy photons [Nat. Photonics **2**(9), 555-559 (2008)] are expected to open up a new science of nonlinear optics of high energy photons. Specifically, lasers of photon energy higher than the plasma frequency of a metal can show new interaction features because they can penetrate deeply into metals without strong reflection. Here we show the observation of ultra-fast switching of vacuum ultra-violet (VUV) light caused by saturable absorption of a solid metal target. A strong gating is observed at energy fluences above $6\text{J}/\text{cm}^2$ at wavelength of 51nm with tin metal thin layers. The ratio of the transmission at high intensity to low intensity is typically greater than 100:1. This means we can design new nonlinear photonic devices such as auto-correlator and pulse slicer for the VUV region.

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References and links

1. T. Shintake, H. Tanaka, T. Hara, T. Tanaka, K. Togawa, M. Yabashi, Y. Otake, Y. Asano, T. Bizen, T. Fukui, S. Goto, A. Higashiya, T. Hirono, N. Hosoda, T. Inagaki, S. Inoue, M. Ishii, Y. Kim, H. Kimura, M. Kitamura, T. Kobayashi, H. Maesaka, T. Masuda, S. Matsui, T. Matsushita, X. Maréchal, M. Nagasono, H. Ohashi, T. Ohata, T. Ohshima, K. Onoe, K. Shirasawa, T. Takagi, S. Takahashi, M. Takeuchi, K. Tamasaku, R. Tanaka, Y. Tanaka, T. Tanikawa, T. Togashi, S. Wu, A. Yamashita, K. Yanagida, C. Zhang, H. Kitamura, and T. Ishikawa, "A compact free-electron laser for generating coherent radiation in the extreme ultraviolet region," Nat. Photonics **2**(9), 555–559 (2008).
2. A. Penzkofer, "Passive Q-switching and Mode-Locking for the generation of nanosecond to femtosecond pulses," Appl. Phys. B **46**(1), 43–60 (1988).
3. H. Kitamura, "Cluster-model study on the K-shell excited states of crystalline lithium under intense laser irradiation," Eur. Phys. J. D **52**(1-3), 147–150 (2009).
4. B. Nagler, U. Zastra, R. R. Fäustlin, S. M. Vinko, T. Whitcher, A. J. Nelson, R. Sobierajski, J. Krzywinski, J. Chalupsky, E. Abreu, S. Bajt, T. Bornath, T. Burian, H. Chapman, J. Cihelka, T. Döppner, S. Düsterer, T. Dzelzainis, M. Fajardo, E. Förster, C. Fortmann, E. Galtier, S. H. Glenzer, S. Göde, G. Gregori, V. Hajkova, P. Heimann, L. Juha, M. Jurek, F. Y. Khattak, A. R. Khorsand, D. Klingner, M. Kozlova, T. Laarmann, H. J. Lee, R. W. Lee, K.-H. Meiwes-Broer, P. Mercere, W. J. Murphy, A. Przystawik, R. Redmer, H. Reinholz, D. Riley, G. Röpke, F. Rosmej, K. Saks, R. Schott, R. Thiele, J. Tiggesbäumker, S. Toilekis, T. Tschentscher, I. Uschmann, H. J. Vollmer, and J. S. Wark, "Turning solid aluminium transparent by intense soft X-ray photoionization," Nat. Phys. **5**(9), 693–696 (2009).
5. B. L. Henke, E. M. Gullikson, and J. C. Davis, "X-ray Interactions: Photoabsorption, Scattering, Transmission, and Reflection at E=50-30,000eV, Z=1-92," At. Data Nucl. Data Tables **54**(2), 181–342 (1993).
6. M. A. Kramer, W. R. Tompkin, and R. W. Boyd, "Nonlinear-optical interactions in fluorescein-doped boric acid glass," Phys. Rev. A **34**(3), 2026–2031 (1986).
7. R. N. Zitter, "Saturated Optical Absorption Through Band Filling Semiconductors," Appl. Phys. Lett. **14**(2), 73–74 (1969).

1. Introduction

Nonlinear response of solid absorbers is commonly used in the visible and infrared range for Q-switching, pulse shortening, and mode-locking [2]. As is well known, metals have a strong free electron response and it is difficult to use metals for such applications in the optical wavelength region. However, above the plasma frequency a metal can transmit light and shows phenomena related to the band gap structure. In this case, inner shell ionization is expected in the solid. Recently, we investigated the condition of such high density inner shell ionization theoretically for Li cluster [3]. This result emphasizes even with inner shell ionization, the cluster is not decomposed if the ionized electrons are located in the conduction band. Observation of transit Al transmission is also reported with L-shell ionization over 10^{16} W cm^{-2} at 92 eV [4]. In this letter, we demonstrate such a nonlinear response of absorption of VUV light by using N shell ionization of tin. In this condition, we can expect lower threshold energy and high contrast of transmission with saturable absorption. We investigate the detailed physics with observations in which we change the thickness of the absorber and wavelength of the VUV light.

2. VUV-free electron laser experiments

VUV light pulses from an FEL [1] source are used in this study. Nominal parameters of this laser are $10\mu\text{J}$ output energy, 51-61nm wavelength with <1% tunability, and <100fs pulse duration. We use tin (Sn) as a target material for the saturable absorber. According to reference [5], Sn has an N-shell edge in this FEL spectral range. The absorption coefficient above and below the edge are $\alpha_1 = 4.4 \times 10^5 \text{ cm}^{-1}$ and $\alpha_2 = 0.74 \times 10^5 \text{ cm}^{-1}$ respectively. These numbers suggest that large changes in transmission can be obtained with this system if N shell electrons are excited with sufficiently intense light and the number density of lower state for the absorption is reduced. That means we expect 1:20 ratio of transmission for an 80nm thick Sn film before and after saturation. With help of general theory of the saturable absorber in visible laser wavelength [6], the threshold of the saturation I_{th} is $h\nu/\eta\sigma\tau$, where $h\nu$ is photon energy η is quantum efficiency for generation of excited state, and τ is relaxation time of the excited state. We don't have an accurate number for τ but when we focus 100fs-10 μJ VUV laser on a $10\mu\text{m}$ spot, the achievable intensity of 10^{14}W/cm^2 is enough high that if $\tau > 10\text{fs}$, we can expect to observe saturation of the absorption. Actually, the optical properties are still quite uncertain in the VUV range. In addition, surface and bulk contamination and amorphous film conditions cause changes of the energy-level structure in the materials. Even with these uncertainties, the expected effect is strong enough and the estimate points to favorable conditions for a test of this idea.

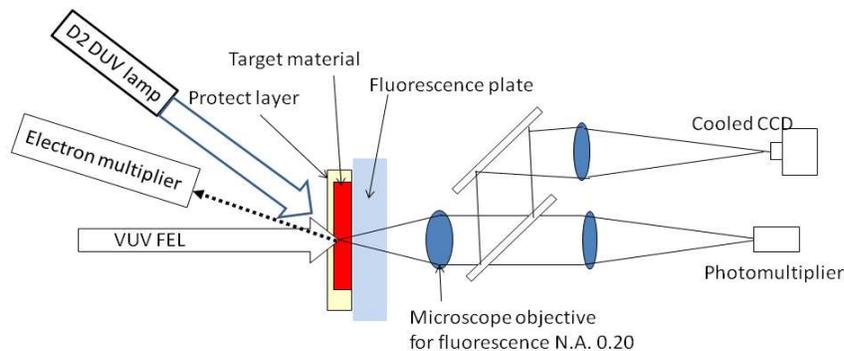


Fig. 1. Schematic drawing of the experimental setup and the target structure for this study. For preventing the surface contamination, we use sandwich target of Au-Sn-SiO₂. Transmission of the VUV laser is monitored by SiO₂ fluorescence while the incident energy is measured by scattered VUV light from the target surface with an electron multiplier. Focusing condition is monitored by a cooled CCD camera. A D₂ DUV lamp is used for removing of the surface contamination after installing the target into high vacuum chamber.

A schematic drawing of the experimental setup is shown in Fig. 1. We use Kirkpatrick-Baez (K-B) microscope optics to focus the VUV laser. The diameter of the spot is $6\mu\text{m}$ (FWHM) and is kept constant during this study. The incident energy on the target was monitored by surface scattered light with an electron multiplier and the energy calibrations are done with a pyroelectric detector. Error in the energy measurement is considered to be 30% in absolute value and 10% for relative value. The target structure for this study is also shown in Fig. 1. In the VUV range, even a thin oxidation contamination layer would strongly affect the experiments. To prevent this problem, we use a multilayered target. The front side of the target is covered with a thin Au film (typical thickness = 5nm) while the rear side is attached to a fluorescent detector material. These layered targets are prepared with a thermal evaporation method and the whole structure is prepared without breaking the vacuum. The estimated loss in the Au layer is about 50% in this thickness. Fused silica glass is used as a fluorescence material. The energy band gap of the SiO_2 is 9.6eV so that in the wavelength range of this study, single photon absorption fluorescence occurs.

For the calibration of this transmission with multilayered target, we prepare three types of targets. These are, (1) bare fused silica substrate, (2) a fused silica substrate coated with only a thin Au layer (5 nm), and (3) a thin Au layer(5nm) + Sn (30, 50, 80nm) + the fused silica. The fluorescence intensity dependence for the Au layer target (2) is almost linear with incident VUV laser energy. The transmission of the Sn layer is decided by signal (3) divided by signal (2) at the corresponding input intensity.

Although we believe this sandwich type target (Au-Sn- SiO_2) idea solves many experimental difficulties (surface contamination of the Sn, difficulty of making a self-supporting ultra-thin film), we should verify that the output fluorescence signal is indeed proportional to the transmitted VUV light. To check this, we also prepared the non-coated fused silica (1) and the results give a good zero point crossing curve. The observed intensity in the case of bare SiO_2 is about half that for the Au coated target. That is a mirror effect for detection of the fluorescence signal from one side: the fluorescence emission is collected with double efficiency for the Au coated sample because the Au- SiO_2 interface is reflecting for visible light. At higher energy range, we observed increasing fluorescence but in the range of this paper, we consider the response is linear within about 15%. Sometimes, in such high energy photon experiments, the effect of photoelectrons should be considered. But in our case, the incident photon energy is 20~25eV and mean free path of such electrons in the metal is about 10nm [7], much less than our Sn target thickness(~80nm). In addition, fused silica requires 9.6eV for excitation of the band gap. Therefore, we consider the photoelectrons have a negligible effect.

3. Nonlinear transmission of the metal

The transmission of the layered target Au-Sn-fused silica as a function of input energy density of 51 nm wavelength is shown in Fig. 2. We observe a rapid change of transmission in the VUV region in the case of 50 and 80nm thickness. The ratio between intensities before and after switching is about 1:100 (above the noise level). The threshold energy density for the 51nm wavelength is $6\text{J}/\text{cm}^2$. When we estimate required energy density E_{req} to excite the atom up to the saturation level whole in this thickness, this number is $E_{\text{req}} = dx * h\nu * N_0 / \eta = 2.3\text{J}/\text{cm}^2$ (with $dx = 50\text{nm}$, $\eta = 0.3$). Considering the absorption inside the Au layer (50%), these numbers are in good agreement with each other. In the case of 30nm thickness, we see the opening intensity of the saturable absorber is much lower than for the 50 and 80nm targets. The penetration of VUV light in the cold condition ($I - I_0 \exp(-\alpha x)$), at least 5~10% energy ($\alpha \sim 10^6 \text{cm}^{-1}$), can reach to 30nm depth point. Therefore, there is a thin-target saturable absorber condition without dynamical penetration during the pulse duration for this thickness. In this case, the opening intensity is determined by $I_{\text{th}} (h\nu/\eta\sigma\tau)$, and if we assume $\tau \sim 130\text{fs}$, the experimental onset time can be explained. Residual absorbance after bleaching seems to be 30% in the case of 30nm. This means that after saturation of the absorption, the absorption coefficient is $1 \times 10^5 \text{cm}^{-1}$. This number roughly matches the initial prediction.

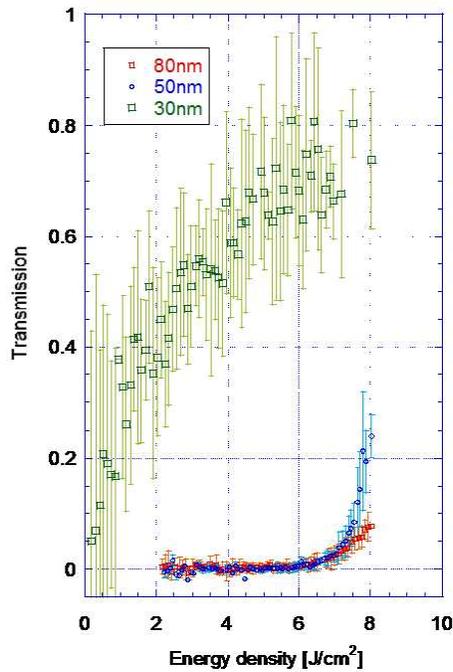


Fig. 2. Large nonlinear response of transmission of the VUV laser. The dot is the average data at the input intensity. The error bar of each data is standard deviation of the measured data.

Figure 3 gives 61nm wavelength results with two different thickness of the Sn layer. The ratio between “switch off” and “switch on” are the same order as that of 51nm result. (The absolute energy calibration for 61 and 51nm has relatively large error. But within 30% energy uncertainty, both the onset intensities are the same.)

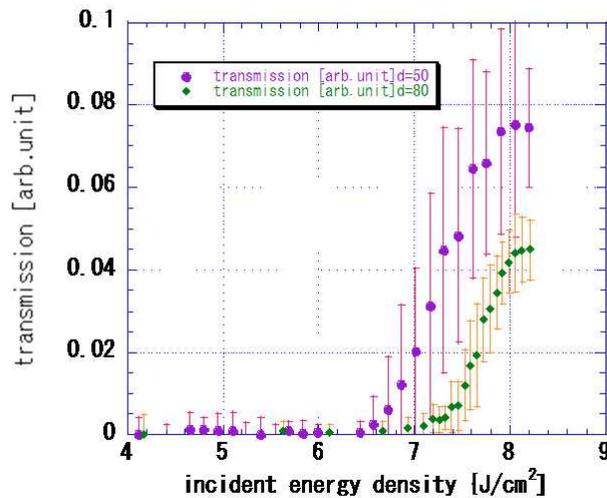


Fig. 3. The switching feature in the case of the 61nm wavelength. There are two different thicknesses of the absorber Sn layer (48nm and 80nm). The ratio of the thresholds for these thickness is 1.1 and differs from the predicted value with a simple edge-shift model in which the ratio should be proportional to the thickness (1.6).

4. Simulations

To understand the experimental data, we use a simple simulation model and estimate the change of the absorption with various parameters. This includes the effective light propagation inside the absorber (like a snow-pile model) and includes spontaneous recombination. The formulas are the following Eqs. (1)–(4):

$$\frac{dI}{dx} = -\sigma_{01}(N_0 - N_1)I - \sigma_{23}(N_2 - N_3)I \quad (1)$$

$$\frac{dN_0}{dt} = -\frac{\eta_{01}}{h\nu}\alpha_{01}I + \frac{N_2}{\tau_{20}} = -\frac{\eta_{01}}{h\nu}\sigma_{01}(N_0 - N_1)I + \frac{N_2}{\tau_{20}} \quad (2)$$

$$\frac{dN_1}{dt} = \frac{\eta_{01}}{h\nu}\alpha_{01}I - \frac{N_1}{\tau_{12}} = \frac{\eta_{01}}{h\nu}\sigma_{01}(N_0 - N_1)I - \frac{N_1}{\tau_{12}} \quad (3)$$

$$\frac{dN_2}{dt} = \frac{N_1}{\tau_{12}} - \frac{\eta_{23}}{h\nu}\alpha_{23}I - \frac{N_2}{\tau_{20}} = \frac{N_1}{\tau_{12}} - \frac{\eta_{23}}{h\nu}\sigma_{23}(N_2 - N_3)I - \frac{N_2}{\tau_{20}} \quad (4)$$

We use a four-level model. N_0 is the ground state population and N_1 is the excited state level of the inner shell ionization of uppermost N-shell electrons. Electrons from N_1 move to a lower energy state (population N_2) with the relaxation time τ_{12} . The energy-level structure of inner-shell ionized atoms at high density is not fully understood. But, the ionization energies of other N-shell electrons are located from 40eV to 100eV in the cold condition. Therefore, we assume there is absorption from the excited state (N_2) to another level whose population is N_3 . The excited state absorption cross-section is taken to be $\sigma_{23}(N_2 \rightarrow N_3)$. Quantitative identification of these levels and determination of their parameters is a task for the future. The α is the absorption coefficient, η is the quantum efficiency to create the excited states with incident photon including other branching loss, $h\nu$ is the photon energy of the incident light, and τ is the spontaneous recombination time. It is not our aim to determine each parameter with this simulation, but with these formulas, we can fit the experimental data with reasonable parameters. One of the fitting results is shown in Fig. 4.

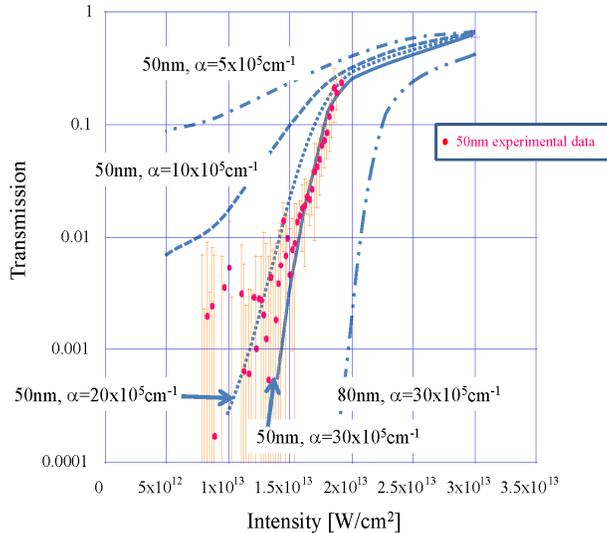


Fig. 4. Simulation results of transmission with various small-signal absorption coefficients. The target thickness is 50nm and 80nm. The all quantum efficiency in this four level model is fixed to 0.3 and the all relaxation time of the first excited state is also fixed to 80fs. We also show the experimental data on this figure.

The model is too simple to describe all the details. For example, we need a more accurate model for light propagation under the steep gradient of propagation parameters. Even with these limitations, the experimental situation can be explained with assumption of $2.0\sim 3.0\times 10^6\text{cm}^{-1}$ absorption coefficient. Although this number is 4~6 times larger than the reference value⁵, an increased absorbance due to contamination introduced during preparation of the films is easily understood. With above model, the speed of bleaching v is estimated with $v = I/h\nu/N$. At the front of the bleaching the penetration into cold material occurs and has a scale length $L = 1/\alpha = 1/N\sigma$. Therefore, the rate of increase of transmission is estimated by $(L/v)^{-1} = N\sigma(I/h\nu/N) = \sigma I/h\nu$. Inserting the numbers, we can expect about 3 fs gating time. This short rise time is obtained due to the high bleaching intensity and the large absorption cross section of small-signal intensity. In addition, it is clear that large absorbance in the small signal condition is important to achieve the large transmission ratio between lower and higher intensity at the threshold. This observed large ratio can provide good prefiltering for high intensity experiments (pedestable cutting in space and time) and measurement of temporal profile of VUV with fast gating method (VUV pump-probe experiments).

Based on an edge shift model in which ionization of N shell electrons causes changed screening of the nuclear charge, we would expect a large difference between 51nm and 61nm according to the ideal cold reference data. However only a small energy difference is observed. Of course, the reference data has a large uncertainty and our target is not ideally pure material. To distinguish from the present experiment, we irradiated a commercial base thin Sn (400nm) foil and checked the small signal transmission. In this case we observed a large difference between 51 and 61nm wavelengths. Our thin samples are amorphous and the band edge structure is typically broadened in an amorphous structure. Additional investigation such as pump-probe experiments will clarify the details.

5. Conclusion

In summary, we have observed a strong nonlinear absorption change of Sn film for wavelengths of 51 and 61nm from a VUV-FEL. The change of the transmission is very rapid and the ratio between before and after switching is about 1:100. The threshold energy of this mechanism is about $6\sim 7\text{J}/\text{cm}^2$ so that we can use this mechanism with good focusing of even a small VUV laser. Although there are still uncertain details about this large nonlinear switching of the absorption, we can use it for new photonic devices in VUV region such as spatial mode cleaning, ultra-pulse slicing to the shorter duration pulse generation.

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