Evaluation of fast EUV scintillator using 13.9 nm x-ray laser

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Abstract. The nickel-like silver x-ray laser with the wavelength of 13.9 nm is used as a high power EUV excitation source to evaluate a scintillator for EUV lithography. The time resolved spectrum of zinc oxide exciton emission at around 380 nm was observed. The determined fluorescence lifetime of 1.1 ns is sufficiently short for characterizing EUV lithography light sources having a few nanoseconds duration. It is also shown that the x-ray laser is an excellent tool for spectroscopic characterization of materials intended for next-generation lithography applications.

1. Introduction
Pulsed EUV sources with high intensity and high spatial coherence promise to be powerful tools for material and biological science. Recently, x-ray lasers are extensively used as coherent extreme ultraviolet (EUV) sources for several measurements, such as scattering [1] and interferometry [2]. The transient collisional excitation (TCE) x-ray lasers have been intensively studied, because they can achieve a high gain of a few tens per centimeter [3, 4] and picosecond pulse duration [5] using a tabletop laser system. The large beam divergence up to 10 mrad, which had been a weak point of the TCE x-ray laser, was dramatically improved to be less than 0.5 mrad using two gain media with oscillator-amplifier configuration [6, 7]. On the other hand, optical technologies around 13.5 nm have been receiving strong interest for next-generation lithography applications [8, 9]. In particular, the development of efficient and fast imaging scintillator devices with sufficient size is a key element for lithographic applications. In these aspects, hydrothermal method grown single crystals of zinc oxide (ZnO) is prominent candidate. ZnO have been intensively studied as light-emitting diode materials [10] and as a result, its growth characteristics have been greatly improved in the aspect of crystalline quality and size of up to 3-inch-diameter [11]. To characterize scintillation property of this material in EUV region, the nickel-like silver TCE laser with the wavelength of 13.9 nm is suitable light source.
Here we describe evaluation of zinc oxide (ZnO) as a fast scintillator for next generation lithography by observation of time resolved fluorescence spectrum excited with 13.9 nm x-ray laser.

2. Experiments

2.1. X-ray laser
The x-ray laser operating at 13.9 nm was employed as the excitation source. The lasing scheme is the 4p-4d transition of the nickel-like silver ion pumped with TCE [7]. The gain medium plasmas of the x-ray laser were generated with a chirped pulse amplification Nd:glass laser system with two beam lines [12]. Each beam consisted of two pulses; a pre-pulse with a pulse duration of 200 ps and a 3 ps heating pulse with a temporal delay of 2 ns from the pre-pulse. The wavelength of pumping laser is 1053 nm. The laser pulses are focused with a line shape of 6.5 mm × 20 μm on flat silver targets with an irradiance of the heating pulse of ~10^15 W/cm². Two targets separated for 20 cm were used to improve the energy and the beam divergence of the x-ray laser; a part of the x-ray laser generated in the first medium is used as the seed x-ray laser, and it is injected into the successive second medium, which is used as an amplifier with calm density gradient [6]. The typical output energy of the x-ray laser emission was 0.5 μJ, and the beam divergence was obtained to be 0.5 × 2 mrad. The pulse duration was 7 ps, which is sufficiently short for this experiment.

2.2. Samples
The single crystal sample of ZnO was grown by hydrothermal method combined with a platinum inner container [11, 13]. Using this method, low defect and large sized single crystals of 3 inch ZnO was successively grown. High-purity and transparent single crystal was sliced with a (0001) surface orientation.

2.3. Measurement of time-resolved spectrum
The experimental setup is shown in Figure 1. The x-ray laser was focused on the sample using a molybdenum/silicon multilayer spherical mirror suitable for 13.9 nm. In spite of the huge photon energy compared with the bandgap, no visible color center was observed after about 50 shots irradiation. The damage threshold for 10 ns and 355 nm pulses is over 1 J/cm². To eliminate continuous emission from the plasma, a 0.2 μm-thick zirconium foil was placed before the EUV mirror. The fluorescence spectrum and the fluorescence lifetime of a sample were measured using the 25 cm-focal-length spectrograph coupled with a streak camera. The temporal resolution of the streak camera is 100 ps in the fastest scanning range. The trigger pulse of the streak camera was provided by

![Figure 1. Experimental setup for the measurement of time resolved spectrum with UV and EUV excitation](image-url)
a pulse generator, which also served as the master clock of the x-ray laser. To compare with EUV excitation, the scintillation properties were also evaluated using the 351 nm third harmonics from the 1053 nm chirped pumping source for the x-ray laser, the ZnO was excited at an energy slightly above the bandgap. The pulse duration of the 351 nm excitation source was measured to be 110 ps.

3. Results and Discussion

Figure 2(a) shows the streak image and the time profile at the peak of the spectrum [14]. The figure is integrated for 3 frames to reduce the noise level, however one shot of x-ray laser was enough to obtain a clear image of time-resolved fluorescence spectrum. The output energy level of the EUV laser was not very stable. The spectral shape and the response time were almost identical even for rather scattered excitation pulse energy, although it was not monitored simultaneously. The time profile of the fluorescence can be expressed by double exponential decay with time constants of 1 ns and 3 ns. The two decay constants have been measured in several works for UV excited ZnO single crystals, the fast decay is the lifetime of free exciton and the slower decay is assigned to be trapped carriers [15]. The corresponding streak image and the time profile of UV excitation is shown in Figure 2(b). In both the excitation conditions, a prominent fluorescence peak of the ZnO exciton transition was observed at around 380 nm. This wavelength is still convenient for high resolution imaging devices, since even BK7 glass is transparent at this wavelength. Moreover, the two decay lifetimes observed in both cases were almost similar regardless of the huge difference in the excitation photon energy. The

![Figure 2](image)

**Figure 2.** Streak camera images and temporal profiles of the fluorescence from ZnO, which are excited by a) x-ray laser pulses (13.9 nm) and b) UV laser pulses (351 nm). The temporal resolution of this measurement is 0.8 ns. The observed profiles can be fitted by double exponential decays described as $I = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$, the solid lines in the figures. The fitting parameters for EUV excitation are $A_1 = 0.65$, $A_2 = 0.35$, $\tau_1 = 1.1$ ns, $\tau_2 = 3.5$ ns, and for UV excitation are $A_1 = 0.60$, $A_2 = 0.40$, $\tau_1 = 0.9$ ns, $\tau_2 = 3.2$ ns. The time constant and ratio of the two decay components are almost identical in both the excitation conditions.
fluorescence lifetime is sufficiently short for the characterization of the laser plasma EUV source with nanoseconds duration for lithographic applications. It is also shown that nickel-like silver x-ray laser is an excellent tool for spectroscopic characterization of materials intended for EUV lithography applications.

4. Conclusion

In conclusion, we have successfully observed the time resolved spectrum of ZnO excited with 13.9 nm x-ray laser. It is shown that nickel-like silver x-ray laser is an ideal tool for characterization of materials intended for next-generation lithography applications. The excellent properties of ZnO as a scintillation material for characterizing EUV lithography light sources are also shown. The availability of large-sized ZnO crystal up to 3-inch is quite attractive for future lithography and imaging applications.

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