Efficient “water window” soft x-ray high-Z plasma source

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Abstract. Unresolved transition array (UTA) is scalable to shorter wavelengths, and we demonstrate a table-top broadband emission “water window” soft x-ray source based on laser-produced plasmas. Resonance emission from multiply charged ions merges to produce intense UTAs in the 2 to 4 nm region, extending below the carbon K edge (4.37 nm). An outline of a microscope design for single-shot live cell imaging is proposed based on a bismuth (Bi) plasma UTA source, coupled to multilayer mirror optics.

1. Introduction
Development of shorter wavelength sources in the extreme ultraviolet (EUV) and soft x-ray spectral regions has been motivated by their application in a number of high profile areas of science and technology. One such topic is the challenge of three-dimensional imaging and single-shot flash photography of microscopic biological structures, such as cells and macromolecules, in vivo [1]. For x-ray microscopy, the x-ray source should emit a sufficient photon flux to expose the image of the biological sample on the detector. To date the most practical light source of high-power, high-brightness x-rays has been radiation from synchrotrons and more recently from free electron lasers (FEL) [2]. Table-top sources using ethanol sprays and liquid nitrogen droplets are being developed for use with zone plates for transmission microscopy. Recently λ = 2.48 nm narrowband emission from a liquid-nitrogen-jet laser-plasma was successfully combined with state-of-the-art normal-incidence multilayer condenser optics and 20-nm zone-plate imaging optics to demonstrate laboratory water-window x-ray microscopy with resolution less than 40 nm [3]. The total collected energy, however, is low, when one combines the narrow line emission with the low reflectivity of the collector mirror. As a result long exposures are needed to record an image and there is not yet published evidence of single-shot exposures using a laboratory-scale source. To overcome the low efficiency imposed by line sources, we propose using high power water-window emission from laser-produced high-Z plasmas, analogous to the scheme used for efficient, high-volume manufacturing EUV sources.

Before discussing the high-power water window source, it is important to summarize the characteristics of efficient UTA light sources used in the 5 to 15 nm region. All are based on n = 4−n = 4 (4d–4f and 4p–4d) transitions that overlap to generate an intense UTA. For efficient 13.5-nm operation, which corresponds to a photon energy hν ≈ 92 eV, it is important to produce an optimum plasma electron temperature of 30–50 eV. The rare-earth elements of gadolinium (Gd, Z = 64) and terbium (Tb, Z = 65) produce strong emission near λ = 6.7 nm (hν = 183 eV) which is maximized at electron temperatures in the 100–120 eV range depending on initial focusing conditions [4-6]. The spectral behavior of Gd and Tb plasmas is expected to be largely similar to that of Sn plasmas, because in the temperature range of interest, both are dominated by 4d open-shell ions. Although the...
conversion efficiency (CE) from the input laser energy to the output in-band EUV emission energy depends on the bandwidth (BW) of the reflection coefficient of the multilayer mirror (MLM), the maximum CEs have been observed to be higher than 1%. Because it moves to shorter wavelength with increasing atomic number, Z, the \( n = 4 - n = 4 \) UTA is expected to lie in the water window if higher Z elements from \( Z = 79 \) (Au) to \( Z = 83 \) (Bi) are used [7]. Higher Z elements such as uranium also emit in the water window but their radioactivity prohibits their use. Much of the previous work on high-Z plasmas has concentrated on the production of quasicontinuum spectra at moderate laser intensities, employing electron temperatures below 100 eV [8,9].

In this paper, we report the demonstration on the efficient "water window" soft x-ray source by strong UTA band emission in laser-produced high-Z plasmas. Our proposed procedure for producing the water window emission is expected to be efficient and scalable in output yield. Our calculations show that a bismuth plasma at an electron temperature in the range 570 to 600 eV radiates strongly near 3.9 nm. At electron temperatures above 800 eV, strong UTA emission around 3.2 nm would be expected.

2. Experimental results and discussion

Because it moves to shorter wavelength with increasing Z, the \( n = 4 - n = 4 \) UTA can be used for other applications, such as transmission x-ray microscopy for biological imaging in the water window (Fig. 1). We have made preliminary studies of the potential of Bi as the "water window" soft x-ray source.

![Figure 1. Calculated position of \( n = 4 - n = 4 \) transitions in key ions in elements from indium (Z = 49) to uranium (Z = 92). The localization of emission near 3.2 and 3.9 nm in Bi is clearly evident.](image)

Figures 2(a) and 2(b) show the time-integrated Bi spectra and the laser intensity dependence of the peak photon energies of \( n = 4 - n = 4 \) and \( n = 4 - n = 5 \) transitions. The position of the \( n = 4 - n = 4 \) transition peak was observed to be unchanged. The position of the \( n = 4 - n = 5 \) peak, on the other hand, shifts to higher photon energy with increasing laser intensity because of the rise in electron temperature and associated charge states. However, the strong emission expected around 3.2 nm, which originates from the \( n = 4 - n = 4 \) UTA, was not observed in this laser intensity region [10].

The emission intensity of the \( n = 4 - n = 5 \) transitions was compared with that of the \( n = 4 - n = 4 \) UTAs. The strong emission at 3.15 nm due to the \( n = 4 - n = 4 \) UTA in Bi plasmas may be coupled with a Sc/Cr MLM with a reflection coefficient of 15% [11]. The variation of this emission with electron temperature was calculated with Cowan's suite of atomic structure codes in order to predict its evolution with increasing laser flux [12]. The predicted spectral evolution as a function of electron temperature is summarized in Fig. 3(a). In this figure the spectra generated for each ion stage are based on excited state populations that allow for electron temperature assuming a Boltzmann distribution. These spectra are then weighted by an ion fraction appropriate to that temperature,
calculated assuming collisional-radiative (CR) equilibrium to yield the distributions shown [13]. Our calculations show that high-Z plasmas, at an electron temperature lower than 700 eV, as shown in Fig. 3(b), radiate strongly near 3.9 nm. In the case of higher electron temperatures, from 800 to 1500 eV, the strongest emission is expected at around 3.2 nm [10], suitable for coupling to Sc/Cr MLMs [11]. Thus for an optimized source, we should produce a plasma at a higher electron temperature plasma of around 1 keV. The intensity of the Bi plasma emission in our experiment was compared with 2.48-nm nitrogen line emission from a Si₃N₄ planar target, in the same experimental setup, and was observed to be 1.2 times higher within a bandwidth of 0.008 nm (FWHM) [14] even though the plasma electron temperature was much lower than the optimum value.

Figure 2. Spectral behaviors of the Bi plasmas in the laser intensity dependence on the observed emission spectra (a), the peak wavelength of the \( n = 4 - n = 4 \) transition (circles) and the \( n = 4 - n = 5 \) transition (rectangles) (b), respectively.

Figure 3. Calculated spectral variation as a function of electron temperature (a) and calculated spectra for electron temperatures higher than 900 eV (b), respectively.

3. Proposal of the UTA source microscope for single shot photograph
Taking the experimental and numerical results into account, we now propose a high brightness, high power water window source for single shot imaging at the laboratory scale [10]. Our proposed method has the advantage that the EUV energy efficiency and atomic number dependence can be scaled based on the fundamental properties of the plasma source. To produce not only a Bi plasma with a high electron temperature of the order of 1 keV but also one that has low density and is optically thin, we should switch to a CO₂ laser operating at a wavelength of 10.6 µm due to the low
critical density of $1 \times 10^{19}$ cm$^{-3}$ attainable, with a pulse duration to permit ionization to the appropriate ion stages (Bi$^{36+}$–Bi$^{45+}$) i.e., 150 ps, while at the same time maintaining a laser intensity of the order of $10^{13}$ W/cm$^2$. In addition, for this proof of principle, we propose a dot Bi target with a diameter less than 10 $\mu$m with a thickness less than 1 $\mu$m to generate a microplasma (to fulfill the requirements for high brightness and a point source) as the expected focal spot size will be the order of 100 $\mu$m in the case of the CO$_2$ laser wavelength of 10.6 $\mu$m. Because of the broadband nature of the emission, zone plate components cannot be used, so one possible solution would be to use a transmission planar x-ray nano-waveguide to image the sample [15]. To achieve high resolution in the recorded image, we should also switch the recording device from x-ray CCD cameras to sensitive EUV resists to overcome the resolution limitation of the CCD pixel size, coupled with Schwarzschild optics, consisting of Sc/Cr MLMs with a reflection coefficient of the order of 15% around 3.2 nm [11]. Although our proposal is based on a simple microscope construction, the key component is the UTA emitted at 3.2 nm from a hot dense Bi plasma point source, combined with Sc/Cr MLMs and sensitive EUV resists based on photochemical reaction [16,17].

4. Summary
In summary, we have demonstrated high-efficiency emission in the water window spectral region based on laser-produced Bi plasmas, and have proposed methods to increase it still further. Resonance emission from multiply charged ions merges to produce intense UTA, extending to wavelengths below the carbon K edge. The overall spectral behavior is well described by simulations. The experimental results also provide an outline for the design concept for single-shot cell imaging with a novel microscope optical system. The method presented here opens the way for applications in next-generation biological science.

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