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Nonlinear Optics with Focused X-ray Lasers

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Abstract

We have investigated the possibility of focusing x-ray lasers with the use of multilayered mirrors or zone plates. The results indicate that x-ray intensities as high as 10^{14} W/cm^2 can be achieved by focusing saturated Ne-like x-ray lasers. These intensities should be adequate for studying nonlinear optical phenomena.

To date x-ray laser research has been primarily motivated by the possibility of producing x-ray holograms of biological specimens[1]. However, with the high powers achievable with current x-ray laser systems we can consider a greater variety of applications. These range from using focused x-ray lasers to irradiate solids and produce high density plasmas to studying nonlinear optical phenomena in the x-ray regime. In this paper we discuss several different approaches to focusing x-ray lasers. The benefits and disadvantages of each in the context of studying such nonlinear phenomena as harmonic generation and four-wave mixing will be discussed.

There are three distinct approaches to focusing soft x-rays. These consist of grazing incidence optics, Fresnel zone plates and normal incidence multilayered mirrors. The first microscope in the x-ray region was built by Kirkpatrick and Baez[2] and used two orthogonal spherical mirrors at grazing incidence. Two mirrors are required because single mirrors suffer from severe astigmatism. An improvement over this configuration was proposed by Wolter and utilizes aspheric mirrors to correct for spherical aberrations. Although 1-2 µm resolution has been demonstrated with Wolter systems their need for accurate aspheric optics makes the cost prohibitive. Furthermore, grazing incidence optics are inherently broadband. For focusing an x-ray laser this is unnecessary and leads to unwanted background.

Fresnel zone plates currently offer the best resolution of any x-ray optic. Manufacturing techniques are now capable of making zone plates with outer ring widths of ~350 Å (the outer ring width is the diffraction limited resolution). An additional advantage of zone plates is that they can operate at wavelengths below the carbon absorption edge. This is important in microscopy applications of x-ray lasers. Unfortunately, however, zone plates suffer from size and throughput limitation. The largest zone plates suitable for x-ray laser applications are less than 100 µm in diameter. This implies that the zone plate has to be a few centimeters from the x-ray laser to collect a significant fraction of the 10 mrad divergence beam. The zone plate would likely not survive the event and require replacement. The diffraction efficiency into first order is calculated to be ~10% but values of 5% are typically observed.

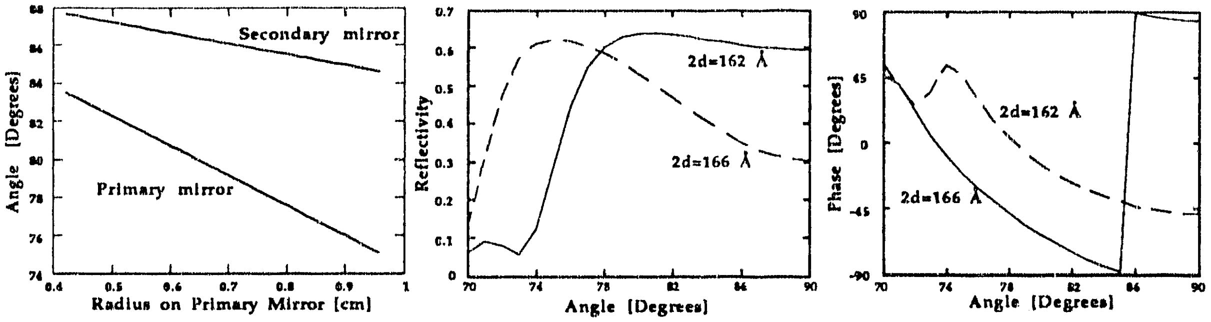

Figure 1 a) Variation in incident angle on 40X Schwazrschild system. The change in reflectivity b) and phase angle c) as a function of incident angle for two different multilayer mirror 2d spacings.

The possibility of focusing x-ray lasers with normal incidence optics has only become feasible in recent years due to the advances in multilayer mirror technology[3]. Curved multilayer mirrors can routinely be coated for high normal incidence reflectivity at wavelengths extending down to the carbon absorption edge (~44.Å). Optic systems with normal incidence optics can use a combination of spherical mirrors to eliminate the aberrations associated with grazing incidence optics. Such a system is the Schwarzschild microscope[4] which makes use of a convex and concave mirror as shown in figure 1. A Schwarzschild microscope was used by Trail[5] at Stanford university to focus 140 Å x-rays from a laser produced plasma. This microscope had demagnification of 100X and achieved spatial resolution of 0.5 µm. Our initial designs for focusing an x-ray laser also considered a 100X demagnification Schwarzschild optics. However, detailed ray tracing of these systems indicated that current multi-layer deposition techniques preclude such high demagnification systems because of the wavelength mismatch between the primary and secondary mirror. That is, the bandpass of each mirror varies across the surface due to the change in multilayer spacing and incident angle. This effect is enhanced for systems with small radius of curvature and monochromatic sources. As a result we have designed a 40X demagnification system for our initial experiments. In figure 1a we illustrate the range of incident angles on the primary and secondary mirrors. A uniform 2d spacing on these optics would greatly reduce the overall throughput as is shown in figure 1b. In order to overcome this Kortright et al.[6] have investigated coating schemes to vary the 2d spacing across the curved mirrors. Their results suggest that mismatches of the order of half of the calculated reflectivity FWHM are possible. This would correspond to a system throughput of >50% of that calculated for perfect mirror overlap. Another consequence of varying incident angle (or 2d spacing) is the change in phase shift across the mirror surface (figure

1c). This can have serious implications for diffraction limited imaging. Phase shifts differences of π occur across the Bragg peak. This necessitates that the coating be adjusted so that the x-ray laser wavelength is always on the same side of the Bragg peak. Measurements of the throughput and focusing capabilities of a Schwarzschild optic are planned for the near future.

As a testbed for focusing and nonlinear optics experiments we have begun a detailed study of the neon-like yttrium x-ray laser. This laser offers several distinct advantages for future applications. Firstly , The 155 Å wavelength is ideally suited for multilayer mirror technology. Mirrors with normal incidence reflectivities as high as 60% have been demonstrated. Secondly, the short wavelength J=2-1 laser line dominates the emission spectrum. This contrasts with other neon-like x-ray lasers[7] where the two J=2-1 transitions are comparable With appropriate multilayer mirrors and filters a near monochromatic source is possible. A recent power measurement using a x-ray diode saturated indicating an x-ray laser power in excess of 5 MW. Using a Schwarzschild 40X demagnification optic we can achieve intensities of 10^{14} W/cm^2 in a ~2 μm diameter spot.

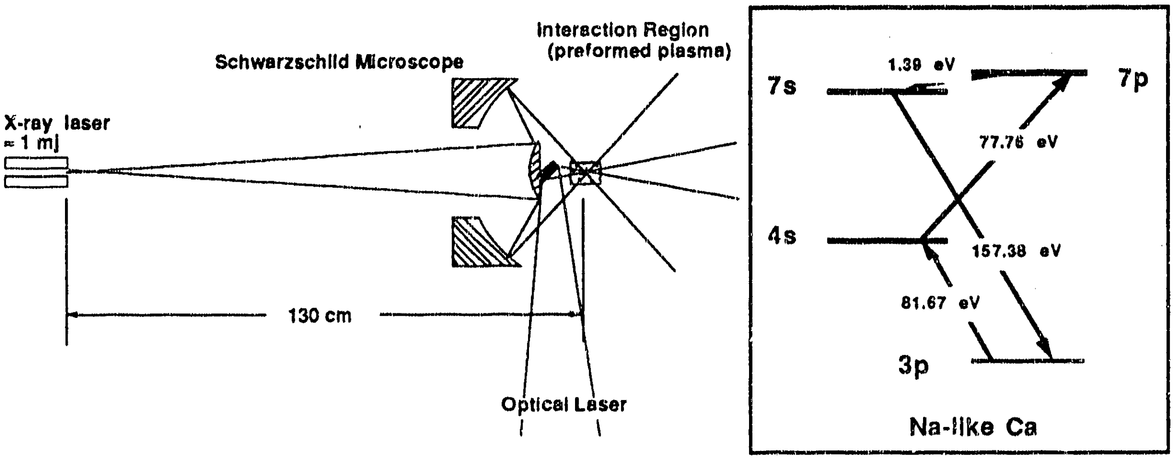

Figure 2 Experimental setup for focusing x-ray laser and energy levels of a system suitable for four wave mixing with a yttrium x-ray laser (80 eV).

A unique application of this system is four wave mixing as an approach for producing a tunable x-ray laser. Figure 2 illustrates this concept. An optical laser beam can be used to preform the plasma and mix with the x-ray laser beam. Numerous experiments have demonstrated multiphoton processes in the near UV by focusing optical lasers through neutral gases[8]. To extend these experiments to the soft x-ray region a plasma is necessary both to reduce absorption and to allow for resonant enhancement of the nonlinear susceptibility. The isotropy of the plasma forces us to consider only odd-order processes of which general four-wave mixing

$$\omega_1 \pm \omega_2 \pm \omega_3 = \omega_4 \qquad (1)$$

is the lowest order. The conversion intensity for such a process scales as

$$I_4 \sim \omega^2 \, |\chi^{(3)}| \, I_1 I_2 I_3 \qquad (2)$$

where $\chi^{(3)} \sim \omega^{-5}$ is the nonlinear susceptibility. The strong scaling with energy requires either high x-ray laser intensities or that $\chi^{(3)}$ be maximized in order to improve conversion. For example, $\chi^{(3)}$ can be increased by several orders of magnitude by selecting a plasma where each photon transition is near resonance. This makes third harmonic generation more difficult since a plasma system with four dipole-connected levels, evenly spaced in energy is difficult to find. It appears therefore that sum/difference frequency conversion with two x-ray photons at ω_1 and an optical photon at ω_3 may offer the best possibility for success. Furthermore, by varying ω_3 this approach can lead to an x-ray laser tunable over a few eV.

Recently, Muendel and Hagelstein[9] studied in detail the conversion efficiency for the process $2\omega_1 - \omega_3 = \omega_4$ in a Na-like potassium plasma. In their scheme resonant enhancement of $\chi^{(3)}$ is produced by using x-ray photons which are near resonance for the 3p-4s and 4s-7p transitions. An infrared photon at ω_3 (1020 nm) then polarizes the 7p-7s producing a field at nearly double the original x-ray laser frequency . They calculate significant increases in $\chi^{(3)}$ at the single and two-photon resonances (three orders of magnitude). Unfortunately photon absorption ultimately limits how close to resonance we can operate. An analog of this system suitable for a yttrium x-ray laser is Na-like Ca (see figure 2). Preliminary experiments to evaluate the feasibility of performing nonlinear optics with this system are planned in the near future.

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