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Multiply ionized carbon plasmas with index of refraction greater than one

J. FILEVICH,[1] J. GRAVA,[1] M. PURVIS,[1] M.C. MARCONI,[1] J.J. ROCCA,[1] J. NILSEN,[2] J. DUNN,[2] AND W.R. JOHNSON[3]

[1]NSF ERC for Extreme Ultraviolet Science and Technology, Colorado State University, Fort Collins, Colorado
[2]Lawrence Livermore National Laboratory, Livermore, California
[3]University of Notre Dame, Indiana

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Abstract

For decades the analysis of interferometry have relied on the approximation that the index of refraction in plasmas is due solely to the free electrons. This general assumption makes the index of refraction always less than one. However, recent soft x-ray laser interferometry experiments with Aluminum plasmas at wavelengths of 14.7 nm and 13.9 nm have shown fringes that bend the opposite direction than would be expected when using that approximation. Analysis of the data demonstrated that this effect is due to bound electrons that contribute significantly to the index of refraction of multiply ionized plasmas, and that this should be encountered in other plasmas at different wavelengths. Recent studies of Silver and Tin plasmas using a 46.9 nm probe beam generated by a Ne-like Ar capillary discharge soft-ray laser identified plasmas with an index of refraction greater than one, as was predicted by computer calculations. In this paper we present new interferometric results obtained with Carbon plasmas at 46.9 nm probe wavelength that clearly show plasma regions with an index of refraction greater than one. Computations suggest that in this case the phenomenon is due to the dominant contribution of bound electrons from doubly ionized carbon ions to the index of refraction. The results reaffirm that bound electrons can strongly influence the index of refraction of numerous plasmas over a broad range of soft x-ray wavelengths.

Keywords: Anomalous index of refraction; Interferometry; Soft X-ray laser

1. INTRODUCTION

For many decades optical interferometers have been used to measure the electron density of plasmas, using the assumption that the index of refraction of plasma is due only to the free electrons (Attwood *et al*., 1978; Tallents, 1984; Griem, 1997), an assumption that implies that the index of refraction in the plasma should always be less than one. With the objective of probing plasmas of higher densities, over the last decade, several interferometers (Da Silva *et al*., 1995; Rocca *et al*., 1999; Filevich *et al*., 2000; Smith *et al*., 2002; Albert *et al*., 1997; Descamps *et al*., 2000; Tang *et al*., 2004; Dunn *et al*., 2005) have been built to perform dense plasma diagnostics, using probe wavelengths in the range of 14 to 72 nm (photon energies 89 to 17 eV), which is a very attractive regime for laser and discharge generated by soft X-ray lasers (Mocek *et al*., 2005; Fiedorowicz, 2005; Neumayer *et al*., 2005; Wagner *et al*., 1996; Kuroda *et al*., 2005). Since the plasmas being studied were highly ionized, the analysis of the experiments done with these sources assumed that only the free electrons contributed significantly to the index of refraction. Recent interferometry experiments of laser-produced Aluminum plasmas conducted using probe wavelengths of 14.7 nm (Filevich *et al*., 2005) and 13.9 nm (Tang *et al*., 2004), observed interference fringes that bent in the opposite direction than was expected, indicating that the index of refraction was greater than one. The analysis of the experiments showed that the anomalous dispersion from the resonance lines and absorption edges of the bound electrons have a large contribution to the index of refraction with the opposite sign as that of the free electrons (Filevich *et al*., 2005; Nilsen & Scofield, 2004). A significant result of the calculations is that the influence of the bound electrons on the index of refraction extends far from the absorption edges and resonance lines, affecting a broad range of wavelengths.

Address correspondence and reprint requests to: Jorge Filevich, 1320 Campus Delivery, Colorado State University, Fort Collins, CO 80523. E-mail: rage@engr.colostate.edu

The traditional formula that assumes only free electron contribution to the index of refraction of a plasma is $n = (1 - N_{elec}/N_{crit})^{1/2}$ where N_{elec} is the electron density of the plasma and N_{crit} is the plasma critical density. At wavelength λ, $N_{crit} = \pi/(r_0\lambda^2)$ where r_0 is the classical electron radius, 2.818×10^{-13} cm. In typical experiments, the electron density is much less than the critical density so the formula above can be approximated by $n = 1 - (N_{elec}/2N_{crit})$. For a plasma of length L, the number of fringe shifts observed in an interferometer equals

$$N_f = \frac{1}{\lambda}\int_0^L (1-n)\,dl$$

For the case of a uniform plasma along the direction of propagation of the probe beam, the above formula simplifies to $N_f = (1 - n)L/\lambda$. Substituting the approximation described above for the index of refraction, the number of fringe shifts N_f equals $(N_{elec}L)/(2\lambda N_{crit})$. To obtain electron density information from the interferogram, the fringe shifts are measured and then converted to electron density using the above approximations. Because the index of refraction is assumed to be always smaller than one, the fringes should always shift in one direction, determined by the geometry of the interferometer. From the anomalous fringe shift results in the interferometry experiments (Filevich *et al.*, 2005; Tang *et al.*, 2004) of the Al plasmas, it is clear that these assumptions used to analyze the VUV to soft X-ray interferometry are not always valid, and that the bound electron contribution have to be taken into account in some cases. We have been exploring different plasmas trying to identify conditions in which the bound electron contribution is important. Aided by computer calculations and confirmed experimentally, we found that at 46.9 nm lowly ionized Silver and Tin plasmas have an index of refraction greater than one. In this paper, we present new results of interferograms obtained in Carbon plasmas that clearly show regions with an index of refraction greater than one, both at early and late times in the evolution. Computer calculations suggest that the main contributors to the index of refraction are doubly ionized carbon atoms.

2. INTERFEROMETER EXPERIMENTS WITH CARBON PLASMAS

The experiments were performed by combining a 46.9 nm table-top Ne-like Ar capillary discharge laser (Benware *et al.*, 1998) with an amplitude division diffraction grating interferometer (DGI) (Filevich *et al.*, 2000, 2004). The DGI is set in a skewed Mach–Zehnder configuration using diffraction gratings as beam splitters. By selecting the blaze angle on the gratings, the light is diffracted with equal intensity (25% of the incident intensity) to zero and first orders, forming the two arms of the interferometer that are reflected at a grazing incidence angle toward a second diffraction grating using two 35 cm long gold-coated mirrors. The second diffraction grating recombines the two beams such that they exit the interferometer propagating with a small angular difference, selected to produce fringes on the spacing required by the experiment. The Carbon target used to generate the plasma was placed to intersect the zero order path of the interferometer at a location between the long mirror and the second diffraction grating. The plasma was imaged with ~25× magnification using Sc/Si multilayer optics (Uspenskii *et al.*, 1998) onto a MicroChannel Plate Charge-Coupled Device (MCP-CCD) detector combination. The compact 46.9 nm Ne-like Ar capillary discharge-pumped laser produces light pulses of ~1 ns duration and energies of ~0.1 mJ. The high brightness of this source helps overcome the self-emission from the laser-created plasma. The very high beam spatial coherence (Liu *et al.*, 2001) produces high quality interferograms with high fringe visibility when combined with the DGI. The soft X-ray laser was laser triggered achieving a measured jitter of ~1–2 ns. The interferometer and the alignment procedure are described in more detail in a previous publication (Filevich *et al.*, 2004).

The plasma was generated by irradiating a 1 mm long, 500 μm in diameter semi-cylindrical 99.99% pure Carbon target with an 800 nm wavelength laser pulse of 120 ps (FWHM) and up to 0.6 J of energy from a Ti:Sapphire laser. A line focus ~1.7 mm long and ~300 μm wide, resulting in an irradiance of $\sim 1 \times 10^{12}$ W cm^{-2} was formed at the target plane using the combination of a 7 m focal length spherical lens, and a variable cylindrical lens used to adjust the beam astigmatism. The line focus shape and intensity at the target plane were monitored on every shot by imaging the reflection off a 4% beam splitter onto a CCD camera. This target geometry combined with a relatively wide line focus irradiation generates a hot dense plasma on the axis of the cavity.

Figure 1 show interferograms corresponding to two different times during the evolution of Carbon plasmas. The 5 ns frame shows interference fringes with maximum shifts on axis due to the convergence of the plasma produced by irradiating the walls of the semi-cylindrical target. The probe beam is strongly absorbed close to the target surface, due to the presence in this region of a large density of lowly charged ions with ionization potential less than the 26.44 eV, the photon energy of the Ar soft X-ray laser. It is noticeable that the fringes closer to the target, at the bottom of the semi-cylindrical groove, shift to the left of the reference fringes (black lines over-imposed on the image), which means that the index of refraction is greater than one. Later in time, in the 15 ns frame in Figure 1, there is not much fringe shifts except on the semicylinder's axis, and these fringes all bend toward the left of the reference fringes. The region where the probe beam is absorbed is larger and now completely fills the target cavity. The region with anomalous fringe shifts is always close to the region with higher absorption, suggesting that the low ionized atoms are the cause of this anomalous index of refraction.

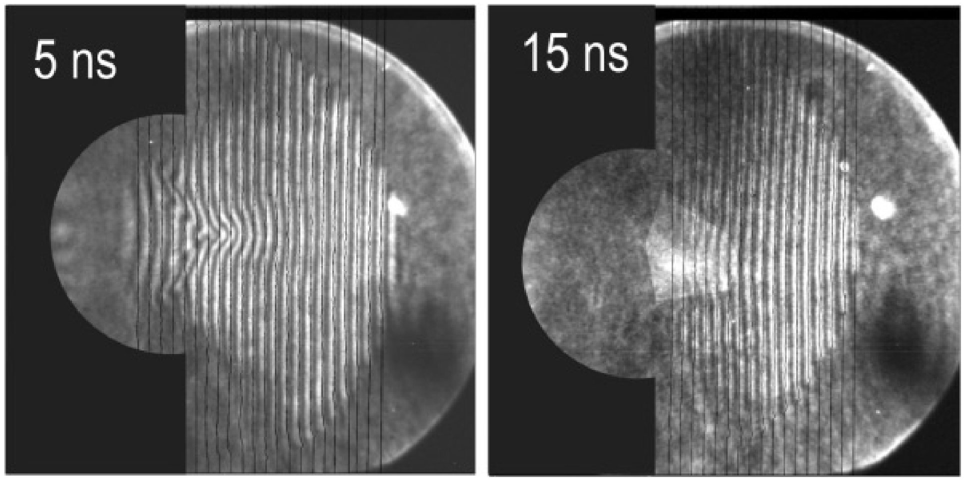

Fig. 1. Soft X-ray interferograms taken at 5 and 15 ns in the evolution of a carbon plasma created inside a 500 μm diameter semi-cylindrical groove. The black lines represent the position of the reference fringes. On the 5 ns shot the anomalous fringe shift are observed at the bottom of the groove while on the 15 ns shot the anomalous fringes are observed on the semi-cylinder's axis.

Figure 2 shows two-dimensional maps of the number of fringe shifts observed in these two interferograms. The 5 ns frame shows that at the axis of the cylindrical groove, the plasma produces approximately one fringe shift which would correspond to $\sim 5 \times 10^{19}$ cm^{-3} if only free electrons contributed to the index of refraction. We also observe that close to the absorption region, inside the groove, and closer to the target, the plasma produces approximately one negative fringe shift, indicating an index of refraction greater than one. Later in time, the 15 ns frame shows very little fringe shifts overall except on the axial region that previously had the highest density. In this case, almost one entire fringe shift is observed (-0.8 fringes).

3. INDEX OF REFRACTION CALCULATIONS

To enable us to calculate the index of refraction for any plasma at any wavelength, we used a modified version of the INFERNO average atom code. The INFERNO code (Liberman, 1982) has been used for many years to calculate the ionization conditions and absorption spectrum of plasmas under a wide variety of conditions. For finite temperatures and densities, the INFERNO code calculates a statistical population for occupation of one-electron Dirac orbitals in the plasma. We use a non-relativistic version of INFERNO in this work to calculate bound and continuum orbitals, and the corresponding self-consistent potential. By applying linear response theory, we obtain an average-atom version of the Kubo–Greenwood equation (Greenwood, 1958; Kubo, 1957) for the frequency-dependent conductivity of the plasma. The imaginary part of the complex dielectric function is proportional to the conductivity. The real part of the dielectric function can be found from its imaginary part using a Kramers–Kronig dispersion relation. The details of the Kubo–Greenwood formula applied to the average-atom model are described in Johnson *et al*. (2006). For modeling purposes, we choose a Carbon plasma with an ion density of 10^{20} cm^{-3}. By varying the temperature of the plasma through the values

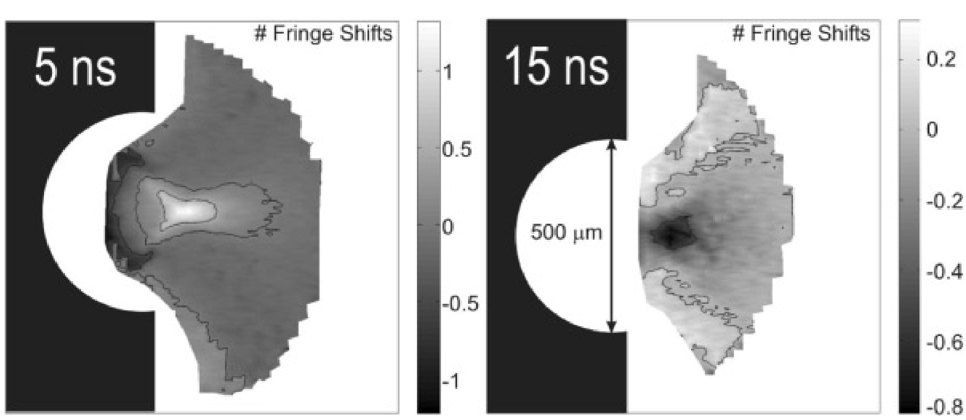

Fig. 2. Number of fringe shifts computed from the interferograms in Figure 1.

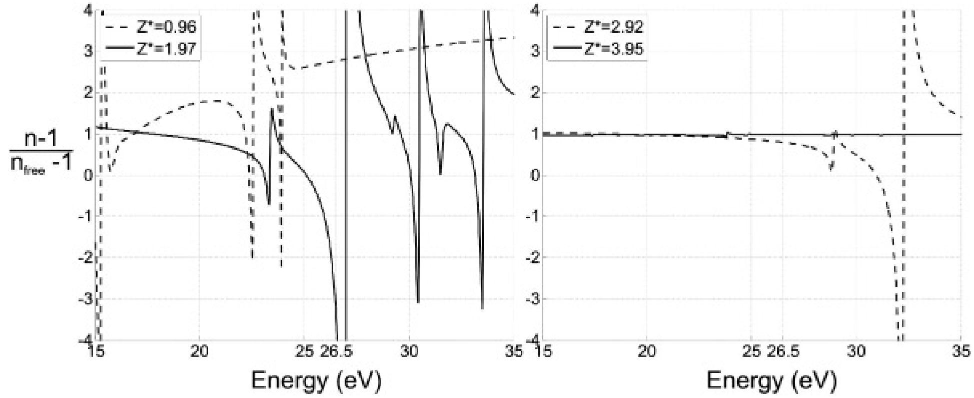

Fig. 3. Index of refraction for Carbon plasmas computed using a modified average atom code based on the INFERNO code. The Ar soft X-ray laser has a photon of 26.44 eV.

3, 6, 10, and 30 eV, we can find the conditions where the Carbon plasma's mean ionization would be $Z^* = 0.96$, 1.97, 2.92, and 3.95. The ionization potentials of neutral Carbon, C^{1+}, C^{2+}, C^{3+}, and C^{4+} are 11.26 eV, 24.38 eV, 47.89 eV, 64.49eV, and 392 eV, respectively. The results of the calculations are plotted in Figure 3. The challenge with calculating these curves is that the exact position where the ratio goes negative is quite sensitive to the energy level structure of the ions. We use experimental energy level data to benchmark a point in the curve and shift the calculated curve accordingly. The ratio of $n - 1$ over $n_{\text{free}} - 1$, plotted in Figure 3, gives an estimate of how far the calculated index of refraction is from the free electron approximation. When the ratio is larger than one, the free and bound electrons are contributing with the same sign to the index of refraction. The calculated ratio is larger than one for singly ionized carbon, but this will not show in the soft X-ray laser interferograms since the absorption edge is at 24.4 eV. The region close to the target probably has a large population of neutral and singly ionized carbon atoms. For doubly ionized carbon, there is a significant amount of structure with the ratio going negative for several energy intervals, in particular, close to the 26.5 eV region. For triply and four times ionized, we see the ratio is approaching one with a strong structure near 30 eV for triply ionized, but without a major contribution from the bound electrons at 26.5 eV. All this suggests that in these experiments, the observed anomalous fringe shifts are due to the dominant contribution of bound electrons from doubly ionized carbon ions to the index of refraction. This underscores the importance of having spectroscopic experimental data together with calculations that predict the index of refraction when performing interferometric diagnostics of plasmas.

4. CONCLUSIONS

For decades the analysis of the plasma diagnostics, such as interferometry, have relied on the approximation that the index of refraction in plasmas is due solely to free-electrons. Analysis of recent soft X-ray interferometric data at both 14.7 nm and 46.9 nm probe wavelengths demonstrated that bound electrons can contribute significantly to the index of refraction of multiply ionized plasmas in the vicinity of absorption lines and edges. The new results of the present study extend the results to Carbon plasmas. The Carbon plasma interferograms clearly show anomalous fringe shifts, both early and late in the plasma evolution. Calculations of the index of refraction suggest that bound electrons in doubly ionized Carbon atoms are the main contributor to the observed anomalous index of refraction. It is important to note that the significance of the bound electron contribution is not limited to plasmas with a low mean ion charge. Nevertheless, most hot plasmas that are many times ionized can be confidently probed using soft X-ray laser interferometry, and it is possible to select the probe wavelengths to avoid the contribution from bound electrons to the index in the particular plasma of interest.

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REFERENCES

Albert, F., Joyeux, D., Jaegle, P., Carillon, A., Chauvineau, J.P., Jamelot, G., Klisnick, A., Lagron, J.C., Phalippou, D., Ros, D., Sebban, S. & Zeitoun, P. (1997). Interferograms obtained with an X-ray laser by means of a wave front division interferometer. *Opt. Commun.* **142**, 184–188.

Attwood, D.T., Sweeney, D.T., Auerbach, J.M. & Lee, P.H.Y. (1978). Interferometric confirmation of radiation-pressure effects in laser-plasma interactions. *Phys. Rev. Lett.* **40**, 184.

Benware, B.R., Macchietto, C.D., Moreno, C.H. & Rocca, J.J. (1998). Demonstration of a high average power tabletop soft x-ray laser. *Phys. Rev. Lett.* **81**, 5804–5807.

Da Silva, L.B., Barbee Jr., T.W., Cauble, R., Celliers, P., Ciarlo, D., Libby, S., London, R.A., Descamps, D., Lyngå, C., Norin, J., L'Hullier, A., Wahlström, C.-G., Hergott, J.-F., Merdji, H., Salières, P., Bellini, M. & Hänsch, T.W. (1995). Fringe formation and coherence of a soft-X-ray laser beam illuminating a Mach–Zehnder interferometer. *Opt. Lett.* **25**, 135.

Descamps, D., Lyngå, C., Norin, J., L'Huillier, A., Wahlström, C.-G., Hergott, J.-F., Merdji, H., Salières, P., Bellini, Hänsch, T.W. (2000). *Opt. Lett.* **25**, 135–137.

Dunn, J., Filevich, J., Smith, R.F., Moon, S.J., Rocca, J.J., Keenan, R., Nilsen, J., Shlyaptsev, V.N., Hunter, J.R., Ng, A. & Marconi, M.C. (2005). Picosecond 14.7 nm interferometry of high intensity laser-produced plasmas. *Laser Part. Beams* **23**, 9–13.

Fiedorowicz, H. (2005). Generation of soft X-rays and extreme ultraviolet (EUV) using a laser-irradiated gas puff target. *Laser Part. Beams* **23**, 365–373.

Filevich, J., Kanizay, K., Marconi, M.C., Chilla, J.L.A. & Rocca, J.J. (2000). Dense plasma diagnostics with an amplitude-division soft-X-ray laser interferometer based on diffraction gratings. *Opt. Lett.* **25**, 356.

Filevich, J., Rocca, J.J., Marconi, M.C., Moon, S.J., Nilsen, J., Scofield, J.H., Dunn, J., Smith, R.F., Keenan, R., Hunter, J.R. & Shlyaptsev, V.N. (2005). Observation of a multiply ionized plasma with index of refraction greater than one. *Phys. Rev. Lett.* **94**, 035005.

Filevich, J., Rocca, J.J., Marconi, M.C., Smith, R.F., Dunn, J., Keenan, R., Hunter, J.R., Moon, S.J., Nilsen, J., Ng, A. & Shlyaptsev, V.N. (2004). Picosecond-resolution soft-X-ray laser plasma interferometry. *Appl. Opt.* **43**, 3938–3946.

Greenwood, D.A. (1958). The Boltzmann equation in the theory of electrical conduction in metals. *Proc. Phys. Soc. London* **715**, 585.

Griem, H.R. (1997). *Principles of Plasma Spectroscopy*. Cambridge, UK: Cambridge University Press.

Johnson, W.R., Guet, C. & Bertsch, G.F. (2006). Optical properties of plasmas based on an average-atom model. *J. Quant. Spectrosc. Radiat. Trans.* **99**, 327–340.

Kubo, R. (1957). Statistical-mechanical theory of irreversible processes. i. general theory and simple applications to magnetic and conduction problems. *J. Phys. Soc. Japan* **12**, 570–586.

Kuroda, H., Suzuki, M., Ganeev, R., Zhang, J., Baba, M., Ozaki, T., Wei, Z.Y. & Zhang, H. (2005). Advanced 20 TW Ti:S laser system for X-ray laser and coherent XUV generation irradiated by ultra-high intensities. *Laser Part. Beams* **23**, 183–186.

Liberman, D.A. (1982). INFERNO: A better model of atoms in dense plasmas. *JQSRT* **27**, 335.

Liu, Y., Seminario, M., Tomasel, F.G., Chang, C., Rocca, J.J. & Attwood, D.T. (2001). Achievement of essentially full spatial coherence in a high-average-power soft-X-ray laser. *Phys. Rev. A* **63**, 033802.

Mocek, T., Sebban, S., Bettaibi, I., Zeitoun, P., Faivre, G., Cros, B., Maynard, G., Butler, A., McKenna, C.M., Spence, D.J., Gonsavles, A., Hooker, S.M., Vorontsov, V., Hallou, S., Fajardo, M., Kazamias, S., Le Pape, S., Mercere, P., Morlens, A.S., Valentin, C. & Balcou, P. (2005). Progress in optic-field-ionization soft X-ray lasers at LOA. *Laser Part. Beams* **23**, 351–356.

Neumayer, P., Bock, R., Borneis, S., Brambrink, E., Brand, H., Caird, J., Campbell, E.M., Gaul, E., Goette, S., Haefner, C., Hahn, T., Heuck, H.M., Hoffmann, D.H.H., Javorkova, D., Kluge, H.J., Kuehl, T., Kunzer, S., Merz, T., Onkels, E., Perry, M.D., Reemts, D., Roth, M., Samek, S., Schaumann, G., Schrader, F., Seelig, W., Tauschwitz, A., Thiel, R., Ursescu, D., Wiewior, P., Wittrock, U. & Zielbauer, B. (2005). Status of PHELIX laser and first experiments. *Laser Part. Beams* **23**, 385–389.

Nilsen, J. & Scofield, J.H. (2004). Plasmas with an index of refraction greater than 1. *Opt. Lett.* **29**, 2677.

Rocca, J.J., Moreno, C.H., Marconi, M.C. & Kanizay, K. (1999). Soft-X-ray laser interferometry of plasma with a tabletop laser and a Lloyd's mirror. *Opt. Lett.* **24**, 420.

Smith, R.F., Dunn, J., Nilsen, J., Shlyaptsev, V.N., Moon, S., Filevich, J., Rocca, J.J., Marconi, M.C., Hunter, J.R. & Barbee, T.W. (2002). Picosecond x-ray laser interferometry of dense plasmas. *Phys. Rev. Lett.* **89**, 065004.

Tallents, G.J. (1984). Interferometry and refraction measurements in plasmas of elliptical cross-section. *J. Phys. D* **17**, 721.

Tang, H., Guilbaud, O., Jamelot, G., Ros, D., Klisnick, A., Joyeux, D., Phalippou, D., Kado, M., Nishikino, M., Kishimoto, M., Sukegawa, K., Ishino, M., Nagashima, K. & Daido, H. (2004). Diagnostics of laser-induced plasma with soft X-ray (13.9 nm) bi-mirror interference microscopy. *Appl. Phys. B* **78**, 975.

Uspenskii, Y.A., Levashov, V.E., Vinogradov, A.V., Fedorenko, A.I., Kondratenko, V.V., Pershin, Y.P., Zubarev, E.N. & Fedotov, V.Y. (1998). High-reflectivity multilayer mirrors for a vacuum-ultraviolet interval of 3550 nm. *Opt. Lett.* **23**, 771–773.

Wagner, T., Eberl, E., Frank, K., Hartmann, W., Hoffmann, D.H.H. & Tkotz, R. (1996). XUV amplification in recombining z-pinch plasma. *Phys. Rev. Lett.* **76**, 3124–3127.