EPJ Web of Conferences **167**, 04001 (2018) https://doi.org/10.1051/epjconf/201816704001
PPLA 2017

Plasma Mirrors for Cleaning Laser Pulses from the Infrared to the Ultraviolet

István B Földes[1,*], *Barnabás* Gilicze[2], *Zsolt* Kovács[1] and *Sándor* Szatmári[2]

[1]Plasma Physics Department, Wigner Research Centre for Physics of the Hungarian Academy of Sciences, H-1525 Budapest P.O.B. 49, Hungary
[2]Department of Experimental Physics, University of Szeged, Dóm tér 9, H-6720 Szeged, Hungary

Abstract. Ultrashort laser pulses are generally preceded by prepulses which – in case of high main pulse intensities - may generate preplasmas on solid surfaces, thus making the initial conditions for the interactions ambiguous. Infrared laser systems applied successfully, with high efficiency self-induced plasma mirrors for improving the contrast of the beam. Short wavelength laser beams however have a larger critical density in the plasma, and due to their deeper penetration the absorption is higher, the reflectivity, and the corresponding plasma mirror efficiency is lower. We show herewith that with carefully planned boundary conditions plasma mirrors can reach up to 70% efficiency even for KrF laser radiation. Our observations can be qualitatively explained by the classical Drude model. The high reflectivity allows the use of plasma mirrors even after the final amplification or before the last amplifier. Different arrangement proposals for its integration to our high power KrF laser system are given as well.

1 Introduction

One can speak about ultrashort laser pulses when the duration of the pulse is less than a picosecond (10^{-12}s). Laser pulses with durations as short as 4-5 fs (1 fs=10^{-15} s), i.e. less than 2 cycles of the electromagnetic pulse can be generated. They are however generally not purely Gaussian pulses, as the short pulse generally follows an amplified spontaneous emission (ASE) pulse with nanosecond duration from the amplifier, and/or a shoulder of a few picosecond due to the incomplete compression after the chirped pulse amplification (CPA) [1]. CPA systems avoid the damage and nonlinearities of the laser amplifier, as the amplification occurs after spectrally selective stretching of the laser pulse. The amplified pulse is then recompressed after the amplifier stage. Most of the solid state laser systems operate with CPA amplification, providing short pulses in the infrared. Excimer lasers however allow ultrashort pulse generation as well, and - as they are pumped by gas discharges - the active gas material allows direct amplification as well, i.e. they are non-CPA system [2].

In case of high-power laser-matter interactions ultrashort pulses allow the generation of intensities exceeding the relativistic limit of >10^{18} W/cm^2 at which the free electrons oscillate with velocities almost the speed of light. Clearly, even a low intensity pedestal of the pulse is sufficient to generate preplasma on the surface of solid targets, thus preventing one to obtain clean boundary conditions. Preplasmas can be detrimental for phenomena as for example high order harmonic generation from the surfaces of solid targets [3].

Plasma mirror or self-induced plasma shuttering [4] is one of the most efficient methods to remove prepulses. In this case the laser beam is focused onto an antireflection-coated transparent glass or quartz target inside a vacuum chamber. The low intensity part of the beam arriving before the main pulse is transmitted through the target. Once the ionization threshold is reached, a high density overdense plasma is generated which reflects the main pulse of the beam with an improved temporal contrast. Clearly more than 2 orders of magnitude improvement can be obtained in a single stage. This has been well applied for the infrared laser systems, in which case reflectivity was obtained up to 80% [5]. Plasma mirror is probably the ultimate method to remove prepulses. It is generally applied after the last amplifier, in the already compressed beam, additionally to the cross polarized wave generation (XPW) method [6], which is - in general - applied between two CPA amplifier stages.

Due to the availability of high reflectivity in the infrared, double plasma mirrors were introduced [7] with 50% efficiency, resulting in 4 orders of magnitude contrast improvement. It must be noted that in case of the CPA systems not only the ASE pedestals are eliminated but the shoulders of a few picosecond duration (caused by the imperfect compression and uncompensated high-order dispersion) are suppressed as well.

Different is the case for the UV lasers. Although KrF systems operate with direct amplification

* Corresponding author: foldes.istvan@wigner.mta.hu

and prepulses from incomplete compression do not appear, the high photon energy is able to modify laser-matter interactions even at lower intensities. The 5 eV photons of the ASE of the KrF amplifiers may generate ions due to photoablation and photoionization of the solid matter even if the prepulse of nanosecond duration has intensity as low as 10^7 W/cm^2 [8]. It means that altogether more than 10-13 orders of magnitude intensity contrast is needed for high intensity interactions, i.e. for 10^{17}-10^{20} W/cm^2. On the other hand, until recently no plasma mirrors were used for short-pulse UV lasers. The reason of it is that the UV radiation of the KrF laser penetrates deeply into the plasma generated on solid surfaces, namely to a critical density which is 16 times higher than for an infrared laser beam with wavelength of 1 μm. If the penetration depth is larger, then the reflectivity is lower because of the collisional absorption in the underdense plasma. A high reflectivity, i.e. 70% efficiency was just recently obtained for laser pulses of 248 nm wavelength [9] which opens the possibility to its application in UV laser systems.

In the case of IR lasers detailed studies of Ziener et al [5] show that plasma mirrors can work well up to quite high laser intensity on the plasma mirror target until nonlinear interactions set in. The maximum applicable intensity depends on the main pulse duration [10], generally it is less than 10^{17} W/cm^2. Therefore the laser beam cannot be tightly focused onto the target, which is positioned in most cases behind the focal plane [11]. Rödel et al [11] obtained three orders of magnitude contrast improvement until 3 ps before the main pulse with a single stage plasma mirror by precise choice of the target coating and the experimental geometry. As the high beam quality on the plasma mirror target is a critical issue, long focal length focusing is often used in order to keep the intensity low. In the case of the double plasma mirror [7] the two plasma mirror targets are inside an f=10 m telescope.

For UV lasers however the ASE prepulse can be relatively strong even if it is minimized by off-axis amplification [2]. Note that for the 248 nm wavelength of the KrF systems the possibilities of filtering the prepulses by nonlinear crystals (Pockels cell or XPW) is strongly limited. It was already shown by the early works of Fedosejevs et al [12] that even the ASE prepulse must be kept low in order to obtain high reflectivity. In the following the methods of optimizing the plasma mirror for the short pulse KrF laser is described in detail which resulted in nearly 70% reflectivity either for 600 fs or for 200 fs ultrashort pulse duration [9].

2 Experimental arrangement

A twin-tube excimer laser system was the basis of the experiments [2]. The first laser tube is an ordinary XeCl laser. It serves as a pumping of the dye-laser chain, which finally gives an ultrashort laser pulse of 500 fs duration on the 496 nm wavelength. After frequency conversion in a BBO crystal it serves as a seed pulse for the second, KrF amplifier. It was used either in a 2-times or in a 3-times off-axis pass configuration providing 10 and 15 mJ pulses of ~500 fs duration, respectively. A further power amplifier has been recently added, in which after 2-passes off-axis amplification up to 80 mJ energy could be obtained (Fig. 1).

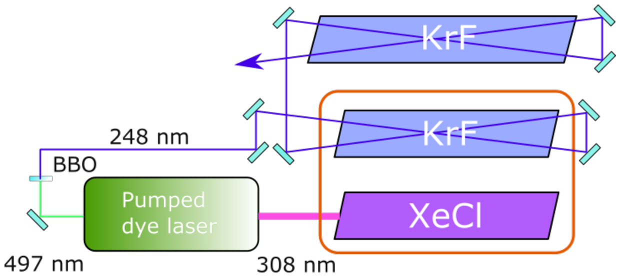

Fig. 1. The scheme of the KrF laser system.

As mentioned above, the only source of prepulses is the ASE of the amplifiers. Off-axis amplification is advantageous in this respect because most of the ASE propagates along the axis of the amplifier [13]. A part of the ASE can be excluded as well, if the pulse to be amplified is coming in the first half of the (discharge) pumping and the corresponding ASE duration. Therefore the first amplifier was used only in a 2-pass configuration, when the energy contrast was better than 100. Taking into account the duration time of the main pulse and that of the ASE this energy contrast corresponds to more than 6 orders of magnitude in the power contrast and up to 10 orders of magnitude intensity contrast in case of tight focusing because the focusability of ASE is much worse than that of the main pulse.

In the case of using the final amplifier a spatial filter is situated between the two amplifier stages. The result is that the energy contrast remains very high (better than 100) but the ASE transmitted by the spatial filter will be the focusable part, therefore the intensity contrast will be not better than 8-9 orders of magnitude, corresponding to $\sim 10^8$–10^9 W/cm^2 ASE intensity in the focal plane resulting in a preplasma caused by photoablation and photoionization. Plasma mirror experiments were carried out after the first amplifier, in which case a modest f/25 or similar focusing was sufficient to investigate and optimize the plasma mirror, and in which case the ASE intensity was less than 10^6 W/cm^2, thus below the threshold of photoablation [8,14].

Part of the experiments were carried out after compressing the laser pulse to 220 fs pulse duration, in which case the laser energy was reduced to 0.5-1 mJ.

The target was an antireflection-coated quartz plate in a vacuum chamber. It was antireflection-coated which was moved by stepper motors to provide fresh surface area for each laser shot. The laser beam was of 15 mm diameter in front of the chamber and it was focused by a 40 cm lens onto the target surface by 12° angle of incidence. At this nearly perpendicular angle of incidence the reflectivity does not depend strongly on the polarization of the laser beam, nevertheless the experiments were carried out both by p- and s-polarized incident radiation.

Care was taken to avoid any beam inhomogeneities on the target, especially that of astigmatism. Therefore the beam profile was monitored by imaging the focal spot to a UV-sensitive Hamamatsu T7040 CCD detector with a 12 times magnification. This imaging was carried out after refocusing the beam reflected by the plasma mirror, too, in order to confirm the applicability of the used configuration.

The incoming and the reflected energy was monitored by fiber coupled energy-meters using peak-hold detection [15]. Note that in the experiments using discharge-pumped lasers fiber-coupling is essential to reduce electrical noises. Energy monitoring was carried out by each shot by splitting out a small part of the beam using a quartz plate. The energy monitor was then calibrated using a Gentec Joulemeter QE50LP. The energy monitors were equipped with a microprocessor, thus the ratio of the incoming and that of the reflected energy could be immediately obtained by the computer.

3 Experimental results

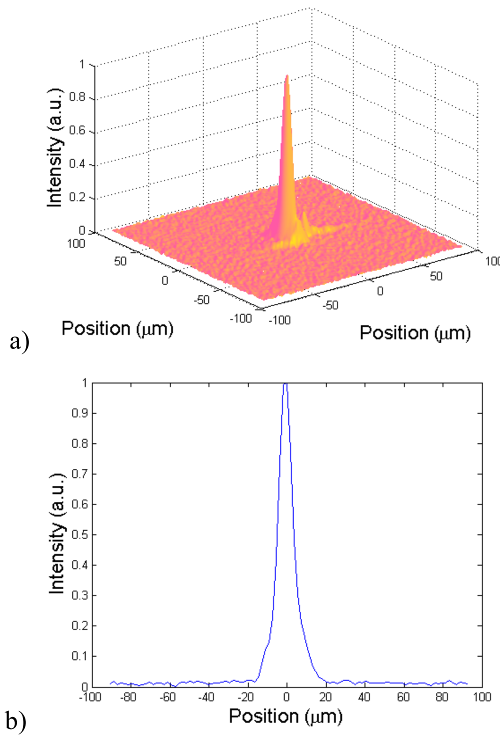

Fig. 2. Image of the 12 times magnified focal spot of the laser (a) and a cross-sectional view (b).

Fig. 2 shows the 2-dimensional focal distribution of the incoming beam and a typical cross-section. The beam is nearly Gaussian, and it corresponds to the focal spot of a 1.5 times diffraction limited rectangular beam. In this case approximately 72% of the laser energy is in the central spot of the Airy-pattern.

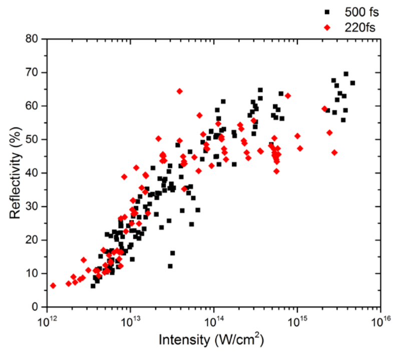

Fig. 3. Intensity dependence of plasma mirror reflectivity. The reflectivity for the s-polarized KrF laser beam is given for 500 fs (black squares) and for 220 fs (red diamonds) pulse duration.

High reflectivity, i.e. more than 60%, in some shots up to 70% was measured both with 500 fs and with 220 fs pulse duration. In the results shown in Fig. 3. s-polarized incoming radiation was used. It must be mentioned that changing the direction of the linear polarization to p-polarization did not affect the results strongly due to the nearly perpendicular angle of incidence. The only difference is that at higher intensities the scatter of data is larger due to the increasing importance of nonlinearities for p-polarized laser beam. It can be seen that below 10^{12} W/cm^2 short-pulse intensity the target remains practically transparent, the reflectivity remains in the noise level. It starts to increase if the intensity reaches 10^{13} W/cm^2 (i.e. above the plasma threshold) and saturates above 10^{14} W/cm^2 remaining approximately constant up 10^{16} W/cm^2. The observed high reflectivity [9] which allows the direct applicability of the plasma mirror for KrF systems can be attributed mainly to the already good initial contrast. In the present case the intensity of the ASE pedestal of 15 ns duration on the target was less than 10^5 W/cm^2. If the prepulse level is higher (e.g. 10^8 W/cm^2 after 3 passes in the amplifier) the reflectivity drops below 50% [16] in agreement with the early observations of Fedosejevs et al. [14]. The nearly perpendicular angle of incidence is needed as well, because in the first plasma-mirror experiments with the KrF laser [17] the angle of incidence was 45°and the reflectivity was as low as 30%. The shot-to-shot variation of reflectivity is attributed to the beam quality nonuniformities due to the shot-to-shot variation of the discharge-pump of the laser. In case the beam quality was lower than illustrated in Fig. 2, then the observable reflectivity could be significantly lower, too. It must be mentioned that the maximum reflectivity is similar in the cases of the two applied pulse durations, especially when taking into account that in the case of the shorter pulse the maximum incident intensity was limited by the size of the compressor optics, therefore the full saturation was not reached.

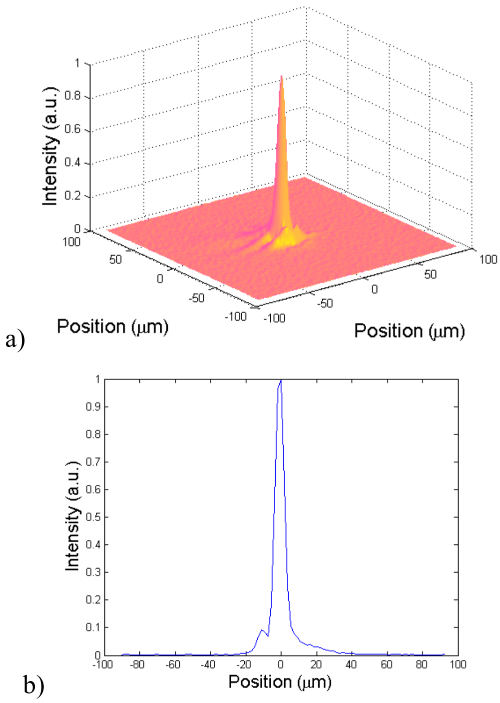

Fig. 4. Image of the 12 times magnified focal spot of the beam after the plasma mirror (a) and a cross-sectional view (b).

The beam quality of the reflected beam is illustrated in Fig. 4. It is similar to that of the incoming beam, as it is well focusable to a 1.75 times Gaussian spot. The energy contained in the central spot is 70%.

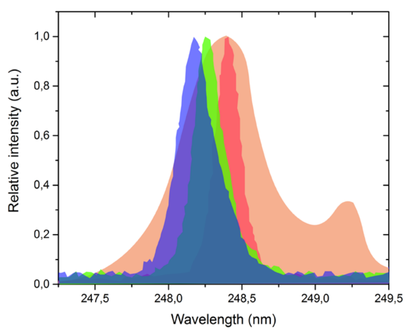

Fig. 5. Laser spectra. The incoming laser beam (red) as compared with the approximate gain bandwidth (salmon) and the reflected pulses at intensities of 2×10^{14} W/cm^2 (green) and 8×10^{15} W/cm^2 (blue).

In case one wants to further amplify the laser beam cleaned by a plasma mirror, it is essential that the reflected and therefore Doppler-shifted beam should be within the gain bandwidth of the amplifier. It is more critical in our case of KrF amplifiers than for the Ti:sapphire laser systems which have much broader bandwidth. Figure 5 shows the laser spectra as compared with the gain spectrum [18]. Clearly the Doppler-shift is increasing with increasing laser intensity [19] but it remains within the spectral range of the gain bandwidth up to the applied maximum intensity of ~10^{16} W/cm^2.

4 Discussion

Plasma reflectivity is generally calculated by applying the Drude model [14] and it gives a rough agreement with our results in case of assuming an average plasma scalelength of L/λ~0.1 during the interactions which gives a reflectivity of 70% corresponding to our experimental observations. Even recent plasma mirror experiments [10] used similarly the Drude model to explain their findings. It must be mentioned however that this model cannot describe the phenomena appropriately.

In the Drude model Fresnel reflection is calculated on the border of the plasma medium which has an index of refraction

$$n_r^2 = 1 - \frac{n_e / n_c}{1 + i\nu_0 / \omega}. \tag{1}$$

Here n_e is the electron density in the high density (overdense) region, $n_c = m_e\omega^2/(4\pi e^2)$ is the critical density, ω is the laser frequency, with m_e electron mass and e charge. The electron-ion collision frequency in the dense region is assigned by υ_0. The Fresnel reflectivity for an angle of incidence of θ_{inc} in the case of s-polarized radiation is

$$R_s = \left| \frac{\sin(\theta_{inc} - \theta_{\text{int}})}{\sin(\theta_{inc} + \theta_{\text{int}})} \right|^2. \tag{2}$$

Here θ_{int} is the complex angle of propagation inside the dense plasma [14] which can be obtained from the generalized Snell law as $\theta_{int} = arcsin[\theta_{inc}/n_r]$. The imaginary part of the index of refraction, and thus the absorption strongly depends on the electron-ion collision frequency. Fedosejevs et al [14] made calculations for different υ_0 values in case of step function vacuum-plasma interface and they obtained high reflectivity when υ_0 was significantly less, typically some tenth of the laser frequency. The electron-ion collision frequency in a plasma is given [20] by

$$\nu_0 = \frac{4(2\pi)^{1/2}}{3} \frac{Z n_e e^4 \ln\Lambda}{m_e^{1/2}(k_B T)^{3/2}} \tag{3}$$

with Z ion charge, T temperature and k_B Boltzmann-constant. It also depends on the $ln\Lambda$ Coulomb logarithm. Clearly, the collisional frequency is strongly dependent on the temperature and the density of the plasma. A special problem arises with the Coulomb logarithm which is derived from the integration limits of the Rutherford scattering of an electron on ions. In the case of very high densities and low temperatures corresponding to the state of the solid matter the classical Coulomb logarithm becomes less than or equal to zero, and then the classical [20] (Spitzer) expression of the Coulomb logarithm cannot be applied any more. In that case namely the plasma is strongly coupled, the potential energy in the electric field is comparable with the kinetic energy of the particles. For this case more accurate quantum mechanical calculations [21] avoid this divergence but further increasing the density the plasma is becoming Fermi degenerate. Due to these problems most textbooks [22, 23] suggest to take the Coulomb logarithm simply equal to 2, a rough estimation. Fedosejevs et al [14] avoided this problem by choosing only some finite discrete values of the electron-ion collision frequency by taking e.g. a typical $\upsilon_0/\omega\sim 0.1$. We also carried out some calculations for steep plasma gradients of our case of 12°angle of incidence. Clearly more than 60% reflectivity was obtained (depending on the temperature) in case of using $\ln\Lambda=2$ and it can be even higher when using the Coulomb logarithm of Mulser et al [21]. When using

$$\ln\Lambda_0 = \ln\frac{\lambda_D}{\lambda_B} + 0.2 + \frac{\lambda_B^2}{4\lambda_D^2} \tag{4}$$

with λ_D Debye length and λ_B de Broglie length, nearly 80% reflectivity was obtained in case of 4eV plasma temperature of the plasma step having a 10 times critical density. Thus our experimental finding can roughly be explained by the theory but in order to a full understanding of the phenomena the exact equation of state and the plasma density and temperature profile is needed at the arrival and during the interaction with the main ultrashort laser pulse.

5 Conclusion

We could demonstrate the high, ~70% efficiency even for the KrF laser system, although its 248 nm wavelength in the ultraviolet causes a deep penetration to the plasma, and therefore generally lower absorption than in the infrared. The method for obtaining it is that we could shorten the interaction length by choosing appropriate initial conditions with shorter scalelength plasmas. Basically there are 2 possible method to integrate it into the laser system In our previous paper [9] we suggested an arrangement in which the plasma mirror is applied in front of the final amplifier. It became possible because although the reflected beam suffers a spectral Doppler shift, the spectrum remains within the gain bandwidth of the final amplifier. In this case the final energy of the laser system will not decrease significantly because the amplifier operates in a saturation regime.

The observed high efficiency of the plasma mirror allows its direct use after the first amplifier, as even in this case more than 60% of the energy can be coupled to the target. We mentioned however that the high reflectivity requires high quality beam, i.e. best is to put the plasma mirror target in focus. In order to avoid nonlinearities the intensity must be limited to the values given above. Therefore in our case of 80 mJ /600 fs laser beam a loose focusing with at least f=5m focusing lens seems to be necessary. Its problem lies in the final size of the laboratory and the long vacuum beamlines. There is however another possibility. As the saturation intensity of the KrF laser is significantly lower than the damage threshold of the optics, it is possible first to reduce the diameter of the beam a telescope, and then to focus it with a shorter focal length focusing optics. Alternatively a galilei telescope may be directly applied to provide a larger focal spot in shorter distance and the near optimal 10^{15} W/cm^2 intensity on the plasma mirror.

We can thus summarize that the observed high reflectivity of plasma mirrors opens the possibility of their application for improving the contrast of lasers even in the ultraviolet.

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 under grant agreement 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission. The authors also acknowledge the support of the Hungarian Scientific Research Fund – OTKA 113222 and the COST MP1208 activity. The technical assistance of Rita Dajka is acknowledged this way, too.

References

1. D. Strickland and G. Mourou, Opt. Commun. **56**, 219 (1985)
2. S. Szatmári, Appl. Phys. B **58**, 211 (1994)
3. C. Thaury and F. Quéré, J. Phys. B **43**, 213001 (2019)
4. H. Kapteyn, M. Murnane, A. Szoke, R. Falcone, Opt. Lett. **16**, 491 (1991)
5. CH. Ziener et al., J. Appl. Phys. **93**, 768 (2003)
6. A. Jullien et al., Appl. Phys. B **84**, 409 (2006)
7. T. Wittmann et al., Rev. Sci. Instrum. **77**, 083109 (2006)
8. I.B. Földes et al., Laser Physics **10**, 264 (2000)
9. B. Gilicze, A. Barna, Zs. Kovács, S. Szatmári, I.B. Földes, Rev. Sci. Instrum. **87**, 083101 (2016)
10. G. Doumy et al., Phys. Rev. E **69**, 026402 (2004)
11. C. Rödel et al., Appl. Phys. B **103**, 295 (2011)
12. R. Fedosejevs, R. Ottmann, R. Sigel, G. Kühnle, S. Szatmári, F.P. Schäfer, Phys. Rev. Lett. **64**, 1250 (1990)
13. G. Almási, S. Szatmári, P. Simon, Opt. Commun. **88**, 231 (1992)
14. R. Fedosejevs, R. Ottmann, R. Sigel, G. Kühnle, S. Szatmári, F.P. Schäfer, Appl. Phys. B **50**, 79 (1990)
15. A. Barna, I.B. Földes, Z. Gingl, R. Mingesz, Metrol. Meas. Syst. **20**, 183 (2013)
16. I.B. Földes, A. Barna, D. Csáti, F.L. Szűcs, S. Szatmári, J. Phys. Conf. Ser. **244**, 032004 (2010)
17. I.B. Földes, D. Csáti, F.L. Szűcs, S. Szatmári, Rad. Effects & Defects in Solids, **165**, 429 (2010)
18. W.T. Silfvast, *Laser Fundamentals* (Cambridge University Press, 2004) ISBN 0-521-83345-0
19. R. Sauerbrey, Phys. Plasmas, **3**, 4712 (1996)
20. S. Atzeni, J. Meyer-ter-Vehn, *The Physics of Inertial Fusion* (Oxford Science Publications, Clarendon Press-Oxford 2004) ISBN 0-198562640
21. P. Mulser, G. Alber, M. Murakami, Phys. Plasmas **21**, 042103 (2014)

22. S. Eliezer, *The interaction of high-power lasers with plasmas* (IOP Bristol and Philadelphia, 2002) ISBN 0 7503 0747 1

23. S. Pfalzner, *An introduction to inertial confinement fusion* (CRC Press Tayleo & Francis New York, London 2006) ISBN 0-7503-0701-3