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**INVESTIGATION OF NONLINEAR PHENOMENA IN LASER  
PLASMAS AT HIGH LASER INTENSITY**

Ph.D. theses

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# 1. Introduction

'Plasma is the 4th phase of matter' this can be heard more and more frequently. Plasma experiments started with investigations of low-pressure-ionised gases, than discharges moved into the focus of more and more studies. Langmuir was the first, who gave a definition of plasma in 1923. He defined the plasma as ionized gas. Nowadays the definition of plasma is the following: Plasma is a quasineutral gas, consisting of charged particles (electrons, ions), moreover the behaviour of this gas is characteristically uniform from the outside. As early as the 1960's, almost together with the invention of lasers, the idea of plasma-generating with high intensity laser radiation appeared. This prediction was soon realized. Laser-plasma physics, the new branch of plasma physics has been developed. Nowadays it is clear that plasma physics and maybe laser-plasma physics will help in the realization of the fusion power plant. The huge source of energy may be realized within a few decades.

At present incredible high intensities can already be reached by focusing of laser radiation. The highest achieved intensity is as high as  $10^{21} \frac{W}{cm^2}$ . If the high-intensity laser beam interacts with the solid state surface, plasma, laser-plasma will appear on the surface. Laser-plasma physics is a field consisting of mainly non-linear processes. Several non-linear processes detected in laser-plasmas are present in other scientific areas, too.

*The aim of the present work was the experimental study of non-linear phenomena occurring in laser plasmas on the solid state surface generated by subpicosecond laser pulse at different intensities.*

The experiments were carried out in the High Intensity Laser Laboratory (HILL) which is at the Department of Experimental Physics of the University of Szeged in collaboration with KFKI Research Institute for Particle and Nuclear Physics Department of Plasma Physics. A part of my work was done in the Max-Planck Institut für Quantenoptik, Garching.

## 2. Antecedents and Objectives

1. Generation of the high order harmonics is a manifestation of the nonlinear behaviour of matter during laser light-matter interaction. The plasma, generated by a high-intensity laser has non-linear optical properties and in this medium the oscillating electron motion is non-linear, too. Certain plasma electrons oscillate near the steep vacuum-plasma boundary. This electron oscillation is anharmonic. High harmonics of the input laser beam is generated by the anharmonically oscillating electrons. Because radiation emitted by the electrons contain electromagnetic wave-components with multiples of the input laser frequency.

At the beginning of the 1970's 2nd harmonics of the input laser pulse in solid state laser plasma has been successfully detected, and then more and more studies aimed at higher harmonics generation and their properties (e.g. propagation, polarization). In 1981 the 20th harmonics of the incoming  $CO_2$  laser pulse was detected by Carman et al. after steepening of the laser-plasma density profile. Recently, by using short laser pulses the scalelength of the solid state plasma decreases, too. The vacuum-plasma boundary profile becomes steep, the plasma gradient will be shorter than the wavelength. In the 1990's subpicosecond lasers

realized short scalelength laser plasmas.

The results of harmonics experiments were different as a function of the pulse-length and intensity. If the incoming laser pulse is as short as  $100fs$  or shorter than this value harmonics were generated only by p-polarized radiation and they propagated into the specular direction. It was a main result of Kohlweyer et al.: the shorter laser wavelength and the better laser intensity contrast results in higher harmonics generation efficiency. Experiments using lasers of several  $100fs$  pulse duration gave different results. Some experiments at intensity value of  $5 \cdot 10^{15} \frac{W}{cm^2}$  were made by the KrF laser of the HILL laboratory. According to their experimental results second and third harmonics of the  $248nm$  incoming p- and s-polarized laser pulse was observed. The harmonics propagated into specular direction and kept the polarization of the incoming laser beam. Norreys and the Rutherford-group generated high harmonics by the high intensity laser VULCAN ( $2.5ps$  pulse duration,  $1053nm$  wavelength). In case of high harmonics generated by intensities of  $10^{19} \frac{W}{cm^2}$  the harmonics were scattered into  $2\pi$  solid angle. These experimental observations were again similar for p- and s-polarized laser beam. The explanation of the scattering of harmonics is probably the rippling of the critical surface of the plasma. Chambers and co-workers also carried out experiments by using a  $1ps$ ,  $248nm$  laser beam of  $10^{16} - 10^{19} \frac{W}{cm^2}$  having a strong prepulse. They observed the 2nd, 3rd and 4th harmonics of the input pulse with the harmonics becoming diffuse above  $10^{16} \frac{W}{cm^2}$  intensity. The cause of this phenomenon was given as the rippling of the critical surface of the plasma.

*The aim of the present work: High harmonics generation on solid state surface by a clean, pre-pulse free focused laser beam with high intensity contrast. Investigation of the propagation and polarization properties of harmonics as a function of the intensity and polarization of the incoming laser beam.*

2. Another important process of the laser-plasma interactions is the absorption of laser light in the plasma, i.e. the absorption mechanism. Until 1990 only a few experimental groups reported absorption measurements of high intensity subpicosecond laser pulses in plasmas. Reflection of the short pulse laser beam ( $160fs$ ,  $616nm$ ) at normal incidence was measured by Murnane et al. According to their experimental results the reflection was around 75%. In the experiments of Milchberg et al. a p-polarized XeCl pulse ( $400fs$ ,  $308nm$ ) generated plasma on the Al target surface. The angle of incidence of the laser beam was  $45^\circ$ . They measured 35% reflection and their results showed that the absorption increased by increasing the angle of incidence of the laser pulse in agreement with absorption theories. For intensities lower than  $10^{15} \frac{W}{cm^2}$  the absorption is mainly collisional. Absorption of the  $250fs$  KrF laser pulse in dense plasma at intensity range of  $10^{12} - 10^{14} \frac{W}{cm^2}$  at normal laser incidence was investigated by Fedosejevs et al. using Al, Au and Cu targets. In their following experiments in the intensity range of  $10^{14} - 2.5 \cdot 10^{15} \frac{W}{cm^2}$  it was found that reflection from the plasma increased with increasing laser intensity. On the other hand plasma reflection decreased by increasing the atomic number of the target materials. At high intensity resonance absorption becomes dominant. Preliminary experiments were carried out 6 years ago by the KrF excimer-dye laser system of the HILL laboratory. The laser intensity reached  $5 \cdot 10^{15} \frac{W}{cm^2}$  for  $45^\circ$  angle of incidence.

In an inhomogeneous laser-plasma the light propagates from low to high densities and it is

strongly reflected where the electrons inhibit the further propagation of the electromagnetic radiation. Because of the fast expansion of the plasma this reflection layer moves quickly, hence the frequency of the reflected light is shifted due to the Doppler-effect. To take into account this effect the expansion velocity of the plasma can be derived. The laser intensity and the degree of absorption determine the plasma temperature which affects the plasma expansion velocity. In a diploma work of Zoltán Juhász the expansion velocity and the temperature of the plasma was determined. In the present work more accurate experiments were carried out to study absorption processes in more detail.

*Doppler-shift of the reflected light from the plasma generated on polystyrene, aluminium, and gold target for intensities between  $5 \cdot 10^{14} - 5 \cdot 10^{15} \frac{W}{cm^2}$  was measured. With the help of the experimental results and by a self-designed local thermodynamic equilibrium based model I determined the absorption coefficients as a function of electron temperature for different materials.*

3. At high focused laser intensity the plasma electrons acquire high energy due to the strong oscillation in the laser field. In this case the plasma electrons interact with a very high intensity laser pulse. It is called relativistic intensity for  $\mu = 10^{-9} \lambda [\mu m] \sqrt{I [\frac{W}{cm^2}]} > 1$ , ( $\lambda$  is the laser wavelength) when the oscillating electrons gain relativistic velocity. The mass of these electrons increases relativistically. Therefore the refractive index of the medium increases on the laser beam axis. The medium thus behaves as a convex lens. Consequently the diameter of the laser beam decreases and the intensity of the laser increases more and more. This process is called 'relativistic self-focusing'. If the high intensity laser is focused into a pre-plasma which is generated by a pre-pulse from the solid surface, then plasma channel or plasma filaments can be generated due to the relativistic self-focusing. These channels play an important role in the fast ignition laser fusion. The high laser intensity accelerates the electrons in these channels along the laser axis. Electron beams up to several MeV energies have been generated by several groups. A Particle-In-Cell (PIC) computer code was developed by Pukhov and Meyer-ter-Vehn which was used to simulate plasma channel generation. In a recent work the interaction between the high intensity laser beam ( $1053nm$ ,  $0.5 - 1ps$ ,  $10^{19} \frac{W}{cm^2}$ ) and plasma was investigated by Tanaka et al. In their experiments the laser pulse was focused into the pre-plasma and the fast electron transport was investigated. They detected  $1MeV$  electrons originating from the plasma channel. Their x-ray pinhole camera gave an image of the plasma channel. Similar experiments started in the Max-Planck Institut für Quantenoptik in Germany (Garching) within the frames of the EURATOM mobility program. I could participate in these experiments.

It is also important to investigate the propagation of fast electrons in the solid matter. The understanding of this phenomenon is necessary for the fast ignition laser fusion in which case the fast electrons propagate to the high density core to ignite fusion.

*The purpose of my investigations was the observation and study of the generated plasma channels caused by self-focusing and the investigation of the propagation of the electron beam in the high density solid matter.*

### 3. Methods

1. Plasma was generated by high laser intensity on solid target in order to investigate harmonics from the solid surface. The laser beam was focused by an off-axis parabolic mirror. The harmonics were studied by using a self-designed and self-built Rowland-type vacuum-ultraviolet spectrometer. The plasma was projected with 1 : 1 magnification onto the detector, which was a double micro-channel plate coated by a phosphor layer on its rear side. A holographic, torodial, reflection grating ( $550l/mm$ ) was the dispersive element of the spectrometer. Single-shot spectra from the plasma were recorded by the spectrometer using a CCD and computer after imaging the phosphor screen.
2. The Doppler-shift of the reflected laser light from the laser plasma was measured by using two SPEX-type monochromators. One of the monochromators monitored the spectrum of the input laser shot to shot as a reference. The spectrum of the reflection from the plasma was recorded by another monochromator. In both monochromators the output slit was substituted by a diode array connected with a PC, thus obtaining a spectrum for each shot. A simple model was developed using a collisional absorption and a local thermodynamic equilibrium in plasma. The plasma absorption coefficient was determined from the Doppler-shift of the laser pulse as a function of the plasma temperature.
3. The study of plasma channels generated in the pre-plasma by the self-focusing of the focused relativistic laser pulse on solid foils in front of the sample foils was carried out by an x-ray pinhole camera. The same x-ray pinhole camera was used to investigate the propagation through solid targets of the fast electrons in front of and behind the sample. The entering pinholes of the pinhole camera were adjustable, simply exchangeable. The detector of the pinhole camera was an x-ray sensitive, cooled CCD camera. The thickness of the foils (aluminium, beryllium) used as filters in the pinhole camera were chosen each time in accordance with the experimental parameters. The results of the x-ray pinhole camera measurements were compared with other diagnostics, i.e. with visible light interferometry, light scattering and an electron spectrometer.

### 4. New scientific results

1. *The 2nd, 3rd and the 4th harmonics (with a 62nm wavelength) of the 248nm wavelength input pulse were detected by using pre-pulse-free p- and s-polarized laser beams having  $1.5 \cdot 10^{17} \frac{W}{cm^2}$  maximum intensity for  $45^\circ$  angle of incidence.*

The diffraction-limited pulse of the subpicosecond KrF hybrid excimer-dye laser system was focused onto a spot of  $2.3\mu m \times 2\mu m$  diameter by an F/2 off-axis parabolic mirror. It was shown that  $36 \pm 1\%$  of the total laser energy fell within the full-width-at-half-maximum (FWHM) of the focus with approximately 77% of the total energy inside the

central lobe of the Airy-pattern. This value is near to the theoretically predicted one of 86%. These results confirm that the laser pulse was diffraction-limited. The obtained intensity was  $2.2 \cdot 10^{17} \frac{W}{cm^2}$  in vacuum, and  $1.5 \cdot 10^{17} \frac{W}{cm^2}$  for  $45^\circ$  angle of incidence. It was found that the intensity of the amplified spontaneous emission (ASE) of the KrF amplifier in the focal plane could be kept below  $10^7 \frac{W}{cm^2}$ . The result is an intensity-contrast ratio of  $10^{10}$ , thus the experiments were carried out in a clean, pre-plasma-free environment [P1, P2, P6].

2. *By examining the angular distribution of the 2nd and 3rd harmonics between  $10^{16} - 1.5 \cdot 10^{17} \frac{W}{cm^2}$  intensities it was found that the harmonics generation becomes diffuse above  $10^{16} \frac{W}{cm^2}$ . The experiment demonstrated that  $2\omega$  and  $3\omega$  radiation were generated with nearly equal efficiency by p- and s-polarized laser pulses at maximum laser intensity. The harmonics did not keep the polarization of the input laser light, i.e. the polarizations were mixed.*

Previous investigations in our laboratory using lower intensities showed the generation of the 2nd and 3rd orders of high harmonics p- and s-polarized laser beam at the intensity of  $5 \cdot 10^{15} \frac{W}{cm^2}$ . The generated harmonics propagated in the specular direction and kept the polarization of the incoming laser. My new results show that these properties change significantly above  $10^{16} \frac{W}{cm^2}$  intensity. The harmonics propagate not only into the specular direction but a significant amount of radiation is scattered diffusely. The polarizations of the harmonics became mixed, too. A possible explanation for the above mentioned experimental results is the density modulation at the critical surface of the laser plasma [P6].

3. *The critical surface of the laser plasma on solid targets generated by the pre-pulse-free excimer laser system becomes rippled at the intensities of  $10^{16} - 1.5 \cdot 10^{17} \frac{W}{cm^2}$ .*

Both diffuse propagation of the harmonics and the mixing of polarizations indicate that the critical surface of the laser plasma is not plane any more but it becomes rippled. As the focused laser pulse was practically pre-pulse-free, it is proved that the critical surface rippling of the plasma was not generated in a preplasma but it was the result of an intrinsic effect within the plasma generated by the ultrashort laser pulse. This observation was verified by the one dimensional PIC and hydrocode simulations. The simulations showed that at such intensities the plasma pressure and the light pressure of the laser are in the same order of magnitude near to the critical surface of the plasma. As the result of the unstable equilibrium and the interaction between the light pressure and the pressure of the expanding plasma-front a Rayleigh-Taylor instability develops on or near to the critical surface of the plasma, thus causing the critical surface rippling during the laser pulse [P6].

4. *By measuring the Doppler-shift of the ultraviolet laser pulse from laser plasmas on solid surfaces and by a simple local thermodynamic equilibrium based model it was demonstrated that the plasma absorption coefficient referring to the targets can be determined*

as a function of the electron temperature. By comparing the experimental data with the results calculated by the model it was verified that within this range of intensity the absorption is mainly collisional.

When increasing the intensity of laser pulse the Doppler-shift of the reflected radiation from the plasma increases, too. By comparing the calculated and measured values of the Doppler-shifts it is shown that the measured values were overestimated by the model for polystyrene target and underestimated for gold target. The outcome verified the previous results, that up to the laser intensity of  $10^{15} \frac{W}{cm^2}$  the absorption is mainly collisional [P3].

5. *Using an x-ray pinhole-camera it was found that the relativistic self-focusing of the  $10^{19} \frac{W}{cm^2}$  intensity of the Ti:Sa laser pulse is accompanied by strong x-ray emission from the plasma. More than  $100\mu m$  long plasma filaments attributable to relativistic self-focusing were shown by the interferograms in the visible range. The electrons accelerated to MeV energies in these channels propagated well collimated in the dense target according to my observations.*

Visible interferograms showed plasma-channels and filaments as a result of relativistic self-focusing of the laser beam in the preformed plasma. The x-ray pinhole images do not show self-focused filaments (due to the low optical density) but they show powerful x-ray emission corresponding to the incidence cone of the incoming laser beam. This intense x-ray emission is the result of the heating and ionizing effects of the ultrashort high intensity pulse in the preformed plasma. The x-ray pinhole-camera on the rear side of the target shows that the high-energy electron beam propagating inside the solid target remains well collimated in the dense matter [P4].

## 5. Publications

### Publications

- [P1] I.B. Földes, G. Kocsis, **E. Rácz**, S. Szatmári, G. Veres:  
*Generation of High Harmonics in Laser Plasmas*,  
Laser and Particle Beams, **21**, pp. 517-521, October 2003.
- [P2] I.B. Földes, K. Gál, G. Kocsis, **E. Rácz**, S. Szatmári, G. Veres:  
*High-harmonics from a UV Laser Plasma*,  
Proceedings of SPIE, **5228**, pp. 473-479, December, 2003.  
Editor(s): Oleg N. Krokhin, Sergey Y.Gus'kov, Yury A. Merkul'ev)
- [P3] G. Veres, G. Kocsis, **E. Rácz**, S. Szatmári:  
*Doppler shift of femtosecond laser pulses from solid density plasmas*,  
Applied Physics B, **78**, pp. 635-638, 2004.

- [P4] M. Kaluza, I.B. Földes, **E. Rácz**, M.I.K. Santala, G.D. Tsakiris and K.-J. Witte:  
*Relativistic self-focusing of fs-laser pulses and their heating effect on the preformed plasma*,  
IEEE Transactions on Plasma Science, Vol. **33**, No. **2**, pp. 480-481., April 2005.
- [P5] I.B. Földes, J. Bohus, K. Gál, B. Hopp, G. Kocsis, N.R. Kresz, **E. Rácz**, T. Suta, T. Smausz, S. Szatmári, Zs. Tóth, G. Veres:  
*Fast electrons for the fast ignitor scheme of inertial confinement fusion*,  
IAEA-TECDOC-1460, Elements of Power Plant Design for Inertial Fusion Energy, pp. 97-105, June 2005.
- [P6] **E. Rácz**, I.B. Földes, G. Kocsis, G. Veres, K. Eidmann, and S. Szatmári:  
*On the effect of surface rippling on the generation of harmonics in laser plasmas*,  
Applied Physics B., in press, (manuscript No: 2039 B), 2005.

## Publications in conference proceedings

1. I.B. Földes, J.S. Bakos, K.Gál, G. Kocsis, **E. Rácz**, S. Szatmári, S. Varró and G. Veres:  
*On Polarization Properties of Harmonic Generated in Laser Plasmas on Solid Surfaces*,  
15th Europhysics Conference on Atomic and Molecular Physics of Ionized Gases, Abstracts (2000) **Vol. 24F**, pp. 380-381, (Eds.: Z. Donkó, L. Jenik, J. Szigeti), Miskolc-Lillafüred, Hungary, 26-30 August 2000.
2. Veres Gábor, Bakos József, Földes István, Gál Kinga, **Rácz Ervin**, Szatmári Sándor, Varró Sándor:  
*Szilárdtest lézerplazmában keltett felharmonikusok polarizációs tulajdonságai*,  
IV. Szimpózium a hazai kvantumelektronikai kutatások eredményeiről, Kvantum-elektronika 2000., pp. P-20, Budapest, 2000.11.03. (in hungarian)
3. **Rácz Ervin**, Földes István, Kocsis Gábor, Veres Gábor, Szatmári Sándor:  
*Felharmonikusok keltése szilárdtest lézerplazmákban nagy lézerintenzitásokon*,  
V. Szimpózium a hazai kvantumelektronikai kutatások eredményeiről, Kvantum-elektronika 2003., ISBN 963 372 6298, pp. P17., Budapest, 2003.10.21. (in hungarian)
4. **E. Rácz**, K. Eidmann, I.B. Földes, G. Kocsis, S. Szatmári, G. Veres:  
*Polarization- and Propagation Properties of High Harmonics for  $10^{17} \frac{W}{cm^2}$  Intensities*,  
31st EPS Conference on Plasma Physics, London, 28th June-2nd July 2004., ECA **Vol. 28G**, P-5.008, 2004.
5. **E. Rácz**, I.B. Földes, and S. Szatmári:  
*Investigation of plasma critical surface rippling by harmonics generation in laser plasmas*,  
International Conference PLASMA-2005 on Research and Applications of Plasmas, Opole, Poland, September 6-9, 2005. (accepted for publication in AIP proceedings)



6. **E. Rácz**, I.B. Földes, L. Ryć:  
*X-ray radiation measurements with photodiodes in plasmas generated by  $10^{17} \frac{W}{cm^2}$  intensity KrF excimer laser pulses*,  
International Conference PLASMA-2005 on Research and Applications of Plasmas,  
Opole, Poland, September 6-9, 2005. (accepted for publication in AIP proceedings)

## Oral presentations and some other publications

1. **Ervin Rácz**:  
*Investigation of generation of high harmonics in laser plasmas at  $10^{17} \frac{W}{cm^2}$  intensities*,  
Second Hungarian Plasma Physics Workshop, MTA KFKI RMKI, 22-24 April 2004.  
(oral presentation)
2. Földes István, **Rácz Ervin**:  
*Harmonikuseltés és lézerplazma spektroszkópia*,  
Lézertea 2004, MTA KFKI RMKI - MFA, Budapest, 2004. november 19. (oral presentation, in hungarian)
3. M. Kaluza, H. Habara, G.D. Tsakiris, K. Eidmann, K.J. Witte, I. Földes, **E. Rácz**:  
*Relativistic Electron Transport in Preformed Overdense Plasma*,  
28th EPS Conference on Controlled Fusion and Plasma Physics, Madeira, 18 - 22 June 2001.
4. K. Witte, U. Andiel, F. Brandl, K. Eidmann, E. Fill, I. Földes, H. Habara, D. Habs, M. Hegelich, R. Mancini, J. Meyer-ter-Vehn, M. Kaluza, S.Karsch, G. Pretzler, **E. Rácz**, T. Schlegel, G. Tsakiris:  
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Second International Conference on Inertial Fusion Sciences and Applications (IFSA 2001), **IFSA0946**: Kyoto Japan, September 9-14, 2001.
5. G. Veres, G. Kocsis, **E. Rácz**, S. Szatmári:  
*Doppler shift of femtosecond laser pulses from solid density plasmas*,  
Ultrashort High-energy radiation and Matter, Conference, Varenna, Italy, October 7-10, 2003, (poster)
6. I.B. Földes and **E. Rácz**:  
*Harmonics generation and critical surface rippling in laser plasmas*,  
Third International Conference on Superstrong Fields in Plasmas 19 - 24 September 2005, Villa Monastero, Varenna (Lc), Italy (poster)