Annual Summary Document Template*

1. Cover Page (1 page):

• Group list (physicists, staff, postdocs, students);

Director of proiect: prof.dr.univ. Niculae N. Pușcaș
Researchers:
Ș.l.dr. Maria-Ana Popovici
Ș.l.dr. Mihai Stafe
Ș.l.dr. Constantin Neguțu
Ș.l.dr. Georgiana Vasile
Ș.l.dr. Romeo Ionică

• Specific scientific focus of group;

The research activity of the group was devoted to the theoretical study of the mechanism of high order harmonic generation in ablation gas jets and the dosimetric estimates of the induced radiation.

• Summary of accomplishments in the last year.

In the last year we performed a theoretical analysis of high order harmonic generation (HHG) in gas jets with high intensity laser pulses. We estimated several laser and target parameters (i.e. focusing parameters- focus length, beam radius at the waist, length of the nonlinear medium, and concentration of the gas jet) involved in efficient conversion of a Nd^{3+} :YAG nanosecond laser radiation to higher frequencies. Also, we evaluated the wave vector mismatches, length of the nonlinear medium and focusing parameters for efficient generation of the fifth and seventh order harmonics generated in mixtures of sodium vapors and xenon. The theoretical findings are in good agreement with the experimental results published in the literature. In the case where the gas jet is produced by laser ablation, the condition for obtaining optimum gas density profile for HHG was evaluated by solving numerically the hydrodynamics of the ablation jet produced by a nanosecond laser pulse. The equations concerning the mass, momentum and energy conservation in the laser-target interaction were solved by using numerical FEM method implemented în MATLAB in order to estimate the ablation gas jet properties as a functions of the target, laser and ambient conditions.

Also, we performed a Monte Carlo simulation study meant to give a dosimetric evaluation of several typical electron sources at ultrahigh intensity laser facilities. We chose source terms which are predicted by theory and PIC simulations and used scaling laws to get the corresponding parameter values at 10 PW, and source terms which have already been measured at 1 PW, in an atempt to cover a wider range of energies and typical energy/spatial distributions. Parallel computations by two of the most widely used Monte Carlo radiation transport codes - FLUKA and GEANT4 allowed for a successful cross-check of the simulations results. “All particle” fluences were also retrieved, their distribution being meaningful for the physical processes which are expected to generate secondaries. Usually these are responsible for the radioprotection problems, as the primary beam can be more or less easily stopped in beamdumps. We also calculated the secondaries energy spectra in order to be able to compare the radiological effects of the selected electron sources at the same position in space, after specific interactions with the same medium. We showed that the level of maximum energy by itself (or the energy distribution) does not determine univoquely the total radiation dose. Similarly, the spatial distribution is just another factor which decides the doses given by one source term or another. Our calculations confirmed that correct predictions of Monte Carlo codes rely entirely on a good definition of the source terms, and this should be a matter of careful consideration where radiation protection issues are concerned.

2. Scientific accomplishments (max. 3 pages) – Results obtained in the last year.

In the last year we performed a theoretical analysis of high order harmonic generation (HHG) in gas jets with high intensity laser pulses. Based on the models existing in literature and considering a driving laser beam with a Gaussian intensity profile in the radial direction, we evaluated several geometrical irradiation parameters for which one can obtain the maximum power of the fifth and seventh harmonic.

In the case of a long focus, the intensity of the fifth harmonic radiation reaches its maximum for $b/L=1/2$, where L is the length of the nonlinear medium (Fig. 1) if the wave vector mismatch for the direct fifth harmonic generation process Δ_{15} and that corresponding to the step processes involving third harmonic generation Δ_{13} are both of them zero ($\Delta_{15}=\Delta_{13}=0$). In the case of tight-focusing of the laser beam considering that the beam is focused in the center of the nonlinear medium $\Delta_{15}=6/b$ for the direct fifth harmonic generation process and $\Delta_{15}=2\Delta_{13}=4/b$ for that corresponding to the step processes involving third harmonic generation process. The above mention conditions may be obtained using gaseous mixtures like: alkali metal vapors (e. g. sodium) and noble gases as buffer (e. g. xenon) with suitable concentrations, N_{Xe}/N_{Na}, suitable length of the nonlinear medium, and focusing parameters (focus length, beam radius at the waist).

---

*Please fill in all the required items and do not alter the template
*1020 hours = 170 average monthly hours x 6 months

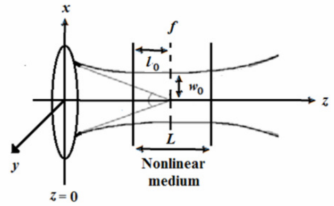
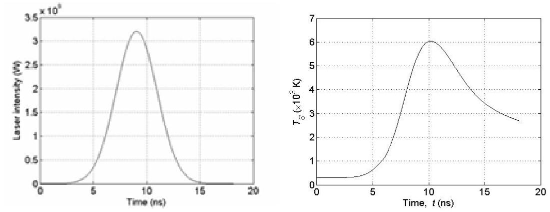


Fig. 1. The propagation of the Gaussian beam through the nonlinear medium.

In the case of a laser radiation with $\lambda = 1.06\,\mu m$ (Nd^{3+}:YAG) and 2.5×10^{8} W pump power, $N_{Xe}/N_{Na} = 32.2$, focus length of about 28 cm the above mentioned calculated parameters are in good agreement with the experimental results being obtained a fifth harmonic radiation with 4 kW power and an efficiency about 5×10^{-6}. The above mention parameters were evaluated in the case of seventh harmonic generation process. In the case of a long focus the intensity of the seventh harmonic radiation reaches its maximum for $b/L = 1/2$, where L is the length of the nonlinear medium (Fig. 2) if the wave vector mismatch for the direct seventh harmonic generation process Δ_{17} and that corresponding to the step processes involving fifth harmonic generation Δ_{15} are both of them zero ($\Delta_{17} = \Delta_{15} = 0$). In the case of tight-focusing of the laser beam considering that the beam is focused in the center of the nonlinear medium $\Delta_{15} = 10/b$ for the direct seventh harmonic generation process and $\Delta_{15} = 4/b$ for that corresponding to the step processes involving fifth harmonic generation process. The above mentioned evaluated parameters are in good agreement with the experimental results obtained in the seventh order harmonic generation [1].

In order to determine the optimum timing for sending the intense driving laser pulse through the non-linear conversion medium, we run simulations to find properties of the ablation plume conversion medium. This is because the harmonics intensity is a function of gas density. The fluence of the focused ablation laser pulse (4.5 ns, 1064 nm) was set to 15 J/cm^2, which gives a peak intensity of ~3GW/cm^2 at the Al target surface. These parameters determine a high density ablation plume, above 10^{19} cm^{-3} as needed in most HHG in gas jets where backing pressure of several bars are used, and low ionization degree (less than 1% as estimated experimentally [2]). The Gaussian temporal profile of the laser pulse is presented in Fig. 2(a) whereas the temporal evolution of the temperature of target surface is presented in Fig. 2(b). The peak temperature of the target surface is ~6000 K, slightly below the critical point of Al, and is reached at ~10 ns, which very close to the 9 ns when the peak of the laser pulse intensity is present. The spatial distribution of the temperature and density of the ablation plume at different times relative to the onset of laser pulse is presented in Fig. 3(a,b). One can see in Fig. 3(a) that the maximum plume temperature of ~3500 K is reached at about 13 ns, 'long' after the peak of the laser pulse intensity. Fig 3(b) depicts the spatial distribution of the plume density at different times. The figure indicates that a density of several 10^{19} cm^{-3}, which is suitable for efficient harmonic generation, can be obtained after the peak of the laser pulse at heights of several tens of microns above the target surface. When the saturated vapor pressure becomes smaller that the pressure of the ablation plume (calculated by using previous data regarding density and temperature of the plume), the ablation velocity becomes negative and re-condensation begins. From the continuity equation we find that the local plume velocity near the target surface becomes also negative due to re-condensation process (see black curve in Fig. 3(c)). Thereby, the evaporation depth, calculated by time integration of the evaporation velocity decays when re-condensation comes into play at times larger than ~13 ns, when maximum of the plume temperature is reached (see fig. 3(d)).



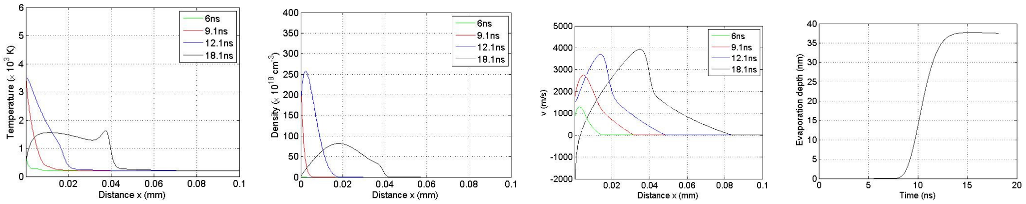
(a) (b)

Fig. 2. (a) Laser pulse intensity profile. (b) Temperature of the target surface.

References

1. *Pulsed Laser Ablation of Solids, Basics, Theory and Applications*, M. Stafe, A. Marcu, N. N. Puscas, Springer Series in Surface Sciences, Vol. 53, Ser. Eds: G. Ertl, Berlin, Germany, (2014).

2. *Ablation plasma spectroscopy for monitoring in real-time the pulsed laser ablation of metals*, M. Stafe and C. Negutu, Plasma Chemistry and Plasma Processing, **32**(3), p. 643-653, (212).



(a) (b) (c) (d)

Fig. 3. (a) Spatial distribution of the ablation plume temperature at different irradiation times. (b) Spatial distribution of the ablation plume density at different times. (c) Spatial distribution of the ablation plume velocity at times. (d) Time dependent evaporation depth of the ablated target.

FLUKA and GEANT4 results

We performed a search of the scientific literature concerning laser acceleration of electrons and protons and we draw the conclusions regarding the characteristics of possible ionizing radiation source terms.

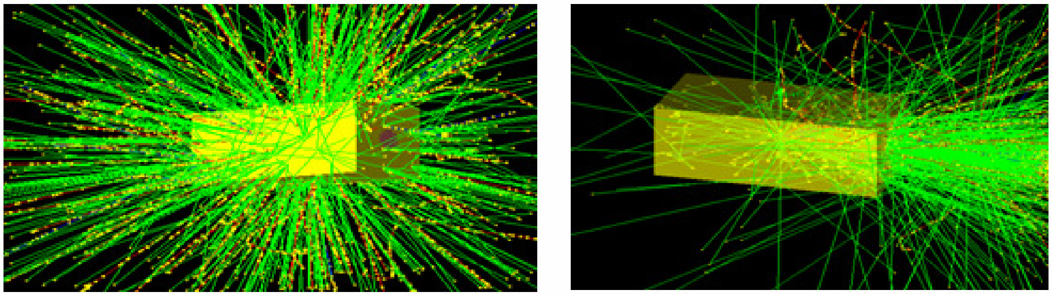


Fig. 4. Particle tracking representation of two electron source terms ST2 (left) and ST3 (right). Neutral particles are represented in green. Interaction chamber and detector are visible.

The scientific papers that were reviewed present theoretical, particle-in-cell simulation and experimental approaches. We organized the information according to the main electron and proton acceleration regimes. A collection o more then 150 research papers is presented in a bibliography which includes relevant papers in each field.

The source terms we selected do not fit into the category of ideally monoenergetic, monodirectional beams. Radiation protection and shielding of such sources are well established due to currently vast experience with conventional accelerators. Instead, we worked with more or less broad energetic and spatial distributions that may be obtained (according to PIC simulations and scaling laws) or even with experimental data. We implemented in FLUKA and GEANT4 seven electron (ST1 – ST4) and proton (ST-P1 – ST-P3) source terms. In Fig. 4 a particle tracking graph representation of ST2 and ST3 obtained by GEANT4 is presented.

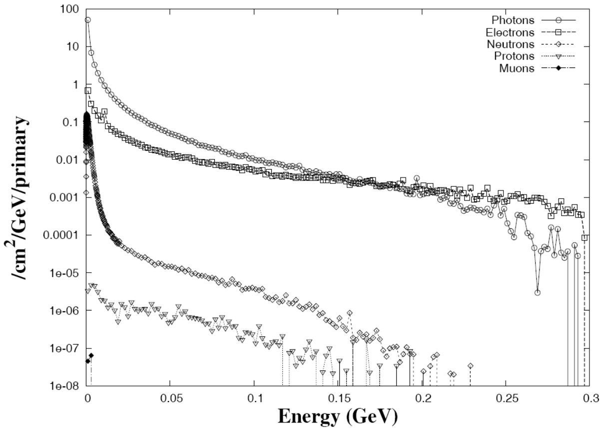


Fig. 5. ST4 - Secondary particles energy spectra at the front face of the "detector " region, in the backward direction

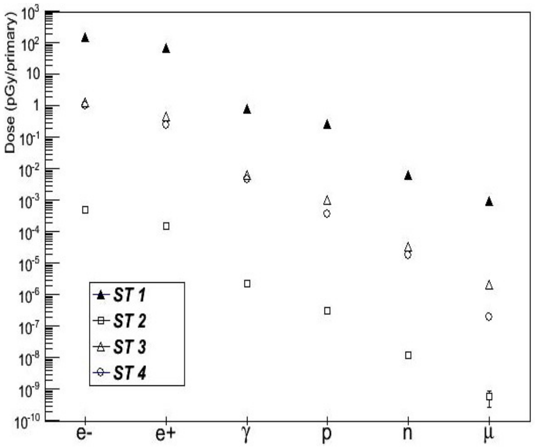


Fig. 6. All particle dose (pGy/primary electron) due to the electron source terms in the "Detector " region.

In FLUKA a set of SOURCE subroutines were written, in order to sample the required energy and spatial distributions of the primary particles. We applied a discrete distribution sampling technique when experimental data were used as such. We also used continuous distribution sampling by inverting the normalized cumulative distribution function or by applying rejection – acceptance method, where the first one was not possible. A range of typical angular distributions were also sampled. The doses due to our choice of electron and proton source terms were obtained by parallel FLUKA and GEANT4 calculations. The simulation setup was minimal and it contained a typical vacuum interaction chamber with extraction window and a “detector” region placed in air. The main results concerning “all particle” doses expressed in pGy/primary particle were presented in comparative data tables. The dosimetric evaluation of the implemented ST encompasses secondary radiation. Energy spectra of secondaries were recorded for each ST (see, for example, Fig. 5). A comparison between different ST was performed. In Fig. 6, for example, the doses due to secondary particles that are generated by the selected electron source terms are presented.

We also scored the “all particle fluence” in the simplified geometry of the simulations. The results were presented as bi-dimensional maps (see Fig. 7 for ST3 electron source (left) and ST-P2 proton source). Our dosimetric evaluations showed that the energetic and spatial distributions compete in defining a complex radiation field which is particular which is particutar too the source term. Choosing the right, typical source terms is an important step for our project. They are going to be used in a more complex, more realistic geometry where the doses are expected to be different, especially due to the primary shielding, the walls of the experimental bunkers.

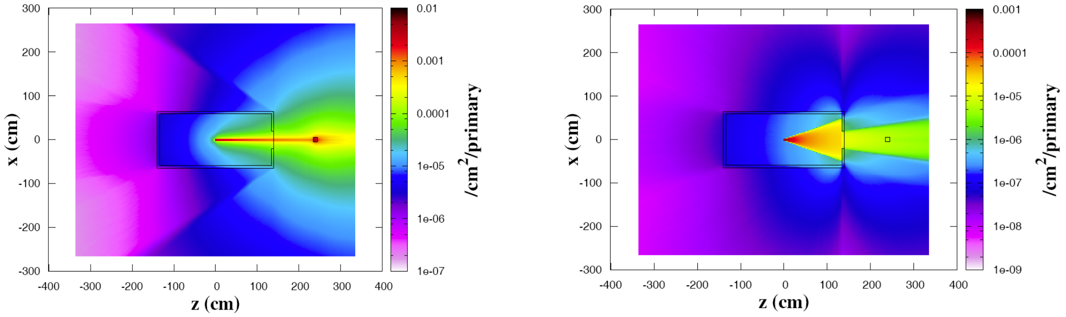


Fig. 7. All particle fluence(particle/cm^2/ primary) generated by ST3 electron source term (left) and ST-P2 proton source term, in the Oxz plane, at y = 0. The source is placed at the center of the interaction chamber.

We also scored the “all particle fluence” in the simplified geometry of the simulations. The results were presented as bi-dimensional maps (see Fig. 7 for ST3 electron source (left) and ST-P2 proton source. Our dosimetric evaluations showed that the energetic and spatial distributions compete in defining a complex radiation field which is particular to the sorce term. Choosing the right, typical source terms is an important step for our project. They are going to be used in a more complex, more realistic geometry were the doses are expected to be different, especially due to the primary shielding, the walls of the experimental bunkers.

3. Group members (table):

- List each member, his/her role in project and the Full Time Equivalent(FTE) % time in project. The FTE formula to be used is: FTE = Total number of worked hours in the last year/1020 hours[†];

No. crt.	Member	Role in project	FTE % time in project
1.	Niculae N. Puşcaş	director of the proiect	9,50
2.	Maria-Ana Popovici	member	13,00
3.	Mihai Stafe	member	13,00
4.	Constantin Neguţu	member	10,10
5.	Georgiana Vasile	member	9,20
6.	Romeo Ionică	member	8,60

- **List of PhD/Master students and current position/job in the institution.**

-

4. Deliverables in the last year related to the project:

- **List of papers (journal or conference proceeding);**
- **1.** ***Preliminary dosimetric evaluation of electron source terms at pw laser systems*****,** Maria-Ana POPOVICI, Romeo IONICĂ, Gh. CATA-DANIL, University “Politehnica” of Bucharest, Scientific Bulletin, Series A: Applied Mathematics and Physics, Vol. **77**, No. 1, 2015, accepted for publication.

[†] 1020 hours = 170 average monthly hours x 6 months

- **2.** ***Analysis of several high order harmonic generation in gaseous media***, M. Stafe, C. Negutu, G. C. Vasile, N. N. Puscas, University “Politehnica” of Bucharest, Scientific Bulletin, Series A: Applied Mathematics and Physics, Vol. **77**, No. 1, 2015, sent for publication.

- **List of talks of group members (title, conference or meeting, date);**

-

- **Other deliverables (patents, booksetc.).**

  -

5. Further group activities (max. 1 page):

- **Collaborations, education, outreach.**

M. Stafe, C. Negutu, G. C. Vasile, N. N. Puscas (director) are members in the project: Optical plasmonic 2 D memories with active chalcogenic glass substrate (MEMOPLAS); 2011-2016, University POLITEHNICA of Bucharest being the lider of the project. The members carried theoretical and experimental work on finding the optimal conditions for resonant coupling of the radiation in plasmonic waveguides with metal-chalcogenide layers.

M. Stafe and C. Negutu are members in the project: Ultrafast laser Facility with Optimized high order harmonics UltraViolet sources (UFOUV); 2011-2016, University POLITEHNICA of Bucharest being the partner of the project. The members carried theoretical study on finding the properties of the gas jet conversion medium, and also on the method of separation of the harmonic emission from the gas jet without metallic filters.

The knowledge acquired during this project is used for teaching two courses in the IALA master program at UPB: 'Laser and Optics' given by Prof. Dr. N.N. Puscas, and 'High Power Lasers Engineering and applications' given by S.l. Dr. Mihai Stafe

6. Financial Report for the last year (see the Annex).

7. Research plan and goals for the next year (max. 1 page).

Stage II. Experiments on the production of laser ablation jets for HHG and simulations of the transport of radiation in zones E1/E6 and E4/E5

Activity II.1. Production of the ablation puffs on different types of solid targets with nanosecond laser pulses; study/report.

Activity II.2. Numerical analysis of the ablation puffs as a function of laser properties and target properties (optical and thermal properties); study/report.

Activity II.3. Experimental characterization of the ablation puffs by shadowgraphy imaging and by optical spectroscopy

Activity II.4. Geometry implementing of E1/E6, E4/E5 experimental areas in FLUKA and GEANT; dose equivalent maps in HPLS experimental areas, corresponding of selected source terms; scientific report.

Activity II.5. Dosimetric estimates of E1/E6, E4/E5 experimental areas with selected source terms; scientific report.

Activity II.6. Dissemination of the results; 3 scientific papers accepted/published in ISI quoted journals and 2 participations at international conferences.

Annex

Financial Report
according to the regulations from H.G. 134/2011

			lei
Type of expenditures		**Year 2014**	
		Value	
		Planned	**Realized**
1	**PERSONNEL EXPENDITURES**, from which:	**64000**	**63119**
	1.1. wages and similar income, according to the law	50181	51510
	1.2. contributions related to salaries and assimilated incomes	13819	11609
2	**LOGISTICS EXPENDITURES**, from which:	**5818**	**6924,98**
	2.1. capital expenditures	5818	0
	2.2. stocks expenditures	0	6749,32
	2.3. expenditures on services performed by third parties, including:	0	175,66
	....	0	0
3	**TRAVEL EXPENDITURES**	**0**	**0**
4	**INDIRECT EXPENDITURES – (OVERHEADS) * (25% from Personnel Expenditures)**	**16000**	**15774,02**
TOTAL EXPENDITURES *(1+2+3+4)*		**85818**	**85818**

* Specify the rate (%) and key of distribution (excluding capital expenditures).

To be filled in for:
- the project leader;
- for each of the parteners (if any);
- for the whole project.