

## Fission of actinides using a tabletop laser

H. SCHWOERER<sup>1</sup>(\*), F. EWALD<sup>1</sup>, R. SAUERBREY<sup>1</sup>, J. GALY<sup>2</sup>,  
J. MAGILL<sup>2</sup>, V. RONDINELLA<sup>2</sup>, R. SCHENKEL<sup>2</sup> and T. BUTZ<sup>3</sup>

<sup>1</sup> *Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität*

*Max-Wien-Platz 1, 07743 Jena, Germany*

<sup>2</sup> *European Commission, Joint Research Centre, Institute for Transuranium Elements  
Postfach 2340, 76125 Karlsruhe, Germany*

<sup>3</sup> *Nukleare Festkörperphysik, Universität Leipzig - Linnéstr. 5, 04103 Leipzig, Germany*

(received 15 July 2002; accepted in final form 8 October 2002)

PACS. 25.85.Jg – Photofission.

PACS. 52.38.Ph – X-ray, gamma-ray and particle generation.

PACS. 25.20.-x – Photonuclear reactions.

**Abstract.** – Powerful tabletop lasers are now available in the laboratory and can be used to induce nuclear reactions. We report the first demonstration of nuclear fission using a high repetition rate tabletop laser with intensities of  $10^{20}$  W/cm<sup>2</sup>. Actinide photo-fission has been achieved in both <sup>238</sup>U and <sup>232</sup>Th from the high-energy bremsstrahlung radiation produced by laser acceleration of electrons. The fission products were identified by time-resolved  $\gamma$ -spectroscopy.

*Introduction.* – Nuclear reactions induced by ultraintense lasers became amenable in the last couple of years by the evolution of experimental relativistic laser plasma physics. Highly sophisticated tabletop laser systems generate ultrashort laser pulses focusable to intensities of  $10^{20}$  W/cm<sup>2</sup>. Matter in the focal spot is instantaneously turned into a hot dense plasma. In this intense light field electrons are accelerated to relativistic energies and the dense plasma heats up to temperatures of several MeV. Maximum detected energies of emitted particles (electrons, positrons, protons, neutrons) and photons (bremsstrahlung) reach tens or even hundreds of MeV [1–4].

Laser interaction with matter at these very high intensities is of relevance in a number of areas ranging from advanced accelerator concepts using plasmas, to astrophysics and inertial confinement fusion. The highly energetic particles and photons emitted by the plasma can induce nuclear reactions such as photofission ( $\gamma, f$ ) or photo-neutron generation ( $\gamma, n$ ) by bremsstrahlung photons [5–9]. These reactions which occur above specific energy thresholds provide a technique for the calibration of bremsstrahlung spectra of laser plasmas in an energy range above a few MeV and therefore allow for the investigation of mechanisms responsible for heating of plasmas to relativistic temperatures.

---

(\*) E-mail: [schwoerer@ioq.uni-jena.de](mailto:schwoerer@ioq.uni-jena.de)

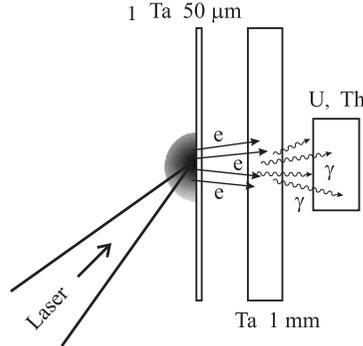


Fig. 1 – Setup of the experiment. The laser beam is focused onto the tantalum target, where it generates a hot plasma on the surface. Electrons accelerated in this plasma enter the second part of the target, which serves as a bremsstrahlung converter. The samples  $^{232}\text{Th}$  and  $^{238}\text{U}$  are positioned directly behind this source of  $\gamma$ -radiation.

An intriguing possibility is to use high-energy bremsstrahlung to photo-transmute the most hazardous components of nuclear waste. These reactions can be investigated with tabletop lasers and irradiation of targets of actinides and fission products. Photo-induced reaction cross-sections for  $(\gamma, n)$  and  $(\gamma, f)$  reactions are of the order 0.1 barn [10]. Although this seems a rather low value, there are some key components of nuclear waste in which photoreaction cross-sections are higher than for neutron capture reactions [10]. The high  $\gamma$ -energies reached with laser-produced bremsstrahlung qualify this source for investigation of photo-induced reaction cross-sections of actinides in comparison to neutron capture cross-sections.

Photofission of uranium by means of bremsstrahlung from laser-produced plasmas was first reported in 2000 by two groups, using large single-shot glass laser systems designed for laser fusion experiments, namely the VULCAN laser at Rutherford Appleton Laboratory (RAL) and NOVA at the Lawrence Livermore National Laboratory (LLNL) [7, 8].

In this contribution, we report the first photofission of  $^{232}\text{Th}$  with laser-generated hard X-rays, and the first laser-induced fission of heavy nuclei with a high repetition rate tabletop laser system.

*Experiment.* – The experiments were performed with the Jena 15 TW laser system. The Ti:sapphire laser system, based on chirped pulse amplification, generates laser pulses with a maximum energy of 1.2 J within 80 fs at a repetition rate of 10 Hz. The high spatial and temporal quality of the beam allows focusing of the pulses to a spot of less than  $5\ \mu\text{m}^2$ , supporting a peak intensity of  $10^{20}\ \text{W}/\text{cm}^2$ .

The laser beam is focused onto a tantalum target under  $45^\circ$  in parallel polarization. This geometry allows for high absorption of laser light into a plasma [11]. The target itself consists of two sheets of tantalum with a thickness of  $50\ \mu\text{m}$  and 1 mm, respectively (fig. 1). At  $10^{20}\ \text{W}/\text{cm}^2$  the laser pulse produces a relativistic plasma on the surface of the first thin sheet. Plasma electrons are accelerated to relativistic energies within the intense laser field. Usually, the energy distribution of relativistic electrons, accelerated by the ponderomotive force of the laser field, is assumed to be Maxwellian with a characteristic temperature given by [12]

$$k_{\text{B}}T_{\text{e}} \simeq 0.511\ \text{MeV} \left[ \left( 1 + \frac{I\lambda^2}{1.37 \times 10^{18}}\ \text{Wcm}^{-2}\mu\text{m}^2 \right)^{1/2} - 1 \right]. \quad (1)$$

From this a hot electron temperature of about 2 MeV is predicted for a laser intensity of

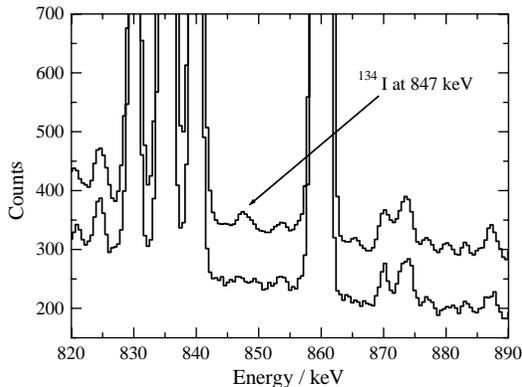


Fig. 2 –  $\gamma$ -spectrum of the irradiated  $^{232}\text{Th}$  sample (upper line). Strong emission occurs from  $^{232}\text{Th}$  decay products, mainly  $^{228}\text{Ac}$  and  $^{208}\text{Tl}$ . Emission from fission products, in this case from  $^{134}\text{I}$  at 847 keV, however, is clearly identifiable. The second line, which is lowered for clarity, represents a background measurement, taken before irradiation of the sample.

$10^{20}$  W/cm<sup>2</sup>. These fast electrons impinge on the second layer of the target, which serves as an efficient bremsstrahlung converter.

Two actinide samples were irradiated in this bremsstrahlung field: a plate of thoriumoxide (12 g) and depleted metal uranium (11 g). The samples were placed directly behind the tantalum target as shown in fig. 1. For each of the samples 10000 laser shots were applied onto the tantalum within 20 min, altogether loading about 5 kJ of laser energy on the bremsstrahlung converter. After irradiation, the actinide samples were taken from the target area to analyse  $\gamma$ -emissions from fission products with a Ge-detector.  $\gamma$ -spectra from the irradiated samples were taken in successive time intervals. In that way it was possible to identify fission products not only by their characteristic  $\gamma$ -spectra but also by their decay constants. As both samples  $^{232}\text{Th}$  and  $^{238}\text{U}$  are already strong  $\gamma$ -emitters due to the presence of relatively short-lived daughters in radioactive equilibrium, great care has to be taken on background measurements before irradiation with the laser-produced  $\gamma$ -photons. The integrated  $\gamma$ -radiation activities of the thorium oxide and uranium samples were  $6 \cdot 10^5$  Bq and  $5 \cdot 10^5$  Bq, respectively.

*Results.* – The  $\gamma$ -spectra of the irradiated thorium and uranium show a number of signatures of typical short-lived fission products with mass numbers around 90 and 130 as expected from  $(\gamma, f)$ -reactions. The isotopes  $^{134}\text{I}$ ,  $^{135}\text{I}$ ,  $^{92}\text{Sr}$ ,  $^{138}\text{Cs}$ , and  $^{128}\text{Sn}$  could be identified from the measured spectra of  $^{232}\text{Th}$ , whereas  $^{134}\text{I}$  and  $^{92}\text{Sr}$  indicate photofission of  $^{238}\text{U}$ . The spectrum of  $^{232}\text{Th}$  shown in fig. 2 shows the  $\gamma$ -line of the fission product  $^{134}\text{I}$  at an energy of

TABLE I – Identified fission products of  $^{232}\text{Th}$ .

Nuclide	$\gamma$ -energy (keV)	Half-life [13]	Measured half-life	Fission yield of $^{232}\text{Th}(n, f)$	Number of fissions
$^{134}\text{I}$	847	52.5 min	57.5 min	$9.96 \cdot 10^{-3}$	$7.5 \cdot 10^7$
$^{92}\text{Sr}$	1384	2.71 h	2.34 h	$4.03 \cdot 10^{-3}$	$6.7 \cdot 10^7$
$^{138}\text{Cs}$	1435	33.4 min	34.7 min	$1.58 \cdot 10^{-3}$	$12 \cdot 10^7$
$^{135}\text{I}$	1260	6.57 h	6.22 h	$2.27 \cdot 10^{-2}$	$4.8 \cdot 10^7$
$^{128}\text{Sn}$	482	59.1 min	64 min	$2.14 \cdot 10^{-3}$	$9.3 \cdot 10^7$

TABLE II – Identified fission products of  $^{238}\text{U}$ .

Nuclide	$\gamma$ -energy (keV)	Half-life [13]	Measured half-life	Fission yield of $^{238}\text{U}(n, f)$	Number of fissions
$^{134}\text{I}$	847	52.5 min	49.6 min	$5.97 \cdot 10^{-3}$	$9.7 \cdot 10^7$
$^{92}\text{Sr}$	1384	2.71 h	2.67 h	$9.76 \cdot 10^{-3}$	$9.2 \cdot 10^7$

847 keV. Correction with the detector efficiency provides the activity of the particular  $\gamma$ -line of the fission product at the time of measurement. The total amount of a certain fission product produced at the end of the irradiation time may then be deduced from this activity, the emission probability of the particular  $\gamma$ -line and the half-life. The total number of photo-induced fissions was calculated independently for each fission product from its activity and its fission yield. The latter is the fraction of a certain isotope generated through fission of the actinide nucleus and can be assumed to be comparable to the fission yield for the neutron-induced reaction of the high-energy (14 MeV) fission of  $^{232}\text{Th}$  or  $^{238}\text{U}$ . Tables I and II summarize the identified isotopes produced by the bremsstrahlung in the  $^{232}\text{Th}$  and  $^{238}\text{U}$  samples. The half-lives which are obtained from the decay measurements, shown in fig. 3, are compared to literature values. For thorium, the total number of fission events at the end of the  $\gamma$ -irradiation is found to be in the order of  $7 \cdot 10^7$  in total or  $7 \cdot 10^3$  per laser shot, *i.e.*  $\sim 10^4$  per 1 J of laser energy. The number of fission events derived for uranium is  $2 \cdot 10^4$  per 1 J. As mentioned previously, the actinide samples are strong  $\gamma$ -emitters, producing a large Compton background. This leads to an error of the activity of 30–50% (see fig. 3). Consequently, the number of fissions which is calculated independently of the different fission products may vary by about a factor of two (see table I).

*Discussion.* – Reactions such as  $(\gamma, n)$  and  $(\gamma, f)$  of  $^{238}\text{U}$  were observed with laser-generated bremsstrahlung at the VULCAN laser system at RAL and the PetaWatt laser system NOVA at LLNL [7, 8]. The laser pulses of these systems achieve similar peak intensities as in Jena, namely above  $10^{19} \text{ W/cm}^2$  and  $10^{20} \text{ W/cm}^2$  at VULCAN and NOVA, respectively, compared to  $10^{20} \text{ W/cm}^2$  in the Jena system. Following eq. (1) and taking into account the wavelengths  $\lambda = 1 \mu\text{m}$  of glass lasers and  $\lambda = 0.8 \mu\text{m}$  of the Ti:sapphire laser, hot electron

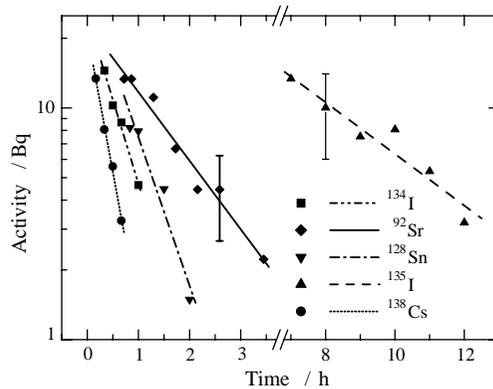


Fig. 3 – Decay characteristics of fission products from bremsstrahlung-induced fission of  $^{232}\text{Th}$ . The deduced half-lives are in good agreement with literature values as shown in table I. Symbols indicate experimental data.

temperatures of 1 MeV, 4 MeV and 2 MeV are expected at VULCAN, NOVA and in Jena, respectively. The temperature of the hot electrons determines the yield of high-energy  $\gamma$ -photons, since the spectrum of bremsstrahlung generated with a Maxwellian distribution of electron energies is again Maxwellian with almost the same slope [14].

However, due to the much longer pulse duration (VULCAN: 1 ps, NOVA: 450 fs) and the comparatively poor focusability of glass laser beams, the required pulse energy on target is significantly higher (50 J and 260 J *vs.* 500 mJ in Jena). For appropriate comparison of the total number of fission events induced in the three experiments, the fissions per laser pulse energy can be considered:  $7 \cdot 10^4$  fissions per joule in the NOVA experiment,  $2 \cdot 10^4$  fissions per joule in RAL and  $10^4$  fissions per joule in Jena. The difference between the glass laser systems can be explained by the somewhat higher electron temperature generated with the PetaWatt laser. The strength of the tabletop Ti:sapphire laser is its compact construction and, in particular the high repetition rate of 10 Hz which can overcompensate the much higher pulse energy of the above-mentioned large facilities. The typical repetition rate of the glass laser systems is  $10^{-3}$  to  $10^{-4}$  Hz.

At present, nuclear activation is the only technique to measure the absolute electron and  $\gamma$ -energy distribution of relativistic laser-produced plasmas. Consequently, the integrated number of  $(\gamma, f)$  events per laser shot is a reliable measure of the integrated cross-section of the bremsstrahlung spectrum above the fission threshold, generated by the interaction of the laser pulse with the tantalum converter. The differential cross-sections of  $(\gamma, f)$ -reactions for  $^{232}\text{Th}$  and  $^{238}\text{U}$  are taken from [15]. They exhibit a threshold at 6.0 and 5.8 MeV, respectively and increase up to the giant resonance around 15 MeV to values of 70 mbarn (thorium) and 170 mbarn (uranium). The ponderomotive acceleration of electrons should lead to an electron temperature of  $T_e \approx 2$  MeV at a laser intensity of  $10^{20}$  W/cm<sup>2</sup> [12]. We assume a somewhat lower temperature of 1 MeV for bremsstrahlung photons within a more or less collimated beam, which is correct for bremsstrahlung produced by relativistic electrons [9]. From the total number of fissions and the fission cross-section we infer a Boltzmann energy distribution for bremsstrahlung photons:

$$n_\gamma = 3 \cdot 10^{10} / \text{MeV} \cdot e^{-E_\gamma / 1 \text{ MeV}} . \quad (2)$$

The total number of photons per laser shot with energies above the threshold around 6 MeV is then found to be  $N_\gamma(E_\gamma > 6 \text{ MeV}) = 7 \cdot 10^7$   $\gamma$ /shot. This number does not yet include the effect of pair production, which becomes a competing process to fission at MeV energies. The number of photons with energies above 500 keV is a lower threshold for the number of relativistic electrons with  $E_e \geq m_0 c^2$ . It is determined to  $N_\gamma(E_\gamma > 500 \text{ keV}) = 2 \cdot 10^{10}$   $\gamma$ /shot. This number leads to a total conversion efficiency from laser light into bremsstrahlung above 500 keV of  $5 \cdot 10^{-3}$ .

*Conclusions.* – We have, for the first time, demonstrated the photofission of  $^{232}\text{Th}$  by laser-produced bremsstrahlung. The fission products were identified by  $\gamma$ -spectroscopy as well as by time-resolved activity measurements. Deduced half-lives show excellent agreement with literature values, thus providing clear evidence of the fission process. The number of fission events has been determined to be about  $7 \cdot 10^3$  per laser shot, *i.e.*  $10^4$  per 1 J of laser energy. These numbers are comparable to those of the high-energy laser systems at RAL and LLNL, pointing out that the much higher repetition rate of the tabletop system is able to compensate for the difference in laser pulse energy. Thus it could be shown that photofission of heavy isotopes as  $^{232}\text{Th}$  and  $^{238}\text{U}$  is possible with a tabletop 15 TeraWatt laser system, which achieves ultra-high intensities of  $10^{20}$  W/cm<sup>2</sup> with a repetition rate of 10 Hz.

It was shown that the usage of high-energy fission barriers can be a very useful means to measure the absolute energy distribution of laser-produced bremsstrahlung. Using a variety of nuclear reactions such as  $(\gamma, n)$ ,  $(\gamma, xn)$ ,  $(\gamma, f)$ , and photoexcitation of long lived isomers  $(\gamma, \gamma')$  with different threshold energies would permit a better characterization of bremsstrahlung and electron spectra.

\* \* \*

We acknowledge the support of M. BETTI, J. JENSTRÖM, J. REBIZANT, W. WAGNER and T. WISS from the Institute for Transuranium Elements as well as B. BELEITES and F. RONNEBERGER from the Friedrich-Schiller-Universität Jena. The work was funded by the Deutsche Forschungsgemeinschaft (Schw 766/2) and the Bundesministerium für Bildung und Forschung (13N7925).

## REFERENCES

- [1] MALKA G. and MIQUEL J. L., *Phys. Rev. Lett.*, **77** (1996) 75.
- [2] GAHN C., TSAKIRIS G. D., PRETZLER G., WITTE K. J., THIROLF P., HABS D., DELFIN C. and WAHLSTRÖM C.-G., *Phys. Plasmas*, **9** (2002) 987.
- [3] HATCHETT S. P., BROWN C. G., COWAN T. E. *et al.*, *Phys. Plasmas*, **7** (2000) 2076.
- [4] PRETZLER G., SAEMANN A., PUKHOV A. *et al.*, *Phys. Rev. E*, **58** (1998) 1165.
- [5] BOYER K., LUK T. S. and RHODES C. K., *Phys. Rev. Lett.*, **60** (1988) 557.
- [6] KEY M. H., CABLE M. D., COWAN T. E. *et al.*, *Phys. Plasmas*, **5** (1998) 1966.
- [7] LEDINGHAM K. W. D., SPENCER I., MCCANNY T. *et al.*, *Phys. Rev. Lett.*, **84** (2000) 899.
- [8] COWAN T. E., HUNT A. W., PHILLIPS T. W. *et al.*, *Phys. Rev. Lett.*, **84** (2000) 903.
- [9] SCHWOERER H., GIBBON P., DÜSTERER S., BEHRENS R., ZIENER C., REICH C. and SAUERBREY R., *Phys. Rev. Lett.*, **86** (2001) 2317.
- [10] *Handbook on Nuclear Activation Data, Technical Report Series*, Vol. **273** (IAEA, Vienna) 1987.
- [11] TEUBNER U., BERGMANN J., VAN WONTERGHEM B., SCHÄFER F. P. and SAUERBREY R., *Phys. Rev. Lett.*, **70** (1993) 794.
- [12] WILKS S. C., KRUEER W. L., TABAK M. and LANGDON A. B., *Phys. Rev. Lett.*, **69** (1992) 1383.
- [13] MAGILL J., *Nuclides.net – An Integrated Environment for Computations on Radionuclides and Their Radiation* (Springer) 2002.
- [14] MCCALL G. H., *J. Phys. D*, **15** (1982) 823.
- [15] CALDWELL J. T., DOWDY E. J., BERMAN B., ALVAREZ R. A. and MEYER P., *Phys. Rev. C*, **21** (1980) 1215.