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Laser transmutation of iodine-129

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Received: 20 August 2003

Published online: 5 September 2003 • © Springer-Verlag 2003

ABSTRACT We report the first successful laser-induced transmutation of ¹²⁹I, one of the key radionuclides in the nuclear fuel cycle. ¹²⁹I with a half-life of 15.7 million years is transmuted into ¹²⁸I with a half-life of 25 min through a (γ , n) reaction using laser-generated Bremsstrahlung. The integral cross-section value for the (γ , n) reaction is determined. These experiments offer a new approach to studying transmutation reactions with neutral and charged particles without resource to nuclear reactors or particle accelerators.

PACS 52.38.Ph; 25.20.Lj

1 Introduction

Very high intensity near-infrared laser radiation, with intensities up to 10^{20} W/cm², can be used to induce a variety of nuclear reactions [1, 2] through the generation of a relativistic plasma, which emits high-energy beams of photons [3–5], neutrons [6–8], electrons [9, 10], protons [11] and ions [12–14]. The maximum detected energies of the emitted particles reach tens or even hundreds of MeV. Of particular interest is the fact that these nuclear reactions can be investigated in the laboratory using tabletop lasers. This development may open a completely new approach to identifying optimum reactions for transmutation of nuclear waste with neutral and charged particles without resource to nuclear reactors or particle accelerators. We report the transmutation of ¹²⁹I through a (γ , n) reaction using laser-generated Bremsstrahlung photons. The cross-section value for the (γ , n) reaction is determined from this experiment to be $> 220_{-100}^{+200}$ mbarn.

2 Iodine-129


Iodine is present in the environment in both stable (¹²⁷I) and radioactive forms. The most important of the radioisotopes is ¹²⁹I. It is produced naturally in the upper atmosphere through cosmic-ray spallation of xenon and by

spontaneous fission in the geosphere. Since the 1950s, the environmental levels of ¹²⁹I have increased due to atmospheric nuclear tests and to the use of nuclear power. These anthropogenic activities have resulted in a global increase of the ¹²⁹I/¹²⁷I ratio by two orders of magnitude from about 1.5×10^{-12} to 10^{-10} [15], although radiological effects remain negligible. Nuclear reactors are now the main sources of anthropogenic ¹²⁹I. Iodine as a fission element is generated in spent fuel at a rate of about 7 kg/GWe per year, with 80% of this inventory in the form of ¹²⁹I and 20% in the form of stable ¹²⁷I [16]. World-wide, based on the currently installed nuclear generating capacity of 360 GWe, this gives rise to an annual production rate of ¹²⁹I of approximately 2000 kg [15–17].

¹²⁹I decays by emitting a beta particle (150 keV), γ -photons (40 keV) and X-ray radiation (30, 34 keV) [18]. Although the low-energy beta particles barely penetrate the outer layers of the skin, the γ - and X-rays are more penetrating and present an external health hazard. More importantly, ¹²⁹I can be incorporated into the body through the food chain, drinking water and breathing air. Up to 30% of the iodine uptake in the body accumulates in the thyroid and is retained with a biological half-life of 120 days. Thyroid tumours can develop from the ionising radiation. Of the long-lived fission products (mainly ¹²⁹I, ⁹⁹Tc, ¹³⁵Cs), ¹²⁹I is the most radiotoxic [18].

Due to its long half-life, high radiotoxicity and mobility, ¹²⁹I is one of the primary risk considerations for several risk analyses and risk assessments in the disposal of nuclear waste. One approach to limit its mobility and potential health risk is to condition the iodine by employing engineered barriers into the disposal system. Their inclusion would significantly reduce the amount of ¹²⁹I released into the environment.

Ideally, however, the iodine released during nuclear fuel reprocessing should be transmuted to a stable product. Current proposals for the transmutation of actinides and long-lived fission products (LLFPs) are based upon neutron transmutation in fast neutron reactors [16, 19]. Through neutron-induced transmutation, for example, ¹²⁹I is transformed to ¹³⁰I, which decays with a half-life of 12 h to the stable gas ¹³⁰Xe. It is not clear, however, that transmutation based on neutrons is the optimum approach for all nuclides. For some problematic nuclides, neutron cross sections are too low to be effective. In addition, isotope mixes of a particular element like e.g. caesium may cause problems, because neutron-

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capture reactions on the lighter caesium isotopes $^{133,134}\text{Cs}$ result in a production of ^{135}Cs , i.e. more ^{135}Cs may be produced rather than destroyed [20].

Alternatively to neutron-induced transmutation, ^{129}I can be photo-excited to emit a single neutron and transmute to ^{128}I , which has a half-life of 25 min. 7% of the ^{128}I nuclei capture an electron and decay to ^{128}Te . The remaining 93% undergo β -decay into the stable ^{128}Xe . This β -decay has a characteristic γ -line at 443 keV with an emission probability of $I_\gamma = 17\%$, and is used to monitor successful photo-transmutation. In our experiment high-energy Bremsstrahlung from relativistic laser-produced plasmas is used to induce the photonuclear $^{129}\text{I}(\gamma, n)^{128}\text{I}$ reaction.

3 Laser-generated γ -source

Relativistic laser plasmas are bright incoherent sources of γ -radiation with energies up to many tens of mega-electronvolts. They can be produced by compact tabletop laser systems delivering laser pulses with joules of energy within less than 100 femtoseconds (fs). In the focus of these laser beams intensities of more than 10^{20} W/cm^2 may be generated. The ponderomotive force of the intense light field, which is the light pressure of the pulse, accelerates the electrons to relativistic energies and pushes them in the forward direction into the target (Fig. 1) [21, 22]. At high energies a Boltzmann-like distribution with a hot-electron temperature T_e is a good approximation. T_e scales with the square root of the laser intensity I . At $I \approx 3 \times 10^{18}\text{ W/cm}^2$ the temperature exceeds the electron rest mass of 511 keV and at 10^{20} W/cm^2 the electron temperature is $T_e \approx 3\text{ MeV}$ [21].

Within the solid target of typically high atomic number like tantalum the hot electrons are stopped and generate Bremsstrahlung. In the extreme relativistic case an exponential energy distribution of electrons leads again to an exponential photon spectrum with the same temperature [23]. In the

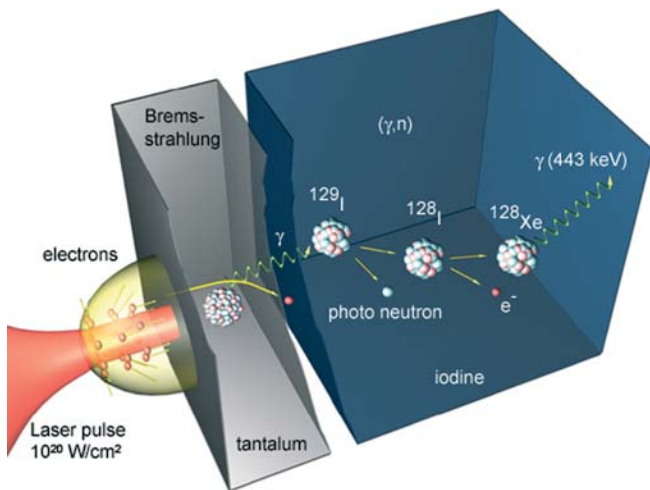


FIGURE 1 Experimental scenario: the high-intensity laser pulse produces a hot plasma on the surface of a tantalum foil. Relativistic electrons are stopped in the tantalum, efficiently generating high-energy Bremsstrahlung. The ^{129}I in the radioactive target is transformed into ^{128}I due to a (γ, n) reaction

intermediate case of electron temperatures of several MeV the photon temperature is slightly lower than T_e [24].

The spectra of the primarily produced electrons and the Bremsstrahlung photons have been investigated by several methods including conventional magnetic electron spectrometers [25], thermoluminescence spectrometers [7, 24] and nuclear activation experiments [9, 26, 27]. With the last technique, several photonuclear reactions are induced and identified. If their cross sections are well known, the γ -spectrum can be deduced. This technique is accepted to yield results with reasonable accuracy for relativistic laser-generated plasmas.

In this paper we demonstrate for the first time that the method of nuclear activation can be used in the opposite way, in order to determine unknown giant resonance cross-section values for photonuclear reactions of radioisotopes. We characterise the spectrum of the γ -emission from the laser plasma via (γ, n) reactions in Ta combined with a thermoluminescence detector based spectrometer in terms of T_γ and absolute photon numbers. Simultaneously we measure the number of photoreactions in the radioisotope ^{129}I and from this we determine the cross section σ_γ .

4 Experimental details

The experiments were performed with the Jena multi TW laser system, which generates pulses with a maximum energy of 1 J within less than 80 fs at a repetition rate of 10 Hz and with a centre wavelength of 800 nm. The pulses are focused onto the target, producing an average intensity of up to 10^{20} W/cm^2 within a focal area of $5\ \mu\text{m}^2$. The primary target is a 2-mm-thick tantalum sheet, which acts as electron source and Bremsstrahlung converter (Fig. 1). The iodine sample (21 g of PbI_2 with 17% of ^{129}I) is placed directly behind the tantalum converter. The target is irradiated with 10 000 laser shots.

After irradiation, γ -radiation from short-lived photoreaction products in the tantalum and iodine samples is detected by two germanium detectors. Spectra are taken in successive time intervals to monitor the characteristic decay times of the radioactive nuclei. With the efficiency calibration of the detectors absolute numbers of laser-induced reactions in units per laser shot are obtained.

5 Results

Figure 2 shows the 443-keV line of the β -decay of ^{128}I into ^{128}Xe . The spectrum is collected during the mean lifetime of ^{128}I after the end of the laser irradiation. The identification of ^{128}I is verified by measuring the decay of the activity in the 443-keV line, as shown in the inset of Fig. 2. The half-life is determined to $28 \pm 5\text{ min}$, which is in good agreement with the tabulated value of 25 min [18]. From the experimental data we calculate the number of (γ, n) reactions in ^{129}I to be $N_1 = 2$ per laser shot.

Simultaneously, γ -spectra of the irradiated tantalum target were measured (Fig. 3). A (γ, n) reaction on ^{181}Ta produces ^{180}Ta , which decays with a probability of 86% by electron capture into ^{180}Hf and with 14% by β^- decay to ^{180}W . The measured decay of the activity perfectly fits the ^{180}Ta lifetime of 8.15 h. From the area of the Hf K_α lines the total number

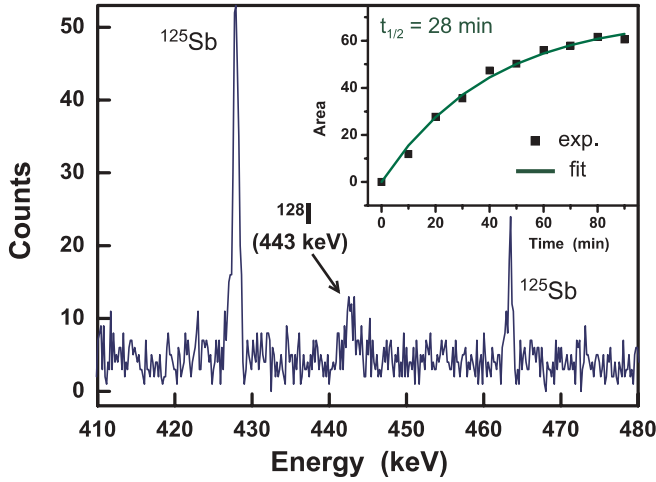


FIGURE 2 γ -spectrum of the iodine sample, integrated over the first 30 min after irradiation with laser-produced Bremsstrahlung. The characteristic 443-keV emission of ^{128}I is clearly visible between two emission lines from ^{125}Sb , which is an impurity in the sample. *Inset:* plotted is the temporal evolution of the integrated area of the 443-keV line of ^{128}I . The data were fitted with $n_1 [1 - \exp(-\ln(2t/\tau))]$, resulting in a half-life of $\tau = 28 \pm 5$ min. This agrees well with the tabulated ^{128}I lifetime of 25 min

of (γ, n) reactions in ^{181}Ta per laser shot is experimentally determined to be $N_{\text{Ta}} = 160$.

We now estimate the photoneutron cross section $\sigma_1(\gamma, n)$ of ^{129}I , see Fig. 4. The procedure is as follows: we first determine the photon spectrum $n(E)$ of the laser in terms of a temperature and an absolute yield. For this we use the additional data on the activation of the thermoluminescence spectrometers (TLDs) and of the tantalum Bremsstrahlung converter. With this photon distribution we then calculate the maximum cross-section value for the ^{129}I (γ, n) reaction.

The spectra of the laser-accelerated electrons and the Bremsstrahlung photons are described by the Boltzmann-like distribution:

$$n_{e,\gamma}(E) = \frac{n_{e,\gamma}(0)}{k_B T_{e,\gamma}} \exp\{-E/k_B T_{e,\gamma}\}. \quad (1)$$

With this assumption the TLD measurement results in a hot-electron temperature of $T_e = 1.7 \text{ MeV}/k_B$ and a photon temperature of $T_\gamma = 1.2 \text{ MeV}/k_B$. The photon yield is determined with the help of the tantalum activation: in contrast to ^{129}I the cross section $\sigma_{\text{Ta}}(\gamma, n)$ of ^{181}Ta is experimentally known to have a Lorentzian-like shape with a threshold energy at $E_{\text{th}} = 7.58 \text{ MeV}$, $\sigma_{\text{max}} = 367 \text{ mbarn}$ at $E_{\text{max}} = 12.7 \text{ MeV}$ and width $\Gamma = 5 \text{ MeV}$ [28]. Using T_γ and the total number N_{Ta} of ^{181}Ta (γ, n) reactions we calculate $n_\gamma(0)$ from

$$N_{\text{Ta}} = \frac{n_{\text{Ta}} d}{k_B T_\gamma} \int \sigma_{\text{Ta}}(\gamma, n) n_\gamma(0) \exp\{-E/k_B T_\gamma\} dE, \quad (2)$$

where n_{Ta} is the density of tantalum atoms in the target and d is the thickness of the target. From (2) it follows that $n_\gamma(0) = 7 \times 10^7$ per MeV.

In the second step we estimate the maximum value of the cross section $\sigma_1(E_{\text{max}})$ from the photon field $n_\gamma(E)$ and the total number N_I of ^{129}I (γ, n) reactions per laser shot. We again assume a Lorentzian-like shape for σ [28]. A reaction threshold of $E_{\text{th}} = 8.8 \text{ MeV}$ was taken from giant resonance

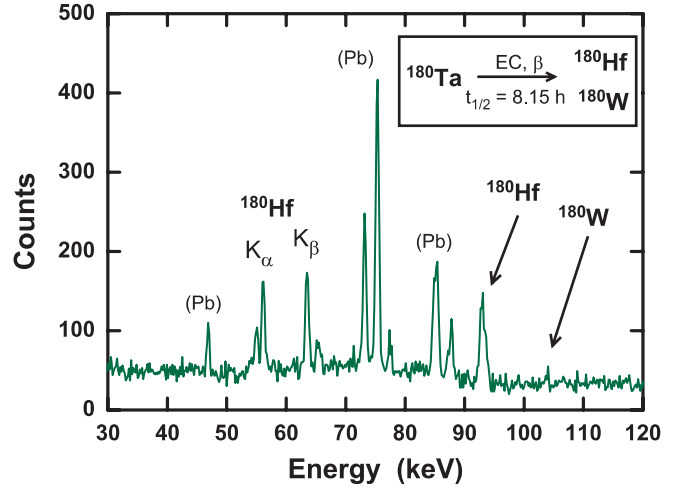


FIGURE 3 γ -spectrum of the tantalum converter plate. ^{181}Ta is transformed into ^{180}Ta by a (γ, n) reaction, which decays with a half-life of 8.15 h into ^{180}Hf and ^{180}W . The X-ray line emission of ^{180}Hf at 56 and 63 keV is clearly visible, as well as two γ -lines of ^{180}Hf and ^{180}W at 93 keV and 103 keV, respectively. The Hf K_α lines were used for the evaluation of the number of induced (γ, n) reactions (the Hf K_β line is superposed by background radiation)

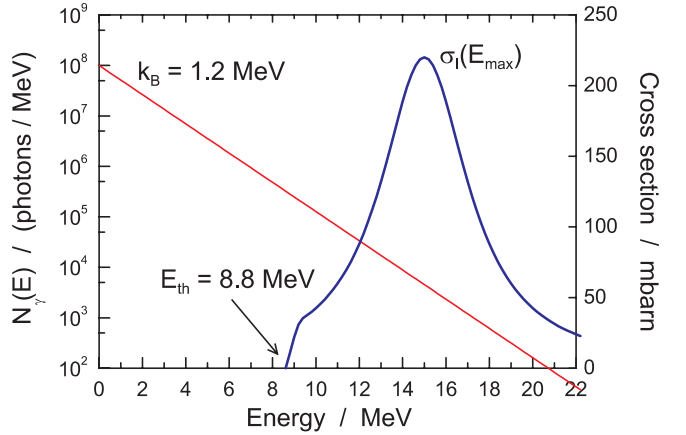


FIGURE 4 Determination of the (γ, n) cross section σ_1 of ^{129}I from the Bremsstrahlung spectrum of the laser-produced plasma (red). We assume a Lorentzian shape of σ_1 with a calculated energy threshold of 8.8 MeV and $E_{\text{max}} = 12.4 \text{ MeV}$. The analysis of the experimental data reveals a cross section $\sigma_1(E_{\text{max}}) = 220 \text{ mbarn}$

dipole models, whereas $E_{\text{max}} = 15 \text{ MeV}$ as well as $\Gamma = 5 \text{ MeV}$ were adopted from ^{127}I data [28]. Using (2) for iodine instead of tantalum and solving it for the cross-section value $\sigma_1(E_{\text{max}})$ at the giant dipole resonance we obtain $\sigma_1(E_{\text{max}}) = 220 \text{ mbarn}$. The value is accurate within a factor of two.

6 Conclusion

In conclusion, we have presented a new method to investigate photonuclear reactions of radioisotopes using laser-generated γ -radiation. In the present work, we describe the laser-induced transmutation of ^{129}I one of the most important radionuclides in the nuclear fuel cycle. However, laser-induced transmutation of radionuclides is not restricted to photoreactions since relativistic laser plasmas can also emit high-energy beams of protons, deuterons, ions and neutrons as projectiles for nuclear reactions. The future develop-

ment of the field will benefit from the currently fast evolution of high-intensity laser technology. Within a few years, compact and efficient laser systems will emerge, capable of producing intensities exceeding 10^{22} W/cm² [29] with repetition rates of 1 shot per minute and higher. These laser pulses will generate electron and photon temperatures in the range of the giant dipole resonance and therefore drastically increase the potential of the described method. These laser experiments may offer a new approach to studying material behaviour under neutral- and charged-particle irradiation without resource to nuclear reactors or particle accelerators.

ACKNOWLEDGEMENTS We would like to thank G. Noguere and A. Leprêtre from the Commissariat à l’Energie Atomique (CEA) Cadarache and Saclay respectively, and P. Rullhusen from the Institute for Reference Materials and Measurements, Geel for providing the iodine samples. We thank J. Rebizant and W. Wagner from the Institute for Transuranium Elements for help with encapsulation of the samples and the transport issues. We acknowledge collaboration with K. Ledingham’s group at the University of Strathclyde, Glasgow, who has successfully repeated the experiment with the VULCAN laser at Rutherford Appleton Laboratory, UK, J. Phys. D: Applied Physics (2003), to be published.

We would also like to thank B. Beleites, F. Ronneberger, K.U. Amthor and W. Ziegler from the FSU as well as R. Behrens from the PTB for technical support.

The work was funded by the German Science Foundation (DFG) and by the German Ministry for Education and Research (BMBF).

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