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RAPID COMMUNICATION

Laser-driven photo-transmutation of ^{129}I—a long-lived nuclear waste product

K W D Ledingham[1,6], J Magill[2], P McKenna[1], J Yang[1], J Galy[2], R Schenkel[2], J Rebizant[2], T McCanny[1], S Shimizu[1], L Robson[1], R P Singhal[3], M S Wei[4], S P D Mangles[4], P Nilson[4], K Krushelnick[4], R J Clarke[5] and P A Norreys[5]

[1] Department of Physics, University of Strathclyde, Glasgow G4 0NG, UK
[2] European Commission, Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, 76125 Karlsruhe, Germany
[3] Department of Physics and Astronomy, University of Glasgow, Glasgow G12 8QQ, UK
[4] Blackett Laboratory, Imperial College, London SW7 2BZ, UK
[5] Central Laser Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX, UK

E-mail: k.ledingham@phys.strath.ac.uk

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Abstract
Intense laser–plasma interactions produce high brightness beams of gamma rays, neutrons and ions and have the potential to deliver accelerating gradients more than 1000 times higher than conventional accelerator technology, and on a tabletop scale. This paper demonstrates one of the exciting applications of this technology, namely for transmutation studies of long-lived radioactive waste. We report the laser-driven photo-transmutation of long-lived ^{129}I with a half-life of 15.7 million years to ^{128}I with a half-life of 25 min. In addition, an integrated cross-section of 97±40 mbarns for the reaction ^{129}I(γ,n)^{128}I is determined from the measured ratio of the (γ,n) induced ^{128}I and ^{126}I activities. The potential for affordable, easy to shield, tabletop laser technology for nuclear transmutation studies is highlighted.

1. Introduction

One of the major problems of the nuclear power industry today is in the management and disposal of high-level radioactive waste. Vitrified high-level waste can be stored for about 50 years before ultimate geological disposal. However, in many countries around the world much research effort is being expended in the possibility of partitioning and transmuting radioactive waste, which can reduce it's toxicity by a factor of 100 [1–3]. Recently a number of roadmaps for nuclear waste transmutation have been produced [4, 5]. Although the most frequently discussed methods involve transmutation by bombardment with neutrons from a reactor, or more recently from a particle accelerator [6], there have been other suggested approaches to this problem, for example laser-driven high-brightness gamma generation for photo-transmutation [7]. In a recent UK report on the transmutation of nuclear waste [8] the importance of launching new ideas in radioactive waste management was emphasized and it is in this context that the present work introduces a novel method of inducing photo-transmutation reactions using bremsstrahlung produced in a target irradiated by an ultrahigh intensity laser.

Compact multi-terawatt and petawatt pulsed high-intensity laser systems have now been developed in a number of laboratories worldwide. Techniques such as chirped pulse amplification [9] have facilitated laser pulses of the order of femtoseconds or picoseconds to be temporally stretched by 3 to 4 orders of magnitude for non-destructive amplification in a chain of optical amplifiers. After amplification the laser pulses are re-compressed to deliver about 10^3–10^5 PW cm^{-2} intensity on target. At these intensities the electron quiver energy in the

[6] Also at AWE plc Aldermaston, Reading RG7 4PR, UK.

laser field is fully relativistic and leads to the generation of high brightness beams of energetic gamma rays, protons, neutrons and heavy ions [10–14].

In this communication we report on the use of the VULCAN petawatt laser to drive the photo-transmutation of long-lived ^{129}I with a half-life of 15.7 million years to ^{128}I with a half-life of 25 min. A laser-generated gamma ray is absorbed into the ^{129}I nucleus, which releases a neutron to undergo transmutation to ^{128}I. This giant dipole resonance (γ,n) reaction is observed in ^{129}I and ^{127}I and the absolute activities of the reaction products ^{128}I and ^{126}I are used for the first time to determine an integrated cross-section for ^{129}I(γ,n)^{128}I, from the known ^{127}I(γ,n)^{126}I cross-section.

2. Experimental arrangement and method

The experiment was performed on the petawatt arm of the VULCAN Nd:glass laser housed at the Rutherford Appleton Laboratory, UK. A 360 J laser pulse, of wavelength $\lambda \sim 1$ μm and duration 0.7 ps, was focused using a 1.8 m focal length off-axis parabolic mirror to a spot size of 5.5 μm diameter, to achieve an intensity I of the order of 5×10^{20} W cm^{-2}. The target was a solid piece of gold with dimensions $5 \times 5 \times 4$ mm. Irradiation of the target with intense laser light pondermotively drives electrons into the target with relativistic energies. The electron energy distribution has been previously shown to be Boltzmann-like [15] and for the p-polarized, 45° laser irradiation arrangement used in this study, kT (the product of the Boltzmann constant and the electron temperature) is (in MeV)

$$kT \sim 0.511\left[\sqrt{\left(1+\frac{I\lambda^2}{6.85\times 10^{17}}\right)}-1\right]$$

where the irradiance $I\lambda^2$ is measured in W cm^{-2}μm^2 [16]. The laser irradiance for this experiment was $\sim 5 \times 10^{20}$ W cm^{-2}μm^2 which would produce a hot electron population with an expected kT of ~6.4 MeV. These hot electrons are stopped in the high-Z gold target, generating bremsstrahlung gamma radiation with a broad energy distribution and similar kT.

In previous experiments the gamma radiation has been shown to be strongly angular dependant with the direction of beam propagation influenced by laser induced preplasma conditions [17]. In the present experiment two iodine samples were positioned along the laser and target normal directions behind the gold target as shown in figure 1. The samples were prepared from waste solution from a fuel processing facility. Iodine was present in the raw waste solution at 1.3 gl^{-1} with isotopic composition 30% ^{127}I, 70% ^{129}I plus short-lived ^{131,133}I. A 90% efficient extraction procedure produced iodine as PbI_2 compound with density 3.24 g cm^{-3}. This powder was then compacted in a 55 mm diameter Al can, which was further encapsulated in a vacuum-tight Plexiglas container. The samples contained 46.2 g and 43.3 g of iodine with isotopic composition 85% ^{129}I and 15% ^{127}I.

After single-shot laser irradiation of the target the characteristic gamma radiation from photo-induced reaction products in the gold target and the iodine samples were measured using two well-shielded intrinsic germanium detectors. The detector efficiency (35%) was calibrated using known sources of ^{152}Eu, ^{57}Co, ^{22}Na, ^{137}Cs and ^{60}Co to facilitate absolute numbers of laser induced reactions to be determined.

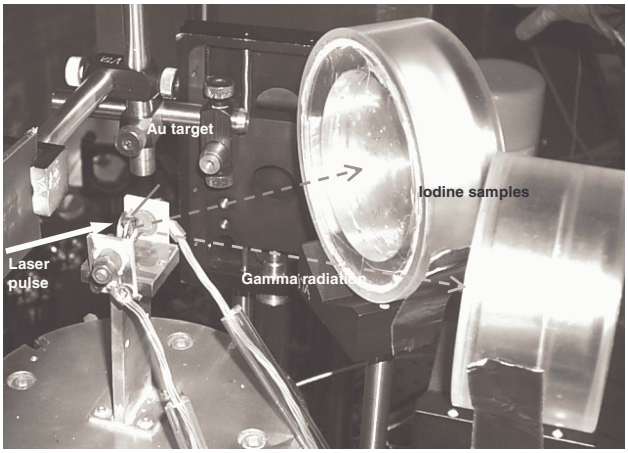

Figure 1. Photograph of the experimental arrangement. The high intensity laser pulse enters from the left of the picture and is focused onto a 4 mm thick Au target. Relativistic electrons from the resulting hot plasma are converted to high-energy bremsstrahlung in the target. Iodine samples encased in Al and Plexiglas are located along the laser and target normal axes. The gamma radiation from the target induces transmutation of the iodine samples through (γ,n) reactions.

3. Results and discussion

Figure 2 shows part of the measured gamma-ray spectrum from the laser-irradiated gold target. The peaks at 332.9 keV and 355.6 keV originate from the decaying isotope ^{196}Au and the peak at 328.4 keV from ^{194}Au, produced by ^{197}Au(γ,n)^{196}Au and ^{197}(γ,3n)^{194}Au reactions. $(6.7 \pm 0.6) \times 10^8$ and $(2.5 \pm 0.4) \times 10^6$ isotopes of ^{196}Au and ^{194}Au were generated respectively. Since the cross-sections for the (γ,n) and (γ,3n) reactions on gold are well known and are peaked at different gamma energies, the bremsstrahlung spectrum can be deduced, using a technique described in detail elsewhere [18]. Assuming a Boltzmann-like energy distribution the kT was determined to be (5.5 ± 0.7) MeV. This is in good agreement with the calculated value of 6.4 MeV for a laser irradiance of $\sim 5 \times 10^{20}$ W cm^{-2}μm^2.

The iodine samples were analysed using the germanium detectors before and after laser irradiation of the gold target. The background spectrum of figure 3 was measured for 3 hours and the principal lines observed resulting from the decay include ^{125}Sb. The background activity from the samples was of the order of 2.4×10^8 Bq. A 1 mm thick Pb filter was inserted between the iodine samples and the detector in order to reduce the detector dead-time due to the high activity of the β emission from the ^{129}I decay and the x-ray radiation at 29.8 keV, 29.4 keV and 33.6 keV and gamma radiation at 39.6 keV. Calculations have shown that the intensity of a line at 450 keV, which is the region of interest for the experiment, is attenuated about 15% by the addition of this filter.

Twenty minutes after laser irradiation the iodine samples were returned to the germanium detectors. Figure 3 shows

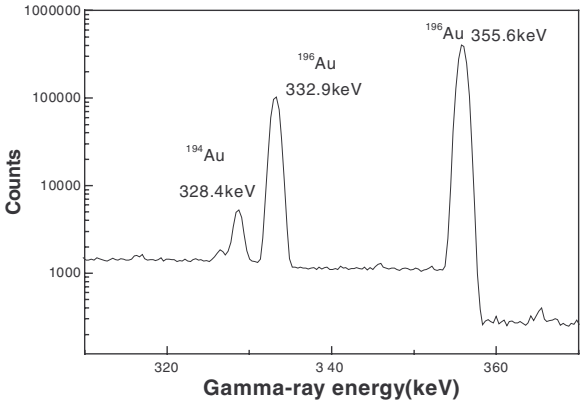

Figure 2. Gamma emission spectrum from the activated gold target after laser irradiation. ^{196}Au and ^{194}Au peaks from (γ,n) and (γ,3n) reactions on ^{197}Au are observed.

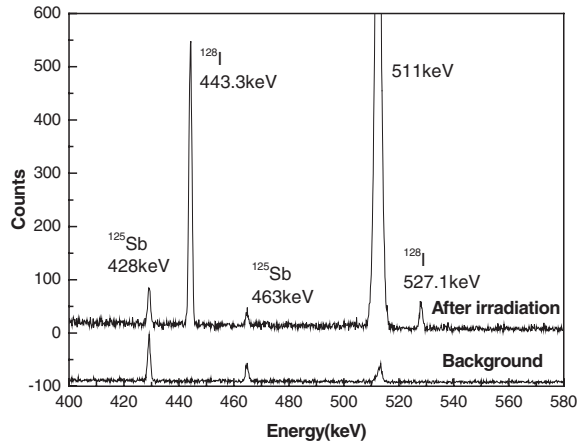

Figure 3. Gamma emission spectra from one of the iodine samples measured before (background) and after laser irradiation of the gold target. Characteristic emission lines of ^{128}I at 443.3 keV and 527.1 keV are clearly observed, alongside peaks from the decay of ^{125}Sb and a peak at ^{511}keV from positron annihilation.

clear peaks at 443.3 keV and 527.1 keV, characteristic of the decay of ^{128}I. Interestingly, both samples had similar peak strengths indicating a wide angular distribution of gamma radiation from the gold target. The decay of ^{128}I was determined by measuring the integrated area of the peaks over successive time intervals, figure 4. The half-lives of the 443.3 keV and 527.1 keV peaks were (25.8±1.0) minutes and (25.5 ± 1.5) minutes respectively, which agree well with the literature value of 25.0 minutes [19]. In addition, the ratio of the area of the 443.3 keV and 527.1 keV peaks averaged over all of the measured spectra is 10.9, in good agreement with the literature value of 10.4. An additional peak at 949.6 keV was weakly observed and is also attributed to the decay of ^{128}I. From the data we have deduced the total activity of ^{128}I produced at the time of the laser shot to be 1323 Bq. This number was determined after correction for the decay branching ratios, attenuation in the 1 mm thick Pb filter (between the sample and the detector) and self-absorption in the Pb-matrix sample. This corresponds to the production of about 2.9×10^6 nuclei of ^{128}I for the 360 J laser shot.

The iodine samples contained 15% of the isotope ^{127}I. Observed weak peaks at 388.6 keV and 666.3 keV in the

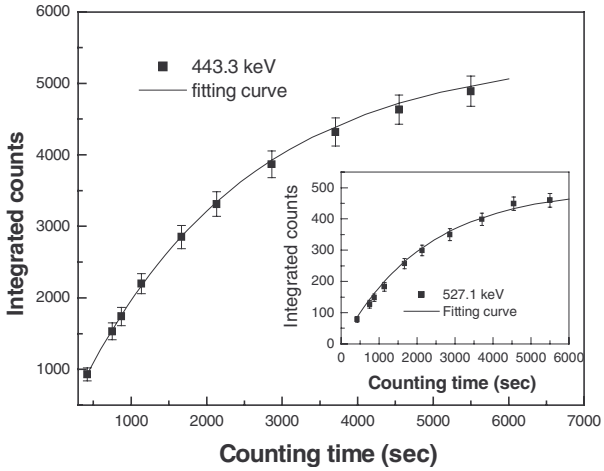

Figure 4. Integrated area of the 443.3 keV emission peak from the decay of ^{128}I measured as a function of time. The measured half-life is (25.8±1.0) minutes. Inset: The measured half-life of the 527.1 keV emission peak was determined to be (25.5±1.5) minutes. Both measurements agree well the literature value of 25 minutes for the half-life of ^{128}I.

measured gamma emission spectra are attributed to the decay of ^{126}I, produced by (γ,n) reactions on ^{127}I. A similar analysis performed on those peaks illustrates the production of 1.7×10^6 ^{126}I nuclei.

Comparison of the numbers of the ^{129}I(γ,n)^{128}I and ^{127}(γ,n)^{126}I reactions induced in the same iodine sample facilitates a ratio of the integrated cross-sections for these reactions to be determined. For a given gamma intensity Φ, the number of nuclei N_{128} of ^{128}I and N_{126} of ^{126}I produced are given by: $N_{128} = \sigma_{129}\Phi N_{129}$ and $N_{126} = \sigma_{127}\Phi N_{127}$, where N_{127} (=3.5x10^{22}) and N_{129} (=1.8x10^{23}) are the initial number of nuclei of ^{127}I and ^{129}I respectively in the sample and σ_{129} and σ_{127} are the cross-sections for the ^{129}I(γ,n)^{128}I and ^{127}I(γ,n)^{126}I reactions respectively. Assuming the Q-value and width of the cross-sections are similar, the ratio of the integrated cross-sections can be written as:

$$\frac{\sigma_{\mathrm{int}}^{^{129}I}}{\sigma_{\mathrm{int}}^{^{127}I}} = \frac{N_{128}}{N_{126}} \cdot \frac{N_{127}}{N_{129}}$$

The integrated cross-section for ^{127}I(γ,n)^{126}I is known, 309 mbarns [20], and the value for the ^{129}I(γ,n)^{128}I reaction was determined using 4 measured spectra to be 97±40 mbarns. This is in good agreement with the theoretical cross-section given in the literature as 110 mbarns [21]. Because the present technique involves a simple ratio of measured peak areas of two isotopes of the same element (irradiated under the same conditions), it is superior to methods employing a convolution of the gamma energy spectrum with an assumed cross-section shape. The main uncertainty in the determined integral cross-section resides in the measured gamma-peak areas.

4. Conclusions

We have demonstrated laser-driven (γ,n) transmutation of an important long lived fission product, ^{129}I, found extensively in radioactive waste, to ^{128}I which decays quickly with a half-life of just 25 minutes. In addition, by comparing the activities of ^{128}I and ^{126}I, and using the known ^{127}I(γ,n)^{126}I cross-section

we have determined the unknown integrated cross-section for the ^{129}I(γ,n)^{128}I reaction. We have shown that a single 360 J laser pulse focused to $\sim 5 \times 10^{20}$ W cm^{-2} can generate about 2.9×10^6 nuclei of ^{128}I. A similar experiment carried out recently on the short-pulsed tabletop laser at the University of Jena [22] at a similar laser intensity, but with pulse energies of about a joule, generated 1.5 nuclei of ^{128}I per laser shot.

A new generation of compact, high-intensity tabletop lasers is currently being designed in a number of laboratories, incorporating novel technology such as optical parametric chirped pulse amplification [23, 24]. Several of these laser systems will be geared towards applications such as medical isotope production and transmutation studies of long-lived isotopes. This paper has introduced laser-driven photo-transmutation as a feasible method for experimental research into transmutation reactions, with potential applications to medicine and nuclear waste management.

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