Potential of a laser-driven source of characteristic $\gamma$-rays for fast detection and identification of nuclear materials

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Abstract. Active interrogation of special nuclear materials (SNM) represents the most promising detection technique in combating their illicit trafficking and diverted use. Among the many interrogation approaches under investigation, the one based on photoinduced fission relies on large prompt and delayed radiation signatures. Pulsed induction (tens of ns) of photofission, in particular, enables to distinguish between prompt and delayed radiation emission, providing additional signatures to identify fissile materials. As the probing radiation, 6- to 7-MeV characteristic $\gamma$-rays produced by the $^{19}$F(p,$\alpha\gamma$)$^{16}$O reaction are of particular interest; in fact, they have energy sufficient to induce fission in SNM, but insufficient to create significant radioactivity via photoneutron activation, especially in lighter elements. In this study, the feasibility of a pulsed source of characteristic $\gamma$-rays based on the interaction of laser-accelerated protons with a fluorine-rich target is assessed. For practical applications, proton energies above 2 MeV, a bunch population of the order of $10^{14}$ particles or higher, and a laser repetition rate of the order of 10 Hz are of interest. The development of ultrahigh-power table-top lasers could greatly contribute to the deployment of this kind of interrogation source for a variety of operations.

1. Introduction
Illicit trafficking and diverted use of SNM—notably highly enriched uranium (HEU)—represent one of the main concerns for the global safety. The techniques used for the detection and identification of nuclear materials can essentially be distinguished between passive and active interrogation. While passive interrogation lies on the detection of radiation emitted through the spontaneous decay of nuclei (e.g., $\gamma$-ray detection and imaging, neutron detection), active interrogation excites nuclei in a target—by either neutrons or $\gamma$-rays—and looks for their radiation signatures (neutrons and/or $\gamma$'s) [1]. One of the most interesting features of active interrogation is represented by the fact that the strength of its response is independent of the inherent activity of the target, whereas it is proportional to the intensity of the probing radiation.

Among the many active interrogation approaches under investigation, the one based on photoinduced fission relies on large prompt and delayed radiation signatures [2]. In particular, pulsed induction of photofission (on a timescale of tens of ns) enables to distinguish between prompt and delayed radiation emission, providing additional signatures to identify fissile materials [3,4]. Moreover, the prompt response leads to an improved signal-to-background ratio in a fast detector. As the probing radiation, 6- to 7-MeV characteristic $\gamma$-rays produced by the $^{19}$F(p,$\alpha\gamma$)$^{16}$O reaction are of
particular interest; in fact, they have energy sufficient to induce fission in SNM, but insufficient to create significant radioactivity via photonuclear activation, especially in lighter elements [4,5]. In stable isotopes, for example, neutron separation energies are more likely to overcome 7 MeV at low rather than at high mass number. Besides the reduced impact in terms of measurement background and radiation protection, this feature also provides the technique with additional discrimination capability between SNM and surrounding ordinary materials. Bremsstrahlung sources, on the contrary, are inherently less selective and provide higher collateral activation.

The most promising experiments on a fast, intense source of mono-energetic $\gamma$-rays via the $^{19}$F(p,$\alpha$$\gamma$)$^{16}$O reaction have so far utilised pinch-reflex ion diodes to generate the proton beam [4,5]. Here, the feasibility of this kind of $\gamma$-source by using laser-driven proton beams [6] is comparatively assessed.

2. Phenomenology

The $^{19}$F(p,$\alpha$$\gamma$)$^{16}$O reaction has a Q-value of 8.115 MeV and proceeds via the population of excited levels of the compound nucleus $^{20}$Ne. These levels have large $\alpha$-decay widths to excited states of $^{16}$O [7]. The reaction cross section (calculated [8]) is plotted as a function of proton energy in figure 1. For proton energies below 4 MeV, transitions to the ground state and four excited states of $^{16}$O mostly occur, through the emission of five $\alpha$-groups. The second, third and fourth excited levels of $^{16}$O almost exclusively decay to the ground state directly, by the emission of $\gamma$-rays with energies of 6.129, 6.917 and 7.116 MeV, respectively. For proton energies above 4 MeV, excitation of higher levels in the residual $^{16}$O nucleus becomes more likely, which however leads—with high probability—to cascade decays through one of the 6.129-, 6.917-, or 7.116-MeV levels [7,9].

The number of photons, $N_{\gamma}$, produced over 4$\pi$ sr by $N_p$ protons impinging on a thick target with an energy distribution $f(E)$ can easily be calculated if the thick-target yield, $Y$, is known as a function of the incident proton energy:

$$N_{\gamma} = 4\pi e N_p \int_0^\infty f(E)Y(E)dE$$

(1)

where $e$ is the elementary charge and $[Y] = C^{-1}$s$^{-1}$. Systematic measurements of total and fractional thick-target $\gamma$-yields for the $^{19}$F(p,$\alpha$$\gamma$)$^{16}$O reaction only exist for proton energies up to 4.0 MeV [9, and references therein]. Results confirm that the total photon yield increases with increasing proton energy, and show how the relative intensity of the individual $\gamma$-lines depends on proton energy. The $\gamma$-emission is found essentially isotropic. In figure 2, total $\gamma$-yields have been plotted for CaF$_2$, MgF$_2$ and SF$_6$ thick targets, using the data of Micklich et al. [9]. These three fluorides are of particular interest as possible proton targets in an actual $\gamma$-source. Curves in figure 2 are generated by a sigmoidal function of the form $Y(E) = A \cdot \left(1 - (1 + \exp((E - E_0)/b_0))^{-1}\right)$, where $A$, $E_0$ and $b_0$ are parameters retrieved from the experiments and tabulated in ref. 9 (we note that the expression for the sigmoid presented there is incorrect). The highest yields have been measured at $E = 4.0$ MeV, and vary from $4.7 \times 10^7$ to $6.0 \times 10^7$ $\mu$C$^{-1}$s$^{-1}$, depending on the fluoride. Similar or even higher values are expected up to $E \approx 9$ MeV (see figure 1), for the sum of the three $\gamma$-lines of interest. Figure 2 also shows that the yield becomes considerable (e.g., within a factor of 10 from the plateau value) only for proton energies above 2 MeV. In what follows, we will retain the energy region highlighted in figure 1—approximately ranging from 2 to 11 MeV—as the most favourable one in terms of photon yield.

Sources with an intensity of at least $10^{12}$ photons per pulse can offer reasonable fluences ($> 10^9$ cm$^{-2}$) on objects at large stand-off (up to 100 m) to provide a detectable signal (prompt/delayed fission signature) [5], on the basis of state-of-the-art sensitivity of the detection techniques and typical SNM target amounts (e.g., 5 kg HEU or 1 kg $^{239}$Pu, with thick metal shielding). Assuming a proton spectrum bound within the 2÷11-MeV RoI, and $<\gamma>_p = 5.0 \times 10^7$ $\mu$C$^{-1}$s$^{-1}$ as the spectrum-averaged value of $Y$ in equation (1), $N_p = 1.0 \times 10^{16}$ is calculated for $N_{\gamma} = 1.0 \times 10^{12}$. 
3. Experimental approaches

3.1 Ion-beam diode

Ion diodes supplied by fast high-voltage generators have been used to produce intense pulsed proton beams for a variety of applications [10]. Schumer et al. [5] have been able to generate a 2-MeV, 270-kA proton beam of 75-ns duration, which resulted in a $\gamma$-ray pulse of 50-ns duration and intensity estimated in $3.1 \times 10^{11}$ photons after impinging on a polytetrafluoroethylene (Teflon) target. The ion diode they used was constituted of a hollow thin-wall cylindrical stainless-steel cathode (12 cm in diameter, connected to ground) and an annular Al anode, connected to a 2-MV pulsed power generator. The anode Al ring was closed by a polyethylene (CH$_2$) foil, while the hollow cathode was closed by a thin polycarbonate disc. By the effect of the high-voltage pulse, electrons emitted from the cathode initially bombard the annular Al anode; as the current increases to several hundred kA, electrons self-pinch onto the polyethylene foil, producing a plasma. Plasma protons are accelerated towards the polycarbonate disc, which they pass through before entering a 1-Torr air region where the beam is charge-neutralized. In this region, the beam is ballistically transported to the Teflon target, where the $\gamma$-ray emission occurs. The Teflon target does not survive the interaction with the proton beam up to a distance of 103 cm from the anode. At this distance it however shows severe damage, characterized by surface melting within a region of 50 cm in diameter. Placing the target at shorter distances would moreover be detrimental to the purpose of active interrogation, because of the intense bremsstrahlung pulse stemming from the anode. This pulse, which can provide unwanted background, has to be well separated in time from the characteristic $\gamma$-ray pulse.

![Figure 1. Cross section for the $^{19}$F(p,$\alpha$)$^{16}$O reaction, as a function of proton energy. Calculated values from the TENDL-2009 library [8] have been plotted. A high-yield region for $\gamma$-ray emission is also indicated.](image1)

![Figure 2. Total thick-target $\gamma$-ray yield (summed over the 6.129-, 6.917- and 7.116-MeV lines) for the $^{19}$F(p,$\alpha$$\gamma$)$^{16}$O reaction in three fluorides of interest, as a function of the incident proton energy. For SF$_6$, the effective proton energy through a 5-$\mu$m W foil which windowed the gas cell is used.](image2)
3.2 Laser-driven proton source

As has been widely demonstrated in the past few years, the interaction of ultrahigh-power laser pulses (intensity of the order of or higher than $10^{19}$ W cm$^{-2}$, sub-ps duration) with thin foils (thickness of $1 \div 100$ μm) is able to produce well collimated (< 10°), ultrashort (few ps) bunches of protons with energy ranging from a few MeV up to several tens of MeV (see e.g. ref. 6 for a review). In the case of metal foils, protons are mostly originated by hydrogen adsorbed onto the target surface and by hydrocarbon surface contamination, and have also been detected at lower laser intensities [11]; nevertheless, polymer foils and metal/polymer composite foils are also used. Among the possible applications of this technique, the proton beam can be directed against a secondary target, placed at a distance of a few cm from the laser target, to induce nuclear reactions. For example, (p,xn) reactions have been demonstrated in a Pb target by McKenna et al. [12].

For applications to a $\gamma$-interrogation-source, the results of Snively et al. [13] are here considered as a benchmark. Following irradiation of a 100-μm CH polymer foil by a 1 PW laser pulse (peak intensity on the target of $3 \times 10^{20}$ W cm$^{-2}$), protons with energy up to 58 MeV were generated. The energy spectrum was found exponential in shape, with a mean energy of 4 MeV. Above 10 MeV, $2 \times 10^{13}$ protons were measured. These figures are typical and have been replicated by other authors using similar experimental conditions [14,15]. From the data of Snively et al. [13] we extrapolate $N_p(E > 2$ MeV) = $1.5 \times 10^{14}$; values up to $5 \times 10^{14}$ do not seem unrealistic by making use of engineered targets.

4. Discussion

With an energy spectrum like that generated by e.g. Snively et al. [13], all protons with $E > 2$ MeV would significantly contribute to the $^{19}$F(p,$\alpha$$^\gamma$)$^{16}$O reaction. In fact, neglecting other kinds of nuclear reactions, protons with energy beyond the RoI of figure 1 will approach that region while slowing down in the target, “climbing up” the cross section curve. With $N_p = 1.5 \times 10^{14}$ and $<Y> = 5.0 \times 10^7 \mu$C$^{-1}$sr$^{-1}$, one calculates $N_{\gamma} = 1.5 \times 10^{10}$ in a single pulse, which is a value already appropriate for interrogation at close and medium distance (e.g., below 10$^4$–20 m).

The recent development of 100-TW class Ti:sapphire lasers characterised by ultrashort pulses (around 30 fs) and operation in repetition mode (10 Hz) [16] shows the potential to bring table-top lasers to emulate the proton-acceleration performances of higher energy, longer pulse-duration, single-shot laser systems taken as a reference in this paper. In connection to active interrogation, one can notice that assuming e.g. $N_p = 5.0 \times 10^{14}$ and $<Y> = 5.0 \times 10^7 \mu$C$^{-1}$sr$^{-1}$, $N_{\gamma} = 1.0 \times 10^{12}$ could be achieved in 1 s with a 20-Hz repetition rate. The constraint on the distance over which a repetitive source could find application comes from the detection threshold of the signature signal on the single pulse; if the single-pulse response is detectable, then integration of the signal over a sequence of pulses is possible. This way, values of $N_{\gamma}$ much higher than those achievable by single-shot sources could be built up in a few seconds. Furthermore, a repetitive source would provide a unique solution for the interrogation of (fast-)moving objects (e.g., means of transport) or series of objects (e.g., baggage on a conveyor belt, or containers on a train). Finally, table-top ultrahigh-power lasers with a repetition rate of the order of 10 Hz could match the features of compactness, operability and portability that an interrogation source should exhibit in situations of practical interest (e.g., inspection at airports, harbours and train stations, or mounting of the source on a mean of transport).

On the contrary, pinch-reflex ion diodes do not seem to show the same potential for development in terms of the abovementioned features. In particular, high-power generators with the characteristics described in section 3.1 are inherently single-shot systems. Repetitive operation of the $\gamma$-ray source developed by Schumer et al. [5] is further complicated by the damage or destruction of the diode foils.

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1 In particular, the (p,n) reaction is possible in $^{19}$F for proton energies above 4.23 MeV, with cross sections between 25 and 60 mb (calculated values [8]) in the energy region ranging from 4.5 to 10 MeV. The cross section rapidly decreases above 10 MeV. Neutron background induced by this reaction should be given careful consideration in experiments and applications.
and of the Teflon target. Moreover, the proton acceleration and transport scheme there applied results in energies close to 2 MeV, which are just at the edge of the high γ-yield region visible in figures 1 and 2. This makes that γ-source quite inefficient (one finds \(N_p = 1.3 \times 10^{17}\) vs. \(N_\gamma = 3.1 \times 10^{11}\); cp. calc. in section 2, \(N_p = 1.0 \times 10^{16}\) vs. \(N_\gamma = 1.0 \times 10^{12}\)). The need to work at higher energies is recognised by the authors themselves; as a matter of fact, 4-MeV, 100-kA proton beams can be delivered by state-of-the-art pulsed power generators [5,17]. Amongst other things, a smaller bunch population and a higher-energy, broader spectrum can help mitigate the thermal and collision-damage loads on the target.

5. Conclusion
Findings of recent experiments on a fast, intense source of characteristic γ-rays based on a pinch-reflex ion diode have been reviewed, in connection to possible application to SNM active interrogation, to the phenomenology of the \(^{19}\text{F}(p,\alpha\gamma)^{16}\text{O}\) reaction used for the production of the γ-radiation, and to the state of the art of laser-driven proton sources. A significant potential has been assessed for these latter in “static” and “dynamic” applications (e.g., remote inspection of containers standing in airports and harbours, or moving on means of transport), inasmuch as proton energies well above 2 MeV, a bunch population of the order of \(10^{14}\) particles or higher, and a repetition rate of the order of 10 Hz are achievable. The development of ultrahigh-power table-top lasers could greatly contribute to reach these targets and to meet the requirements in terms of deployability –including portability– of the interrogation source.

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