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Light response of YAP:Ce and $LaBr_3$:Ce scintillators to 4-30 MeV protons for applications to Telescope Proton Recoil Neutron Spectrometers

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ABSTRACT

Two thin inorganic scintillators based on YAP:Ce and $LaBr_3$:Ce crystals have been tested with protons in the 4-8 MeV energy range at the Uppsala tandem accelerator and in the 8-26 MeV energy range at the Legnaro tandem accelerator. The crystals have been calibrated *in situ* with ^{137}Cs and ^{60}Co γ-ray sources. The relative light yields of protons with respect to gammas have been measured and are here reported to be (95.6 ± 0.3) % and (79.6 ± 0.4) % for YAP:Ce and $LaBr_3$:Ce, respectively. The results open up to the development of a Telescope Proton Recoil spectrometer based on either of the two crystals as alternative to a silicon based spectrometer for applications to high neutron fluxes.

1. Introduction

In recent years there has been increasing interest in fast neutron spectroscopy for both thermonuclear fusion experiments [1-11], where neutrons are the carriers of the energy released by the fusion reactions, and spallation sources [12-16], where there is need to mimic fast atmospheric neutrons to study their effects on micro-electronics components. The development of dedicated instrumentation must match the requirements of each research domain. In particular, for diagnosing a fusion plasma, one has to measure at high resolution the details of a quasi-mono-energetic spectrum, which is peaked at 14 MeV for deuterium-tritium and at 2.5 MeV for deuterium plasmas. On the other hand, spallation sources have a white neutron spectrum extending up to hundreds or thousands of MeV (i.e. the energy of the proton accelerator). In both cases, neutron spectrometers should combine high resolution and high counting rate (MHz) spectroscopy capabilities.

Neutron spectroscopy measurements in the MeV range are however a difficult task, as they require the development of dedicated instruments. An interesting option is the *so called* Telescope Proton Recoil (TPR) spectrometer, as it can be made compact and has a high energy resolution. The TPR diagnostic principle is based on neutron-to-proton conversion via elastic scattering on hydrogen nuclei of an hydrogenated target. The scattered proton energy can be measured and related to the incoming neutron energy, provided that the recoil angle is known [17].

Concerning fusion applications, the TPR represents a valid alternative to Magnetic Proton Recoil (MPR) spectrometers in experimental arrangements where there are space limitations [18-20]. A prototype of a non-magnetic TPR spectrometer equipped with a Si detector has been developed at Uppsala University for fusion applications and simulation studies indicated that an excellent energy resolution for the 14 MeV peak of the neutron spectrum (<5%) can be achieved with such device [21]. Inorganic scintillators, based on $LaBr_3$:Ce [22-26] and YAP:Ce crystals [27-30], have been proposed as alternative to Si detectors. These crystals have been chosen for their good resolution (due to the large light yield, which is 63000 photons-per-MeV for $LaBr_3$ [22] and 20000 to 25000 photons-per-MeV for YAP [29]) combined with fast scintillation time, which is 16 ns and 27 ns, respectively, and their resilience to damage at high neutron fluxes, unlike silicon detectors. Energy resolution measurements, carried out at Uppsala Tandem Accelerator in the 4-8 MeV energy range on $LaBr_3$:Ce and YAP:Ce, indicated that a peak energy resolution better than 2% can be achieved when these crystals are irradiated with 8 MeV protons [31].

Concerning spallation sources applications, inorganic scintillators are the natural choice, since Si detectors cannot be made thick enough to stop protons at those energies (several tens of MeV). A TPR prototype based on a YAP:Ce scintillator has been tested at the ISIS neutron source of the Rutherford-Appleton Laboratory (UK), and has delivered measurements in the energy range 30-80 MeV [32]. The same scintillator has been used in coincidence with a Si detector for ΔE-E measurements, and a time coincidence windows below 10 ns was possible thanks to fast signals [33].

In this context, the YAP and $LaBr_3$ light yields from protons are crucial parameters in the design of a TPR and they are not available in the literature. Only a crude estimation can be found in Ref.34, where the proton-to-gamma relative light yield is surmised to be ≈ 3/5 for YAP crystals at energies of few tens of MeV, relative to gamma-rays of same energy. The aim of our experiment is therefore to measure the relative proton/gamma light yield of YAP and $LaBr_3$ crystals for protons in the 4-30 MeV energy range.

2. Experimental

Two thin inorganic scintillators (Fig. 1) based on $LaBr_3$:Ce (25 mm diameter, 2.5 mm thickness) and YAP:Ce (25 mm diameter, 2 mm thickness) have been tested. Since the $LaBr_3$:Ce crystal is hygroscopic, it was encapsulated and equipped with a thin Be entrance window of 127 μm thickness, resulting in a small energy loss for the incoming protons (see later). The YAP:Ce crystal is not hygroscopic and was only covered with a thin Al layer (20 μm) for the optimization of the light collection. The thickness of the two crystals was optimized to stop protons up to 26 MeV. The range of 26 MeV protons, according to SRIM calculations [35], is 2.43 mm for $LaBr_3$ and 1.81 mm for YAP crystal (Fig. 2).

Measurements have been performed on these crystals in the 4-8 MeV energy range at the Uppsala Tandem Accelerator and in the 8-26 MeV energy range at the Legnaro Tandem Accelerator (INFN Legnaro National Laboratory - LNL).

In Legnaro each of the two crystals was coupled to an eight-dynode Photo Multiplier Tube (PMTs) by Hamamatsu (model R6231-100) [36]. At the Uppsala Tandem Accelerator, on the other hand, a Silicon PhotoMultiplier (SiPM) has been used for the $LaBr_3$ crystal [37,38].

The fast signals, both from PMTs or from SiPM, were fed directly to a 1GS/s – 10 bit digitizer (CAEN DT5751), in Uppsala, and 500Ms/s -14 bit digitizer (CAEN DT5730), in Legnaro. As previously demonstrated, the use of digital acquisition allows for high rate measurements (up to few MHz) with little degradation of the energy resolution [39,40], which is among the needs of neutron measurements with TPR spectrometers. For this reason, although the counting rates in our experiment did not exceed a few kHz, we decided to adopt a measurement setup suitable for high counting rate measurements and the pulse height spectra were reconstructed by off-line and online algorithms for DT5751 and DT5730, respectively, based on pulse fitting or charge integration [39-40].

The electron equivalent energy scales (in units of MeVee, MeV electron equivalent) of the spectra were obtained by *in situ* calibrations with γ-ray ^{137}Cs and ^{60}Co sources. Examples of measured Pulse Height Spectra can be found in Ref.26. A linear relation between the measured pulse height in channels and the MeVee was found for both detectors, as presented in the calibration curve of Fig.3.

Both the Uppsala and Legnaro accelerators provided a collimated proton beam of intensity ranging from 1 to 10 nA. The experiment was performed using a Rutherford scattering configuration [41] on a gold foil target (~3μm), which was necessary to significantly reduce the proton current on the detector, that would otherwise result in counting rates well beyond the MHz range that can be coped with by the device. Moreover, the Rutherford configuration allows simultaneous measurements with different detectors at different positions. In this experiment the $LaBr_3$:Ce and YAP:Ce detectors were placed at +45° and -45° with respect to the incident direction of the proton beam, respectively. Fig. 4 is a picture of the Legnaro irradiation chamber where the experiment was performed.

Fig.1 Pictures of thin YAP:Ce (a) and $LaBr_3$:Ce (b) crystals coupled to PMTs and used in the measurements described in the text

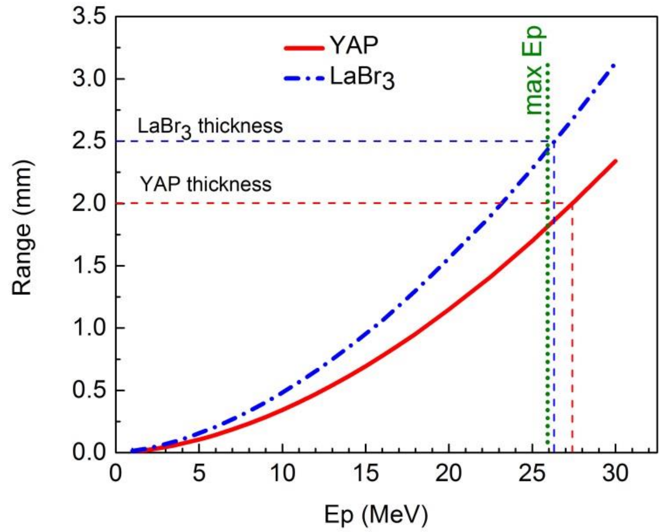

Fig. 2 Range of 1-30 MeV protons in YAP:Ce (a) and $LaBr_3$:Ce (b) crystals. Dashed lines correspond to an energy range equal to the actual thickness of the crystals. Vertical dotted line corresponds to 26 MeV, the maximum proton energy used in the experiments.

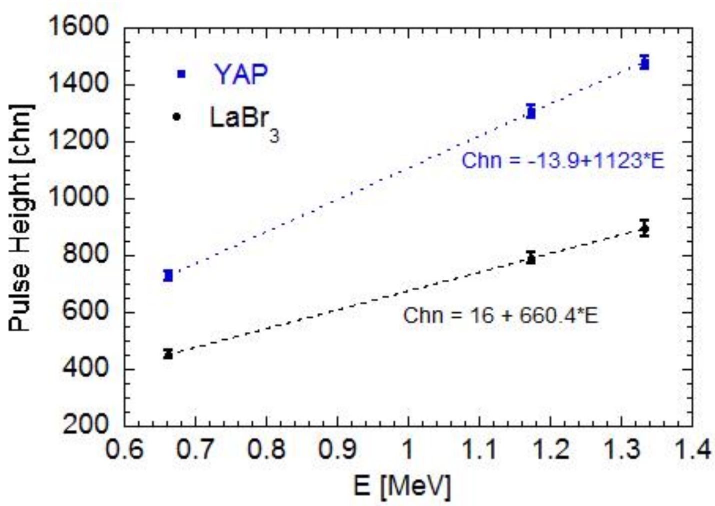

Fig. 3 Measured calibration lines for YAP:Ce and $LaBr_3$:Ce scintillators using γ-rays from ^{137}Cs and ^{60}Co sources.

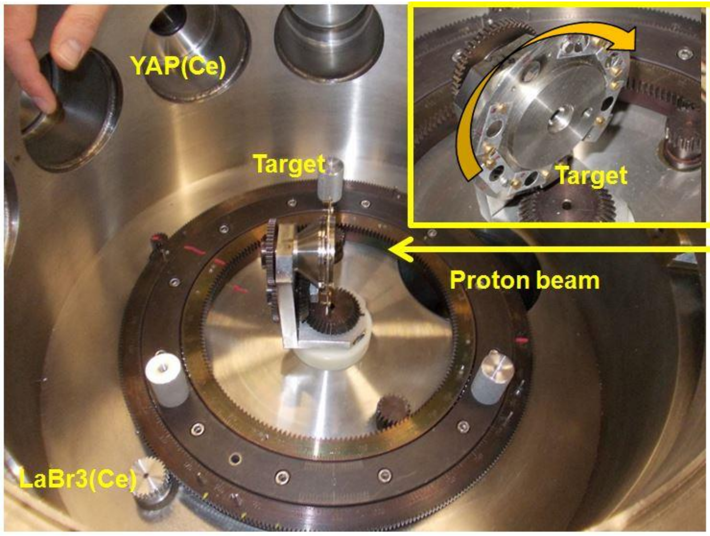

Fig. 4 Picture of the irradiation chamber at the Legnaro tandem accelerator. The proton beam direction and the positions of the target (gold foil) for the Rutherford scattering experiments are indicated, together with the positions of the two detectors. In the upper right corner the zoom shows the rotating sample holder.

3. **Data analysis and results.**

Pulse Height Spectra (PHS) have been measured and protons with full energy, from elastic scattering on gold, resulted in well-defined peaks. In order to estimate the relative proton/gamma light yield of the two scintillator crystals, the electron equivalent energy of the proton peaks in the PHS have been plotted (see e.g. Fig.7) against the nominal energy of the proton beam.

PHS are shown in Fig.5 at selected energies, i.e. 8 MeV and 20 MeV. One can clearly notice the full energy peak and, at lower energies, the background due to gamma-rays and inelastic reactions. PHS from the Uppsala experiment have been presented in Ref. 31, where measurements have given a proton energy resolution better than 2% at 8 MeV. On the other hand, in the Legnaro experiment a non-Gaussian shape was systematically found for the proton peak (as one can notice from Fig.5), which can be interpreted as due to a poor collimation of the incident proton beam, which broadens the peaks, and did not allow us to evaluate the resolution of the two detectors at the proton peak for energies above 8 MeV.

In order to take into account the energy loss in the entrance window of each detector, a correction to the nominal proton energy as known from the accelerating Voltage of the Tandems, has been introduced using SRIM calculations with 5000 ions. Fig. 6 shows the proton energy loss for the two detectors. As can be noticed, the proton energy loss in the Be window used to encapsulate the hygroscopic $LaBr_3$:Ce crystal is larger than that in the Al foil used to cover the YAP:Ce crystal. This effect is a reflexion of the different thickness of the two windows, which is somewhat off-set by the fact that Be has a lower stopping power, due to lower Z.

The resulting plot of the relative light yield of the two scintillators is shown in Fig. 7, where the pulse-height amplitudes are expressed in electron equivalent energy (MeVee) as a function of the nominal proton energy. The relative light output, or electron equivalent Energy (E_{ee}), of YAP:Ce scintillator versus the incoming proton energy (E_p) is linear (R = 0.9995) and, quite interestingly, very close to that from γ-rays (or electrons) of same energy. The best fit to data is given by

$$YAP : E_{ee} = (0.956 \pm 0.003)E_p \quad (1)$$

The relative light yield of the $LaBr_3$:Ce scintillator is lower than the YAP:Ce one but also linear (R= 0.9998) as a function of the proton energy, i.e.

$$LaBr_3 : E_{ee} = (0.796 \pm 0.004)E_p \quad (2)$$

Both results are encouraging in view of the implementation of a TPR system based on YAP and/or $LaBr_3$ inorganic scintillators as further discussed later on.

A remark on uncertainties is due at this point. The average position associated to the proton peaks has been obtained directly from the Pulse Height data and the error on the centroid, $err=\sigma/\sqrt{N}$, can be assumed to be one of several contributions to the uncertainty; here σ is the standard deviation of the peak. However, the larger contributions to the uncertainty are given (1) by the propagation of errors from the calibration, which is extrapolated from the low energy of the ^{137}Cs and ^{60}Co γ-ray lines (0.67, 1.1 and 1.3 MeV) up to 26 MeV; and (2) by systematic errors, such as the fact that the PMT gain does not return exactly to its value when the PMT is first switched off and then on again. This was necessary since, for example, the energy calibration with radioactive sources was done in air but, during the pump down of the chamber before the start of the proton experiment, the PMT had to be switched off to avoid damage. Another possible contribution to the systematic uncertainty is the fact that the temperature may not have been constant for the whole experiment, thus affecting the PMT and SiPM gains between different proton irradiations. All these contributions are difficult to evaluate *a priori*, and were therefore calculated *a posteriori* from the dispersion of the experimental points with respect to the linear fit of the data. This gave absolute errors on the data of 0.25 MeVee and 0.18 MeVee for YAP and $LaBr_3$, respectively, as well as the errors on the relative light yields of eq. 1 and 2. Note that the error bars are not shown in Fig. 7 because they are of the same size as the bullets.

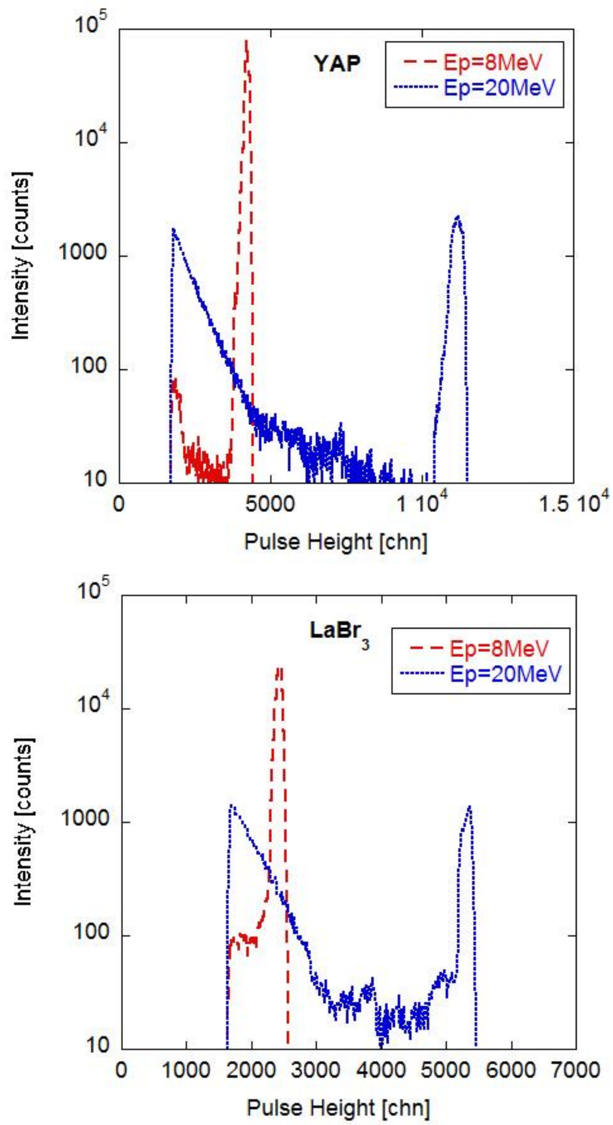

Fig.5 Measured Pulse Height Spectra for YAP (top) and $LaBr_3$ (bottom) at selected proton energies (E_p = 8 MeV and E_p = 20 MeV).

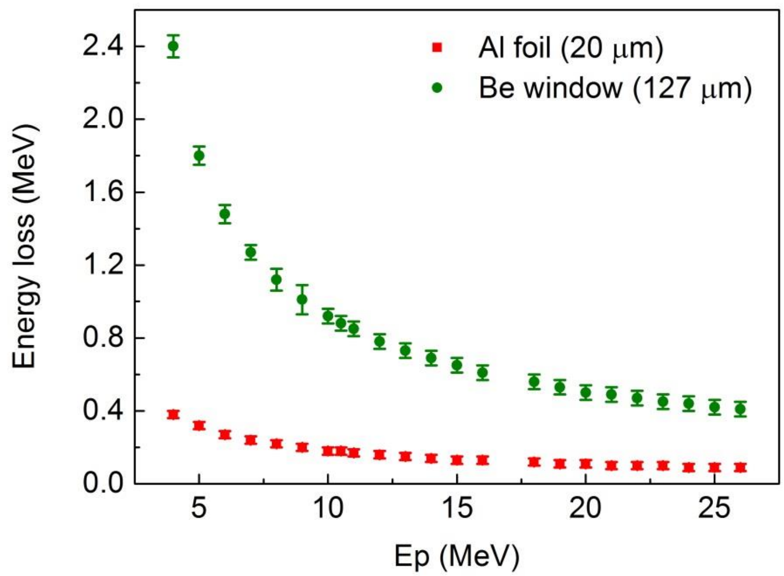

Fig. 6 Energy loss for 4-26 MeV protons in the Al foil and Be windows of the YAP and LaBr3 detectors, as calculated with SRIM [30].

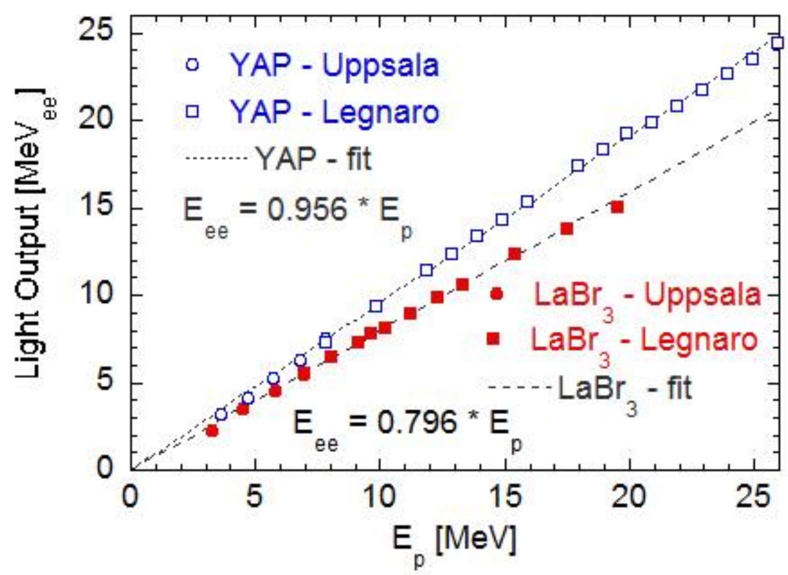

Fig. 7 Measured Light Output for YAP:Ce and $LaBr_3$ crystals, expressed in MeVee, as a function of the nominal proton energy. A fit to both datasets with a line from the origin is also shown.

4. Discussion

In its traditional implementation a TPR spectrometer is made of annular silicon detectors which are placed along a ring at a fixed angle with respect to the incoming neutron beam. Silicon is typically chosen as the material for the proton recoil spectrometer since, being a semiconductor, it provides excellent energy resolution properties (typically, less than 1% at MeV proton energies), often without any dedicated optimization effort. On the other hand, as already pointed out, there are also limitations to the implementation of silicon TPR systems for application to both fusion and spallation sources. These are due to the poor resilience of Silicon detectors to damage at high neutron fluxes and the limited thickness at which these devices are fabricated, which prevents their application in a TPR to neutron energies greater than, say, 30 MeV. An important parameter that needs to be considered is, however, the signal to background ratio. Neutron measurements are always carried out in a significant gamma-ray background (of the same order of magnitude as the neutron signal) that, both in fusion and spallation sources applications, comes from the interaction of neutrons with materials in the experimental environment. Silicon detectors, given their limited thickness, have little sensitivity to the gamma-ray background and offer, at the same time, an efficiency approaching 100% to MeV range protons. Both features provide an excellent signal to background ratio to a silicon TPR and have been so far thought to overcome the limitations mentioned above. The main objection against the implementation of a TPR based on inorganic crystals is actually based on the signal-to-background ratio that can be achieved with such device, which in turn depends on the relative light yield to protons with respect to gamma-rays of same energy. In fact, the peak resulting from the interaction of recoil protons with the device must be sufficiently separated from the gamma-ray background generated by neutron interactions with materials of the experimental environment, which typically has a spectrum that does not exceed E_{ee} = 10 MeV. This means that, for example, a gamma-to-proton relative light of, at least, 0.7 would be desired to completely separate the 14 MeV neutron peak from the background in the measured energy spectrum for fusion applications. Our experimental results on the relative light output of YAP and $LaBr_3$, unlike the more conservative estimation available in the literature [34], show that such requirement is met, thus opening up to the development of a detailed TPR design based on inorganic crystal for fusion applications, which is presently ongoing and will be reported in forthcoming publications. Concerning the use of a YAP or $LaBr_3$ based TPR for measurements of the neutron spectrum from spallation sources over a broad energy range up to 100 MeV, there is here need to qualify the linearity of the response of such device to proton detection. In other words, we must determine the upper value at which there is still a linear relationship between proton energy and equivalent electron energy. In this paper we have shown that a linear relation holds up to E_p = 25 MeV, which is already of practical interest for applications. Based on our data, however, an even higher energy limit is plausible and will be investigated in future measurements.

5. Conclusions

Light output measurements have been performed with thin YAP:Ce and $LaBr_3$:Ce scintillators using protons at energies between 4 and 26 MeV. Both scintillators show a linear response in this energy range. In particular, the relative proton/gamma light yield of the YAP:Ce crystal was measured to be (95.6 ± 0.3) %, which is higher than the measured value for the $LaBr_3$:Ce crystal, which was found to be (79.6 ± 0.4) %. Both crystals are thus good candidate components for a TPR spectrometer based on inorganic crystals in fusion and spallation source applications, given their large light yield and fast scintillation constants.

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