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Measurements of $\gamma$-ray emission induced by protons on fluorine and lithium

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Abstract

We measured the $\gamma$-ray yields of the reactions $^{19}$F(p,$p'\gamma$)$^{19}$F ($E_{\gamma} = 0.110, 0.197, 1.24, 1.35, 1.36$ MeV), $^{19}$F(p,$\alpha\gamma$)$^{16}$O ($E_{\gamma} = 6.13, 6.92, 7.12$ MeV), $^7$Li(p,$n\gamma$)$^7$Be ($E_{\gamma} = 429$ keV) and $^7$Li(p,$p'\gamma$)$^7$Li ($E_{\gamma} = 478$ keV) for proton energies from 3.0 to 5.7 MeV using a 50 $\mu$g/cm$^2$ LiF target evaporated on a self-supporting thin C film. The $\gamma$-rays were detected by a 38% relative efficiency Ge detector placed at an angle of 135$^\circ$ with respect to the beam direction. Absolute $\gamma$-ray differential cross-sections were obtained for all the listed reactions with an overall uncertainty of $\pm15\%$.

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1. Introduction

Proton induced $\gamma$-ray emission (PIGE) is commonly used for the analysis of light elements, typically as a complementary technique to proton induced X-ray emission (PIXE). In particular, the analysis of fluorine is based on the detection of 110 and 197 keV $\gamma$-radiation from the $^{19}$F(p,$p'\gamma$)$^{19}$F reaction [1], whereas for the lithium the 478 $\gamma$-ray from the $^7$Li(p,$p'\gamma$)$^7$Li reaction is the most frequently used ($^7$Li is the most abundant lithium isotope, 92.47%) [2]. Accurate values of the cross-sections for prompt $\gamma$-ray emission are needed for quantitative PIGE analysis not relying on the use of reference standards [3]. Some papers have been published, even recently, concerning this topic, typically for proton energies lower than 3 MeV [4–8], whereas only a few deal with higher energies [6,7], anyway no higher than 4.5 MeV.

Apart from the aforementioned reactions, it is important to know the excitation functions for the emission of higher energy $\gamma$-rays which can contribute negatively on a measurement, as in the case of PIXE analysis of airborne particulate matter collected on Teflon filters [9], where the fluorine content of Teflon gives rise in the PIXE spectra to a strong Compton $\gamma$-ray background – due to the emission of 6–7 MeV $\gamma$-rays from the $^{19}$F(p,$\alpha\gamma$)$^{16}$O reaction – which worsens PIXE detection limits for medium-high $Z$ elements.

In order to provide basic data for practical applications of PIGE in ion beam analysis, this paper deals with the measurements of differential cross-sections for $\gamma$ reactions on $^7$Li and $^{19}$F – namely $^{19}$F(p,$p'\gamma$)$^{19}$F ($E_{\gamma} = 0.110, 0.197, 1.24, 1.35, 1.36$ MeV), $^{19}$F(p,$\alpha\gamma$)$^{16}$O ($E_{\gamma} = 6.13, 6.92, 7.12$ MeV), $^7$Li(p,$n\gamma$)$^7$Be ($E_{\gamma} = 429$ keV) and $^7$Li(p,$p'\gamma$)$^7$Li
to dimensions of 3.0 mm² by two sets of rectangular slits located at 0.7 m and 2.7 m upstream from the target. The target consisted in a 49 g/cm² Au layer, further coated with a 19 µg/cm² C target. In order to obtain the excitation functions and cross-sections, we normalized the results by the Rutherford elastic backscattering of protons from Au, adopting a procedure [4] not relying on the knowledge of the absolute number of incident protons (see below).

The γ radiation was detected by a 61.0 mm × 61.5 mm Ge detector connected to the vacuum of the target chamber, placed at an angle of 135° to the beam axis, 207.5 mm distant from the target and subtending an angle of about 17°. The nominal efficiency and resolution of the detector are 38% and 2.0 keV FWHM energy resolution (particle detector solid angle), ±10% (γ detector absolute efficiency). Adding linearly these contributions to the statistical errors (less than ± 2%), a ±15% conservative estimate for the uncertainty of the various differential cross-sections is obtained.

The measured differential cross-sections are presented in Figs. 1–3; in the energy range of the present experiment the cross-sections are dominated by broad resonances and, in the case of lithium, show very regular trends. In general, our results agree well in shape with the other data available in the literature (always presented in graphical form). In particular, the cross-section values for both 429 and 478 keV γ-rays from lithium (Fig. 1) are in good agreement with the literature values.

The contributions to the error in the absolute values of the differential cross-sections are: ±1.0% (Rutherford cross-section on Au), ±1.0% (area density ratio), ±1.5% (particle detector solid angle), ±10% (γ detector absolute efficiency). Adding linearly these contributions to the statistical errors (less than ± 2%), a ±15% conservative estimate for the uncertainty of the various differential cross-sections is obtained.

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\( (E_x = 478 \text{ keV}) \) – in the proton energy range 3.0–5.7 MeV at \( \theta_{\text{lab}} = 135° \).

2. Experimental

The experimental work was conducted at the 5 MV Tandetron accelerator at the CMAM in Madrid [10]. The measurements were performed at proton energies from 3.0 to 5.7 MeV with a 25 keV step; the uncertainty on the bombarding energy is better than ±0.1% [11]. The proton beam entered the target chamber after being collimated to dimensions of 3.0 × 3.0 mm² by two sets of rectangular slits located at 0.7 m and 2.7 m upstream from the target. The target consisted in a 49 g/cm² Au layer, further coated with a 19 µg/cm² C layer. In order to obtain the excitation functions and cross-sections, we normalized the results by the Rutherford elastic backscattering of protons from Au, adopting a procedure [4] not relying on the knowledge of the absolute number of incident protons (see below).

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The elastic scattered protons were measured by a silicon surface barrier detector (50 mm² area, 500 µm thickness and 12 keV FWHM energy resolution) placed at a backward angle of 150°; the subtended solid angle was 2.51 ± 0.04 msr, while the spread in the scattering angle due to beam size on the target and detector finite aperture was 1.5°.

During the measurements proton beam currents were in the range 20–40 nA, depending on beam energy, in order to keep the count rate low enough to reduce pile-up effects. Dead time corrections were always less than 15% for the γ detector, whereas they were negligible for the particle detector (<0.5%). Each measurement was allowed to continue until obtaining at least 3000 counts in the elastic scattering peak of protons on Au.

3. Data reduction and results

The various absolute γ-ray cross-section values \( \sigma_x(E_{\text{LiF}}, \theta) \) at the energy \( E_{\text{LiF}} \) and detection angle \( \theta \) were deduced from the following equation:

\[
\sigma_x(E_{\text{LiF}}, \theta) = \sigma_{R,\text{Au}}(E_{\text{Au}}, \phi) \cdot \frac{A_x}{A_{\text{Au}}} \cdot \frac{\Delta \Omega_p}{\Delta \Omega_{\gamma,\text{Li}}} \cdot r,
\]

where \( \sigma_{R,\text{Au}}(E_{\text{Au}}, \phi) \) is the proton Rutherford cross-section from Au at the energy \( E_{\text{Au}} \) and scattering angle \( \phi \) (in the energy region of the present experiment the backscattering of protons on Au nuclei is purely Coulomb and the electron screening effect is negligible); \( A_x \) is the area of the detected γ-ray peak; \( A_{\text{Au}} \) is the area of the proton elastic scattering peak on gold obtained from a simultaneous backscattering spectrum; \( \Delta \Omega_p \) is the solid angle of the particle detector; \( \Delta \Omega_{\gamma,\text{Li}} \) and \( \epsilon_\gamma \) are the solid angle and the efficiency of the γ detector, respectively; \( r \) is the ratio between the area density of Au and \(^{12}\text{C}(\text{LiF}) \) (or \(^{7}\text{Li}(\text{LiF}) \)) atoms in the target. Mean proton energies \( E_{\text{Au}} \) and \( E_{\text{LiF}} \) in the target layers were calculated taking into account the appropriate energy loss in Au and LiF film.

The product of the γ detector solid angle and efficiency – the absolute efficiency – was calculated by geometric modelling of the detector, assuming that all the relevant data provided by the manufacturer (position and dimension of the Ge crystal, thickness of the cap) are correct. The resulting detector efficiency was verified by comparison with the yields of several γ-ray lines from \(^{133}\text{Ba} \) and \(^{22}\text{Na} \) radioactive sources (energies between 81 keV and 1.27 MeV) placed at the target position, and the results agreed within 10%.

The area density ratio \( r \) has been obtained by 1.8 MeV alpha particle RBS spectra of the sample. The target stability was verified by repeating these measurements several times during the runs and no changes were observed. Also, reproducibility checks at the same proton energies were performed periodically and the results were always in agreement by better than 3%.

The contributions to the error in the absolute values of the differential cross-sections are: ±1.0% (Rutherford cross-section on Au), ±1.0% (area density ratio), ±1.5% (particle detector solid angle), ±10% (γ detector absolute efficiency). Adding linearly these contributions to the statistical errors (less than ± 2%), a ±15% conservative estimate for the uncertainty of the various differential cross-sections is obtained.

The measured differential cross-sections are presented in Figs. 1–3; in the energy range of the present experiment the cross-sections are dominated by broad resonances and, in the case of lithium, show very regular trends. In general, our results agree well in shape with the other data available in the literature (always presented in graphical form). In particular, the cross-section values for both 429 and 478 keV γ-rays from lithium (Fig. 1) are in good agreement with the literature values.

Fig. 1. Differential cross-sections for the production of 429 and 478 keV γ-rays from the reactions \(^{7}\text{Li}(p,p\gamma)^{7}\text{Be} \) and \(^{7}\text{Li}(p,p\gamma)^{7}\text{Li} \) respectively (\( \theta_{\text{lab}} = 135° \)).
with those measured by Boni et al. [6] (quoted uncertainty 10%). On the contrary, comparing our data on $^{19}$F(p,p'c)$^{19}$F (Fig. 2) with those again from [6], our results for the 110 keV line are ~25% lower than theirs while for the 197 keV line they are ~35% lower; these discrepancies are anyway very similar to those reported by Jesus et al. [4] in a different energy region, thus hinting to some systematic error in these data from [6]. Comparing our results with those from Ranken et al. [7] (quoted uncertainty 10–15%), we find that for both the 110 and 197 keV line they are in good agreement with theirs within the quoted experimental uncertainties.

The differential cross-section for the high energy γ-rays (6.1, 6.9, 7.1 MeV) from the $^{19}$F(p,xγ)$^{16}$O reaction (Fig. 3) was obtained for the whole group, not for the individual peaks, setting a window in the energy spectrum of the γ-radiation between 5 and 7.2 MeV and calculating the mean γ detector absolute efficiency over this energy range. Our data are similar in shape to the relative yields given by Willard et al. [12] and the energies of the resonances agree with the values listed by Tilley et al. [13]. However, the measured differential cross-section is 30% lower than that from [7], and this might be due to an overestimate of the detector efficiency or to the cross-section normalization procedure adopted in [7] as well.

The limited extension of this paper makes it impossible to insert the present cross-section values in tabular form for practical use in PIGE analysis; however, the data are available from the authors upon request.

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