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Measurement of gamma-ray production cross sections in Li and F induced by protons from 810 to 3700 keV

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ABSTRACT

Differential cross sections for the reaction channels $^{7}Li(p,n\gamma_{1\text{-}0})^{7}Be$ (Eγ = 429 keV), $^{7}Li(p,p\gamma_{1\text{-}0})$ (Eγ = 478 keV), $^{19}F(p,p\gamma_{1\text{-}0})^{19}F$ (Eγ = 110 keV), $^{19}F(p,p\gamma_{2\text{-}0})^{19}F$ (Eγ = 197 keV) have been measured in the proton energy range 810–3700 keV, with an energy step of 10 keV. The reactions channels $^{19}F(p,p\gamma_{3\text{-}1})^{19}F$ (Eγ = 1236 keV), $^{19}F(p,p\gamma_{4\text{-}1})^{19}F$ (Eγ = 1349 keV), $^{19}F(p,p\gamma_{5\text{-}2})^{19}F$ (Eγ = 1357 keV) have also been investigated in the proton energy range 1915–3225 keV. The experimental set-up, the uncertainty budget, the comparison with other data and the progress assured by new data in the analytical use of standard-less Particle Induced Gamma-ray Emission (PIGE) are discussed.

1. Introduction

Particle Induced Gamma-ray Emission (PIGE) is an IBA (Ion Beam Analysis) technique that uses MeV ions (to a large extent protons) to probe samples and characterize their atomic/isotopic content by detecting the characteristic prompt gamma-rays produced by the interaction of the ion with the nuclei in the sample. Due to much lower attenuation of gamma-rays with respect to other reaction products and to the convenient spacing of gamma-ray lines, this technique has a high potential in the multi-elemental detection of low Z elements (Li to P). The relevance of PIGE has grown together with the development of gamma-ray detectors, the major step ahead being produced by the use of the high resolution of Ge detectors, from the pioneering works of the 1960s [1–5], to the major breakthroughs of the 1980's. A methodology has been developed for both PIGE bulk [6,7] and thin target analysis [8–10] and a database of cross sections and thick target yields has begun to be constructed [11–16]. As concerns quantitation, the comparison with a standard continues to be used today [17,18] as a fast procedure, the accuracy of which depends on the similarity of the standard and the unknown sample. In alternative, a standard-less quantitative analysis protocol for bulk samples, was developed and is being constantly improved to allow the quantitation [19–21] through an iterative calculation that takes into account both the gamma-ray production cross sections and the ions energy loss in the sample. The perspectives offered by a standard-less method and therefore the analytical potential of PIGE has been greatly improved as a result of an International Atomic Energy Agency (IAEA) coordinated research project (CRP) aimed at the measurement and validation of cross section in small energy steps. This has recently produced three times more cross-section data than available before, regarding Li, F, Na, Mg, Al, Si. This work is part of that CRP and concerns Li and F, with particular attention in the extension of Li cross sections to high energies, to assure detectability deeper from the surface, an important issue in the study of nuclear-fusion related materials. For Li and F four reaction channels have normally been considered for analytic purposes: $^{7}Li(p,n\gamma_{1\text{-}0})^{7}Be$ (Eγ = 429KeV), $^{7}Li(p,p\gamma_{1\text{-}0})$ (Eγ = 478 keV), $^{19}F(p,p\gamma_{1\text{-}0})^{19}F$ (Eγ = 110 keV), $^{19}F(p,p\gamma_{2\text{-}0})^{19}F$ (Eγ = 197 keV). The reactions channels $^{19}F(p,p\gamma_{3\text{-}1})^{19}F$ (Eγ = 1236 keV), $^{19}F(p,p\gamma_{4\text{-}1})^{19}F$ (Eγ = 1349 keV), $^{19}F(p,p\gamma_{5\text{-}2})^{19}F$ (Eγ = 1357 keV) have also been investigated, the last two being analyzed together since the two photons cannot be separated in the detector.

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2. Experimental set-up

PIGE from LiF targets (of natural isotopic composition) has been studied at the Centro de Micro Análisis de Materiales (CMAM) of the Universidad Autónoma de Madrid, in 10 keV steps (or lower over sharp resonances), from 810 to 3700 keV. A few widely spaced points have been measured to check matching with existing higher energy points. Contrary to most of the other cases in the CRP, our cross sections have been directly extracted from measured quantities: yields, target thickness, collected charge, detector absolute efficiency and not relative to the RBS cross section on a heavy nucleus (Ag or Au). The accelerator is the high current Tandetron of 5MV by HVEE (High Voltage Engineering Europe) using the coaxial Cockcroft-Walton power supply system and equipped with a Generating Volt meter (GVM) for the terminal voltage (TV) control. Its design guarantees a more reliable operation, low ripple and high stability and accuracy of the TV. TV calibration has been regularly repeated using the resonances of the reaction $27Al(p,\gamma)^{28}Si$ at 991.86, 1317.14 and 1799.75 keV and of the reaction $14\,N(p,p'\gamma)^{14}N$ at 3903 and 3996 keV. The TV calibration coefficient (1.01875) is determined with an error better than 0.7 parts per mil that correspond to a maximum error of 2.5 keV at 3700 keV proton energy.

The reactions are produced inside a 25 cm diameter vacuum chamber. Given that the proton energy is always below 5 MeV and the current limited to a few tens nA for a power to be dissipated not exceeding 1 W, our tantalum Faraday cup (FC) with secondary electron suppression is adequate to measure the number of incident protons with an error of ±2.5%, essentially due to the uncertainty in the secondary electrons suppression and not to the charge integration system. The secondary electron energy is of the order of a few ten eV and their flux is proportional to the cosine of the emission angle with respect to the surface normal. Their escape increases the positive current in the circuit and generates a systematic overestimation of the charge. Secondary emission is reduced by assuring that the cup diameter/length ratio is the lowest possible (in our case it is only 0.75 due to the compact reaction chamber design) and by a suppression ring in front of the FC maintained at -180 V. The gamma detector is a 6 cm × 6 cm reverse electrode germanium detector (REGe), sitting at 135 degrees from the beam direction and having a polymer entrance window. The most relevant feature of the detector for the measurements of cross sections is the absolute efficiency, which is energy dependent. If we consider a radioactive source, this is defined as the inverse of the ratio between the number of gamma-rays emitted in the entire solid angle and the number of gamma-rays detected at full energy. This has been measured in our case by a set of calibrated sources: ^{133}Ba (activity error = 1.3%), ^{152}Eu (2.0%), ^{137}Cs (2.1%) and ^{60}Co (1.4%), covering energy range from 53.1622 to 1408.013 keV. The absolute efficiency curve is given in Fig. 1. It has to be remarked that, due to the adoption of a polymer window the efficiency of our detector decreases continuously from the minimum energy contrary to other systems where the efficiency curve has a maximum at the 121.7817 keV of ^{152E}u. The points are fitted with a polynomial function and the error of the interpolated efficiency is estimated at ±1.9% at the low energies and ±4.6% at the higher energies. The high absolute efficiency allowed us to operate with low currents (max 30 nA, max deadtime 0.6% in all measurements), still being able to reach rapidly a high statistics in each of the four main gamma-ray peaks (average collection time 400 s).

Two lithium fluoride targets, of natural isotopic composition, produced by INFN-Legnaro (Italy), were used for our experiment: first a sandwich of Au/LiF/C (34.4 μg/cm^2 ^{19}F + 11.7 μg/cm^2 ^7Li) and then a sandwich of LiF/Ag/C (17.4 μg/cm^2 ^{19}F + 5.9 μg/cm^2 ^7Li).

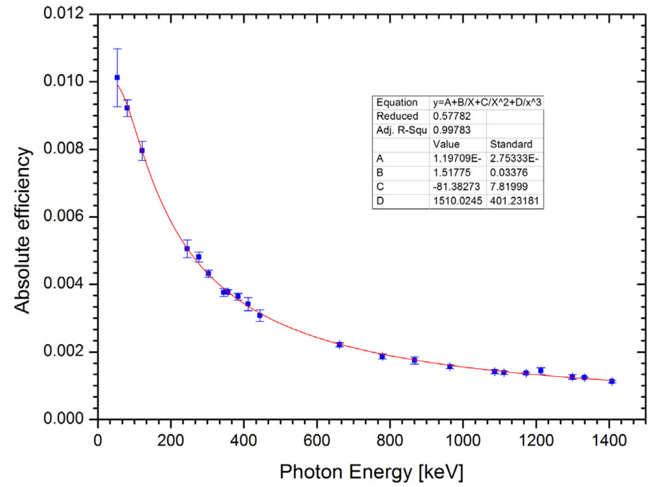

Fig. 1. The absolute detector efficiency measured with calibrated ^{133}Ba, ^{152}Eu, ^{137}Cs and ^{60}Co gamma-ray sources.

Fluorine was measured by alpha particle EBS with an error on the thickness of ±3% and the ^7Li content was calculated assuming stoichiometric ratio to ^{19}F. The energy loss in the gold layer (19.1 μg/cm^2) of the Au/LiF/C target is 1.3 keV for protons of 850 keV and goes down to 0.6 keV at 3700 keV. The shift was not corrected for the plots. Preamplifier signals have been processed by an ORTEC 572A amplifier and a Fast Comtec 7072 dual analog to digital converter. Spectra have been collected and analyzed with a Fast Comtec MP4 multi-parameter system. The fitting error for the ^{19}F peaks at 110, 197, 1233, 1349 + 1357 keV is respectively ±1.5%, ±1.2%, ±4% and ±4%. The fitting error for the ^7Li peaks at 429 and 478 keV is respectively 4% and 1%.

3. Differential cross sections

The differential cross sections for the reaction channels above mentioned are given by:

$$\frac{d\sigma_\gamma(E_0,\theta)}{d\Omega} = \frac{Y_\gamma(E_0,\theta)}{N_p N_T \varepsilon_{abs}(E_\gamma)\cdot 4\pi} \qquad N_p = \frac{Q}{e} \qquad N_T = \frac{N_0 \rho dx}{A} \qquad (1)$$

where E_0 is the incoming proton energy, N_p is the number of incident protons, Q is the total collected charge, N_T is the number of target atoms, $\rho \cdot dx$ is the target areal density, N_0 is the Avogadro number, e is the unit charge, $\varepsilon_{abs}(E\gamma)$ is the detector absolute efficiency and $Y\gamma\ (E_0,\theta)$ is the photon yield. All quantities on the right side are measured and the cross section is directly extracted from Eq. (1). Their error is obtained by adding quadratically the errors of each measured quantity, given in Section 2.

The results for the two low energy ^{19}F gamma-ray emissions are reported in Fig. 2. Our data are compared to those of references [8,22–26]. The results for the three high energy ^{19}F gamma-ray emissions are reported in Fig. 3. Our data are compared to those of references [22,24]. In these cases our data analysis has been performed only from 1915 to 3225 keV. The results for the two gamma-ray emissions from ^7Li are reported in Fig. 4 for the reaction $^7Li(p,n\gamma_{1\text{-}0})^7Be$ (Eγ = 429KeV), that has been measured from threshold to 3700 keV and in Fig. 5 for the reaction $^7Li(p,p'\gamma_{1\text{-}0})$ (Eγ = 478 keV), measured from 810 to 3700 keV. Our data are compared to those of references [8,19,22,27,28]. In both cases sparse points up to 5000 keV have been measured, as in the case of the two low energy gamma emissions of ^{19}F.

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