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Indirect method to measure differential cross-sections for nuclear reaction analysis

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ABSTRACT

Accurate knowledge of differential cross-sections is essential for studying concentrations of light elements in heavy substrates by nuclear reaction analysis (NRA). In this paper we present an indirect method of determining excitation functions of NRA taking the nuclear reaction $^{19}F(p,\alpha_0)^{16}O$ as an example. The methodology is demonstrated in detail for a backscattering angle of 150° using a bulk CaF_2 sample and proton beam energies of 1335, 1535, 1735 and 1935 keV. The differential cross section is directly extracted from the yield of backscattered reaction particles for an energy range of 800–1900 keV. To validate the procedure, a further fluorine-containing sample (NaF) was measured for comparison. The method was additionally applied to backscattering angles of 90° and 170°. The results obtained were compared with known data found in literature and a good agreement is observed. The approach presented here offers the possibility for measuring the excitation functions in a simple manner for individual measurement configurations. The method can be applied if the detected reaction particles have larger energies than particles that are elastically backscattered from the heavy matrix and if the cross section does not exhibit a fine structure with a width of the resonances which is below the energy uncertainty caused by detector resolution and energy straggling.

1. Introduction

Modern ion beam analysis (IBA) comprises general concepts that use all possible scattering and reaction processes between accelerated ions with moderate kinetic energies of about 0.5–100 MeV and target atoms to determine even small elemental concentrations [1]. One possibility to measure light elements in a compound solid is offered by nuclear reaction analysis (NRA) [2]. NRA is an important complement to Rutherford backscattering spectrometry (RBS), as RBS is a preferred method to measure heavy elements in a compound substrate (see e.g., [3]). NRA can be used for obtaining concentration distributions of atomic species close to the surface and over depths up to few micrometres in solid materials, provided that a suitable nuclear reaction exists. Detection does not depend on electron configuration and binding state of the element of interest.

A general key parameter for application of IBA-techniques is the differential reaction cross-section which indicates the probability of the occurrence of the corresponding reaction per solid angle range. However, the differential reaction cross-section does not only depend on the energy of the incident ion, it also depends on the detection angle of the experimental setup. The correct knowledge of the differential cross-section as a function of the ion energy (i.e., the so called excitation function) for the utilised experimental condition is crucial for the use of any IBA-technique to obtain reliable concentration values. The excitation functions for many nuclear reactions were measured and catalogued in the past and are freely available e.g., in the ion beam analysis data library (IBANDL, see [4] and references therein).

The common procedure for obtaining excitation functions is measuring a thin film of the element of interest and varying the energy of the incident ions as described elsewhere (see e.g., [5–7]). This procedure is extremely time-consuming and in many modern experimental systems not always applicable, e.g., in focused systems and micro-beam applications. Therefore different approaches were developed in the past, which avoid the need for a detailed scan of the beam energy by using bulk materials and calculate the cross-section directly from a spectrum of a material with well-known composition. In principle two different methods were applied in the literature. The first method as described by Barradas et al. [8,9] utilises the Bayesian inference method of the computer software NDF to fit cross-sections to the experimental spectra of bulk samples. This method is even on modern computers extremely time-consuming [8]. Another approach was first introduced by

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Källman et al. [10]. Therein the cross-section was determined directly from the yield of reaction particles by measuring a bulk sample with larger energy steps. It was applied for measuring elastic scattering cross-section of α-particles. In this paper a similar approach is used for determination of excitation functions of nuclear reactions with the incident and reaction particles being different.

The procedure for indirect determination of the differential cross-section versus energy is illustrated by means of a nuclear reaction for detection of ^{19}F atoms which is given by:

$$^{19}\mathrm{F} + \mathrm{p} \rightarrow {}^{16}\mathrm{O} + \alpha + \mathrm{Q} \quad \text{(abbreviated as } {}^{19}\mathrm{F}(\mathrm{p},\alpha_0){}^{16}\mathrm{O}).$$

Excitation functions of the reaction ^{19}F(p,α$_0$)^{16}O have been studied by several authors (see Fig. 1). Golicheff et al. [6] studied the cross-section for a backscattering angle of 150° and proton energies of 500–1800 keV and in a more detailed study for energies between 1000 and 1400 keV. Lerner and Marion [5] studied yield curves for ^{19}F(p,α$_0$)^{16}O and determined cross-sections at a backscattering angle of 90°. Dieumegard et al. [7] measured the cross-section for this reaction at 90° and 150° in great detail over a proton energy range of 300-2000 keV, by means of an automatic energy scanning system. More recently a detailed study for the energy range between 600 and 1000 keV and backscattering at 150° was performed by Lombardo et al. [11]. Paneta et al. [12] investigated a broad range of reactions including the one under consideration here but for high proton energies up to about 4 MeV.

The high Q-value of the ^{19}F(p,α$_0$)^{16}O reaction ensures that the α-particles are very well separated from protons elastically backscattered from the substrate. This means, the signal of the α-particles is detected in a range of the energy spectrum where background noise can be neglected. It is a prerequisite for the method presented here that the detected reaction particles have larger energies than the incoming particles elastically backscattered from the matrix. Additionally it has to be mentioned that the method cannot resolve fine structures of the excitation function with the width of resonances being below the energy uncertainty caused by detector resolution and energy straggling (see Section 3). But even if fine structures do occur, the method can be used to obtain a survey of the energy dependence of the cross section for identifying energy ranges which need to be investigated by other methods with higher energy resolution. If the two prerequisites mentioned above are fulfilled, the method can be easily applied in any ion beam laboratory with only a software for calculation of NRA spectra being needed.

2. Experimental

The measurements were carried out using the 3 MV Tandetron accelerator JULIA at the Institut für Festkörperphysik in Jena. The beam energy of incoming protons, E_{inc}, was 1335, 1535, 1735 and 1935 keV. Calibration of the beam energy was checked by analysing several resonances in elastic scattering of protons on ^{12}C and ^{28}Si targets at a backscattering angle of 170°. The uncertainty of the beam energy is 0.4%. Beam currents were between 5 and 8 nA. The diameter of the beam spot was approximately 1 mm. The number of protons that hit the target was determined by charge integration of the incident particle beam while using a Faraday cup with a voltage of 500 V.

The nuclear reaction particles (α-particles) and backscattered protons were detected by Passivated Implanted Planar Silicon (PIPS) detectors. The measurements were carried out at three different backscattering angles θ of 90°, 150° and 170°. In Fig. 2 the position of the detectors for the different angles is shown. For $\theta = 150°$ and $\theta = 170°$ the incident angle was 0°, while for $\theta = 90°$ the incident angle was 45°. The distance between sample surface and detector was measured using a mechanical instrument with a vernier slide gauge. The fixed standard detector at $\theta = 170°$ is located (112.4 ± 0.5) mm away from the sample and has a surface area of 50 mm^2 (as given by the manufacturer). This yields a solid angle of (3.96 ± 0.03) msr. The detector placed at $\theta = 150°$ or $\theta = 90°$ was (70.0 ± 0.5) mm away from the sample. This detector is mounted on a rotatable holder and its angular position was determined using the laser beam inside the beam line along the path of the ions and the goniometer of the target holder. A slit aperture was placed in front of this detector for better definition of the backscattering angle. This reduces the active detector area to (16.4 ± 0.2) mm^2 and a solid angle of (3.35 ± 0.09) msr is obtained.

For the processing of differential cross-sections using the nuclear reaction ^{19}F(p,α$_0$)^{16}O, a sample with a known concentration of fluorine is needed. Here a CaF_2 target was used to determine the differential cross-section σ_d assuming that the concentration of fluorine as a function of depth is homogeneous and corresponds to the stoichiometry of the material. To verify the results obtained, a second fluorine-containing reference sample, NaF, was measured. Owing to the energy loss of protons, the samples can be examined in a depth range of 0 to about 18 μm, depending on the composition of the sample and the backscattering angle.

RBS spectra for protons backscattered on CaF_2 were calculated with the program NDF [13] using own spectra-handling software. Because for Ca atoms no resonances occur in the proton energy range of interest here, fitting the spectrum height in the Ca part of the spectra was used to determine the solid angle. For the four beam energies applied a mean value of 4.07 msr at 170° and 3.44 msr at 150° and 90° is obtained. The deviation from the geometrically measured values of the solid angle is due to uncertainties in the charge integration. The uncertainty of the charge-solid angle product is 3%.

The energy per channel of the multi-channel-analyser was calibrated using the energy of protons backscattered on Ca surface atoms and the energy of α-particles from the nuclear reaction with ^{19}F surface atoms for the applied beam energies. An estimation of the effect of pulse height defects [14] induces an uncertainty in the

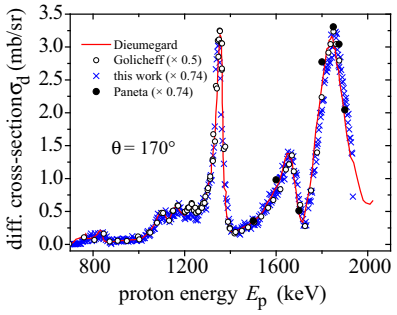

Fig. 1. Differential cross-section σ_d versus proton energy E_p (excitation function) of the nuclear reaction ^{19}F(p,α$_0$)^{16}O and a backscattering angle of $\theta = 150°$ [6]. The data measured by Golicheff et al. [6], Paneta et al. [12] and in this work are normalised to that measured by Dieumegard et al. [7] using the factors given in the legend.

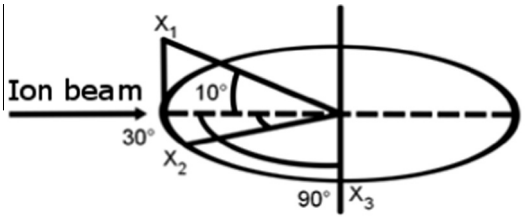

Fig. 2. Position of the PIPS detectors for the different backscattering angles: X_1: $\theta = 170°$, X_2: $\theta = 150°$ and X_3: $\theta = 90°$.