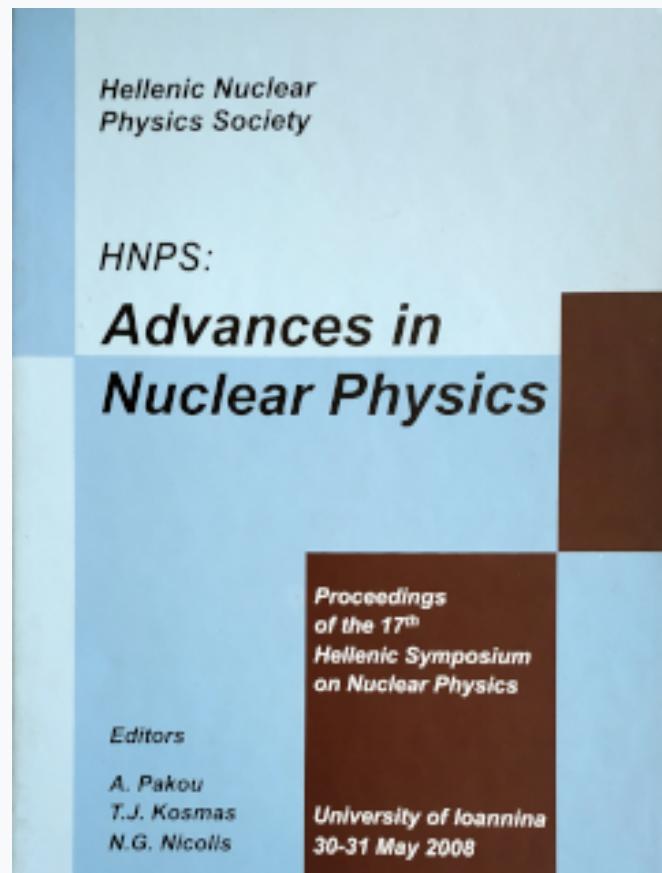


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# A Review on Recent Developments in Deuteron Induced Reactions Enhancing NRA Capabilities

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## Abstract

Nuclear Reaction Analysis (NRA) is well established as one of the principal IBA methods nowadays. Among the most important NRA characteristics are its high isotopic selectivity, its enhanced sensitivity for many nuclides, the capability of least destructive depth profiling and the possibility of simultaneous analysis of more than one light element in near-surface layers of materials. Moreover, in the particular case when deuterium is used as probing beam, critical advantages for NRA studies emerge. As NRA quantifies individual light isotopes absolutely, and can depth profile with nanometer resolution, it is the most suitable ion beam technique for the determination of the concentration and depth profiling of light elements in complex matrices. However, as already pointed out in the recent literature, the application of NRA to the determination of the concentration and the depth profiling of light elements is frequently impeded by the lack of adequate and/or reliable experimental differential cross section data. It is the ambition of the present work to contribute in the fields of differential cross section measurements, as well as of data evaluation and general theoretical analysis.

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## Introduction and General Overview

Interactions of MeV ions with matter permit off-line analysis of light elements in solids (**CPAA** – Charged Particle Activation Analysis – implying the use of high resolution  $\gamma$ -ray spectroscopy) and in-beam microanalysis and imaging of sample constituents using excited X-rays (**PIXE** – Proton Induced X-ray

Emission),  $\gamma$ -rays from nuclear reactions (**PIGE** – Proton Induced Gamma-ray Emission), outgoing nuclear reaction particles (**NRA** – Nuclear Reaction Analysis, implying emitted charged particles), elastically scattered ions (**BS** – Backscattering, including **RBS** – Rutherford Backscattering Spectroscopy, implying potential scattering, **NBS** – Nuclear Backscattering Spectroscopy, implying non-Rutherford cross-sections, and – in most cases – the implementation of the resonant mechanism in elastic scattering, and **ERDA** – Elastic Recoil Detection Analysis), or visible and infrared emissions from the sample. The ion beam can also be channelled down crystal axes and planes (channeling related phenomena). Many of these techniques can be combined with channelling to study the lattice location of species. In general, MeV light ions are very penetrating while producing very little sample damage. These qualities permit the *in situ*, least destructive analysis and imaging of buried structures such as solid and fluid inclusions in minerals. All the above mentioned techniques are integral parts of Ion Beam Analysis (**IBA**), a strong, interdisciplinary field [1-3], combining Nuclear Physics and Material Science, which is continuously enriched and evolving. Among IBA techniques, RBS, NBS, ERDA, resonant PIGE and NRA are more commonly implemented for elemental depth profiling.

In a more detailed approach, Nuclear Reaction Analysis (NRA) constitutes by itself a general category of techniques [4, 5] involving nuclear reactions between a target nucleus and beam particle. Above a certain energy of the incident beam, which depends on the incident particle and the target nucleus ( $\sim$ 0.3–1 MeV for light elements), the backscattered (forward scattered or recoiled) particles and other energetic particle species appear in the detected spectrum. When the latter are detected, they usually provide information that is not available by RBS. When a layer of a light element is positioned on top of a heavy substrate, then the RBS spectrum of the former is superimposed on the corresponding one of the latter. Since the Rutherford cross section increases with the square of the atomic number, the light element's signal will be seen against a huge background. While NBS measurements could solve this problem only for a limited number of cases, NRA, through the detection of charged particles, could in principle provide the required answers.

Among the most important NRA characteristics are its high isotopic selectivity, its enhanced sensitivity for many nuclides, the capability of least destructive depth profiling and the possibility of simultaneous analysis of more than one light element in near-surface layers of materials. Moreover, in the particular case when deuterium is used as probing beam, critical advantages for NRA studies emerge, due to: a) the simultaneous excitation of most light elements (e.g. O, N, C, F, Al, Mg and S) usually co-existing in complex matrices, either as main constituents or as impurities, b) the enhanced sensitivity and accuracy, mainly due to the generally large cross sections of the deuteron-induced nuclear reactions and c) the applicability of rather simple accelerator

setups, such as even small VdG machines. These advantages are, of course, offered at the expense of background interference in certain cases (e.g. peak overlaps, 3–body reaction kinematics). Also, certain radiation safety precautions are mandatory due to the emitted neutrons from (d,n) reactions (on the target elements and structural materials), and/or deuteron breakup (for deuteron beam energies above 2.2 MeV).

As NRA quantifies individual light isotopes absolutely, and can depth profile with nanometer resolution, it is the most suitable ion beam technique for the determination of the concentration and depth profiling of light elements in complex matrices. However, as already pointed out in the recent literature, the application of NRA to the determination of the concentration and the depth profiling of light elements is frequently impeded by the lack of adequate and/or reliable experimental differential cross section data.

The principal objectives of this study are: (a) the measurement, with the highest possible precision, of the differential cross sections of a number of deuteron and proton induced reactions on light element isotopes ( $^{10,11}\text{B}$ ,  $^{14}\text{N}$ ,  $^{6,7}\text{Li}$ ,  $^{19}\text{F}$ ) absolutely necessary for Nuclear Reaction Analysis (NRA), at suitable (low) energies and at multiple backward angles, and (b) their subsequent theoretical analysis and evaluation, which will solve the main problems in the implementation of NRA.

The selection of the reactions to be studied has been performed taking into account the availability, reliability and necessity of the corresponding cross section data. Boron, lithium, nitrogen and fluorine are very common elements, even in small quantities, in a variety of matrices. Their presence is evident in the particle spectra obtained during profiling of other light elements, especially carbon and oxygen (e.g. when using deuteron beams). Therefore, the high precision determination and profiling of these elements is of extreme importance for technological applications (e.g. semiconductor industry, biomedical and environmental research etc). Thus, as far as the experimental part is concerned, the anticipated outcome of this work will be the availability, to all members of the Ion Beam Analysis (IBA) community, of several, critical for NRA, differential cross sections for proton and deuteron induced reactions, still lacking in literature.

Moreover, the theoretical analysis and evaluation of such reactions (based both on the available experimental points and on R-matrix calculations and on other existing models) is of vital importance for practical NRA applications. Differential cross section datasets exhibit differences of more than 20%, for the same beam energy range, even in the best studied cases (e.g.  $^{12}\text{C}$ ,  $^{16}\text{O}$ ), thus impeding high precision depth profiling [6]. The aim of this part of the project is the production of reliable cross section datasets for all proposed reactions in a sufficiently wide energy interval at any scattering angle.

These datasets will evidently become the standard choice in all commonly used NRA algorithms (e.g. SIMNRA, WinDF etc.). However, such an analysis/evaluation presents many interesting theoretical challenges [7]. There are several reasons that enhance the complexity of this problem, such as e.g. the electric charge asymmetry of the deuteron, the effects of a multiple projectile–target exchange of nucleons, the existence of possible additional direct exchange processes (knock–out and heavy stripping) and the problem of taking into account close lying resonances interference. Also, in the case of deuteron induced reactions, an appropriate theoretical treatment has to take into account simultaneously all the open reaction channels, namely the (d,d), (d,p), (d,α) as well as the indirect influence of the (d,n) one. Thus, a theoretical analysis and subsequent evaluation requires a strong coordinated effort in both the experimental and the theoretical fields.

The scientific validity and importance of this study has been recognized by the International Atomic Energy Agency (IAEA, CRP ‘Development of a Reference Database for Ion Beam Analysis’[8], 2006–2009) recently. It has to be emphasized that the existing research agreement with IAEA covers only a small part of the measurements described in the current manuscript. It is evident, however, that there is a strong, ongoing effort, for IBA and NRA studies; thus the anticipated outcomes of the present work will have a significant impact in both the experimental (enhanced NRA capabilities of multiple light element depth profiling in various complex matrices) and the theoretical field (clarification of certain details in the treatment of multiple open reaction channels).

## Methodology and Schedule

The experiments are carried out at the Institute of Nuclear Physics of N.C.S.R. ‘Demokritos’, using the 5.5 MV TN–11 HV Tandem Accelerator. For the main course of experiments a motor–driven large goniometer setup is mandatory. This goniometer has an accuracy of  $0.01^\circ$  in detector and/or target positioning, and is capable of sustaining simultaneously 16 single Si surface barrier detectors, or 8  $\Delta E/E$  solid state detector telescopes for particle identification (depending on the reaction studied), associated with standard or fast NIM/CAMAC electronics (including all appropriate supporting units and ADCs) respectively. It also allows for target cooling with water or methanol through a closed circuit during data acquisition. The needs for high vacuum, efficient charge collection and several standards (e.g. an absolutely calibrated α–source, and samples suitable for stopping power testing) must always be fulfilled. At least two full working weeks of beam time are required for each isotope and reaction. In certain cases measurements are also performed in the forward direction, in order to facilitate the theoretical calculations. Following each measurement, the experimental data are analyzed at N.T.U.A. using appropriate software packages for spectrum analysis. The theoretical code for the

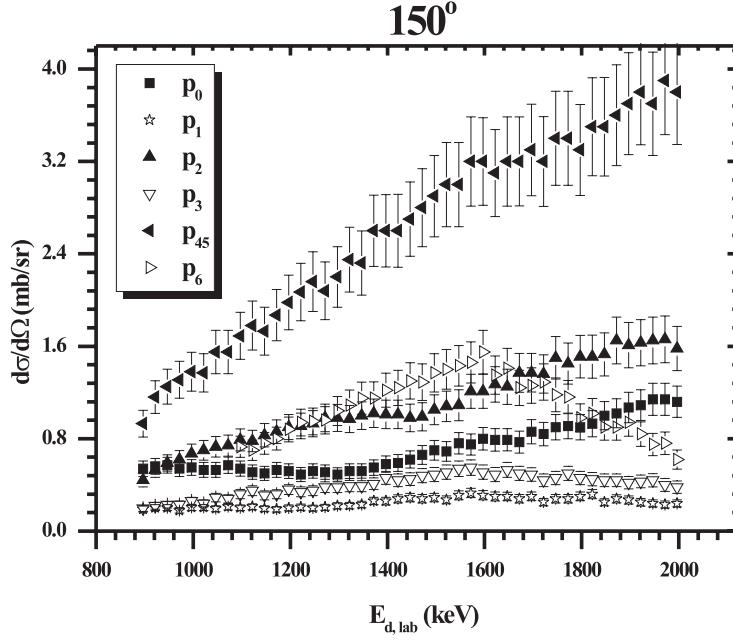


Fig. 1. Differential cross section values (mb/sr) of the  $^{10}\text{B}(\text{d},\text{p}_{0,1,2,3,45,6})^{11}\text{B}$  reactions at  $150^\circ$ , for  $E_{d,\text{lab}}=900\text{--}2000$  keV. The combined experimental errors are included in the graphs. The errors along the x-axis (energy ripple of  $\pm 2$  keV) are not visible due to the adopted scale.

analysis and the evaluation of the results has not yet been developed but work is in progress in this field. It has to be noted here, that our group has recently studied the  $\text{d}+^{12}\text{C}$ ,  $\text{d}+^{10}\text{B}$ , and  $\text{d}+^{32}\text{S}$  systems at several backward detector angles, in the energy range  $E_{d,\text{lab}}=900\text{--}2000$  keV. As a consequence, more than 5000 new differential cross section values have been added to IBANDL ([www-nds.iaea.org/ibandl/](http://www-nds.iaea.org/ibandl/)). A characteristic example of such measurements is illustrated in fig. 1 for the  $\text{d}+^{10}\text{B}$  case, with all the analyzed levels at  $150^\circ$ .

A brief outline of only the programmed experimental time schedule for a five-year period starting in 2007, would be as follows:

**Year 1:** Study of the  $^{11}\text{B}(\text{d},\text{p})$  and  $^{11}\text{B}(\text{p},\alpha)$  reactions using natural and/or enriched thin boron targets evaporated on or implanted (IBAD) in thin self-supporting substrates. The main experimental problems are connected to the target construction, to the unstable resulting nucleus  $^8\text{Be} \rightarrow 2\alpha$  in the case of the  $^{11}\text{B}(\text{p},\alpha)$  reaction (3-body kinematics), as well as to the low Q-value ( $\sim 1146$  keV) of the  $^{11}\text{B}(\text{d},\text{p}_0)$  reaction. In this latter case, the use of foils in

front of the detectors in order to avoid the elastic scattering from the matrix will be avoided. All the reactions will be measured at 4-8 different backward and forward angles in appropriate angular steps, with an average energy step of 25 keV (or smaller in the vicinity of resonances).

Year 2: Study of the  $^{14}\text{N}(\text{d},\text{p})$  and  $^{14}\text{N}(\text{d},\alpha)$  and  $^{14}\text{N}(\text{d},\text{d})$  reactions using self-supporting ultra thin  $\text{Si}_3\text{N}_4$  targets. The main experimental problems in the measurements of these reactions are connected to the requirement of implementing thick Si surface barrier detectors for the  $^{14}\text{N}(\text{d},\text{p}_0)$  reaction and to the partial possible overlap of proton and alpha groups (use of  $\Delta\text{E}/\text{E}$  telescopes). Measurements will be carried out also at certain angles in the forward direction for theoretical purposes. In this case, following the pioneer works of the past at  $150^\circ$  [9, 10], all the reactions will be measured at 6/12 different backward/forward angles, at  $E_{d,\text{lab}}=900\text{--}2000$  keV, with an average energy step of 15–25 keV (or smaller in the vicinity of resonances).

Year 3: Study of the  $^{19}\text{F}(\text{d},\text{p})$  and  $^{19}\text{F}(\text{d},\alpha)$  and  $^6\text{Li}(\text{d},\text{p})$  and  $^6\text{Li}(\text{d},\alpha)$  reactions using thin  $^{19}\text{F}$  and  $^6\text{Li}$  enriched targets evaporated on or implanted in thin self-supporting substrates. The main experimental problems are connected to the relatively low differential cross sections involved, especially in the cases of the  $^{19}\text{F}(\text{d},\alpha_0)$  and  $^{19}\text{F}(\text{d},\alpha_1)$  reactions, as shown in the past at  $156^\circ$  [11]. For all the reactions studied, the  $(\text{d},\text{n})$  reaction channels are open (and particularly strong due to the high Q-value in the case of fluorine). An effort will be made to measure the reactions at 8/8 different backward/forward angles, at  $E_{d,\text{lab}}=900\text{--}2000$  keV, with an average energy step of 25 keV (or smaller in the vicinity of resonances).

Year 4: Study of the  $^{19}\text{F}(\text{p},\alpha)$  and  $^7\text{Li}(\text{p},\alpha)$  reactions using thin  $^{19}\text{F}$  and  $^7\text{Li}$  natural targets evaporated on or implanted in thin self-supporting substrates. The main experimental problems are connected to the large number of existing resonances in the case of the  $^{19}\text{F}(\text{p},\alpha)$  reaction, as well as its possible interference with the  $^7\text{Li}(\text{p},\alpha)$  one for certain proton energies. An effort will be made to measure the differential cross sections for at least 8/8 different backward/forward angles, at  $E_{p,\text{lab}}=900\text{--}3500$  keV, with an average energy step of 25 keV (or smaller in the vicinity of resonances).

Year 5: Study of the most difficult and least promising cases for NRA purposes, which would nonetheless complete the elemental studies, namely of the  $^{10}\text{B}(\text{p},\alpha)$ ,  $^6\text{Li}(\text{p},^4\text{He})^3\text{He}$  and  $^6\text{Li}(\text{p},^3\text{He})^4\text{He}$  reactions using once more thin  $^{10}\text{B}$  and  $^6\text{Li}$  isotopically enriched composite targets. The main experimental problems are related to the relatively low Q-values involved. An effort will be made to measure the reactions at 8/8 different backward/forward angles in steps of 10 degrees, at  $E_{p,\text{lab}}=1000\text{--}4000$  keV, with an average energy step of 25 keV (or smaller in the vicinity of resonances).

The theoretical analysis will advance in parallel during the period 2007-2011.

## Conclusions

The measurement of differential cross sections, suitable for NRA purposes constitutes an exciting field for low energy nuclear physics, which provides important challenges on both the experimental and the theoretical fields. Our group aims at providing an important amount of accurate and reliable data over the next five-year period. These data would be of extreme importance to the scientific community.

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