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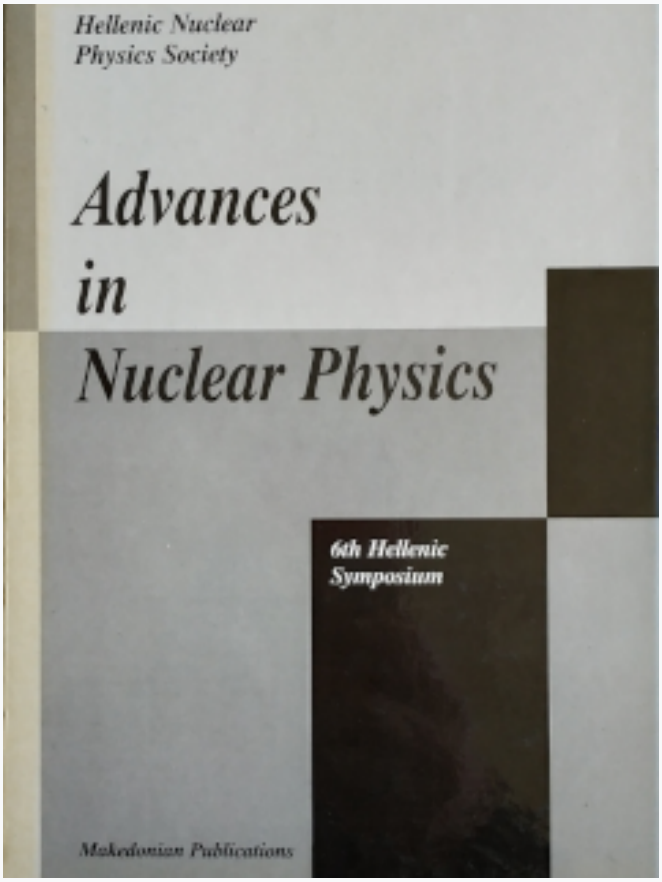

Determination of light elements using the RBS method

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Abstract

The RBS (Rutherford Backscattering Spectroscopy) method is a classical method which has been successfully used for the study and analysis of surface elements. The strong points as well as the weaknesses of the technique are briefly outlined and emphasis is given in the determination of light elements. A method is proposed in order to overcome the problem and the first results and prospects for the future are analysed.

1 Introduction

The RBS (Rutherford Backscattering Spectroscopy) method is a classical nuclear technique for the study and analysis of surface elements. The corresponding theory was developed by Geiger and Marsden (1913) but it was standardized by K. Allison around 1960, owing to the evolution of solid state detectors.

The principle of the method is the following: A well-collimated beam of particles with mass M_1 and charge Z_1 falls on a homogeneous target with corresponding M_2, Z_2 and the backscattered particles within the solid angle $\Delta\Omega$ are registered by a silicon surface barrier detector. Then, the experimental spectrum can in principle be described as shown in fig.1 by the following equations:

$$E_s = E_o \cdot K^2$$

$$K = \frac{M_1 \cdot \cos(\vartheta)}{M_1+M_2} + \sqrt{\left(\frac{M_1 \cdot \cos(\vartheta)}{M_1+M_2}\right)^2 + \frac{M_2-M_1}{M_1+M_2}}$$

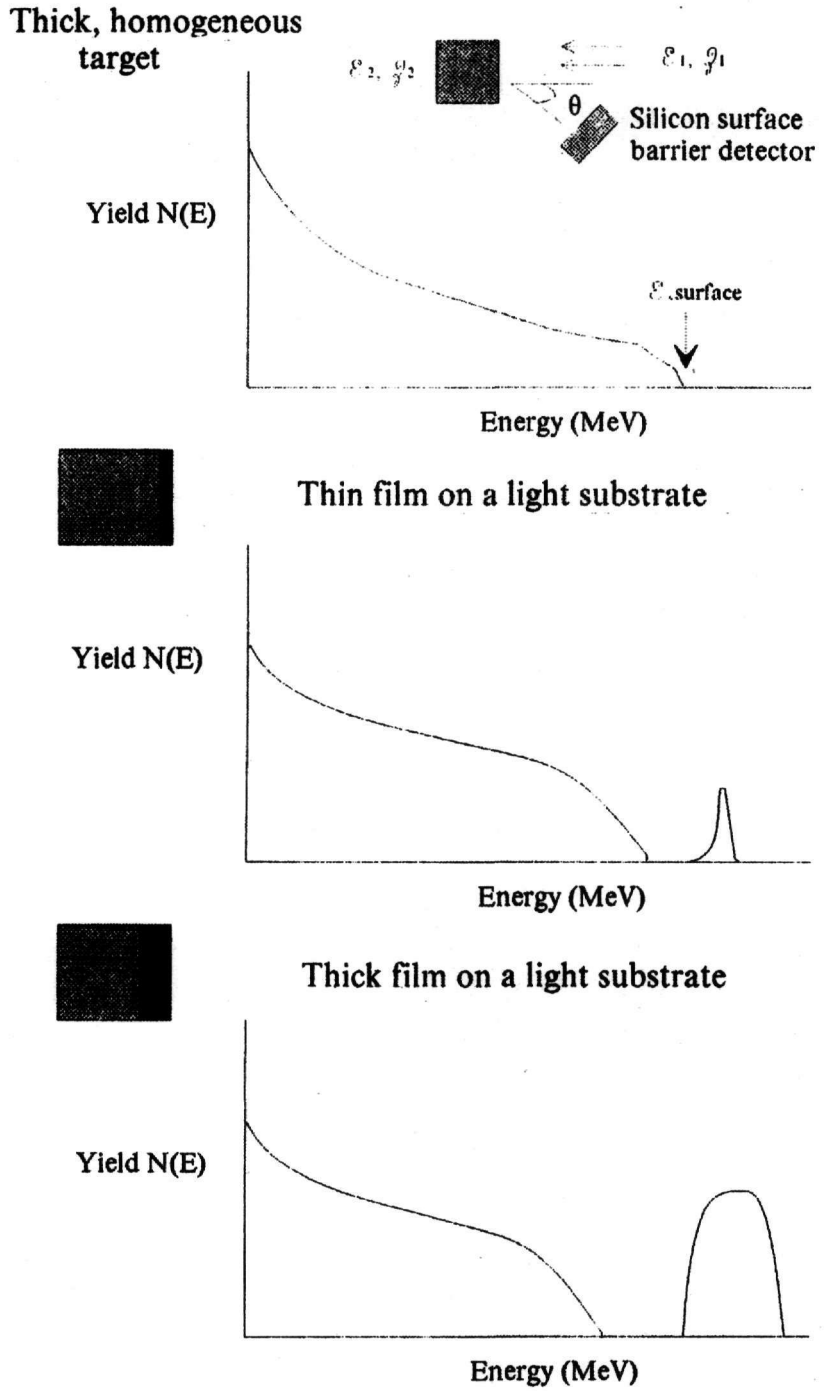

Fig.1-3 Principle of the RBS method.

$$Yield = N_s \cdot \left(\frac{d\sigma}{d\Omega}\right)_R \cdot \Delta\Omega \cdot Q$$

where E_s is the energy of the particles backscattered from the surface of the target, E_o is the initial energy of the beam, ϑ is the scattering angle between the normal to the target surface and the detector, K is the kinematic factor, Q is the total accumulated charge and N_s is the integrated number of particles falling on the target surface. It can be proved that these basic equations can be enhanced in order to correctly describe the experimental yield up to a certain depth below the surface of the target.

Among the many advantages of the RBS method we would like to cite its ability to provide information concerning the depth composition of a target both quantitatively and qualitatively, its great sensitivity in thin film analysis when a light substrate is used (100-200 Å), its use in crystallography and also the fact that the measurements are acquired relatively quickly and the destruction of the samples is normally avoided.

However, this technique requires a specific preparation of the samples before the irradiation in order to avoid roughness effects (and also certain precautions should be taken against contaminations either through dust or oxidization), it requires qualitative *a priori* information about the chemical composition of the target and it has smaller sensitivity for the detection of light elements.

A particular emphasis is put recently on the precise determination of carbon, oxygen, nitrogen and boron because of their important industrial applications (e.g. hydrated carbon in steel, oxidations and absorptions, nitrogen in metals and ceramic materials, T_iB_x stoichiometry etc.).

2 Experimental setup

The experimental setup at N.C.S.R. "Demokritos" includes a goniometer system (C.E.&A.) which permits experiments for backscattering spectroscopy as well as channelling for oriented crystalline targets. It is composed of a vacuum chamber, a four-axis goniometer with the appropriate motor drivers and controller, a fixed laser pointer for the determination of the precise beam-target orientation and the corresponding electronics. Data acquisition (including current integration, multichannel analyzer card) and analysis (with the corresponding software) is performed via an ordinary personal computer[@]. The accuracy of our measurements (including several systematic errors like finite solid angle corrections, imperfect charge collection, changes in the detector resolution etc.) is estimated to be in the order of 7-10%.

3 Indirect method for the determination of light elements

It is possible to determine the stoichiometry of a certain light element in an indirect way if additional information concerning the quantitative proportions of any heavy elements present in the target is available (e.g. with the use of XRF measurements in addition to the RBS ones). The experimental spectra are simulated with RUMP, whose code takes into consideration only the Rutherford formula for the cross section, ignoring any nuclear effects. The simulated spectra which describe the "supposed" composition of the targets are very sensitive to the presence of light elements because then, the corresponding average stopping power of a given layer decreases and so does its apparent "thickness" in the spectrum as well as the total yield. On the other hand, if one tries to fit the experimental data by overestimating the thickness of a given layer the simulation fails completely to describe the subsequent layers.

The main disadvantage of the above described method is the existence of many free parameters which can make the analysis either complicated or even impossible. Mainly, the possible existence of more than one light elements and a variation of their composition with the depth inside the target can cause inaccuracies of the order of 20% or more. Also in the case of a multi-layered sample the analysis and weighing of all the factors present can become quite time consuming, thus limiting the usefulness of the method.

An interesting example of the power of the method is shown in fig.4. It concerns a thin Co-Tb film grown through evaporation on a silicon wafer substrate. The film demonstrated interesting magnetic qualities and thus the determination of its composition was imperative. It was bombarded with 1.5 MeV protons and its complicated multi-layered structure is illustrated in the following table. The results shown were obtained after a detailed χ^2 minimization and correspond to atomic proportions.

TABLE 1.

Layer	Thickness (Å)	Co	Tb	O	Cr	Si
1	50	0	0	0	1	0
2	16121.8	1	0.58	1.32	0	0
3	4612.15	1	0.60	2.24	0	0
4	300000	0	0	0	0	1

4 Direct methods for the determination of light elements - Results and discussion

Many techniques have been developed in order to achieve a direct quantitative evaluation of light elements in the last twenty years. In general, nuclear reactions – e.g. (p, γ), (α, γ), (d, α), (d, p) – are utilized [1] which are sensitive to one component only. Although they have proved quite successful, they present certain disadvantages; that is, they are useful only for certain materials and beam-target conditions, they don't give information for the whole target structure, the corresponding cross sections are several orders of magnitude smaller than the elastic one – thus requiring longer exposure periods – and finally they require a radical transformation of the standard detector-goniometer setup. A more promising method which has been exploited lately is ERDA (Elastic Recoil Detection Analysis) which has great sensitivity for light element detection and provides an adequate mass separation for most applications; nevertheless it is much more demanding in accelerator capabilities, requires complex experimental devices, the depth profiling is severely limited due to the use of very heavy ions, often resulting also in the destruction of the samples, and the straggling factor becomes very important. Although ERDA's sensitivity for heavy ions is significantly poor, some impressive results have been published lately [2] although the accuracy is lower than the standard RBS one.

For all the above cited reasons it seems that the most promising and convenient approach to the problem could be the extensive use of the nuclear resonance phenomenon which does not require any changes in the standard apparatus, and only minor ones in the software analysis package commonly used (RUMP). The reason for this phenomenon is the constructive as well as destructive interference between the elastic cross section of the compound nucleus formed and the elastic cross section for direct scattering (Rutherford case). The former is symmetric around 90° relative to the beam, while the latter shows the well known $1/\sin^4(\theta/2)$ dependence, thus it is significantly reduced in the case of backscattering angles. As a result, the total elastic cross section can become 30 to 150 times larger than σ_R, a factor which enhances the sensitivity and permits the detection of even low light element concentrations. Another advantage of this technique is that the inelastic cross sections which always accompany the resonance phenomena are 1 to 3 orders of magnitude smaller than the elastic ones, causing inaccuracies of less than 1% [3].

For the occurrence of the phenomenon different beams can be used, the most common ones being protons and α particles. The use of each beam presents certain advantages. For the same E_o, protons permit sample analysis at a greater depth (ranging from 5 to 10 times deeper), the straggling factor is reduced and greater sensitivity ($\geq$ 10 times) for the detection of light elements

can be achieved, while α particles due to their greater mass provide better resolution as far as energy and depth are concerned.

In the following table several well known resonances for oxygen in the range of a few MeV are presented in the case of protons as well as α particles.

TABLE 2.

Protons:		α particles:	
Energy (in MeV)	Γ (in keV)	Energy (in MeV)	Γ (in keV)
2.66	20	**2.490**	**24**
3.47	3	3.045	10
4.354	240	3.090	5
4.787	1630	3.380	10
5.231	72.5	3.885	3

An example of the above method is shown in fig. 5 in the case of an YBaCuO superconductor grown with evaporation on an MgO substrate. The target was bombarded with 2.5 MeV α particles. Although the simulation using the "indirect method" reproduces the experimental spectrum with remarkable accuracy in the case of heavy elements, it fails considerably to reproduce the oxygen peak caused by the 2.49 MeV resonance as shown in table 2, thus enhancing the uncertainty concerning the surface oxygen concentration which greatly determines the superconductor properties. The simulated structure is presented in table 3 and it is directly comparable to the theoretical one: $Y_1 Ba_2 Cu_3 O_7$ on $Mg_1 O_1$.

TABLE 3.

Layer	Thickness (in Å)	Y	Ba	Cu	Mg	O
1	1929	1	1.845	2.365	0	6.152
2	100000	0	0	0	1.1	0.9

Our current goal is to combine, correlate and analyze several cross section data concerning protons and alphas for different nuclear resonances and backscattering angles, to add – where necessary – measurements acquired *in situ* and thus to adapt the RUMP code in order to make it capable of simulating the concentrations of more than one light elements with special emphasis put on samples containing C, O, N or B. The tests of the method's accuracy will be performed using well calibrated targets of known stoichiometry e.g. titani-

um nitrites provided by the Beschleunigerlaboratorium at Garching (Munich, Germany). Our results will be the subject of a future publication.

5 Conclusions

The nuclear resonance analysis seems to be the most promising method for the detection and correct evaluation of any concentrations of light elements. Difficulties are expected from the steady background caused by elastic scattering on heavier elements, from the change of the shape of resonances with the penetration depth inside the target – mainly due to energy straggling – and from the theoretical problems concerning the mathematical treatment of overlapping resonances. The examination of the above mentioned problems is still in progress.

Despite the difficulties however, we hope to establish a reliable system for the quantitative analysis of light elements exploiting the present experimental setup at N.C.S.R. "Demokritos" in the near future.

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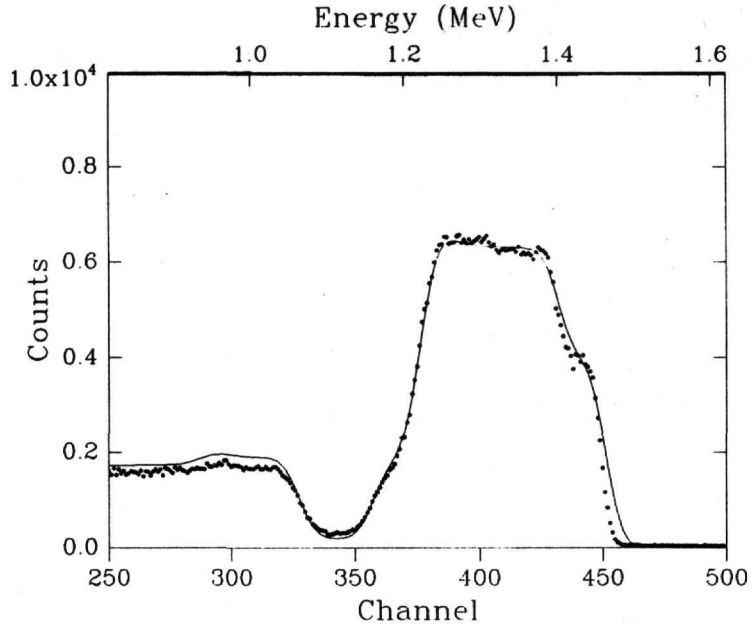

Fig.4 Co-Tb film on Si (using 1.5 MeV protons, dots for the experimental points, solid line for the simulation).

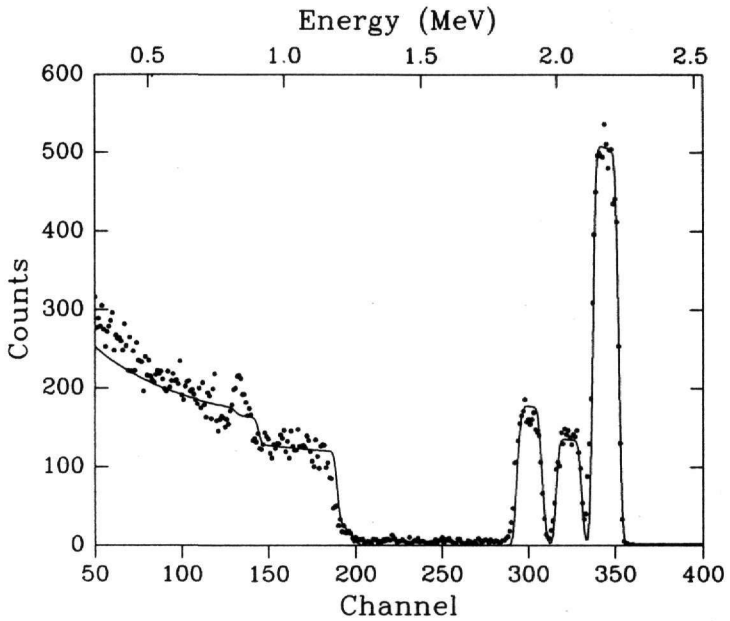

Fig. 5 YBCO film on MgO (using 2.5 MeV α)