

APPLICATION OF Li ION BEAM OBTAINED BY THE CESIUM SPUTTERING SOURCE FOR RBS ANALYSIS OF OPTICAL LAYERS

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High energy Li ions of multiple charge states (Li^+ , Li^{2+} and Li^{3+}) were produced in a tandem accelerator from a source of negative ions by a cesium sputtering (SNICS) ion source. The negative ion current of Li ions up to 32 nA was obtained in the pre-acceleration stage after the inflection magnet and retractable aperture. The current of the high energy ion beam obtained after acceleration and used for RBS spectra measurements was up to 3 nA. The Li target cone production procedure and design are presented in this work. Initial experiments using Li RBS were performed and their results indicate enhanced analytical application capabilities of RBS for an optical coating analysis, compared to the conventional proton RBS.

Keywords: ion beam, accelerator, Cs sputtering, RBS

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1. Introduction

Rutherford backscattering spectroscopy is widely used to analyse the structure and elemental composition of surface layers of solids [1]. Usually the He^+ beam is used for RBS experiments due to relatively good mass and depth resolution [2]. It is known that using the proton beam for RBS increases the analysed depth, but resolution by mass is decreased [3]. The advantages of using ions heavier than He^+ for the RBS analysis with a noticeable improvement of mass resolution and sensitivity in RBS are well known [4]. E. Norbeck had shown that the distribution of elements as a function of distance from the surface is more clearly revealed with ^7Li than ^4He [5]. On the other hand, Hiroaki noted that the energy resolution of the detector generally decreases as the projectile mass increases if a silicon surface-barrier detector is used [6]. Possible solution of this problem is the use of higher projectile energy in order to overcome the decrease in energy resolution. The Hiroaki group also used 8 MeV carbon and oxygen ions to improve mass resolution, depth resolution, and sensitivity.

A general Ionex Corporation Model 4110A 1MV accelerator in the Center for Physical Sciences and Technology (CPST) is equipped with a Model 834 Hiconex Cs ion sputter source [7]. Very low He^- yields are reported from cesium sputtering sources [8] and accurate depth profiling requires better mass resolution than proton RBS has to offer. According

to M. Mayer [9], the best alternative for the conventional He RBS is to use the lithium ion beam because identical or better depth resolution with Li than with He can be achieved in the RBS analysis, combined with enhanced mass resolution and sensitivity. It was decided to manufacture a Li target cone for a cesium sputtering ion source for the purpose of improving analytical application of RBS.

One of analytical applications of RBS is quality measurement of optical coatings [10]. The quality of coatings of optical elements is one of the most important issues for any optical system. The proper choice of the type of coatings and their structure gives the possibility to improve parameters of the optical system making light transmission more effective, reducing light reflections and scattering, also suppressing, choosing or transforming light polarization, etc. Development of sophisticated optical coatings includes several important steps: choosing the proper materials, coating design, coating technology, substrates, etc. To obtain the high quality of required optical elements, one must ensure precise thickness, chemical composition and stoichiometry of layers.

2. Experiment

Cesium ions in a Hiconex 834 Cs ion sputter source are formed by tungsten surface ionization. Accelerated Cs^+ ions are used to bombard a cesiated inner surface of a conical target canal lined with the beam

material [7]. Beam ions are sputtered from the surface of the cone by cesium and produce low velocity atoms which charge exchange in the deposited cesium surface layer. Negative ions are extracted from the exit end of the target cone and are accelerated to ground potential, acquiring energy up to 30 keV. Twelve target cones may be positioned into the primary beam without breaking vacuum for rapid, convenient change of beam species. Actuation is provided by an air cylinder operating a ratchet and pawl mechanism through bellows. The tandem accelerator is equipped with a gas stripper instead of foils, so the system vacuum during operation is about 2×10^{-5} mbar. N_2 5.0 gas is used in the tandem gas stripper. The ion current is measured with two Faraday cups: one before the acceleration system and other after the high energy magnet before the measurement chamber. All measurements were done with extraction voltage of 25 kV in the ion source and terminal voltages of 700 kV and 750 kV. The terminal voltage is monitored via a voltage divider and the energy of the beam was calibrated using ${}^7\text{Li}(p,n){}^7\text{Be}$ nuclear reaction with the 1.881 MeV threshold.

According to R. Middleton and Mayer [8, 9], the most suitable material to produce the Li^- ion beam in SNICS is lithium hydroxide, prepared from lithium metal, with the lifetime of 4 to 8 hours depending on the negative ion current. For routinely performed RBS measurements one needs a steady beam producing a target cone with as long lifetime as possible. It was suggested [8] that lithium metal should have the longest cone lifetime, although the beam yield is reported significantly lower – $0.5 \mu\text{A}$. The predrilled titanium cone was used as a support cone for the lithium target (Fig. 1). A 1 mm thick lithium layer was manufactured by pressing lithium metal into the titanium cone in the Ar atmosphere, then rap-

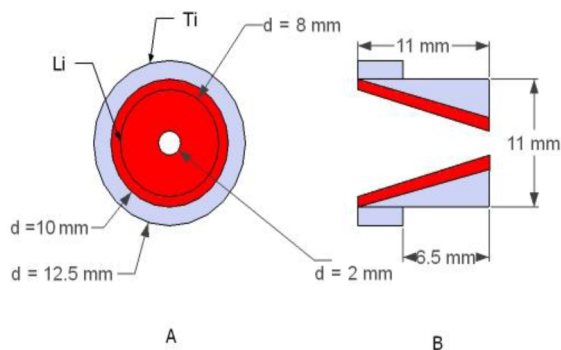


Fig. 1. The titanium support cone with the pressed in lithium metal layer of 1 mm thickness. A – top view (beam entrance side), B – horizontal section profile.

idly placed in SNICS and evacuated to the pressure of 2×10^{-5} mbar, to avoid oxidation of the lithium surface and of cesium in the SNICS source.

The negative ion current from SNICS is a function of target cone composition, suppressor voltage, cesium ion flux and ionizer power. The rate of cesium vapour flow influences the cesium positive ion current and cesium deposition on the target cone surface [11]. Therefore, the temperature of the cesium reservoir is usually controlled at the optimal level slightly below 180°C .

Ion mass analysis using the high energy magnet and preliminary results of RBS experiments using the Li beam was performed to confirm and characterize Li^+ , Li^{2+} and Li^{3+} beams. A collimating aperture of 2 mm in diameter was placed 10 cm in front of the target during RBS experiments. A Canberra DSA-1000 Multichannel analyzer and an Ortec Model 142A preamplifier were used with a surface barrier detector (AMETEK CR-015-050-100, 15 keV alpha resolution) at a scattering angle of 135° for RBS data acquisition.

The optical coating, manufactured in CPST by ion beam sputtering technique, was selected as the target for initial RBS experiments. Optical layers, consisting of 200 nm SiO_2 on the top of 200 nm Nb_2O_5 , were sputtered on the fused silica substrate.

3. Results and discussion

Li ion beams were characterized after 8 hours of operation in order to cesiate the inner surface of the conical Li target. The Li^- ion current of 29 nA was obtained with the following parameters of SNICS: ionizer current 28 A, suppressor voltage 4 kV, cesium boiler temperature 180°C and extraction voltage 25 kV. The inflection magnet (bending radius of 59.26 cm at 30°) current was scanned from 0 to 25 A and the negative ion beam yield from the Li target was measured in the pre-accelerated stage by a Faraday cup after the aperture of 5 mm in diameter.

Dependence of the negative ion beam current from the Li target on the inflection magnet current is presented in Fig. 2. H^- , H^{2-} and H^{3-} peaks can be identified in the recorded spectra at the region of low inflection magnets current. Usually hydrogen gas is sprayed directly on the titanium target cone in SNICS to produce H^- beam, so the presence of these peaks can be explained by partial contamination of the Li cone surface and by the leaking needle-type valve of the hydrogen gas fed system.

The obtained results show that the Li^- beam is intensive and well separated from others, therefore being a good choice for further acceleration. Li^{2-} ion yield

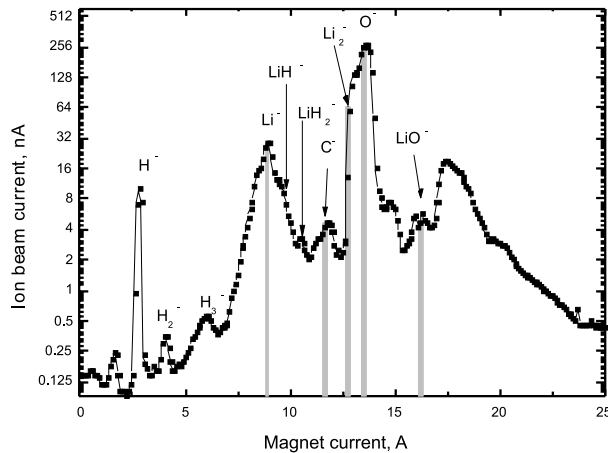


Fig. 2. Negative ion beam current yields from the Li target obtained in the pre-accelerated stage by scanning inflection magnet current. Position of columns indicates the current of the inflection magnet, calculated by Eq. for different mass species: Li^- , C^- , Li_2^- , O^- and LiO^- .

could not be estimated properly due to high intensity of the O^- yield and poor mass resolution of the inflection magnet. Moreover, the yield of Li^{2-} peak is reported more than one order lower than that of Li^- [8] in SNICS. Intensity of the LiO^- molecular ion was expected to produce low positive ion yields in the accelerated stage, so only the Li^- peak was accelerated.

Ion beam yields, obtained by the scanning inflection magnet current, correspond well with other results [8]. Species of ion beam mass was assigned using a relation between kinetic ion energy and magnetic field (bending radius of ion $F_B = qvB = mv^2/r$):

$$\frac{m}{q} = \frac{B^2 r^2}{2U},$$

where m is the mass of accelerated ion, q is the charge state of that ion, B is the magnetic field, r is the bending radius and U is a potential difference for the region of acceleration.

Different positive Li ion charge state yields at different terminal voltage (700 kV and 750 kV) after acceleration are presented in Fig. 3. The Faraday cup measures charge, but not the charge state of the ion. If the ion is ionised more than one time, the current by particles is lower than the current measured by in-

duced charge. In this case we have Li^{3+} , Li^{2+} and Li^+ charge states, so the current, measured by the Faraday cup by charge, has to be divided by 3, 2 and 1, respectively, to obtain current by particle. The number of incident ions is one of the most important parameters in RBS. Particle current yields of the Li^{3+} , Li^{2+} and Li^+ charge state at the 700 kV terminal voltage were measured by the second Faraday cup and reached 0.03 nA, 0.95 nA and 1.25 nA, respectively. This makes 1%, 43% and 56% from the total yield of particles for the Li^{3+} , Li^{2+} and Li^+ charge state in the accelerated stage. When the terminal voltage is increased, the yield for higher charge state increases: 2%, 49% and 49% for Li^{3+} , Li^{2+} and Li^+ ions from the total yield (0.07 nA, 1.5 nA and 1.48 nA of particle current, respectively). This result corresponds well with the published results in the work of the Kiisk group from Lund University [12]. The Li^{3+} ion beam would be preferable for RBS experiments due to higher energy, but the intensity of this beam is not sufficient (Fig. 3) to perform RBS experiments, and the penetration depth of Li^- ions of 1.5 MeV energy would be too low for a proper analysis of the layer structure.

RBS experiments with optical coating were performed using H^+ (1500 keV) and Li^{2+} (2800 keV) ions beams. In the spectra depicted in Fig. 4, the highest

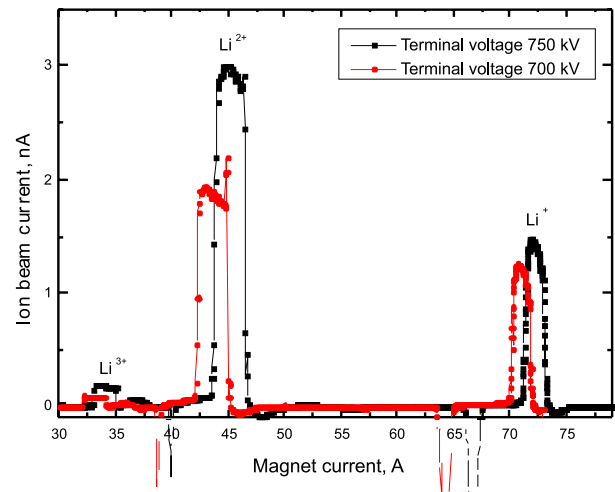


Fig. 3. Positive ion beam yields of different charge state from the Li target, measured by the Faraday cup in the accelerated stage.

Table. Molecular composition of optical layers.

Layer	Thickness, 10^{15} at/cm ²	Uncertainty, 10^{15} at/cm ²	SiO ₂ conc., %	Nb ₂ O ₅ conc., %
1	1570	7	100	0
2	468	2	37	63
3	1896	4	0	100
4	Substrate		100	0

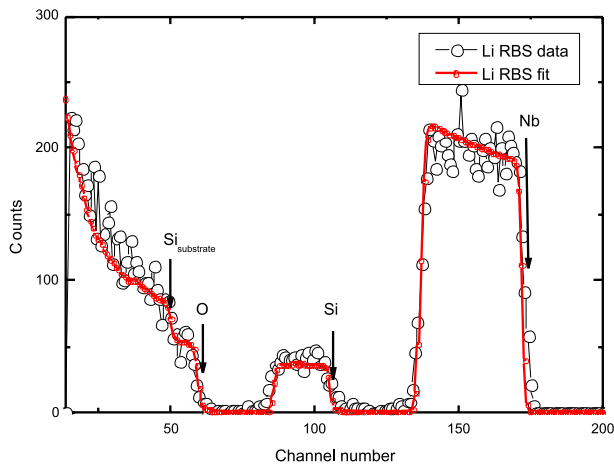


Fig. 4. The backscattered spectrum of the Li^{2+} ion beam of 2800 keV energy from optical coating and fitting with the *DataFurnace* code.

backscattered particle energy is recorded from niobium, the width of the peak is related to the thickness of the layer and the intensity is related to the concentration of the element. Although silicon dioxide is on the surface of the coating, due to lower kinematic factor backscattered particles have lower energy than those from niobium. Only minor difference can be seen in the lower energy part of the spectrum between energies of backscattered particles from oxygen in the coating layer and from silicon in the substrate. Due to multiple scattering and energy straggling in the coating it is difficult to estimate oxygen content in the coating. This is not the case when protons are used (Fig. 5): particles penetrate deeper into the substrate and only high energy part of the spectrum represents niobium, silicon and oxygen from the coating.

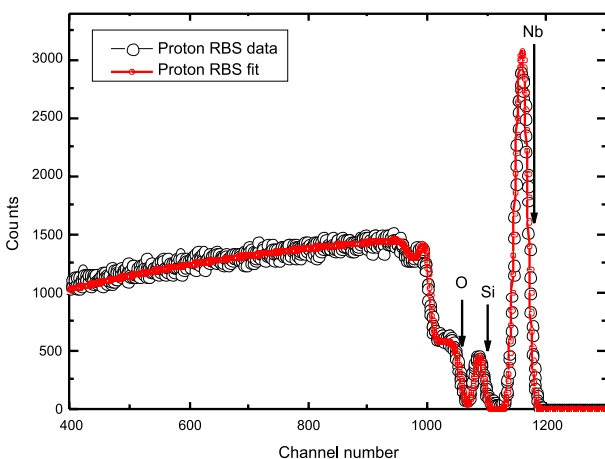


Fig. 5. The backscattered spectrum of the proton beam of 1500 keV energy from optical coating and fitting with the *DataFurnace* code.

Better mass separation can be seen when comparing the corresponding RBS spectra presented in Figs. 4 and 5. RBS spectra were collected at several Li ion beam energies: 2175 keV, 2275 keV, 2425 keV, 2550 keV, 2675 keV and 2800 keV. The collected charge on the sample slightly varied with the increasing terminal voltage and was 120 nC, 120 nC, 130 nC, 50 nC, 70 nC and 80 nC, respectively, for each energy beam. The *DataFurnace* ion beam analysis code was used to extract depth profiles from backscattered spectra [13]. By default *DataFurnace* uses ZBL2000 stopping powers [14]. Depth profiles of irradiated optical coating are presented in Figs. 6 and 7. Depth profiles in RBS are in 10^{15} at/cm² units. Values in nm use weighted averages of atomic densities and are meaningless except for pure layers. To get actual depth in nm, one needs to use different profiling techniques, such as SIMS or profiling with a lateral microbeam.

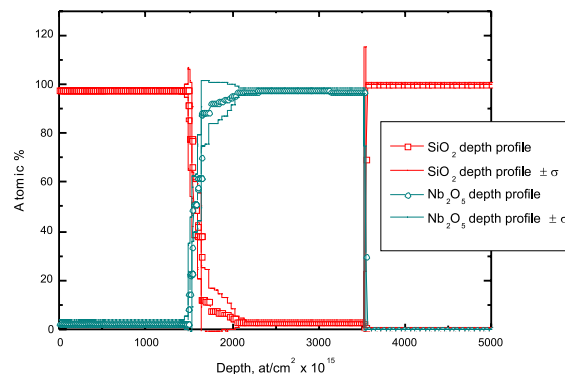


Fig. 6. The depth profile extracted from proton and lithium backscattered spectra with the *DataFurnace* code. The depicted profiles have uncertainty margins $\pm\sigma$, so the actual structure of layers can be imagined more clearly.

Energy resolution for protons is about 13 keV at 1.5 MeV and for lithium ions it is about 24 keV at the 2.8 MeV energy beam (1.85 times worse than H resolution). Although energy resolution is decreased almost 2 times when using lithium ions, the overall sensitivity due to better mass resolution increases. The Li ion beam offers better mass separation and surface depth resolution [9]. The maximal beam energy of Li^{2+} beam (2800 keV) is too low to estimate the oxygen content in the optical coating, but from Fig. 7 it is clear that the niobium content in the optical layer can be estimated with enhanced sensitivity. Analysis of RBS spectra obtained by the proton beam indicates possibility of layer intermixing, which was

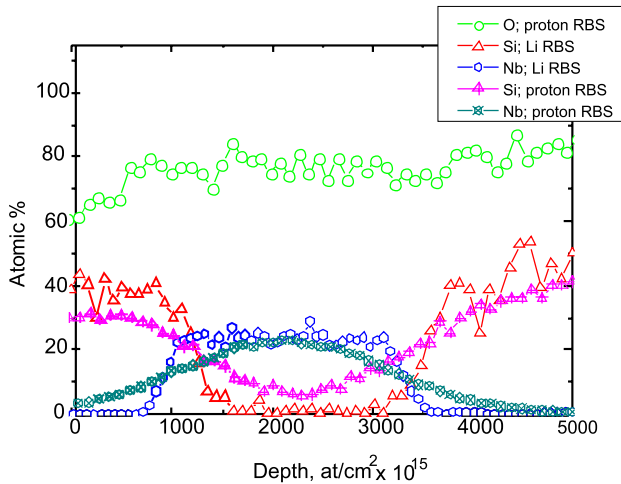


Fig. 7. The elemental depth profile extracted using the *DataFurnace* code from proton and lithium backscattered spectra separately/independently.

corrected by RBS spectra obtained by the Li beam. Four layers (last of them – fused silica substrate) were assigned to backscattered spectra by simultaneous fitting proton and lithium ion backscattered spectra with the *DataFurnace* code. Results of the analysis (Fig. 7) still indicate SiO_2 and Nb_2O_5 intermixing in the second layer of $468 \cdot 10^{15} \text{ at/cm}^2$ thickness, but the $+\sigma$ profile of SiO_2 and the $-\sigma$ profile of Nb_2O_5 are without layer intermixing. The composition of the optical coating is presented in the Table.

4. Conclusions

Li^- ions were successfully extracted from the Cs sputtering source and different charge state (Li^+ , Li^{2+} , Li^{3+}) of the positive Li ion beam was obtained after acceleration in a Tandetron 4110A accelerator. The most intensive Li^{2+} beam current reached up to 3 nA and was chosen to perform initial Li RBS experiments. The yield of the Li^{2+} ion beam was sufficient enough to estimate thicknesses of optical layers with enhanced precision, but not enough to eliminate uncertainties in the evaluation of elemental profiles of the optical coating. The performed analysis of RBS spectra shows that Nb_2O_5 layer intermixing with SiO_2 is doubtful within the margins of the error ($\pm\sigma$). Although energy resolution of the surface barrier detector for Li ions is worse than for protons, overall analytical application of RBS was improved due to better mass resolution, offered by the Li beam and simultaneous experimental data treatment with the *DataFurnace* code. Combined ion beam analysis using proton and Li produced in SNICS sources are viable alternatives to the helium ion beam that requires a duoplasmatron type source.

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CEZIO DULKINIMO ŠALTINYJE IŠGAUTO LI JONŲ PLUOŠTELIO PANAUDOJIMAS OPTINIŲ DANGŲ RBS ANALIZEI

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Santrauka

Kietųjų kūnų paviršių analizei gali būti naudojama Rezerfordo atgalinės sklaidos spektroskopija (RBS). Tandemo tipo dalelių greitintuvuose, kurie kompleksuojami su neigiamų jonų cezio dulkinimo šaltiniais, įprastai tokiems tyrimams naudojamo He⁻ pluoštelio išėiga yra labai maža. Norint praplėsti protonų RBS analitines galimybes, neigiamų jonų šaltinyje išgautas Li⁻ jonų srautas, kurio srovė siekė 32 nA. Greitinant Li⁻ jonus, užregistruotos visos galimos teigiamos ličio jonų krūvio būsenos (Li⁺, Li²⁺, Li³⁺). Atsižvelgiant į skirtingų būsenų įgreitintų jonų srovę bei dalelės siekį medžiagoje, RBS eksperimentams atlikti buvo pasirinktas Li²⁺ srautas ir iširta jonapluoščio dulkinimo būdu suformuota danga, sudaryta iš 200 nm SiO₂ ir

200 nm Nb₂O₅ sluoksnių, užgarintų ant amorfinio silicio padėklo. Nors detektoriaus energijos skyra naudojant Li pluoštelį suprastėja iki 24 keV, palyginti su 13 keV, kai naudojamas protonų pluoštelis, tačiau dėl geresnės masių skyros pasiekiami tikslesni rezultatai: niobio ir silicio elementiniai profiliai dangoje atskiriami aiškiau. Gautų Li²⁺ jonų dalelių srautas pakankamai intensyvus dangoje esančių sluoksnių storiams bei jų stochiometrijai nustatyti, tačiau nepakankamas elementinių profilių vertinimo paklaidoms sumažinti. SiO₂ ir Nb₂O₅ sluoksnių persiklojimas mažai tikėtinas dėl persiklojančių paklaidų ($\pm\sigma$). Naudojant *DataFurnace* jonų analizės programinį paketą, galima apjungti protonų bei ličio jonų RBS eksperimentus ir taip tiksliau įvertinti tiriamų objektų paviršinę sandarą.