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Preparation of isotopic antimony targets

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Abstract Thin self-supporting ^{123}Sb targets were needed for studies using GAMMASPHERE investigating transverse wobbling in the highly-deformed triaxial nucleus ^{135}Pr. The experiment was carried out using the ^{123}Sb(^{16}O,4n)^{135}Pr reaction with the 80 MeV ^{16}O beam provided by the ATLAS accelerator facility. In particle–particle coincidence measurements ^{121}Sb targets were irradiated with a 332 MeV ^{28}Si beam from ATLAS to measure evaporation residues and fission. The antimony targets were prepared self-supporting by the method of physical vapor deposition onto polished glass substrates or on various backing materials. Target thicknesses on the order of 500–1,000 μg/cm^2 were obtained and used for the experiments. Details of the target production and performance in beam will be discussed.

Keywords Antimony · Thermal evaporation · Backings · Target thickness

Introduction and motivation

The Physics Division of Argonne National Laboratory (ANL) maintains a heavy-ion accelerator user facility, ATLAS (Argonne Tandem Linac Accelerator System), available to provide beam times for approved nuclear physics experiments. Commissioned in 1985, it became the world's first superconducting accelerator based on niobium split-ring RF resonating structures [1]. Its continuous-wave (CW) operation can provide beams of any species from protons through uranium up to energies approaching 10 MeV per nucleon. Available instrumentation surrounding the target positions includes a large multi-purpose scattering chamber, a split-pole spectrograph, the HELIOS solenoidal spectrometer and GAMMASPHERE, a 110 Compton-suppressed HPGe detector array used for high resolution gamma-ray spectroscopy [2]. Built by collaboration at Lawrence Berkeley National Laboratory (LBNL) this array has been currently in place at ANL since 2003. Coupled with the Fragment Mass Analyzer (FMA) for measuring recoils based on their mass-to-charge ration (m/q) [3], this unique combination provides powerful instrumentation for a multitude of nuclear physics applications.

Recently, a study requiring self-supporting ^{121}Sb targets was undertaken to measure particle–particle coincidence events at low energy arising from non-fissile composite nuclei [4]. The experiment, investigating the evaporation residues from the reaction using a 332 MeV ^{28}Si beam, collected detailed coincidence data using the magnetic spectrograph and taking advantage of the excellent beam timing capabilities provided by ATLAS. Several particle and fission fragment detectors were mounted in the target scattering chamber attached to the spectrograph which allowed measurement of the evaporation residues and fission using light charged particles as a reaction probe.

A second experimental investigation demanding self-supporting isotopic antimony targets was to investigate the interesting phenomenon of traverse wobbling [5] in medium spin bands in the nucleus ^{135}Pr. Detailed spectroscopic

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studies requiring angular distribution measurements of the two-phonon wobbling band were carried out using GAMMASPHERE, employing the ^{123}Sb(^{16}O,4n)^{135}Pr reaction at a beam energy of 80 MeV to populate the medium spin bands in this nucleus. Using a low-mass projectile afforded an enhanced probability of populating the two-phonon wobbling band and allowed investigations of the anharmonicities expected near the critical spin.

A follow-up to these studies was performed at the Tata Institute of Fundamental Research in India using their 14UD Pelletron Accelerator and the Indian National Gamma Array (INGA) array of 20 Compton-suppressed clover HPGe detectors. For this experiment the recoils are stopped and therefore a thick gold backing was required for the ^{123}Sb targets.

Antimony target preparation

The element stibium, Sb, commonly known as antimony has been found in compounds, particularly in its native form stibnite (Sb_2S_3) since ancient times and in the metallic form beginning in the 17th century [6]. Metallic antimony is extremely brittle, bluish-white and crystalline in texture. Long a favorite of the alchemists, antimony has dual forms, the shiny metal luster as well as a gray powder. The black sulphide, which is the biblical Jezebel's kohl [7], can shift to orange with no special apparatus required, giving rise to its name *anti monos*, meaning against singleness. In modern times it is finding increasing use in semiconductors for making diodes, infrared detectors and other devices.

Web Elements provides the following physical properties for antimony: an atomic wt. of 121.76, m.p. 903.78 K and b.p. 1,860 K, with a density of 6.697 g/cc. Important for thin film deposition, antimony exhibits a substantial vapor pressure at low temperatures similar to those of Li and Sr. In addition, it has a thermal conductivity of 0.24 W/cm/K and therefore care should be taken regarding target heating under beam irradiation. Found in nature, antimony consists of two isotopes: ^{121}Sb at 57.21 % natural abundance and ^{123}Sb with a natural abundance of 42.79 %. These isotopes are available commercially in the metallic form.

Perusing the INTDS bibliography [8] we find but a few entries for antimony target preparation beginning with the early electroplating work onto Cu, Au, Ni and Fe backings done at AERE, Harwell [9]. Self-supporting foils were able to be obtained using dissolvable plastic substrates. Kelley and Dropesky [10] employed implantation using the Los Alamos National Laboratory isotope separator onto various metal backings. At Argonne, Thomas [11] produced Sb "sandwiched" targets on lead backings by the method of physical vapor deposition (PVD). While in München, Maier-Komor [12] exploited this technique for high efficiency sulphiding of antimony for preparing ($Sb_2^{36}S$,$Sb^{36}S$) targets. Single crystal growth of U and Pu antimony compounds was accomplished by levitation melting at IRMM [13]. Finally, Ueta and Engel [14] at the University of Sao Paulo reported difficulty with antimony target preparation onto gold backings due to low deposition efficiency.

Sb-121 self-supporting targets

Targets of ^{121}Sb of thicknesses from 300 to 500 μg/cm^2 were needed for experiments performed using the Enge split-pole spectrograph at the ATLAS facility. As the measurements involved characterization of the evaporation residues, it was highly desirable to produce self-supporting foils. While Ramsay [15] recommends CsI as a substrate, several alternates were also investigated, including Teepol (Sigma-Aldrich Chemie GmbH, Industriestrasse 25, CH-9471 Buchs SG, Switzerland), NaCl and betaine sucrose [16]. Practice evaporations were first undertaken within an NRC Model 3117 diffusion-pumped evaporator system (see, for example, Ref. [17]) using high-purity antimony (chunks) available commercially (Sigma-Aldrich Chemical, PO Box 14508, St. Louis, MO 63178 USA). The method employed was resistive heating using a closed Ta "pinhole" source boat with a current of approx. 100 A from a 10 V supply. The distance of the thermal source to the target substrates was 8.0 cm. It is important to note that due to the high vapor pressure, the evaporation rate must be kept extremely low. The deposition rate was monitored using a Kronos quartz crystal film thickness oscillator. The best results from these attempts yielded targets using CsI as a parting agent, the CsI first being deposited to 100 μg/cm^2 in a separate evaporation. The foils obtained had to be floated very slowly. Deposits on betaine however, could be floated quite easily and reproducibly, though the targets themselves did not exhibit a shiny metallic luster. These also had a tendency to rupture while drying.

Backed and supported Sb-121 targets

In order to assure successful target production, deposits on backings were also investigated. Gursky [17] recommends formvar as a support, though this was not attempted. The best method found was using collodion-dipped slides with Teepol as the release agent. These targets appeared nice and shiny and were easily floated and handled. Targets of 500 μg/cm^2 ^{121}Sb on 15 μg/cm^2 carbon backings were also prepared.

Sb-123 self-supporting targets

Targets of self-supporting ^{123}Sb of thicknesses from 500 to 1,000 μg/cm^2 were needed for high-spin gamma-ray

spectroscopy studies using GAMMASPHERE. Testing via the PVD method determined again that successful targets could be obtained using substrates of betaine and collodion-dipped Teepol slides. The collodion was then carefully removed from the target foil using several droplets of methanol applied gently to the back of the targets. RBS 35 (Pierce, PO Box 117, Rockford, IL 61105 USA) was also employed as a parting agent with moderate success. For these evaporations a Type C10-Ta crucible heated in a ME-1 crucible oven (RD Mathis Co., PO Box 92916, Long Beach, CA 90809 USA) was employed. The distance of the source to target slides was 8.0 cm. A photograph of the deposition set-up is shown in Fig. 1. It was realized after target manufacture that as an aid to target survivability in beam, a thin aluminum layer (15 μg/cm^2) should be applied to these targets to aid in heat conduction.

Thick gold backings

After the successful studies carried out using these self-supporting ^{123}Sb targets, a second experiment was to be undertaken requiring a thick backing in order to stop the recoiling nuclei. Two targets, previously prepared, were mounted in the Intlvac Model NANOCT2I1 cryo-pumped evaporator system [18], and a gold layer was slowly deposited. This method is highly inefficient, while the reverse, depositing onto the gold, would require additional isotope. A RD Mathis Type S19C-TA thermal source boat was employed with a distance to the targets of 6 cm. In addition to the quartz oscillator deposition monitor, a small glass witness disk was placed next to each target for additional gravimetric thickness determination.

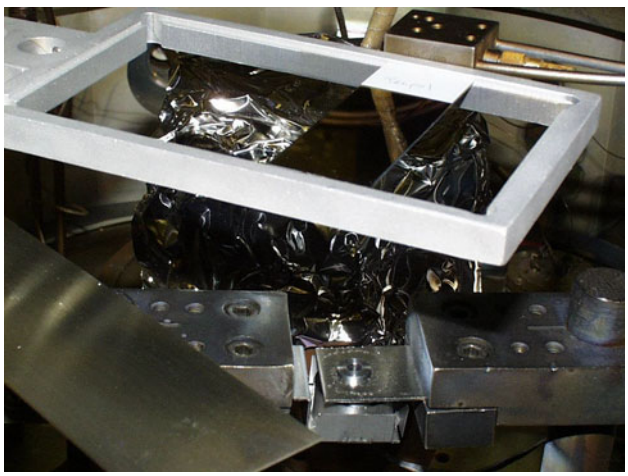

Fig. 1 Photograph showing the deposition set-up within the evaporator with the source crucible below the suspended glass slide substrate. Behind the slide can be seen the quartz crystal deposition monitor

Experimental results

The self-supporting and collodion supported ^{121}Sb targets were used in an ATLAS experiment employing a ^{28}Si beam at several energies from 238 to 420 MeV. The beam intensity was 5 pnA. The antimony isotope was obtained in the metallic form from the Isotope Sales Office at Oak Ridge National Laboratory (ORNL). Separated using the Calutrons, the enrichment provided was 99.57 % ^{121}Sb. Table 1 lists the various targets and thicknesses produced. These targets performed well in beam, including those prepared on collodion.

For a recent experiment carried out using GAMMASPHERE, a number of ^{123}Sb self-supported targets were prepared (see Table 1). For these as well as for the previous targets, the thickness values were determined by the method of α-particle energy loss [19]. The beam provided by the ATLAS accelerator was 80 MeV ^{16}O with a beam current of 3 pnA. The isotope was provided as a metal powder (ISOFLEX USA, PO Box 29475, San Francisco, CA 94129 USA) and had an enrichment of 98.28 % ^{123}Sb. An additional covering layer of 15 μg/cm^2 of aluminum had been applied in an effort to achieve additional thermal conductivity for the absorbed heat induced by the bombarding particle flux. All targets performed satisfactorily during the 4 days of experimental beam time.

Finally, an additional study was carried out at the Tata Institute of Fundamental Research with two of these targets having an additional thick gold backing deposit of 20 mg/cm^2 evaporated by PVD from a thermal source. An 80 MeV^{16}O beam was provided by their 14UD Pelletron Accelerator for these polarization measurements, employing the ^{123}Sb(^{16}O,4n)^{135}Pr reaction. The targets survived the beam bombardment well at an intensity of 3 pnA.

Table 1 Thicknesses of prepared ^{121}Sb and ^{123}Sb targets (self-supported or on various backings)

ISOTOPE	Target thickness (μg/cm^2)	Backing (thickness)
^{121}Sb	430	Collodion
	430	Collodion
	430	Collodion
	507	Carbon 16 μg/cm^2
	507	Carbon 16 μg/cm^2
^{123}Sb	499	
	553	
	674	Aluminum 14 μg/cm^2
	697	Aluminum 14 μg/cm^2
	761	Aluminum 14 μg/cm^2
	797	
	630	Gold 20.3 mg/cm^2
	634	Gold 22.8 mg/cm^2

Conclusion

In conclusion, self-supporting isotopic antimony targets were prepared by the method of vapor deposition from a thermal source for experiments carried out at using the ATLAS accelerator at ANL. Thicknesses of 0.4 mg/cm^2 ^{121}Sb were obtained, in some instances reinforced with a layer of collodion. Self-supporting targets of 0.5–0.8 mg/cm^2 ^{123}Sb were also produced for studies using GAMMASPHERE. In addition, ^{123}Sb targets were prepared with thick gold backings for experiments undertaken at the Tata Institute of Fundamental Research in Mumbai, India, using the INGA facility.

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