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Development of high-resolution ERDA with double MCP system and determination of detection limit for H and D

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ABSTRACT

A double MCP (micro channel plate) system is developed for the improvement of detection limit in HERDA (high-resolution elastic recoil detection analysis) for hydrogen (H) and its isotope. One MCP detects the recoils of hydrogen (or deuterium (D)), the other one detects the secondary electrons emitted from stopper foil. While the dark current noise is reduced down to 2%, the detection efficiency still remains as 80%. The improved detection limit for H and D achieved $\sim 6.3 \times 10^{19}$ atoms/cm^3 ($\sim$0.06 at.%) for 360 s and $\sim 2.3 \times 10^{20}$ atoms/cm^3 for 114 s, respectively. It is also found that the contributions of recoils of other light elements to the detection limit are crucial from the comparison with the previous study, in which both H and D are included in the samples. The developed system successfully determines the D depth profile in a SiO_2 thin film (t = 5 nm) caused by the D_2 annealing.

1. Introduction

High-resolution elastic recoil detection analysis (HERDA), which consists of a 90° magnetic spectrometer and a position sensitive detector [1], is a potential method to determine the absolute quantity of light elements on surfaces or at interfaces of devices. While HERDA shows the fairly good depth resolution as ~mono-atomic layer, the detection limit for the light elements still has margin to be improved. The dominant origins of the background in HERDA, which determine the detection limit of the method, are (1) the dark current in Micro Channel Plate (MCP) and (2) detected particles of non-interest and the stray particles. Hashimoto *et al.* successfully removed the background due to the dark current in MCP with an elegant way, in which they used two MCPs to coincidently detect the recoils and the secondary electron from the stopper foil. This method was firstly tried in high-resolution Rutherford backscattering spectrometry (HRBS) [2]. By taking the coincidence between the two MCPs, they reduced the detection limit for arsenic (As) implanted in a Si wafer from ~50 ppm down to ~10 ppm. The same method was also applied to HERDA by the same group for boron (B) detection in a Si wafer. The coincidence method improved the detection limit for B in Si from ~1 at.% down to 0.008 at.% [3].

One of the advantages of ion beam analysis, in particular of HERDA, is the absolute quantitativity for hydrogen (H) in solids. It would be a naive question how this coincidence measurement with the double MCP system improves the detection limit for hydrogen in HERDA. In the present paper, we report the improvement of the detection limit for hydrogen and deuterium (D) with the double MCP system. The details of the developed system and the detection efficiency depending on the coating materials on the stopper foil will be shown. Furthermore the obtained better detection limit for H and D and some applications to the samples, which include dilute H and D atoms, are discussed.

2. Experimentals

2.1. Detectors

Fig. 1(a) and (b) show the picture of the detection system and the simulation [4] of secondary electron tracks emitted from a Mylar film (stopper foil). The ions of recoils enter into the detection system from the left hand side of Fig. 1(b) as indicated by an arrow. The ions penetrate a Cu mesh and the Mylar film (t = 0.5 μm), and then they are detected by MCP-1. The transmission of the Cu mesh is ~90%. When an ion penetrates the Mylar film, some secondary electrons are emitted. The secondary electrons emitted backward are accelerated by the electric potential difference between the Cu mesh and the Mylar film. The Mylar film is given a metal (Al and Au) coating with the thickness

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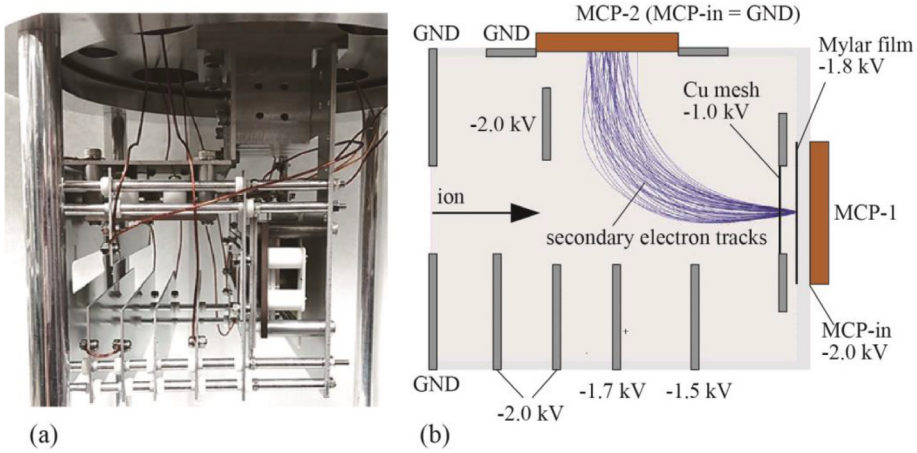

Fig. 1. (a) The picture of the developed detection system. (b) The schematics of the arrangement of electrodes and potentials. The simulations of the secondary electron tracks are shown by blue lines. The details of the beam line are described in the previous paper [5]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

of ~20 nm for two reasons. One is the formation of the uniform electric potential distribution, the other one is the enhancement of secondary electron emission. The later topic will be discussed in the section of results. The electric potential difference between the Mylar film and the surface of MCP-1 is put to avoid the detection of the forward emitted secondary electrons. The secondary electrons passed the Cu mesh are guided toward MCP-2 by the electrodes. The arrangement of the electrodes is similar to that designed by Hashimoto *et al.* [2], but the shape of each electrode is improved to the much simpler one. The potential put on each electrode is denoted in Fig. 1(b).

2.2. Experimental conditions and samples

The new detection system was installed in the detection chamber of HERDA beamline in 1 MV Tandetron in UTTAC (University of Tsukuba, Tandem Accelerator Complex). The details of the beam line are described in the previous paper [5]. The 0.5 MeV $^{16}O^{+}$ ion was used as the probe beam. The probe beam was shaped as 1 mm × 1 mm by a double slit system. The beam dosage was normalized by a beam chopper, which consists of a rotating ϕ0.2 Cu wire, and the beam current was calibrated by monitoring the current ratio of the chopper and a Faraday cup. The recoil angle was set at 30° with respect to the beam direction, while the beam incident angle was set at 75° from the surface normal.

For the determination of the detection limit of the developed system, hydrogenated amorphous carbon film (a-C:H film) grown with chemical vapor deposition (CVD) on a Si wafer was used as a standard sample. The a-C:H film thickness was ~160 nm, and the hydrogen concentration was determined as n_H ~3.5 × 10^{22} atoms/cm^3 by means of conventional RBS/ERDA system in our laboratory [5,6] and a simulation with SIMNRA [7]. For the observation of D, we used another amorphous carbon film including the both H and D (a-C:H + D). The concentration ratio of H and D was similarly determined by RBS/ERDA as ~2.9:1 [8]. We note that the stopper foil (Mylar) with the thickness of 1 μm was used for the D detection, while the stopper foil for the H detection has the thickness of 0.5 μm.

For the application of the developed system with the improved detection limit, we prepared well-defined SiO_2 films (t = 5 nm) with a dry process on a Si wafer. Apart from the as-grown Si oxide film as a reference, two films with subsequent annealing treatments at 673 K for 30 min in H_2 and/or D_2 with the atmospheric pressure are prepared.

3. Results and discussions

3.1. Detection efficiency depending on coating materials on Mylar film

One of the problems in the development of the double MCP system for H and/or D is how keeps the detection efficiency with suppressing the dark current noise in MCPs. In other words, we have to gain the secondary electrons emitted from the stopper foil as many as possible. In general, compared with other elements, the penetration of H and/or D through materials induces much smaller number of secondary electrons. This fact would significantly decrease the detection efficiency of H and D in the double MCP system, so that it is not straightforward to make the detection limit for H and D better.

We can simply calculate transmission of the secondary electrons by using the combination of the Cu mesh (~90%) and the aperture ratio of MCP-2 (~60%). The decrease of the detection efficiency due to the geometry of the parts would be thus simply estimated as ~54%, if the one penetration of H or D induced more less 1 secondary electron from the Mylar film. This value sounds rather serious to improve the detection limit. Nevertheless, Hashimoto *et al.* kept the almost same detection efficiency as well as the normal HRBS and HERDA [2,3], because the He^{+} ions and the B^{+} ions induced an enough number of the secondary elections in the penetration of the Mylar film.

One of the solutions to increase the secondary electrons is to search coating materials put on the stopper foil. In this sense, two meanings are required for the coating materials: (1) construction of uniform electric potential, (2) enhancement of secondary electron emission. As for the second meaning, while the theory of secondary electron emission is not simple, phenomenologically the yield of secondary electron depends on the stopping power of the target [9]. We compared Al and Au as coating materials, which have the stopping power for ~80 keV H^{+} of ~12 eV/Å and ~20 eV/Å, respectively [10]. Fig. 2 shows the HERDA spectra taken on the same a-C:H film with and without coincidence (the Mylar coating is Al) and with coincidence (the Mylar coating is Al and Au). We can see that the yield of detected recoils decreases down to ~80% with using the coincidence system. The value of ~80% is significantly larger than that estimated from the apparatus geometry ~54%. This means that the average of secondary electron emission from the stopper foil is ~2.1 for single H^{+} ion [9]. However the difference of the yield of recoils between the Al and Au coating on the Mylar film is negligible. This might be interpreted by the natural oxidation of the Al coat surface. The stopping power of Al_2O_3 for 80 keV H^{+} is 21 eV/Å is quite close to that of Au. After this confirmation on the coating material dependence of the detection efficiency, we have adopted Al for the coating material on the Mylar film.

Finally in this subsection, we mention some points, for which the readers might have questions. As for the oxidation of the Al coat on the Mylar film, we believe that only a few or several mono-layers of the Al coat surface are oxidized and the enough part in the depth direction of the coat is still metallic. One of the reasons is that we observed the good enough HERDA spectra also with MCP-2 (for the secondary electrons) quite similar to that with MCP-1 (for the H ions). This implies that the electric field guiding the secondary electrons to MCP-2 is well constructed as designed. Next, the detection efficiency of 80% for H, we