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Use of a duoplasmatron ion source for negative ion generation

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ABSTRACT

The use of electronegative species as primary ions considerably enhances the emission of positive secondary ions in SIMS. Considering furthermore that negative primary ions can be required due to instrumental configurations (e.g. the Cameca NanoSIMS 50 requires an opposite polarity of the primary and secondary ions), O^- ion bombardment is employed in SIMS analysis. These O^- ions are typically created in a duoplasmatron source, which suffers however from its low brightness and which is thus not suited for high resolution imaging applications. The development of new (electro)negative ion sources is thus necessary to optimize the analysis of electropositive elements in terms of lateral resolution and sensitivity.

In this paper, we present the performance of a duoplasmatron ion source generating F^-, Cl^-, Br^- and I^- ion beams. In particular, we experimentally determine on a dedicated test bench the brightness of the source in the F^-, Cl^-, Br^- and I^- modes as a function of the gas pressure, the magnetic field strength and the arc current in the source. The obtained results are compared to the performances of the duoplasmatron in the standard O^- mode. In this context, a five times higher brightness was found for F^- (200 A/cm^2 sr) compared to the standard O^- (42 A/cm^2 sr).

1. Introduction

Dynamic Secondary Ion Mass Spectrometry (D-SIMS) is an extremely powerful analysis technique for surfaces and thin films. The emission of secondary ions is very sensitive to the chemical state of the sample surface. An increase in the sensitivity of electropositive elements can be induced by bombardment with electronegative primary ions.

The optical configuration of the Cameca NanoSIMS 50 (superimposed axes of the primary and secondary ions) requires an opposite polarity of the primary and secondary ions.

For these reasons, O^- ion bombardment is employed on the NanoSIMS 50 for the analysis of positive secondary ions. These O^- ions are generated in a duoplasmatron ion source, which is based on the original design of Manfred von Ardenne [1]. The duoplasmatron ion source suffers however from an important drawback, especially in the O^- mode: its brightness B is low (B = 25–50 A/cm^2 sr [2]). Small beam diameters with sufficient ion currents for imaging applications can therefore not be obtained. The analytical potential of the NanoSIMS 50 in terms of lateral resolution is thus not available for positive secondary ions. This hampers a considerable number of applications of the NanoSIMS 50 in biology, medicine and material science for which the detection of (electro)positive elements is required.

In this paper, we study the performances of the duoplasmatron ion source run with different gas feeds in order to generate halogen ion beams other than O^-, namely F^-, Cl^-, Br^- and I^-. In contrast to earlier works aiming at the generation of halogen ions that used techniques such as the hot surface ionization or ion generation by RF plasma [3,4], we used an unmodified cold cathode duoplasmatron [5] from Cameca. General descriptions of negative ion formation can be found in [3,4,6]. The beam characteristics (ion current, beam diameter and brightness) are determined as a function of the plasma properties such as the magnetic field strength, the arc current and the pressure [7,8]. The obtained performances are compared to those obtained with an O^- ion beam.

2. Experimental

2.1. Generation of the ion beams in a cold cathode duoplasmatron

Start up problems and instabilities of the plasma in the duoplasmatron were noticed when using the halogen containing gases as reported in [9]. The use of argon as support gas did not significantly improve the stability of the plasma [6]. As the duoplasmatron ion source, which we used for this study, was initially conceived to work with oxygen, this support gas was found to be the best choice to stabilize the plasma.

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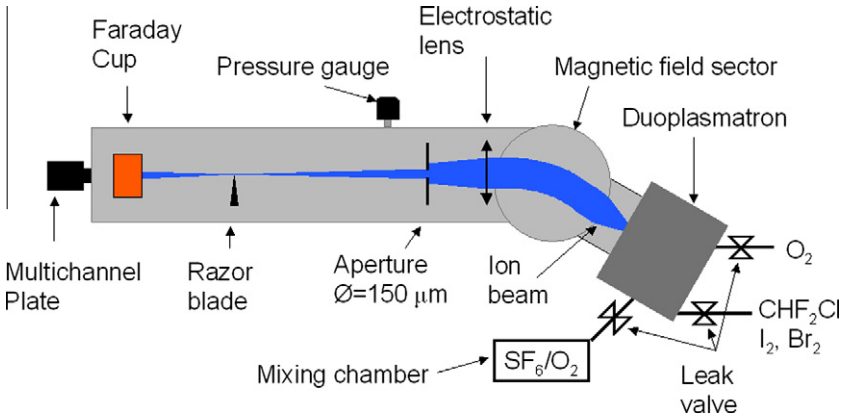

Fig. 1. Schematic overview of the test bench and ion beam trajectory.

To generate F^- ions, we used SF_6 gas in order to avoid toxic and corrosive F_2. The duoplasmatron was connected to a small chamber in which the O_2 and the SF_6 gases were mixed with an adjustable ratio (Fig. 1). The ratio of SF_6 was kept below 30%, otherwise the plasma became unstable [10]. The highest beam intensity was found for a mixing ratio of 80% O_2 and 20% SF_6. The total pressure in the chamber was 1 bar. It is supposed that the F^- ion formation in the O_2–SF_6 plasma is mainly induced by dissociative attachment of an electron to the SF_6 molecule. At electron energies below 2.5 eV, the F^- ion is the predominant negative ion fragment produced in this process. At lower energies, the SF_6 decays to SF_5^- ions and F atoms, as reported in [11].

For Cl^-, O_2 and CHF_2Cl (initial pressure: 1 bar), which was used to avoid toxic and corrosive Cl_2, were injected separately into the duoplasmatron (Fig. 1). The O_2 and CHF_2Cl gas flows were regulated with two leak valves installed on the gas inlet manifold of the duoplasmatron. Using this two valves configuration, reproducible (but unknown) CHF_2Cl/O_2 ratios could be obtained. It is supposed that the Cl^- ion is the dominant anion for electron attachment processes in the CHF_2Cl molecule because the electron affinity of the Cl atom generally exceeds the C–Cl bond dissociation energy [12]. Furthermore the molecule fragmentation into Cl^- and CHF_2^+ ions is predominant due to its low dissociation energy [13]. As chlorine has two isotopes with a ratio of 75.7% ^{35}Cl and 24.3% ^{37}Cl, only the $^{35}Cl^-$ ion current was used to determine the Cl^- ion brightness of the ion source.

For the generation of Br^-, liquid bromine was placed in a stainless steel reservoir and evaporated under vacuum conditions before being injected into the duoplasmatron. The reservoir was heated to ~60 °C during the process. A two valves configuration was used to adjust reproducible (but unknown) bromine/oxygen ratios. The negative ion formation of diatomic halogen molecules such as Br_2 is explained by dissociative attachment of low-energy electrons [14]. As the isotopic ratio of bromine is 50.6% ^{79}Br–49.4% ^{81}Br, only half of the Br^- ions extracted from the duoplasmatron were measured to determine the brightness. In our case we choose the $^{79}Br^-$ isotope to determine the brightness.

For I^-, we used the same set-up as for Br^- by replacing liquid Br_2 by solid I_2. Iodine ion generation using a duoplasmatron has been reported by Liebl [9]. As Iodine has only one isotope, all the I^- ions generated in the source were measured to determine the brightness.

As it is well known that the negative ion density of the plasma in a duoplasmatron is higher at the edges of the plasma, the extraction of the negative ions could be enhanced by shifting the intermediate electrode with respect to the axis of the anode aperture. As no correlation of the position of the intermediate electrode and the other plasma parameters was found, the optimal position of the intermediate electrode for the extraction of the ions was kept constant during the measurements of the ion current as a function of the afore mentioned parameters.

2.2. Test bench

The properties of the duoplasmatron generating the different halogen negative ions were experimentally determined on a test-bench (Fig. 1). The total gas pressure, the magnetic field used to compress the plasma near the extraction cathode and the arc current were the main parameters influencing the brightness of the ion source. As the total gas pressure in the duoplasmatron could not be measured directly, a pressure gauge connected to the test chamber was used as a reference. Likewise the measurement of the magnetic field strength at the plasma region was not possible. The intensity of the magnetic field, generated by a coil surrounding the plasma, was therefore referenced by the voltage applied to the coil V_{coil}, which was variable between 0 V and 100 V. The upper and lower limits of the applied voltage guaranteeing a stable plasma were found experimentally for the different plasma gases. The arc current in the plasma could be varied between 0 and 150 mA. The ions generated in the duoplasmatron, which was held at −10 kV, were extracted and accelerated to an energy of 10 keV by means of a grounded extraction electrode.

The ion beam was then mass filtered by a magnetic field sector according to the mass/charge ratio. At the exit of the magnetic field sector, the beam was focused into the so-called measurement plane by an electrostatic lens. As no shift of the crossover position along the beam axis was found when changing the different parameters of the ion source, the voltage applied onto the electrostatic lens was kept constant. Furthermore, the beam was limited by an aperture of 150 μm in diameter situated at 380 mm from the measurement plane. This aperture had the double role of defining the beam half angle α (2×10^{-4} rad in the described configuration) needed to calculate the brightness and minimizing spherical aberrations. The beam diameter d was determined in the measurement plane using a grounded razor blade mounted on a movable stage with micrometric resolution. The beam diameter was taken to be the distance between the positions of the razor blade shading the beam to 10% and 90% of its total ion intensity. The intensity I of the ion current was measured with a movable cylindrical Faraday cup with electron repelling electrode biased at −50 V, which was situated at 150 mm behind the razor blade and which was connected to a pico-amperemeter. A microchannel plate assembly placed behind the Faraday cup was used for the optical alignment