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Time-of-flight MeV-SIMS with beam induced secondary electron trigger

Martina Schulte-Borchers *, Max Döbeli, Arnold Milenko Müller, Matthias George, Hans-Arno Synal

Ion Beam Physics, ETH Zurich, Otto-Stern-Weg 5, CH-8093 Zürich, Switzerland

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ABSTRACT

A new Time-of-flight MeV Secondary Ion Mass Spectrometry (MeV-SIMS) setup was developed to be used with a capillary microprobe for molecular imaging with heavy primary ions at MeV energies. Due to the low output current of the ion collimating capillary a Time-of-flight (ToF) measurement method with high duty cycle is necessary. Secondary electrons from the sample surface and transmitted ions were studied as start signals. They enable measurements with a continuous primary beam and unpulsed ToF spectrometer. Tests with various primary ion beams and sample types have shown that a secondary electron signal is obtained from 30% to 40% of incident MeV particles. This provides a ToF start signal with considerably better time resolution than the one obtained from transmitted primary ions detected in a radiation hard gas ionization detector. Beam induced secondary electrons therefore allow for MeV-SIMS measurements with reasonable mass resolution at primary ion beam currents in the low fA range.

1. Introduction

Molecular imaging by mass spectrometric techniques is an indispensable tool in biomedical research (e.g. [1,2]). Secondary ion yields for macromolecules and large molecular fragments are strongly enhanced if fast and heavy primary ion beams are used in Secondary Ion Mass Spectrometry (SIMS) [3–5]. Nearly intact molecules have been reported to leave the surface under these conditions, which considerably facilitates identification of chemical species. In past years, the use of MeV ions for ToF-SIMS has been investigated for molecular imaging with very promising results and the technique has been applied to imaging of biological samples [6,7].

One major difficulty in MeV-SIMS for imaging is the limited focusing ability of typical magnetic quadrupole lenses for heavy and fast primary beams. To a certain extent electrostatic lenses can be applied at the expense of lower focusing quality. As an alternative to ion optical lens systems, a MeV-SIMS setup based on a capillary microprobe [8] was developed at ETH Zurich. Such a capillary acts as a collimator for a wide variety of primary beams including very heavy and fast ions, which are the most promising candidates to exploit the full potential of accelerator based MeV-SIMS. Beam spot diameters down to about 1 μm are possible, but as a result of the strong collimation the achieved beam current is relatively low. However, currents in the fA to pA range, usually obtained with capillaries, are sufficient for MeV-SIMS due to the high secondary ion yields.

For ToF mass spectrometry, on the other hand, a precise start signal as an estimate for impact time of the primary ion on the sample or a pulsed secondary mass spectrometer is necessary. Several ToF MeV-SIMS systems use a fast primary beam chopper to produce a start signal, e.g. [5,9,10]. The time resolution is then defined by the duration of the primary particle pulse, which can be as short as a few nanoseconds for modern devices [9]. A big drawback of all pulsed systems is their small duty cycle since the flight times of heavy secondary ions are several tens of microseconds and the waiting period between beam bunches has to be larger than this. For highly focused beams with low brightness and especially for low intensity primary species such as molecular or cluster ions, the DC currents generated in tandem accelerators are already reduced to the fA level due to the charge stripping process [11]. Additional beam pulsing results in count rates that are often too low for reasonable imaging applications. For a capillary microbeam the same problem applies, since the primary beam is additionally reduced by several orders of magnitude by collimation directly in front of the sample. Bunched spectrometers [12] are a valid alternative to primary beam pulsing as they can be used with DC primary beams and analyze secondary ions with dramatically improved duty cycle. They are, however, an important cost factor in an MeV-SIMS system and require careful adaptation in a customized environment due to their advanced sophistication. Therefore an alternative high duty cycle ToF trigger technique is desirable for imaging with a capillary microprobe. In early systems working with fission sources the recoil nucleus was used to pro-

* Corresponding author.
E-mail address: marschul@phys.ethz.ch (M. Schulte-Borchers).

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duce the ToF start signal [3] which is no option in accelerator based setups. For thin samples the transmitted primary ion can be detected [13]. However, this approach complicates sample preparation and limits the applicability of the technique. In addition, the primary ion detector can suffer from radiation damage very fast. In the case of negative secondary ion detection, the secondary electrons emitted by the sample surface can be used to produce a fast start signal in the stop detector of the ToF spectrometer [6,9]. Alternatively also photons produced by primary ion impact have been shown to present fast start signals for high resolution ToF measurements [14], but the design of an experiment with reliably high light detection efficiency is rather difficult and the background in spectra taken with a photon trigger is therefore increased.

In the present work we introduce the method of secondary electrons from the sample to be used as ToF start signal in positive ion MeV-SIMS. The setup was designed to allow extracting secondary electrons on one side of the sample, while positive secondary ions are extracted to the opposite side. The results of this new technique are compared with beam pulsing and transmitted ions as start signals in the same ToF MeV-SIMS system at the Laboratory of Ion Beam Physics at ETH Zurich. Performance of the setup is evaluated in terms of efficiency and time resolution with different start signals. The full details of the new MeV-SIMS set-up including imaging capabilities will be presented in a later publication.

2. Experimental

All experiments were conducted at ETH Zurich in the Laboratory of Ion Beam Physics, whose 6 MV tandem accelerator facility is used for Accelerator Mass Spectrometry (AMS) and materials analysis and modification [15,16]. The MeV-SIMS set-up is attached to a part of the AMS beam line which includes only electrostatic ion optical elements on the high energy side of the accelerator. This allows to use ion beams of high mass at low charge states, such as accelerated C_{60} clusters of up to 18 MeV [11] and monoatomic heavy ions like iodine or gold of up to nearly 80 MeV. In addition, beam pulsing is available from AMS applications [17]. For the higher timing requirements of MeV-SIMS, pulse length was adjusted to be as short as possible. Its performance will be evaluated later.

Dedicated for MeV-SIMS, a new beamline and UHV chamber (see Fig. 1) was added to the 0° port of the high energy AMS

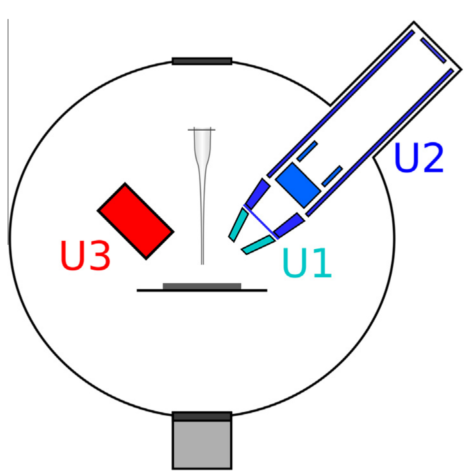

Fig. 1. Top view of the new MeV-SIMS chamber, with the primary beam coming from the top. The sample is placed on the piezo table in the middle. In front of the sample are an electron detector (left) with positive voltage U_3 and a Time-of-flight tube (right) with voltages U_1 and U_2. Behind the sample is a gas ionization detector.

magnet. Without baking, the chamber reaches a base pressure around $1 \cdot 10^{-7}$ mbar and includes a load lock for a tray with five samples. The setup is designed for imaging with a capillary microbeam and a piezo raster stage [8]. In order to simplify the study of different ToF trigger modes the capillary was not mounted for the measurements presented here.

For optimization of simultaneous positive secondary ion and secondary electron extraction the inner setup of the chamber was simulated in detail using SIMION ion trajectory software [18] version 8.0.3. Primary ions hit the sample along the surface normal, while the extracting nozzle of the ToF tube and the electron detector are on either side 45° from the primary beam in the horizontal plane. For electron extraction and detection a channeltron (SJUTS KBL505) is used with positive bias voltage of a few kV. The ToF spectrometer has a two stage extraction system with adjustable negative voltages of −1 to −2 kV at the nozzle and −5 to −6 kV at the second stage. Behind the two extraction stages a set of electrostatic XY steerers is placed for correction of the ion flight path direction. This is necessary due to the angled extraction. The whole flight tube has an isolated metal liner which can be held at a separate potential. For the experiments described here the liner was electrically connected to the second extraction stage. A chevron pair of micro channel plates (Hamamatsu F4293-07) with 20 mm active diameter serves as stop detector at the end of the 455 mm long flight tube. The positive voltage at the electron detector and the negative voltage at the ToF nozzle have to be carefully adjusted. Since the electric field direction is perpendicular to the surface for conducting samples the field strength at the point of impact of the primary ion beam on the sample has to be low to allow the simultaneous extraction of positively and negatively charged particles.

Furthermore a radiation hard gas ionization detector [19] is mounted in transmission geometry behind the sample for precise measurement of the incoming particle rate in the case of thin samples. Thick samples have to be periodically removed to measure the primary ion rate. The transmission detector can be used as an alternative start signal for thin samples.

The signals of all three detectors as well as the timing signal of the beam pulsing system are connected to a four channel CAEN DT5751 digitizer [20]. Due to the acquisition rate of 1 GS/s per channel, the intrinsic timing resolution without interpolation is 1 ns for acquired spectra. This fully digital approach allows the recording of timestamps (and optionally pulse heights) for all four signals in parallel via a custom made LabVIEW application in a straightforward manner. This application controls data acquisition and data analysis simultaneously, including the evaluation of multi-stop events.

3. Measurement modes

The system was designed to enable multiple modes for MeV-SIMS measurements, depending on the sample. This makes a thorough evaluation of the performance possible.

(a) Pulsing mode: Primary beam is pulsed and the pulsing signal is used as start for the Time-of-flight measurement.
(b) Electron start mode: Start time is measured by secondary electrons from the sample, which are detected nanoseconds after the primary ion hit.
(c) Transmission mode: For thin samples, primary ions can be detected in the transmission detector behind the sample.

In all cases, positive secondary ions are extracted into the nozzle of the Time-of-flight tube and detected in the MCP detector, independently of the start measurement mode. For thin samples