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First results on ion micro-tomography at LIPSION

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ABSTRACT

At the nanoprobe LIPSION ion micro-tomography can be used to determine the 3D distribution information of a sample's mass density and elemental composition. For ion micro-tomography the two analytical techniques scanning transmission ion microscopy tomography (STIM-T) and particle induced X-ray emission tomography (PIXE-T) are combined. The required data are collected in two consecutive series of measurements, during which the sample is rotated by 180°/360° in small steps. Because all ions have to traverse the sample, the upper limit of the sample size is given by the range of the ions in the material. The tomogram is obtained using the discrete image space reconstruction algorithm (DISRA) by Sakellariou (1997) [1]. This algorithm iteratively corrects a sketchy initial tomogram estimated from the experimental reconstruction – obtained by backprojection of filtered projections (BFP) – and an a priori elemental composition. The necessary correction factors are calculated comparing the reconstruction of the experimental data with the reconstruction of simulated data. For the simulated data sets of STIM projections and PIXE maps are computed from the tomogram. These data sets are proceeded with the BFP algorithm to get simulated reconstruction data. Using the DISRA for ion micro-tomography, one can benefit from the high resolution of STIM-T by transferring it to the elemental distribution given by PIXE-T. This article presents first results of this technique applied on a phantom at the LIPSION facility.

1. Introduction

The technique of ion micro-tomography (IMT) provides 3D information about a sample's mass density and elemental composition. Although applications still remain scarce, reaching a spatial resolution of less than a 1 μm this technique becomes more and more interesting for lots of applications in materials research as well as life sciences. Whereas some micro-tomographical techniques need to cut the sample into slices, the sole preparation step for IMT is to mount the sample freestanding, thus enabling a non-destructive analysis. The very recent development of limited angle tomography [2] with ion beams [3] even allows mounting the samples on thin foils or Si_3N_4-windows. Despite the larger efforts – compared to commercially available X-ray tomography devices – the additional elemental information as well as the better density resolution demonstrate the advantages of this technique.

Ion micro-tomography combines the two analytical techniques scanning transmission ion microscopy tomography (STIM-T) and particle induced X-ray emission tomography (PIXE-T). In combination with a computer algorithm suited to ion micro-tomography to reconstruct the data, the technique can benefit from the high spatial resolution of STIM-T by transferring it the elemental distribution given by PIXE-T. Nevertheless, the spatial resolution in STIM and PIXE mode need to be comparable. This is available at the high energy nanoprobe in the LIPSION laboratory [4,5].

The required data are collected in two consecutive series of measurements, during which the sample is rotated in small steps. The large range of high energy ions in matter allows the measurement of information deep inside the sample. The only restriction is that the ions have to traverse the sample, giving an upper limit of typically some tens of micrometers using 2.25 MeV protons.

Although the need for tomography was mainly driven by medical applications, the principle can easily be transferred to focused ion beam techniques. Starting with Cormack and Koehler in 1976 (128 MeV proton beam with a diameter of 2 mm) [6], this technique was progressed to ion micro beams by Pontau and Fischer [7,8]. In the 1990s computational power and the availability of sub-micron ion beams boost the possibilities of ion tomography, reaching practical biological applications [9–11]. This experience of other laboratories [12,13] (especially Melbourne) paved the way for STIM-T experiments at the Leipzig high energy nanoprobe LIPSION [11,14]. Whereas STIM-T nowadays is well established in several groups [12–16], there remained some obstacles regarding PIXE-T [17,18]. In contrast to STIM-T, where simple median filtering of few detected energy loss events suffices for adequate statistics, the physical phenomena in PIXE-T are more complex. Besides

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variations of the X-ray production cross section along an ion's trajectory, a major problem to obtain quantitative information in PIXE-T is the need to correct the X-ray yields because of attenuation and attenuation variations inside the sample. Since X-rays emitted from one point in the sample may undergo different attenuations on their path to the detector, the total detector solid angle needs to be subdivided for simulation purposes, thus taking into account the cone-beam geometry of the X-ray detection. To overcome this problem caused by the sample geometry and local composition variations in quantitative analysis, a successive approximation to the real composition of the sample is necessary. A reconstruction code including the most important physical effects as well as taking into consideration convergence criteria of the iteration process was proposed and tested by Sakellariou's DISRA [1,19].

This article enlightens the procedure to derive a 3D tomogram from the measured data using a phantom consisting of a ZnO micro-wire and oxide grains and introduces to the interactive, real-time 3D visualization possibilities of the IDL software [20].

2. Experimental and data processing

Due to different experimental setups for STIM and PIXE, the measurements for ion micro-tomography have to be conducted in two series of experiments. Thereby, the sample is rotated stepwise over an angular range of 180° (STIM) and 360° (PIXE). Each projection is done by scanning a parallel ion beam over the sample and detecting the desired information.

Mathematically, with the measurement a Radon transform of the specimen is performed. Thus, the original data can be obtained by the inverse operation. The method of choice is the BFP, an algorithm where the filter is Fourier transformed and then convoluted with the sinograms. Since discrete Fourier transformations always produce sidelobes a data set without disturbances is desired.

2.1. Sample preparation

The phantom was prepared as follows: Since the range of 2.25 MeV protons in ZnO is close to 30 μm, a thick ZnO micro-wire of about 13 μm with a thin, triangular flag at its side was chosen from a pile. Simple electrostatics attached the wire to a steel needle, where it was fixed under a stereo-microscope with varnish by hand using a wooden toothpick. The varnish (division of superconductivity and magnetism, Universität Leipzig) is soluble in alcohol, thus the consistency was easily controllable. A mixture of varnish with oxide grains (SiO_2, MnO_2, Co_3O_4, Fe_2O_3, and $K_2Cr_2O_7$) was prepared on an optical microscope slide. Again, using the toothpick a droplet of this mixture was put on the tip of the micro-wire. The needle is attached to a tomography sample holder and, with the help of the integrated microscope in the specimen chamber, is shifted and tilted to align the rotational axis with respect to the long axis of the specimen. During the experiment the projection angle of the specimen axis is controlled via a piezo driven rotary stage with an encoder resolution of 0.0001°. In case the specimen moves out of the field of view during rotation piezo driven linear stages in *x*- and *z*-direction with an encoder resolution of 0.1 μm and a stepper motor driven y stage (51,200 steps per revolution with 1 mm screw pitch) are used to correct the specimen position.

2.2. Data acquisition

Since the expected sample damage by a STIM-T experiment is negligible (fluence of ~50 protons per μm^2), the STIM projection set was recorded first. The residual energy of the protons after traversing the sample was detected using a windowless PIN photodiode (Hamamatsu 1223-01) with a resolution of 12 keV at full width at half-maximum (FWHM). The preamplifier Amptek A250 was mounted just beside the PIN-diode on the same circuit board directly into the vacuum chamber.

The STIM projection set was measured over a 180° angular range with a step width of 1° resulting in 181 projections. The scan width of $40 \times 40\ \mu m^2$ was covered by 128×128 pixels. As object diaphragm micro-slits were used, thus the measurements were performed at a count rate of the PIN-diode of about 3000–4000 ions per second. Recording five events per pixel, the pure measurement time of all projections took about an hour. As the photodiode suffers from damage during the experiment due to ion implantation into the depletion area, the diode needs to be shifted slightly from time to time to expose a fresh region.

The set of PIXE maps was recorded with an EG&G Ortec HPGe-detector of the IGLET-X series with a detector area size of 95 mm^2 at a crystal thickness of 10 mm. The measurement set covers an angular range of 360° in 22 steps (step width 16.363°). Each measurement was performed at a beam current of 200 pA to avoid electric charging of the sample. The scan width was again 40×40 μm now covered by 64×64 pixels. To apply the charge of 60 pC per pixel, the beam was scanned 200 times over the sample to reduce external influences and sample damage. This took about 20 min pure measurement time per map. The fast scan speed did not lead to any disturbances in the maps, due to a novel glass beam tube inside the scan coils. Since in usual beam tubes fast alternating magnetic fields induce eddy-currents the changing magnetic field is delayed. The new glass beam tube is coated with a silver layer inside. This layer avoids charging effects in the tube affecting the beam and due to linearly scratched paths circumvents the eddy-currents.

2.3. Pre-processing

At the entrance point to the DISRA iteration procedure fully preprocessed data are needed. That is, aligned energy loss data for STIM and aligned PIXE maps with element marks of the contained elements.

First, an overall energy spectrum of the STIM data is extracted from the event files, where each detected event is stored with its position and energy channel. Then, by identifying a maximum channel number, the spectra are cut off for further processing. Thus artificial counts caused by pile up are removed. Now, the projections are median filtered, excluding spurious and random events to contribute to the measured energy loss. With appropriate calibration parameters the channel data are converted to energy loss. Since the detector damage causes artifacts due to locally reduced sensitivity a threshold has been set. Pixel with an energy loss below this value are set to zero energy loss, thus artifacts due to detector damage are removed. From case to case one has to decide whether thresholding is necessary, because real, small energy loss data might get lost. If still some data artifacts exist in the projection data, they can be removed by an algorithm searching for spikes and plateaus. The parameters for this algorithm need to be adjusted manually to find the best result. On the one hand side, the corrections enhance the consistency of the reconstruction. On the other hand side, each correction step of the data may remove real data and reduce the accuracy of the reconstruction. Thus, one has to adjust the parameters carefully.

The alignment of the STIM projections has been done manually, since the automatic alignment program did not succeed. However, the automatic alignment of the sinograms using the center-of-mass shifted on a sine curve enhanced the quality of the reconstruction. This was the final step for the STIM data pre-processing.

Since the DISRA needs the data of each beam position consecutively, the list mode PIXE files had to be sorted, due to the recoding