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► To cite this version:

Mathieu Lalande, Pierre Salou, Arnaud Houel, Thierry Been, Thierry Birou, et al.. Nanoscale multiply charged focused ion beam platform for surface modification, implantation and analysis. *Review of Scientific Instruments*, 2022, 93 (4), pp.043703. 10.1063/5.0078914 . hal-03634593

HAL Id: hal-03634593

<https://hal.science/hal-03634593>

Submitted on 7 Apr 2022

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Nanoscale multiply charged focused ion beam platform for surface modification, implantation and analysis.

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(Dated: 7 February 2022)

The PELIICAEN (Platform for the Study of Ion Implantation Controlled and Analyzed at the Nanometric Scale) setup is a unique device, both for all of its in situ ultra-high vacuum equipment (focused ion beams (FIB) column, Secondary Electron Microscope (SEM), Atomic Force and Scanning Tunnelling Microscope (AFM/STM)), and for its nanostructuring performances on material. The setup has been recently equipped with its own electron cyclotron resonance ion sources, a new position-controlled platform using pneumatic vibration insulators and a fast pulsing device. Its performances were then deeply improved, providing access to a large choice of ions, an adjustable ion implantation depth up to few hundreds of nanometres, an image resolution down to 25 nm and an ion beam size on sample down to 100 nm. With all these equipment, the PELIICAEN setup is in the international foreground to perform and analyse ion implantation and surface modification.

I. INTRODUCTION

Nanoscience and nanotechnology are in the center of our technology and their progress has a direct impact on our society. Focused ion beam (FIB) is a crucial tool for the research and development of this technology, allowing elaboration, modification and characterization of material and components at the nanometric scale¹⁻⁴. The last decade has seen the emergence and the development of new domains in nanoscience such as spintronic^{5,6}, opto-electronic with the elaboration of quantum dots⁷⁻¹⁰ and single ion implantation¹¹⁻¹⁴. These new fields require localized implantation of elements at depths that are not commonly used in previous technology. Depending on the fluence, FIB irradiation can induce almost no damage to the irradiated area or make it completely amorphous since the ion current density directly affects the damage recovery dynamics of the sample^{15,16}. From the industrial point of view, the use of successive masks and implantation sequences could be easily adapted to these new needs. However, in the R&D phase and prototyping, the beam provided by today's FIB column shows limitations in terms of available ion species and energies¹. Liquid Metal Ion Source (LMIS) are used on conventional FIB, due to their ease of use and their outstanding brightness. Most of them are producing Ga⁺ ions, but with the development of mass-selecting FIB, separable alloys source became more and more popular, providing access to a larger range of ion species^{7,17}. Moreover, electron beam ion sources (EBIS) were tested on FIB column^{18,19}, producing tens of picoAmp beams of multiply charged ions with diameters of a few micrometers. The goal of the PELIICAEN project is to provide a platform for implantation, nanostructuring, and in situ characterization responding to the requirements of new nanotechnologies.

The use of electron cyclotron resonance ion sources (ECRIS) is appropriate for the production of a large range of

ions over a wide range of energies. This type of plasma source allows the production of ions from the injection of neutrals in many ways such as gas injection (atomic, molecular or metallic ions from volatile compounds), evaporation or sputtering from solid elements. If the plasma conditions allow successive ionization, the produced ions can be multiply charged, which allows reaching higher kinetic energy than singly charged ions for the same accelerating high voltage. Multiply charged ions can also transfer their potential energy, which is directly linked to their charge state (for example 8 keV for Xe²⁵⁺), to the sample surface^{20,21}. By coupling an ECRIS and a FIB column, the PELIICAEN set up provides access to a large range of ion beams, with adjustable ion implantation depths up to a few hundreds of nanometers, at a nanometric position precision.

A first experiment using multiply charged focused ion beams showed promising results²². In order to improve the beam size, to simplify the platform operation and to increase the available beam time, we designed a new autonomous FIB column based on a dedicated 10 GHz ECRIS.

II. ION SOURCE AND EVOL COLUMN

A. Setup description

The PELIICAEN setup is a platform dedicated to the study of surface modification. To do so, several equipments are placed in coincidence on the sample surface (12 mm working distance) in an ultra-high vacuum environment ($P < 5 \times 10^{-10}$ mbar): a Scanning Electron Microscope (SEM), a Secondary Electron Detector (SED), a probe microscope for Atomic Force and Scanning Tunneling Microscopy (AFM/STM) and a Focused Ion Beam (FIB) column (COBRA-FIB - Orsay Physics). The latter was previ-

ously attached to the ARIBE low energy (5-15 keV/q) beam line²³ at Grand Accélérateur National d'Ions Lourds (GANIL) in Caen (France)²².

This setup allowed the study of surface nanostructuring induced by multiply charged ions. However, the mechanical vibrations and electromagnetic background limited the achievable resolution. In order to overcome these limitations and to increase the available beam time, the layout of the setup was modified. The whole system was placed on a position-controlled platform using pneumatic vibration insulators. The COBRA-FIB column was upgraded into the EVOL column (by the company Orsay Physics) to host its own beam injector and a new fast pulsing device, for a 2 m total beamline length. Therefore, the maximum mechanical vibrations of the setup are now 1.5 and 20 $\mu\text{m/s}$ RMS respectively below and above 15 Hz, and the maximum electro-magnetic field is 0.5 mG peak to peak (50/60 Hz sinus).

The EVOL column is described on FIG. 1. One can see the beam injector and the beam pulsing part that were added to a standard gallium FIB column, COBRA-FIB. The beam injector is composed of a 10 GHz full permanent magnet Electron Cyclotron Ion Source (ECRIS), Nanogan²⁴, that can provide a wide range of ions species (H to Bi) by using different methods to inject neutrals (by gas injection, by evaporation in the source using a micro-oven, or for refractory elements by sputtering the element inside the plasma). The charge state of the produced ions start from singly charged to midsly charged ions (e.g. Ar^{8+} , Xe^{17+} , Bi^{19+}). The source can be biased from 2 to 30 kV, and a biased puller electrode allows the extraction at low energy. The produced beam is focused by an electrostatic lens into a Wien Filter for charge separation.

The Wien Filter has a 0.73 T magnetic field over approximately 60 mm that is produced by permanent magnets. It allows the beam selection at 30 keV for masses as low as He^{2+} and permits the separation of multiply charged heavy atoms. FIG. 2 shows a Bi beam spectrum measured at the entrance of the pulsing system. One can see the separation of the different charge states from Bi^{6+} to Bi^{16+} and other elements from water pollution and support gas (oxygen). Finally, an electrostatic quadrupole steerer can adjust the beam position at the entrance of the pulser.

The latter is composed of exchangeable collimators with 200 μm and 600 μm internal diameter apertures as well as a Faraday cup dedicated for the beam injector tuning. A focusing lens, an electrostatic octopole steerer and 10 μm to 800 μm exchangeable collimators shape the beam at the input of the blanking system. This electrostatic device, designed by Orsay Physics, is positioned upstream a third stage of exchangeable collimators in order to suppress any tail that can be observed on beam spots when the beam is pulsed by conventional blanking systems^{25,26}. Indeed, this effect can be predominant and problematic with either a fast pulsing rate or a short pixel exposure time relative to the blanking time. Thus, this device pulses the beam with a low voltage (15 V) and with a minimal pulse length of 1.5 ns at a repetition rate up to 1 MHz. The output beam, pulsed or not, is guided to the COBRA-FIB column for focusing and scanning. A wide range of focused ion beams has already been produced (H,

TABLE I. Resolution performances with several ions beams.

Beam	Energy	Resolution
He^+	20 keV	25 nm
Ar^+	20 keV	35 nm
O^{2+}	40 keV	36 nm
Xe^{2+}	40 keV	60 nm
Xe^{10+}	100 keV	90 nm*
Bi^{17+}	475 keV	200 nm*

* Best achieved resolution before optimisation of the ion transport

He, N, O, Ne, Ar, Xe, Ta, Bi) with an energy ranging from 2-532 keV.

B. Performances

Thanks to the modifications previously described, the setup capabilities are deeply improved. First of all, the new platform using pneumatic vibration insulators allows a good mechanical stability of the whole system, thus improving the SEM resolution down to 12 nm, as shown on FIG. 3. This high imaging resolution allows precise positioning and control of the sample to be modified by the ion beam. For resolution calculations we use the 20/80 Gaussian method.

Regarding the ion beam, the two most important improvements are the spot size and the image resolution. Resolutions obtained with different ions are presented in Table I. Thanks to the new ECRIS and the EVOL column, the resolution improved down to 25 nm with a He^+ beam at 20 keV (measured on silver paint and presented in FIG. 4), and 35 nm with an O^{2+} ions at 40 keV. Furthermore, the combination of the ECRIS and the EVOL column allows the focalisation of multiply charged ions. We thus already achieved resolutions of 90 nm for Xe^{10+} ions and 200 nm for Bi^{17+} ions, with further improvement since the ion transport was optimised after obtaining these two results. Such resolutions ensure a very good accuracy for experiments such as ion implantation, regardless of the ion nature or energy. As a direct consequence, the spot size is also improved down to 100 nm for Ar^+ beams at 20 keV (previously 470 nm with 90 keV O^{6+} ions on the ARIBE facility²²). This result was obtained by SEM measurement of a spot made on Silicon by a 4.17×10^{17} ions/cm² fluence.

The other improvements due to the installation of the ECRIS are listed in Table II in comparison with those obtained at the ARIBE facility. The ECRIS coupled with the EVOL column provides a strong increase of intensities and fluxes measured on the target, as well as a larger range of source voltages. The latter then giving access to a larger range of ions energies.

All of these improvements provide a really wide range of experiments with simply or multiply charged ions in a broad energy spectrum and with nanometric precision, such as ion implantation, amorphization and sputtering.

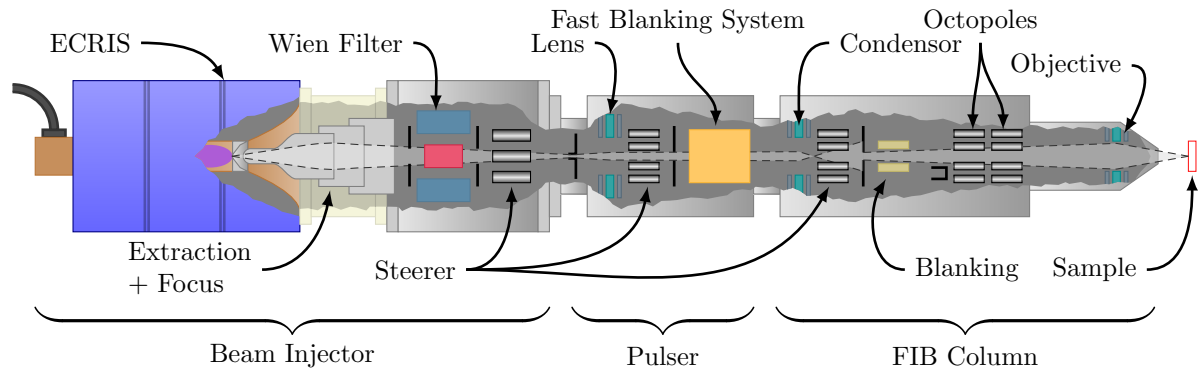


FIG. 1. Schematic of the EVOL FIB column.

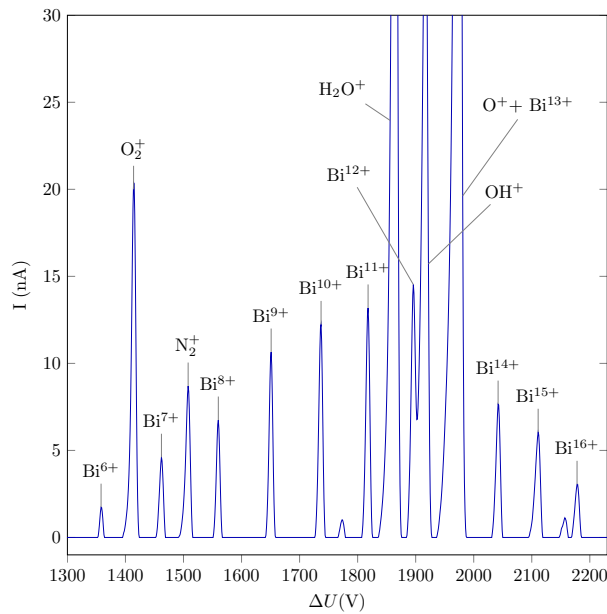
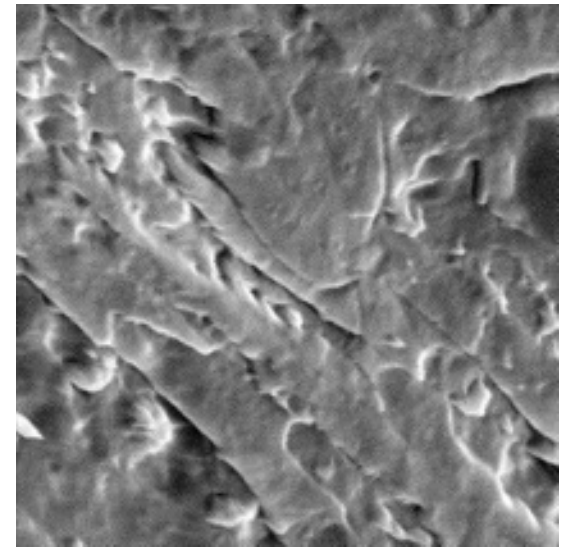
FIG. 2. Spectrum of a Bismuth beam measured at the entrance of the pulsing system. ΔU corresponds to the voltage difference applied on the Wien filter electrodes.

TABLE II. Comparison of the ARIBE and EVOL beamlines.

	ARIBE beamline	EVOL beamline
Best achieved spot size (nm)	470	100
Best resolution for ionic imaging (nm)	800	25
Maximal intensity on target (pAe)	200	5000
Maximal flux on target (ions/cm ² /s)	10 ¹³	10 ¹⁷
Source voltage range (kV)	5 – 17	2 – 30
Average beam adjustment time (h)	6	1
Beam availability dedicated to PELIICAEN (days/year)	20	200

FIG. 3. SEM image of a Mo sample surface with a 5 μm field of view at 10 keV with 12 nm of resolution.

C. Milling and ion implantation

In this section we will present typical examples of ion implantation and sputtering in order to illustrate the setup capabilities. FIG. 5 presents different milled patterns of $50 \times 50 \mu\text{m}$ length made by O^+ ions at 20 keV. The matrix of spots on the top left pattern presents the pulsing performances of the setup putting forward the precise ion milling provided by the EVOL column.

Experiments requiring precision can already benefit from real time in situ analysis inside the PELIICAEN setup²². However when additional analysis techniques are needed, samples can be moved to other platforms. This requires the surface modifications to be easily located. To do so, specific patterns can be milled to facilitate tracking and positioning of the sample in order to perform ex situ analysis. In FIG. 5 a $200 \times 200 \mu\text{m}$ cross was engraved by Ar^+ ions at 20 keV. This kind of cross can be easily found by several techniques, even by an optical microscope, allowing precise positioning

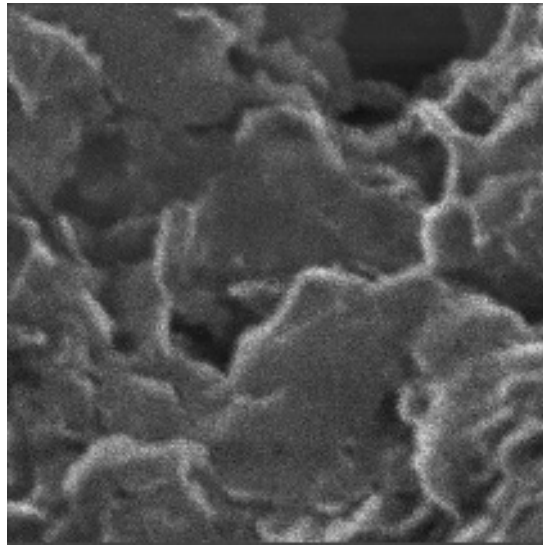


FIG. 4. He⁺ ion beam at 20 keV image of silver paint with 25 nm of resolution with a field of view of 4.1 μm .

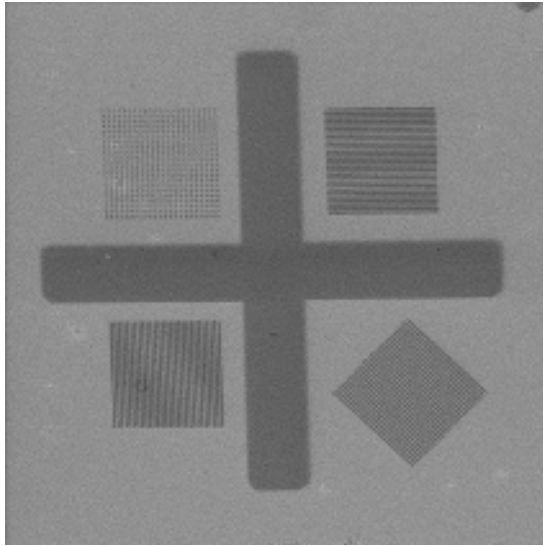


FIG. 5. SEM image (field of view of 250 μm) of a milling made on an LSMO sample by O⁺ ions at 20 keV with different patterns: matrix of dots on the top left, horizontal lines on the top right, vertical ones on the bottom left and oblique ones on the bottom right. The 200 \times 200 μm cross is engraved by Ar⁺ ions at 20 keV.

of the sample to locally analyse.

Ion sputtering can also be made on the sample surface, as the image from FIG. 6 shows. This cross was obtained with a 5 min irradiation by Ar⁺ ions at 20 keV on a Si sample, with a total fluence of 3.3×10^{18} ions/cm. The high fluxes produced by the ECRIS allow strong and deep milling in a short experiment time, for both marking and drilling.

These irradiations illustrate the performances of the innovative coupling of the ECRIS with the EVOL column, to get both accuracy for precise surface modification and high intensity to engrave position markers.

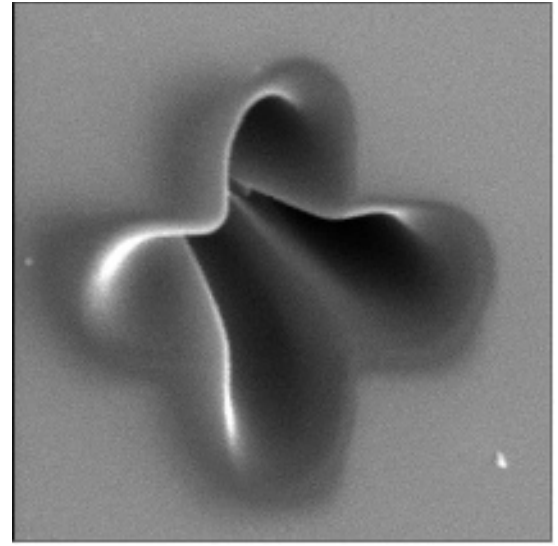


FIG. 6. SEM image (field of view of 9.2 μm) of a cross obtained by Ar⁺ ions milling at 20 keV on Si sample, with a fluence of 3.3×10^{18} ions/cm.

III. CONCLUSION AND OUTLOOK

The PELIICAEN setup has been recently upgraded with a fast pulsing device and an ECRIS, directly connected to its standard COBRA-FIB column. The system can now provide multiply charged ion beams of a broad range of species (H to Bi) and energies (2 to 532 keV), with fluxes up to 10^{17} ions/cm²/s, and focused down to a 100 nm spot size (20 keV Ar⁺). These performances allow implantation and structuration of materials with nanometric precision, strengthening studies on materials with fundamental research purposes, while paving the way for new technologies. As an example, one can cite quantum technologies (group IV and V elements implanted in diamond and in silicon, respectively), the development of single photon emission sources (implantation of rare earth elements), and microelectronics with its constantly increasing needs of precise nanostructuration.

In situ and real-time characterizations such as topographic, and conductivity imaging at the nanoscale can already be performed on the setup. An upgrade of the scanning probe microscope is in progress and will provide new modes of scanning measurements in order to open up to new applications, by providing access to new physical properties that can be modified by ion irradiations²⁷. One can cite magnetic, piezoresistance, kelvin probe and optical analysis. Moreover, a micro-channel plate detector has recently been added to the setup. We are currently working on the measurement of the backscattered ions energy by time of flight, in order to perform Low and Medium Energy Ions Scattering (LEIS/MEIS) analysis with the sub-micrometric resolution provided by the EVOL FIB column. This technique provides information on the chemical analysis of the sample extreme surface (few atomic layers). In a more distant future and with the addition of an ion extraction device near the sample, the PELIICAEN setup could also complete its already wide range of nano-characterizations

with the collection of secondary ions to perform nano-SIMS analysis.

IV. ACKNOWLEDGMENTS

These equipments were funded by the French National Research Agency (ANR) in the Peliicaen Project (ANR-12-NANO-008) and the LabCom CiCLOp (ANR-18-LCV3-0005-01), by Région Normandie RIN Recherche and European Union ERDF funds (EVOL-Peliicaen[17P04263]), and by the RADIATE project under the Grant Agreement 824096 from the EU Research and Innovation programme HORIZON 2020.

V. DATA AVAILABILITY

The data that supports the findings of this study are available within the article.

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