LIGHT DETECTION WITH SPECTRAL ANALYSIS AT THE LEGNARO NUCLEAR MICROPROBE: APPLICATIONS IN MATERIAL AND EARTH SCIENCES

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Abstract

Ion beam induced luminescence (IBIL or IL) is the emission of light as the result of energetic ions bombardment. Although the importance of IL technique, particularly if combined with other IBA (ion beam analytical) techniques, has been widely proven, very few apparatuses to analyse light emission spectra have been installed at the microbeam facilities.

We present here the new IL apparatus installed at the Legnaro (LNL) 2 MeV proton microbeam facility. This system allows for the spatial mapping of a monochromatic signal and the collection of the luminescence spectra of samples. Light collection is performed by using a retractable parabolic mirror located at a very short distance from the sample, with a small aperture to allow the ion beam to hit the sample. Accurate positioning of the retractable mirror directly coupled to a chamber mounted high-resolution monochromator allows for high light collection efficiency. The IL technique can thus be performed in low ion beam current experiments (e.g. IBICC) and to reduce the build-up of ion induced defects in luminescent materials. The design of the system assures that IL can be used with low beam currents (<1 pA) with the consequent reduction of the radiation damage, which often occurs during ionoluminescence measurements.

A summary of some meaningful results is presented. The combination of IL/PIXE was used to characterise natural silica glass, known as Libyan Desert Glass, and cubic BN grains; polycrystalline CVD diamond has been studied by a synergetic combination of IBICC/IL technique.

Scheme of IL apparatus

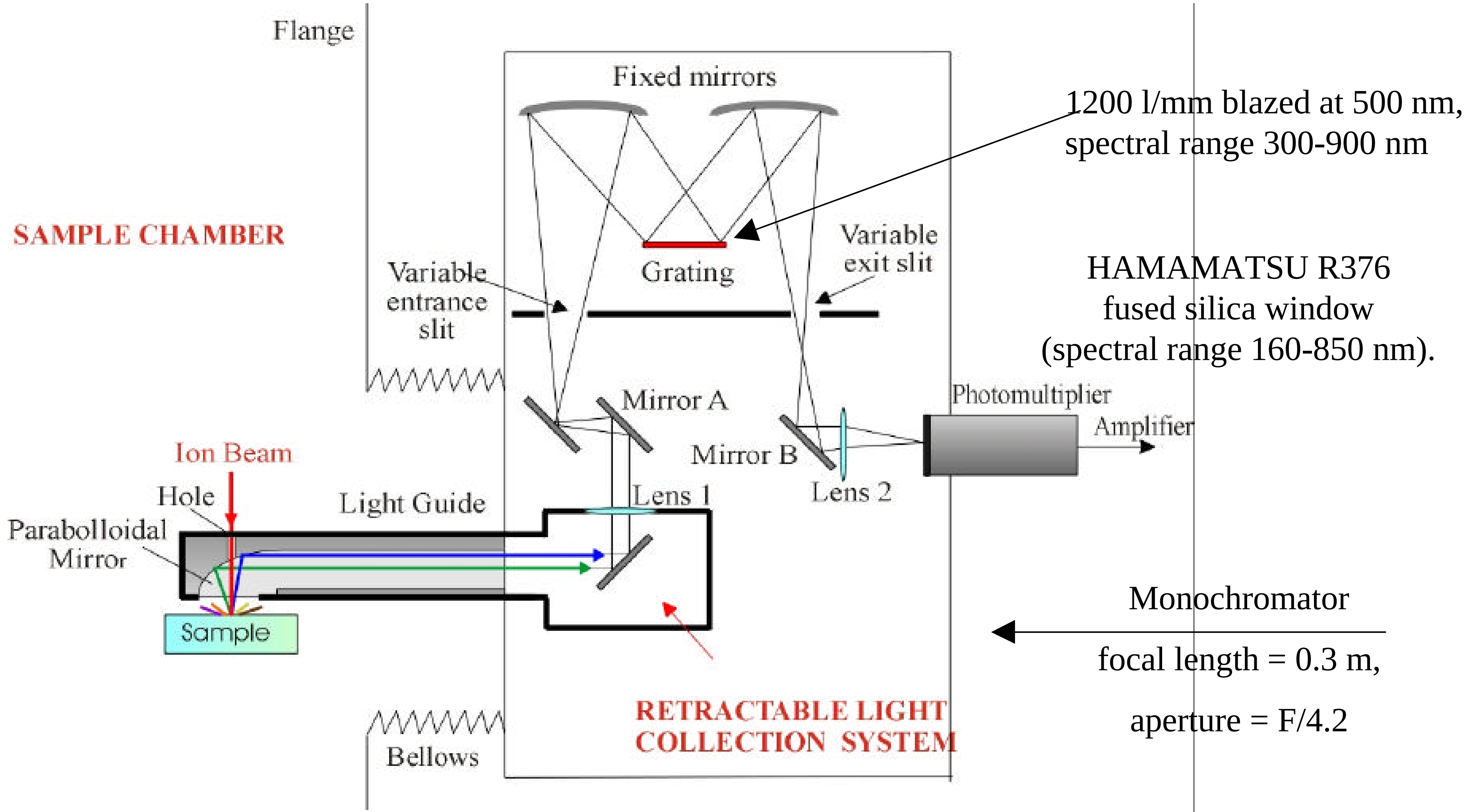


Fig.1. The optical collection system (schematic) in monochromatic mode. By rotating mirror A 90° counterclockwise and retracting mirror B, the system works in panchromatic mode by directly projecting the collected light from the input lens to the output lens.

The IL apparatus installed at LNL

It is a modified version of MONO-CL produced by Oxford Instruments for high resolution cathodoluminescence .

Light collection: retractable (up to 75 mm) module with a parabolic aluminium mirror (LCE:80%), waveguide, quartz lens.

Operation: monochromatic/panchromatic mode

Linear dispersion of the grating: 2 nm/mm; maximum resolution: 0.05 nm.

ACQUISITION

IL Spectroscopy: Oxford Instruments Photo Amplifier system (PA3) for power supplies, signal amplification and stepper motor drive electronics for the monochromator.

IL imaging: amplification electronic chain (based on the AMPTEK 250 charge sensitive preamplifier and AMPTEK 275 hybrid differential OP-amp) which is integrated into the existing microprobe data acquisition system.

The IL system can be easily moved from the ion microbeam analysis chamber and installed into a SEM (StereoScan 420, LEICA) for high-resolution cathodoluminescence (CL) imaging and spectroscopy.

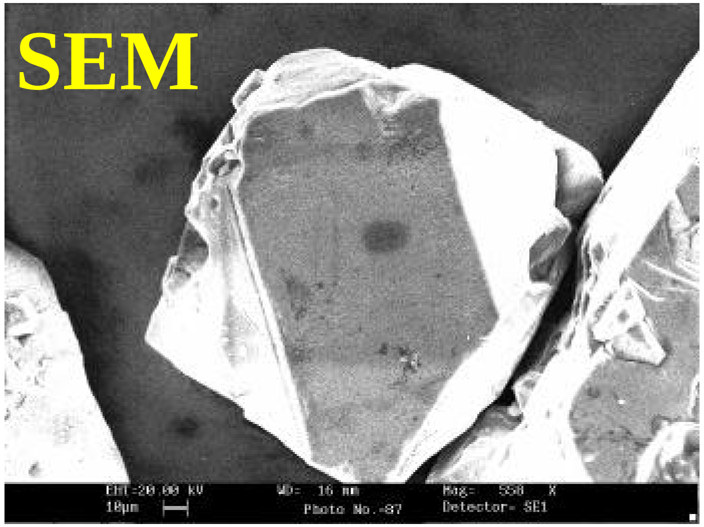
Cubic boron nitride (cBN)

Properties: cBN has physical and chemical properties isomorphic to those of diamond: **extreme hardness, wide optical gap (6 eV), chemical inertness, high electrical resistivity and high thermal conductivity.**

It can be doped both p and n type and shows high thermal stability at high temperature and a lower solubility on ferrous material than diamond.

Applications: hard coating and semiconductor layers in high temperature electronic devices.

Sample details: amber c-BN powders for grinding application obtained from graphitic BN (h-BN) by means of high pressure and high temperature techniques and using Ca_3N_2+LiF+NH_4F as catalyst.



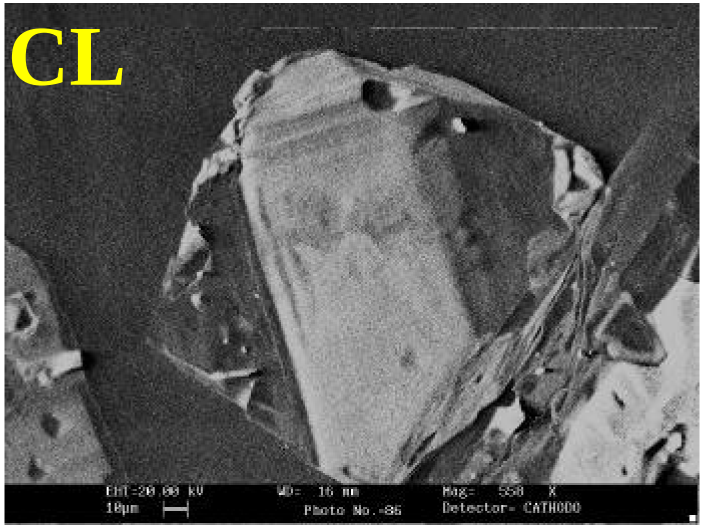


Fig.2

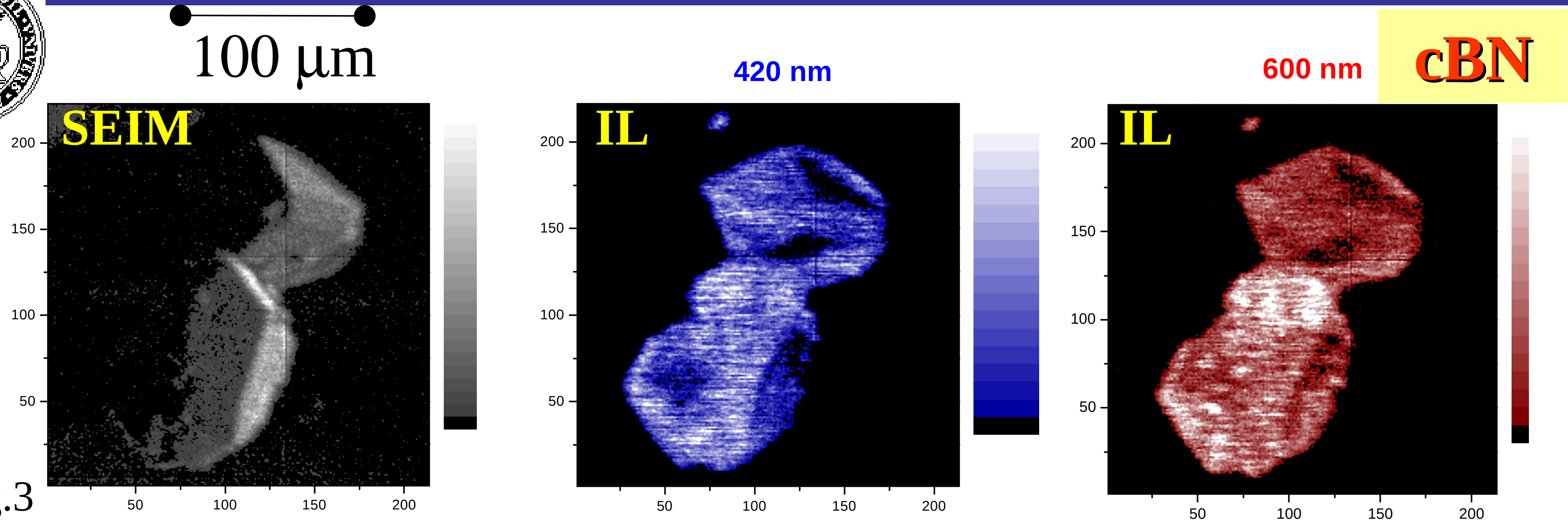


Fig.3

IL maps of a c-BN grain: (a) panchromatic, (b) at a fixed wavelength of 420 nm and (c) at 600 nm.

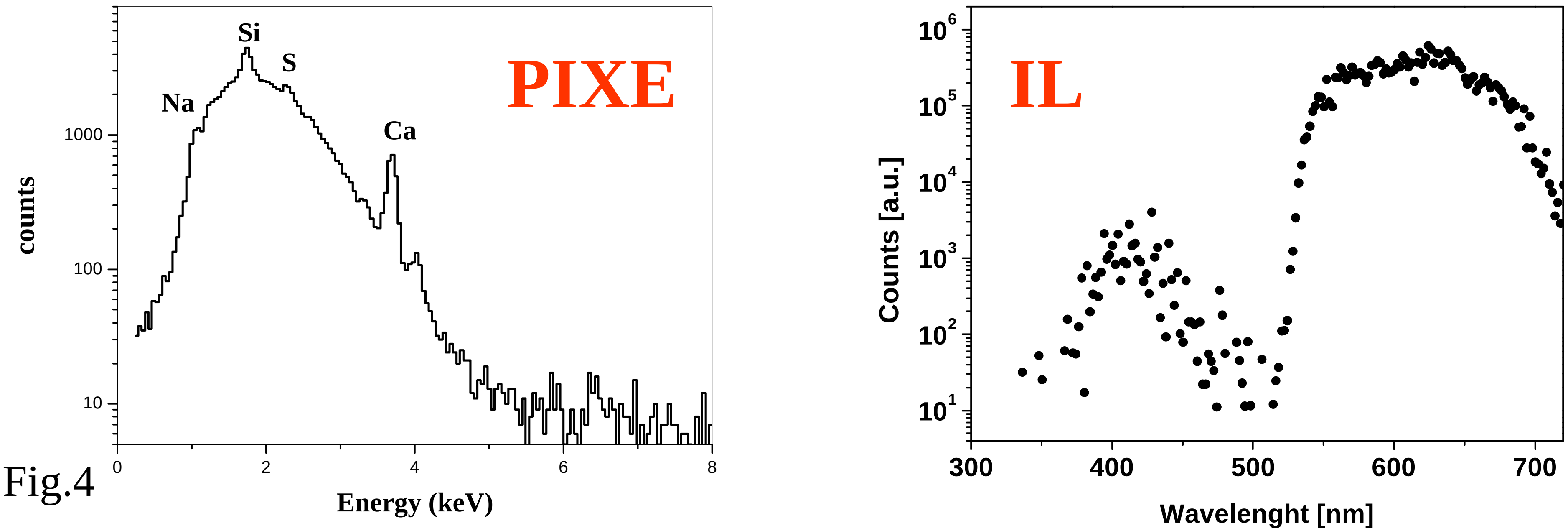
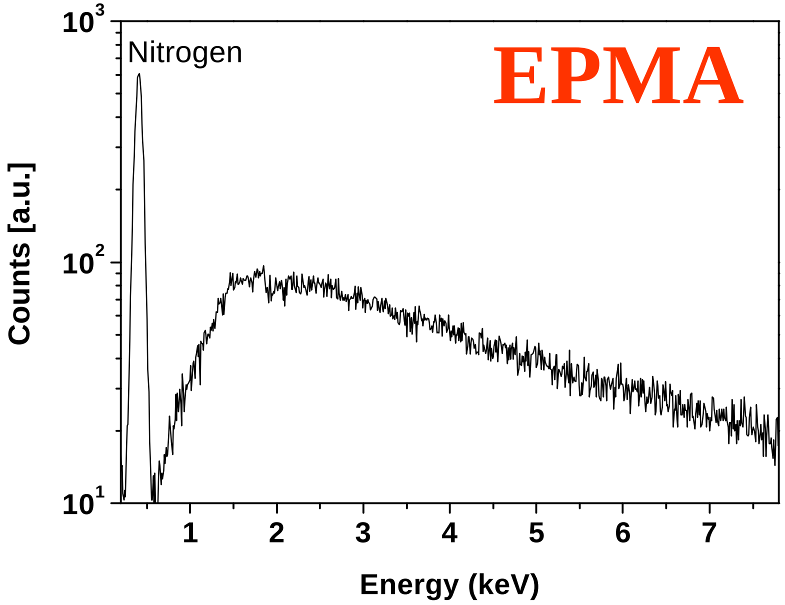
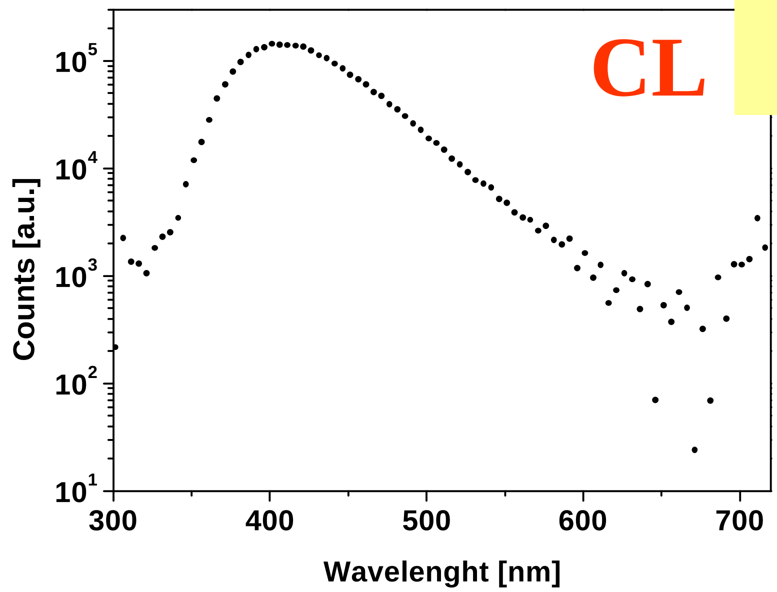


Fig.4

Fig.5





cBN

IL: two main bands centred at 420 nm and 600 nm.

CL: a broad band around 420 nm. The CL spectrum is the average over several grains.

PIXE

Ca: due to the precursor (Ca_3N_2) used during the synthesis.
Na, Si and Al are the main impurities of the originary h-BN.
S: is due to the tar in the carbon tube; traces of sulphate remain on the sink surface after the various chemical treatments for c-BN separation.

EPMA: only N and S

Interpretation: We can postulate that the luminescence is generated in the bulk of the material (the penetration range of protons is about 24 μm and the penetration of 20 keV electrons is of the order of some micrometers) and is probably related to Ca which acts as activator.

Libyan Desert Glass (LDG)

is a natural glass composed of nearly pure silica (98 wt %).
Discovered in 1932 by P.Clayton and L.Spencer in the Great Sand See in the Western Desert of Egypt, one of Earth's most remote and inhospitable regions.

The formation of this glass, because of its unusual composition, has been considered mysterious for a long time.
Probably it is the result of a meteorite impact in the Sahara sands, which ejected melted sand thrown into space and then shattered like broken glass upon re-impact.

"It seemed easier to assume that it had simply fallen from the sky"
(Spencer 1937).

A pectoral with the Sun god represented by both the scarab and the falcon which are fused as one. Above the scarab there is the bark of the Moon with the eye of Horus representing the Moon. Composed of gold, lapis lazuli, calcite, turquoise and **glass**, it was found on the mummy of Tutankhamun.

LDG raw material was employed to carve the central motif.

LDG

IL map centered at 400 nm

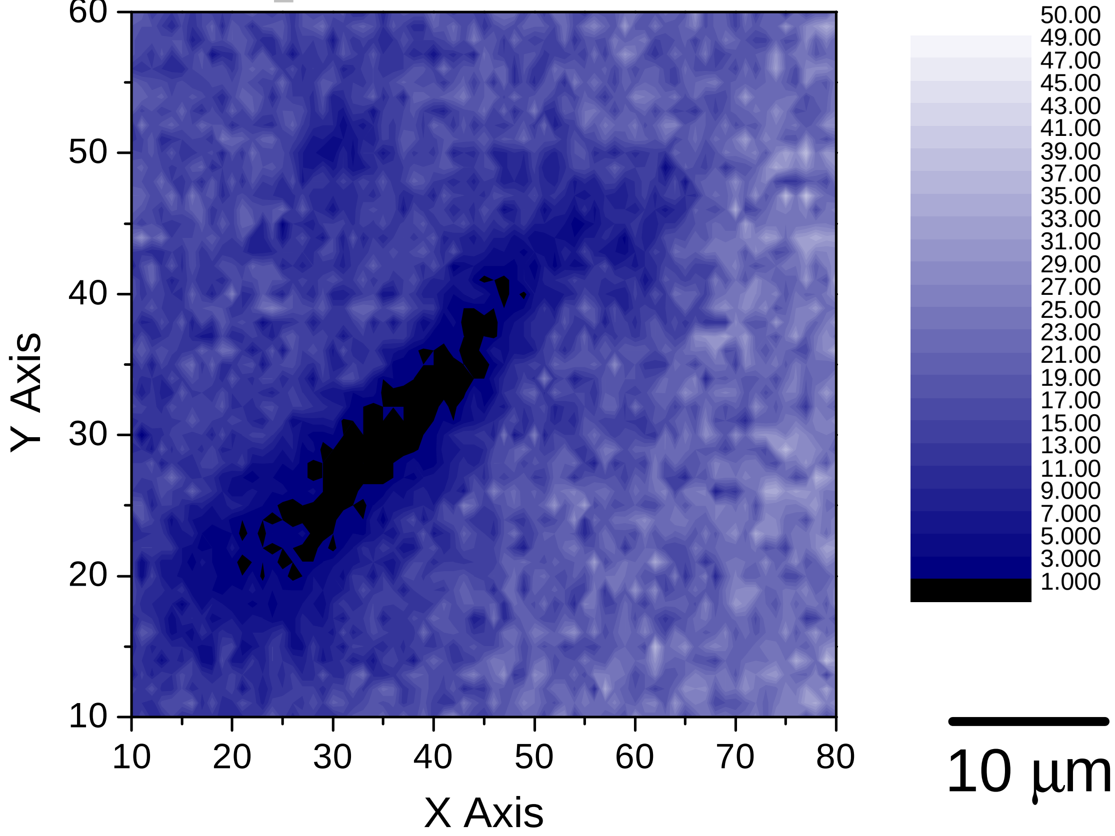


Fig.6

IL: the matrix shows an **intense blue luminescence**. The black little area in the centre of the map is much less luminescent than the matrix and its spectrum does not present the broad intense blue band that characterises the luminescence of LDG.

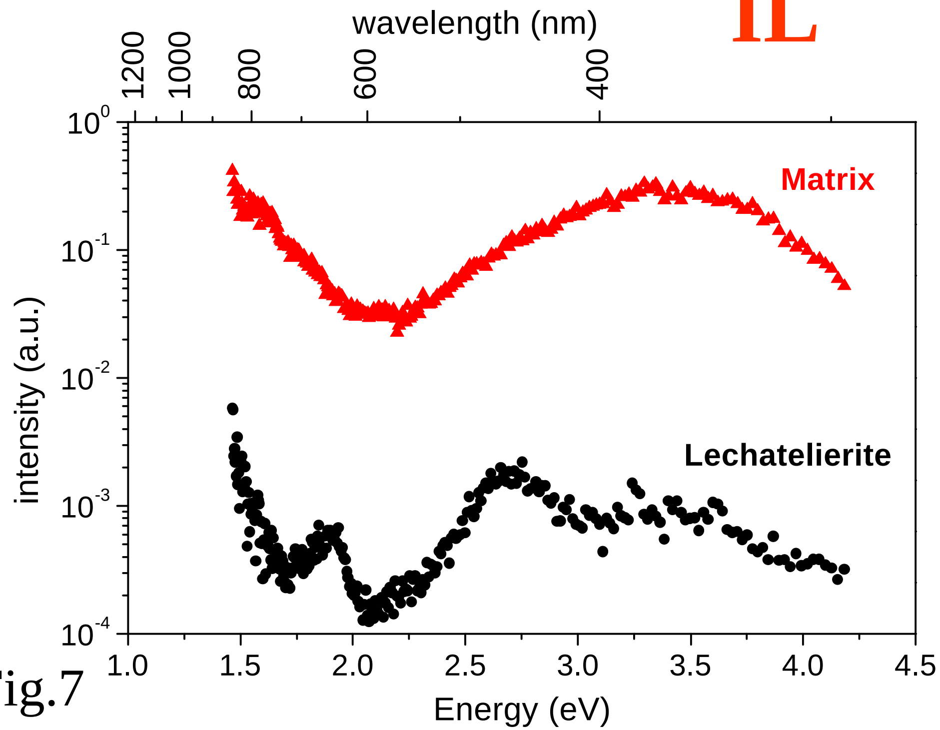
The colour scale is proportional to the pulse counts.

PIXE: **Matrix: Al, Ti, Fe, ; “Black region”: none**

LDG

Interpretation: the granular structure is composed of pure silica (lechatelierite protonucleus) enclosed in impure silica matrix. **Origin:** quartz grains of the precursor rock (sand or sandstone) vitrified at very high temperature, as suggested by a direct comparison with a local fulgurite and a substrate rock (Nubian Sandstone)[1]. It corroborates the interpretation of the LDG as the result of a meteorite impact in the Sahara sands, which ejected melted sand thrown into space and then shattered like broken glass upon re-impact.

IL



PIXE

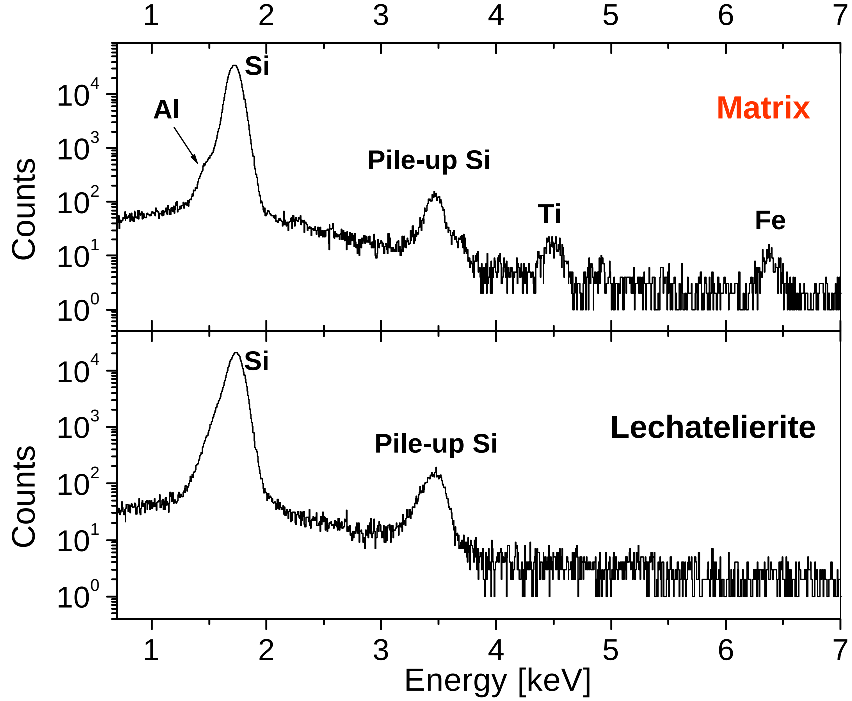


Fig.7

[1]Piacenza B: Proceedings of the “Silica ‘96” Meeting, Bologna 1997.

CVD diamond

A Norton CVD diamond detector 1x1x0.04 cm^3 was analysed by means of the coupled IBICC/IL technique. The IL spectrum of the sample (Fig.8) is peaked at the A-band of diamond. IBICC and IL panchromatic maps are then shown (Fig.9). Two different IBICC measurements were performed. The former (*IBICC Dark*) was carried out under dark conditions. The electrodes are localised at y=25 (anode, bias voltage +400 V) and y=125 (grounded). The growth surface is at the anode. The IBICC map shows the typical columnar structure illustrated in numerous previous papers. The colour scale is relevant to the average collection efficiency of each pixel. The collection efficiency profile of such a map, shown at the bottom, highlights the presence of a non-constant collection length as a function of depth. The peak occurring at channel 30 (which corresponds to the anode) can be attributed to a polarisation field generated at the anode, which reduces the applied electric field and, as a consequence, also the amplitude of the charge pulses collected at the electrode. Two solutions of such a problem have been adopted: pre-irradiating the sample before the measurement ("priming effect") or generating a space charge limited current from one of the two contacts. The latter solution was proposed by Koslov[2] and consists in injecting carriers that recombine with the trapped carriers of opposite sign.

The map *IBICC Light* shows that illuminating the sample during the measurement can produce a similar reduction of space charge.

[2]S.F.Koslov, IEEE Trans on Nucl. Science, Vol. NS22, (1975), 160

CVD diamond

The photocurrent induced by illumination was sufficient to passivate the charge trapped during the IBICC measurement (due to the high resisistivity of the material, the photocurrent induces a negligible electronic noise). The charge efficiency profile shows an almost linear behaviour, which is in good agreement with data obtained in pre-irradiated ("primed" or "pumped") samples.

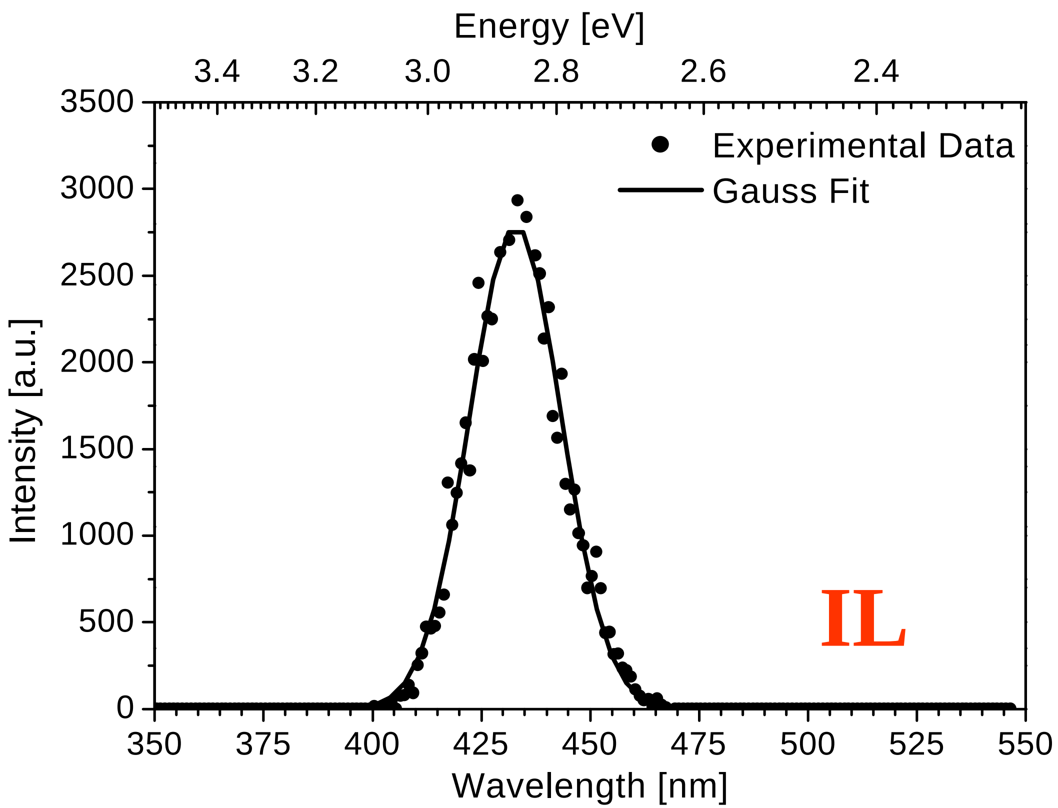
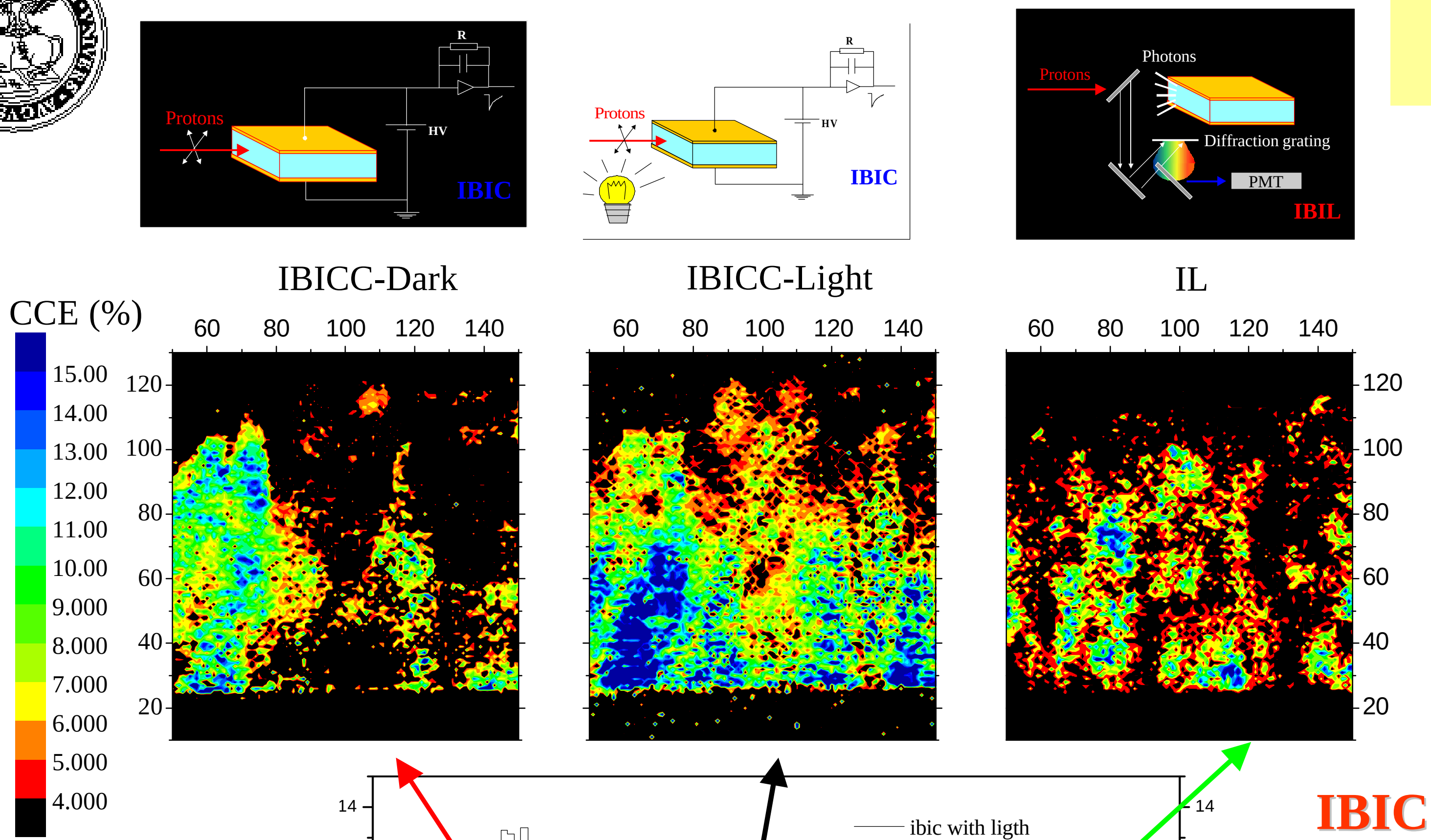


Fig.8

This behaviour is due to a linear reduction of the collection length vs the sample thickness.

It is interesting to compare the IBICC profiles with the IL profiles obtained in the same region using the same low ion microbeam current (lower than 1000 protons/s) in order not to damage the sample. A good level of complementarity can be found by comparing the first and the third map shown in fig.9. Moreover, the improvement of the charge collection efficiency is more evident in luminescent regions with respect to "dark" regions.

CVD diamond



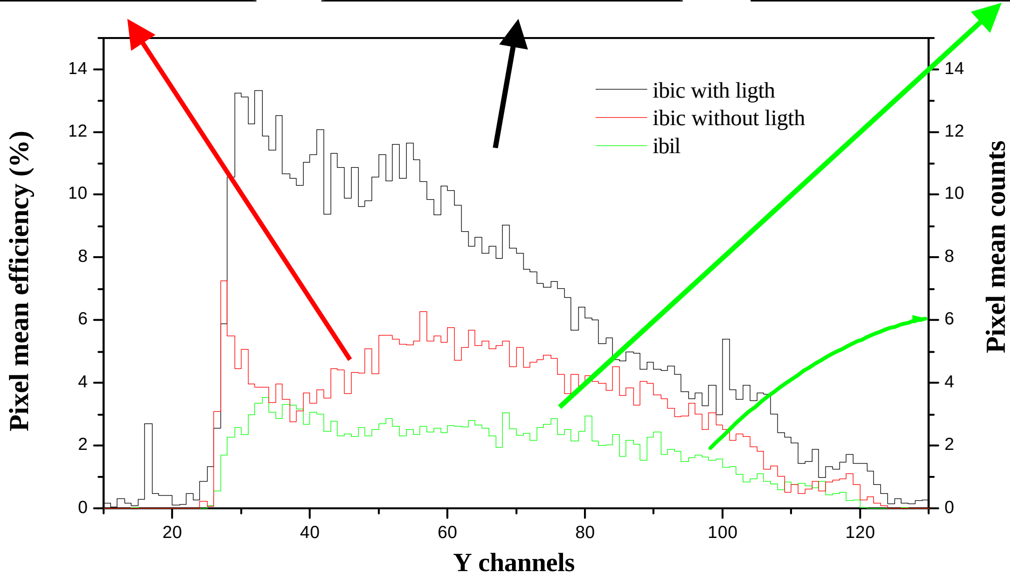


Fig.9

IBICC: Columnar structure; illumination improves the homogeneity of the lateral IBICC map.

IL: Only “blue A-Band”; the luminescent regions are related to regions where illumination is more effective on IBICC maps.

Conclusions

• Even considering IL a “semi-quantitative” IBA technique, the analytical combination of PIXE or IBICC and IL is useful to study structural and opto-electronic properties of materials and devices.

• The IL apparatus at the LNL allows the chromatic analysis of luminescence to be performed with high photon collection efficiency. This feature is necessary to perform coupled IBICC/IL measurements in wide band gap semiconductors as diamond.

• The more commonly used PIXE/IL coupling is suitable to individuate structures in the bulk of c-BN grains and lechatelierite protonuclei in Libyan desert glass which are not clearly visible using other "traditional" nuclear microbeam techniques.