# Condensed Matter Physics

*Frederico Gama Carvalho*

The Condensed Matter Physics Group aims to contribute to the development and characterisation of new materials, using radiation as a tool to investigate as well as to modify the material systems of interest. The group activities consist of materials processing, characterisation and research and also design, construction, installation and exploitation of experimental equipment for neutron scattering and X-ray diffraction. The materials systems under investigation include natural stones, such as topaz, high temperature materials and organic-inorganic hybrid materials. The instruments installed or in installation at the Research Reactor are a **Neutron Time-of-Flight Diffractometer, ETV, a Two-Axis Diffractometer** with a "banana" multidetector, DIDE, a **Small Angle Neutron Scattering Instrument**, EPA. At the High Temperature Materials Laboratory a **high-resolution X-ray diffractometer**, the Hotbird, was commissioned being now operational.

As concerns the installation of instruments, the main progress during 2000 occurred with DIDE. The remote control electronics, which was developed in collaboration with the LLB (Saclay), was received, and the shielding was installed and tested, which proved to be suitable. The assembly of the components required for the instrument to go into test phase is now being completed. Also related with instrumentation, a new project was approved and funded by the national **Foundation for Science and Technology** to construct a prototype equipment that will optimise the performance of small angle neutron scattering instruments.

The Group's activity has been developed by means of a dynamic interaction with the international neutron scattering community and the possibility of access to higher flux neutron sources. During 2000 neutron scattering measurements have been performed at **LLB (Saclay)** and a new collaboration project started with the **Budapest Neutron Centre** in the framework of a Co-ordinated Research Project launched by the International Atomic Energy Agency.

2000 was the first year of activity of the **High Temperature Materials Laboratory, MA$^3$T**. After the installation of the high-resolution X-ray diffractometer, the Hotbird, in the end of 1999, some additional hardware and software have been developed. Some were centered in tuning and improving performance, e.g. background reduction, developing software procedures, etc. However, one deserves emphasis, the addition of a crystal analyser as the third axis of the diffractometer. Now the Hotbird can operate in two different geometries: double-crystal and triple-axis diffractometer.

The Hotbird has been intensively used to respond to the demands of beam time from local and external research groups, to solve specific problems that cannot be tackled by conventional diffractometers, e.g. nanometer thick films that require grazing incidence geometry, etc.

A large part of the work occursed within collaboration projects with Portuguese and foreign University groups. Work on single crystalline materials was centered on semiconductor devices (thin multilayers, optoelectronic devices) and high-temperature aeronautical coatings. Several polycrystalline materials have been studied, namely magnetic multilayer sensors, nano-precipitates in implanted materials, ceramic powders, natural ornamental stones, etc.

# Research Team

## Researchers

- F.Carvalho (**Group Leader**) (Coordinator Researcher) (90%)
- J.Salgado (Coordinator Researcher) (30%)
- F.M.A. Margaça (Principal Researcher)
- A.N. Falcão (Aux. Researcher)
- A.D. Sequeira (Aux. Researcher)

## Students

- Pedro Ferreira (**PhD** Student)
- Nuno Franco (**MSc** Student)
- Iiro Rihimaki(**MSc** Student)
- Alexandre Santos
- Joana Santos Sousa (BIC PRAXIS XXI)
- José António Saraiva

- José Luís Sequeira (undergraduate students)
- David Silva (undergraduate students)
- Miguel Carrapiço (undergraduate students)

# Publications

| | |
|---|---|
| Journals: | 7 and 4 in press |
| Proceedings: | 1 in press |
| Conf. Communications: | 8 |

# Funding

| | | $\times 10^3$ PTE |
|---|---|---|
| **Research Projects:** | [a] | 5881 |
| **ITN:** | [b] | 1121 |
| **TOTAL:** | | 7002 |

**(a)** $\times 10^3$ PTE

- Microstructure Investigation of Organic-Inorganic Hybrid Materials (PRAXIS/P/CTM/12212/98) (Mar. 1999-Mar. 2001) ($14\,000 \times 10^3$ PTE $\rightarrow$ ITN/$7620 \times 10^3$)
  ITN/Co-ordinator**: Fernanda Margaça**, Partner: Univ. Aveiro (M.I. Salvado)..... 4200
- Study and Design of a New Generation of SANS Instruments: Smaller, Modular, with Enhanced Capabilities and Low Servicing Requirements (IAEA/11361/Regular Budget Fund) (ITN/5000 USD - Renewed annually).
  ITN/Co-ordinator**: Frederico Carvalho** Partner: Research Institute for Solid State Physics and Optics, Budapest, Hungary (Lazlo Cser)..... 567
- Characterisation and Modelling of Physical Properties of High-Temperature Coated Superalloys (PRAXIS/PCEX/P/FIS/21/96) (1997-2001) ($40\,000 \times 10^3$ PTE $\rightarrow$ ITN/$33\,900 \times 10^3$ PTE)
  ITN/Co-ordinator**: António Sequeira**, Partner: IST (Rui Vilar) ..... 1114
- Development of a Variable Geometry Neutron Multichannel Collimator (PROCTI/1999/FIS/34449) (Oct.2000- Oct. 2002) ($6000 \times 10^3$ PTE)
  ITN/Co-ordinator**: A.N. Falcão** ..... —
- Electrical Assisted LFZ Processing for Tailoring Textured Ceramic Materials (CTM/35492/99-00) (2000-2003) ($15\,000 \times 10^3$ PTE $\rightarrow$ ITN/$1000 \times 10^3$ PTE)
  Co-ordinator: Florinda Mendes da Costa (Univ. Aveiro), Partners: ITN (**António Sequeira**)..... —
- Neutron Beam Time Utilisation via UE 5th R&D Framework Programme "Human Potential-Access to Research Infrastructures" (one week of beam time for two researchers)

[b] From the Services of the Nuclear Instruments and Methods Group.

# Compositional Dependence of the Strain Free Optical Band Gap in $In_xGa_{1-x}N$ Layers

A.D. Sequeira, S. Pereira[1], E. Alves[2] and N. Franco

## Objectives

The aim of the current project is to study the strain effect on the optical properties of epitaxial single layers of $In_xGa_{1-x}N$.

## Results

Light emitting devices based upon group III nitride semiconductors have been impressively fast developing in the last years. High efficiency and long lived light emitting diodes, covering almost the entire visible region, as well as commercially available blue laser diodes, are based on the ternary alloy $In_xGa_{1-x}N$. The emission wavelength in these devices is tuned by adjusting indium composition in the active layer.
The accurate knowledge of the In mole fraction, combined with high-resolution X-ray diffraction measurements of the lattice parameters, allowed evaluating the strain for each sample. Indium contents were measured without the influence of strain by Rutherford backscattering spectrometry (RBS).
Optical bandgaps were determined by absorption spectroscopy and corrected for strain. Following this approach, the strain free dependence of the optical bandgap in $In_xGa_{1-x}N$ alloys was established. Our results indicate a linear, $Eg(x)=3.42-3.86x$ eV, "anomalous" dependence of the energy gap, at room temperature, with In content for relaxed material. Extension of this behaviour to higher concentrations is discussed on the basis of reported experimental results.

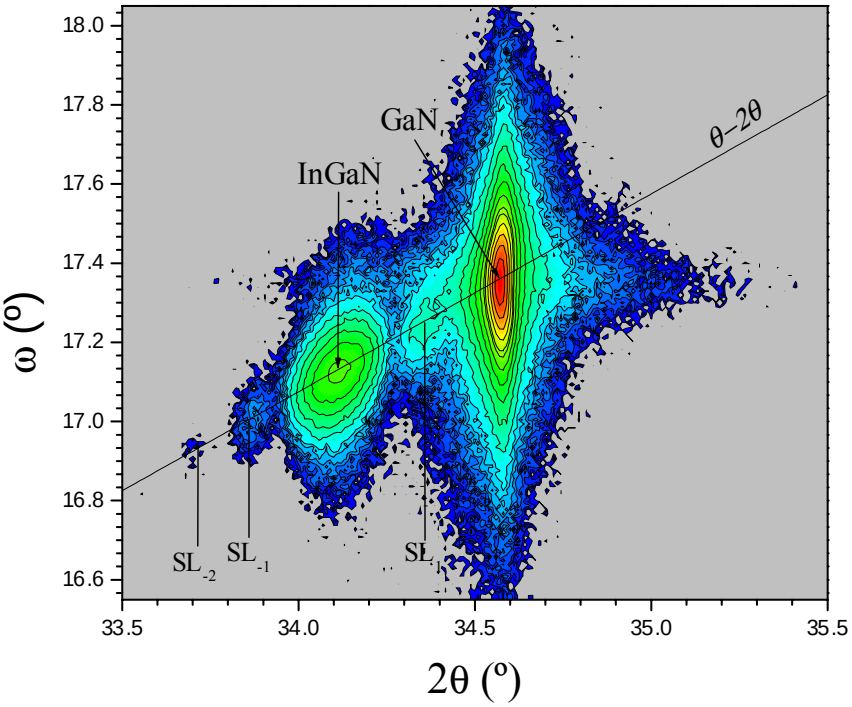



Fig. Ω-2θ Reciprocal space map of the (0002) GaN and $In_xGa_{1-x}N$ satellite peaks. From the angular separation between the peaks the strain in the surface layer is calculated.

## Published (or in press) work

[1] Pereira, S., Correia, M.R., Monteiro, T., Pereira, E., Alves, E., Sequeira, A.D., Franco, N., Compositional Dependence of the strain free optical band gap in $In_xGa_{1-x}N$ layers, *Appl. Phys. Letters*, in press.

[2] Pereira, S., Correia, M.R., Pereira, E., Trager-Cowan, C., Sweeney, F., Edwards, P.R., O'Donnell, K.P., Alves, E., Alves, L.C., Sequeira, A.D., Franco, N., Strain and compositional Analysis of InGaN/GaN layers, *Mater. Res. Soc. Meeting*, Boston PE, Nov. 2000.

[1] Departamento de Física, Universidade de Aveiro, 3810-193 Aveiro, Portugal
[2] Nuclear Solid State Physics using Ion Beams Group.

# Study of Strain Development on Ca, Fe and Mn Implanted GaN and its Evolution upon Annealing

C. Liu, E. Alves, A.D. Sequeira, N. Franco, M.F. da Silva[1], J.C. Soares[1]

## Objectives

The aim of the current project is to determine the strain and damage produced on GaN during the implantation process of several species (Ca, Fe, Mn, etc.), the dependence on implanted dose and its evolution under different annealing conditions.

## Results

**Ca implanted GaN.** The ion-beam-induced amorphization of GaN was studied by using 180 keV $Ar^+$ and $Ca^+$ implantation at –150 °C and room temperature, respectively. The structure of the GaN films before and after implantation was characterized by RRS/channeling and X-ray diffraction (see Figure). Lattice expansion due to ion implantation was found, and excessive lattice expansion results in local collapse of the GaN lattice into amorphous clusters. Accumulation of these amorphous zones should be the mechanism of amorphization. It was concluded that GaN is extremely resistant to amorphization compared to other semiconductors.

**Fe implanted GaN.** Fe ions were homogeneously implanted at room temperature in GaN films with an energy of 150 keV at doses of $1\times10^{15}$ and $1\times10^{16}$ $cm^{-2}$, respectively. A two-step annealing (650 °C 15 min and then 1000 °C 2 min) was performed to remove the implantation-induced damage and to drive the dopants into the lattice sites. The structure of GaN films before and after the implantation as well as at each stage of the annealing was characterized by high resolution XRD and RBS/channeling combined with particle induced x-ray emission. The $Fe^+$ implanted GaN films exhibits an expanded lattice by the appearance of a satellite peak. After the two-step annealing, the lattice distortion does not fully recover. The damage is not annealed out and the dopants are not completely activated.

**Mn implanted GaN.** This study is still in progress.

## Published (or in press) work

[1] Liu, C., Wenzel, A., Rauschenbach, B., Alves, E., Sequeira, A.D., Franco, N., da Silva, M.F., Soares, J.C., Fan, X.J., Amorphization of GaN by ion implantation. *Nuclear Instruments and Methods* (2000), in press.

[2] Liu, C., Alves, E., Sequeira, A.D., Franco, N., da Silva, M.F., Soares, J.C., Fe ion implantation in GaN: Damage, annealing and lattice site location, *J. Appl. Phys*, in press.

[3] Alves, E., Liu, C., Sequeira, A.D., Franco, N., Waerenborgh, J.C., da Silva, M.F., Soares, J.C., Study of Fe Implanted GaN, *12th International Conference on Ion Beam Modification of Materials, IBMM 2000*, Canela – Rio Grande do Sul, Brazil, Sept. 3-8, 2000.

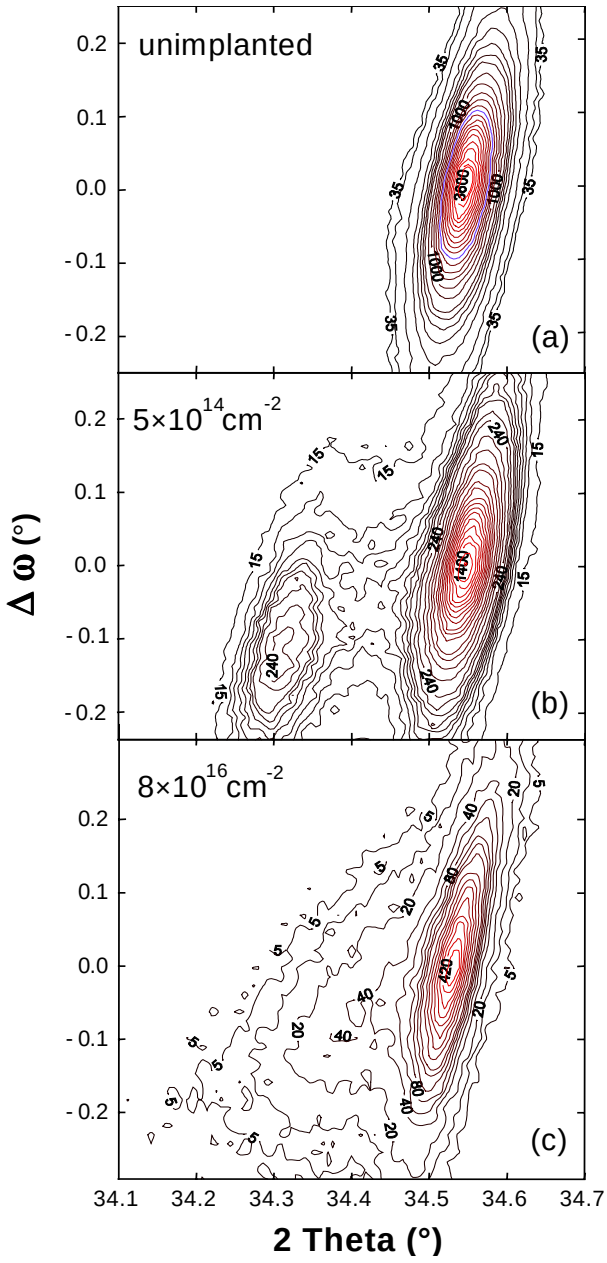



Fig. - XRD ω-2θ reciprocal space maps of the GaN (0002) peak for samples unimplanted (a), after 180 keV $Ca^+$ implantation at room temperature for the doses of $5\times10^{14}$ $cm^{-2}$ (b) and $8\times10^{16}$ $cm^{-2}$ (c).

[1] Nuclear Solid State Physics using Ion Beams Group.

# Structural characterisation of $Si_{1-x}Ge_x/Si_{1-y}Ge_y/Si(100)$ heterostructures

A.D. Sequeira, N. Franco, N. Barradas M. Myronov[1], O.A. Mironov[1] and E. Parker[1]

## Objectives

The aim of the current project is to determine structural parameters of Si1-xGex/$Si_{1-y}Ge_y$/Si(100) p-type modulation doped heterostructures using high-resolution X-ray diffraction and RBS. It is of particular importance to determine the influence of annealing at temperatures. It is of particular importance to determine the influence of annealing in the range of 600-900 ºC on their structural properties.

## Further work

The characterisation of $Si_{1-x}Ge_x$ / $Si_{1-y}Ge_y$/Si(100) has started and will be pursued. This will require, apart from the high angular resolution, also high spatial resolution (very small beam sizes in the micrometer region) to allow the study the fine surface device structure.

## Results

One of the main problems in growing $Si_{1-x}Ge_x$ epilayers on a Si(001) substrate is the lattice mismatch, which increases from 0 to 4.2% as x varies from 0 to 1. The larger x becomes, the thinner the $Si_{1-x}Ge_x$ epilayer has to be grown in order to prevent misfit dislocations from relaxing the strain. One of the possibilities to obtain high Ge concentrations, while retaining strain in the $Si_{1-x}Ge_x$ epilayer, is to use relaxed $Si_{1-y}Ge_y$ substrate with the bulk lattice constant of the $Si_{1-y}Ge_y$. This allows strained $Si_{1-x}Ge_x$ or Ge epilayers to be grown on a underlying Si(001) substrate. Such substrates are termed virtual substrates (VS).

The heterostructures were grown on Si(001) substrates by solid source MBE. The active layers of modulation doped heterostructures inclusive $Si_{0.2}Ge_{0.8}$ channel were grown on $Si_{0.7}Ge_{0.3}$/Si(001) virtual substrate. The structural properties were determined using high-resolution XRD and RBS.

These complex materials are difficult to characterise owing to its fine multilayered structure. In particular the high Ge content $Si_{0.2}Ge_{0.8}$ channel layer buried between $Si_{0.7}Ge_{0.3}$ layers has a typical thickness of only 10nm.

Figure 1a) illustrates a contour map of the reciprocal space map of a typical $Si_{0.2}Ge_{0.8}$ / $Si_{0.7}Ge_{0.3}$/Si(100) heterostructure. On the far right one can observe the Bragg peak from the Si(001) substrate, in middle the peak corresponding to $Si_{0.7}Ge_{0.3}$ relaxed layers and on the left hand side the signal correspondent to the $Si_{0.2}Ge_{0.8}$ strained layer.

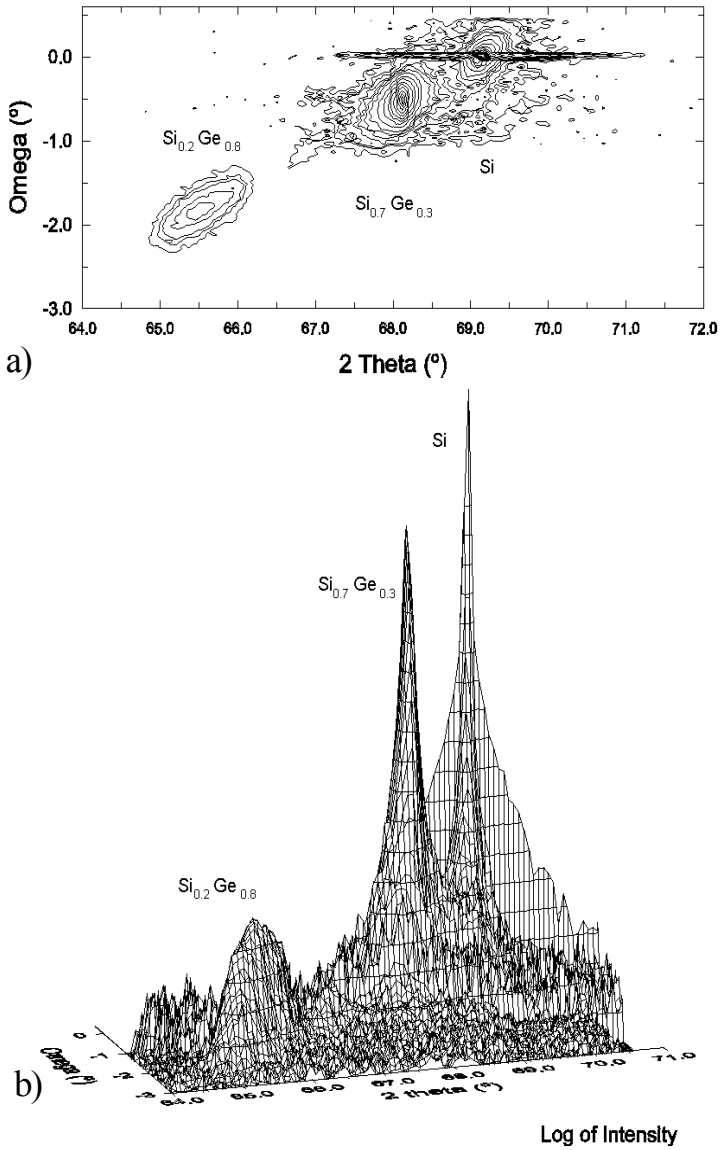



Fig. – Reciprocal space mapping of a $Si_{1-x}Ge_x/Si_{1-y}Ge_y/Si(100$ heterostructure where the channel layer ($Si_{0.2}Ge_{0.8}$) is only 10 nn

[1] Department of Physics, University of Warwick, Coventry, CV47AL, UK.

# Reconstruction of High-Temperature Single Crystalline Aeronautical Components by Laser Cladding and its Testing using Thermo-Mechanical Fatigue

A.D. Sequeira, P. Ferreira[1], R. Vilar[1] and J. Bressers[2]

## Objectives

The aim of the study was to produce and characterise CoNiCrAlY polycrystalline coatings on single crystalline superalloys with low porosity and good oxidation resistance at high temperature, using laser cladding. It is very importance to produce coatings free of defects (pores, inclusions) since these defects are associated with crack initiation and accelerate thermal degradation of the coating and subsequently the substrate.

## Results

Coatings were deposited initially on polycrystalline superalloy samples and at a later stage in single crystalline substrates. The coatings have been characterised by microscopic techniques as well as X-ray diffraction, Rutherford backscattering and Channelling. To our surprise we have managed to go beyond that initial idea and produced single crystalline coatings that are coherent with the substrate with only a small mismatch. This process is not yet being used in industry and is crucial to allow to repair the very expensive single crystalline turbine blades.

Additionally, this technique proved to be an interesting alternative to the conventional deposition techniques (spraying) since it allows, in a single operation, to produce coatings with thickness from 50 to 1000 μm, free from pores and inclusions and excellent adhesion. This may prove to be competitive with the current commercial materials. The laser process is also an alternative to the conventional furnace thermal treatments used to improve adhesion of the coating. In fact, the high cooling speeds observed in these processes produce very fine microstructures, supersaturated solid solutions and metastable compounds, with exceptional functional characteristics. Since the energy density used is several orders of magnitude higher then the ones used in conventional methods, the time of interaction is very short and the energy transfer to the substrate reduced.

## Published work

[1] Sequeira, A.D., Moretto, P., Bressers, J., Residual Stress and Microstructure of CoNiCrAlY coated SRR-99 upon Thermo-mechanical Fatigue, *Materials Science Forum* **Vol. 321-324** (2000) 748-753.

## Further work

We intend to improve the deposition process and extend it to more complex shaped samples. Contacts with industry were initiated to use the process commercially.

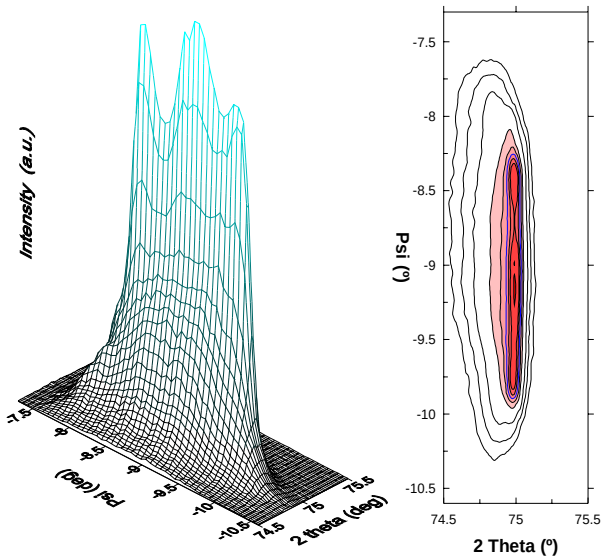



**Fig**. ψ–2θ Reciprocal space mapping of the coating that proved coherent with the single crystalline substrate.

[1] Instituto Superior Técnico, Av. Rovisco Pais, 1096 Lisboa Codex, Portugal
[2] Institute for Advanced Materials, PO BOX 2, 1755 ZG Petten, The Netherlands.

# Study of Strain and Amorphisation of Ge/Si Multilayers Upon Ion Implantation

E. Alves[1], A.D. Sequeira, N. Franco, M.F. da Silva[1], J.C. Soares[1], N.A. Sobolev[2], M.C. Carmo[2]

## Objectives

In this project one intends to investigate the amorphization of SiGe multilayers with different periods under argon irradiation using RBS and high-resolution X-ray diffraction.

## Results

Different Ge/Si superlattices were irradiated with 150 keV Ar ions at room temperature with fluences in the range of $10^{12}$ to $5\times10^{15}$ $cm^{-2}$. Defect production was studied with Rutherford backscattering/channeling spectrometry and X-ray diffraction (XRD). The evolution of the damage with ion fluence reveals the existence of three distinct regimes. During the first regime the concentration of defects increases slowly until the minimum yield reaches a value of ~20%. The onset of the second regime occurs at a fluence of ~$2\times10^{13}$ $cm^{-2}$ and is characterized by a fast buildup of the damage. This regime develops in a narrow interval of fluences ($2\times10^{13}$ - $5\times10^{13}$ $cm^{-2}$) and leads to a saturation of the measured damage. The third regime corresponds to the formation of a continuous amorphous layer that widens with the increase of fluence. A large asymmetry of the diffuse X-ray scattering arises near the critical dose of the crystalline-to-amorphous transition and then relaxes again. The presence of three regimes was also observed in pure silicon irradiated simultaneously with the superlattices, but with a damage saturation threshold nearly one order of magnitude higher ($3\times10^{14}$ $cm^{-2}$). We suppose the driving force of the transition is the excess of strain in the implanted region.

[1] Nuclear Solid State Physics using Ion Beams Group.
[2] Departamento de Física, Universidade de Aveiro, 3810-193 Aveiro, Portugal.

## Published (or in press) work

[1] Alves, E., Sequeira, A.D., Franco, N., da Silva, M.F., Soares, J.C., Sobolev, N.A., Carmo, M.C., Coherent Amorphization of Ge/Si Multilayers With Ion Beams, *Nuclear Instruments and Methods* (2000), in press.

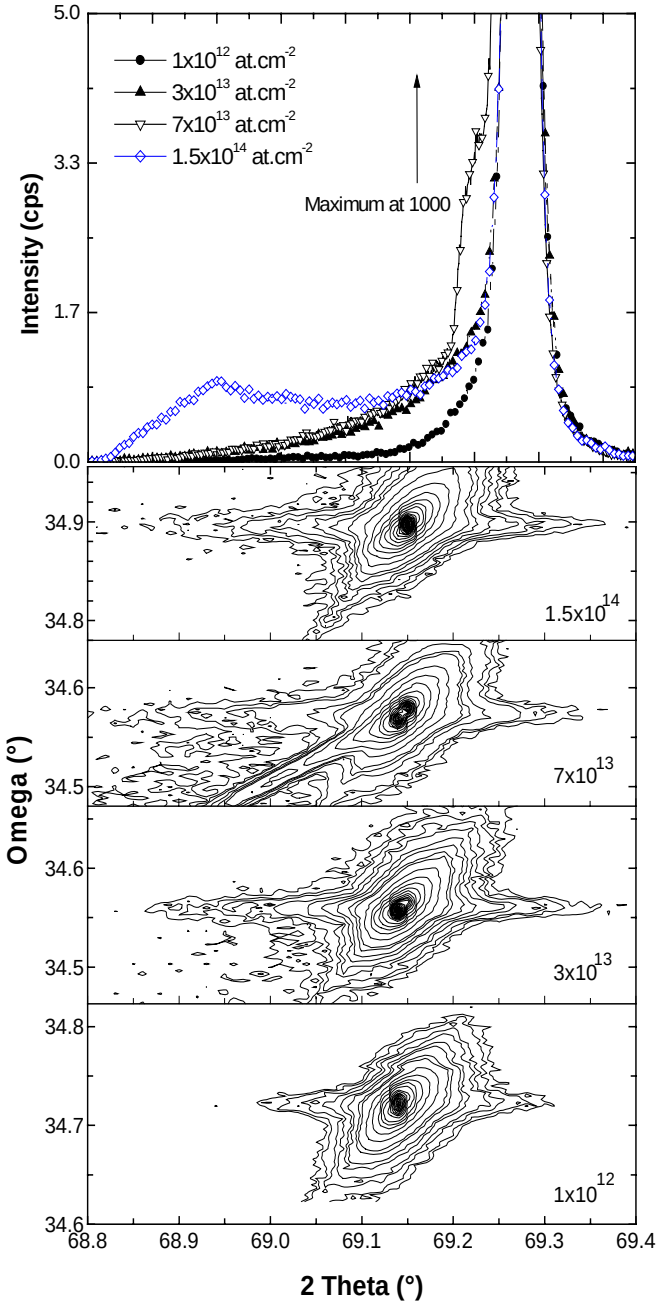



Fig. – Strain produced induced on the multilayer as a function of the implanted dose: a) sequence of 'Integral ω-scans' from the (400) diffraction planes and b) ω-2θ reciprocal space maps around the (400) reflection.

# Characterisation of the Geometrical Pore Arrangement on Silica Grades MCM-41

A.D. Sequeira, M.M.L. Ribeiro Carrott[1], A.J. Estêvão Candeias[1], P.J.M. Carrott[1], P.I. Ravikovitch[2] and A.V. Neimark[2]

## Objectives

The well defined pore structure presented by MCM-41 materials has made them unique candidates for fundamental studies aimed at testing standard adsorption isotherm analysis methods for the characterization of porous solids for predicting phase equilibrium in confined geometries, adsorption in pores and pore size distributions. The aim of the current project is to correlate the adsorption properties of these amorphous materials with the dimensions of the hexagonal lattice structure into which the pores organize themselves.

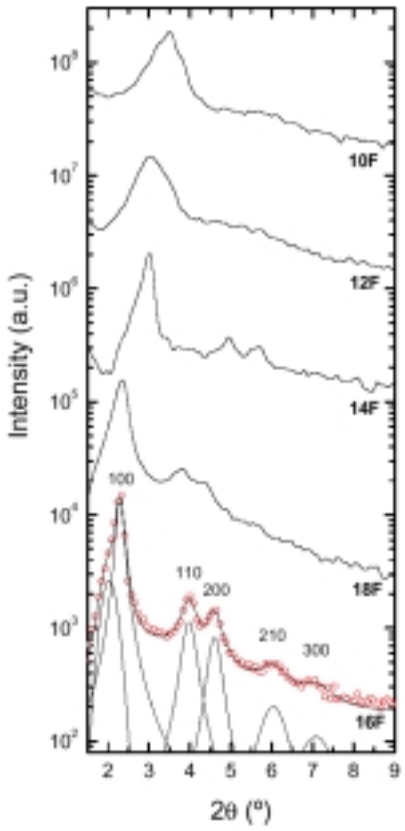



Fig. - XRD spectra of three of the MCM-41 samples. Note that the main peaks appear at very small diffraction angles, making this type of experiment rather unconventional.

## Results

A systematic study of the adsorption of several adsorptives in a series of samples with different pore sizes was performed. The determination of the pore structure was performed using grazing incidence X-ray diffraction. The measurements are delicate due to the small scattering angles of the main peaks which are very close to the incident beam.

Analysis of the results indicates that the density of the nitrogen in the pores is higher than that of the normal liquid. In addition, the results show that in order to calculate the surface area of the pore walls, which is needed in order to calculate the hydraulic pore width, a value for the cross sectional area of the nitrogen molecule lower than the normal value must also be used.

## Communication

[1] Candeias, A.J.E., Ribeiro Carrott, M.M.L., Carrott, P.J.M., Unger, K.K., Sequeira, A.D., "Adsorção de azoto e de compostos orgânicos em MCM-41", *XXV Reunión Ibérica de Adsorción*, Pamplona, 18-20 de Setembro de 2000.

## Further work

Uncertainity still remains with regard to the absolute values of pore volume and surface area and more experimental evidence is still needed to confirm if the mean density of nitrogen adsorbed at 77K in the pores is in fact higher than in the normal liquid state. Therefore, one intends to continue to pursue this objective.

[1] Department of Chemistry, University of Évora, Colégio Luís António Verney, Évora, Portugal
[2] TRI/Princeton, 601 Prospect Avenue, Princeton, New Jersey, USA.

# Study of Spin Dependent Tunnel Junctions by X-ray Diffraction and RBS – Composition and Phase Analysis

A.D. Sequeira, N. Franco, J.C. Soares[1], M.F. Da Silva[1], S. Cardoso[2] and P.P. Freitas[2]

## Objectives

Spin dependent tunnel junctions are multilayered materials with complex structure. One aims to determine the phases present and their evolution upon annealing. Owing to the very fine multilayred structure their characterisation is difficult even with grazing incidence techniques. This study would not be possible without the used the Triple-axis Diffractometer Geometry that was used for the first time here (see the summary entitled "From double-crystal to triple-axis geometry" two pages below).

## Results

The magnetic multilayer sensor samples have the following nominal structure, where the numbers indicate its thickness in angstrom (Ta90/NiFe70/CoFe30/Al9/Fe25/CoFe40/MnIr250/Ta 30). The samples were analysed before and after annealing at 385ºC using grazing incidence triple-axis X-ray diffractometry. The depth profile of the sample was measured with RBS.

Albeit the very thin thickness of the films it was possible to obtain a spectra identifying several phases present namely $IrMn_3$, FeNi, CoFe or Fe, and Ta. Some changes are apparent after the annealing, namely a net reduction of the full-width half-maximum of the Bragg peaks is observed indicating that recrystallization occurs during annealing.

## Further Work

Further work will be carried out with different samples to clarify the influence of the microstructure on the magnetic properties of the devices.

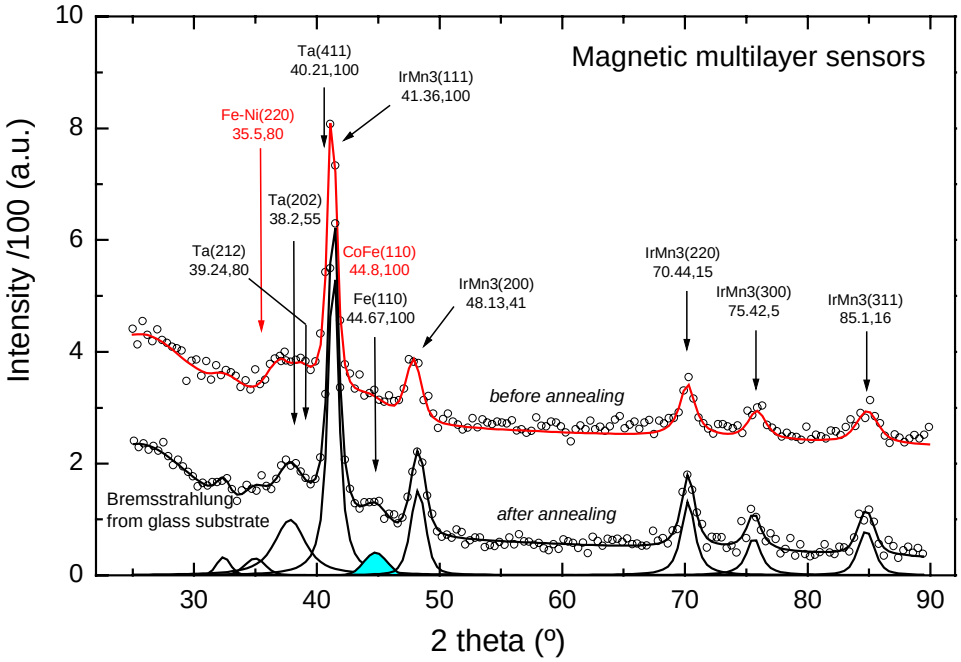



Fig. – Spectra for two samples before (upper curve) and after (lower) annealing at 385ºC. The solid lines correspond to the fitted curves. To visualize the two spectra without superposition they have been shifted by adding and subtracting 100 counts on each spectra respectively.

[1] Nuclear Solid State Physics using Ion Beams Group.
[2] INESC, R. Alves Redol 9, 1º, 1000 Lisboa, Portugal.

# Generalization of the Classic Formalism for Double Crystal Diffractometers to Any Type of Source

N.R. Pinhão[1], I. Rihimaki and A.D. Sequeira

## Objectives

The aim of this project is to generalise the formalism that describes diffractometers in order to allow their optimisation in any configuration needed. This consists in finding the diffractometer geometry that leads to the highest detector count-rate for any given value of the resolution. Depending on the type of experiment, different configurations are needed demanding an optimised diffractometer design.

## Results

Fourty years ago, a series of articles by Caglioti *et al*. have studied thoroughly the influence of geometrical factors on the resolution and luminosity of neutron diffractometers. Since then the 'Caglioti formalism' has been applied to all type of diffractometers independently of the type of source used. However, the above formalism is valid only for 'white' sources and can not be applied to X-ray diffractometers using 'monochromatic' sources.

In the current project, the mathematical expressions of the Bragg peak intensity and its full width half maximum (FWHM) were obtained for a general source and considered in terms of the angular divergences of the incident and diffracted beams and of the mosaicity of both the monochromator and the sample. The general results were then applied taking into account the actual diffractometer components. Namely, the use of a position sensitive detector and different monochromator crystals were considered.

Contrarily to white sources, monochromatic sources have a sharp line profile that can be approximated by a Gaussian distribution with standard deviation given by the natural width. Since the FWHM (W) for a 'monochromatic' source is always smaller than for a 'white' the relative difference, $(W_w-W_m)/W_w$ between the two cases can be as high as 95% as can be seen in figure 1.

## Published work

[1] Margaça, F.M., Pinhão, N.R. and Sequeira, A.D.. Design Optimisation of a High-temperature X-ray Diffractometer for in-situ Residual Stress Analysis and Lattice Mismatch Determination. *Materials Science Forum* Vol. **322** (2000)168.

[2] Pinhão, N.R., Riihimäki, I., Sequeira., A.D., Geometrical analysis of Double Crystal Diffractometers – Extension of the Classical Formalism to X-ray Sources, *XII National Conference of Physics - FISICA 2000*, Figueira da Foz, Portugal, Sept. 2000.

## Further work

Experimental determinations of the diffractometer basic parameters (FWHM, intensity, etc) under a set of different conditions are under way (see Figure) and being compared with the calculated ones in order to confirm the correctness of the new formalism.

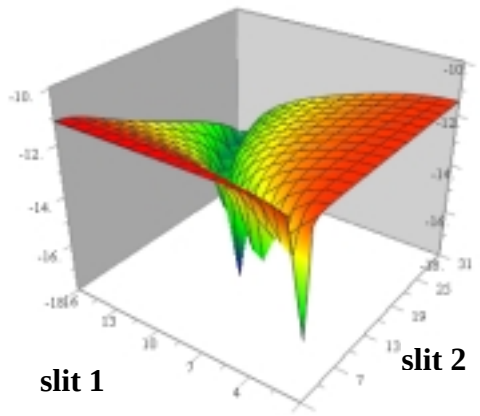



Fig. 1 – Relative difference between Caglioti FWHM and the first order correction.

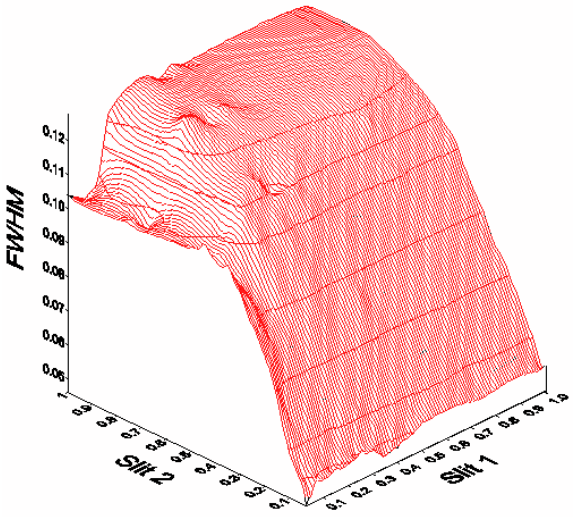



Fig. – Experimental determination the FWHM of the Hotbird beam at the sample position as a function of the divergencies defined by slit 1 and slit 2.

[1] Nuclear Instruments and Methods Group.

# From Double-Crystal to Triple-Axis Geometry - Development of a Crystal Analyser Stage and Corresponding Software for the Hotbird

A.D. Sequeira, N. Franco, J. Teixeira[1]

## Objectives

The aim of the current project is to extend the capabilities of the Hotbird diffractometer towards higher resolution through the implementation of a crystal analyser as the third axis of the diffractometer.

## Results

The implementation of the third axis of the Hotbird involved the project of mechanical components and the development of software in LabView. The most expensive component of such a system is the single-crystal analyser. A graphite single crystal was selected owing to its high efficiency. This precious part was kindly offered by Dr. José Teixeira from the Laboratoire Léon-Brillouin, Saclay, France. All the experimental software procedures were altered to accommodate the new geometry (an example is shown in the Fig.).

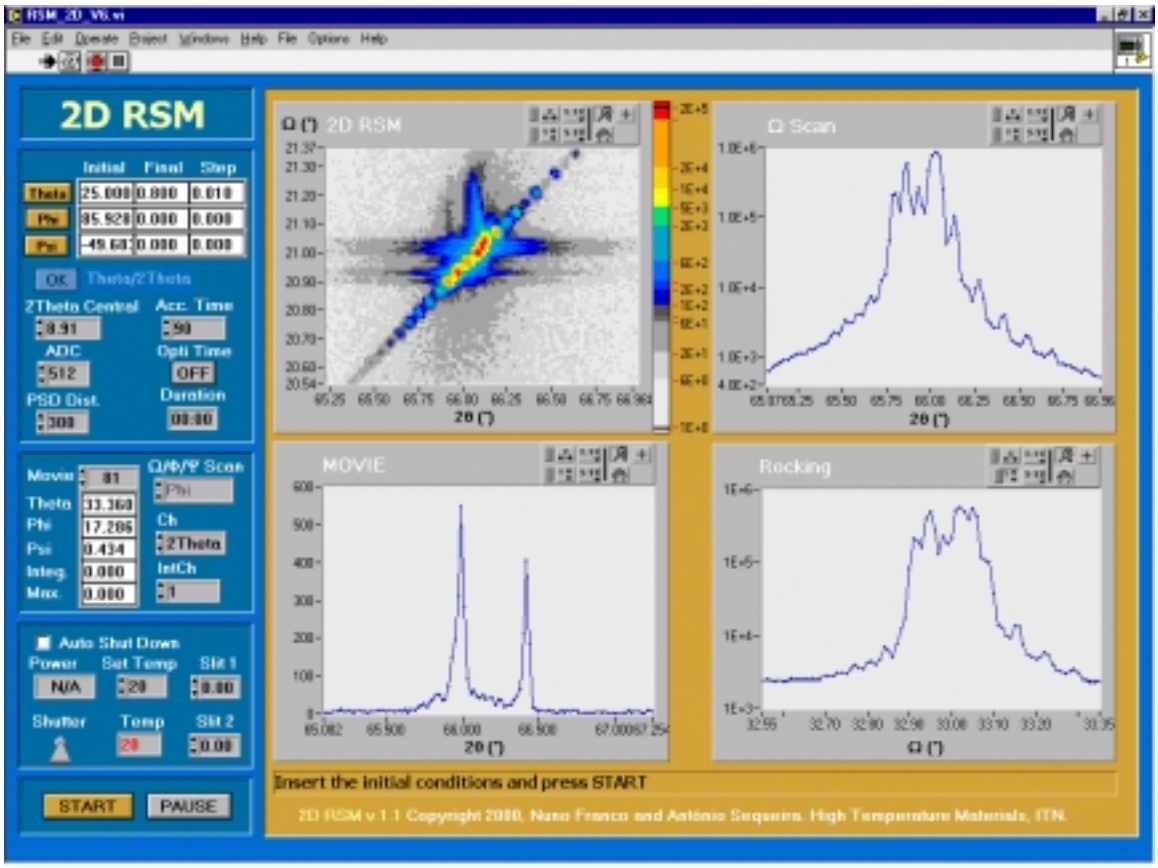
## Published (or in press) work

[1] Sequeira, A.D. , Franco, N., Neves., J., High-Resolution and High-Temperature Double-Crystal X-ray Diffractometer for *in-situ* Studies - the Hotbird, *Mat. Sci. Forum* 6pp., Accepted for publication.

[2] Franco, N., Sequeira, A.D., Software Package for the Remote Control and Data Acquisition from a High-temperature X-ray Diffractometer The Hotbird, *Mat. Sci. Forum* 6pp., Accepted for publication.

## Further work

Currently, additional experimental procedures are being adapted to the new geometry.




[1] Laboratoire Léon-Brillouin, CEA/CNRS, Saclay, France.

# Microstructure Investigation of Organic-Inorganic Hybrid Materials*

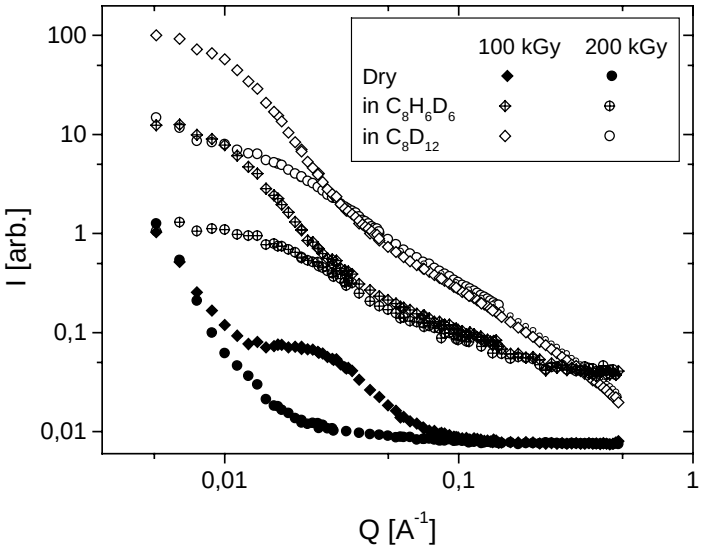
F.M.A. Margaça, A.N. Falcão, J.S. Sousa, I.M.M. Salvado[1], A.M.R. Mendonça[1], J.Teixeira[2], M. Misheva[3], F.G. Carvalho

## Objectives

Preparation and characterisation of materials with an inorganic constituent composition of (100-x)SiO2.xMO2 (M=Ti, Zr) covalently linked with an organic component of polymeric material such as PDMS, to carry out a systematic investigation of the hybrids so obtained in order to find the dominant relationships between microstructure (mainly at the nanoscale), macroscopic properties and processing conditions.

## Results

The preparation of new silica-based hybrid materials has been carried out both by the alkoxides method of the sol-gel process and by irradiation of the precursors using the $^{60}$Co Gamma Irradiation Unit, UTR. The latter is performed to promote radiation induced cross-linking. Several samples have been irradiated, prepared with TEOS and PDMS in different concentrations. Samples with different inorganic compositions, with zirconia and titania in different concentrations, have also been prepared by the alkoxides method. Results of the systematic investigation of the microstructure of the materials inorganic component, using Small Angle Neutron Scattering, SANS, and Positron Anihilation Spectroscopy, PAS, are being published [1-3]. The nanostructure investigation of some of the hybrid samples has been performed using the SANS instrument PAXE [4] at Laboratoire Léon-Brillouin, in November 2000. Data analysis is in progress. A preliminary analysis show that the scattered intensities are very much dependent on the processing conditions. This can be observed in the figure where scattered intensities obtained from samples prepared by irradiation to two different doses (100 kGy and 200 kGy) are displayed. Irradiated hybrid samples were measured as dry and immersed in a good solvent of the polymer. From the five different values of H/D isotopic substitution in the solvent, for each sample, two are shown (fully deuterated solvent and 50% H/D substitution). In addition the plot also shows scattering spectra obtained from the dry samples.




## Published (or in press) work

[1] Miranda Salvado, I.M., Santos Sousa, J., Margaça, F.M.A. and Teixeira, J., Structure of $SiO_2$ gels prepared with different water contents *Physica B* 276-278 (2000) 388.

[2] Misheva, M., Djourelov, N., Margaça, F.M.A., Miranda Salvado, I.M., Positronium decay study of zirconia-silica sol-gels, *J. Non-Cryst. Solids,* **272** (2000) 209.

[3] Misheva, M., Djourelov, N., Margaça, F.M.A., Miranda Salvado, I.M., Positronium decay study of porous structure of sol-gel prepared $SiO_2$: influence of Ph, *J. Non-Cryst. Solids,* (2000) in press.

[4] Falcão. A.N. and Mendonça, A.M.R., SANS investigation of organic-inorganic hybrid xerogels with contrast variation, LLB Progress Report, Dec. 2000.

## Further work

Systematic characterisation of the existing hybrid samples needs to be done as concerns the mechanical properties. The influence of irradiation dose (covering a wider range) and that of polymer molecular weight will be investigated.

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# Production and Characterisation of Colour Centres in Alumino-Silicate Crystals: Topaz

M. Carrapiço, A.N. Falcão, R.C. da Silva[1], L. Santos[2]

## Objectives

To produce colour centres in natural colourless topaz by means of gamma-ray and neutron beam irradiation, and ion-beam implantation. To characterise the colour centres created.

## Work in 2000

Samples were prepared by irradiation with mixed neutron and gamma–ray beams and annealed to produce stable blue colours. RBS, RBS-C, and EPR experiments were performed on colourless and coloured samples in order to investigate the role played by hydrogen in the production of colour. The results obtained show correlation between the fraction of OH–F substitution and colour intensity.
Some of the results obtained were published in two papers [1,2], two other papers [3,4] were presented at a conference and a student started his University Degree thesis.

## Published (or in press) work

[1] Marques, C., Falcão, A. N., da Silva, R. C., Alves, E., Annealing behaviour of natural topaz implanted with W and Cr Ions, *Nuclear Instruments and Methods in Physics Research* B 166-167 (2000) 204.

[2] Marques, C., Santos, L., Falcão, A. N., da Silva, R. C., Alves, E., Luminescence studies in colour centres produced in natural topaz, *Journal of Luminescence* **87-89** (2000) 583.

[3] Marques, C., Carrapiço, M., Santos, L., Falcão, A. N., da Silva, R. C., Alves, E., Production of colour centres in topaz by gamma–ray and neutron irradiation, *Com. Pres. XXII Nac. Phys. Conf.*, Coimbra (Sept. 2000).

[4] Marques, C., Falcão, A. N., da Silva, R. C., Alves, E., Implantation of transition metals in colourless topaz, *Oral Com. Pres. XXII Nac. Phys. Conf.*, Coimbra (Sept. 2000).

## Further work

The correlation structure – colour in gamma-ray and neutron beam irradiated topaz will be further investigated. Techniques like RBS, RBS-C, X-ray diffraction, SEM and TEM, EPR as well as micro-Raman spectroscopy will be used. Initial ion-implantation work will continue choosing different implantation conditions (temperature, energy) to avoid destruction of the surface structure and implanting new ions. The re-crystallisation process of the damaged surface observed in the previously studied samples will be investigated as well as the role played by the implanted ions on the final colour.

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# Development of a Converging Multichannel Collimator (CMC)

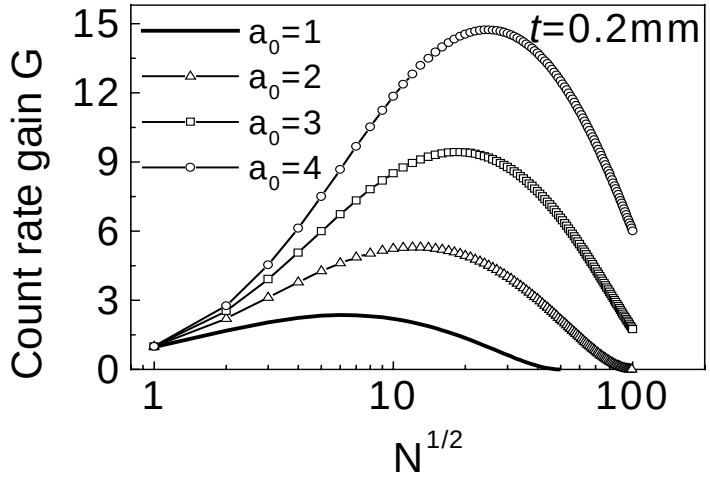
A.N.Falcão, F.M.A. Margaça, J. F. Salgado, F. G. Carvalho

## Objectives

To design and construct a prototype of a variable geometry multichannel collimator with converging walls. First a fixed geometry unit will be produced and tested. Than a manually operated unit will de developed and, finally, a fully automatic operated unit will be produced.

## Results

The effect of the use of different collimator types in the performance of small angle scattering instruments (SANS) was recently addressed [1]. It was shown that the use of converging multichannel collimators (splitting the neutron source, bringing it closer to sample and allowing larger sample areas) can lead to considerable count rates gains at fixed resolution relative to the classical single collimator arrangement. A project was written and submitted for financial support to Fundação para Ciência e Tecnologia. Upon approval (project PROCT/ 1999/FIS/34449), design of the fixed geometry unit has started.




Count rate gain of an optimised SANS facility equipped with a CMC. N, is the number of CMC channels; t, the thickness of the channel walls; and $a_0 = (A_{NS}^0)^{1/2} / \Delta l$, where $A_{NS}^0$ is the available neutron source area and Δl is the detector pixel size.

## Published work

[1] Margaça, F.M.A., Falcão, A.N., Salgado, F.J., Carvalho, F.G., "Multichannel Collimation for SANS Instruments", *Physica B* **276-278** (2000) 189.

## Further Work

Design, construct and test a prototype of a fixed geometry multichannel collimator with converging walls.

# Instruments for Neutron Scattering at the Portuguese Research Reactor

A. N. Falcão, F. M. A. Margaça, J. F. Salgado, F. G. Carvalho

## Objectives

Exploitation of the Time-of-Flight Diffractometer, ETV, and installation of both the Two-Axis Diffactometer, DIDE, and the Small Angle Neutron Scattering Instrument, EPA.

## Results

The installation of DIDE was pushed forward following the visit of the International Advisory Board, in September. The remote control electronics, which was developed in collaboration with the LLB (Saclay), was received in October, and the shielding was installed and tested in November. The result of the test proved that the shielding is adequate. Presently (December), the assembly of the components required for the instrument to go into test phase is being done. First tests of the instrument are scheduled for January of 2001, and will be carried out in collaboration with the LLB. In 2000, the project spent a total of 1466 kPTE. The overall budget now amounts to 27.8 MPTE (26.4 of which allocated in 1998).

The set-up of ETV was changed. The $2\theta=45^{\circ}$ detector bank was removed and the corresponding detectors added to the $2\theta=90^{\circ}$ detector bank. A new structure holding the detectors was made and the electronics renewed. The instrument was used for training and educational purposes in general – experimental work included in introductory lectures to neutron scattering (collaboration with the Physics Department of the Faculdade de Ciências of the Lisbon University); introduction to neutron diffraction techniques to students from Lisbon University (collaboration in a Master Course with the Physics Department of Faculdade de Ciências); and to students from Instituto Superior Técnico of the Technical University of Lisbon, and from the Physics Department of the Aveiro University.

As concerns EPA, it was defined in September that the safety report was to be produced by our group. Radiation fields and shielding were studied in collaboration with the Nuclear Instrumentation group. Radiation induced energy deposition and resulting heating of the materials were estimated with the aid of Monte Carlo Simulation techniques. Assemblage/disassemblage procedures have been elaborated. The full safety report should be delivered soon.

View of the shielding that sur-rounds the monochromator of the two – axis diffractometer, DIDE.

## Further work

To complete the installation of DIDE the leveling of the floor will have to be done, a furnace and a cryostat will have to be purchased. Also, but depending on the evaluation to be done during the test phase, filtering devices are predicted to be necessary to purchase.
Installation, test and commissioning of EPA.