

Materials Characterization with Radioactive Nuclear Techniques

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A laboratory infrastructure on materials characterization is maintained and developed at ISOLDE-CERN by the Nuclear Solid State Physics group of ITN and CFNUL. ISOLDE is a European, and unique facility in the world, where more than 750 radioactive isotopes of 80 elements are produced and delivered as ion beams of high elemental and isotopic purity. In this context nuclear techniques such as Emission Channeling (EC) and Perturbed Angular Correlations (PAC) provide (atomic scale) information complementary to the material analysis capabilities available at ITN. The ITN-CFNUL infrastructure and related projects are refereed and reevaluated each year within the scope of FCT-supported CERN projects. The scientific work in 2006 was currently centered in three research subjects, which have been approved with beam time at ISOLDE by the ISOLDE Scientific Committee:

a) IS368 "Lattice Location of Transition Metals and Rare Earths in Semiconductors". Within this subject, the lattice sites of impurities in technologically relevant semiconductors (e.g. Si, Ge, ZnO, GaN) and oxides (e.g. SrTiO₃, BaTiO₃) are studied by means of the emission channelling technique.

b) IS360 "Studies of High-Tc Superconductors doped with radioactive isotopes". The PAC technique is used to study the atomic ordering of fluorine and oxygen dopants at the Hg planes of the first three members of the HgBa₂Ca_{n-1}Cu_nO_{2n+2+δ} high-Tc family of superconductors. The aim is to understand if dopants ordering and consequent lattice deformations are related or unrelated with charge ordering stripe formation at the superconducting planes.

c) IS390 "Studies of colossal magnetoresistive oxides with radioactive isotopes". PAC is used to probe local

lattice deformations and relaxation of the Mn³⁺O₆ octahedra on manganites as a function of doping and temperature. In this way phase coexistence and polaron dynamics are studied, which are local phenomena that are correlated with charge transport mechanisms in giant magnetoresistive materials.

The group has also accomplished technical development of position-sensitive Si pad detectors to implement fast readout of self-triggered detectors. On-line EC experiments with short lived isotopes shall run in 2007.

Since the obtained information, i.e., the precise lattice location and rms displacements of impurities, or the incoherent relaxation of elements in crystalline solids, are not accessible by more traditional techniques, the radioactive methods have also the potential for being applied to different subjects or new materials.

Together with the development of the new detector, a sophisticated new experimental chamber has been developed that will allow new EC experiments at low measurement temperatures, to provide high precision insight of phenomena such as element relaxation in semiconductors and superconductors.

Of interdisciplinary nature, these activities integrate and initiate young students, from different backgrounds and universities, in applied nuclear physics. With shared work between the different environments of ITN, CFNUL and ISOLDE-CERN, there participate students and senior researchers from the universities of Lisbon, Aveiro, Porto, Braga, as well as from Leuven, and Bonn. Presently, six Ph.D., one M.Sc. and three diploma students accomplish their work using this infrastructure, within the scientific proposals. Two students have finished Ph.D. in 2006, one of such still working as post-Doc in the group.

Researchers^(*)

J.G.M. CORREIA, Principal Researcher

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Emission channelling lattice location studies

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 and the ISOLDE collaboration¹

Objectives

The aim of this work is to study the lattice location of dopants and impurities in technologically relevant semiconductors and oxides by means of electron emission channelling (EC) from radioactive isotopes. With this technique information is available for very low dopants concentrations and independent from the host lattice elemental composition. The experiments are carried out using the ITN/CFNUL infrastructure installed at CERN's ISOLDE facility.

Results

1. Lattice location of implanted Ca and Sr in GaN

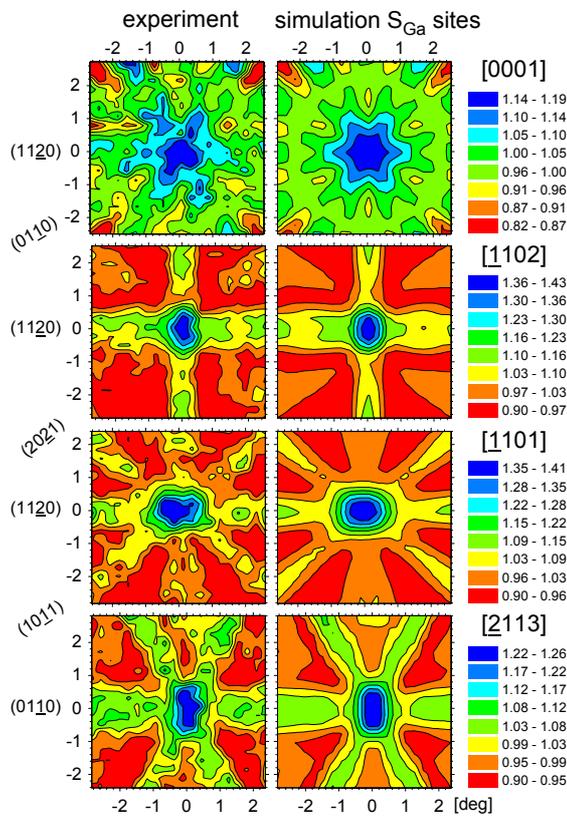


Fig. 1: Experimental EC patterns from ⁴⁵Ca in GaN (left) and simulations for Ca on Ga sites (right).

Using the emission channelling technique, the angular distributions of β^- -particles emitted by the radioactive

isotopes ⁴⁵Ca and ⁸⁹Sr implanted into thin films of single-crystalline wurtzite GaN were monitored with a position-sensitive detector. The experiments give direct evidence that ~90% of Ca and ~60% of Sr atoms occupied substitutional Ga sites with root mean square displacements of the order of 0.15-0.30 Å, i.e., larger than the expected thermal vibration amplitude of 0.074 Å. Annealing the Ca implanted samples at 1100-1350 °C in high-pressure N₂ atmosphere resulted in a better incorporation into the substitutional Ga site. The Sr implanted sample showed a small decrease in rms displacements for vacuum annealing up to 900 °C, while the substitutional fraction remained nearly constant. The annealing behaviour of rms displacements can explain why annealing temperatures above 1100 °C are needed to achieve electrical and optical activations, despite the fact that the majority of the acceptors are already located on Ga sites immediately after ion implantation.

2. Emission channelling with short-lived isotopes

Due to count rate and noise-related limitations of the detection systems, electron emission channelling experiments using position-sensitive detection were restricted to isotopes with half lives above 6 h and electron energies above 40 keV. Recently, major technical developments have been realized and new equipment has been implemented which has allowed these limitations to be overcome. The main new development supported by the ITN Sacavém/CFNUL Lisbon group in collaboration with CERN, was the implementation of self-triggering readout chips for position-sensitive Si pad detectors. These new readout systems allow count rates of several kHz, sufficient to measure samples in the MBq range, and with energies below 40 keV. A scientific proposal for lattice location studies using short-lived isotopes produced at CERN's ISOLDE facility has been approved by the CERN research board on 29.11.06 with 20 shifts of beam time. The physics case of the presented proposal will be to extend the successful work in the fundamentally and technologically relevant field of the lattice location of transition metals and dopants in semiconductors using as new probes the isotopes ⁶⁵Ni (2.5 h), ⁶¹Co (1.6 h), and ²⁷Mg (9.46 min).

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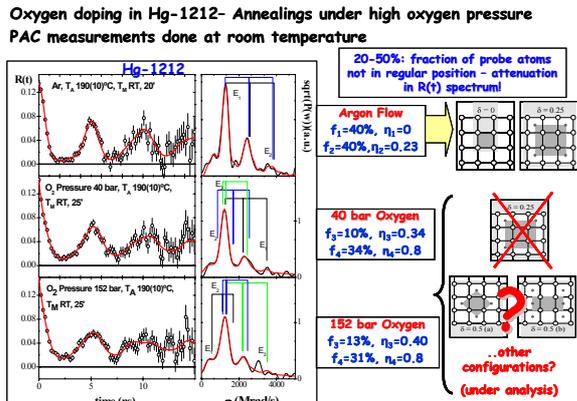
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IS360 experiment – Atomic ordering of high concentrations of oxygen in HgBa₂Cu_nCa_{n-1}O_{2n+2+δ} for n = 2,3 high-Tc superconductors

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Collective ordering of highly oxygen doped samples of HgBa₂Cu_nCa_{n-1}O_{2n+2+δ} (n = 2,3 Hg1212, Hg1223) were investigated with the perturbed angular correlation (PAC) technique by measuring the electric field gradients induced at ^{199m}Hg nuclei. The experiments were performed at different annealing conditions, under Ar flow or O₂ pressure up to 152 bars. In comparison with the data and calculations already published for equivalent fluorine doping in Hg1201 (n = 1), the preliminary analysis hints that at high concentrations the oxygen atoms order in different way, other than the atomic-like stripes found for fluorine. In addition, these experiments have been performed at different temperatures, above and below the superconducting transition, which have revealed further differences in the charge distribution of the Hg surroundings. A full set of PAC, SQUID (Tc) and X-ray data has been obtained that is now being analyzed to be further compared to first principle calculations of charge density in these materials obtained for different oxygen configurations.

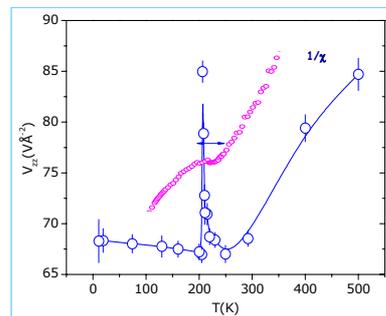


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IS390 experiment – Studies of Free percolative phase transition on ferromagnetic insulator manganites

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We report atomic scale studies of the Pr-CaMnO₃ system using PAC spectroscopy. No macroscopic measurements were up to date able to detect electric polarization in the Pr-CaMnO₃ (PCMO) systems. The present set of data provides first experimental evidence for local electric dipoles in charge ordered PCMO. We hint that the electric dipole is localized to a small region (nm?) and that the ferroelectric behaviour occurs below the CO/orbital ordering phase transition.

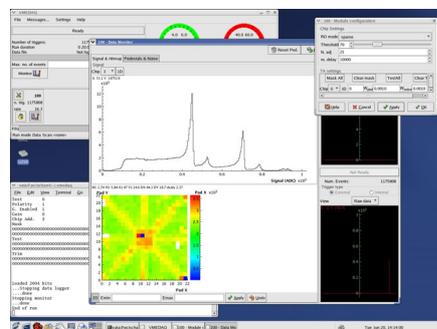


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R&D development – new self-triggered Si pad detectors for position sensitive electron detection

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This work is made in collaboration with CERN’s Compton camera project on new positron emission tomography (PET) devices <http://192.171.198.135/In/lettr/PSDT.pdf>, whose technology and detectors also fulfill the requirements for electron emission channeling experiments. In 2006 the readout program was developed to the 1 mm thick 22×22 Si pad detector, mounted on a newly designed printed circuit board equipped with fast VATAGP3 preamplifier chips. The figure shows the acquisition program visual interface during first tests with ¹¹¹In implanted SrTiO₃, of the new detector to be installed online in 2007.



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