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 Large Scale Facility where more than 750 isotopes and 80 elements are produced and delivered as ion beams of high elemental and isotopic purity, which is unique in the world. In this context nuclear techniques such as Emission Channeling (EC) and Perturbed Angular Correlations (PAC) provide complementary (atomic scale) information 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 2007 was centered in three research subjects that have been approved with beam time by the ISOLDE Scientific Committee:

a) IS453 "Emission Channeling Lattice Site Location Experiments with Short-Lived Isotopes". The lattice sites of impurities in technologically relevant semiconductors (e.g. Si, Ge, ZnO, GaN) and oxides (e.g. $SrTiO_3$, $KTaO_3$) are studied by means of the EC technique. New probes from elements that only possess short-lived radioactive isotopes are now available, as is later detailed in this report.

b) IS390 "Studies of colossal magnetoresistive oxides with radioactive isotopes". PAC is used to study multi-ferroic $RMnO_3$ manganites as a function of element R and temperature to study local phenomena that are correlated with the coexistence of ferroelectricity, ferromagnetism and ferroelasticity.

c) IS360 "Studies of High-Tc Superconductors doped with radioactive isotopes". PAC is used to study the atomic distribution of oxygen dopants at high concentration at the Hg planes of the $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, high-Tc superconductors. The aim is to learn where O_δ sits (and dopes) depending on concentration. This is accompanied by modern first principle density charge simulations that reveal the importance of spin interactions on site occupancy and lattice relaxation.

With respect to the work topics during 2007, priority was given to the commissioning of the new emission-channeling chamber, the new fast Si pad-detector, the development of the data treatment program for on-line experiments, and first successful data-taking in June 2007. Regarding the EC on-line setup, 2008 will be still a year of experimental consolidation, with the planned integration of a cooling station (30 K-300 K). The overall effort for further development of the EC technique is well justified since the precise lattice location and rms displacements of impurities in crystalline materials are not accessible by more traditional methods. In a different context, radioactive hyperfine techniques are very useful for studying electrical and magnetic properties at the nanoscopic scale. During 2007 an extra physics push was made to learn and handle new state-of-the-art charge density simulation programs that include relativistic and spin interactions to provide a better understanding of systems with strong electronic ⇔ atomic correlations as occurs within the manganites.
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, ISEL as well as from Leuven in Belgium. Presently, seven Ph.D. students, one M.Sc. and one diploma student accomplish their work using this infrastructure, within the scientific proposals and R&D projects, testing new experimental and physics concepts.

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IS-453 experiment: Emission channeling lattice location studies

U. Wahl, J.G. Correia, A.C. Marques, C.P. Marques, E. Rita, E. Alves, S. Decoster, A. Vantomme, M.R. da Silva, J.P. Araújo, L. Pereira, J.C. Soares, and the ISOLDE collaboration[1]

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 dopant 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. Amphoteric arsenic in GaN

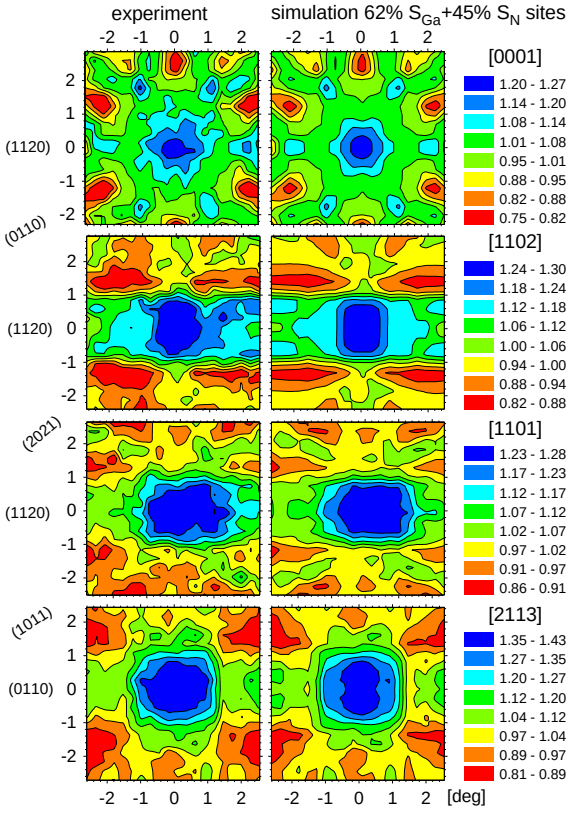

Fig. 1: experimental EC patterns from ^{73}As in GaN (left) and simulations for 62% of As on Ga and 45% on N sites (right).

We have determined the lattice location of implanted arsenic in GaN by means of conversion electron emission channelling from radioactive ^{73}As (80.3 d). Already in the as-implanted state but also following annealing up to 900°C it was found that similar fractions of As occupied substitutional Ga and substitutional N sites. We have thus given direct evidence that As is an amphoteric impurity in GaN, thus settling the long-standing question as to whether it prefers cation or anion sites. The amphoteric character of As provides additional aspects to be taken into account for an explanantion of the so-called "miscibility gap" in ternary $GaAs_{1-x}N_x$ compounds, which cannot be grown with a single phase for values of x in the range $0.1 < x < 0.99$. The lattice location of As in GaN contrasts with its site preference in ZnO where we found it to occupy mainly substitutional Zn sites with only minor fractions on O sites, a fact which can be explained by the smaller size mismatch between As^{3-} and N^{3-} ions (2.22 Å *vs* 1.71 Å).

2. Emission channeling with short-lived isotopes

Following last year's report on the implementation of self-triggering readout chips for position-sensitive Si pad detectors (supported by the ITN Sacavém/CFNUL Lisbon group in collaboration with CERN) and on the development of an emission channelling on-line setup, we report here on first results with this new equipment. The setup was used on-line during the Mn beam time in June 2007, mounted at the LA2 beam line of ISOLDE and equipped with the new detectors, which performed well and allowed to reach data-talking rates above 3 kHz. Two isotopes were successfully used for β^- emission channelling experiments for the first time: ^{56}Mn (2.58 h) and ^{61}Co. While ^{56}Mn was available directly, ^{61}Co was obtained by means of implanting the short-lived precursor isotope ^{61}Mn and exploiting the decay chain ^{61}Mn (4.6 s) $\rightarrow$ ^{61}Fe (6 min) $\rightarrow$ ^{61}Co (1.6 h). We were thus able to determine the lattice location of Mn in GaN and of Co in ZnO in the as-implanted state and following annealing up to 900 °C. In both cases it was found that the transition metals preferred substitutional cation (i.e. Ga or Zn) sites.

Published work

U. Wahl, J.G. Correia, J.P. Araújo, E. Rita, and J.C. Soares: "Amphoteric arsenic in GaN", Applied Physics Letters 90 (2007) 181934/1-3.

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IS360 experiment – Oxygen occupancy in $HgBa_2Cu_2Ca_1O_{6+\delta}$ high-Tc superconductors

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We found evidence for O_δ occupying (unexpected) interstitial positions along the edge of the Hg square in $HgBa_2Cu_2Ca_1O_{6+\delta}$ (Hg1212) on samples heavily doped, under pressure, with oxygen. This was first revealed by an electric field gradient (EFG) with relatively low Vzz and high $\eta \sim 1$ induced at ^{199m}Hg nuclei, measured with Perturbed Angular Correlation (PAC). Then a major effort was made in 2007 to run simulations with a new program, WIEN2k that uses density functional theory with the local spin density approximation. This requires quite large supercells and lengthy computer time for the present physics cases. Since this position for O_δ cannot be stable alone, even if it will reproduce the experimental EFG results, we simulate several oxygen configurations to learn that the stability is ruled by local strong deformations in correlation with the known elongation of the *a* lattice parameter, which can only be properly simulated with the inclusion of spin density functionals. It hints as if oxygen can be promoted to such interstitial abnormal sites if, in adjacent rows the centers of the Hg squares are heavily loaded with oxygen, and extra deformations take place when more oxygen atoms are forced in. Once simulations are completed, this work might explain the origin of controversial diffraction data that hinted such an additional position for the dopant without being able to confirm or explain it.

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

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A family of multiferroic systems $RMnO_3$ perovskite manganites (R = Eu, Gd, Ho, Y, Er and Lu) have been prepared with the sol-gel technique. On these systems the issue is to understand the microscopic phenomenology that causes coexistence of ferroelectricity, ferromagnetism and ferroelasticity as a function of R and temperature. PAC studies have been performed allover the series to measure the Electric Field Gradient / Magnetic Hyperfine Field of ^{111m}Cd/^{111}cd atoms at R sites, as a function of the ionic radius of the element R and for some cases under different temperature conditions from 10 K to 473 K. The first data and analysis show that there are two competing local configurations on the lattice, from which only one shows a correlation of the η (asymmetry EFG parameter) with transition temperature as a function of R. These first results point to the separation of microscopic phases, maybe associated to local deformations, with the macroscopic properties.

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R&D development – new Si pixelated detectors for position sensitive electron detection

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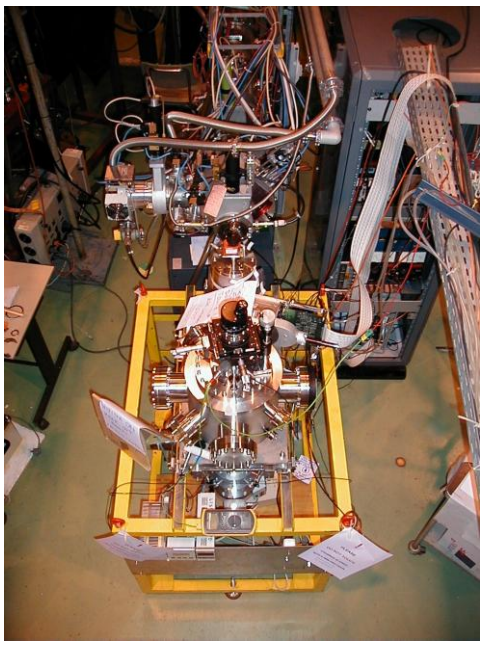

The commissioning of the on-line chamber and fast Si-pad detector has been successfully achieved, with data taking. Looking forward to the future, we have submitted a research proposal to the MEDIPIX detector collaboration at CERN (http://medipix.web.cern.ch/MEDIPIX/) that got approved in early December. These detectors have 256×256 pixels covering 15×15mm^2, self common mode compensation and the bias and full readout is provided via a single USB cable. The collaboration will do a prototype for emission channeling purposes with four of such detectors covering an active surface of 30×30mm^2, that we expect to commission during the second half of 2008. The software is fully developed and can easily be adapted to our type of data analysis.

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