Materials Characterization with Radioactive Nuclear Techniques

João Guilherme Martins Correia

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 1000 isotopes and isobars of 80 elements are produced and delivered as ion beams of high elemental and isotopic purity, which is a unique feature 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 ion beam laboratories. This infrastructure and projects are refereed and reevaluated each year within the scope of FCT-supported CERN projects. In 2010 the scientific work was centered in the following research subjects approved by the ISOLDE Scientific Committee:

a) IS453 (U.Wahl) "Emission Channeling Lattice Location Experiments with Short-Lived Isotopes". The lattice sites of dopants and impurities in scientifically and technologically relevant semiconductors are studied with the emission channeling technique at highly diluted concentrations. During 2010, ^{56}Mn (2.58 h) was used to study the lattice site of the transition metal Mn implanted into ZnO, Si, Ge, GaAs, $SrTiO_3$ and AlN. The ^{24}Na (14.95h) alkaline isotope was used to study p- and n-Si doping. Being Mg an essential p-type dopant in semiconductors, we have pursued lattice location studies on GaN, AlN and InN using ^{27}Mg (9.45m). Looking forward for lattice site location of Be a first demonstrative experiment was performed in GaN with the short lived isotope ^{11}Be (13.8s).

b) IS487 (V. Amaral) "Study of Local Correlations of Magnetic and Multiferroic Compounds". PAC is used to study a large variety of multiferroic $RMnO_3$ (R= rare-earth) manganites and cromites $ACrO_2$ (A=Ag,Cu) as a function of the elements R, A, and of temperature. By combining PAC data with first principle simulations (f.p.s.) of charge density distributions on these materials, local phenomena that correlate the coexistence of ferroelectricity, ferromagnetism and ferroelasticity are studied. During 2010 PAC experiments were performed in several multiferroic oxides (manganite, nickelite, chromite) pnictides and chalcogenides. The experimental studies are accompanied by the theoretical analysis and calculation of the hyperfine parameters using *ab initio* methods for determining the electronic structure as well as the modeling of the relevant physical properties.

c) IS481 (K. Lorenz) "The role of In in III-nitride ternary semiconductors", have combined γ–γ with e^--γ PAC using the ^{111m}Cd/^{111}Cd and the ^{117}Cd/^{117}In isotopes. Results obtained in 2010 have confirmed the role of the In dopant, as being predominantly stabilizing defects in GaN, InN and AlN.

In what R&D projects are concerned, first alpha particle detection tests were successfully done with a highly pixelated 256x256 14×14mm^2 Si detector (TimePix). This setup will be mounted at the ITN accelerator to be tested on RBS/Channeling experiments using H^+ and He^{2+} beams. In parallel, the new high-resolution goniometer from Panmure, dedicated to on-line electron EC experiments with short-lived isotopes, has been mounted on a (very) special sustaining cradle to be commissioned on-line at the ISOLDE GHM-beam line in 2011.

Of interdisciplinary nature, these activities integrate and initiate 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 (Belgium) and Bonn(Germany). During 2010 one MSc student defended his thesis; five Portuguese and one German PhD students performed work for their thesis and one new PhD student is about to start work, using this infrastructure within the scientific proposals and R&D projects.

Research Team

Researchers

J.G.M. CORREIA, Princ.
U. WAHL, Princ. (15%)
K. LORENZ, Aux. (10%)

Students

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Collaborators

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IS453 experiment: Emission channeling lattice location studies

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and the ISOLDE collaboration[5]

Results

1. Lattice location of Mn in GaAs

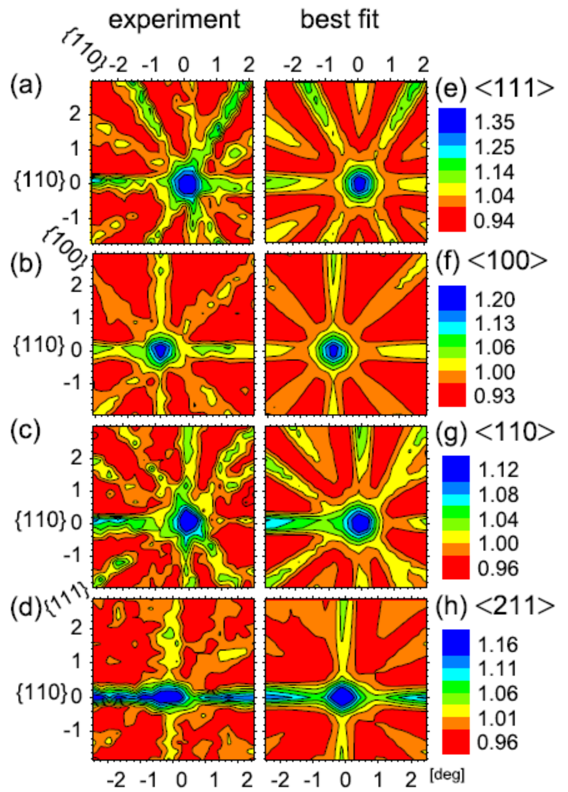

Fig. 1: (a)-(d) Angular emission patterns of β^- particles from ^{56}Mn in GaAs in the vicinity of <111>, <100>, <110> and <211> crystallographic directions following annealing at 300 °C. (e)-(h) best fits of theoretical patterns for a combination of 71% of ^{56}Mn on substitutional S_{Ga} and 29% on interstitial T_{As} sites.

The ternary compound $Ga_{1-x}Mn_xAs$ represents the best understood dilute magnetic semiconductor system which exhibits ferromagnetism, although the maximum Curie temperature T_C realized (around 200 K) is currently still considerably below room temperature. Whereas it is well-known that T_C in $Ga_{1-x}Mn_xAs$ increases with increasing Mn fraction x and that Mn mostly replaces Ga atoms, it was suggested in the literature that there is a limit imposed on T_c by the amount of Mn atoms that are incorporated interstitially. However, the exact location of interstitial Mn was unknown and its thermal stability and diffusion behaviour were uncertain as well. We have studied the lattice location of ^{56}Mn implanted into p^+-GaAs:Zn. Fig. 1 compares the normalized experimental ^{56}Mn β^- emission yields along the four major directions with the best fits of theoretical patterns, which were obtained for a combination of 71% of ^{56}Mn on substitutional S_{Ga} and 29% on tetrahedral interstitial T_{As} sites. Measuring the emission channeling effects as a function of annealing temperature showed that the interstitial Mn fraction of ~30% was converted to substitutional Mn only for annealing temperatures above 400 °C, which is contrary to the claim reported in the literature that Mn interstitials anneal by means of outdiffusion from the sample at temperatures around 200 °C.

2. Emission channeling with short-lived isotopes

Using our emission channeling on-line setup at the ISOLDE GHM beamline, we participated in three beam times using short-lived radioactive isotopes in 2010. During the Mg beam time we determined the lattice location of the potential acceptor ^{27}Mg (9.5 min) in InN, and Mg was found mainly on substitutional In sites. Due to beam sharing with a nuclear physics experiment, exceptionally 10 days were available for emission channeling experiments with Mn isotopes, and lattice location data was obtained for ^{56}Mn in $SrTiO_3$, InN, *si*-GaAs, n^+-GaAs, $Ga_{0.99}Mn_{0.01}As$, $Ga_{0.95}Mn_{0.05}As$, Ge, *p*-Si, p^+-Si, ^{59}Mn in p^+-GaAs and 3C-SiC , as well as a high angular resolution measurement of ^{56}Mn in ZnO. The bulk of this data is currently being analyzed and the results can therefore not yet be entirely included in this report. However, the *si*-GaAs, n^+-GaAs and GaMnAs experiments confirmed the results described above for ^{56}Mn in p^+-GaAs. First test for the use of ^{11}Be (13.8 s) in β^- emission channeling experiments in GaN were undertaken during a Be beam time, showing that this isotope can be used for lattice location purposes, however, however that it suffers from some background problems due to its high β^- endpoint energy of 11.5 MeV.

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IS487 experiment – Hyperfine Interactions in MnAs studied by Perturbed Angular Correlations of γ-Rays using the probe 77Br→77Se and first principles calculations for MnAs and other Mn pnictides.

J.N. Gonçalves[1], V.S. Amara1[1], J.G. Correia, A.M.L. Lopes[2]

New systems that exhibit simultaneous magnetocaloric, magnetoelastic and magnetoresistance effects are particularly challenging, looking forward to understand their properties for immediate applications. Hyperfine techniques offer unique tools, which the Perturbed Angular Correlation (PAC) Technique is particularly appropriate to probe at a nanoscopic scale, in the surroundings of highly diluted impurity elements, microscopic phase transitions involved in these materials phenomenology. The case study here presented resumes the use of radioactive ^{77}Br/^{77}Se PAC probe to study MnAs phase transitions, which work has just been accepted by Physical Review B, i.e., the MnAs compound shows a first-order transition at $T_C \approx 42$ ºC, and a second-order transition at $T_t \approx 120$ ºC. The first-order transition, with structural (hexagonal-orthorhombic), magnetic (FM-PM) and electrical conductivity changes, is associated to magnetocaloric, magnetoelastic, and magnetoresistance effects. We report a study in a large temperature range from −196 up to 140 ºC, using the γ−γ perturbed angular correlations method with the radioactive probe ^{77}Br→^{77}Se, produced at the ISOLDE-CERN facility. The electric field gradients and magnetic hyperfine fields are determined across the first- and second-order phase transitions encompassing the pure and mixed phase regimes in cooling and heating cycles. The temperature irreversibility of the 1st order phase transition is seen locally, at the nanoscopic scale sensitivity of the hyperfine field, by its hysteresis, detailing and complementing information obtained with macroscopic measurements (magnetization and X-ray powder diffraction). To interpret the results, hyperfine parameters were obtained with first-principles spin-polarized density functional calculations using the generalized gradient approximation with the full potential (L)APW+lo method (WIEN2K code) by considering the Se probe at both Mn and As sites. A clear assignment of the probe location at the As site is made and complemented with the calculated densities of states and local magnetic moments. We model electronic and magnetic properties of the chemically similar MnSb and MnBi compounds, complementing previous calculations.

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Implementation of "ab initio" Perturbed Angular Correlation Observables for Analysis of Fluctuating Quadrupole Interactions

M.B. Barbosa[1,2], J.G. Correia. J.P.E. Araújo[2]

The PAC technique offers a privileged way to look at time transient phenomena involving changes of electronic density at the nanosecond time scale, among which polarons and charge ordering could be featured within a certain temperature range. Still, a proper analysis modeling was missing due to the difficult formalism and heavy computer power required. This year M. Barbosa presented his M.Sc. thesis on the development and implementation of a ab-initio simulation program based on stochastic Hamiltonians that reproduce electronic density fluctuations during the measurement time, allowing data analysis to polycrystalline and single-crystalline systems with dynamic interactions. This program was briefly tested on the experimental case of ^{181}Hf:GaN. In the near future we aim to adapt the code to parallel computing and three transition stochastic states, necessary to interpret some physical problems.

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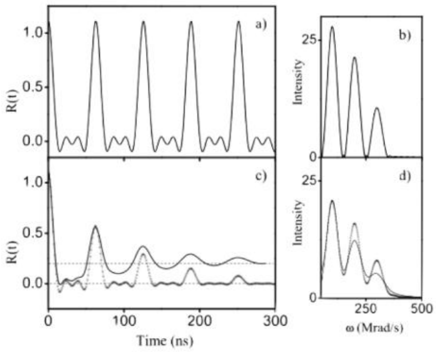

Legend: (a) Schematic representation of R(t) with no attenuation, η = 0, with the correspondent Fourier transform on (b). (c) Representation of R(t) for the same frequency as in (a) with EFG static distribution attenuation (top curve) and, with attenuation due to fast EFG fluctuations (bottom curve), with the correspondent Fourier transform represented on (d). In in the case of damping due to dynamic interactions the ω1, ω2, ω3 amplitudes decrease by the same factor while for a damping originated by static-like frequency distribution the attenuation is proportional to the frequency itself. The R(t) additionally shows an exponentially-like vanishing a_0, as a function of time, for dynamic interactions.

IS481 experiment – An In-defect complex as a possible explanation for high luminous efficacy of InGaN and AlInN based devices

P. Kessler[1], K. Lorenz, S.M.C. Miranda, J. G. Correia, K. Johnston[2], R. Vianden[1] and the ISOLDE collaboration

The role of indium in GaN and AlN films was investigated with the γ– γ and e-γ perturbed angular correlation (PAC) methods. Using the PAC probe ^{111}In/^{111}Cd, in addition to indium on substitutional cation sites a large fraction of probes is found in a distinctly different microscopic environment, which was attributed to the formation of an indium nitrogen-vacancy (V_N) complex. The influence of electron capture induced *after-effects* was ruled out by additional measurements with the PAC probes ^{111m}Cd and ^{117}Cd on differently doped GaN samples. It is further shown that V_N is not bound to substitutional Cd impurities, suggesting that the In- V_N complex formation is a particularity of In in GaN and AlN. In fact, after the decay of ^{111}In to ^{111}Cd, below a critical temperature, VN remains nearby to the probe atom during the measurement, since VN has not enough energy to leave the cadmium within the lifetime of the initial excited state of ^{111}Cd. This originates a specific EFG with a temperature dependent attenuation that allowed developing a preliminary model to interpret the trapping and release of V_N in In doped AlN and GaN.

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R&D development –TimePix detectors for emission channeling and / or He^{2+} RBS/Channeling studies

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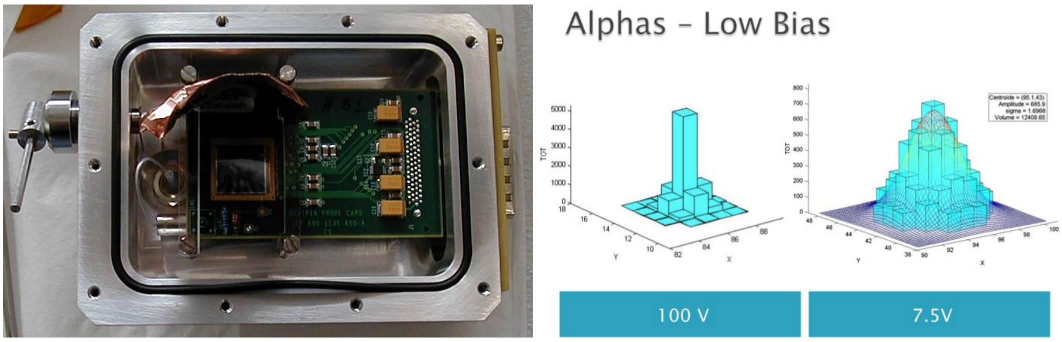
Looking forward to the future, where short-lived isotopes with high beta energies require improved position detection resolution, since December 2007 that we are working with the MEDIPIX CERN collaboration at CERN (http://medipix.web.cern.ch/MEDIPIX/). In 2010 we have developed the mounting and tested one of such detectors with 256x256 pixels covering ~15x15mm2, with alpha particle emitters aiming emission-channeling experiments. The system, which is shown in the figure below (left), is now ready to be tested on-line at a He^{2+} beam line of the Rutherford Back-Scattering Channeling (RBS/C) ITN - accelerator. The figure further shows on the right, the pixel response to single impacts of 5.6MeV alpha particles from a ^{241}Am source for two bias voltages. With the detector biased at 100 V, the charge generated by an alpha particle hit is confined to a few pixels only, while it spreads over a considerable number of pixels at the lower detector bias of 7.5 V. However, this effect is actually beneficial: since the charge spread is exactly Gaussian, a Gaussian fit to the charge distribution at low bias provides sub-pixel position resolution, being the integral used for energy determination.



Legend: (left) timepix detector implemented on the special housing developed by us, to be mounted at the ITN-Sacavém accelerator beam line. (righ) Alpha particle event taken as a function of bias voltage; the spectra show on z the energy deposition (TOT) signal deposited per pixel on a single event. Fitting the spectra with a pure 2D Gaussian, provides the full energy (integral) and the precise (x,y) event hit coordinates (centroid of the Gaussian).

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