Materials Characterization with Radioactive Nuclear Techniques

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A **laboratory infrastructure** on materials characterization is maintained and developed at ISOLDE-CERN by researchers 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 ion beam laboratories. 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 2008 was centered in three research subjects approved by the ISOLDE Scientific Committee:

a) IS453 "Emission Channeling Lattice Site Location Experiments with Short-Lived Isotopes". The lattice sites of impurities in scientifically and 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. Elements of interest, which can only be characterized by using their short-lived isotopes are now detectable.

b) IS390 "Studies of colossal magnetoresistive oxides with radioactive isotopes". PAC is continuously used to study a large variety os multi(anti)ferroic $RMnO_3$ (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.

c) IS360 "Studies of High-Tc Superconductors doped with radioactive isotopes". PAC was 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 was to learn the O_δ atomic configurations depending on concentration and temperature. This project was closed in 2008 after finishing f.p.s. of charge density distributions that allowed to interpret the experimental data, revealing that the oxygen atomic configurations change with temperature above and below Tc. An extensive paper will be published in 2009.

2008 was a year of consolidation and new achievements on R&D projects. The new emission-channeling chamber using the fast Si pad-detector setup was refined and reused with success. There we have further implemented a cooling station (50 K – 300 K) and first low temperature test EC experiments were successfully achieved with ^{111}In implanted into InP. Additionally, first electron tests on a highly pixilated 256x256 14x14mm^2 Si detector (MEDIPIX) were done with a source of 73As: ZnO sample.

In a different context, the radioactive hyperfine technique PAC was very useful to study at the nanoscopic scale electrical and magnetic properties. 2008 was a rich year in both experimental, with new materials being studied, and simulation achievements. We point the fact that our group is now able to use, autonomously, complex charge density simulation programs, which are essential providing the data interpretation on the studied systems.

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, ISEL as well as from Leuven in Belgium. On 2008 one PhD student defended her thesis, one is finishing the writing, six other PhD and three MSci students performed their work using this infrastructure within the scientific proposals and R&D projects.

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IS-453 experiment: Emission channeling lattice location studies

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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 channeling (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. Lattice location of implanted Er in Ge

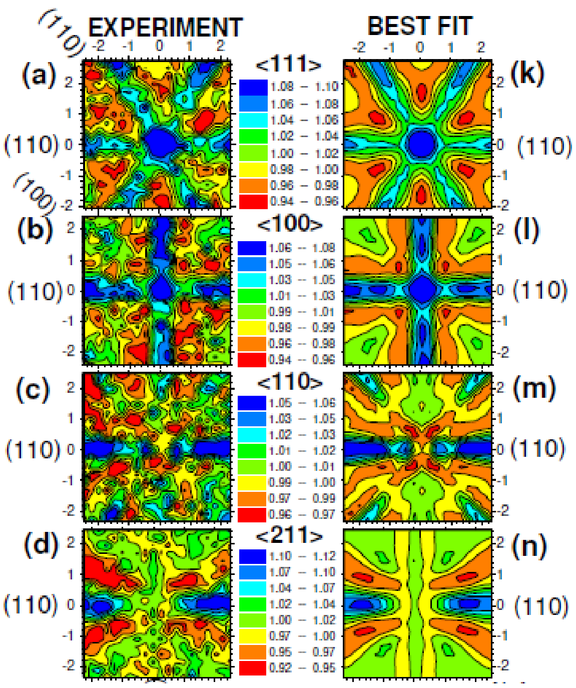

Figure: Experimental EC patterns from ^{167m}Er in Ge (left) and simulations for a combination of 20% of Er on T , 16% on BC and the rest on random sites (right).

We have used conversion electron emission channeling to determine the lattice location of ^{167m}Er (2.28 s) in Ge after 60 keV room temperature implantation of the precursor isotope ^{167}Tm (9.25 d). We found direct experimental evidence of Er atoms located on the tetrahedral interstitial site (T) and on the bond-centered (BC) site. Whereas Er is expected to occupy the interstitial T site in a diamond crystal structure (as was previously determined from emission channeling in the case of ^{167m}Er Si), the observation of bond-centered Er in Ge is more surprising and believed to be related to the so-called Er-vacancy defect in the split-vacancy complex configuration, which is formed when an Er atom pairs with a Ge double-vacancy. Such complexes have been theoretically proposed to exist for a number of oversized transition metal impurities in Ge and our results strongly suggest that these predictions also apply to the case of rare earths.

2. Emission channeling with short-lived isotopes

Using our new emission channeling on-line setup which is equipped with a fast position-sensitive Si pad detector and was commissioned in 2007 followed by its first on-line run at the ISOLDE facility, a second set of lattice location experiments with short-lived radioactive isotopes were done during this year's Mn beam time. We were able to determine the lattice sites of implanted ^{56}Mn (2.6 h) in ZnO, Ge and GaAs, and of ^{61}Co (1.6 h) in GaN during the run. We found that Mn in ZnO prefers substitutional Zn sites and Mn in Ge substitutional Ge sites. Mn in GaAs occupies substitutional sites as well, but distinguishing between Ga and As sites will only be possible after careful analysis due to the very similar nuclear charges of these two elements. While ^{56}Mn was implanted directly, for the study of Co in GaN we exploited the decay chain ^{61}Mn(4.6 s) $\rightarrow$ ^{61}Fe (6 min) $\rightarrow$ ^{61}Co (1.6 h), i.e. ^{61}Mn was implanted and after a waiting period of 30 min lattice location was done on ^{61}Co. We found that Co occupies substitutional Ga sites in GaN. In addition, by means of using the decay chain ^{59}Mn (0.71 s) $\rightarrow$ ^{59}Fe (45 d), we prepared samples of $SrTiO_3$ and $KTaO_3$ doped with ^{59}Fe, for which the lattice location of Fe is being determined during the following months.

Published work

S. Decoster, B. De Vries, A. Vantomme, U. Wahl and J.G. Correia: Experimental evidence of tetrahedral interstitial and bond-centered Er in Ge, Appl. Phys-Lett. 93 (2008) 141907.

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IS390 experiment – Studies of local phase transitions on multiferroic compounds.

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In 2008 we have published in a PRL a detailed study of the electric field gradient (EFG) across the $Pr_{1\_x}Ca_xMnO_3$ phase diagram and of its temperature dependence. The EFG behaves differently on samples outside or inside the charge order (CO) regime. In particularly, the EFG temperature dependence evidences a new and discontinuous phase transition occurring over the broad CO region of the phase diagram. Such prominent features revealed to be associated with polar atomic vibrations, which eventually lead to spontaneous local electric polarization below the CO transition. Following the same experimental procedures we are now looking in detail at the temperature dependence of the EFGs on multiferroic systems $YMnO_3$ and $EuMnO_3$ and of the newly synthesized $AgCrO_2$ as a function of temperature down to 10 K. The aim is to study other cases where there is coexistence of magnetic and electric polarization along the phase diagrams, looking forward to find the extension, looking at the nature of the different local regimes. For all samples the data is excellent, in the sense that it has enough precision for evidencing local transitions at different regions of the phase diagrams for each sample. Again these results point the coexistence of multiple microscopic phases, somehow hindered at measurements of only macroscopic properties. The data is presently under analysis, refining the fits and performing first principles calculations of the EFGs for different local arrangements.

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R&D development – concept design, manufacture and commissioning of a cooling station for EC studies

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Since about ten years that lattice location experiments with the EC technique have not been performed at low temperatures. In spite of the fact that one of the old goniometer setups is equipped with a He flow cryostat, there were several inhibiting factors for regularly doing such experiments: 1) it is very heavy (~50kg), requiring the use of a crane each time samples need to be changed or the cryoshield needs to be turned to cope with the sample axis, 2) it is actually installed on a building without He recovery line and 3) it cannot be mounted on-line. With the commissioning of the new fast detector and on-line chamber the motivation to go below RT become very appealing, where the samples could be implanted at cryogenic temperatures to study annealing stages of impurities below RT. Furthermore, on materials with low structural symmetry we might attempt to probe the anisotropy of vibrations, or position r.m.s shifts of the elements / impurities as a function of temperature, e.g., upon electric and magnetic phase transitions. Successfully cases of such experiments would be of great interest on probing microscopic local effects. With these aims in mind we have developed and successfully commissioned a cooling station using a closed cycle He refrigerator and a cryogenic shielding system that is coupled to our "all-day-use" simple goniometer. In this first approach we could achieve a sample temperature down to 50K. We have studied a first test case prepared by ion implantation of ^{111}In on InP. Once In is a member of the lattice it was easy to anneal out the implantation damage at 300^{o}C. Additionally, since InP has a low Debye temperature it is expected (as were measured) large variations of the vibration r.m.s. amplitudes as a function of temperature from RT down to 50K. The data is presently under analysis but, as new information from these experiments, we guess so far that on systems with low Debye coefficients, i.e., large r.m.s vibration amplitudes the dechanneling plays a stronger effect than what is actually considered in the present manybeam EC simulations. 2009 will be a year of analysis, experiments, and consolidation and improvement of the low temperature system, aiming to achieve 30K.

Still on the field of EC R&D, we report first EC tests of electron detection with a MEDIPIX 256x256 channels, 14x14mm^2, 300μm thick detector. We used a ^{73}As : ZnO source and nicely measured the EC electron anisotropy spectrum for the face axis <0001> for the 42 keV electrons. A tentative spectrum using only 12keV electrons was done, with some channeling effect being visible, but the probable detection of x-rays and the lack of knowledge of how should look such low energy electron spectra hindered further conclusions. New tests using TIMEPIX (energy resolved detectors) will be further done on 2009 on a special chamber developed by us, equipped with a cooling system on real experimental conditions.

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