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 unique facility in the world where more than 750 isotopes and 75 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 complementary information to the materials analysis capabilities available at the ITN home institute. The ITN-CFNUL infrastructure and related projects are refereed and reevaluated each year within the scope of FCT-supported CERN projects. The scientific work is 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. $SrTiO_3$, $BaTiO_3$) are studied by means of the emission channeling 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 $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, high-Tc family of superconductors. The aim is to understand if dopant 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 $Mn^{3+}O_6$ 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 magnetoresitive materials.

The group is also involved in technical research and development of position-sensitive Si pad detectors. The aim is to implement fast, self-triggered detectors which will allow performing EC experiments on-line with short lived isotopes, which cannot be accomplished with the present set of detectors. The first of the new detectors is scheduled to run in 2006.

Since the obtained information (i.e. the precise lattice location and rms displacements of impurities in crystalline solids, or impurity interactions with point defects or with local lattice deformations) is not accessible by other techniques, radioactive methods also have the potential for being applied to different subjects or new materials. It is intended that in the near future new EC and PAC experiments will provide high precision insight on low temperature phenomena such as element relaxation in semiconductors and superconductors, or on charge diffusion processes in insulators.

At CFNUL we support and form young engineers working in nuclear electronics, with the aim of developing and implementing new detectors with LYSO scintillators, for improved energy resolution in complicated PAC cascades.

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. Actually, five Ph.D., one M.Sc. and three diploma students accomplish their work within this infrastructure and scientific proposals

Research Team

Researchers

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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 emission channelling (EC) from radioactive isotopes. The experiments are carried out using the ITN/CFNUL infrastructure installed at CERN's ISOLDE facility.

Results

1. Arsenic as a Zn-site impurity in ZnO

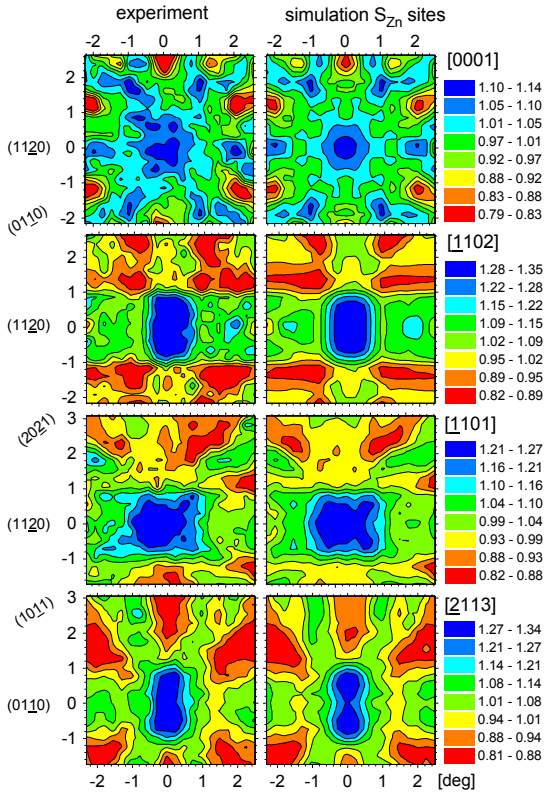

Fig. 1: Experimental EC patterns from ^{73}As in ZnO (left) and simulations for As on Zn sites (right).

Arsenic has been reported in the literature as one of the few p-type dopants in the technologically promising II-VI semiconductor ZnO. However, there is an ongoing debate whether the p-type character is due to As simply replacing O atoms or to the formation of more complicated defect complexes, possibly involving As on Zn sites. We have determined the lattice location of implanted As in ZnO by means of conversion electron emission channelling from radioactive ^{73}As. In contrast to what one might expect from its nature as a group V element, we find that As does not occupy substitutional O sites but in its large majority substitutional Zn sites. Arsenic in ZnO (and probably also in GaN) is thus an interesting example for an impurity in a semiconductor where the major impurity lattice site is determined by atomic size and electro-negativity rather than its position in the periodic system.

2. Lattice sites of implanted Fe in Si

Fe represents, together with other transition metals such as Cu, Ni or Co, one of the most dangerous contaminants in Si processing technology. However, little has been known on possible lattice sites of Fe in Si. Using β^- emission channelling from the radioactive isotope ^{59}Fe implanted into Si single crystals at fluences from 1.4×10^{12} cm^{-2} to 1×10^{14} cm^{-2}, we identified Fe on three distinct sites: ideal substitutional, displaced substitutional and displaced tetrahedral interstitial. Whereas displaced substitutional Fe was dominating for annealing temperatures below 500°C, annealing between 500-700°C caused the majority of Fe to occupy displaced tetrahedral interstitial sites. Ideal substitutional positions were increasingly populated following annealing at 800°C and above.

3. Lattice location of implants in $SrTiO_3$

$SrTiO_3$ is a semiconducting, perovskite-type oxide with a band gap of 3.2 eV and a high bulk dielectric constant. Its optical, magnetic or electrical properties can be modified by doping, allowing for some possible applications in microelectronics. Since implantation damage in $SrTiO_3$ is removed around 400°C, ion implantation has the potential to represent an attractive approach for doping, however, little has been known whether implants are incorporated on proper lattice sites. In that respect and in order to further investigate the annealing behaviour of implanted $SrTiO_3$, we are currently carrying out lattice location studies of a number of implanted impurities such as Cu, Ag, Fe and Sr. First results indicate that the group Ib impurities Cu and Ag mainly substitute for Sr atoms while Fe prefers Ti sites.

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IS360 experiment – Atomic ordering of fluorine dopant in $HgBa_2CuO_{4+\delta}$ and of oxygen in $HgBa_2CaCu_2O_{6+\delta}$ high-Tc superconductors

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Lattice sites and collective ordering of fluorine atoms in oxygen-reduced samples of $HgBa_2CuO_4$, Hg1201, and of oxygen doped samples of $HgBa_2CaCu_2O_{6+d}$, Hg1212, were investigated with the perturbed angular correlation technique by measuring the electric field gradients induced at ^{199m}Hg nuclei. For the Hg1201:F doped samples the experimental data were interpreted with the help of ab-initio calculations of charge distributions for different fluorine configurations in $Hg_mBa_{2m}Cu_mO4_mF_n$, supercells. Internal parameters were allowed to relax, to cancel residual atomic forces due to the dopant. The experimental results show clearly that fluorine occupies only the center of the mercury mesh. For a fluorine content $\delta F > \sim 0.35$ best agreement with theoretical data is obtained under the assumption that fluorine shows a tendency toward ordering along interstitial rows parallel to a, b. No strong ordering of the transferred charge at the copper planes is present in the computed charge density, even when the fluorine atoms order at high concentrations. Therefore, it is concluded that charge stripes observed at the superconducting Cu planes do not seem correlated to the dopant atomistic behavior. For the oxygen doped Hg1212 samples, a new sol-gel method of preparing the precursors by replacing the acrylamide transport agent by urea was implemented with success. After high pressure mercurization the right Hg1212 phase was obtained with high purity and the PAC experiments were done under oxygen pressure. Still under analysis, these results hint the dopant reordering as a function of temperature and concentration – work to be accomplished in 2006.

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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 ferromagnetic insulator manganite $LaMnO_{3.12}$ using PAC spectroscopy. Data analysis reveals a nanoscopic transition from an undistorted to a Jahn-Teller- distorted local environment upon cooling. The percolation thresholds of the two local environments enclose a macroscopic structural transition (Rhombohedric-Orthorhombic). Two distinct regimes of JT-distortions were found: a high temperature regime where uncorrelated polaron clusters with severe distortions of the $Mn^{3+}O_6$ octahedra survive up to T ~ 800 K and a low temperature regime where correlated regions have a weaker JT distorted symmetry. Such work is being extended to several other doping stoichiometry and $^{16}O/^{18}O$ isotopic ratios.

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

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This development is made in collaboration with CERN's Compton camera project on new positron emission tomography (PET) devices http://xray.web.cern.ch/xray/publications/Lyon2000PaperCERNPreprint.pdf, whose technology and detectors also fulfill the requirements for electron emission channeling experiments. In 2005 a 1 mm thick 22×22 Si pad detector was mounted on a newly designed printed circuit board equipped with fast VATAGP3 preamplifier chips. The tests of the VME readout methodology and the readout chain in "sparse" (single channel readout) and "serial" (full readout) modes are now completed. The available data acquisition software for the Compton camera project was modified to better serve the emission channeling purposes for on-line/off-line experiments. Several hardware tests were done with different X-ray sources to study gain linearity, energy resolution (~1.5 keV FWHM at ~17 keV) and to optimize the low energy trigger threshold. The technical design of the detector housing and of a new implantation chamber has been accomplished and will be executed early 2006.

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