

Studies of High- T_C Superconductors Doped with Radioactive Isotopes

REQUEST OF ADDITIONAL BEAM TIME TO IS360 - ADDENDUM 1 -

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ABSTRACT

The IS360 collaboration performs local studies on relevant structural problems of high- T_C superconductors by doping these with radioactive isotopes of appropriate elements. The three main topics are:

- 1) $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta}$: The electric field gradients (EFG) at the Hg site in Hg1201 ($n=1$) provide the local information for characterising the oxygen atoms (O_δ) which go to the Hg-planes and dope the superconducting CuO_2 planes with double-hole charge carriers. EFG measurements at different temperatures have revealed that the charge distribution in the Hg surroundings becomes asymmetric at low temperature. These studies should be continued to further investigate the electronic / structural phenomena that influence, in particular, the Hg neighbourhood. Experiments are proposed to microscopically characterise the F^- and O_δ^{2-} single and double-hole dopant ions in Hg1201, Hg1212 ($n=2$) and Hg1223 ($n=3$), as well as possibly the Cu^{2+} impurity in the Hg planes.
- 2) $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (YBCO): Lattice site, stability and diffusion studies of Hg implanted in YBCO thin films have been successfully performed. The new experiments proposed are an attempt to characterise the lattice site of Ca and Ag ions, and possibly the oxygen dynamics in doped YBCO thin films.
- 3) Infinite Layers Cuprates (ILC): Thin films of $(\text{CaCuO}_2)_n$ and $(\text{SrCuO}_2)_n$ shall be doped with radioactive nuclei of the earth-alkaline (IIA-group) that decay to alkali elements. The purpose is to observe the induction of charge carriers due to the nuclear transmutation. We propose to further investigate the microscopic magnetic properties of the recently developed (atomically engineered) nano-structures of BiSrCaCuO imbedded in ILC materials, which show room temperature resistivity below $10^{-9} \Omega\cdot\text{cm}$.

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1 Introduction

Point-like defects and impurity elements are known to play a significant role in the generation of charge carriers in the high- T_c superconductor cuprates (HTSc) [1, 2, 3]. In many cases the presence of defects can induce structural and local distortions that appear to be as important for the effectiveness of the doping as the impurity valence. The understanding of the structural and electronic mechanisms that regulate the charge transfer from the charge reservoir block layers [1] to the superconducting CuO_2 planes therefore represents a major aspect of the present experimental research. Recently, the observation of charge inhomogeneities in the CuO_2 layers has led to intensive research and models that point to the existence of local and non-linear charge ordering in these compounds [see ref. 4 for a review]. Such phenomena are currently thought to be due to the segregation of holes away from regions of intense local moments. They lead to the formation of “stripes”, i.e. alternating regions at the nanometer scale with different structural and electronic properties. So far it is not known under which conditions such phenomena occur within the materials or whether they are intrinsically related with high- T_c superconductivity, either inhibiting or enhancing it.

By developing the sample preparation procedures, the specific experimental equipment and techniques the IS360 collaboration at ISOLDE is obtaining atomic scale insight into some of the structural and electronic problems in High T_c Superconductors. The use of the γ - γ and e - γ Perturbed Angular Correlation (PAC) techniques [5, 6, 7] working on excited nuclear states, has opened the possibility of probing the local atomic environment via the electric field gradients (EFGs). Also elements which do not have adequate stable isotopes for applying standard techniques like Nuclear Quadrupole Resonance (NQR) can be used. By studying the EFGs, which depend on the probe element surrounding charge distribution and symmetry, we get information about the behaviour of impurity elements and defects that interact with the probing atoms and affect the injection of charge carriers. As a natural output of the technique we further probe local charge asymmetries, which have shown to appear at low temperatures in the Hg family of HTSc compounds. The experimental results of IS360 are compared with physical models and hyperfine fields determined from first-principles band structure calculations. The use of the Emission Channeling (EC) [8, 9] technique allows to determine the lattice site of the implanted probe atoms. Electric/Magnetic measurements of the transport properties and a large variety of standard crystallographic techniques offered by the home laboratories are used to fully characterise the samples.

This addendum first reports the results obtained for each family of high- T_c superconductor materials studied. For each HTSc class it presents the motivation and discusses the experimental program to be continued. A reference list includes the publications and students theses that have originated from this proposal.

Finally we would like to point out that the availability of beams of radioactive isotopes of a big variety of elements with high purity and yields makes the ISOLDE laboratory the unique facility where this experimental program could be envisaged and executed.

2 Results and Work Perspectives

2.1 The $\text{Hg}_1\text{Ba}_2\text{Ca}_{(n-1)}\text{Cu}_n\text{O}_{(2n+2+\delta)}$ compounds

Problem recall:

The $\text{Hg}_1\text{Ba}_2\text{Ca}_{(n-1)}\text{Cu}_n\text{O}_{(2n+2+\delta)}$ family of HTSc compounds has a simple tetragonal structure (Figure 1) and shows (for $n=3$) the highest critical temperature $T_c = 135$ K at ambient pressure or $T_c = 153,157$ K under high-pressure [10]. These facts have motivated an intense research on structural properties and their relationship with superconductivity. It was soon realised, however, that the Hg planes are particularly disordered, being often Hg deficient. There are many reports of impurities replacing Hg, particularly Cu^{2+} and CO_3^{2-} ions. The non-stoichiometric oxygen, O_δ , which is the main impurity that regulates T_c by injecting holes in the CuO_2 planes, also goes to the Hg planes [11]. Not yet understood is the fact that the optimum doping of 0.16 holes per CuO_2 plane is only achieved for O_δ concentrations 50% higher than what would be theoretically needed, assuming that each O_δ ion occupies unique lattice sites and contributes with -2 charges. Recently O_δ has been substituted by F⁻ ions, which have similar chemical affinity to oxygen but differ by one electronic charge. Again it was observed that in order to achieve the effective hole doping a very high fluorine concentration must be present in the material [12].

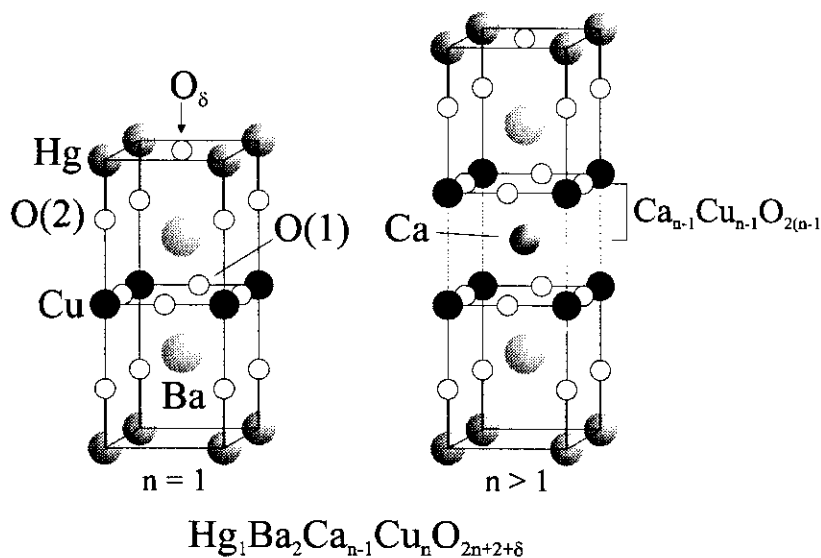


Figure 1 Lattice representation of the Hg-HTSc. Left, ($n=1$, no Ca planes); Right, $n>1$ leads to the intercalation of Ca and CuO_2 planes.

During the last few years neutron diffraction studies performed on powder samples [13] as well as more local methods such as EXAFS [14] have pointed to the existence of local distortions in these compounds. So far it is not clear if and how such effects are linked to the superconductivity mechanism or if they are simply due to impurity chemical effects. Advances in crystal chemistry and probing techniques have created a wealth of information that has led to the concept of spin and charge inhomogeneities (“stripes”) in the field of superconductivity. Stripes are defined as local and non-linear phenomena, which are due to the expulsion of holes from regions of strong local moments. The models suggested propose that holes and spins prefer to live separately in the HTSc materials. Coulomb repulsion, however, frustrates this desire and “stripes” appear as a compromise. These effects should then generate local distortions, which can propagate away from the CuO_2 planes. The resulting zones with different local structural behaviour are difficult to resolve by crystallographic techniques.

We have performed:

PAC measurements with ^{199m}Hg in $\text{HgBa}_2\text{CuO}_{4+\delta}$ ($n=1$, Hg1201) under Ar or O_2 annealings have revealed that the EFGs at the Hg site are very sensitive to the O_δ doping concentration. PAC could provide a local identification of O_δ atoms sitting in the center of the Hg plane and revealed the existence of other yet unidentified oxygen-related defects near the Hg planes (Figure 2) [15, 16]. The interpretation of the EFG data was based on first-principle band structure calculations of the charge distribution in undoped and oxygen doped Hg1201 lattice structures. These calculations have confirmed that Hg is tightly bound to the apical oxygen, whose contributions dominate the EFG at the Hg site. In Hg1201 samples where PAC detects higher oxygen content the EFGs have decreased to lower values, indicating an elongation of the dumbbell of Hg with the apical oxygen. The calculations have further shown that the EFG at the Ba site is extremely sensitive to the O_δ presence. This fact suggests that an experimental effort should be made to measure the EFGs at the Ba site, in order to probe the changes of the Ba local charge density and symmetry as a function of doping and temperature.

The temperature dependence of the EFG at the Hg site has been further studied in Hg1201 at low temperatures. It was found that the asymmetry parameter (η) becomes non-zero already below 160K (Figure 3). Subsequently the charge distribution near the O(2)-Hg-O(2) apical chain becomes non-axially symmetric at low temperatures [17, 18]. This effect occurs in a temperature range where structural anomalies, possibly related with the superconducting transition have previously been observed.

In addendum, IS360 proposes:

To proceed with the systematic study of the EFG at the Hg site as a function of temperature in Hg1201 ($n=1$), Hg1212 ($n=2$) and Hg1223 ($n=3$) for different doping levels with ^{199m}Hg (42.6 m) \rightarrow ^{199}Hg . The aim is to characterise the observed local charge density asymmetries as a function of doping and temperature, in an attempt to relate this microscopic observation with the macroscopic properties of the material all over the superconducting and normal states.

In continuation of our microscopic characterisation of defects we intend to perform PAC experiments in F and Cu doped Hg1201. We expect to detect F-related defects and identify the F lattice site as was possible earlier for O_δ [15, 17]. First PAC experiments with the new ^{80m}Br (4h) \rightarrow ^{80}Br e^- - γ PAC isotope, that has recently been developed at ISOLDE, should be performed.. Br is an isovalent element to F that can be expected to occupy the same site in the Hg1201 lattice, thus permitting to obtain complementary information to that obtained with PAC experiments at the Hg site. Hg1201 samples where Hg is partially replaced by Cu shall also be studied in an attempt to characterise this defect in the Hg neighbourhood.

The Ba site should be highly sensitive to the doping charge carrier transfer mechanism as predicted by the first principle EFG and cluster structural calculations [15, 19]. Moreover, charge inhomogeneities (stripes) in the CuO_2 layer should also strongly affect on the EFG of the Ba site. We therefore propose to initiate studies in Hg1201 with the ^{133}Ba (10y) \rightarrow ^{133}Cs γ - γ PAC probe isotope.

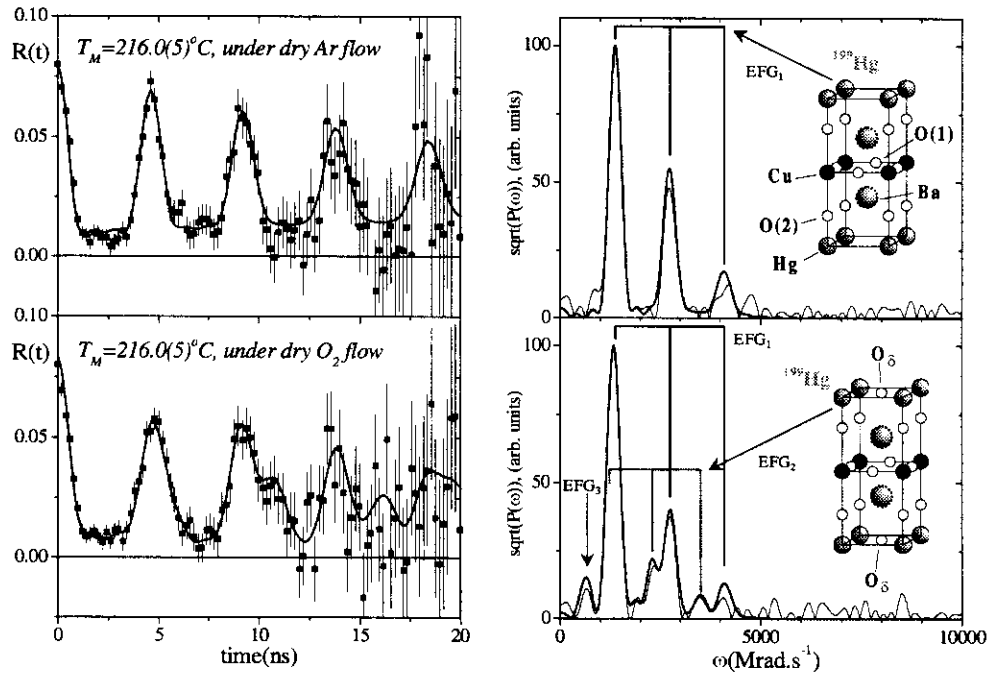


Figure 2 (left) PAC time spectra $R(t)$ and (right) the corresponding Fourier transforms (FT) of the $R(t)$ functions. In the $R(t)$ and the FT spectra the thicker lines represent the fit function and the FT of the fit function, respectively. When annealing under Ar flow, the spectra ($R(t)$ and FT top figures) show a frequency triplet that characterises a highly symmetric EFG $_1$, corresponding to ^{199}Hg nuclei placed on perfect sites with the tetragonal symmetry of the Hg1201 lattice. When annealing under O_2 flow, the spectra ($R(t)$ and FT bottom figures) show two frequency triplets that correspond to two slightly different EFGs. One is still the same highly symmetric EFG $_1$, the second one being a non-symmetric EFG $_2$, which is due to the ^{199}Hg nuclei that interact with O_δ single atoms. Still unidentified oxygen-defects complexes are revealed by a third EFG $_3$.

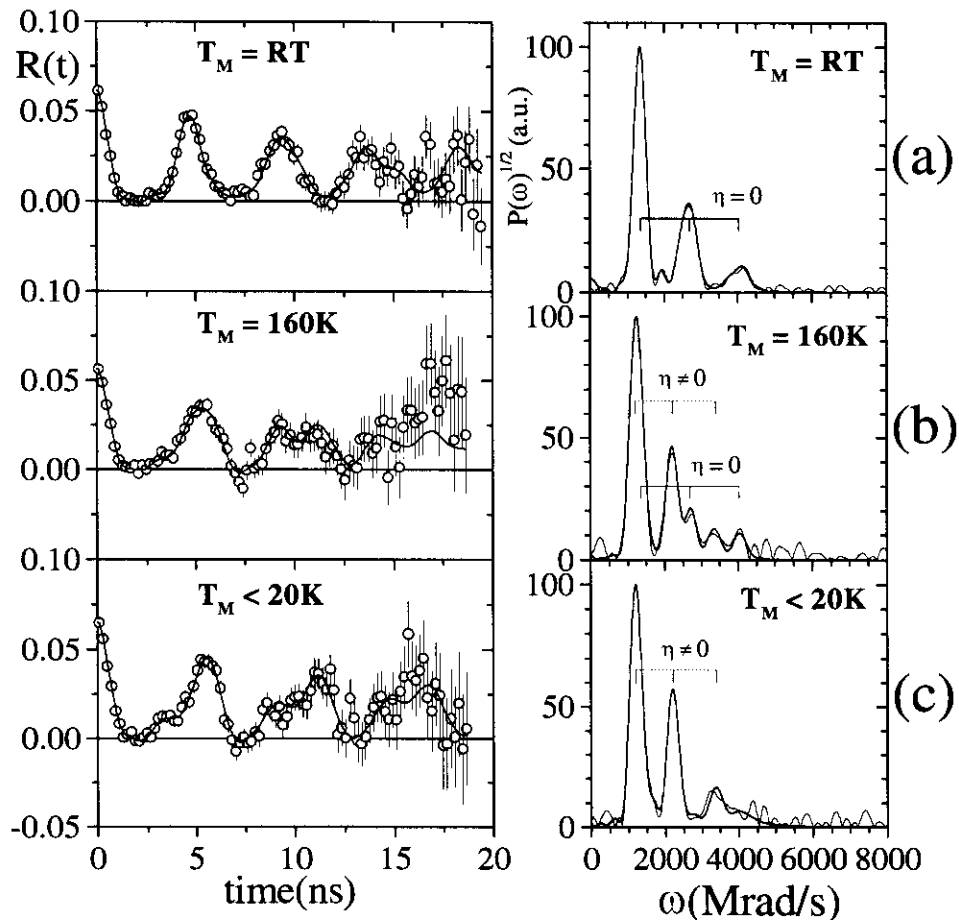


Figure 3 PAC experimental spectra measured after annealing under Ar atmosphere (a) at room temperature (RT), (b) at 160K and (c) below 20K. The spectra show clear differences between the RT and lower temperature measurements. These are due to the appearance of asymmetric EFGs at low temperatures, e.g., $\eta \neq 0$, already at 160K.

2.2 Hg and Ca doping of the $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ compound

Problem recall:

The $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ (YBCO) compound is one of the best studied and interesting HTSc materials for applications. Doping studies have shown that T_C is always lowered except when Au or Hg are incorporated, which increase T_C by 2 K [20] or 10 K [21]. The Hg site, in particular, has never been identified in the YBCO lattice and the presence of different crystalline phases could not be excluded since Hg was added during sintering by changing the reactants composition and stoichiometry.

We have performed:

Combined $e^- \gamma$ PAC (Figure 4) and electron channeling (EC) (Figure 5) studies of $^{197\text{m}}\text{Hg}$ implanted at low doses into high-quality thin YBCO films. The PAC and the EC data analysis and simulations have revealed that Hg is located in Cu(1) sites. The crystalline quality and T_C have been controlled in the as-grown state and after the implantation and subsequent annealing treatments, with X-ray, Rutherford Backscattering/Channeling (RBS/C) and a.c. susceptibility / resistivity measurements. Diffusion coefficients have been measured and Hg was found to be highly stable in the YBCO lattice up to 450°C . Oxygen was further found to hinder the Hg out diffusion [22, 23, 24, 25, 26, 27]. Recent $e^- \gamma$ PAC experiments revealed that a small attenuation that is still observed in several $R(t)$ spectra at room temperature, completely vanishes below 60K. This is probably related with the mobility of the non stoichiometric oxygen in the YBCO orthorhombic phase.

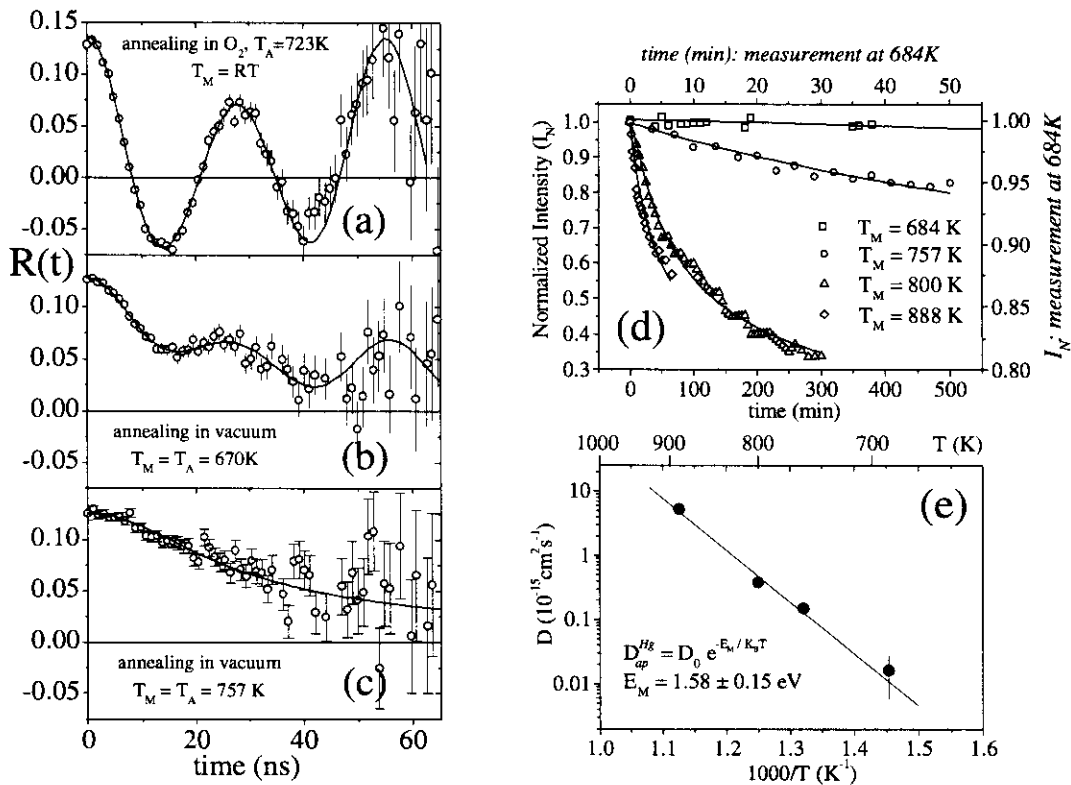


Figure 4 $e^- \gamma$ PAC $R(t)$ experimental functions: (a) measured at RT that reveals one unique lattice site for Hg bound to two oxygen atoms aligned along $\langle 001 \rangle$, (b) $R(t)$ spectrum taken at $T_M = 670\text{K}$ when diffusion starts, (c) $R(t)$ spectrum taken at $T_M = 757\text{K}$. When diffusing through the YBCO lattice the Hg atoms are no more bound to oxygen and interact with much weaker EFGs; (d) normalised $e^- \gamma$ coincidence count rate corrected for the $^{197\text{m}}\text{Hg}$ half-life (I_N) and plotted as a function of annealing time. (e) Arrhenius plot of the apparent Hg diffusion coefficient.

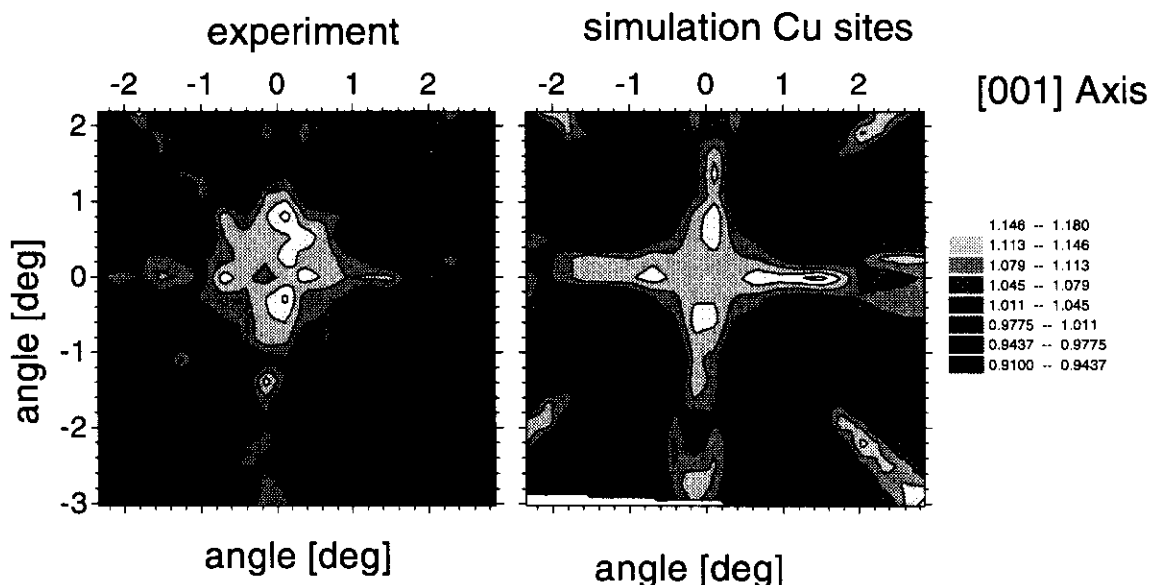


Figure 5 (left) Normalised emission yield of conversion electrons from the decay of ^{197m}Hg ($T_{1/2}=24\text{h}$) as a function of the angle towards the $\langle 001 \rangle$ direction. The right pattern shows a 2-dimensional EC simulation pattern obtained for Hg replacing Cu (1) atoms. The EC analysis and simulations are undergoing, which shall be published in 2001 [26].

In addendum:

The electronic transport properties of grain and thin-film interfaces in high- T superconductors are of crucial importance for numerous applications of these materials, varying from electronic devices to large-current carrying cables and tapes. In particular, electronic band-bending effects, which cause the formation of depletion layers near grain boundaries, are constantly present in the HTSc materials. Such depletion layers have modified carrier densities and for strong enough depletion the diamagnetic superconducting state is driven into the antiferromagnetic insulating state. In YBCO it has been found that by doping during growth with Ca a maximum concentration of 0.3 hole per unit cell can be achieved which compensates band-bending effects and contributes to strongly increase the critical current at low temperatures, relative to the Ca-free material [28]. More recently [29], a substantial increase of the YBCO critical current densities at 77 K was achieved by selectively overdoping the grain boundaries of a mixture of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}/\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ superlattices. In this material Ca preferably diffuses to the grain boundary regions achieving the necessary doping to reduce the band bending and increase the critical current. So far, only based on the similarity of ionic radius, it is assumed that Ca^{2+} firstly replaces Y^{3+} and that for increasing Ca content it starts replacing Ba^{2+} [30]. Also important in Ca doped YBCO, is the oxygen dynamics since the creation of oxygen vacancies in such non stoichiometric materials can inhibit the desired overdoping. Ag is also being used to increase the critical current density of YBCO in melt-textured composites [31], where it further contributes to significantly improve the mechanical properties. However, the superconducting properties of YBCO-Ag composites are still controversial, once the observed critical current increase is probably due to the formation of Ag_2O precipitates that act as pinning centers [32].

IS360 proposes:

To study the dynamics of the mobile oxygen in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{Y}_{1-y}\text{Ca}_y\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$ thin films by using ^{197}Hg as the $e^- \gamma$ PAC probe. Detailed measurements performed, as a function of temperature, in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{YBa}_2\text{Cu}_3\text{O}_6$ shall provide the necessary systematic on this important aspect.

To study the lattice site location of Ca implanted $Y_{1-x}Ca_xBa_2Cu_3O_7$, EC experiments shall be done after implantation of ^{45}Ca (163d) \rightarrow ^{45}Sc and subsequent annealing of $Y_{1-x}Ca_xBa_2Cu_3O_7$ high quality, well characterised thin films grown along the c axis (001) and the a axis (100) (lattice projection schema are shown in Figure 6). Surface grown (100) thin films are needed to distinguish the Ca occupation at Y or Ba rows once that for other higher index axis the EC patterns are more difficult to simulate. The existence of two mirror symmetry planes allows considerable simplification on the emission channeling pattern simulations. These experiments should provide information on the Ca lattice site as a function of Ca concentration. Looking forward to characterise the microscopic environment of Ag in YBCO-Ag composites, we propose to use the ^{111}Ag (7.45d) \rightarrow ^{111}Cd PAC and EC probe isotope.

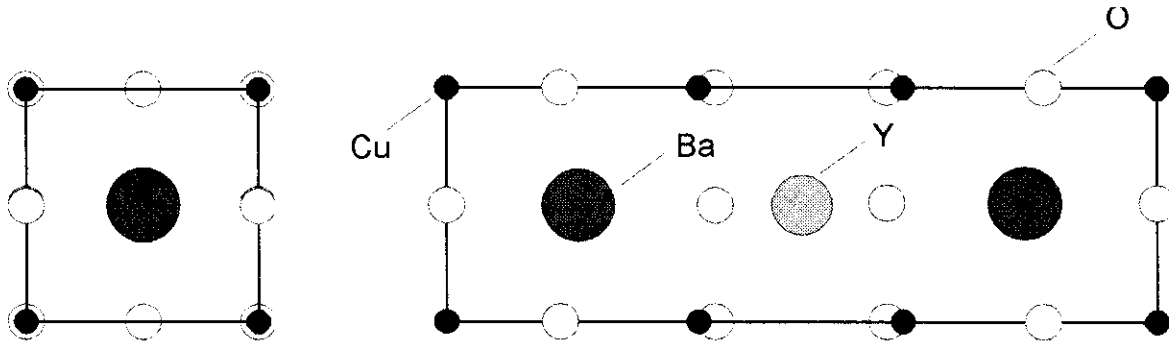


Figure 6: YBCO lattice projection of the plane perpendicular to the <001> (left) and to the <100> axis (right).

2.3 The doping of the Infinite Layer Cuprates

Problem recall:

The Infinite Layer Cuprates (ILC) are the simplest structures of the HTSc compounds, being only made of CuO_2 planes which alternate with planes of earth-alkaline elements (Ca, Sr and Ba) (1 and references therein). Consequently, the ideal ILC structures are not expected to generate superconducting carriers once the charge reservoir layers are absent. However, superconductivity ($T_C=110$ K) has been observed after introducing vacancies at Sr-Ca sites in $((Ca_x,Sr_x)_{1-y}CuO_2)_n$ [33] or when doping with rare-earth elements at the Sr site ($T_C = 40$ K) [34]. Puzzling results have been observed in single crystals where superconductivity has been detected although the structural analysis did not show any cation deficit or parasitic phases [35]. In fact, it is not well known if the superconductivity is due to impurity elements, point-like defects or to deficient Ca, Sr or Ba extended defects.

In the past few years the use of sophisticated techniques, like molecular beam epitaxy (MBE), has allowed the preparation of high-quality ILCs thin films as well new ILC-based atomic engineered structures. Among such we mention the intergrowth of $BiSrCaCuO$ nano-structures embedded in $SrCaCuO$ ILC matrix. These materials show room temperature resistivity below 10^{-9} $\Omega.cm$ and non-linear I (V) current / voltage relationship that suggest a superconductor behaviour [36]. So far, susceptibility measurements have not been able to show relevant diamagnetic properties because of the interference of the substrate signal.

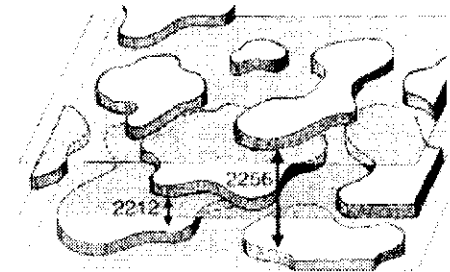


Figure 7 Artistic representation of the $BiSrCaCuO$ nano structures embedded in $SrCaCuO$ ILC matrix.

IS360 proposes:

The possibility of observing p-type superconductor carriers due to valence changes in the ILC earth-alkaline layers, by replacing Ca, Sr or Ba (2+) with alkaline ions (1+) would contribute to identify point-like doping centers. By doping thin films of $(R(\text{CuO}_2))_n$, $R = \text{Ca}$ and Sr, with $^{85}\text{Sr}(64.8 \text{ d}) \rightarrow ^{85}\text{Rb}$ and performing the measurement of the superconducting properties as a function of time will allow to distinguish between the effects caused by the doping and the ones caused by the residual implantation damage. The implantation control will be performed with EC experiments on $^{89}\text{Sr}(50.5\text{d}) \rightarrow ^{89}\text{Y}$ and $^{86}\text{Rb}(18.8\text{d}) \rightarrow ^{86}\text{Sr}$, with PAC on $^{79}\text{Rb}(23\text{m}) \rightarrow ^{79}\text{Kr}(35\text{h})$ and with RBS/C and X-ray experiments. In the BiSrCaCuO-ILCs nanostructures it is our aim to implant low concentration of $^{79}\text{Rb}(23\text{m}) \rightarrow ^{79}\text{Kr}(35\text{h})$, $^{197\text{m}}\text{Hg}(224\text{h}) \rightarrow ^{197}\text{Hg}$ and $^{80\text{m}}\text{Br}(4\text{h}) \rightarrow ^{80}\text{Br}$ PAC isotopes, which could reveal the presence of magnetic fields to be studied as a function of temperature.

Comment:

Figure 8 shows an emission channeling experiment performed on ^{45}Ca implanted $\text{Sr}_{0.9}\text{La}_{0.1}\text{CuO}_2$ (La doped Sr-ILC) during the $^{45}\text{Ca}(163\text{d}) \rightarrow ^{45}\text{Sc}$ decay. This test aims to learn about the possibility of introducing Ca (isovalent to Sr) into regular lattice sites by ion implantation. In spite of the fact that the spectra show a significant fraction of the Ca in regular sites of the lattice, the film had bad epitaxy quality what explains the poor anisotropy of the EC patterns in off surface directions and inhibited further detailed analysis. In fact, during the previous years the collaboration has faced difficulties on regularly obtaining high-quality reproducible samples and when shipping the implanted samples to foreign universities (Porto/Portugal and Texas/EUA). The reception of the samples has been delayed several times and the measurement of their electric/magnetic properties could not be performed on time.

Such difficulties have been recently surmounted with the new collaboration of the Laboratory of Surfaces et Supraconducteurs, C.N.R.S. (PARIS) which is currently and reliably producing high quality thin film samples of the ILC and the BiSrCaCuO - ILC compounds with the MBE technique. Paris is easily reachable from CERN where the full characterisation of these samples shall be done before and after they are doped at ISOLDE. Additionally, IS360 has permanently installed at ISOLDE a four-contact probe resistivity measurement that is coupled to a closed cycle He refrigerator and allows in-situ and complementar resistivity measurements.

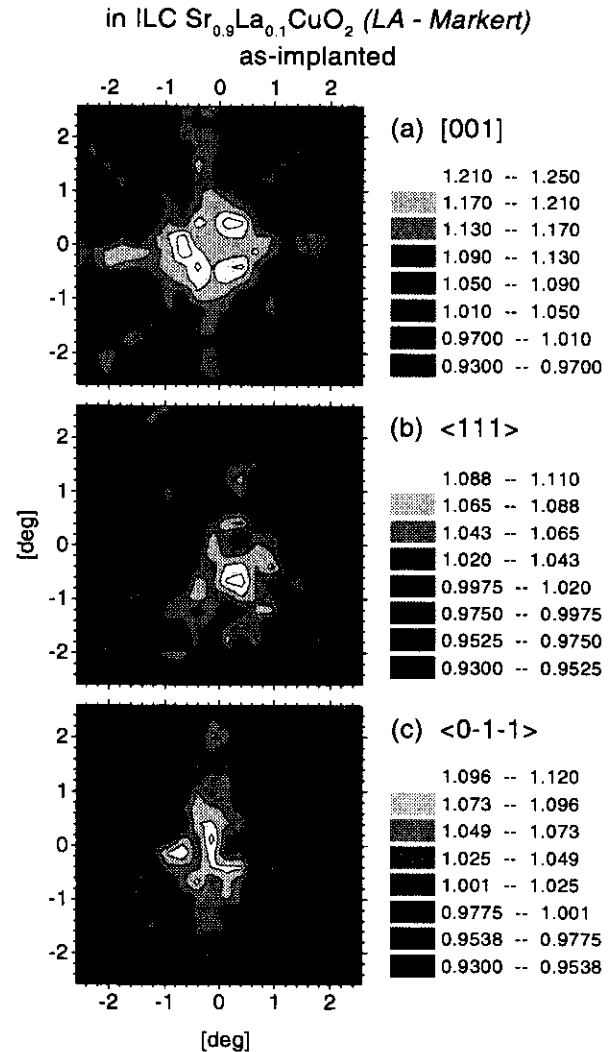


Figure 8 Normalised emission yield of β^- emitted from the decay of $^{45}\text{Ca}(163\text{d}) \rightarrow ^{45}\text{Sc}$ as a function of the angle towards the $\langle 001 \rangle$, $\langle 111 \rangle$ and $\langle 0-1-1 \rangle$ directions.

3 Experimental Facilities

3.1 Sample production and characterisation

Table I: Where and how the samples are produced

family	samples	Laboratory			
		Grenoble (CNRS)	Moscow (State Univ.)	Paris (CNRS)	Max Plank Institute
Hg12(n-1)n n=1,2,3	Pellets and powder	High Pressure Solid State Reaction	Sealed Quartz tubes with O2 controlled pressure [37]	-	-
ILCs and BiSrCaCuO/ILC	Thin films	-	-	Molecular Beam Epitaxy	-
Ca and Ag doped YBaCuO	Thin films	-	-	-	Pulsed Laser Ablation

All samples are characterised at the home institutes, before and after the experiments with radioactive isotopes. Available characterisation techniques are: X-ray diffraction, Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM) and High Resolution TEM and Rutherford Backscattering/Channeling (RBS/C), which allow to monitor the sample's crystalline structure, orientation, composition, as well as the characterisation of the implantation profile and residual damage from the ion implantation and annealing procedures. The transport properties are measured at the home-institutes. In thin films T_c is controlled by measuring the resistivity with the four-probe contact technique and magnetic susceptibility measurements, performed with the χ_{ac} susceptibility or SQUID techniques. In most powder samples only the χ_{ac} susceptibility will be measured. For in-situ control of some of the thin film samples, IS360 has installed at ISOLDE a four-contact probe resistivity measurement that is coupled to a closed cycle He refrigerator.

3.2 Nuclear techniques working with radioactive isotopes

3.2.1 Perturbed Angular Correlations (PAC) and Emission Channeling (EC)

Both the γ - γ and e^- - γ PAC techniques are well established at ISOLDE. Table II shows the PAC spectrometers which are permanently installed at CERN and the employed isotopes. All spectrometers are equipped with sample holders that can be heated (up to 800 K) and cooled (below 20 K).

Table II: PAC spectrometers

Isotope	PAC spectrometer (provenience)	measuring atmosphere	ISOLDE beam coupling
^{199m}Hg (42.6 m) \rightarrow ^{199}Hg	γ - γ (Leipzig) + e^- - γ	vacuum / gas flow	off-line
^{197m}Hg (24h) \rightarrow ^{197}Hg	e^- - γ (Lisbon)	vacuum	off-line
^{133}Ba (10y) \rightarrow ^{133}Cs	γ - γ (Leipzig)	vacuum / gas flow	off-line
^{111}Ag (7.45d) \rightarrow ^{111}Cd	γ - γ (Leipzig)	vacuum / gas flow	off-line
^{80m}Br (4h) \rightarrow ^{80}Br	e^- - γ (Lisbon)	vacuum	off-line
^{79}Rb (23m) \rightarrow ^{79}Kr (35h)	γ - γ (Leipzig)	vacuum / gas flow	on-line

The EC experiments are done using the two-dimensional Si pad detector developed at CERN [9]. All experiments are carried off-line using, particularly, the new emission channeling setup that was afforded by the Lisbon group, which consists of a two-axes goniometer equipped with in-situ heating up to 1200 K.

3.2.2 Correlation of the probing isotopes with the experimental techniques

Table III: Radioactive isotopes and techniques

isotope	annealing	γ - γ PAC	e^- - γ PAC	EC (*)	E&M (**)
^{199m}Hg (42.6m) ↓ ^{199}Hg	✓	✓	✓	-	-
^{197m}Hg (23.8h) ↓ ^{197}Hg (64.1h) ↓ ^{197}Au	✓	-	✓	✓(c.e.)	-
^{133}Ba (10y) ↓ ^{133}Cs	✓	✓	-	-	-
^{111}Ag (7.45d) ↓ ^{111}Cd	✓	✓	-	✓(β^-)	✓
^{89}Sr (50.5d) ↓ ^{89}Y	✓	-	-	✓(β^-)	✓
^{86}Rb (18.8d) ↓ ^{86}Sr	✓	-	-	✓(β^-)	✓
^{85}Sr (64.8d) ↓ ^{85}Rb	✓	-	-	-	✓
^{80m}Br (4h) ↓ ^{80}Br	✓	-	✓	-	-
^{79}Rb (23m) ↓ ^{79}Kr (35h) ↓ ^{79}Br	✓ on-line	✓	-	-	-
^{45}Ca (163d) ↓ ^{45}Sc	✓	-	-	✓(β^-)	✓

(*) Electron channeling performed with conversion electron (c.e.) or β^- .

(**) Experiments where the electrical and magnetic properties are measured as a function of (decay) time.

3.3.1 Equipment and Laboratories

-The e^- - γ spectrometer setup occupies about 9m^2 . Due to its weight (~ 1000 Kg) and different components the alignment procedure is a delicate time-consuming task (~ 1 week) and the spectrometer should not be moved between runs. Due to the installation of the REX accelerator at the ISOLDE hall no beam line or empty space was left to run the e^- - γ PAC spectrometer on-line or off-line. This avoided, particularly, that experiments with the short lived ^{127}Ba (13m) \rightarrow ^{127}Cs (6.2h) could be made. However, during the 1998/1999 shutdown this situation was partially solved by the combined efforts of the EP Division, TIS-Radioprotection and the ISOLDE management that provided the access to a radioactive "class C" laboratory in building 304/R-012. To all of them we address special thanks. In there the e^- - γ PAC spectrometer, the new EC Lisbon setup, the 6-gamma

detector PAC spectrometer from the Leipzig group [38] and the closed cycle refrigerator that is equipped with a four probe head for performing resistivity measurements are mounted and running full time. These setups are often shared by different solid state physics approved experiments.

-During the Hg run the 6-gamma detector PAC spectrometer should operate (as usually) in building 3-1-039. This room is a radioactive laboratory specially ventilated, further equipped with glove boxes (where the HTSc are stored and handled) and several gas huts. Such conditions are required when performing PAC experiments on delicate samples to be annealed and measured under gas flow.

-During the ^{79}Rb run one on-line position is needed for mounting the 6-detector PAC spectrometer.

-Collections of the long-lived isotopes for EC and for measuring electrical and magnetic properties will be done in the “old” Solid State implantation chamber or in the “new” ISOLDE collection point.

-Several furnace systems exist already at ISOLDE for annealing treatments under vacuum or under 1bar gas flow. When annealing under (Ar, N₂, O₂) gas flow the furnaces are equipped with activated charcoal filters that trap volatile elements.

3.3.2 Beam time request

Table IV: Beam time request of 26 shifts for the next two years.

type of samples	isotope	intensity [at/ μC]	target material	ion source	Shifts
Hg-HTCs	$^{199\text{m}}\text{Hg}$	$\sim 3 \cdot 10^8$	molten Pb	Hot plasma	10
Bi-ILC	$^{197\text{m}}\text{Hg}, ^{199\text{m}}\text{Hg}$	$\sim 5 \cdot 10^8$	molten Pb	“	2
Hg-HTCs	^{133}Ba	$\sim 10^8$	Ta foil	W-surface ion.	1
YBCO	^{111}Ag	$\sim 3 \cdot 10^9$	UC ₂ or ThC	Laser (Ag)	1
	^{89}Sr	$> 10^8$	Nb foil, ZrO fibbers or UC ₂	surface ion.	1
ILC	^{86}Rb	$\sim 3 \cdot 10^8$	Nb foil or ZrO fibbers	“	1
	^{85}Sr	$\sim 2 \cdot 10^9$	Nb foil	“	2
Hg-HTCs, Bi-ILC	$^{80\text{m}}\text{Br}$	$\sim 10^9$	Nb foil or ZrO fibbers	Hot plasma	2
ILC, Bi-ILC	^{79}Rb	$\sim 10^9$	Nb foil or ZrO fibbers	“	4
YBCO and ILC	^{45}Ca (as ^{45}K)	$\sim 10^8$	UC ₂ or ThC	W-surface ion.	2

Most of our sample preparations require collection times of the order of 5 to 60 min per sample, every 4 to 24 hours measurement periods. Therefore all beam times should be shared with other users. Only the on-line $\gamma\text{-}\gamma$ PAC measurements with ^{79}Rb require dedicated beam line periods of about three hours per sample.

4 References:

Students Thesis:

Diploma work	1997 - 1997 - João Pedro Esteves de Araújo	- IFIMU Porto / Portugal
Travail de Stage	1998 - 1998 - Magali Brunet	- LPM-INSA Lyon / France
MsC. Thesis	1999 - 2001 - Fernando do Espírito Santo	- DCUA Aveiro / Portugal
Ph. D. Thesis	2001 - “ “ “ “	
Ph. D. Thesis	1998 - 2001 - João Pedro Esteves de Araújo	- IFIMUP Porto / Portugal
Ph. D. Thesis	2001 - - Armandina Maria Lima Lopes	- DFUA Aveiro / Portugal

Legend: Underlined references were produced under this proposal concepts.

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