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Perturbed angular correlations at ISOLDE: A 40 years young technique

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The idea that “new-is-small” is a paradigm propelling industries and research: new materials for new applications and new technologies. Precise and efficient characterization techniques are, therefore, required to make the “new” and the “small”, understandable, applicable, and reliable. Within this concept, Time Differential Perturbed Angular Correlations, TDPAC, appears as one of the most exotic and efficient techniques to characterize materials and is celebrating 40 years at ISOLDE, CERN. In this overview we explore the TDPAC measurement possibilities at ISOLDE-CERN for solid state physics research with a rich potential due to the wide number of available radioactive probe elements, delivered with great purity and high yield. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.4994249>

The increasing usage of nanomaterials or nano-scaled materials in high-tech applications requires that the underlying physical phenomena are studied on an atomic scale. This has to be done with methods not influencing the subject of interest itself. Consequently, it becomes more and more difficult for the characterization of smaller and smaller systems. In this concept, single atomic probes are becoming more and more useful and important. A single atom in the system gives all information which is needed. In fact, tip-less techniques are required, not interfering with the system under study. Interestingly, such an approach has long been realized using nuclear probes, where radioactive nuclei interact with their surroundings in a solid on an atomic scale and transmit this information via their radioactive decay complementing current macroscopic techniques. Being a hyperfine interactions technique, unique or complementing Mössbauer effect, Nuclear Magnetic Resonance, and Nuclear Orientation, TDPAC probes atomic scale phenomena of (bio)molecules, gases, and solid state materials from 1 K up to 1500 K.

The Resonance Ionization Laser Ion Source (RILIS)¹ is used to increase the beam purity and efficiency at ISOLDE.² This leads to the diversity of research subjects and applications of the TDPAC technique.³⁻⁵ Inherently, the experiments require the introduction of typically 10^{10} – 10^{12} radioactive probe atoms into the material under study.

Different methods have been applied worldwide for improving isotope incorporation in order to achieve a diluted condition. In this way, ion implantation,^{6,7} diffusion,⁸ irradiation,⁹ chemistry,⁵ including sol-gel,¹⁰⁻¹² and in-growth methods¹³ are being used. Furthermore, the probe concentration is as low as around 10^{10} atoms, so that interference with properties of the sample and doping levels can be excluded. When the active elements are introduced by implantation additional perturbations to the TDPAC signal possibly arise due to local damage. Thermal annealing is well known to remove such radiation damage very effectively leaving the probe site unperturbed afterwards.

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ISOLDE offers a large number of different radioactive isotopes that can be used for TDPAC and, therefore, the most suited element/isotope can be chosen to study atomic phenomenology of a sample at its one scale. Samples can be prepared directly by ion implantation or by chemical methods, being the only laboratory in the world with the instrumentation to carry out gamma-gamma, (γ - γ), conversion electron-gamma, (e_c - γ), and beta-gamma (β - γ), TDPAC measurements. Furthermore, the measurement facilities at ISOLDE comprise several digital^{14–17} and conventional analogue experimental setups with 4 or 6 gamma-detectors. One electron-gamma¹⁸ and one beta-gamma spectrometer are also available. A (4-/6-) detectors TDPAC-spectrometer, produces 12/30 coincidence spectra, which are not all similar. There are a set of similar spectra taken with detectors at 90 degrees and another set of spectra taken at 180 degrees. These two sets of spectra are combined to build the $R(t)$ anisotropy ratio function, i.e., the observable containing all the information regarding the coupling of the nuclear moments with the hyperfine magnetic field and electric field gradient. This experimental $R(t)$ function is fitted by a theoretical function using a specific software, such as NNfit^{19,20} or Nightmare,²¹ generally custom-made and adapted to the studied problem. It considers magnetic and quadrupole interactions and dynamic effects aiming to extract information at the nanoscale. This concerns local magnetism, probe lattice site, local deformations, electronic distribution, and its dynamics in the neighborhood of the probe in molecules, on surfaces, and in bulk matter. All the experimental results can be then interpreted by comparison with different possible configurations of environments around the probe nuclei in the host materials, generally using first principles methods based on Density Functional Theory (DFT) available in several commercial or research dedicated packages.²²

In fact, TDPAC measurements have a very local character and can provide important information on the environment around the probe nuclei. However, additional macroscopic measurements are necessary to characterize the samples in terms of major crystal structure, electromagnetic phase transitions, morphology, stoichiometry and composition. In addition, using the ideal isotope, key microscopic features can be investigated, such as lattice location, diffusion, interaction with defects present in the neighborhood of the probe atom, magnetic properties, percolation phenomena leading to structural and magnetic phase transitions, dopant incorporation, and transport phenomena, such as those in bulk nanomaterials and near surfaces or interfaces. TDPAC experiments just perfectly match this concept. Details about the technique can then be found in the Refs. 23–28. Figure 1 illustrates the principle of the TDPAC technique.

Conventional TDPAC experiments aim to find the probability emission of two successive gamma-rays emitted in cascade. Since both the magnetic and the electric hyperfine interactions exercise a torque on the angular momentum of the intermediate state, the population of the m states is reordered. Detecting the first gamma sets a reference direction to where the population of the m -states in the intermediate state is unequally distributed. The second gamma detection is generally anisotropic in space and is angularly correlated with respect to the first. Taking profit from the existence of an extranuclear electric field gradient (EFG) and/or magnetic field (B_{hyp}), which sets the hyperfine interaction, the TDPAC spectra show the resulting time dependent redistribution of the m -substates population. In other words, the spectra register the perturbation that changes the angular correlation

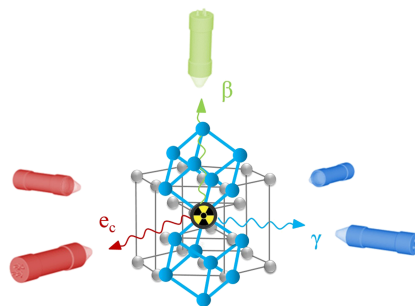


FIG. 1. Principle of the TDPAC technique: the probe atom is placed inside the material under research and emits radiation in cascade, which is detected by scintillators.

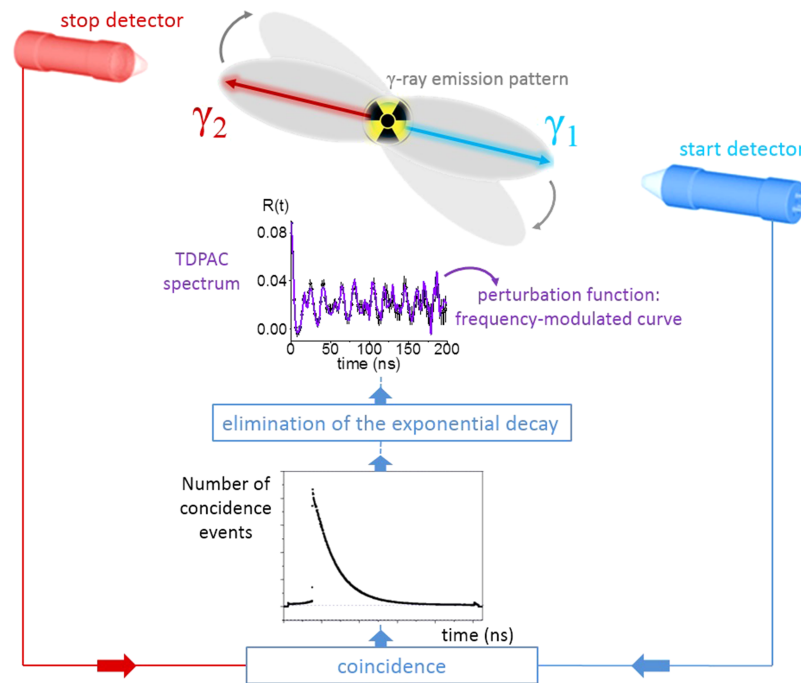


FIG. 2. Basic representation of the γ - γ TDPAC technique: detection and coincidence of the rotating emission gamma-cascade pattern, elimination of the exponential decay and generation of frequency-modulated TDPAC curve of the intermediate state.

function and characterize the charge asymmetry and magnetic fields aimed to be probed. From such *time-shaping* of the angular anisotropy of the γ , e_C , β radiations it is possible to determine the hyperfine field B_{hyp} and field gradient EFG.

Figure 2 shows the schematic representation of the TDPAC measurement. Transient phenomena occurring at the time scale of the measurements can also be observed and studied. In a simple way, the measurement of the TDPAC observable function consists of a time differential analysis of the variation of the angular anisotropy of γ , or e or β , radiation followed by a gamma-ray on the particular decay cascade.

Usually, nuclei that decay through a cascade of two gamma rays are used, in which the energies, multipolarity, spins, magnetic, and quadrupole moments are well known.²⁹ A big asset of being at ISOLDE is the possibility to test the feasibility of new TDPAC isotopes and determine the still unknown nuclear parameters³⁰ of new probes from potentially interesting elements. Lattice distortion and defects,^{31,32} polaron excitations,³³ orbital ordering,³⁴ diffusion³⁵ and/or electronic polarization can be investigated as a function of temperature,²⁰ magnetic field³⁶ or compressive loads.³⁷ These are a few examples of use of the worldwide well-established TDPAC technique.

At ISOLDE, many different gamma-gamma isotopes can be produced. The conventional probes are ^{111}In ,³⁸ $^{111\text{m}}\text{Cd}$,³⁹ and ^{111}Ag .⁴⁰ More exotic examples comprise $^{199\text{m}}\text{Hg}$,⁴¹ $^{204\text{m}}\text{Pb}$,⁴² ^{117}Cd ,⁴³ and $^{68\text{m}}\text{Cu}$.³⁰ ^{204}Pb also appears as a very effective exotic probe, because of its greater sensitivity to deviations from axial symmetry than other usual TDPAC probes.⁴⁴ Also the long half-life of 260 ns should allow to measure very weak EFGs characterized by low frequencies in the TDPAC observable function. Interesting, ideal target conditions opens interesting perspectives for studies profiting from the possibility of simultaneous ^{111}In and $^{111\text{m}}\text{Cd}$ implantation, only possible at ISOLDE.³⁸ Moreover, the TDPAC laboratory is able to measure all suitable isotopes, independently of ISOLDE production possibilities. This includes ^{181}Hf that can be implanted at the Bonn Radioisotope Separator (BONIS) located at Helmholtz-Institut für Strahlen-und Kernphysik (HISKP), which is an ion implanter used for the investigation of materials for about 50 years.^{45,46}

The beta-gamma TDPAC measurement is based on parity violation due to the weak interaction responsible for the beta decay.⁴⁷⁻⁵⁰ For standard γ - γ or e_C - γ TDPAC experiments, where the electromagnetic force is the only responsible interaction for the anisotropy function, this is described

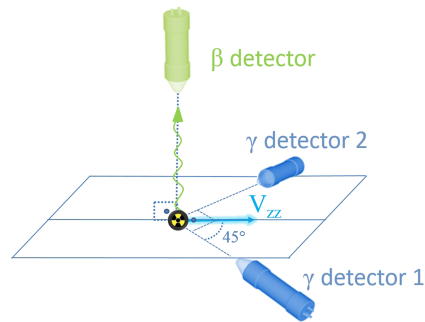


FIG. 3. Basic schema of the β - γ TDPAC setup: the different precessions of the γ emission pattern can be determined by a suitable arrangement of β and γ detectors, which are perpendicular to one another. This measurement allows the determination of the V_{zz} sign.

by even cosine functions. For β - γ angular correlations the odd sine function prevails as the time observable. From such measurements it is possible to determine the sign of the V_{zz} component of EFG, provided the sign of the quadrupole moment is known.⁴⁷ However, only single crystals can be used, since the EFG principal system of axes must be properly oriented regarding a special geometry alignment of the beta and gamma detectors,^{47,50} as shown in Figure 3. For such experiments the statistics is generally poor, since only two, β - γ detector combinations are used. Consequently, this technique has not often been used. It was mainly required when the sign of the EFG components was a determinant for the interpretation of defect complexes, such as the work performed in recent times at the HSKP, University of Bonn.^{47,50} Recently, the Bonn detector setup has been transferred to ISOLDE where several β - γ TDPAC cases can easily be tested. In this concept, the $^{111}\text{Ag}/\text{Cd}$ probe has often been used in solid state and soft-matter/chemistry experiments. It is introduced into the samples via diffusion, implantation, or chemical reactions. ^{111}Ag is obtained at ISOLDE or at reactors for measuring γ - γ and β - γ TDPAC. Only 7% of ^{111}Ag decay to the $3/2^+$ with 342 keV, first state of the appropriate γ - γ TDPAC cascade, in which the probing $5/2^+$, 245 keV excited state on ^{111}Cd is the same nuclear level as when using $^{111}\text{In}/\text{Cd}$ or $^{111\text{m}}\text{Cd}/\text{Cd}$ probes. However, the 7.5 days half-life of the ^{111}Ag nucleus, the 84 ns half-life of the probing state, and the magnitude of the quadrupole moment $Q = 0.77$ (12) b⁵⁰ are clearly enough to provide a high quality signal that outweighs the poor statistics. Figure 4 illustrates different patterns of the hyperfine splitting for γ - γ or β - γ cascades. The partial nuclear polarization characteristic of the β -decay causes a non-uniform population of the magnetic substates in comparison to the γ - γ cascade.⁴⁷

In addition, $^{115}\text{Cd}/\text{In}$ with 53.46 hours half-life, is also well produced at ISOLDE. Its decay cascade starts by an allowed beta decay ($I = 1, P = 0$),⁴⁷ with 3.3% feeding to the $3/2^+$, 828.58 keV, 5.78 ns probing state of ^{115}In with $Q = -0.60$ b. By means of β - γ TDPAC using the radioactive probes ^{111}Ag and ^{115}Cd ^{47,50} the sign determination of the electric field gradient of the GaN and AlN lattices were successfully obtained demonstrating the feasibility of the technique.

As already mentioned, the conventional γ - γ TDPAC works by detecting two sequential gamma quanta, emitted upon decay of a cascade from an excited state. Alternatively, a conversion electron (*c.e.* or e_c) emitted in competition with the gamma ray of a certain transition of the decay cascade can be used and correlated with another gamma ray or another c.e. from a different cascade transition.⁵¹ Therefore, e_c - γ or γ - e_c , e_{c1} - e_{c2} TDPAC are experimental ways to complement conventional



FIG. 4. Schematic illustration of different hyperfine splitting patterns for the intermediate state of β and γ cascades.

measurements by extending the number of available probe nuclei, such as $^{73}\text{As}/\text{Ge}$, $^{77}\text{Br}/\text{Se}$, $^{80\text{m}}\text{Br}/\text{Br}$, $^{197\text{m}}\text{Hg}/\text{Hg}$, $^{119\text{m}}\text{Sn}/\text{Sn}$ and $^{127}\text{Ba}/\text{Cs}$. Conversion electron TDPAC was intensively used in the seventies at the HISKP - University of Bonn using the Kleinheinz-Siegbahn spectrometer⁵² that was later upgraded and installed at ISOLDE.^{53–55} This spectrometer consists of an array of two magnetic lenses of Siegbahn type for detection of conversion electrons^{52–55} and two BaF_2 scintillators for gamma detection, arranged in a plane as a standard 4-detector machine as shown in Figure 5. The samples are placed inside a vacuum chamber at the center of the detectors, with the implanted side facing both magnetic lenses. In the present setup samples can be measured as a function of temperature from 30 K up to 900 K.

Examples of probe nuclei, where only the conversion electron method works, are $^{73}\text{As}/\text{Ge}$ (80.3 d)⁵³ and $^{119\text{m}}\text{Sn}/\text{Sn}$ (293 d).⁵⁵ The $^{73}\text{As}/\text{Ge}$ electron capture nuclear transmutation provides a unique case of a semiconductor TDPAC probe. Via its 100% decay to a first 66.7 keV long lived (0.5 s) state on ^{73}Ge , allowing the full reconstruction of the electronic shells, the decay follows a first 53 keV M2 gamma ray, being the second E2 transition of 13.3 keV fully converted. In this case, it is necessary to use γ - e_c coincidences to measure the perturbation function. The 2.86 microseconds half-life of the intermediate 13.3 keV state provides high resolution which can be used to identify very low electric field gradients or magnetic fields.⁵³

The particular case of the isomeric $^{119\text{m}}\text{Sn}/\text{Sn}$ decay requires the use of e_c - γ coincidence experiments due to the very high conversion coefficient of the first 65 keV M4 transition.⁵⁵ This allows studying hyperfine interactions combined with complementary Mössbauer experiments on the same probe. This can be particularly interesting in measurements performed as a function of temperature, since the quality of the TDPAC signal is not affected by temperature.

Last but not least, there is a unique feature of e_c - γ experiments that does not manifest itself on γ - e_c and γ - γ suitable cascades, being a unique way to induce excited electron holes in the atomic shell of the probe atom before the measurement starts, and to look at the subsequent electronic recombination mechanisms. The observation window is limited by the time resolution of the experiment, typically of 1 ns. Moreover, the observation of transient fields depends on the electron mobility and carrier concentration in the host – but as well on the relationship – probe-impurity-host, particularly from the existence of long lived local electronic excited states which can be probed in this way.⁵⁶ If recombination processes are too fast, like in metals and some semiconducting materials, e_c - γ and γ - γ methods show no differences on the observable functions.^{57,58} On nuclei where both e - γ and γ - γ experiments can be performed, such as $^{111\text{m}}\text{Cd}/\text{Cd}$, $^{199}\text{Hg}/\text{Hg}$, $^{80\text{m}}\text{Br}/\text{Br}$, $^{181}\text{Hf}/\text{Ta}$, $^{127}\text{Cs}/\text{Ba}$, $^{100}\text{Pd}/^{100}\text{Rh}$, these studies provide a unique and unequivocal identification of carrier mobility and lifetimes of electronic excited states at the probe atom.^{56–58} Figure 6 shows the pictures from 1999 of one analogue TDPAC setup that is still used at ISOLDE nowadays. It has been made for two systems of 4-detectors. At the time of writing, ISOLDE has received one more digital setup from the University of Göttingen, which is shown in Figure 7. This machine¹⁶ is already in use and is one more contribution from the Federal Ministry of Education and Research (BMBF) for allowing the community to apply ever-more exotic isotopes to solid state physics.

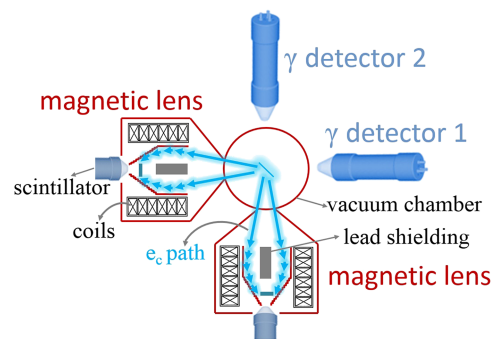


FIG. 5. Basic representation of the current e - γ TDPAC setup: a vacuum chamber, two magnetic lenses, two gamma detectors and plastic scintillators coupled to photomultiplier tubes. A favourable complement to the γ - γ TDPAC method.

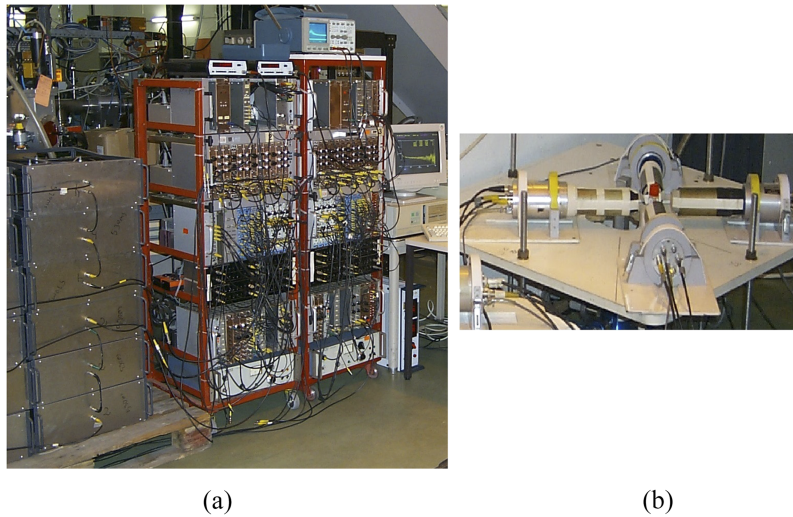


FIG. 6. Analogue TDPAC setup at the ISOLDE hall made for two systems of 4-detectors. Photos taken in 1999. (a) Electronic system with delay boxes; (b) one of the four detectors setup.

Concluding, TDPAC offers a unique probing method – at the (sub)nanoscale - where each radioactive nucleus *talks* by itself without the intervention of external tips and provides fundamental information on properties of materials under study. The whole process works at low concentrations (ppm) of the radioactive probing element. The TDPAC world at ISOLDE³ stands on individuals and collaborations with the know-how and expertise of both material and nuclear applied sciences, the whole relying on ISOLDE-CERN, a unique radioactive beam facility providing a multitude of elements and isotopes adequate to the use and development of the TDPAC technique in ideal experimental conditions for over 40 years.^{59–62}

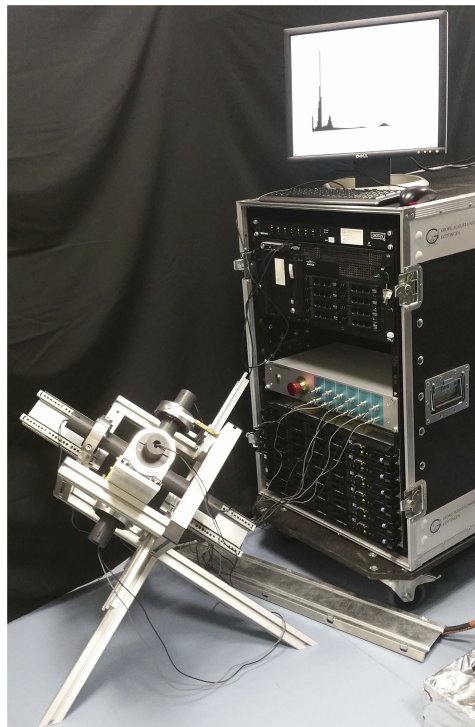


FIG. 7. Digital, user-friendly and compact TDPAC setup in operation at the ISOLDE solid state physics laboratory. Photo taken in 2017.

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