Joint Institute for Nuclear Research

Laboratory of Nuclear Problem

Final Report on the INTEREST Program

In-situ setup of the positron lifetime principle in a coincidence scheme

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Participation period:

November 05 – December 14

Winter Session 2024

Abstract

Positron annihilation spectroscopy (PAS) is a powerful method for investigating the defect structure of materials by analysing the positron lifetime in specific environments. The positron lifetime is influenced by the electron density, where larger defects correspond to lower electron densities and extended positron lifetimes. This study employs a coincidence scheme to measure positron lifetimes, utilizing two BaF2 detectors for high-resolution gamma detection. The first detector captures the start signal from a 1.27 MeV gamma photon emitted during positron production (using the radioactive isotope ^{22}Na), while the second detector records the stop signal from one of the 511 keV gamma photons produced during positron annihilation. The difference between the start and stop times determines the positron lifetime, excluding thermalization time. In the case of dynamic, in-situ defect evolution studies, achieving high time resolution is critical. The need to capture sufficient events within minutes necessitates the use of a beta+ active isotope with a short half-life, generated during proton irradiation of a specific material. To achieve this, a 3.5 MeV proton beam irradiates a carefully selected target material, converting it into a positron-emitting isotope. Careful consideration is given to the material thickness to ensure proton penetration into the sample, thereby inducing defects while simultaneously generating positrons for lifetime measurements. The setup allows real-time monitoring of defect formation and evolution during irradiation. Data acquisition involves photomultipliers to amplify detector signals, an analog-to-digital converter (ADC) for signal processing, and specialized software for spectrum analysis. The resulting annihilation spectra are analyzed to extract positron lifetimes and provide insights into the defect structures and their evolution. This methodology bridges the gap between static defect analysis and dynamic, in-situ measurements, enabling a comprehensive understanding of defect behaviour in materials under irradiation conditions.

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1. Introduction

Defect analysis in materials plays a crucial role in understanding structural integrity, defect distribution, and material performance. One effective technique for rapid defect profiling involves the use of positron annihilation spectroscopy (PAS), where positrons emitted from a beta-plus (β^+) emitting isotope interact with the sample to provide insight into defect characteristics. The aim is to prepare an in situ screening assay for the diagnosis of defect evolution during irradiation. To enable this process, specific nuclear reactions are induced using proton irradiation on a thin layer of a chosen material, which generates the desired positron-emitting isotope and is deposited on the surface of the sample itself. This section describes the conditions required for this method, including proton energy, material properties, and isotope characteristics, and discusses their significance in defect analysis.

The fundamental requirement for generating positron-emitting isotopes is the interaction between 3.5 MeV protons and a thin layer of target material applied to the sample surface. Protons with this energy can penetrate the material and induce specific nuclear reactions that lead to the formation of β^+ emitting isotopes. These positrons are subsequently used to study structural defects in the sample via their annihilation process. The current EG-5 facility (to which we are aiming to apply an in-situ diagnostic instrument) is currently capable of producing protons with a maximum energy of 3.5 MeV, and for this reason we are limiting ourselves to this range.

To facilitate positron emission, a thin layer of a specific material must be applied to the sample surface. This layer acts as a target for proton irradiation, producing β^+ emitting isotopes when bombarded with 3.5 MeV protons. The thickness of the applied material is a critical parameter in this process, as it must be thin enough—typically on the order of a few micrometres, but also in the required amount to allow sufficient production of the $\beta+$ isotope, while allowing most of the protons to pass into the sample under study, where they generate the defects of interest. Therefore, the applied layer must strike a balance—thin enough to facilitate positron escape yet sufficiently thick to produce an adequate quantity of β^+ emitting isotopes.

The nuclear reactions induced by 3.5 MeV protons generate β^+ emitting isotopes with a specific half-life, the proton decays into a neutron releasing a positron e^+ (the antimatter equivalent of an electron) and a neutrino ν,

$$p \rightarrow n + e^+ + \nu$$
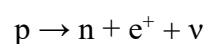
that can only occur within the nucleus with the energy available from the decay transition. Selecting isotopes with a short half-life is crucial for effective and rapid defect profiling. A small half-life ensures large statistics in a short time, enabling real-time defect analysis and quick scanning of the sample. Therefore, selecting short-lived isotopes is essential to ensure rapid, precise, and safe defect profiling in positron annihilation spectroscopy.

2. Experimental Setup and Method

The experimental setup for in situ defect analysis under radiation exposure utilizes positron lifetime spectroscopy (PLS) combined with a coincidence scheme to detection, monitoring and characterize the structural defects induced by radiation in materials. This method allows for precise measurement of defect formation in real time, providing valuable data on the material microstructure and the evolution of structural changes due to radiation exposure.

The proton beam used in this experimental setup is sourced from the EG-5 accelerator, which is capable of accelerating protons to energies of up to 3.5 MeV. Proton irradiation serves two key purposes in the system. First, it induces defects in the sample material, specifically point defects such as vacancies and dislocations within the crystal lattice. This defect formation forms the basis for the subsequent positron lifetime analysis. Second, the proton beam interacts with a thin film of a specific material applied to the sample surface. This interaction results in nuclear reactions, such as the following reactions:

$$^{13}C + p \rightarrow {}^{13}N + n \qquad \text{Half-life } \sim 10 \text{ minutes}$$

$$^{64}Ni + p \rightarrow {}^{64}Cu + n \qquad \text{Half-life } \sim 12.7 \text{ hours}$$

These reactions produce β^+ isotopes that decay by emitting positrons. The short half-lives of the isotopes used—such as nitrogen-13 with a half-life of approximately 10 minutes and copper-64 with a half-life of around 12.7 hours—ensure that data collection is rapid, which is essential for dynamic in situ measurements.

The positron lifetime measurement itself is based on a coincidence detection scheme, which uses two detectors to capture the gamma rays emitted during the annihilation process. The first detector captures the "start" signal, a gamma ray emitted simultaneously with the positron during β^+ decay. The second detector captures the "stop" signal, which is a gamma

ray produced when the positron annihilates with an electron in the sample. The time difference between the detection of these two gamma rays corresponds to the positron lifetime, which is a critical parameter for defect analysis. This setup requires precise synchronization of the detectors and the detection of the gamma rays to ensure accurate timing measurements. To achieve high precision in the timing measurements, the detectors are typically paired with BaF2 crystals, known for their fast recovery time and excellent time resolution. The gamma rays interacting with these crystals produce light pulses, which are then amplified by photomultiplier tubes (PMTs). The PMTs convert the light signals into electrical signals, which are further processed by an analog-to-digital converter (ADC). The digitized signals are analyzed by specialized software, which generates a positron lifetime spectrum. This spectrum provides detailed information about the material's defect structures, with longer positron lifetimes indicating regions of lower electron density, such as vacancies or voids. The entire system thus facilitates the non-destructive analysis of radiation-induced defects in materials in real time, making it an invaluable tool for material characterization.
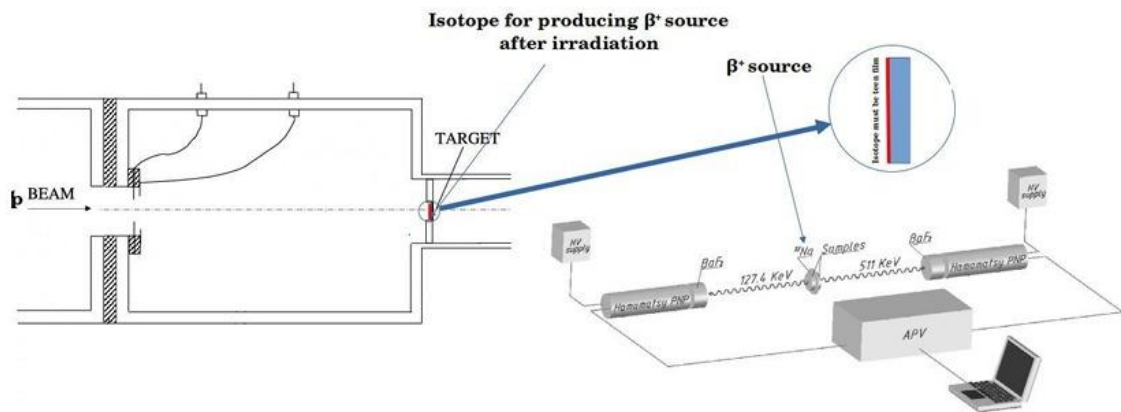

Figure 1: Beta beam irradiation setup for β⁺ source production and detection

3. Analysing the defects using Positron Annihilation

Once the β⁺emitting isotope is generated and positrons are emitted, the analysis of positron annihilation provides critical insights into the defect profile of the sample. Positrons interact with electrons in the material, leading to annihilation and the emission of gamma rays, typically two photons of 511 keV energy. By analyzing key parameters such as positron lifetime (the time from emission to annihilation), the annihilation energy spectrum, and the angular correlation of annihilation photons, valuable information can be extracted. This includes identifying the defect type, such as vacancies, voids, and dislocations in the crystal

lattice; determining defect size and density, where positron lifetimes are sensitive to defect size since larger defects trap positrons for longer durations; and understanding material changes, particularly structural modifications caused by irradiation or other processes. Positron annihilation spectroscopy serves as a non-destructive and highly sensitive technique for defect analysis, making it indispensable for both research and industrial applications where assessing material integrity and behaviour is essential.

When a positron interacts with the material being studied, it loses energy through inelastic scattering, ionization, and electron excitation. The deceleration process occurs over a few picoseconds, after which the thermalized positron diffuses in a delocalized state for several hundred picoseconds before annihilating with an electron. In regions with vacancy defects, the electron density and electrostatic repulsion of the missing positive ions are lower compared to defect-free areas of the lattice. As a result, there is a higher probability of positron capture at these defect sites, leading to a longer lifetime than in a defect-free lattice. By measuring the positron lifetime, we can gain insights into the concentration and type of vacancy defects, a principle at the heart of positron annihilation lifetime spectroscopy (PALS).

PAS techniques typically include three main methods: positron lifetime spectroscopy, Doppler broadening analysis, and angular correlation measurements. While positron lifetime measurements reveal information about the electron concentration at the annihilation site, Doppler broadening and angular correlation provide insights into the electron momentum distribution. The combination of these techniques allows for a comprehensive understanding of the electronic configuration of the material under study, as reflected in the positron annihilation parameters.

PAS methods are often used in conjunction with other material analysis techniques such as Raman spectroscopy, X-ray diffraction (XRD), and neutron diffraction (ND).

4. Understanding the effect of material defects on positron lifetime

In a defect-free crystal, the electron density is uniform and high, enabling the positron to encounter electrons quickly, resulting in a short lifetime. For most defect-free crystals, the positron lifetime typically ranges between 100 to 150 picoseconds, serving as a reference for identifying deviations caused by structural irregularities. However, when defects such as vacancies, voids, dislocations, or grain boundaries are present, the local electron density decreases significantly. These defects create regions where the positron becomes "trapped," encountering fewer electrons and consequently taking a longer time to annihilate. This

prolonged positron lifetime provides valuable information about the defect structure, including defect size—larger defects provide more space and fewer electrons, increasing the lifetime; defect type—vacancies, voids, and other defects exhibit characteristic lifetime signatures; and defect concentration—the degree of positron trapping correlates with defect density. In summary, the presence of defects increases the positron lifetime, and by analyzing the lifetime spectrum, it is possible to determine the type, size, and distribution of defects. This makes PAS a highly sensitive and non-destructive tool for studying microstructural imperfections in crystalline materials.

5. Two-Component Density Functional Theory (TCDFT) and Electron-Positron Interaction

To model and describe the interaction between positrons and electrons in a material, Two-Component Density Functional Theory (TCDFT) is employed. TCDFT extends traditional density functional theory (DFT) by incorporating both electron and positron densities as separate components, allowing for a more accurate description of their mutual interaction and correlation energy.

The total energy functional in TCDFT is expressed as:

$$E[\rho_e,\rho_p]=T_e[\rho_e]+T_p[\rho_p]+E_{ee}[\rho_e]+E_{ep}[\rho_e,\rho_p]+E_{pp}[\rho_p]+\int V_{ext}(r)\rho(r)dr$$

where:

- ρ_e and ρ_p are the electron and positron densities, respectively,
- $T_e[\rho_e]$ and $T_p[\rho_p]$ are the kinetic energy terms of electrons and positrons,
- $E_{ee}[\rho_e]$ represents the electron-electron Coulomb interaction,
- $E_{pp}[\rho_p]$ accounts for positron-positron interactions (negligible in most cases),
- $E_{ep}[\rho_e,\rho_p]$ is the electron-positron correlation energy, which describes the mutual interaction between the two particles, and
- V_{ext} represents the external potential acting on the system.

The **electron-positron correlation energy** $E_{ep}[\rho_e,\rho_p]$ is critical in TCDFT as it captures the annihilation behaviour of the positron with electrons in the material. This energy term reflects the positron's preference to reside in regions of lower electron density (such as defects), where its interaction with electrons is reduced, leading to an increase in its lifetime.

The annihilation rate, which is proportional to the electron-positron overlap, is often expressed using the enhancement factor $\gamma(r)$, which modifies the electron density to account for positron screening effects. The equation for the annihilation rate is:

$$\lambda = \pi r_0^2 c \int \gamma(r) \rho_e(r) \rho_p(r) dr,$$

where:

- r_0 is the classical electron radius,
- c is the speed of light,
- λ is the annihilation rate

In essence, TCDFT provides a theoretical framework to describe and calculate the interaction between electrons and positrons, enabling the interpretation of experimental positron lifetime data and the identification of defects within materials.

6. Positron Lifetime Detection in Materials Using a Coincidence Scheme

The detection of positron lifetime in materials is facilitated by a coincidence scheme that utilizes two detectors to capture the start and stop signals of the annihilation process. This setup enables the precise measurement of the time interval between positron creation and annihilation, providing critical information about the material's microstructure.

In this coincidence scheme, the start signal is generated when a positron is produced in the material, and a gamma quantum of energy 1.27 MeV (when used 22Na) is emitted almost simultaneously. This gamma photon is detected by the first detector, acting as the "start" signal. The stop signal occurs when the positron annihilates after interacting with the sample, producing two 511 keV gamma photons. One of these photons is detected by the second detector, marking the "stop" signal. The time difference between these two signals corresponds to the positron lifetime, a key parameter for defect analysis.

The detection process relies on **photomultipliers (PMTs)** and **analog-to-digital converters (ADC)**. PMTs amplify the weak light signals generated when gamma photons interact with the detectors. These signals are converted into electrical pulses through a series of dynodes, creating an amplification effect known as the "avalanche" process. The amplified signal is then sent to the ADC, which digitizes the signal and forwards the data to specialized software for further analysis. This process generates a positron lifetime spectrum, which is used to extract valuable information about the material's defects.

BaF2 detectors are integral to the coincidence scheme, thanks to their fast recovery time and excellent time resolution. The collected data from the start-stop time intervals is

processed to create a lifetime spectrum, which is then analyzed using specialized software to fit functions that approximate the observed positron lifetimes. This analysis helps identify defect structures in the material, with longer positron lifetimes indicating regions of lower electron density, such as vacancies, voids, or dislocations.

In dynamic measurements, particularly when monitoring real-time defect evolution, short-lived β^+-emitting isotopes play a critical role. These isotopes, generated during proton irradiation (3.5 MeV), provide the necessary data for in-situ experiments. However, if the target material's applied layer is too thick, the protons may not penetrate deeply enough to interact with the sample, limiting the creation of defects and positron interaction. Thus, careful control over the target layer thickness of isotope is essential to obtain accurate, dynamic defect profiles.

7. Obstacles, Efficiency Optimization, and Close Nuclear Reactions

Developing an *in situ* diagnostic system for analyzing defective structures under radiation exposure presents several challenges. One of the primary issues is the high background contribution from nuclear reactions and gamma radiations, which can introduce significant noise into the measurement process. For instance, when using natural nickel for the

$$64Ni(p,n)64Cu \ (^{64}Ni+p\rightarrow^{64}Cu+n)$$
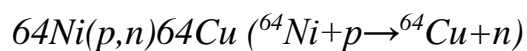
reaction, unwanted secondary nuclear reactions may distort the spectrum, making signal isolation difficult (Laptev et al., 2021). Another major challenge lies in the synchronization of detectors. The proposed system relies on BaF2 detectors for positron lifetime spectrometry, which requires precise start-stop coordination. Without accurate synchronization, valid positron signals can be lost in the background, reducing the system's overall efficiency (Bordulev et al., 2022). Material-specific issues further complicate the process. In the case of the 13C(p,n)13N

$$(^{13}C+p\rightarrow^{13}N+n)$$
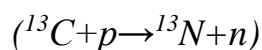
reaction, condensation of CO2 at extremely low temperatures (~215 K) is necessary to form the isotope. While effective, this method becomes impractical when high-temperature conditions are required for testing. On the other hand, using enriched 64Ni is costly, and relying on natural nickel introduces additional reactions that degrade the signal quality (Li et al., 2021).

Finally, implementing advanced double or triple coincidence schemes adds another layer of complexity. These schemes, while effective at reducing noise, require additional

detectors, robust synchronization, and advanced data processing, all of which add financial and technical burdens to the project (Kudiyarov et al., 2023).

To address these challenges, several strategies are proposed to enhance the efficiency and reliability of the *in situ* diagnostic system. One of the key solutions is the use of **double and triple coincidence schemes**. By incorporating multiple detectors and validating positron events through coincidence triggers, background noise can be significantly reduced. A triple coincidence scheme adds another layer of filtering, ensuring that only valid signals are processed, even in high-background environments (Lotkov et al., 2020).

Expanding the detector array is another crucial step toward improving efficiency. Adding more BaF2 detectors not only increases the system's detection coverage but also enhances resolution and reduces the chance of missing valid events. Proper synchronization of these detectors will ensure that signals are captured accurately and processed without significant delays (Laptev et al., 2023).

The choice of nuclear reactions must also be optimized to balance performance and cost. While the 13C(p,n)13N reaction is ideal for short-term measurements due to its 10-minute half-life, its temperature limitations restrict its use in high-temperature experiments. For longer studies, the 64Ni(p,n)64Cu reaction offers a half-life of 12.7 hours, but it requires careful management to minimize secondary reactions. Alternative solid-state forms of carbon or optimized use of natural nickel may provide a middle ground (Bordulev et al., 2021).

Advanced software development will also play a significant role in improving efficiency. By integrating real-time signal processing and noise reduction algorithms, the system can isolate valid signals more effectively. Machine learning techniques can further assist in distinguishing between background noise and meaningful data, enabling faster and more accurate analysis (Li et al., 2022).

To optimize EG-5 accelerator performance, several key areas require attention. Precise beam focusing and alignment minimize energy loss. Upgrading RF power systems enhances energy coupling. Optimizing the ion source and target improves particle production and interaction. Maintaining a high-quality vacuum reduces scattering. Upgrading power supplies and implementing energy recovery systems minimize energy waste. Regular maintenance and advanced control systems ensure optimal performance and efficiency.

The selection of nuclear reactions directly influences the overall performance and statistical reliability of the system. The 13C(p,n)13N reaction is highly efficient for short-term

diagnostics due to its short half-life and minimal residual activity. However, its reliance on low-temperature CO_2 condensation limits its practical use in high-temperature environments, making it unsuitable for certain materials (Laptev et al., 2023).

In contrast, the 64Ni(p,n)64Cu reaction provides a longer half-life, making it ideal for extended measurements. However, the cost of enriching 64Ni is significant, and using natural nickel introduces multiple close nuclear reactions. These secondary reactions produce overlapping gamma signals, which can complicate the spectrum and hinder accurate analysis. In such cases, implementing double or triple coincidence schemes becomes essential to filter out unwanted signals and improve the quality of the measurements (Kudiyarov et al., 2023).

To minimize the impact of secondary reactions, it is important to carefully manage the experimental setup. Optimization of isotope usage, coupled with precise detector synchronization and background subtraction methods, will ensure that the system operates with maximum efficiency and reliability (Lotkov et al., 2020).

To overcome the identified obstacles and enhance the performance of the system, the following strategies are recommended: First, implementing double and triple coincidence schemes will help reduce background noise and improve the clarity of the signals. Second, expanding the detector array is essential to increase coverage and ensure better synchronization of the start-stop signals. Third, optimizing nuclear reaction management by balancing cost, temperature conditions, and isotope performance will improve the overall efficiency of the system. Additionally, developing advanced software for real-time signal processing, noise reduction, and data analysis will enhance the accuracy of measurements. Finally, fine-tuning thermal and hydraulic systems is crucial to ensure consistent operation under varying experimental conditions. By addressing these challenges, the in-situ diagnostic system will achieve higher efficiency, minimized background interference, and reliable defect analysis in materials exposed to radiation.

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