REALIZATION OF POSITRON ANNIHILATION SPECTROSCOPY AT LEPTA FACILITY

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Abstract

The Positron Annihilation Spectroscopy (PAS) unit was created as a part of LEPTA project at JINR in Dubna. Currently works performed in PAS laboratory focus on studies of defects in solid state, especially on studying radiation damages in novel materials and semiconductors as a part of the international project "Novel Semiconductor for Fundamental and Applied Research". This report aims to present a current status of realization and progress in PAS methods at LEPTA facility at JINR.

INTRODUCTION

Positrons are used in materials science to study open volume defects such as vacancies, vacancy clusters and dislocations. Several positron annihilation spectroscopy (PAS) techniques exist. These methods are based on detection of the 511 keV gamma quantum. The first method is the analysis of the Doppler broadening of annihilation line and provides information about defect concentration. Both annihilation quanta can be observed. Coincidence observation of two quanta gives additional information about the environment around defect. The second method is based on lifetime concept, which allows us to distinguish type of defects. Nowadays, positron beams are of great interest for materials science. Using a low energy, monoenergetic beam it is possible to control the positron penetration depth, see Fig. 1, from the sample surface to a depth of several microns. Thus, the beam can be used to characterize thin films, analysis of surface modification, studying influence of ions on matter etc.
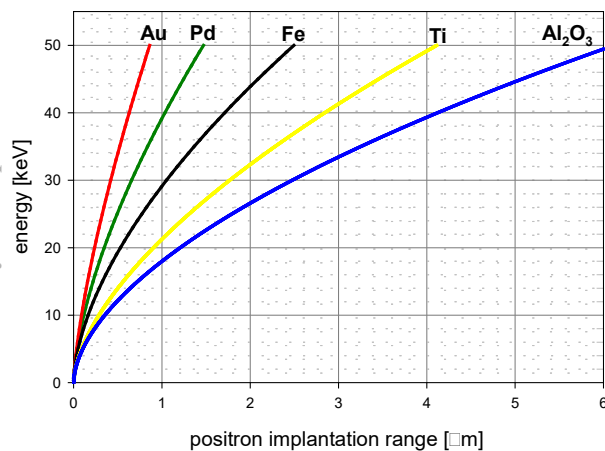
THE DOPPLER BROADENING OF ANNIHILATION GAMMA LINE

The basics of method rely on registration energies of annihilation quanta. Due to Doppler's phenomenon quantum energies are changed according to the formula:

$$E_\gamma \cong mc^2 + E_B \pm \frac{p_\| c}{2}, \qquad (1)$$

where E_b is the energy of positron-electron pair coupling and $p_\|$ is a component longitudinal to annihilation quanta direction of the pair's momentum. It is worth emphasising that positron's momentum is negligibly small in relation to electron's momentum, therefore it is usually omitted in deliberations.

The heart of Doppler broadening spectroscopy is a detector which requires a high energetic resolution. Typical available germanium detectors allow us to take measurements with resolution equal to 1-2 keV around 511 keV energy. The scheme of Doppler broadening spectrometer unit working in PAS laboratory at JINR is shown in Fig. 2. The important parameters of high purity germanium Detector (HPGe) made by Baltic Scientific Instruments working at LEPTA facility is gathered in Table 1. Next year also the second detector should appear in PAS laboratory, which will allow to perform coincidence measurements.
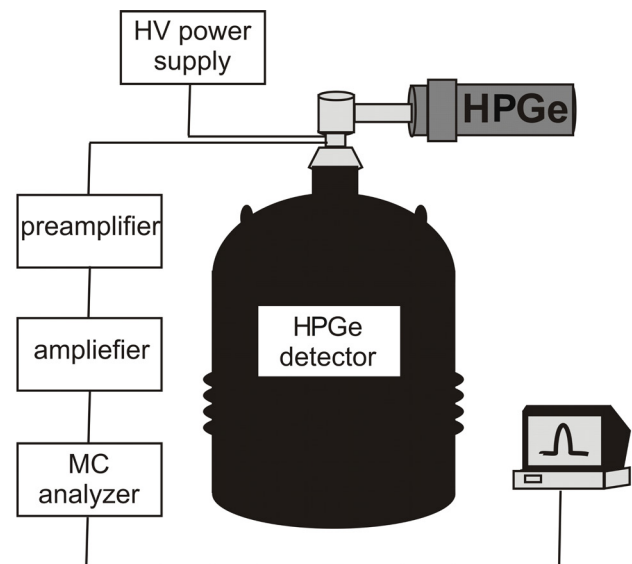

Figure 1: Dependency of positron energy on mean positron implantation depth [1].

Figure 2: The scheme of Doppler broadening of annihilation gamma line 511 keV spectrometer.

Table 1: Main Parameters of HPGe Detector

Feature	Value
relative registration efficiency at 1.33 MeV-photon	30%
energy resolution FWHM for energy 511 keV	<1250 eV
peak to Compton ratio	58:1
energy range of detector operation intensity	40 ÷ 10 000 keV

In practice, information about concentration of defects is received from analysis of the shape of annihilation line, by calculating two important shape parameters, called S and W. Exemplary annihilation line as well as the rule for calculating both parameters are presented in Fig. 3.
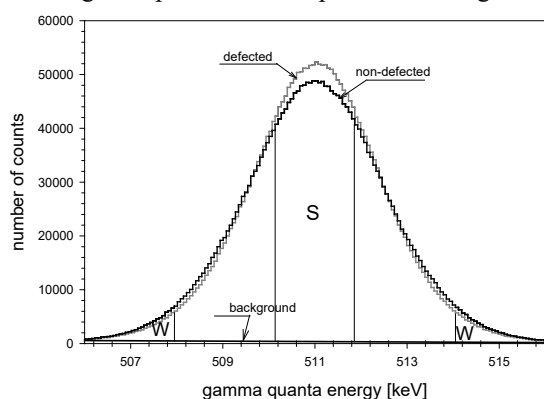

Figure 3: The annihilation lines with marked areas defining parameters S and W measured in stainless steel. The grey line comes from the defected (by sliding) sample, while the black line represents the non-defected sample.

S parameter defines proportion of annihilation of positrons with low-momentum electrons. It is closely related to concentration of defects in a material. It is defined as ratio of surface area under the central part of the 511 keV line to total surface area under this line. Areas are usually selected so that their ratio was approximately 0,5. Value of S parameter for trapped in defect positron is smaller than for a non-defected sample due to lower probability of annihilation with high-momentum core electrons during occupying vacancy position. In other words, less defected sample gives smaller broadening of the 511 keV line and bigger S parameter value. The second parameter, the so called W parameter is defined as ratio of surface area under the wing part of 511 keV line to total surface area. It is related to annihilation of positrons with high-momentum electrons and it provides information about chemical environment of the defect. Both S and W parameters are calculated after background reduction. Calculations are made by special computer software.

This method could be used to obtain information about defect concentration, its profile and defected zones range.

In case of surface damaged in dozens of micrometres, i.e during cutting process standard measurements including isotope ^{22}Na could be used. Such results for cutting with various techniques, using milling, laser and water stream, AISI 304 austenitic stainless steel is presented in Fig. 4 [2]. It this case decreasing dependency of S parameter could be observed, which means that the defect profile in sample also decreases. For milling and water stream cutting defect profile is almost linear. However, the determined damaged zone range, the region where S parameter is bigger than value of S parameter obtained for well annealed sample, strongly depends on used cutting techniques and for water cutting is equal about 50 μm and for milling 150 μm. In case of laser cutting bulk value was not reached, despite 350 μm of the sample was etched. It turns out that positron spectroscopy is very useful in such studies.
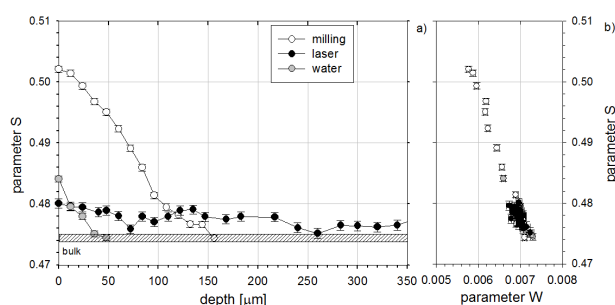

Figure 4: Dependency of S parameter on the depth below the surface for the 304 AISI stainless steel samples cut with various techniques: milling, laser and water stream. The hatched region represents the bulk value of S parameter obtained for the well-annealed RF sample. On the right, S parameter versus W parameter values from the Doppler broadening spectra are depicted. [2]

To study thin films, radiation damages a standard technique employing fast positrons implanted directly from isotope ^{22}Na source could not be used because most of positrons will penetrate the deeper, not interesting region of sample. For this case a slow positron beam with several keV energies should be applied. Slow positrons can be obtained through moderation of fast positron emitted from ^{22}Na. At JINR the moderation process is done using frozen neon. The exemplary studies of radiation damages for the iron samples irradiated with 167 MeV Xe^{26+} heavy ions with different doses from 10^{12} to 10^{14} ions/cm^2 is shown in Fig. 5 [3]. Characteristic decreasing from surface to deeper region dependency of S parameter could be observed. It is linked with diffusion of positrons to surface, which has a higher value of S parameter due to presence of oxide and surface imperfection. In deeper region, i.e. over 400 nm, the saturation of S parameter dependency could be observed. Increment of ions dose implanted to the samples gives higher value of the S parameter saturation value. The full analysis of these results is available in work [3]. It should be highlighted that positron spectroscopy is also very important in such studies.
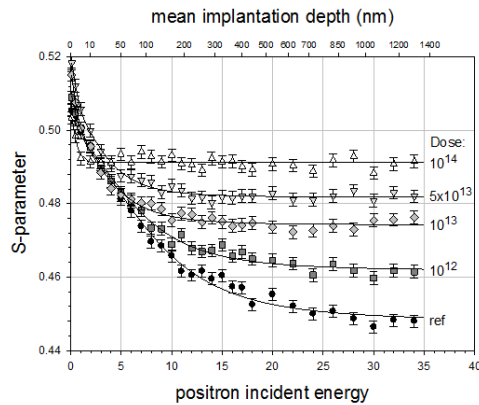
Figure 5: Variable energy positron measurements for the iron samples irradiated with 167 MeV Xe^{26+} heavy ions with different doses from 10^{12} to 10^{14} ions/cm^2 [3].

THE POSITRON LIFETIME SPECTROSCOPY

Positron lifetime spectroscopy measures the elapsed time between the implantation of the positron into the material and the emission of annihilation radiation. It is related with local electron density n_e and could be estimated as:

$$\overline{\tau} = \frac{1}{\pi r_0^2 c n_e}, \qquad (2)$$

where c is light velocity, r_0 electron radius. When positrons are trapped in open-volume defects, such as vacancies and their agglomerates, positrons stay in the locally reduced electron density of the defect. This gives longer positron lifetimes. The calculations of positron lifetime for metals and other materials show a link between number of vacancies in a cluster and measured positron lifetime. When number of vacancies rises, the positron lifetime increases too. This relation one can obtain using ab initio theoretical calculations. The performed calculations using ABINIT program for zirconium are shown in Fig. 6 [4]. Such results allow to assign the type of defect to measured experimental positron lifetime.
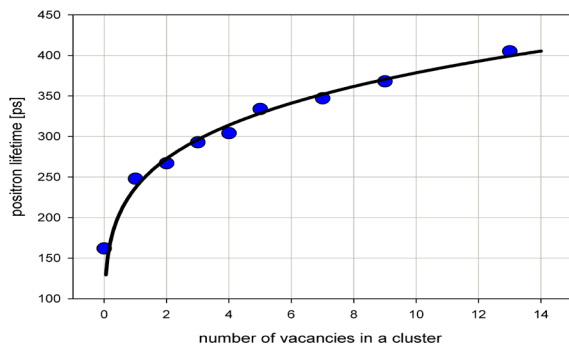

Figure 6: Positron lifetime as a function of the number of vacancies in a cluster Zr calculated using ABINIT code [3].

The positron lifetime spectroscopy measurements will start at JINR until year 2017. The collected equipment includes digital single processor APU-8002-RU and two photomultiplayers PMT H3370-50 with BaF_2 monocrystals. The scheme of positron lifetime measurements system is shown in Fig. 7.
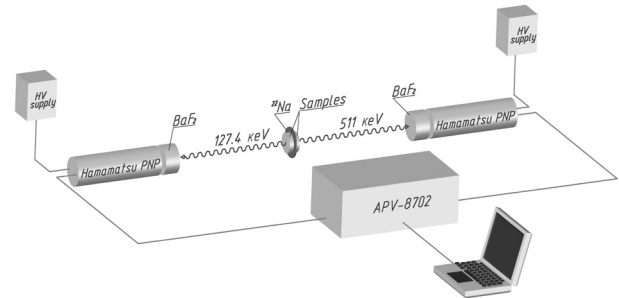

Figure 7: Scheme of positron lifetime measurement system.

SUMMARY

Positron annihilation spectroscopy measurements constitute an effective way to study surface and subsurface defects. It could be used for determination of the concentration and determination of the defect type. Currently installed apparatus allows to perform standard Doppler broadening and Variable Positron Beam measurements. Until next year positron lifetime measurements will start at LEPTA facility. In the nearest future we intend to expand our measuring system on Coincidence Doppler Spectroscopy, which would significantly reduce the background in the spectrum.

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