Introduction to Experimental setup of two-dimensional Positron Annihilation Lifetime and Doppler Broadening Spectroscopy (2D-PALS+DBS)

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1. Introduction

Korea Atomic Energy Research Institute have studied the defect identification in materials with positron annihilation spectroscopy. Currently, Coincidence Doppler Broadening Spectroscopy (CDBS) [1, 2] and Positron Annihilation Lifetime Spectroscopy (PALS) had been performing. CDBS consists of two High Purity Ge (HPGe) detectors that possess a high energy resolution and the coincidence setup reduces background [2]. PALS system has two plastic scintillators which has fast time constant and high time resolution. ^{22}Na positron source decays through the β^+ process, producing a positron and a neutrino, leaving an excited ^{22}Ne nucleus that rapidly decays a 1.27 MeV γ photon. As shown in Fig. 1, positron annihilation spectroscopies are selectively sensitive to vacancy-type defects. The positron lifetime and Doppler broadening can be calculated by knowning the corresponding electronic structure of the solid when the positron is annihilated in a solid. The positron annihilation rate, λ, the inverse of the positron lifetime τ, is known to be proportional to the superposition of the electron and positron desities.

$$\lambda = \frac{1}{\tau} = \pi r_e^2 c \int |\psi_+(\boldsymbol{r})|^2 \, n\_(\boldsymbol{r}) d\boldsymbol{r}, \qquad (1)$$

where r_e is the electron radius, c is the velocity of light, $n_-(\mathbf{r})$ is the electron density, and ψ_+ is the positron wave function. The momentum distribution $\rho(\mathbf{p})$ of the annihilation photon can be described all electron wave functions ψ_i overlapping with the positron. Approximately, it can be written in the form

$$\rho(\boldsymbol{p}) = \pi r_e^2 c \sum_i \left| \int d\boldsymbol{r} e^{-i\boldsymbol{p}\cdot\boldsymbol{r}} \psi_+(\boldsymbol{r}) \psi_i(\boldsymbol{r}) \right|^2, \qquad (2)$$

where the summation goes over occupied electron states [3]. The electron density is locally reduced in vacancy defects and it is reflected in the longer positron lifetime than in the defect-free lattice. Therefore, positron lifetime measurement is to investigate vacancy defects in materials. Positron annihilation in the vacancy defect also results in a change in the momentum distribution, which is investigated by Doppler broadening spectroscopy. The momentum distribution of valence electron annihilation is slightly narrower due to the lower electron density. In addition, positrons localized in vacancies have reduced overlap with the ionic core, which greatly reduces annihilation by core electrons with high momentum. Therefore, we propose an experimental setup that can simultaneously measure the two spectroscopic phenomena in order to examine the correlation between PALS and DBS from vacancy defects in solids.

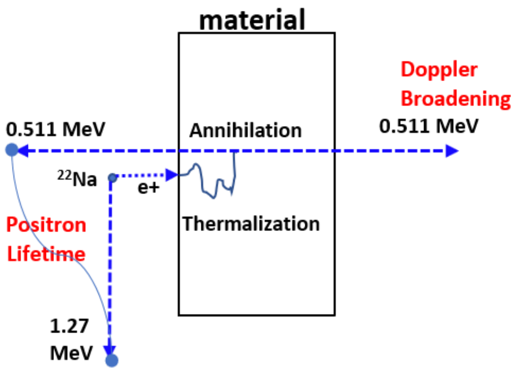

Fig. 1. PALS and DBS phenonon in a solid by using a radioactive ^{22}Na positron source.

2. Experimental Set-up

2.1 Positron Annihillation Lifetime Spectroscopy

PALS measurement is performed by detecting two photons that use as “start” and “stop” signals with two scintillating detectors coupled with a photomultiplier tube. As shown in Fig. 2, two BaF2 scintillators are installed, providing a significantly higher detection efficiency and better resolution.

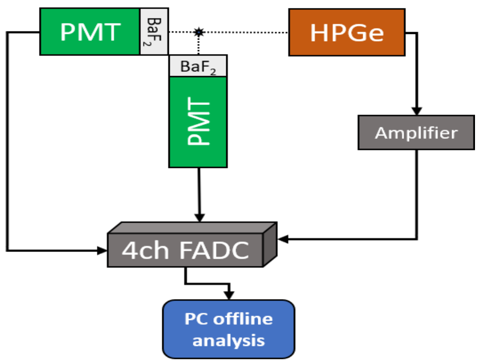

Fig. 2. Simple scheme of PALS + DBS experimental setup and the data acqusition system.

The probability of a positron to be alive at time t is obtained as

$$n(t) = n_B(t) + n_V(t) + n_{st}(t) = \sum_i I_i e^{\lambda_i t} \qquad (3)$$

where the probability of a positron being in the free state n_B and in a trpped state at vacancy defect n_V and at shallow trap n_{st}. With Eq. (3), the experimental positron lifetime spectrum can be expressed as the convolution of the preferably Gaussina resolution function *R(t)* with the sum of exponential functions:

$$N(t) = \int_{-\infty}^{\infty} ds R(s) \left(-\frac{dn}{dt}(t-s) \right) \qquad (4)$$

In practice, the experimental data can be fitted by Eq. (4). The fitting parameters include the intensities I_i and annihilation rates λ_i of the lifetime components.

2.2 Doppler Broadening Spectroscopy

One annihillation photon is detected by HPGe detector with high energy resolution. The motion of electron-positron annihillation causes a Doppler shift in the annihillation radiation $\Delta E = cp_L/2$, where p_L is the longitudinal momentum component of the pair in the direction of the 511 keV photon emission. The typical resolution of the HPGe detector is about 1-1.5 keV at 511 keV. This is a significant number compared to the 2-3 keV total width of the annihillation peak, indicating that the experimental line shape is strongly dependent on the detector resolution. Also, the peak-to-background ratio is very important parameter for Doppler broadening mesurement. Therefore, two collinear detectors and coincidence conditions can significantly both parameters. Fig. 2 shows DBS experimental setup (one HPGe detector and another BaF_2 scintillating detector to gate the HPGe detector). Even if the peak-to-background ratio can be much improved when two HPGe detectors are used, BaF2 scintillating detector is selected because it has much higher time resolution for PALS measurement. the shape parameters of the annihillation peak are used to characterize the 511 keV line. The low-momentum shape parameter *S* is defined as the ratio of counts in the central region of the annihillation line to the total number of the counts in the line. The high momentum parameter *W* is the fraction of the counts in the wing regins of the line.

2.3 Data Acquistion and Offline Analysis

The data acquisition is based on a flash Analogue-to-Digital converter (FADC) with a sampling rate of 500 MHz system produced by Notice Korea and the offline data analysis on the ROOT framework [5]. FADC500 four-channel were utilised to process the amplifier outputs of the HPGe-detector, the tow anode outputs of the scintillation detectors. In Fig. 2 a schematic of the detector signal acquisition is given. In the offline analysis the events are checked to see if event contains the starting pulse and the stopping pulse. The positron lifetime is calculated as the timing difference between both pulses using a digital constant fraction discrimination (CFD) [6].

3. Summary and Plan

In previous studies [7], it has been well documented that the *S* and *W* parameters from DBS are time-averaged (τ_{av}) quantities and thus behave similarly to the average positron lifetime. As a consequence, the vacancy concentration can also be determined from the S and W parameters. Furthermore, through the combination of positron lifetime and Doppler broadening results, various correlations between τ_{av}, *S*, and *W* can be studied

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