Performance Optimization Through Offline Analysis of Age-Momentum Correlation Spectroscopy Using a Fast Digitizer

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1. Introduction

In materials science, understanding the microstructure of materials, including free volumes and vacancy structures, is crucial for analyzing various material properties. In materials containing free volume, positrons can combine with electrons to form positronium (Ps), which exists in two states: para-positronium (para-Ps) and ortho-positronium (ortho-Ps), generated in a specific ratio.

Ps formed in materials provides valuable information about both the free volume characteristics and the chemical environment surrounding it. Specifically, the positron lifetime reflects information about the size and concentration of free volumes, while the momentum of electrons participating in Ps formation creates Doppler broadening phenomena that contain chemical information about the material environment.

However, conventional measurement techniques such as positron annihilation lifetime spectroscopy (PALS) or Doppler broadening spectroscopy (DBS) alone are insufficient to simultaneously characterize both aspects. Each method only provides partial information about either the free volume or the surrounding chemical environment, but not both simultaneously in a correlated manner.

The Age-Momentum Correlation (AMOC) spectroscopy technique addresses this limitation by simultaneously measuring positron lifetime and Doppler broadening information. This correlation allows for a more precise analysis of electron density and nanoscale structural changes within materials. AMOC is particularly valuable for studying the chemical environment of lattice vacancies and nanostructures, which play a crucial role in atomic diffusion, plastic deformation, and structural phase transitions [1-3].

For AMOC spectroscopy to be most effective, excellent time resolution is essential. Improved time resolution enables more accurate correlation between signals, allowing for a more precise understanding of subtle structural changes in materials. Therefore, optimizing the measurement conditions to achieve the best possible time resolution is a key factor in enhancing the applicability and accuracy of AMOC spectroscopy.

In this study, the digital Constant Fraction Discriminator (dCFD) method [4] is applied to investigate variations in time resolution according to different constant fractions and delay times. The focus is placed on deriving optimal measurement conditions to improve the accuracy of AMOC measurements, thereby contributing to the application of AMOC systems in various materials science fields.

2. Methods and Results

The data acquisition (DAQ) system used in this study was composed of a flash analog-to-digital converter (FADC500, Notice Korea) and a server. The sampling frequency of FADC was 500 MHz. The maximum allowable voltage of the FADC was 2500 mV (12 bit), and the width of a single bin was 0.61 ($2{,}500/2^{12}$) mV/bit. Pulse waveform data were sampled and collected at 2-ns intervals. The digitized data were stored in the DAQ server and utilized for offline analysis, which was performed using the C^{++} programming language. The AMOC measurement was conducted by acquiring three coincidence data of the prompt gamma-ray from beta decay of the positron source and positron annihilation gamma-rays.

For the AMOC measurement, a ^{22}Na positron source with an activity of approximately 100 μCi was used. The measurement was carried out with the source sandwiched between two 7.6 μm Kapton foils. One of the gamma rays emitted from ^{22}Na was detected using a cylindrical $CeBr_3$ scintillator with 25 mm × 25 mm dimensions. This gamma ray was either the positron-annihilation ray (511 keV) or the prompt gamma-ray (1,274 keV). Each $CeBr_3$ scintillator was optically coupled to a photomultiplier tube (R329-02, Hamamatsu Photonics, K. K.) with optical grease. A pair of scintillation detectors were equipped with the PMT bases (E5859-01, Hamamatsu Photonics, K. K.). A high-purity Ge detector (HPGe) with an energy resolution of 1.94 keV@1,332 keV Co-60 was aligned collinearly with the stop detector to detect annihilation rays simultaneously. A ^{60}Co source was used to measure the instrument time resolution. Figure 1 shows the configuration of the AMOC system.

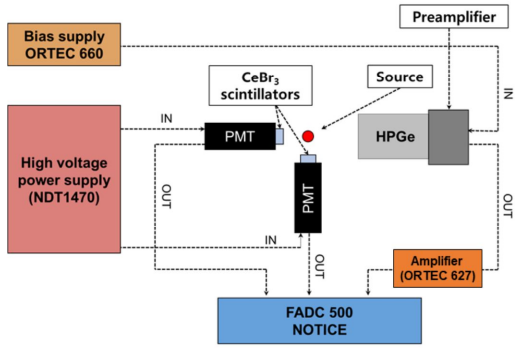

Fig. 1 . The configuration of the AMOC system was constructed using a high-speed digitizer. PMT: photomultiplier tube, HPGe: high-purity Ge.

The anode output of the $CeBr_3$ detector was converted from a negative pulse to a positive pulse for analysis. The digitized data were processed using the dCFD method to perform timing analysis. The unipolar output of the $CeBr_3$ detector was transformed into a bipolar signal according to the constant fraction and delay time set in the dCFD method. The processed bipolar signal was analyzed using linear fitting and cubic spline fitting techniques to determine the point at which the sampled data reached zero. Figure 2 shows the signal of the bipolar pulse processed by the dCFD method.

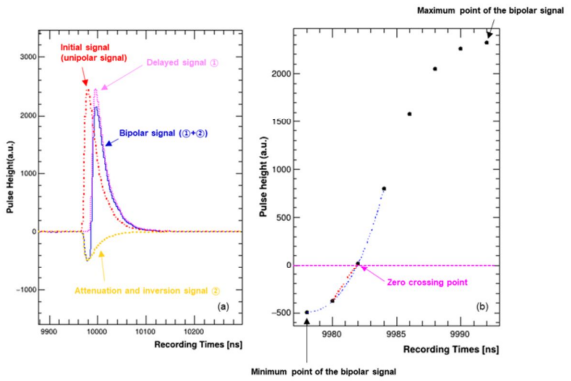

Fig. 2. The dCFD (Digital Constant Fraction discrimination) method processes the bipolar signal and determines the timing. (a) The transformation of the initial unipolar signal into a bipolar signal through delay and attenuation is illustrated. (b) The method of determining timing using the minimum point, maximum point, and zero crossing point of the bipolar signal is presented.

3. Result and Discussion

The timing measurement performance was evaluated based on different constant fraction and delay time values. The linear fitting and cubic spline fitting were applied and analyzed for comparison to achieve this (Fig. 3).

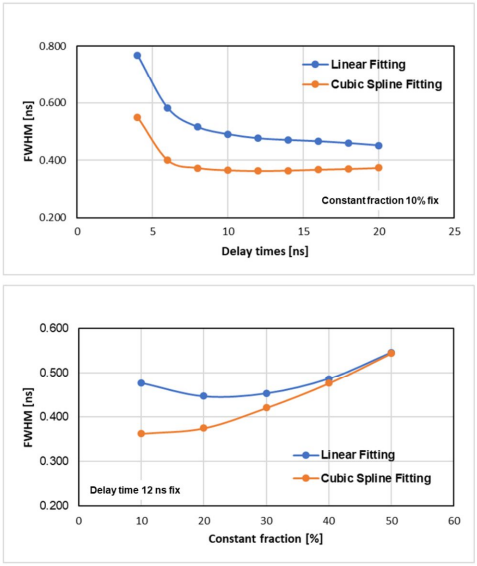

Fig. 3. Comparison of Time Resolution Based on Delay Time and Constant Fraction Using Linear and Cubic Spline Fitting

The effect of varying the delay time was examined with the constant fraction fixed at 10%. The results showed that cubic spline fitting provided an FWHM (full width at half maximum) value approximately 23.7% lower than linear fitting, confirming its effectiveness in improving time resolution. The minimum FWHM value of 0.363 ns was recorded at a delay time of 12 ns. When the delay time exceeded 12 ns, the FWHM value no longer improved and showed a stabilization trend.

The time resolution was analyzed by varying the constant fraction with the delay time fixed at 12 ns. The lowest FWHM value of 0.363 ns was observed when the constant fraction was set at 10%. As the constant fraction increased, the FWHM value gradually increased, and a sharp rise was observed beyond 30%. For example, when the constant fraction was 30%, the FWHM value was 0.420 ns, but it increased to 0.476 ns at 40% and 0.544 ns at 50%. This result suggested that a large constant fraction could degrade time resolution due to signal attenuation.

These findings confirmed that the optimal conditions for achieving the best time resolution were a delay time of 12 ns and a constant fraction of 10%. These optimal conditions were determined for the dCFD method in the AMOC measurement system, contributing to more accurate AMOC measurements.

4. Conclusions

In this study, the optimal conditions for the dCFD method in the AMOC measurement system were determined, confirming that time resolution could be improved. The analysis results showed that setting the delay time to 12 ns and the constant fraction to 10% provided the lowest FWHM value of 0.363 ns, demonstrating optimal performance.

This optimization enables AMOC spectroscopy to simultaneously analyze both the free volume characteristics and the chemical environment in materials with greater precision, overcoming the limitations of conventional PALS or DBS measurements used individually.

Through this improved time resolution, more precise AMOC analysis becomes possible, and it is expected to be utilized in various experimental environments where accurate characterization of materials' microstructure is crucial for understanding atomic diffusion, plastic deformation, and structural phase transitions.

ACKNOWLEDGEMENT

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