Effect of Constraining the Source Lifetime Parameter during Least-square-fit Analysis on Positron Lifetime Measurements

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We examined the effect of constraining the positron source lifetime parameter during a least-square fitting (LSF) of positron annihilation lifetime (PAL) data on the resultant lifetimes, as determined from the fit result. The effect was assessed using calculated PAL spectra and also experimentally-obtained spectra for single-crystal silicon with different measurement statistics, in order to discuss the uncertainty of the resultant lifetime for a sample having a relatively short lifetime of around 220 ps. The obtained results suggest that constraining the positron source lifetime during the LSF analysis results in a relatively minor uncertainty of the sample lifetime in comparison with that due to the measurement statistics.

1. Introduction

The positron annihilation lifetime (PAL) technique is a powerful tool for exploring open spaces at the atomic level. In PAL measurements, a frequency distribution of the measured time interval between a start signal (obtained via a $\gamma$ ray emitted synchronously with the positron) and the corresponding stop signal due to annihilation of the positron is acquired using a lifetime measurement system with $\gamma$-ray detectors (solid scintillators coupled to photomultiplier tubes). The accumulated PAL data is generally fitted using a model function including multiple decaying exponentials, with each exponential component giving a mean annihilation rate, which is the inverse of the lifetime for that component. For bulk PAL measurements radioactive $^{22}$NaCl sealed with Kapton films is generally used as the positron source [1, 2]. Hence, the recorded PAL data consists of at least two lifetime components due to the annihilation of positrons in the sample and the Kapton film.

When data analysis is performed through least-square curve fitting, the source lifetime of Kapton may vary by up to 7 ps due to the irradiation effect of the emitted positrons. Therefore the constrained value for the source component may deviate from its actual value, which may give rise to an ill-conditioned fit during the analysis. In this work, we examined the effect of constraining the Kapton lifetime parameter for the least-square fitting (LSF) to the PAL data on the resultant lifetimes obtained from the fit result. We first estimated a change of the fit results, obtained with and without parameter constraining during LSF, as a function of the input value of the source lifetime using mathematically-calculated PAL data. The effect was then assessed using experimentally-obtained PAL data for single-crystal silicon with different measurement statistics, in order to discuss the measurement uncertainty of the resultant lifetime for a sample having a relatively short lifetime of around 220 ps.
2. Calculations and Experiments

2.1 Mathematical model and conditions for calculated PAL data

PAL data \( y(T) \) is expressed by multiplying the PAL spectrum \( f(T) \) by the channel time interval \( \Delta T > 0 \). \( f(T) \) is generally described by the following formula,

\[
f(T) = A \int_{-\infty}^{\infty} R(t) \Theta(T - t) C(T - t) dt + B,
\]

where \( \Theta(t) \) represents the Heaviside-step function, \( A \) is a standardization coefficient, and \( B \) is a background, respectively. \( R(t) \) and \( C(t) \) are formulated as follows,

\[
R(t) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{t^2}{2\sigma^2}\right), \text{ and}
\]

\[
C(t) = \sum_{i=1}^{N} \frac{I_i}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right).
\]

Here, \( \tau_i, I_i, \sigma, \) and \( N \) denote the positron lifetime of \( i \)-th component, its relative intensity, the system time resolution in standard deviation, and the number of the lifetime components, respectively.

To estimate the constraint effect on the LSF analysis, we mathematically calculated a set of PAL data with \( \tau_{1,\text{in}} = 220 \) ps \( (I_{1,\text{in}} = 80 \%) \) for the sample component and different \( \tau_{2,\text{in}} \) from 360 ps to 400 ps with an interval of 10 ps for the source component. The other parameters, that is, FWHM \( = \sqrt{2 \ln 2} \sigma \) of \( R(t) \), the total accumulated counts, and the background counts per channel \( B\Delta T \), were set to 150 ps, 2 million counts, and 20 counts \cdot CH^{-1}, respectively. These calculated data are provided in Section 3.1 for discussion.

2.2 PAL measurements

Experimental PAL data for single-crystal silicon were measured using a PAL measurement system, consisting of BaF\textsubscript{2} scintillators (a trapezoid pillar shape an upper diameter of 30 mm, a lower diameter of 40 mm and a height of 20 mm, Oyo Koken Kogyo, Japan), photomultiplier tubes (H3378-51, Hamamatsu Photonics) and a digital storage oscilloscope (WaveRunner 610Zi, Teledyne LeCroy Japan). 1-mm-thick silicon samples in a plane size of 15 mm \( \times \) 15 mm were purchased from Shin-Etsu Chemical, Japan. A radioactive \( ^{22}\text{Na} \) positron source sealed with 7.5-\textmu m-thick Kapton films was used for the present work.

2.3 Data analysis

Based on the above mentioned mathematical model, both the calculated and experimentally-obtained PAL data were analyzed by a least-square fitting with the PATFIT program [4] (LSF analysis), to deduce each component of the average lifetime \( \tau \) of the positrons and the corresponding relative intensity \( I \) (the count ratio of each component).

3. Results and discussion

3.1 Effect of constraining the source lifetime during the LSF analysis

A set of the calculated PAL data was analyzed through LSF with and without constraining the 2nd lifetime to 380 ps as the Kapton film. Figure 1 shows the variation of the resultant 1st lifetime \( \tau_{1,\text{out}} \) as a function of \( \tau_{2,\text{in}} \) provided to the calculated data. While for \( \tau_{1,\text{out}} \) without parameter constraining \( (\tau_{1,\text{wo}}) \) the values well agree with the provided lifetime i.e. 220 ps, those analyzed with parameter...
Input lifetime for Kapton, $\tau_{2,\text{in}}$ [ps]

Resultant lifetime for sample, $\tau_{1,\text{out}}$ [ps]

Fig. 1. Variation of $\tau_{1,\text{out}}$ analyzed with and without constraining the 2nd lifetime as a function of $\tau_{2,\text{in}}$. The broken line represents the result of a linear fit for the data with parameter constraining. The error bars denote the twice width of the respective standard errors from the LSF analysis.

constraining ($\tau_{1,\text{out}}^w$) linearly decrease with increasing $\tau_{2,\text{in}}$. A linear fit to the data of $\tau_{1,\text{out}}^w$ versus $\tau_{2,\text{in}}$ provides the following equation,

$$\tau_{1,\text{out}}^w = 220 - 0.223(\tau_{2,\text{in}} - 380) \text{ [ps]}. \quad (4)$$

This equation indicates that a change of $\tau_{2,\text{in}}$ for the PALS data by 5 ps leads to a shift of the resultant $\tau_{1,\text{out}}$ of $\sim 1$ ps (relative value: 0.5 %), substantially smaller than the standard error of 2.5 ps at the present LSF analysis. This suggests that constraining the Kapton source lifetime during the LSF analysis causes only relatively minor uncertainty in the shorter lifetime in comparison with the uncertainty due to the present statistics of 2 M counts.

3.2 PAL measurements for single-crystal silicon

During LSF analysis, uncertainty in the fit results can be reduced by using accurate reference values for the initial fit parameters. This means that constraining the source component to a proper value may be advantageous to attain lower fitting error of the obtained lifetime even for PAL data with lower counting statistics. Figure 2 shows PAL data for single-crystal silicon, experimentally measured with different accumulation statistics, that is, 2 M counts and 20 k counts. As one can see from the data with the higher statistics, two lifetime components are recognizable, reasonably ascribed to positron annihilation in silicon and Kapton film. Lifetime parameters obtained through LSF analysis for both spectra are summarized in Table I (LSF #1 and #2 for 2 M counts, and LSF #3 and #4 for 20 k counts).

For each spectrum the LSF analysis was conducted with and without constraining the 2nd lifetime to 380 ps for the Kapton source. For both LSF #1 and #2 with 2 M counts, $\tau_1$ with and without parameter constraining are obtained with standard uncertainties less than 2 ps (relative value: 1 %). This illustrates that a total accumulation of 2 M counts is enough to estimate the sample lifetime with sufficiently small error in the present condition. On the other hand, the result of LSF #3 obtained without parameter constraining provides much larger uncertainty for $\tau_1$ due to the poor statistics.
Fig. 2. PAL data for the single-crystal silicon, experimentally measured with different accumulation statistics (red dots: 2 M counts, blue dots: 20 k counts).

Table I. Results of the LSF analysis for the experimentally-obtained PAL data.

<table>
<thead>
<tr>
<th>LSF #</th>
<th>Counts</th>
<th>Constraint</th>
<th>$\tau_1$ [ps]</th>
<th>$\tau_2$ [ps]</th>
<th>$I_1$ [%]</th>
<th>$I_2$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2 M</td>
<td>NO</td>
<td>223.5 (1.9)</td>
<td>379.1 (14.6)</td>
<td>88.2 (2.0)</td>
<td>11.8 (2.0)</td>
</tr>
<tr>
<td>2</td>
<td>2 M</td>
<td>YES</td>
<td>223.6 (0.7)</td>
<td>380 (--</td>
<td>88.4 (0.3)</td>
<td>11.6 (0.3)</td>
</tr>
<tr>
<td>3</td>
<td>20 k</td>
<td>NO</td>
<td>223.8 (16.2)</td>
<td>399.6 (173.5)</td>
<td>90.8 (15.9)</td>
<td>9.2 (15.9)</td>
</tr>
<tr>
<td>4</td>
<td>20 k</td>
<td>YES</td>
<td>221.9 (6.0)</td>
<td>380 (--</td>
<td>88.7 (2.8)</td>
<td>11.3 (2.8)</td>
</tr>
</tbody>
</table>

The values in parentheses represent the respective standard errors.

Fig. 3. Variation of $\tau_1$ for the data of 20 k counts, analyzed with and without constraining the 2nd lifetime, as a function of measurement number. The broken line represents the average value of 219 ps for $\tau_1$ analyzed with parameter constraining. The solid line is drawn to guide the eye.
In contrast, for LSF #4 analyzed with the constrained 2nd lifetime, \( \tau_1 \) is obtained with a standard uncertainty of 6 ps, regarded as an acceptable error, and is in good agreement with that obtained with higher statistics.

The effect of constraining the 2nd parameter on the resultant \( \tau_1 \) with lower statistics was further examined in terms of measurement repeatability. Figure 3 shows the variation of \( \tau_1 \) analyzed with and without constraining the 2nd lifetime as a function of measurement number. The variance of \( \tau_1 \) with parameter constraining is significantly reduced in comparison with that without parameter constraining, in which each standard deviation is \( \sim 6.3 \) ps and \( \sim 28 \) ps for \( \tau_1 \) with and without parameter constraining, respectively. This indicates that constraining the source lifetime is an effective method to attain the sample lifetime with higher precision from PAL data with relatively lower statistics.

4. Summary

We have examined the effect of constraining the source lifetime parameter during least-square fitting (LSF analysis) of positron annihilation lifetime (PAL) data on the resultant sample lifetime. The obtained results suggested that constraining the Kapton positron source lifetime during LSF analysis results in relatively minor uncertainty of the sample lifetime in comparison with that due to the measurement statistics. We have also assessed the constraint effect using experimental PAL data for single-crystal silicon with different measurement statistics. It was demonstrated that constraining the source lifetime is an effective method to attain the sample lifetime with higher precision from PAL data with relatively low counting statistics, indicating that the PAL technique is applicable to in-line measurements for product inspection in industry, requiring short measurement times [5].

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