Doppler broadening of positron annihilation radiation: fitting the coincidence spectrum

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Abstract

The profile of the annihilation radiation in aluminum using positrons from a $^{22}$Na source was observed in a two-HPGe-detectors arrangement. The coincidence photon energy spectrum was fitted using a model function, accounting for both Doppler broadening and detector system response. Intensities of the thermalized positron annihilation with band, 2p, 2s, and 1s electrons, and in-flight positron annihilation were fitted. The binding energies of the 1s, 2s, and 2p electrons and the Fermi cutoff parameters of the band electrons were also fitted.

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1. Introduction

Positrons injected into materials are rapidly thermalized before annihilation with surrounding electrons, when, in most cases, two gamma-rays are emitted. These gamma-rays are Doppler shifted due to the center of mass velocity of the electron–positron pair. Since the center of mass velocity depends on the electron momentum, the measurement of the gamma-ray energies can provide information about the electron momentum distribution. This technique has been successfully used in the study of materials, especially after the introduction of a second detector to the experimental setup by Lynn et al. [1].

The energies of the emitted photons are given by

$$E_{\pm} = \frac{m_0 c^2}{2} - \frac{B_i}{2} \pm \frac{p_z c}{2}$$

where $m_0 c^2$ is the electron rest energy, $B_i$ is the binding energy of the annihilated electron, and $p_z$...
is the longitudinal component of the electron
momentum.

When just one detector is used, the annihilation
radiation yields a gamma-ray peak at 511 keV
broadened both by the Doppler effect given by

$$\frac{\sqrt{\langle p^2 \rangle} c}{2}$$

(2)

where $\langle \rangle$ stands for mean value, and by the
detector response function. When two detectors
are used, a coincidence energy spectrum with a
peak at $511 \text{ keV} \times 511 \text{ keV}$ is obtained. Calling $E_1$
and $E_2$ the main axes of the coincidence energy
spectrum, the detector system response function
broadens the peak along the $E_1 = E_2$ direction,
while along the $E_1 + E_2 \approx 2mc^2$ direction both
the Doppler effect and the detector response
contribute to the broadening. Since electrons of
different shells can be annihilated, the coincidence
peak is a superposition of different peaks slightly
shifted along the $E_1 = E_2$ direction by the
electron binding energies.

Usually, in the analysis of coincidence experi-
ments, the two-dimensional coincidence spectrum
is reduced to a projected one-dimensional spec-
trum by calculating the marginal distribution
along the $E_1 + E_2 = \text{constant}$ direction, that is,
the projection of the data in the coincidence
spectrum in a perpendicular plane along one of
its diagonals. In that procedure, the horizontal
axis is usually given in units of the electron
momentum (see, for instance, Refs. [2–4]). Com-
paring this projection with the results provided by
just one detector, some improvements are ob-
tained; the peak to background ratio is increased
by a few orders of magnitude and the Doppler
broadening to combined detectors resolution ratio
is increased by $\sqrt{2}$.

In spite of these improvements, some informa-
tion is lost after the projection. When projected,
some events due to the detectors secondary effects
that were out of the region of interest in the
coincidence spectrum are interwoven with the
main signal. Also, events due to positron annihi-
alation with conduction and bound electrons, which
were slightly separated by the binding energy in
the two-dimensional coincidence spectrum, are
summed together. Thus, the analysis of the two-
dimensional energy spectrum can reveal details
that are hidden in the one-dimensional projection.
In this paper we choose to analyze the coincidence
spectrum.

The Doppler-broadened gamma-ray spectra are
usually analyzed as follows. In a first step, the
electron momentum distribution and the expected
gamma-ray spectrum are theoretically calculated.
Next, one calculates the convolution of the
obtained spectrum with the system response
function, which is compared with the projected
spectrum. In this paper we opted for another
procedure. In a first step, the convolution of the
detector response function with empirical func-
tions to represent the gamma-rays emitted after
annihilation was calculated; all these functions
were parameterized. Next, the parameters were
fitted to the coincidence spectrum using the least-
squares method. The advantage of this approach is
that the fitted parameters can be compared with
other experimental results and theoretical values.
The handicap of the method is the difficulty in
choosing suitable functions to model the gamma-
ray energy distribution.

The main aim of this paper is to give a
contribution to the statistical analysis of the
experimental data of the Doppler broadening of
positron annihilation radiation.

2. Experiment and the model function

2.1. Experimental

The annihilation gamma-rays were measured by
two HPGe detectors in diametrically opposed
positions and separated by 15 cm, with a $4 \times 10^5\text{ Bq}$ $^{22}\text{Na}$ source placed between two 2 mm thick
aluminum sheets (99.999% pure and annealed for
6 h at $600^\circ\text{C}$). The coincidence time window was
set to 180 ns. An $^{192}\text{Ir}$ source was simultaneously
measured to provide the detector energy calibra-
tions and to follow any energy drift during the
experiment. The measurement run lasted for 200 h,
when $1.5 \times 10^7$ events in the peak region were
accumulated. The contour plot of the coincidence
spectrum is presented in Fig. 1, where the crest
along the line $E_1 + E_2 \approx 1022 \text{ keV}$ is mainly due
to at-rest positron annihilation with core and band electrons. The ridges parallel to the principal axes come from the coincidence between an annihilation gamma-ray with a Compton scattered gamma-ray, coming from either the other annihilation radiation or the 1274 keV gamma-ray following $^{22}$Na decay. The complete explanation of the spectrum, however, requires many more details, given below.

2.2. Model function

The measured spectrum was modeled by a function which was fitted, after convolution with the detector system response function (see Section 2.3), to the experimental histogram. The model included at-rest positron annihilation with core and band electrons, and in-flight positron annihilation. The model function was determined from a qualitative analysis of the experimental data and published theoretical results [2–4].

Positron annihilation with band electrons was fitted by three arcs of parabola and one Gaussian along the line $E_1 + E_2 = 1022$ keV, given by

$$f_b = \sum_{i=1}^{3} C_i (E_1 - E_2 - \alpha_i)(E_1 - E_2 + \alpha_i) + A_i e^{-(E_1 - E_2^2)/2\sigma_i^2}$$

along the line $E_1 + E_2 + B_1s = 2m_0c^2$, where $B_1s$ is the 1s electron binding energy. The fitted parameters are $A_{1s}$, $\sigma_{1s}$, and $B_{1s}$.

Positron annihilation with 2s electrons has a dominant maximum in the region $p < 20 \times 10^{-3}m_0c$ and a large tail that extends beyond $p \sim 60 \times 10^{-3}m_0c$ [2–4]. In consequence, we represent this annihilation by two Gaussians

$$f_{2s} = \frac{A_{2s} e^{-(E_1 - E_2^2)/2\sigma_{2s}^2}}{\sqrt{2\pi}\sigma_{2s}} + \frac{A'_{2s} e^{-(E_1 - E_2^2)/2\sigma'_{2s}^2}}{\sqrt{2\pi}\sigma'_{2s}}$$

along the line $E_1 + E_2 + B_{2s} = 2m_0c^2$, where $B_{2s}$ is the binding energy of the 2s electrons. $B_{2s}$, $A_{2s}$, $A'_{2s}$, $\sigma_{2s}$, and $\sigma'_{2s}$ were fitted.

Positron annihilation with 2p electrons has just a maximum in the analyzed region [2,3]. Thus, it was fitted by just one Gaussian

$$f_{2p} = \frac{A_{2p} e^{-(E_1 - E_2^2)/2\sigma_{2p}^2}}{\sqrt{2\pi}\sigma_{2p}}$$

along the line $E_1 + E_2 + B_{2p} = 2m_0c^2$. In this case we fitted $A_{2p}$, $\sigma_{2p}$, and the binding energy $B_{2p}$. Since positron annihilation radiation with $2p_{1/2}$ and $2p_{3/2}$ electrons show the same dependence on the electron momentum, they cannot be separated in the fit.
In-flight positron annihilation has been observed in recent experiments [5–9] and was considered in the fitting. When a positron annihilates in-flight with a low-momentum electron, two gamma-rays are emitted with energies \( E_1 \) and \( E_2 \) given by
\[
E_{1,2} = m_0 c^2 + \frac{p^2}{2m_0} - B \pm \frac{p_{\parallel} c}{2},
\]
where \( p \) is the total positron momentum, \( p_{\parallel} \) is its component parallel to the gamma-ray emission direction, and \( B \) is the electron binding energy. When \( p_{\parallel} \) takes its extreme value \( p \), then \( E_1 \) and \( E_2 \) take their extreme values, too. Neglecting \( B \) in Eq. (7), the extreme values of \( E_1 \) and \( E_2 \) are given by
\[
E_{1,2}^m = m_0 c^2 + \frac{p^2}{4m_0} \pm \frac{pc}{2}.
\]
This pair of equations can be written as
\[
\left( E_1^m - \frac{3}{2} m_0 c^2 \right)^2 + \left( E_2^m - \frac{3}{2} m_0 c^2 \right)^2 = \frac{p^4}{8m_0^2} + \frac{m_0^2 c^4}{2}.
\]
Consequently, for small positron momentum \( (p \ll mc) \), corresponding to points near the 511 keV \( \times 511 \) keV peak, this equation describes a circular arc which is barely seen in Fig. 1. Fig. 2 shows a projection of the spectrum along the radial distance, measured from the center of the circle \( E_1 = E_2 = 3m_0 c^2/2 \), summing the counts in the regions identified in the figure inset. This one-dimensional projected spectrum shows a clear bump just above 1022 keV, confirming the existence of a circular ridge in the coincidence spectrum.

Thus, the last component included in the fit is the in-flight positron annihilation, empirically approximated by
\[
f_\text{f} = \frac{A_f e^{-2d_\text{f} d/(E_1-E_2)^2/2\sigma_\text{f}^2}}{\sqrt{2\pi\sigma_\text{f}}}
\]
for points \((E_1, E_2)\) inside the circle given by Eq. (9), where \( d \) is the distance (in energy) between \((E_1, E_2)\) and the circular arc given by
\[
d = \frac{m_0 c^2}{\sqrt{2}} - \sqrt{\left( E_1 - \frac{3m_0 c^2}{2} \right)^2 + \left( E_2 - \frac{3m_0 c^2}{2} \right)^2}.
\]

For points outside the circle the contribution of in-flight positron annihilation is zero. The empirical parameters \( A_f, \lambda, \) and \( \sigma_\text{f} \) were fitted.

Fig. 2. Radial projection of the selected regions (shown in the inset) of the coincidence spectrum; see text for definition of the radial coordinate.
2.3. Response function and background

The detectors response functions are given by a Gaussian part and a non-Gaussian part. The Gaussian part has two parameters corresponding to the detectors’ resolution. The non-Gaussian part has two parameters corresponding to ballistic defects and incomplete charge collection, was fitted by two internal \((E < E_c)\) and two external \((E > E_c)\) exponential queues for each detector, with a total of 16 parameters.

Two internal and two external ridges along the lines \(E_1 = 511\) keV and \(E_2 = 511\) keV, with amplitudes proportional to the peak intensity, were included in the fit to take into account photons from the scattering of annihilation or 1274 keV gamma-rays in coincidence with an annihilation gamma-ray, corresponding to four intensity parameters. It was assumed that the number of gamma-rays in coincidence with an annihilation \(73\) eV, \(0.118\) keV, and \(1.56\) keV [13]. The adjusted Fermi cutoff parameters agree well with their recommended values, respectively \(73\) eV, \(0.118\) keV, and \(1.56\) keV [13]. The adjusted Fermi cutoff parameters agree with the values given in Ref. [4].

The relative intensities of positron annihilation with band and core electrons agree with other works [2–4]. The fitted binding energies of \(2p\), \(2s\), and \(1s\) electrons, \(70(9)\) eV, \(0.12(5)\) keV, and \(1.45(13)\) keV, agree well with their recommended values, respectively \(73\) eV, \(0.118\) keV, and \(1.56\) keV [13]. The obtained \(\chi^2\) value was 6.9615 \times 10^4 for 6.2448 \times 10^4 degrees of freedom (52 parameters fitted to 250 \times 250 channels), giving a reduced \(\chi^2\) of 1.11.

The most interesting results are given in Table 1.

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Fig. 3 shows the fitted coincidence spectrum. Fig. 4 shows the fitted and experimental projected Doppler profiles, including the \(1s\), \(2s\), \(2p\), \(1s\) electron annihilations, and in-flight positron annihilation components.

### Table 1

| Selection of fitted parameters: relative intensity of positron annihilation with band, \(2p\), \(2s\), and \(1s\) electrons; in-flight annihilation intensity; the three Fermi cutoff parameters; and \(2p\), \(2s\), and \(1s\) electron binding energies |
|---|---|
| Band annihilation (%) | 91.9(13) |
| \(2p\) annihilation (%) | 5.42(32) |
| \(2s\) annihilation (%) | 2.6(12) |
| \(1s\) annihilation (%) | 0.0145(25) |
| In-flight annihilation* (%) | 0.066(3) |
| \(\sigma_1(10^{-3}m_0c)\) | 6.820(8) |
| \(\sigma_2(10^{-3}m_0c)\) | 10.87(9) |
| \(\sigma_3(10^{-3}m_0c)\) | 16.24(11) |
| \(2p\) binding energy (keV) | 0.070(9) |
| \(2s\) binding energy (keV) | 0.12(5) |
| \(1s\) binding energy (keV) | 1.45(13) |

*In the fitted region.
The squares of the relative residues given by

\[ \Delta_{ij} = \frac{(n_{ij} - F_{ij})^2}{F_{ij}} \]  

are shown in Fig. 5, where no special feature stands out against the statistical fluctuation.

4. Discussion and conclusion

The annihilation components fitted to the observed peak span several orders of magnitude and require 52 different parameters in the least-squares procedure, which were described in Sections 2.2 and 2.3. The experimental data allowed a reliable determination of almost all the fitted parameters; the most difficult cases and the exceptions will be noted below.

Surprising as it may seem, the 1s bound electron annihilation intensity, although blurred with the strong band electron annihilation intensity, can be extracted precisely. This happens because an important fraction of gamma-rays from these events falls in an energy region where there are not many events from band electron annihilation. First, the 1s annihilation gamma-ray energies are displaced from the dominant events, annihilation with band electrons distributed around the line \( E_1 + E_2 \approx 1022 \text{ keV} \), by the binding energy, therefore lying around the line \( E_1 + E_2 = 2m_0c^2 - B_{1s} \). Additionally, the Doppler shift for annihilation with 1s electrons tends to be larger than with band electrons due to the higher momentum of the electron, resulting in many annihilation gamma-rays with energies very different from 511 keV.

Fig. 5. Contour plot of the relative residue spectrum.
and counting statistics of the experiment allowed fair observation of the phenomenon. From the statistical point of view, this is reflected in a large difference in $\chi^2$ when including or not 1s annihilation in the model function; in this experiment, $\chi^2$ decreased by 480 when reducing the number of degrees of freedom by three, corresponding to the inclusion of the parameters $A_{1s}$, $\sigma_{1s}$, and $B_{1s}$ (see Eq. (4)) in the fit.

Two other weak components, representing phenomena secondary to this work, could also be fitted because they show up themselves in energy regions away from the dominant peak. One of them describe the simultaneous detection by each detector of one annihilation photon scattered in the other, which are located along $E_1 + E_2 = 1022$ keV, and can be clearly observed only far from the main peak. The other is the in-flight positron annihilation, also very weak, which was resolved from the other structures due to its typical circular profile in the gamma–gamma coincidence energy plan, extending beyond the region dominated by the 511 keV × 511 keV peak.

The parameters that cannot be extracted reliably are the intensities and widths of the two Gaussian functions fitted to take into account annihilation with 2s electrons, described by Eq. (5). However, there is no reason to observe annihilation with 2p and 1s electrons and not with 2s electrons; since the fitted parameters agree well with expectations, it was kept in the fitting procedure. On account of the node in the radial wave function of 2s electrons, the momentum wave function must have two components, which led to the choice of two Gaussians to model the annihilation intensity.

In conclusion, we have found that a complete statistical analysis of the coincidence Doppler annihilation radiation spectrum is possible and provides some parameters and their uncertainties that are difficult to obtain from a projected spectrum. The electron binding energies and Fermi cutoff parameters were determined and the obtained results agree well with published values, corroborating the procedure. Better model functions will improve this procedure.

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References


