Nuclear Instruments and Methods in Physics Research A 609 (2009) 244–249

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Statistical analysis of the Doppler broadening coincidence spectrum of electron–positron annihilation radiation in silicon

E. do Nascimento [a,b,*], O. Helene [a], V.R. Vanin [a], M.T.F. da Cruz [a,1], M. Moralles [c]

[a] Instituto de Física, Universidade de São Paulo, CP 66318, CEP 05315-970, São Paulo, SP, Brazil
[b] Facultad de Física, Universitat de Barcelona, CEP 08028, Barcelona, Spain
[c] Instituto de Pesquisas Energéticas e Nucleares-IPEN/CNEN-SP, CEP 05508-000, São Paulo, SP, Brazil

ARTICLE INFO

Article history:
Received 27 March 2008
Received in revised form
21 July 2009
Accepted 28 July 2009
Available online 4 August 2009

Keywords:
Doppler broadening
Positron annihilation
Least-squares method
Silicon

ABSTRACT

We report a statistical analysis of Doppler broadening coincidence data of electron–positron annihilation radiation in silicon using a ^{22}Na source. The Doppler broadening coincidence spectrum was fit using a model function that included positron annihilation at rest with 1s, 2s, 2p, and valence band electrons. In-flight positron annihilation was also fit. The response functions of the detectors accounted for backscattering, combinations of Compton effects, pileup, ballistic deficit, and pulse-shaping problems. The procedure allows the quantitative determination of positron annihilation with core and valence electron intensities as well as their standard deviations directly from the experimental spectrum. The results obtained for the core and valence band electron annihilation intensities were 2.56(9)% and 97.44(9)%, respectively. These intensities are consistent with published experimental data treated by conventional analysis methods. This new procedure has the advantage of allowing one to distinguish additional effects from those associated with the detection system response function.

1. Introduction

Positron annihilation spectroscopy is a well-established technique that has been extensively used to probe condensed matter systems [1,2]. With the introduction of a second Ge detector enabling the measurement of both annihilation photons [3], Doppler broadening spectroscopy takes advantage of coincidence techniques, adding characteristics of high selectivity of the detected signal and great improvement of the analysis procedure. A gain of $\sim\sqrt{2}$ in the combined energy resolution for the observation of the annihilation peak and an important reduction in the background are obtained when compared with measurements taken with only one Ge detector. Another important feature that has not received much attention is that in these coincidence measurements one obtains two-dimensional (2D) energy spectra. Hence, the Doppler broadening coincidence spectrum (DBCS) shows effects of the electron–positron annihilation process and the detection system, which can be broken down into their components because they are distributed throughout the 2D spectrum. In the annihilation processes, both energy and momentum are conserved [1]; however, precise measurements in solids [4–6] have demonstrated that electron–positron annihilation radiation has an energy deficit, with the total photon energy below $2m_0c^2$. This deficit is due to the binding energies of the annihilated electrons [6,7] and this information is always present in the photons of annihilations that occur in surface and bulk metals or semiconductors, as well as information related to the momentum of electron–positron pairs. However, these properties have never been used for the analysis of the Doppler broadening spectrum.

The behavior of positrons near nuclei is essentially independent of the atomic environment and, therefore, the overlaps of their wavefunctions with the various core states of an atom in a solid are well represented in a purely atomic description [8]. On the other hand, information about the atomic environment can be obtained from the contribution of the valence/conduction band to Doppler broadening in the annihilation photon spectrum. Thus, in this work, we have applied a statistical treatment similar to that used for the analysis of the DBCS measured in aluminum [9–11], where a model function was fit to the experimental spectrum using a non-linear least-squares method to get information about the intensities of positron annihilation with 1s, 2s, 2p, and valence band electrons in silicon. The fitting procedure is similar to the method widely used in positron lifetime spectroscopy, for which several computer programs based on non-linear fitting routines are available [1].

In this work, we performed measurements on a Czochralski-grown silicon crystal (Cz-Si) sample. Silicon is a semiconductor of great technological importance [12–19] and has an electronic

* Corresponding author at: Instituto de Física, Universidade de São Paulo, CP 66318, CEP 05315-970, São Paulo, SP, Brazil.
E-mail address: eduardon@if.usp.br (E. do. Nascimento).
[1] Deceased.

0168-9002/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2009.07.051

configuration that differs from that of aluminum by one excess electron, requiring modifications in the description of core electrons. This choice was due to the fact that, at the beginning of semiconductor studies with positrons, the use of models developed for metals and applied to semiconductors led to false results and mistaken interpretations about semiconductors [1]. As in the case of aluminum, our goal in this work is to accomplish a realistic description of the DBCS of electron–positron annihilation photons in silicon that, together with information on the response function of the detection system, allows one to directly obtain the annihilation rates with all electrons and their momentum distributions from this spectrum.

2. Method

2.1. General

In positron annihilation experiments, the vast majority of positrons penetrate the sample, thermalize very quickly (within a few ps) [1], and diffuse before annihilation. In DBCS, both annihilation photons are measured in coincidence using two Ge detectors positioned collinearly with the sample.

Let E_1 and E_2 be the energies of the annihilation photons observed by detectors 1 and 2, respectively. For thermalized positrons, the energies of the emitted photons of the electron–positron annihilation are given by

$$E_{1,2} = m_0c^2 - \frac{B_i}{2} \pm \frac{p_zc}{2} \tag{1}$$

where m_0c^2 is the electron rest mass energy, B_i the electron binding energy, c the speed of light, and p_z the momentum component of the positron–electron pair in the direction of the emitted annihilation photons. It is useful to define two other energies: the sum, $E_1+E_2 = 2m_0c^2-B_i$, and the difference, $E_1-E_2 = p_zc$. The sum energy is equal to the total energy of the electron–positron system before annihilation. The Doppler broadening profile is determined with the help of the energy difference, where the energy of one annihilation photon is Doppler shifted upwards by $+p_zc/2$, and that of the other photon is Doppler shifted downwards by $-p_zc/2$. The energies of both annihilation photons and the time interval between their detections are recorded, and the event is stored in a 2D array [9–11].

Fig. 1 shows the contour plot of the experimental DBCS obtained from a measurement performed with a positron source

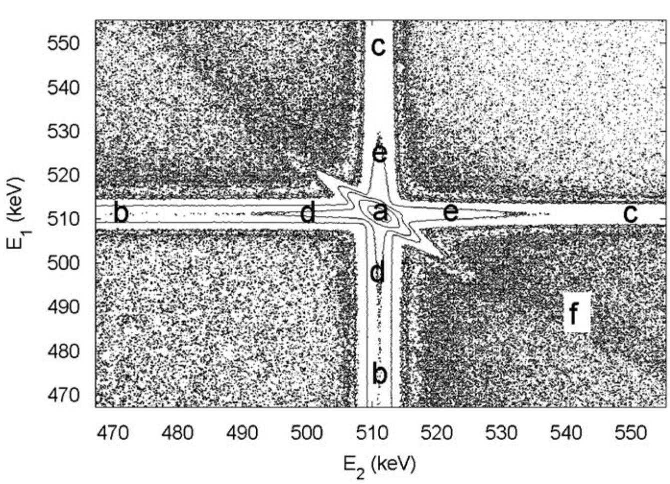

Fig. 1. Contour plot of the experimental DBCS of annihilation radiation in silicon (Cz-Si). E_1 denotes the energy measured by detector 1 and E_2 that by detector 2.

deposited between two silicon disks. In this figure, the DBCS may be interpreted as follows: the peak identified as a has a broadening along the line $E_1+E_2 = 2m_0c^2$ due to Doppler shift of the annihilation radiation. Furthermore, at both ends of this line there are subtle structures corresponding to backscattered annihilation photons. The two ridges extending from the peak towards lower energies ($E_1 < m_0c^2$ or $E_2 < m_0c^2$), identified as b, are called internal ridges, mainly due to coincidence events where one of the annihilation photons suffers Compton scattering. Since we have used ^{22}Na as the positron source, which emits a 1274 keV gamma-ray in its decay, ridges are formed towards higher energies. These are the external ridges (c in Fig. 1), which are due to the coincidence of one annihilation photon with a Compton-scattered 1274 keV gamma-ray. The spectrum also contains exponential tails due to a ballistic deficit, pulse-shaping problems (d in Fig. 1), and pileup (e in Fig. 1).

Although most positrons thermalize before annihilation, there is a small fraction that annihilate before thermalizing. This is the so-called in-flight positron annihilation. When positrons annihilate in flight with a low-momentum electron two photons with energies E_1 and E_2 are emitted and the total energy is given by

$$E_1 + E_2 = 2m_0c^2 + K_+ - B_i \tag{2}$$

where K_+ is the positron kinetic energy at the instant of annihilation. With a small number of events and a larger energy spread [20–24], in-flight positron annihilation is usually masked by the background arising from thermalized positron annihilation. These annihilations are distributed along a circular arc near the 511–511 keV peak in the DBCS (f in Fig. 1).

Fig. 1 also shows a background between the internal ridges that is mainly associated with Compton scattering of both photons in coincidence. There is even more information in this spectrum, although too subtle to be visually noticed, as the various distributions related to annihilation with core electrons are shifted off the line $E_1+E_2 = 2m_0c^2$ by their binding energies as discussed below.

2.2. Experimental setup

A 10 μCi (3.7×10^5 Bq) ^{22}NaCl source was placed between two silicon (Cz-Si) disks, both 2 mm thick. The crystals were positioned with their orientation (111) collinear with the axes of the detectors. Positron lifetime measurements were performed by means of the 1274 keV–511 keV coincidence using a spectrometer with one plastic scintillator and one BaF_2 detector. This system had a time resolution of 360 ps. Three lifetime components plus background were fit to a spectrum with 1.1×10^7 counts. The lifetime components were exponential functions convoluted with a Gaussian function that approximately describes the response function of the time measurements obtained with the spectrometer. The reduced χ^2 obtained in the fit performed in the time range −150 ps to 18 ns was 1.07 with 1357 degrees of freedom. The results indicate that 98.38(20)% of the positrons decay with the characteristic lifetime of annihilation in the silicon bulk. The other components have lifetimes of 552(20) ps and 3.06(19) ns with intensities of 1.36(16)% and 0.26(4)%, respectively, corresponding to minor annihilation contributions in NaCl surfaces and defects. These results confirmed the good quality of the silicon crystal and indicated successful NaCl deposition onto the sample.

The coincident Doppler broadening measurement was performed using a detection system that consisted of two HPGe Ortec detectors (detector 1 with 50 cm^3 and detector 2 with 164 cm^3), analog-to-digital converters (ADC-Ortec AD413), a spectroscopy amplifier (Ortec 673), a fast-filter amplifier (FFA-Ortec 579), a