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High-momentum analysis in Doppler spectroscopy

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ABSTRACT

Unique information about the chemical vicinity of positron annihilation sites is provided by the contribution of high electron momenta to the Doppler spectrum, since this momentum range is characteristic for the annihilation with core electrons and hence element specific. However, the corresponding energy region in the spectrum is overlaid by a huge background caused by the annihilation radiation itself and the Compton spectrum of other gamma lines having an energy above 511 keV. Usually these backgrounds are reduced by measuring both annihilation quanta in coincidence.

By mathematically analyzing the background contributions, we open another possibility to obtain the high-momentum region employing one single germanium detector. A necessary precondition is employing either background-free positron beams or a low-background positron source, e.g. ^{68}Ge, instead of the widely used positron emitter ^{22}Na. The ^{68}Ge-source emits positrons with an endpoint energy of about 1.9 MeV, where as the contribution of gamma quanta having higher energies than the annihilation radiation at 511 keV is negligible low.

When analyzing spectra from metals and semiconductors according to the described background subtraction, the same information contained in the momentum range up to $35 \times 10^{-3} m_0c$ or beyond can be extracted, as if the spectra were measured employing a coincidence setup with two Ge-detectors.

1. Introduction

Positron annihilation spectroscopy (PAS) is an unique tool with an outstanding high sensitivity for the non-destructive detection of open-volume defects metals [1,2] and semiconductors [3]. The Doppler broadening of the annihilation radiation provides information about the momenta of the annihilation electron–positron pairs (Doppler broadening spectroscopy, DBS). In the ideal lattice, a thermalized positron resides mainly in the interstitial regions repelled by the positively charged nuclei, and its wave function overlaps partly with valence and partly with core electrons. Localized in an open-volume defect the positron annihilates predominantly with valence electrons. This is reflected in a significant change of the shape of the 511 keV annihilation peak. The contribution of electrons having low momenta correlated to the annihilation with valence electrons is quantified by the shape parameter *S*, which is defined as the ratio of a central peak area to the total integral of the peak [4]. The contribution of high electron momenta provides unique information about the chemical surrounding of positron annihilation sites, since this momentum range is characteristic for core electrons being element specific. This part of the spectrum can also be characterized by a shape parameter, the *W*-parameter, which is the ratio of the wing region of the peak to the total peak integral [5].

Unfortunately, the high-momentum region in the Doppler spectrum is overlaid by a background being up to three orders of magnitude higher than the signal. This is due to the background caused by the annihilation peak itself and Compton scattering of gammas higher than 511 keV as well. Traditionally, this background is reduced by measuring both annihilation quanta in coincidence employing a pair of Ge-detectors arranged opposite to each other (coincidence Doppler broadening spectroscopy, CDBS) [6–8].

But even when the Doppler spectrum is obtained by one single Ge-detector, the chemical information can be extracted by analyzing and subtracting the background employing a sufficiently well-suited mathematical model (high-momentum analysis, HMA). In the momentum range up to $35 \times 10^{-3} m_0c$, the results are equivalent with data obtained by CDBS [9] and numerical calculations [10]. When the preconditions described below are considered, this technique can be applied to any Doppler spectrometer.

2. Preconditions for HMA

Technically, the HMA method describes and subtracts the background caused by the annihilation radiation itself. Hence, the

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raw Doppler data should be free of background caused by any gammas with higher energies than 511 keV. This can be achieved by either using a positron beam, where the detector is shielded against γ-radiation emitted by the positron source or by using a positron source which has a clean γ-spectrum with only a low contribution of high energy γ-radiation. One good candidate for that purpose is ^{68}Ge. The commonly used positron emitter ^{22}Na cannot be employed since the prompt gamma at 1275 keV produces a background not analyzable by HMA.

^{68}Ge decays with a reasonable half-life into ^{68}Ga (100% EC, $\tau_{1/2}$ = 270.8 d) which decays via positron emission into ^{68}Zn (90% β^+, $\tau_{1/2}$ = 68.4 m). Even though, the strongest gamma line of ^{68}Ga has an energy of 1077.4 keV, it can be ignored since it only contributes with 3% to the total decay [11,12]. ^{68}Ge can be produced at a cyclotron facility by a (d,3n) reaction from natural gallium. This reaction has its threshold at 14.1 MeV and with 27 MeV deuterons the maximum cross-section of 550 mB is reached [13]. Using accelerated deuterons the isotope ^{69}Ge is produced with a three times higher cross-section. On account of its comparatively short half-life of 39 h, it has vanished after 4 weeks. When a gallium phosphide (GaP) wafer is used as target for irradiation, the resulting positron source is well suited for laboratory purposes, since it is solid state and contamination-free and can be employed without coverage. The β-spectrum of ^{68}Ga has its endpoint energy at 1.89 MeV which is significantly higher than that of ^{22}Na (544 keV). This results in a deeper penetration of the positrons into the sample, which has to be concerned when designing an experiment.

Any drift of the electronics during data acquisition will artificially broaden the annihilation line and can result in an unusable spectrum. To avoid this, a stabilization is required. This can be realized in an accurate way by a combination of an analog hardware stabilization on the gamma decay of a suitable isotope and a digital stabilization after data acquisition. For the former, ^{7}Be is an ideal isotope having just one gamma line at 477.6 keV, which can be measured simultaneously to the annihilation radiation without disturbing the annihilation peak. For the latter, the post-processing of the data can be done by writing the spectra to hard disk in minute intervals and a subsequent adding of the data into one file after a digital gain/zero correction on both the ^{7}Be line and the annihilation line.

Before the background can be removed from the Doppler spectrum, the differential efficiency of the Ge-detector has to be corrected. The efficiency is a non-linear function of energy Eff(E), which can be measured by obtaining the γ-spectrum of an isotope having a bunch of γ-lines over a broad range of energies, e.g. ^{152}Eu. According to [14], the experimental function Eff(E) – which is obtained by comparing the measured intensities to the known values – can be parameterized around 511 keV by an exponential function of energy. As Fig. 1 shows, Eff(E) varies more than ±5% in the region of interest and, hence, cannot be ignored. When preparing the spectra for HMA, the corrected intensities are calculated by dividing the measured intensities with Eff(E) normalized to Eff(511 keV).

3. Background subtraction

One fundamental aspect of a gamma spectrometer is its finite energy resolution arising from the Fano-factor and statistical noise produced by the electronics. This results in a Gaussian distribution of all γ-events recorded. Fig. 2 gives a schematic overview of the shapes of a γ-line and the background components described below in an hypothetic ideal Ge-detector with zero energy resolution and a real Ge-detector. The transition between both can be understood as a convolution with Gaussian noise.

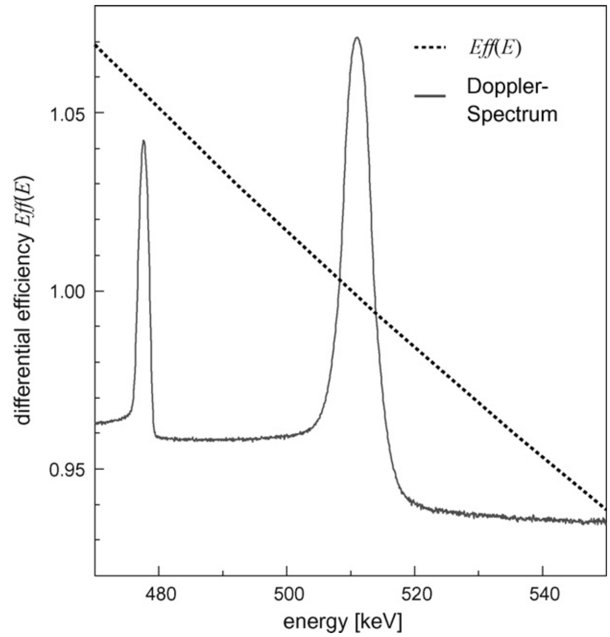

Fig. 1. Measured differential efficiency curve Eff(E) around 511 keV for a HPGe-detector (dotted line). The region of interest in the Doppler spectrum containing the annihilation peak and the ^{7}Be monitor line is indicated as well.

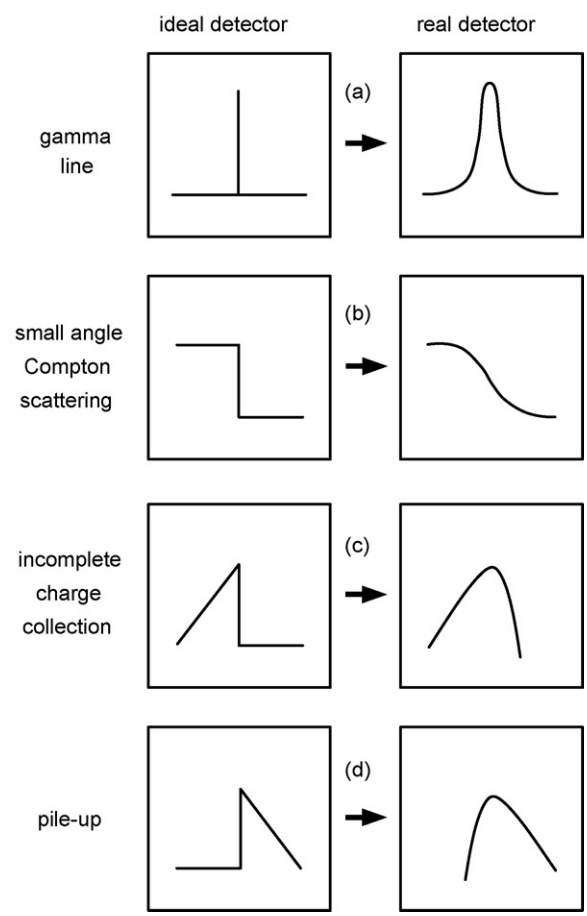

Fig. 2. Sketch of a γ-line and background components in a hypothetic ideal Ge-detector having zero energy resolution and in a real detector. The arrows indicate convolution with Gaussian noise.