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Coincidence Doppler-broadening Measurements of Positron Annihilation in Transition Metals

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1. Introduction

Since 1940s, some experimental techniques based on positron annihilation have been widely used to study properties of solid. Among them, Doppler-broadening spectroscopy (DOBS) plays an important role to obtain elemental information on the surroundings of the positron annihilation site. The annihilation radiation contains information on the electron momentum distribution at the annihilation site, as a result of momentum conservation during the annihilation process [1-3]. A drawback of DOBS is the low positron annihilation probability of the signal with electrons of high momentum. In 1977, Lynn et al. improved the DOBS by adding a second detector in order to observe the second annihilation quantum in coincidence. In coincidence Doppler-broadening spectroscopy (CDOB), the peak to background ratio is drastically reduced by three orders of magnitude.

We have constructed a two-detector spectrometer for measurements of Doppler-broadened positron annihilation spectra. In order to identify chemical elements on surroundings of the annihilation site, basically it is necessary to obtain the annihilation signal from the pure material. In this study, we have investigated 8 transition metals (Ti, V, Cr, Fe, Co, Ni, Cu, Zn) and analyzed the CDOB data using the DualNTW and CDB program.

2. Experimental Setup

2.1 Coincidence Doppler-broadening Spectroscopy

For the measurement of the Doppler-broadened annihilation gamma-rays, 20 μCi of ^{22}Na-source was sandwiched between the two equal-thickness nickel foils (2.5 μm thick & 8 x 8 mm^2 area). As a result of β^+-decay, positrons are emitted from ^{22}Na isotope. The positrons penetrate the sample, thermalize, diffuse, and annihilate with electrons of sample. Since the momentum should be conserved in the annihilation process, the momentum of the electron-positron pair is transferred to the annihilation radiation pair.

For the measurement of the energy broadening of annihilation radiations, high-resolution energy-dispersive detector system, shown in Figure. 1, was used. Two liquid-nitrogen-cooled HPGe detectors of high efficiency were equipped for the registration of the collinear coincidence γ-quanta. The energy resolution of each detector was calibrated to be 1.1 keV (full width at half maximum, FWHM) at the 514 keV line of ^{85}Sr. The annihilation photons cause a charge separation that is converted by a preamplifier into an electrical pulse. And this signal can be registered after amplification in two-dimensional multi-channel analyzer (Labo, NT24-DUAL).

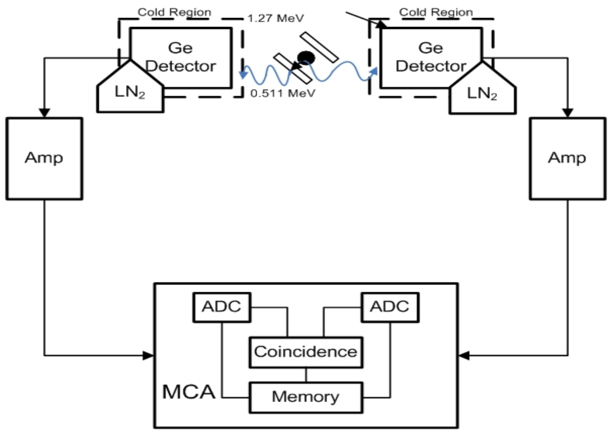

Figure 1. Scheme of coincidence Doppler-broadening spectroscopy system. (Amp: amplifier, ADC: analog-digital converter, MCA: multi-channel analyzer, LN_2: Liquid Nitrogen)

The quantitative evaluation was carried out with specific line shape parameters. The S (shape) parameter is defined as the area of the central low-momentum part of the spectrum, A_s, divided by the area below the whole curve A_0. The W (wing) parameter is taken in a high-momentum region far from the center. It is calculated as the area of the curve in a fixed energy interval A_w, divided by A_0.

$$S=\frac{A_S}{A_0},\ A_S=\int_{E_0-E_S}^{E_0+E_S} N_D dE\ ,\qquad W=\frac{A_W}{A_0},\ A_W=\int_{E_1}^{E_2} N_D dE$$

In this study, S region was set to 511±0.95 keV (≒ $P_L<4\text{x}10^{-3}\ m_oC$) and W region was set from 511+ 4.6 to 511+6.7 keV (≒$18\text{x}10^{-3} < P_L<30\text{x}10^{-3}\ m_oC$). But the absolute values of the parameters are meaningless, because they depend on the positrons of the windows. Only the changes of the parameters are of importance.

To compare the differences in the positron annihilation process depending on materials, we used ratio-curves obtained dividing the measured curves by the reference element [3, 4, 5]. The overall energy resolution of the system was estimated to be ~ 0.9 keV (FWHM), which corresponds to the momentum resolution of ~ 3.52×10^{-3} m_oC, from the cross-section along $E_1 - E_2 = CP_L$ direction.

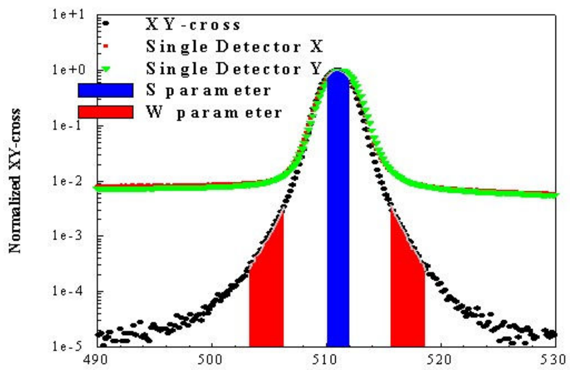

Figure 2. Doppler-broadening spectra for pure Ti.

The equipment is kept in at a constant room temperature to reduce the electronic drift. We used the DualNTW and CDB program to analyze the CDB data.

2.2 Samples Examined

Eight metal samples were prepared to check out the validity of our system. The list of these elements and characteristics of the measured samples are shown in Table 1. Specimens of 10×10×1 mm^3 were measured for 80000 s, resulting in the peak counts of at least 0.8 x 10^6 in the 1024x1024 coincidence matrix.

Sample	Purity	Group	Electron configuration	# of valence electrons
^{22}Ti	99.995	4	$(1s^2)(2s^22p^6)(3s^23p^6)3d^24s^2$	4
^{23}V	99.9	5	$(1s^2)(2s^22p^6)(3s^23p^6)3d^34s^2$	5
^{24}Cr	99.95	6	$(1s^2)(2s^22p^6)(3s^23p^6)3d^54s^1$	6
^{26}Fe	99.95	8	$(1s^2)(2s^22p^6)(3s^23p^6)3d^64s^2$	8
^{27}Co	99.999	9	$(1s^2)(2s^22p^6)(3s^23p^6)3d^74s^2$	9
^{28}Ni	99.99	10	$(1s^2)(2s^22p^6)(3s^23p^6)3d^84s^2$	10
^{29}Cu	99.997	11	$(1s^2)(2s^22p^6)(3s^23p^6)3d^94s^2$	11
^{30}Zn	99.995	12	$(1s^2)(2s^22p^6)(3s^23p^6)3d^{10}4s^2$	12

3. Results

3.1 S (shape) - and W (wing) - parameter

It is known that W parameter is more sensitive to the chemical surroundings of the annihilation site than the S parameter, because the core electrons having a high momentum make significant contributions to large energy deviations from the annihilation energy of 511 keV. In this experiment, we could obtain such results. As shown in Figure 3, we fiybd that the W parameter increase with the number of valence electrons. This means that the W (high momentum, core electron) parameter has characteristic properties of elements. However, those values depend on the system resolution. Therefore, it is necessary to compare them with reference spectrum.

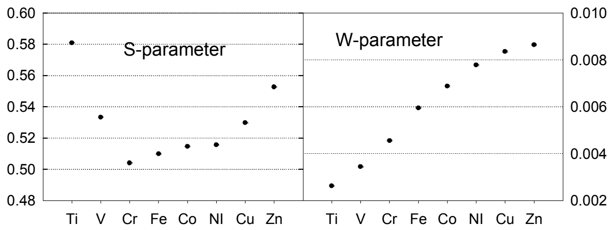

Figure 3. The CDB ratio spectra for different Cr concentration after normalizing relative to Fe.

3.2 Ratio-curves normalized to Ti

In this study, we chose Ti annihilation spectra as a reference. Because the number of Ti valence electrons is low, we can clearly compare the shape of ratio-curves. The results are shown in Figure 4. The shape is similar to the published results [3, 4, 5].

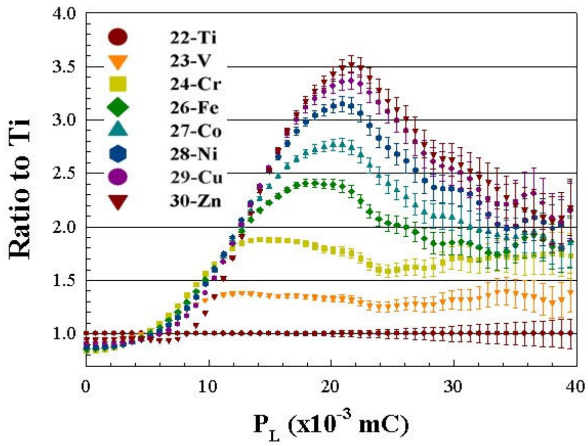

Figure 4. The CDB ratio spectra for different Cr concentration after normalizing relative to Fe.

4. Conclusion

The coincidence Doppler-broadening spectroscopy measurement techniques were applied to investigate 3-d metal elements and to set up our Doppler-system. This spectrometer can be used for various objectives such as the detection of the chemical impurities or gaining of information about the electronic structure of metals. In this study, we could verify the system validity and reproducibility by measuring CDOB of transition metals.

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