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Electron Structure Study through Positron Annihilation Spectra for a Feasibility Evaluation of an Elemental Specificity

Yongmin Kim[a,b], Junhyun Kwon[a], Changsun Kang[b], Whungwhoe Kim[a]
a Korea Atomic Energy Research Institute, 150 Deokjin, Yuseong, Daejeon 305-353
b Seoul National University, San 56-1, Sillim-dong, Gwank-gu, Seoul, 151-742, Korea
Corresponding Author:altair2@snu.ac.krs

1. Introduction

As an antiparticle of an electron, positrons can be used to investigate the microstructure of a material [1]. Positron annihilation techniques are the spectroscopies of the photons coming out from an annihilation of positrons and electrons. The result of the electron-positron reaction is an annihilation, i.e. both particles disappear. However, the momentum of the e^+-e^- pair is carried away by the annihilation radiation. Based on these results, an annihilation radiations are energy-shifted. The energy shift appears as a broadening of the annihilation line. Therefore the elemental information in the vicinity of the positron annihilation site can be obtained from a Doppler broadening spectroscopy (DBS) by measuring the local electron momentum distribution in a material. We have constructed a two-detector system for the measurement of the coincidence Doppler broadening (CDB) spectra, which enhances the peak to background ratio by five orders of a magnitude over the conventional method. In the present work, we obtained high momentum spectra by using a CDB spectroscopy to investigate the elemental specificity for pure metals and to clarify the chemical elements surrounding on annihilation site.

2. Experimental Setup

^{22}Ti, ^{23}V, ^{24}Cr, ^{26}Fe, ^{27}Co, ^{28}Ni, ^{29}Cu, ^{30}Zn transition metal samples were prepared to check out the elemental specificity of our CDB system. These samples have the same number of core electrons $(1s^2)(2s^2 2p^6)(3s^2 3p^6)$ while the number of valence electrons is different each other (4: $3d^2 4s^2$ ~ 12: $3d^{10} 4s^2$). And the positron source was prepared by placing a small amount of aqueous ^{22}NaCl on a thin nickel foil of 2.2 mg/cm^2 (2.5 μm) thick with an area of 8x8 mm^2. After each drop was dried, the source was sealed with a nickel foil of the same size. The positron source consisted of approximately 20 μCi of ^{22}Na.

For the CDB measurements, we used a collinear setup of two HPGe detectors. The detectors had an energy resolution of 1.1 keV (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. After an amplification, this pulse signal can be registered in a two-dimensional multi-channel analyzer (Labo, NT24- DUAL). The measurement for each sample was continued for about 22 hours, which corresponds to total counts of at least 6 x 10^7. The selection of the coincidence events that fulfill the condition $2m_oC^2$- 2.4 keV < E_T < $2m_oC^2$ + 2.4 keV, results in a significant improvement in the peak to background ratio over the conventional one-detector measurements. All the resolution values of the measured CDB spectra are within (0.88 ± 0.05) keV, which corresponds to a momentum resolution of ~ 3.44x10^{-3} m_oc.

Two-dimensional array of the counting rates measured from the CDB spectra are presented in Fig. 1. The diagonal line of Fig. 1 is used as the CDB spectra. Examples of the CDB spectra of Si, Ti, and Fe are shown in Fig. 2 to demonstrate the characteristic shape of each sample.

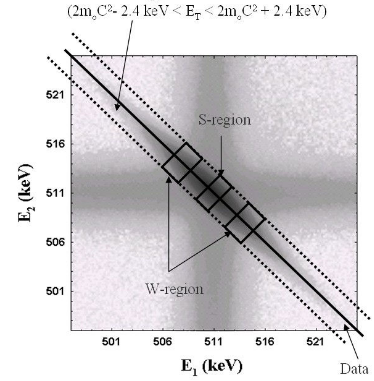

Figure 1. Two-dimensional array of counting rates measured from the two Doppler broadening spectra of well annealed Fe

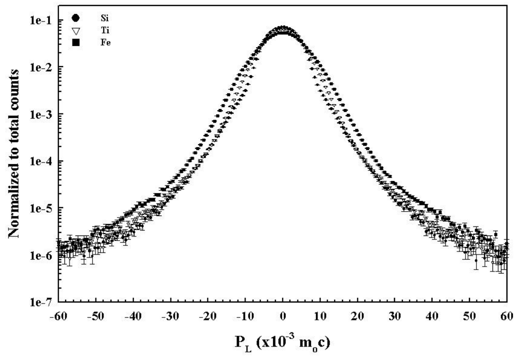

Figure 2. The coincidence Doppler broadening spectra from two-dimensional array of Si, Ti, and Fe. Each spectra has own characteristic shape

3. Data and Result

Usually, the measurements of the Doppler spectra are displayed by plotting the ratio curves for clarifying the differences in the spectra between elements [2, 3]. In this study, the CDB ratio curve was obtained by normalizing the CDB momentum distribution of the sample to the CDB momentum distribution of a pure Ti sample as in Fig. 3.

It is of interest to investigate the behavior of the ratio curve depending on the materials, particularly those close to each other in the Periodic table. Such ratio curves contain one or more characteristic peak, of which their position, shape, and magnitude can be used to identify a chemical species. This implies that the high momentum part of CDB spectra can be used to investigate an elemental specificity. It is clearly seen that the curve increases in the range of ($12 < P_L < 20$) x10^{-3} m_oc particularly in 3-d elements from Fig. 3.

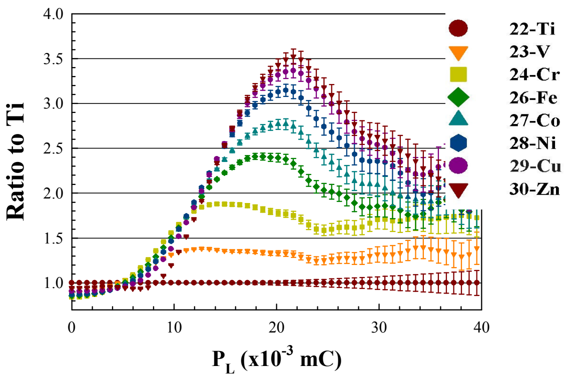

Figure 4. The CDB ratio spectra for transition metals after normalizing relative to Ti.

Based on these features, an attempt has been made to regress the CDB spectra in the range of ($12 < P_L < 20$) x10^{-3} m_oc by using the Sigmaplot program. The CDB spectra normalized to the total counts in the extracted region are shown in Fig. 4.

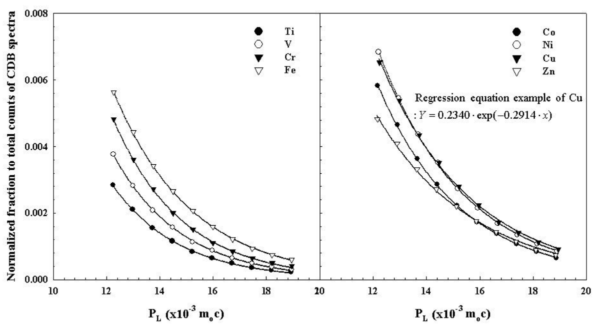

Figure 4. The calculated regression plot for the CDB spectra for 3-d transition metal.

The spectra observed in this region was fitted by using two parameters of an exponential regression equation as

$$Y = A \exp(-B \cdot X) \qquad (1)$$

The constant B can be regarded as the decay constant of the spectrum, which determines the curve shape For 3-d metals, there is a decreasing tendency of the constant B with an increasing number of the valence electrons as shown in Fig. 4.

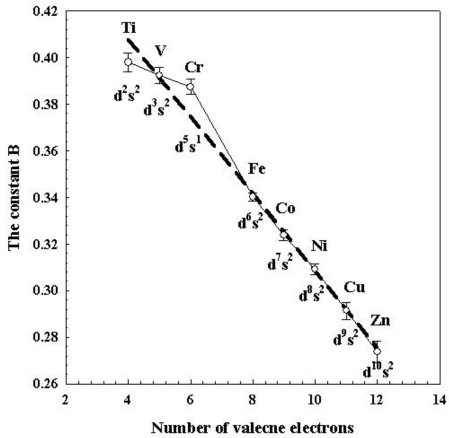

Figure 4. The correlation between the constant B and the number of valence electrons.

4. Conclusion

It is known that annihilations with valence electrons produce smaller Doppler shifts (20 x $10^{-3}m_0c$) than annihilations with core electrons [4]. Accordingly the high fraction of valence electron annihilation makes small decline shape of Doppler broadening spectra . We could find results that are consistent with these facts. Considering the relation of decay constant B and spectra shape that larger B makes sharp decline shape, the tendency of the constant B can be used to distinguish different elements in decay equation (1).

Therefore we verified that the feasibility of the specific momentum part of the Doppler-broadened annihilation as elemental evaluation tools.

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