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Coincidence Doppler Broadening Study of Elemental Metals

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Twenty pure elemental metal samples have been studied with a coincidence Doppler broadening system (CDB). The results show the relationship between the CDB spectra and the electronic structure of these samples. The experimental results are compared to simple theoretical predictions, which show that the high-momentum part of the Doppler-broadening spectra can be used to distinguish different elements.

Key words: Positron annihilation, Coincidence Doppler broadening, Metal, Elemental specificity**I. INTRODUCTION**

Positron annihilation spectroscopy (PAS) is a powerful tool for studying defects in solids. It has been widely used because of its non-destructive and highly sensitive characteristics [1,2]. The most frequently used techniques to detect the properties of open volume defects and their interaction with the medium are the lifetime, the Doppler broadening (DB) techniques, angular correlation of positron annihilation radiation (ACAR) and so on. A positron lifetime spectrum reveals electron density at the annihilation site. DB and ACAR are sensitive methods used to determine the density distribution of electrons in momentum space. DB is always distorted by background events, which come from Compton scattering, incomplete charge collection, pulse pile-up and environment radiation. The ratio of peak to background (PB) in these experiments is poor, about 10^3 . In 1977, Lynn *et al.* developed a coincident set-up to detect both of the annihilation photons simultaneously. This increased the ratio of PB to an order of 10^5 and improved the relative energy resolution by a factor of $\sqrt{2}$ [3]. This method is the so-called coincidence Doppler broadening (CDB), and it has been used in many fields recently [4,5]. With a more detailed analysis of the CDB spectrum one can glean more information about the defect site. The wave function of a trapped positron overlaps with the atoms surrounding the defect and these atoms can be identified by analyzing the high momentum part of the annihilation spectrum.

The possibility of identifying not only the type of open volume defect, but also the atoms that surround the defect, makes the positron probe even more powerful. In order to identify atoms that surround an open volume defect, we need accurate information about the core annihilation signal from these atoms. Some prior

research was done on pure elements but the picture is far from complete. Pressing ahead, we measured 20 pure elements using CDB methods and discussed the lineshape in detail.

II. EXPERIMENTAL

The twenty pure elements we investigated, were purchased from Alfa-Aesar. All samples were cut into 10 mm×10 mm sections by electroetching. Further details of the samples are provided in Table I.

TABLE I Details of the samples

Sample	Purity/%	Thickness/mm
Mg	99.90	1.0
Ti	99.70	0.89
Cr	100.00	2–3
Co	99.85	1.0
Cu	99.99	1.0
Zr	99.20	1.0
Mo	99.95	1.0
In	100.00	1.0
Hf	99.50	0.75
W	99.95	1.0
Al	99.99	1.0
V	99.50	1.0
Fe	99.99	0.5
Ni	99.50	0.787
Zn	100.00	1.0
Nb	99.80	1.0
Ag	99.90	1.0
Sn	99.80	1.0
Ta	99.95	1.0
Pb	100.00	0.5

The CDB experiments were performed at room temperature. For each sample, about 10 million counts were

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accumulated in the spectrum, with a counting rate of 80 cps. The details of the CDB measurement system were previously documented in our early works [6].

The experimental results are grouped according to their position in the Periodic Table. The ratio curves, obtained by dividing the measured curves by the curve of a reference element, of each group are compared with the calculated ones. The calculation program was developed by Helsinki University of Technology [7,8].

The momentum distributions are calculated separately for each electron state described as ψ_j according to the following equation

$$\rho_j(P) = \pi r_e^2 c \mu_j^2(0) \left| \int dr e^{-iP \cdot r} \psi_+(r) \psi_j(r) \right|^2 \quad (1)$$

where r_e is the classical electron radius, ψ_+ is the positron wave function. The term $\mu_j^2(0)$ is called the state-dependent enhancement factor and it is a constant related to the annihilation rate of the state j as

$$\mu_j^2 = \frac{\lambda_j}{\lambda_j^{\text{IPM}}} \quad (2)$$

In the above, the annihilation rate λ_j is calculated by LDA (Local-Density Approximation) or GGA (Generalized-Gradient Approximation) as

$$\lambda_j = \pi r_0^2 c \int dr \gamma^{\text{LDA,GGA}}(r) |\psi_+(r)|^2 |\psi_j(r)|^2 \quad (3)$$

where γ is the corresponding enhancement factor, and γ^{IPM} is the annihilation rate within the independent particle model. The final momentum distribution is then obtained by summing ρ_j , as in Eq.(1), over all electron states.

A parametrized positron wave function is used in the calculation of momentum distributions,

$$\begin{aligned} |\psi_+\rangle &\approx a_0 + a_1 \left[\text{erf}\left(\frac{r}{a_2}\right) \right]^{a_3} \\ &= a_0 + a_1 \left[\frac{1}{\sqrt{2\pi}} \int_{-\infty}^r e^{-\left(\frac{r}{\sqrt{2}a_2}\right)^2} dr \right]^{a_3} \end{aligned} \quad (4)$$

where r is the distance from the center of the atomic sphere and a_0 , a_1 , a_2 , and a_3 are fitted parameters.

III. RESULTS AND DISCUSSION

The groups, arranged according to Period number, are as follows: Mg, Al(Fig.1 (a) and (b)), Ti, V, Cr, Fe, Co, Ni, Cu, Zn (Fig.1 (c) and (d)), Zr, Nb, Mo, Ag, In, Sn (Fig.1 (e) and (f)), Hf, Ta, W, Pb (Fig.1 (g) and (h)). The left parts of Fig.1 are the calculated ratio curves and the right parts are the experiments ones. The calculations and experimental results are qualitatively consistent in all the groups.

The Mg and Al curves in Fig.1 (a) and (b) show a single peak around $8 \times 10^{-3} m_0 c$. In this area, the lineshape of Doppler broadening depends mainly on the contributions of the positron annihilation with the electrons of 3p shell. The peak in the ratio curve can be correlated to the 3p electrons.

In Fig.1 (c) and (d), the ratio curves for the transition metals of the fourth row in the Periodic Table are reported. The main difference in the elements of this group is the number of 3d electrons. The Ti curve was selected as the reference curve in this group. The elements of the series are characterized by a peak between $10 \times 10^{-3} m_0 c$ and $35 \times 10^{-3} m_0 c$, with increasing intensity and width moving from V to Cu. Prior calculations have shown that in this momentum range the dominant contribution is due to the positron annihilation with 3d electrons [9]. Extrapolating these results we can infer that the increase in the peak intensity can be correlated with the increase of the annihilation of positrons with 3d electrons: V, Cr, Fe, Co, Ni, Cu have 3, 5, 6, 7, 8, 10 electrons in the 3d orbital respectively. The Zn curve does not follow the trend observed in the ratio curves from V to Cu: it is very broad and lower than the Cu curve. As we know, Zn has two electrons in the 4s orbital, while Cu has only one. The calculation indicates that the annihilation possibility of positrons with 4s electrons are 32% and 61% in Cu and Zn respectively. The increase in the annihilation of positrons with 4s electrons decreases the intensity of peak in the ratio curve.

The ratio curves for the elements in the fifth row in the Periodic Table in Fig.1 (e) and (f) are more complicated. The elements of this series: Zr, Nb, Mo, Ag, In, Sn have different 5s and 5p electrons except for the 4d electrons. As we can see, in the low-momentum range ($< 5 \times 10^{-3} m_0 c$) In and Sn curves are higher than others, this is due to the increase of 5p electrons. Nb, Mo, Ag have 4, 5, 10 electrons in the 4d orbital respectively, and the intensity and width of the peak increase with the increase of the 4d electrons.

The characteristic of the curves for Hf, Ta, W, Pb can be explained in the same way. There are many differences between the calculated and measured results for this series, as we can see from Fig.1 (g) and (h). For the high- z elements, our proposed model seems too simple.

In Fig.2, we grouped the samples by the same column in the Periodic Table. Al was selected as the reference sample. We found that all the groups had the same trend. In Fig.2 (a), there is a peak near $10 \times 10^{-3} m_0 c$ in Ti, Zr, Hf curves respectively. These peaks can be correlated with the annihilation of positrons by d electrons. As for the Hf curve, there is broad peak above $25 \times 10^{-3} m_0 c$, this is due to the annihilation of positrons with 4f electrons.

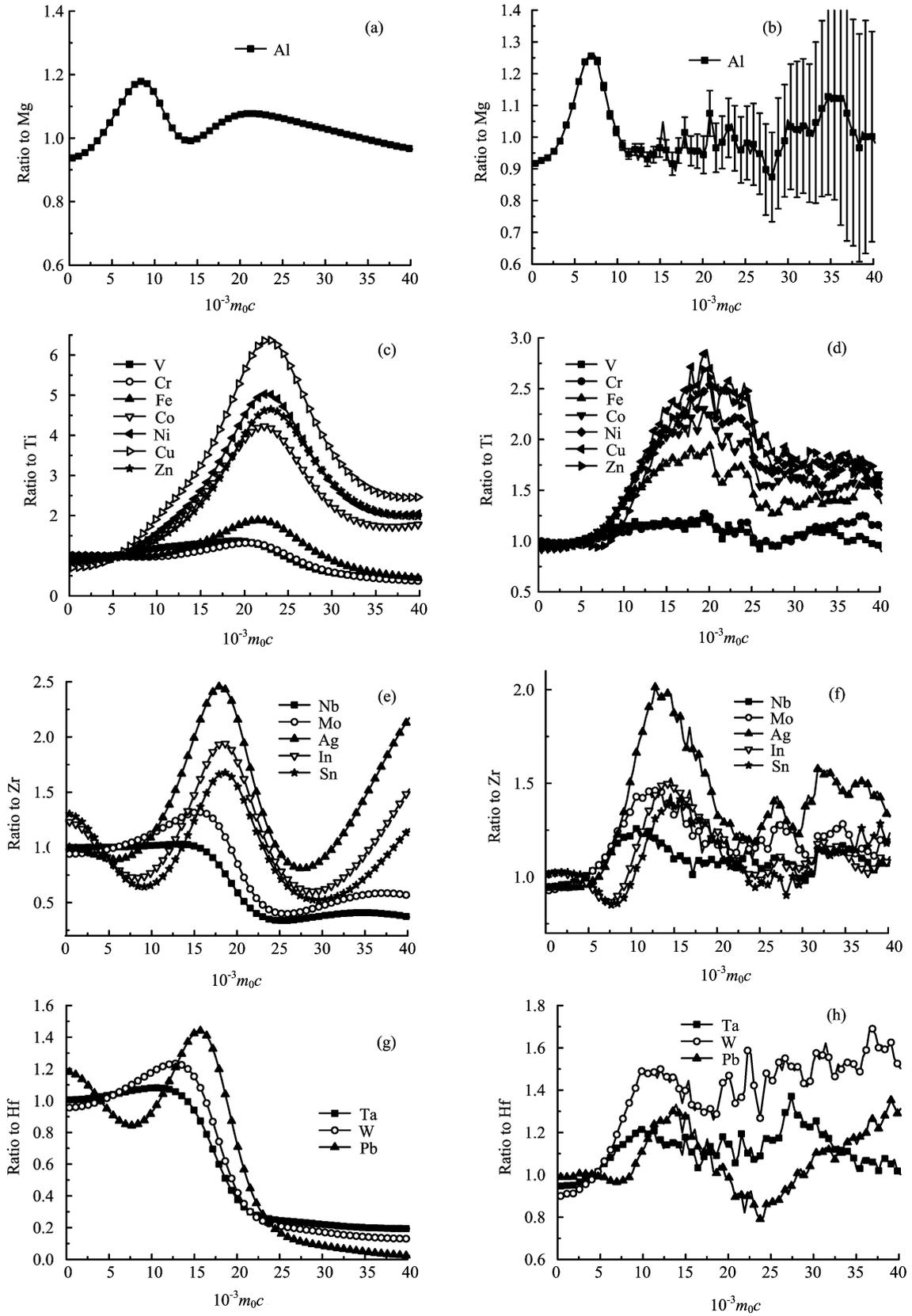


FIG. 1 Ratio curves of the groups according to the Periodic number. (a), (c), (e), (g) are calculated results; (b), (d), (f), (h) are experimental results.

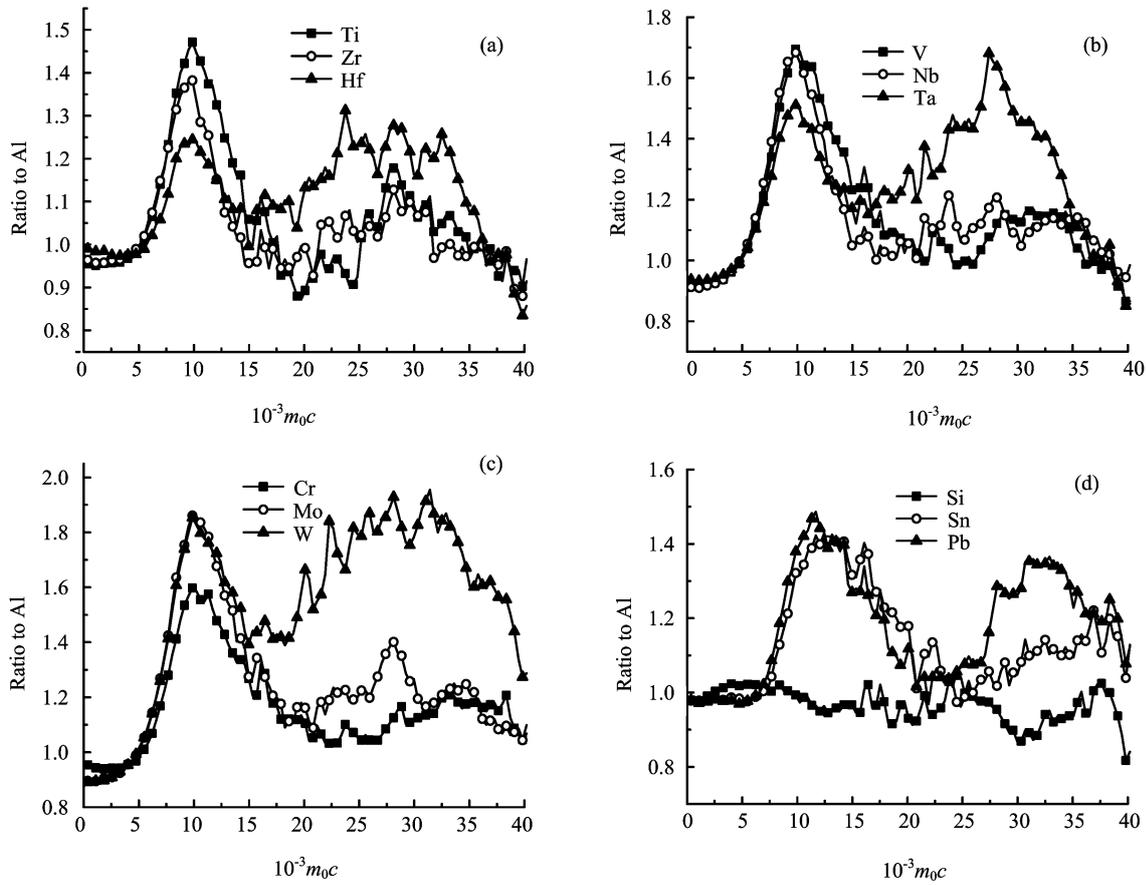


FIG. 2 Ratio curves of the groups according to the Periodic column.

IV. CONCLUSIONS

Herein we have reported the CDB profiles of twenty types of pure elemental metals. The results show that each element is characterized by features seen in the ratio curves: one or two peaks with different intensities and positions. These features can be explained by the electron configuration of each element.

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- [2] P. J. Schultz and K. G. Lynn, *Rev. Mod. Phys.* **60**, 701 (1988).
- [3] K. G. Lynn, *et al.* *Phys. Rev. Lett.* **38**, 241 (1977).
- [4] P. A. Kumar, *et al.* *Phys. Rev. Lett.* **77**, 2097 (1996).
- [5] R. S. Brusa, *et al.* *Nucl. Instrum. Method. B* **194**, 519 (2002).
- [6] W. Kong, *et al.* *Nucl. Instrum. Method. B* **225**, 623 (2004).
- [7] M. J. Puska, *J. Phys. C: Condens. Matter.* **1**, 6081 (1989).
- [8] Laboratory of Physics, *A Program to Model Positron States and Annihilation in Solids*, Helsinki University of Technology: Doppler, (2003).
- [9] V. J. Ghosh, *et al.* *Phys. Rev. B* **61**, 10092 (2000).

[1] M. J. Puska and R. M. Nieminen, *Rev. Mod. Phys.* **66**, 841 (1994).