The Compton profiles of tantalum

1993 J. Phys.: Condens. Matter 5 5371

(http://iopscience.iop.org/0953-8984/5/30/016)

J. Phys.: Condens. Matter 5 (1993) 5371–5376. Printed in the UK

The Compton profiles of tantalum

Chu-Nan Chang, Yu-Mei Shu, Chuhn-Chuh Chen, and Huey-Fen Liu
Department of Physics, National Taiwan Normal University, Taipei, Taiwan

Received 8 March 1993, in final form 19 April 1993

Abstract. The isotropic Compton profiles of tantalum were measured by a 59.54 keV γ-ray Compton profile spectrometer. Comparisons with the renormalized free-atom model and band structure calculations have been made. The valence electron configuration is found close to $5d^46s^1$, and the band structure calculations give a sharper Compton profile. The Compton profiles of V, Nb and Ta normalized to their respective Fermi momentum have also been compared and discussed. The d electrons are seen to play a more significant role than the orthogonality effect, as observed in the IVA group elements.

1. Introduction

In recent years γ-ray Compton backscattering experiments in solids have become highly accurate, thus providing reliable results about the momentum distribution of the electronic ground state. In the impulse approximation [1, 2], the Compton profile (CP) is the projection of the electronic ground-state momentum density on the scattering vector. This technique has been used to investigate the electronic structure of many transition metals [3] which are important in alloys, oxides and other compounds due to the peculiar property of the d electrons.

The Compton profiles (CP) of VB metals—vanadium and niobium—have been examined experimentally and theoretically by many authors. Theoretically the Compton profiles for vanadium have been calculated through the band structure calculation by Wakoh and co-workers using KKR and the augmented plane wave (APW) method [4, 5] and by Laurent and co-workers using the linear combination of the atomic orbital method [6]. For niobium, Wakoh and co-workers obtained CP by the APW method [7]. In [5–7], the effective potential used is a modified $X\alpha$ potential. Papanicolaou and co-workers used a self-consistent semi-relativistic (neglecting the spin–orbital interaction) APW method in the local density approximation of Hedin and Lundqvist to obtain the CP of vanadium and niobium [8], tantalum and tungsten [9]. All these theoretical values have been compared to the experimental data on vanadium [10] and niobium [7, 11, 12], except tantalum. All of them show that the theoretical values are larger than the experimental values for small momentum, q, and smaller for large momentum. The discrepancies were partly attributed to the electron correlation correction, which transfers electrons to larger q, and to some non-local exchange-correlation effects, which may broaden the CP, by Papanicolaou and co-workers [8]. Mittal and co-workers [13] expressed their concern about the spin–orbital interaction effect which were neglected in the APW calculation by Papanicolaou and co-workers [8, 9].

In this paper, we report our isotropic experimental CP data for tantalum by using 59.54 keV γ-rays. The data will be discussed on the basis of the renormalized free-atom

0953-8984/93/305371+06$07.50

model [14]. Discussions will be also made for the CP of V, Nb and Ta on the normalized momentum scale P/P_F, where P_F is the Fermi momentum. The discrepancies between the experiment and the band structure theory for V, Nb and Ta will be compared and discussed.

2. The experiment

An annular source of Am^{241} about 1 Ci was used for 59.54 keV γ-ray scattering measurements. Both the target and the radioactive source were housed in a vacuum chamber of 10^{-6} torr. The backscattering γ-rays passing through a beryllium window holding the vacuum were detected by a HPGe detector. The overall momentum resolution of this set-up was 0.50 au.

The target was a 0.2mm, 99.9% pure tantalum foil (polycrystalline, Marz grade). The Compton scattering spectrum and the spectral line of the 5.9 keV x-ray of Fe^{55} source was automatically recorded every 30 minutes by a PC-controlled MCA. The latter was used as a system stability check. The drift of the Compton peak was estimated to be less than 0.5 per channel. In this experiment, each channel was equal to 32 eV.

The Compton scattering spectrum with peak counts greater than 10^4 was measured. The tails of the tantalum K x-rays extended to the Compton spectrum were corrected by using a pure tantalum K x-rays spectrum generated by 320 keV γ-rays (from a 50μCi Cr^{51} source irradiation). Similarly the Ge K escape peak buried under the Compton peak was also corrected by using a 10μCi Am^{241} spectrum. Both of these corrections were less than 0.1%.

The total transfer energy corresponding to the Compton peak (48.3 keV) was 11.2 keV, but the binding energy of K-shell electrons, L_I-shell electrons, and L_{II}-shell electrons was respectively 57.41 keV, 11.68 keV and 11.13 keV. Thus these electrons were excluded from the analysis of the upper half of the Compton spectrum. However, the binding energy of L_{III}-shell electrons was 9.88 keV, the corresponding position of the Compton spectrum as shown in figure 1 is 49.66 keV, about 2.08 au in the momentum space. In this case we included the L_{III}-shell electrons in our data analysis only in the region of $0 \sim 2.0$ au.

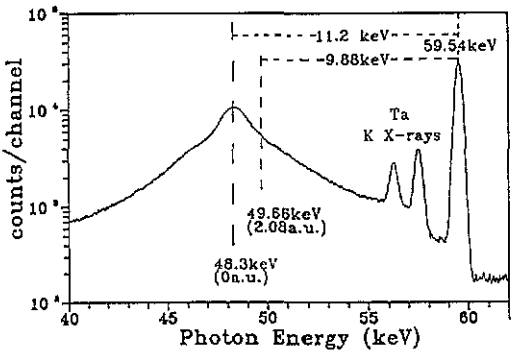

Figure 1. The Compton backscattering spectrum of Ta by using 59.54 keV γ-rays.

A standard Fourier transformation method was used to deconvolute the Compton spectrum. A soft-cut method similar to that used in [15] was used to eliminate the noise amplification caused by the low counting statistics of the high-energy portion of the Compton spectrum. The residual instrumentation function (RIF) so obtained was used to convolute the theoretical values when comparisons between theory and experiment were made. The

Monte Carlo method of Felsteiner and co-workers [16] was used to do the multiscattering correction. However, the intensity of the second scattering was found to be less than 0.2% of the first scattering, which were quite small relative to the counting statistic errors.

Table 1. The Compton profile of Ta valence electrons. The theoretical values of MT–APW and RFA have been convoluted with the RIF. The Compton profile of Ta core electrons (without RIF convolution) is also shown. The normalization area in $0 \sim 6.0$ au is 24.361. The errors shown at some momenta are statistical errors.

P_z (au)	Exp.	MT–APW (Papanicolaou)	RFA ($5d^46s^1$)	Core	RIF
0.0	2.42 ± 0.09	2.55	2.43	6.93	1.3725
0.1	2.41	2.53	2.39	6.87	1.3179
0.2	2.34	2.47	2.31	6.83	1.0636
0.3	2.24	2.38	2.17	6.82	0.6887
0.4	2.10	2.26	2.00	6.75	0.3016
0.5	1.93	2.10	1.82	6.65	0.0012
0.6	1.75	1.91	1.63	6.53	0.1682
0.7	1.58	1.71	1.45	6.39	0.2015
0.8	1.40	1.49	1.28	6.22	0.1440
1.0	1.08 ± 0.08	1.07	1.01	5.85	0.0225
1.2	0.81	0.70	0.80	5.46	0.0589
1.4	0.61	0.44	0.60	5.08	0.0058
1.6	0.46	0.27	0.43	4.74	0.0207
1.8	0.36	0.18	0.30	4.47	0.0070
2.0	0.28 ± 0.06	0.12	0.20	4.23	0.0061
3.0	0.01 ± 0.05	0.04	0.04	3.49	0.0007
4.0	0.03	0.011	0.021	2.83	
5.0	0.02	0.006	0.017	2.19	
6.0	0.02 ± 0.03	0.005	0.012	1.69	

The Compton profiles of tantalum are listed in table 1. Note that a smooth procedure has been used to interpolate the experimental data at the appropriate values of P_z. The theoretical values of [9] and the renormalized free-atom (RFA) model calculation with the electron configuration of $5d^46s^1$ are shown for comparison. The theoretical values have been convoluted by the residual instrumentation function (RIF) which are also listed in the table for the 59.54 keV Compton profile spectrometer.

3. Discussions

As shown in figure 2, the RFA model calculation with an electron configuration of $5d^44s^1$ is the most suitable one to describe the tantalum metal. In the RFA model calculation, as people usually do for other transition metals, only the 6s atomic electron wavefunction is renormalized to the Wigner–Seitz sphere. The other electrons are treated as those in the free atom and their Compton profiles can be obtained directly from the calculation in [17]. The electron configuration of tantalum metal is thus similar to those found in vanadium ($3d^44s^1$) [14] and niobium ($4d^{4.4}5s^{0.6}$) [11]. These configurations are very close to those of band structure calculations of [18], where it was shown that $3d^{3.98}4s^{0.61}3p^{0.41}$ for vanadium, $4d^{4.1}5s^{0.64}5p^{0.25}$ for niobium and $5d^{3.78}6s^{0.85}6p^{0.36}$ for tantalum.

Vanadium, niobium and tantalum are in the VB group and all have a BCC lattice structure with lattice constants of 5.714 53 au, 6.236 10 au and 6.247 06 au respectively.

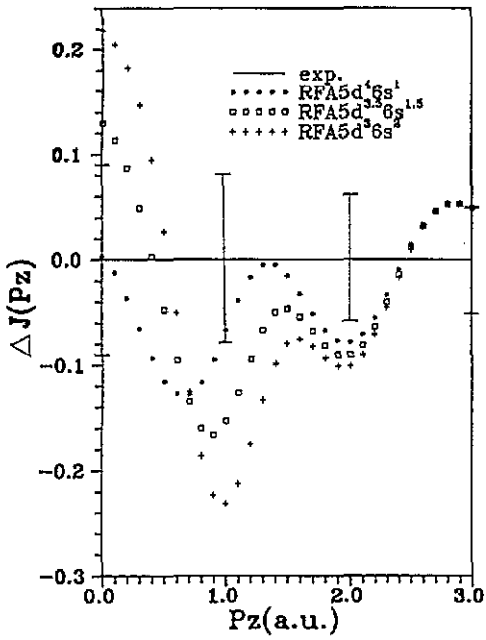

Figure 2. The difference of Ta Compton profiles between experiment J_{exp} and the RFA model J_{th} with various electron configurations, $\Delta J = J_{th} - J_{exp}$. Statistical error bars are shown for some data points.

It is interesting to see their Compton profiles normalized to their Fermi momenta, which are 1.166 36 au, 1.068 81 au and 1.067 02 au for V, Nb and Ta respectively. Figure 3 shows the normalized Compton profiles of the valence electrons for V, Nb and Ta. Here the data on V and Ta are our experimental data. The data on V have been demonstrated [19] to agree very well with those of [10]. The data on Nb are taken from [11]. The trend of these normalized valence electron Compton profiles is different from those of IVA group: diamond, silicon and germanium [20]. They are attributed to the effect of the orthogonality of the valence electrons to the core electrons, and therefore the normalized valence electron Compton profile decreases at low momentum from diamond to germanium as the orthogonality increases. This is not the case, as seen in figure 3. We suggest that the d electrons may play a significant role, i.e. the localization of the d electrons decreases according to the sequence of 3d, 4d and 5d. This effect may outweigh the orthogonality effect when the atomic number increases. This localization property can readily be seen from the RFA calculation, where the wavefunction of d electrons in a free-atom state is used. Since 86.3% of the 3d wavefunction in the vanadium atom, 81.3% of the 4d wavefunction in the niobium atom, and 78.1% of the 5d wavefunction in the tantalum atom are contained in the Wigner–Seitz sphere, the relatively good agreements between the experiments and the RFA calculations indicate that the RFA treats the 3d electrons in V as more localized than the 4d electrons in Nb and even more localized than the 5d electrons in Ta. Increased localization causes a broader Compton profile. This trend is shown in figure 3.

We have examined the three theoretical approaches of a band structure calculation for the Compton profile of V, namely the APW method of [4, 5], the LCAO method of [6, 21] and the muffin-tin APW method (MT–APW) of [8]. They are close to each other, but all overestimated the CP of V at low momentum when compared with experiment, with the APW of [4, 5] agreeing with the experiment slightly better than the other two methods. This overestimation by the band structure theory was also seen by Wakoh and co-workers for Nb [7]. The electron correlation correction has been considered by many authors [10, 21–25]

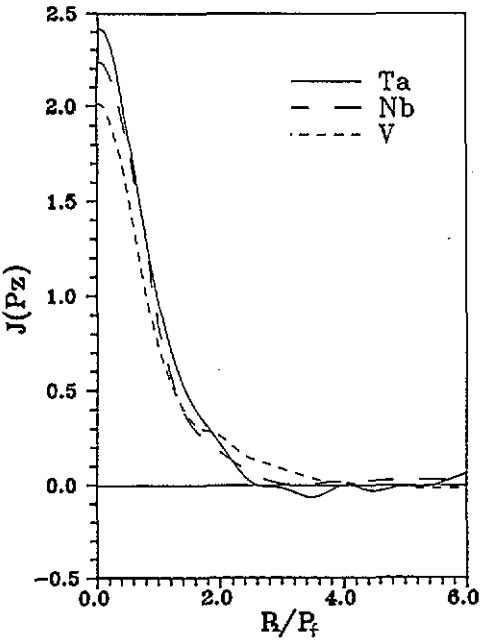

Figure 3. The experimental Compton profiles of V, Nb and Ta normalized to their respective Fermi momenta $P_F = 1.16636$ au, 1.06881 au and 1.06702 au.

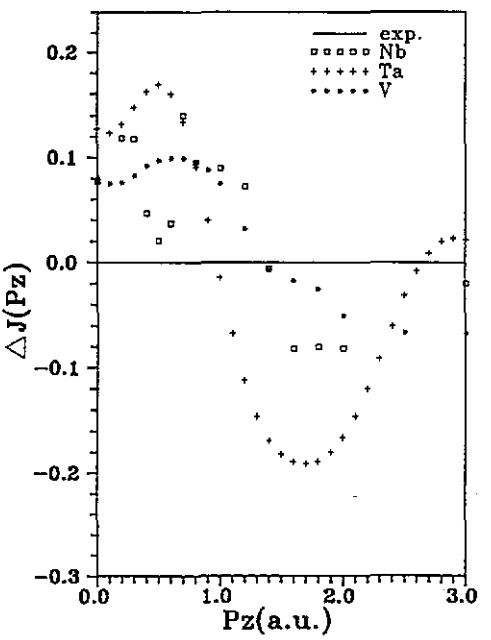

Figure 4. The difference of Compton profiles between experiment J_{exp} and theory (MT–APW) J_{th}, $\Delta J = J_{th} - J_{exp}$ for V, Nb and Ta. The experimental values of V and Ta are our data while those of Nb are taken from [11].

to bring the theory closer to the experiment.

The MT–APW method was used to derive all the Compton profiles of V, Nb [8] and Ta [9]. We are interested in seeing their comparison with the experiment. Here, only isotropic Compton profiles will be compared due to the lack of anisotropic CP data for Ta. Therefore, all theoretical anisotropic values have been averaged to the isotropic values by Houston's method as suggested in [26]. The results are shown in figure 4. The data on V and Ta are from our work, while the data of Nb is taken from [11].

A gradual increase of the discrepancy at low momentum between the theory and the experiment is seen in figure 4 for V, Nb and Ta. This feature may be discussed in terms of the electron correlation effect, the non-local exchange-correction effect and the spin–orbital coupling effect. All of them were neglected in the MT–APW calculation of Papanicolaou and co-workers.

The electron correlation correction in the scope of the Lam–Platzman theory [27] has been calculated for V and Nb within the local density approximation and indeed increases from V to Nb [8]. The non-local exchange-correlation effects, which are not included in the local density approximation calculation of [8, 9], may also play a role since the delocalization of the d electrons, as previously mentioned, increases from V to Ta. As for the spin–orbital interaction, Bacalis and co-workers [28] have performed the investigation through the Hamiltonian of $H = \xi l \cdot s$. The coupling constant ξ was found, as expected, to increase with the mass of the element and so did the average splitting of the d state. A larger correction of the spin–orbital interaction for CP may appear for the larger mass of the element. We feel that all these three effects, the electron correlation correction, the non-

local exchange effect and the spin–orbital interaction correction, are necessary to correct the band structure theory to bring it closer to experiment.

4. Summary

We presented in this paper the isotropic experimental Compton profile of Ta and discussed it on the basis of the renormalized free-atom model. The electron configuration $5d^4 6s^1$ is found to be appropriate for Ta. The extension of the wavefunction of the d electrons is shown to increase with the mass of the elements from V, Nb and Ta when the momentum-normalized CP of V, Nb and Ta are compared. The electron correlation correction, the non-local exchange effect, and the spin–orbital interaction correction appear to increase with the mass of the elements and are necessary to bring the band structure theory closer to experiment for V, Nb and Ta.

Acknowledgments

This work is partly supported by the National Science Council of the Republic of China, under the project no. NSC 81-0208-M003-018.

References

[1] Eisenberger P and Reed W A 1974 *Phys. Rev.* B **9** 3237
[2] Ribberfors R 1975 *Phys. Rev.* B **12** 2067
[3] Chang C N, Lee S B and Chen C C 1991 *J. Phys. Soc. Japan* **60** 4253
Chang C N, Chen C C and Liu H F 1992 *J. Phys.: Condens. Matter* **4** 10445
[4] Wakoh S and Yamashita J 1973 *J. Phys. Soc. Japan* **35** 1406
[5] Wakoh S, Kubo Y and Yamashita J 1976 *J. Phys. Soc. Japan* **40** 1043
Wakoh S, Kubo Y and Yamashita J 1975 *J. Phys. Soc. Japan* **38** 416
[6] Laurent D G, Wang C S and Callaway J 1978 *Phys. Rev.* B **17** 455
[7] Wakoh S, Fukamachi T, Hosoya S and Yamashita J 1975 *J. Phys. Soc. Japan* **38** 1601
[8] Papanicolaou N I, Bacalis N C and Papaconstantopoulos D A 1987 *Z. Phys.* B **65** 453
[9] Papanicolaou N I, Bacalis N C and Papaconstantopoulos D A 1986 *Phys. Stat. Sol.(b)* **137** 597
[10] Rollason A J, Holt R S and Cooper M J 1983 *Phil. Mag.* B **47** 51
[11] Tomak M, Singh Hanuman, Sharma B K and Manninen S 1985 *Phys. Stat. Sol.(b)* **127** 221
[12] Pattison P, Cooper M, Holt R, Schneider J R and Stump N 1977 *Z. Phys.* B **27** 205
[13] Mittal U, Sharma B K, Mohammad F M and Ahuja B L 1988 *Phys. Rev.* B **38** 12208
[14] Berggren K-F 1972 *Phys. Rev.* B **6** 2156
[15] Paatero P, Manninen S and Paakkari T 1974 *Phil. Mag.* **30** 1281
[16] Felsteiner J, Pattison P and Cooper M 1974 *Phil. Mag.* **30** 537
[17] Biggs F, Mendelson L B and Mann J B *At. Data Nucl. Data Tables* **16** 201
[18] Papaconstantopoulos D A 1986 *Handbook of the Band Structure of Elemental Solids* (New York: Plenum)
[19] Chang C N, Chen C C and Liu H F 1992 *J. Phys.: Condens. Matter* **4** 10445
[20] Reed W A and Eisenberger P 1971 *Phys. Rev.* B **6** 4596
[21] Cardwell D A and Cooper M J 1989 *J. Phys.: Condens. Matter* **1** 541
[22] Bauer G E W and Schneider R 1983 *Z. Phys.* B **54** 17
[23] Bauer G E W and Schneider R 1985 *Phys. Rev.* B **19** 6038
[24] Rollason A J, Schneider J, Laundy D, Holt R S and Cooper M J 1987 *J. Phys. F: Met. Phys.* **17** 1105
[25] Sundararajan V, Kanhere D G and Singru R M 1992 *Phys. Rev.* B **46** 7857
[26] Betts D D, Bhatia A B and Wyman Max 1956 *Phys. Rev.* **104** 37
[27] Lam L and Platzman P 1974 *Phys. Rev.* B **9** 5122
[28] Bacalis N C, Blathras K, Thomaides P and Papaconstantopoulos D A 1985 *Phys. Rev.* B **32** 4849