CHINESE JOURNAL OF PHYSICS VOL. 33, NO. 2 APRIL 1995

The Directional Compton Profiles of V and Ta

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(Received December 5, 1994; revised manuscript received January 5, 1995)

The directional Compton profiles of single crystals of vanadium and tantalum symmetrical to their [111],[110], and [100] directions were measured with a Compton profile spectrometer with an annular source and y-rays at 59.54 keV. The main features of the observed profiles of vanadium and tantalum agree with the predictions of the band structure theory and are interpreted in terms of the geometry of the Fermi surfaces. The theory overestimated the Compton profiles of both V and Ta.

PACS. 71.25.Pi– Band structure of crystalline metals.
PACS. 78.70.Ck– X-ray scattering.

I. INTRODUCTION

The directional Compton profile along the $\hat{e}$ direction of a crystal is expressed, according to the impulse approximation [1], as

$$J(P_z,\hat{e}) = \sum_P N(\vec{P})\delta(\vec{P}\cdot\hat{e} - P_z), \tag{1.1}$$

in which N(P) is the momentum distribution of the electronic ground state

$$N(P) = \sum_{n,occupied} \sum_k^{BZ} \sum_G \delta_{\vec{P},\vec{k}+\vec{G}} \chi_n(\vec{k},\vec{P})^2/N, \tag{1.2}$$

where n is the band index, $\chi_n(\vec{k},\vec{P})$ the Fourier transform of wave functions $\Psi_n(\vec{k},\vec{r})$, $\vec{k}$ the wave vector, G the reciprocal lattice vector, N a normalization factor and BZ the Brillouin zone. This profile is a measure of the momentum distribution of the electronic ground state projected on a certain crystal direction $\hat{e}$. Experimentally, the Compton profile $J(P_z,\hat{e})$ is measured through the double-differential Compton scattering cross sections [2]

$$\frac{d^2\sigma}{d\Omega d\omega_2} = C(\omega_1,\omega_2,\theta,P_z)J(P_z), \tag{1.3}$$

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in which ω_1 and ω_2 are respectively the frequencies of the incident and scattered photons, θ is the scattering angle and P_z is the electron momentum projected along the scattering vector that can be aligned with the crystal direction $\hat{e}$. P_z is expressed as

$$P_z = -\frac{E_1(\omega_1 - \omega_2) - h\omega_1\omega_2(1 - \cos\theta)}{C[\omega_1^2 \text{ t } \omega_2^2 - 2\omega_1\omega_2\cos\theta]^{1/2}}, \tag{1.4}$$

where E_1 is the electron energy that is approximated as the rest mass energy of the electron, m_0c^2, without significant error. The factor $C(\omega_1, \omega_2, \theta, P_z)$ presented in Ref. [2] is

$$C(\omega_1, \omega_2, P_z, \theta) = \left[\frac{e^2}{4\pi^2 m_0^2 c^2}\right]^2 \left[\frac{\omega_1(1 + P_z/m_0c)}{\omega_2(1 - P_z/m_0c)} + \frac{\omega_2(1 - P_z/m_0c)}{\omega_1(1 + P_z/m_0c)}\right] \times \frac{\omega_2 m_0 c}{2\omega_1[(\omega_1^2 + \omega_2^2 - 2\omega_1\omega_2\cos\theta)^{1/2} + (P_z/m_0c)(\omega_1 - \omega_2)]}. \tag{1.5}$$

The Compton profile spectrometer with 7 rays from an annular radioactive source is generally used to measure the isotropic Compton profile. Because of the symmetrical character of the Fermi surface that was related strongly to features of directional Compton profiles by Papanicolaou et al. [3], we used our spectrometer with 7 rays at 59.54 keV to measure the directional Compton profiles of single crystals of vanadium and tantalum symmetrical to their crystal directions [111],[110], and [100].

The Compton profiles of vanadium, niobium and tantalum were examined both experimentally and theoretically by many authors. The Compton profiles of vanadium were calculated through the band structure by Wakoh et al. using Korringa, Kohn, and Rostoker method and the augmented plane wave method [4,5], and by Laurent *et al.* using a linear combination of atomic orbitals [6]. For niobium, Wakoh *et al.* also used the augmented plane wave method [7]. In Refs. [5-7], the effective potential used is a modified X_α potential. Papanicolaou *et al.* used a self-consistent semi-relativistic (neglecting spin-orbital interaction) method with augmented plane waves in the local density approximation of Hedin and Lundgvist to derive the Compton profiles of vanadium and niobium [8], and tantalum and tungsten [3]. All these theoretical values are compared to experimental data on vanadium [9], niobium [7,10,11], and tantalum [12]. The former two have directional Compton profiles and the latter has an isotropic one. All show theoretical values larger than experimental values for small momentum, P_z, and smaller for large momentum. The discrepancies are partly attributed to a correction for electron correlation, which transfers electrons to larger P_z, and to non-local exchange-correlation effects, which may broaden the profile, by Papanicolaou et al. [8]. Mittal *et al.* [13] expressed their concern about neglect of spin-orbital interaction in the calculations by Papanicolaou *et al.* [3,8]. In our previous work on the isotropic Compton profiles of V, Nb and Ta [12], we concluded that the correction for electron correlation, the non-local exchange-correlation effects, and the spin-orbital interaction appear to increase with the mass of the elements and are necessary

to bring the theoretical results from band structure closer to experimental data of V, Nb and Ta.

In order to gain insight into this matter, experimental data for the directional Compton profile of Ta seem essential. We thus made these measurements on V and Ta with our Compton profile spectrometer and γ rays at 59.54 keV. The measurement of V enables verification of the method because directional Compton profiles of V were measured at high resolution with a synchrotron radiation source at 60 keV in combination with a single-crystal spectrometer [4].

II. EXPERIMENTS

The detailed experimental arrangement and procedures of data reduction resembled those in our previous work to measure isotropic Compton profiles [12,15]. Here we summarize important features.

An annular source of Am^{241} about 1 Ci provided γ rays at 59.54 keV for scattering measurements. Both the target and the radioactive source were housed in a vacuum chamber at 10^{-4} Pa. The backscattering γ rays passing a beryllium window to maintain the vacuum were detected with a HPGe detector. The overall momentum resolution was 0.50 atomic units.

Targets of both vanadium and tantalum were single crystals with (100), (110) and (111) orientation and 99.99% purity. They are disks of diameter 12 mm and thickness 5 mm for vanadium, and diameter 12 mm and thickness 2 mm for tantalum. The crystal plane of the target was placed perpendicular to the direction of y-ray scattering, shown in Fig. 1. As the scattering angle of our spectrometer was 174° (Fig. 1), the scattering vector lay on a conical surface 3.3 ± 1.5" relative to the direction of the scattered γ rays. The overall angular uncertainty of the scattered γ rays was ±1.5°. Hence the directional Compton profiles were measured along the direction symmetrical to the crystal direction.

The Compton scattering spectrum and the X ray spectral line at 5.9 keV of an Fe^{55} source were automatically recorded every 30 min with a multichannel analyser controlled with a microcomputer. The X ray measurement served as a test of the stability of the system. The drift of the Compton maximum was estimated to be less than 0.5 channel. In this experiment, each channel was equal to 32 eV. The Compton scattering spectra were measured with peak counts greater than 4×10^4 for vanadium and 1 x 10" for tantalum.

A standard Fourier transformation was used to deconvolute the Compton spectrum. A soft-cut method similar to that used in Ref. [16] was used to eliminate noise amplification caused by the small counting statistics of the high-energy portion of the Compton spectrum. The residual instrumentation function so obtained was used to convolute the theoretical values when comparisons between theory and experiment were made. The directional

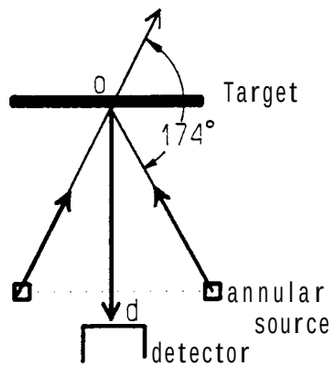

FIG. 1. Sketch of the experimental arrangement; the crystal surface orientations (111), (110) and (100) are perpendicular to the y-ray scattering direction od.

Compton profiles of vanadium and tantalum and their residual instrument functions are listed in Table I. The differences of the latter between vanadium and tantalum arise mainly from the counting statistics. The ratios of signal to noise were 650:1 for V and 77:1 for Ta. No correction for multiple scattering was made for these data as most effect of multiple scattering was eliminated when the differences between directional Compton profiles were made. The superscript "s" on Compton profile J in Table I denotes that these directional profiles are not exactly the directional Compton profiles at the respective crystal directions of [111], [110] and [100], but on directions 3.3 ± 1.5" symmetrical to them as described above. A smoothing procedure was used to interpolate experimental data at appropriate values of P_z.

III. DISCUSSION

III-l. Vanadium

Our directional Compton profiles of vanadium (Fig. 2) are somewhat sharper than those of Shiotani et al. [14] which were measured with synchrotron radiation at 60 keV and a single-crystal spectrometer, consistent with our data being directional profiles about 3.3 ±1.5° away from the respective crystal directions; the main structures of Compton profiles related to the geometry of Fermi surfaces as described elsewhere [3]. For both V and Ta, the jungle-gym tubes in the third Brillouin zone contribute most. According to Fig. 1 of Ref. [17], the jungle-gym tube of vanadium extends from the origin along

TABLE I. Directional Compton profiles 3.31 1.5° symmetrical to the [111], [110] and [100] crystal directions of V and Ta. The residual instrument function of V and Ta are also shown. The errors are statistical.

P_z(a.u.)	J^s_{111}	J^s_{110}	J^s_{100}	RIF	J^s_{111}	J^s_{111}	J^s_{100}	RIF
0		5.01 ± 0.02	5.08 ± 0.02		8.98 ± 0.09		8.81 ± 0.09	
0.1	5.12 5.11 ± 0.02	4.97	5.01	1.47 1.39	8.87	8.95 ± 0.09	8.73	1.17 1.14
0.3	5.04	4.89	4.91	0.60	8.72	8.67	8.61	0.98
0.4	4.93 4.79	4.78		0.17	8.51	8.47	8.45	0.74
	4.60		4.76		8.27			0.46
0.5	4.39	4.63	4.59	-0.11	7.98	8.23	8.25	0.22
0.6		4.43	4.38	-0.21		7.97	8.00	0.04
0.7	4.16	4.19	4.15	-0.16	7.68	7.68	7.72	-0.07
0.8	3.91	3.94	3.91	-0.07	7.36	7.38	7.41	-0.11
0.9	3.66	3.67	3.67	0.02	7.03	7.07	7.08	-0.10
1.0	3.41 ± 0.02	3.42 ± 0.02	3.43 ± 0.02	0.06	6.71 ± 0.08	6.76 ± 0.08	6.75 ± 0.08	-0.07
1.2	2.95	2.96	2.98	0.02	6.10	6.13	6.12	-0.01
	2.56	2.57		-0.02		5.56	5.57	0.02
1.6	2.23	2.24	2.28	-0.01	5.07	5.08	5.11	0.01
1.8	1.94	1.94	1.94	0.01	4.66	4.68	4.72	0.00
2.0	1.69 ± 0.01	1.70 ± 0.01	1.70 ± 0.01		4.33 ± 0.06	4.33 ± 0.06	4.36 ± 0.06	0.00
3.0				0.00		3.26 ± 0.05	3.27 ± 0.05	
4.0	0.97 ± 0.01	0.97 ± 0.01	0.97 ± 0.01	0.00 0.00	3.28 ± 0.05	2.66	2.68	0.00 0.00
	0.66	0.66	0.66		2.63			

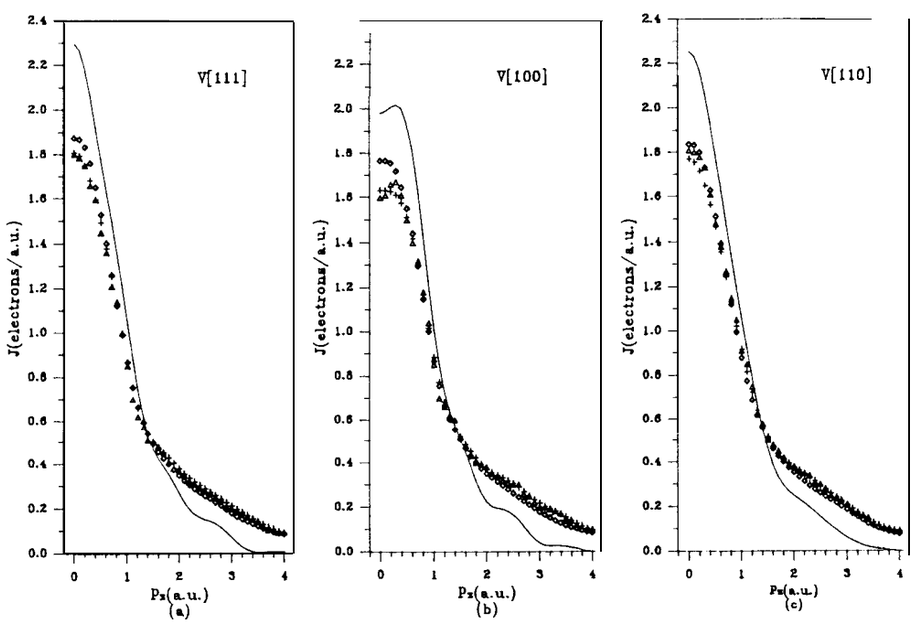

FIG. 2. Directional Compton profiles of V : present work (0), data of Shiotani *et* al. without (A) and with (+) our convolution, and theoretical results convoluted with residual instrument function (solid line).

$\Gamma \rightarrow H$, i.e. the [100] direction, and bears sixfold symmetry with respect to the origin. The effect of the jungle-gym tube decreases if the profiles are not exactly at the [100] direction. The actual differences may be larger than those shown in Fig. 2, because our profiles have poorer momentum resolution ($\simeq$ 0.50 a.u.) than those of Shiotani *et al.* ($\simeq$ 0.12 a.u.) [14]. We thus convoluted the Compton profiles of Shiotani *et al.* with a residual instrument function generated for momentum resolution $\sqrt{0.50^2 - 0.12^2}$ = 0.49 a.u. These results are shown in Fig. 2. The concaves near $P_z = 0$ in our data in the [100] direction are apparently absent because of the poorer resolution of our spectrometer. This smoothing effect is also seen in the theoretical Compton profiles [8] convoluted with our instrument function, also shown in Fig. 2 for comparison.

The differences of profiles $J_{111-100}$, $J_{110-100}$ and $J_{111-110}$ appear in Fig. 3. The data of Shiotani *et al.* [14] and the theoretical results [8] convoluted with the proper residual instrument function appear also for comparison. The overestimates of the theory are clear even though the main features agree satisfactorily. Our data show fine structure less sharp than that of Shiotani *et al.* [14]. This effect may be also be largely due to the directional

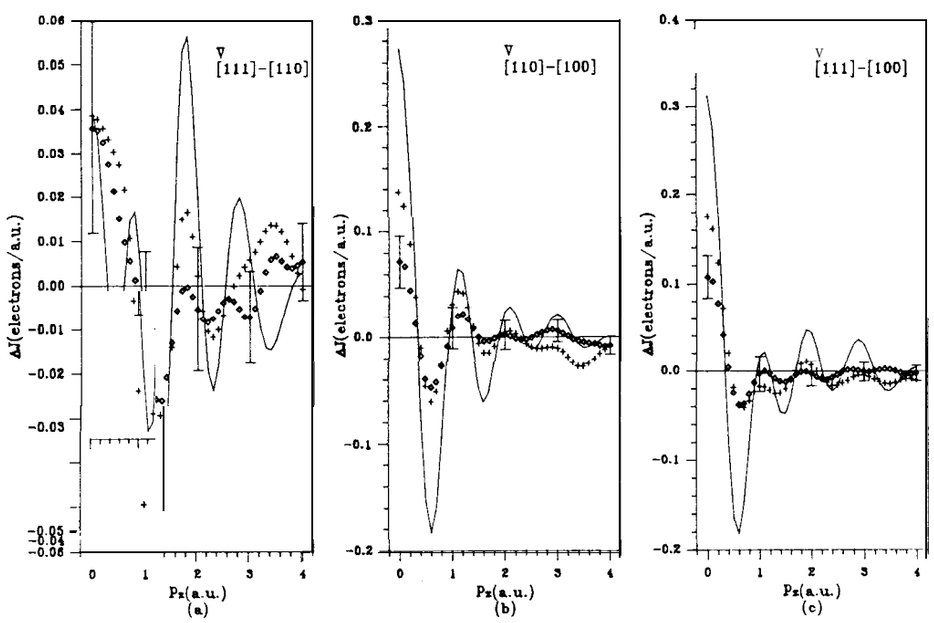

FIG. 3. Differences of Compton profiles $J_{111-100}$, $J_{110-100}$ and $J_{111-110}$ of V; present work (◇), data of Shiotani *et al.* convoluted with our instrument function (+) and the theoretical results similarly convoluted (solid line).

differences mentioned above, and to the poorer resolution and smaller counting statistics of our data.

The positive and negative oscillations of $J_{110} - J_{100}$, $J_{111} - J_{100}$ and $J_{111} - J_{110}$ are discussed by Papanicolaou et *al.* for Ta [3] according to the geometry of the Fermi surfaces. As the geometry of Fermi surfaces of V is similar to that of Ta, we expect the first positive oscillation of $J_{110} - J_{100}$ at $P_z = \frac{2\pi}{\sqrt{2}a} = 0.78$ a.u. and $J_{111} - J_{100}$ at $P_z = \frac{2\pi}{\sqrt{3}a} = 0.63$ a.u. These features are well described according both the theory [8] and the experimental data in Fig. 3. The $J_{111} - J_{110}$ profiles are smaller than those of $J_{110} - J_{100}$ and $J_{111} - J_{100}$; statistically meaningful analysis of the data can be made only with higher counting statistics, but our data show some main features of the profiles $J_{111} - J_{110}$.

III-P. Tantalum

The directional Compton profiles 3.3 ± 1.5" symmetrical to [111], [110] and [100] for single crystals of tantalum are presented in Table I. Fig. 4 shows the isotropic profiles obtained from these directional profiles on applying Houston's method [18] and the isotropic

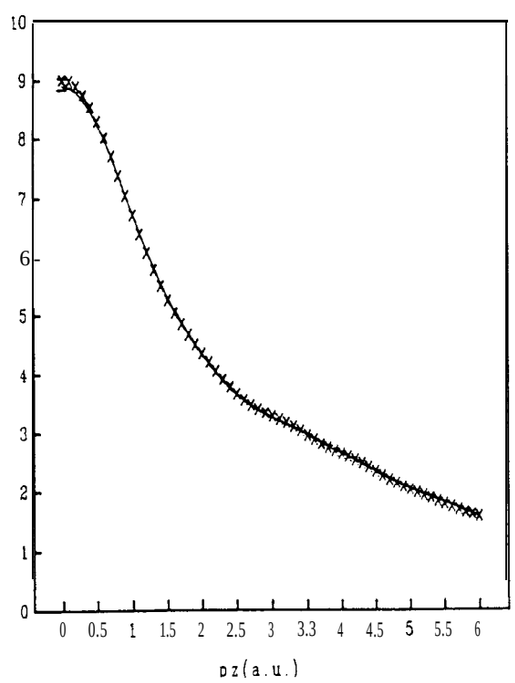

FIG. 4. Isotropic Compton profiles of Ta obtained from a polycrystalline sample (x) and single crystals (solid line) (see test for details).

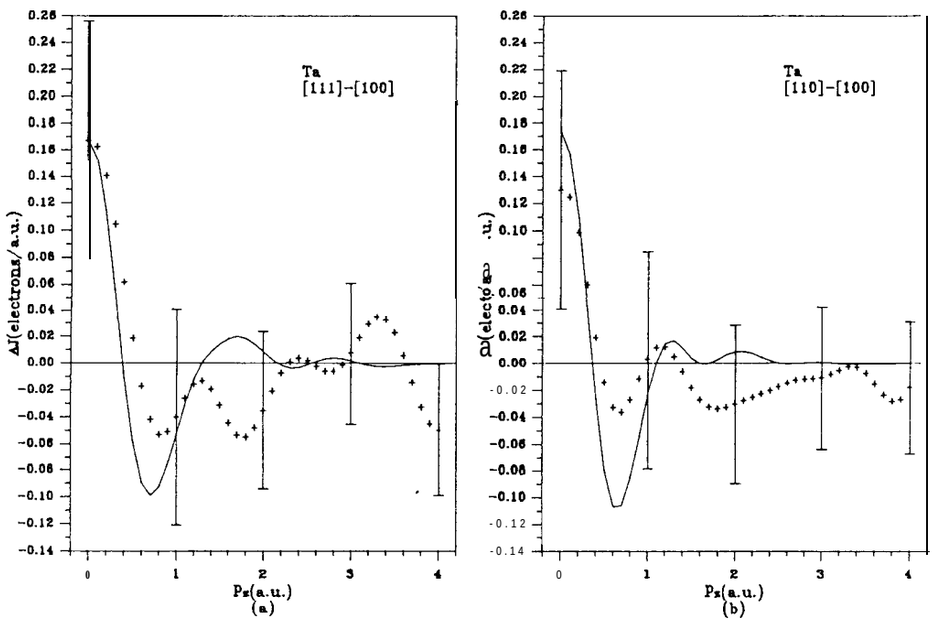

FIG. 5. Differences of profiles $J_{111-100}$ and $J_{110-100}$ of Ta : experimental data (+) and theoretical results convoluted with our residual instrument function (solid line).

profile previously obtained with a polycrystalline sample. They agree satisfactorily within statistical errors. The differences of profiles $J_{111} - J_{100}$ and $J_{110} - J_{100}$ are shown in Fig. 5; the profiles $J_{111} - J_{110}$ are not shown because of the small values within the error bars. Results of theoretical calculations [3] convoluted with a residual instrument function are also shown for comparison. The overestimation of the theory, similarly to those of V [9,14] and Nb [7,10,11], appears also for this high Z element Ta. The first negative oscillations in the differences of profiles $J_{111-100}$ and $J_{110-100}$ agree satisfactorily with the theory, interpreted from the geometry of the Fermi surfaces of Ta [3]. The first positive oscillations are not identified clearly because statistical error bars are large beyond $P_z = 1.0$ a.u.

Vanadium, niobium and tantalum in group 5 of the periodic chart of the elements all have a body-centred cubic structure with lattice parameters 5.71453, 6.23610 and 6.24'706 a.u. respectively. Their directional Compton profiles normalized to their Fermi momenta 1.16636 a.u.,1.06881a.u., and 1.06702a.u. for V, Nb and Ta, respectively, appear in Fig. 6. The trend of these normalized profiles is similar to the normalized isotropic profiles [12], i.e. Ta exhibited the sharpest profiles, Nb less sharp, and V broadest. This trend is attributed to localization of d electrons which increases according to the sequence 5d(Ta),

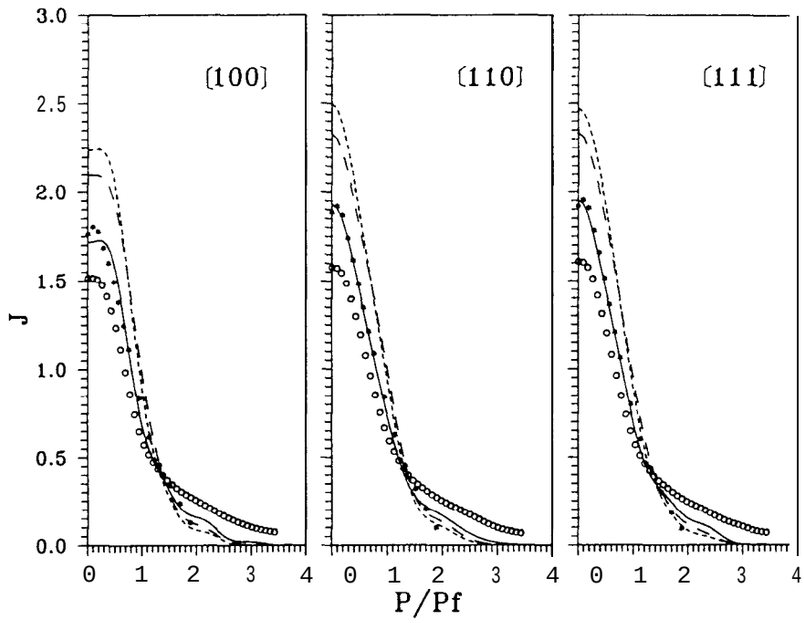

FIG. 6. Normalized directional Compton profiles of V(o o o) and Ta (***). Results of a band calculation for V(-), Nb(- -) and Ta(-- -) are also shown.

4d(Nb), and 3d(V). The results of a band calculation [3] show the same trend but a relatively larger value in the region of small momentum (Fig. 6).

IV. SUMMARY

Here we present results of directional Compton profiles measured with a y-ray spectrometer and an annular source. Data of V and Ta agree with main features predicted according to the band structure theory [3] and are interpreted in terms of the geometry of Fermi surfaces [17]. Overestimations of the theory are common to V and Ta. The effects of electron correlation, the non-local exchange correlation and spin-orbital interaction, suggested in our previous work on isotropic Compton profiles of V, Nb and Ta [12], may have to be taken into consideration to account for these discrepancies.

ACKNOWLEDGMENT

We thank the National Science Council of the Republic of China (NSC 83-0208-M003-16) for support.

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