Study of Electron Momentum Distribution and Compton profiles of β-Mn

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Abstract

Compton profile of transition metal (β-Mn) was calculated employing the renormalized-free-atom (RFA) and free electron models, choosing $3d^{6-x}$-$4s^{1-x}$ configuration, whereas (x=0 to 1 step 0.1). This result was compared with experiment values. It is seen that the RFA calculation in $3d^6 4s^1$ gives a better agreement with the experiment. This theoretical Compton profile data have been used to compute the cohesive energy of Manganese for the first time and compared it with available data.

دراسة توزيع الزخم الالكتروني وشكل منحني كومبتن في β−Mn

المستخلص

تم حساب شكل منحني كومبتن لمتعدد البلورات (β−Mn) والتي ينتمي الى سلسلة (3d) باستخدام النموذجين (اعادة معايرة الذرة الحرة والكترون الحر)، حيث تم اختيار عدد من الترتيبات الكترونية ($3d^{6-x}$ $4s^{1-x}$) اي (x=0,0.1,0.2,0.3,0.4,0.5,0.6,0.7,0.8,0.9,1). قورنت النتائج التي حصلنا عليها نظريا مع البيانات التجريبية السابقة والتي استخدمت فيها مصدر مشع Am^{241} بطاقة 60 KeV. لاحظنا افضل توافق جيد بين حسابتنا بواسطة استخدام نموذج(RFA)عند ترتيب الكتروني $3d^6$ $4s^1$. وكذلك ايضا حسبت طاقة التماسك في العنصر المنغيز وقورنت مع الحسابات المتوفرة.

1. Introduction

It is known that the Compton profile, J (P_z), can provide information about the projection of electronic momentum distribution on the scattering wave vector [1]. Within the impulse approximation, J (P_z) is given by:

$$J(P_z) = \iint \rho(\vec{P})\, dp_x dp_y \quad (1)$$

Where $p_x and p_y$ are the momentum components in $x and y$ directions while the z direction is parallel to the resultant of the incident and scattered wave vectors, $\rho(\vec{P})$ momentum density [2].

Mn belongs to the 3d transition metals and exists in many allotropic forms, most of which have complicated structures [3].In the last decade Compton scattering has been recognized as a powerful tool to study electron structure in light and medium transition metals [4-7], experimental results were predicted reasonably well at medium and high-momentum (p_z> 3.0 a.u) by the free atom profiles. In the low momentum region more refined calculation employing both the band structure as well as simple renormalized free atom (RFA) models can explain the Compton line shapes. Among 3d metals, very little work has been reported on Mn. The first Compton profile measurement on polycrystalline Mn and observed that the measured values were much flatter than the convoluted free atom values at low momenta[8].Compton profile were measured for α and β-Mn and determined the 4s band occupancies as 0.93 and 1.12 respectively[9].Their J(0) values in α and β-Phase different by about 2% and were both < 5 e/au, while a change of 2% in J(0) is rather large, their J(0) values are significantly lower than those observed for all other 3d- metals and thus do not follow the trend of constancy pointed out by [10].The electronic momentum distribution and Compton profiles of silver has been measured by [11].Also Study of electronic momentum distribution and Compton profiles of europium [12].

$3d^{5.6}4s^{1.4}$, $3d^{5.7}4s^{1.3}$) and after convolution electron configuration become ($3d^{5.8}4s^{1.2}$, $3d^{6}4s^{1}$, $3d^{5.7}4s^{1.3}$).Best agreement between our theoretical results and experimental [13] is found for ($3d^{6}4s^{1}$).In§2 we present the details of theoretical calculation .In§3and 4 described the result and discussion, conclusions. Objective of the study

is due to the shortage of refine calculation of electronic momentum density (β-Mn).

2. Calculation

A) Renormalized – free-atom (RFA) model:

The renormalized - atom approach was the firstly to be used by [14]. In the RFA model one starts with the free –atom wave function, truncates them at the Wigner-Sites (WS) Sphere and renormalizes the wave function to one within this Sphere to preserve charge neutrality .The effect of such renormalization in the case of (β-Mn) turned out to be the largest for 4s electron because only 38% of the wave function is contained in the WS Sphere. In contrast, this number is 96% for 3d wave function .Thus, only 4s electron were treated in the RFA scheme.

For bcc metals, the Compton profile $J_{4s}(p_z)$ for 4s electrons, can be written by as [15]:

$$J_{4s}(p_z) = 4\pi \sum_{n=0}^{\infty} |\Psi_0^c(K_n)|^2 \, G_n(p_z) \qquad (2)$$

Where K_n is a reciprocal lattice vector andp_z the projection of electron momentum along the scattering vector direction.

$\Psi_0^c(K_n)$ is the Fourier transform of the RFA wave function $\Phi_0^c(r)$.

(S_Electrons): The procedure for computing Compton profile is *already* published and here we rewrite this equation for the sake of completeness. Following [16] the momentum transform of a Bloch function (for the unhybridised outermost *electrons)* for the cubic structures is given by

$$\Psi_{\vec{K}}(\vec{P}) = N\delta(p - \vec{K}_1\vec{K}_n)\Psi_{\vec{K}}^c(\vec{p}) \qquad (3)$$

Here N is the total number of atoms, the transform $\Psi_{\vec{K}}^c(\vec{p})$ is defined as

$$\Psi_{\vec{K}}^c(\vec{p}) = (2\pi)^{-3/2} \int e^{-\overrightarrow{ip}.\vec{r}} \, \Psi_{\vec{K}}(\vec{r}) d\vec{r} \qquad (4)$$

Where the integration is over the Wigner-Seitz polyhedron. In the conventional cell approximation

$$\Psi_{\vec{K}}(\vec{r}) = e^{i\vec{K}.\vec{r}}\Psi_{\vec{K}=0}(\vec{r}) \quad (5)$$

When $\vec{K}_n = \vec{P} - \vec{K}$, then $\vec{K} = 0$

$$\Psi^c_{\vec{K}}(\vec{P}) = \Psi^c_0(\vec{K}_n) \quad (6)$$

For

$$K_n = 0\Psi^c_0(0) = (2\pi)^{\frac{1}{2}} \int_0^{r_0} dr\, r^2 \Phi^c_0(r) \quad (7)$$

And for $K_n \neq 0$

$$\Psi^c_0(K_n) = (\frac{2}{\pi})^{1/2} K_n^{-1} \int_0^{r_0} drr\sin(K_n r)\,[\Phi^c_0(r) - \Phi^c_0(r_0)] \quad (8)$$

The auxiliary function $G_n(P_z)$ in (1) is given as

For n=0

$$G_0(P_z) = \begin{cases} \frac{1}{2}(P_F^2 - P_Z^2)P_z \leq P_F \\ 0 \qquad otherwise \end{cases} \quad (9)$$

$$\text{For n}\neq 0 G_n(P_z) = \begin{cases} 0 & P_z > K_n + P_F \\ \tilde{G}_n(P_z) P_z \in (K_n - P_F, K_n + P_F) \\ \tilde{G}_n(K_n - P_F) P_Z < K_n - P_F \end{cases} \quad (10)$$

Where the auxiliary function $\tilde{G}_n(P_z)$ is defined as

$$\tilde{G}_n(P_z) = N_n \Big\{(P_F^2 - K_n^2)(K_n + P_F - P_z) - \frac{1}{3}[(K_n + P_F)^3 - P_z^3] + K_n[(K_n + P_F)^2 - P_z^2]\Big\} /4K_n \quad (11)$$

N_n is the number of points in the nth shell in the reciprocal space,as regards the wave function for 4s electrons, the free atom Hartree - Fock wave function was taken from tables of [17].The Compton profile $J_{4s}(P_z)$ was then calculated using equation (2) to (6) for several cases choosing various 3d-4s configuration. The values of the Compton profile of 3d electrons and other inner electrons were taken from [18]. All the theoretical Compton profiles were normalized to an area of 11.28 electrons. As

usual in all 15 shortest reciprocal lattice vectors were considered.

B) Free electron–based model profile:

In case of an isotropic momentum distribution, eq (1) reduces to the well-known form:

$$J_{4s}(p_z) = 2\pi \int_{p_z}^{\infty} dp\, \rho(\vec{p}) p \qquad (12)$$

If we consider the valence electrons in a metal as a non-interacting electron gas, then the momentum density by:

$$\rho(\vec{P}) = constant = \frac{n}{\frac{4}{3}\pi p_F^3} \qquad (13)$$

Where n is the number of free electrons per site and p_F is the Fermi momentum. Substitution of $\rho(p)$ from eq.(13) to eq.(12) gives

$$J_{4s}(p_z) = \frac{3n}{4p_F^3}(p_F^2 - p_z^2) \qquad for\ p_z \le p_F \qquad (14)$$

The free electron Compton profile is then an inverted parabola including discontinuities of the first derivative at $\pm p_F$ [1].Using eq.(14), we have also calculated the free electron Compton profile for 4s electron of Mn. To get a total profile in the momentum range 0 to +7 a.u.,the Compton profile for core electrons($1s^2$ to $3d^5$) were directly taken from the tables of Biggs et al [18].

C) Cohesive energy:

The cohesive energy E_{Coh} which is defined as the difference between the total ground –state energy of the solid and the energy of the individual isolated atoms can be calculated from Compton profile data [19] using following relation:

$$E_{Coh} = \int_0^{p_{max}} p_z^2 [J_s(p_z) - J_{FA}(p_z)] dp_z \qquad (15)$$

Where the $J_s(p_z)$ and $J_{FA}(p_z)$ refer to solid state and free atom profiles, respectively. In our calculation, p_{max} was taken as infinite. The values of $J_s(p_z)$ were taken from the present RFA calculation which represents the solid-state phase of Mn and those for $J_{FA}(p_z)$ from free atom Compton

profile tables[18].the contribution core electrons are same in the$J_s(p_z)$ and$J_{FA}(p_z)$ and hence cancel out in the difference seen in Eq(15) .

3. Results and Discussion

The results theoretical Compton profile for three different Configuration for β-Mn, namely ($3d^{5.8}4s^{1.2}$, $3d^{6}4s^{1}$, $3d^{5.7}4s^{1.3}$) and the free electron profile, all results compared with experimental data [13] given in table 1.In the low momentum region $p_z < 1.4$.It is seen that the experimental values higher than the theoretical (RFA) value but those for $p_z > 1.6$ the experiment values were found to be smaller than from theoretical. In momentum region $p_z > 3$ the experimental and the theoretical values using (RFA, FE and FA) models are nearly same because in this region the contribution is essentially due to inner core electrons. Moreover, these electrons remain nearly unaffected during the formation of solids and hence their Compton profiles can be expected to be described accurately by their atomic values. In order to investigate the effect of varying 3d-4s electron distribution, in Fig (1) we plot the various theoretical and experimental values. We observe that in the beginning from (p_z =0 to 0.4)au, the RFA value for($3d^{5.8}4s^{1.2}$,$3d^{5.7}4s^{1.3}$) are lower than $3d^{6}4s^{1}$ results but between (0.5 to 0.8)au, the trend is reversed and the $3d^{5.8}4s^{1.2}$,$3d^{5.7}4s^{1.3}$ values are higher than $3d^{6}4s^{1}$.Also in $p_z = 1\ to\ 3$ au, the $3d^{6}4s^{1}$ the larger from $3d^{5.8}4s^{1.2}$,$3d^{5.7}4s^{1.3}$,the latter in region$p_z > 3\ au,$ higher than from $3d^{6}4s^{1}$. From table (1) and Fig (1), it can see that the convoluted (free electron and free atom) model profile gives a very poor agreement with the experiment, which may be due to its unrealistic assumption.

Comparison between Free electron and Free atom, it is seen in low momentum Free electron ($3d^{5}4s^{2}$) higher than the Free atom ($3d^{5}4s^{2}$) but in part between p_z (0.3 and 0.8) the trends get reversed and the free electron values are somewhat larger than the free atom .At $p_z > 0.9au,$ both models values to become similar.

Figure (2) shows the difference between theoretical (after convolution) and experimental profiles in β-Mn. It can be seen in the low momentum that ΔJ($3d^{6}4s^{1}$-Expt) larger than fromΔJ ($3d^{5.8}4s^{1.2}$-Expt and $3d^{5.7}4s^{1.3}$-Expt), as well as the ΔJ ($3d^{5.8}4s^{1.2}$-Expt) and ($3d^{5.7}4s^{1.3}$-Expt) have similar values only in low momentum different, but (Free atom - Expt and Free Electron - Expt) are nearly the same where $p_z > 1\ au.$. Also in the high momentum transfer region(p_z >4

au), experimental values are very close to corresponding theoretical data .It is known that the contribution of valence electron is very small in this region and hence, most of the contribution may be due to the inner-core electrons. These inner-core electrons are reasonably described by the free-atom values. In order to determine the best configuration electrons, the total square deviation $\sum_0^{7\ a.u.}|\Delta J|^2$ was obtained for each cases .The values founded were (0.0.5393, 0.5416 and 0.5419) for ($3d^6 4s^1$,$3d^{5.8} 4s^{1.2}$,$3d^{5.7} 4s^{1.3}$) configuration respectively. Thus $3d^6 4s^1$seems to be the best configuration. From this we can observe by effect of convolution the theoretical values.

The purpose of the computation of cohesive energy was to see the applicability of the RFA scheme in reproducing the cohesive of transition metals. The value of calculated cohesive energy (with $p_{max} = 2\ au$). Table (2) show comparison between our theoretical by RFA model , experiment[13] and another data. A choice of low value ofp_{max} is justified because ,to a good approximation ,after this value the major contribution in the theoretical and experimental profile is expected only due to core electrons, which almost remain unaffected in formation of solids. This is evident from the core contribution plotted in Fig (2).

4. Conclusion

In this work compared has been compared theoretical values with experimental results [13].The RFA model shows good agreement with the experiment in the $3d^6 4s^1$ configuration, but Compton profiles value using free electron model higher those experimental. Evidently, there is a need for a relativistic band structure calculation to interpret the Compton profile data. In table (3) illustrate the comparison between theoretical results using (RFA)model with previous works [13,21] in the process transfer charge of shells(s,d).The cohesive energy of Manganese computed by (RFA) model and comparison with another results [13,20].

Table 1: Theoretical results compared with experimental value [13] for β-Mn. All the quantities in atomic. The profiles are normalized so that the integral from 0 to 7.0 au, is 11.28 electrons.

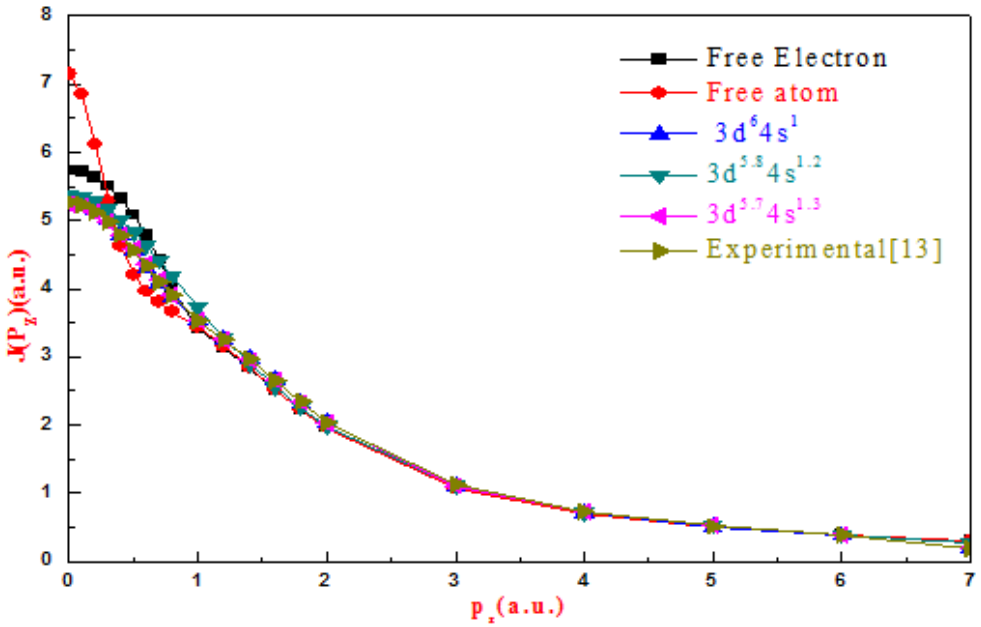
P_z(au)	J(P_z)(e/au)					
	Free electron ($3d^5 4s^2$)	Free atom ($3d^5 4s^2$)	Theory(RFA)			**Experiment [13]**
			Core + $3d^6 4s^1$	Core + $3d^{5.8} 4s^{1.2}$	Core + $3d^{5.7} 4s^{1.3}$	
0.0	5.737	7.1556	5.314	5.253	5.224	**5.371**
0.1	5.71	6.8496	5.279	5.223	5.194	**5.351**
0.2	5.634	6.1176	5.17	5.128	5.108	**5.283**
0.3	5.502	5.2856	5.002	4.981	4.969	**5.16**
0.4	5.314	4.6286	4.79	4.789	4.787	**5.01**
0.5	5.078	4.2084	4.55	4.568	4.576	**4.834**
0.6	4.778	3.9536	4.312	4.337	4.347	**4.629**
0.7	4.426	3.798	4.075	4.106	4.12	**4.416**
0.8	3.994	3.6596	3.875	3.896	3.905	**4.192**
1.0	3.429	3.4364	3.539	3.538	3.538	**3.73**
1.2	3.147	3.1548	3.26	3.247	3.24	**3.282**
1.4	2.837	2.839	2.973	2.962	2.955	**2.895**
1.6	2.518	2.5128	2.656	2.653	2.647	**2.565**
1.8	2.236	2.2242	2.333	2.331	2.328	**2.262**
2	1.964	1.949	2.035	2.025	2.029	**1.981**
3	1.078	1.064	1.111	1.11	1.107	**1.104**
4	0.701	0.692	0.714	0.725	0.715	**0.717**
5	0.514	0.508	0.509	0.512	0.515	**0.525**
6	0.392	0.388	0.378	0.384	0.388	**0.388**
7	0.301	0.298	0.194	0.195	0.195	**0.279**

Table 2: Cohesive energy of Manganese (E_{Coh}). 1 au, of Energy is equivalent to27.212 eV .

Reference	E_{Coh}**(in eV)**
Our theoretical(RFA)	8.04
Experiment[13]	8.24
Brooks and Johansson[20]	8.16

Table 3. Charge occupancies in β-Mn.

Reference	Q_{3d}	Q_{4s}
Das .G.P and Shhni .V.C.[21]	5.88	**1.12**
Ahuja.B.L.et al [13]	5.4	**1.6**
Present work	6	**1**



Fig(1). Comparison of theoretical results with experimental [13] Compton profiles for β-Mn.

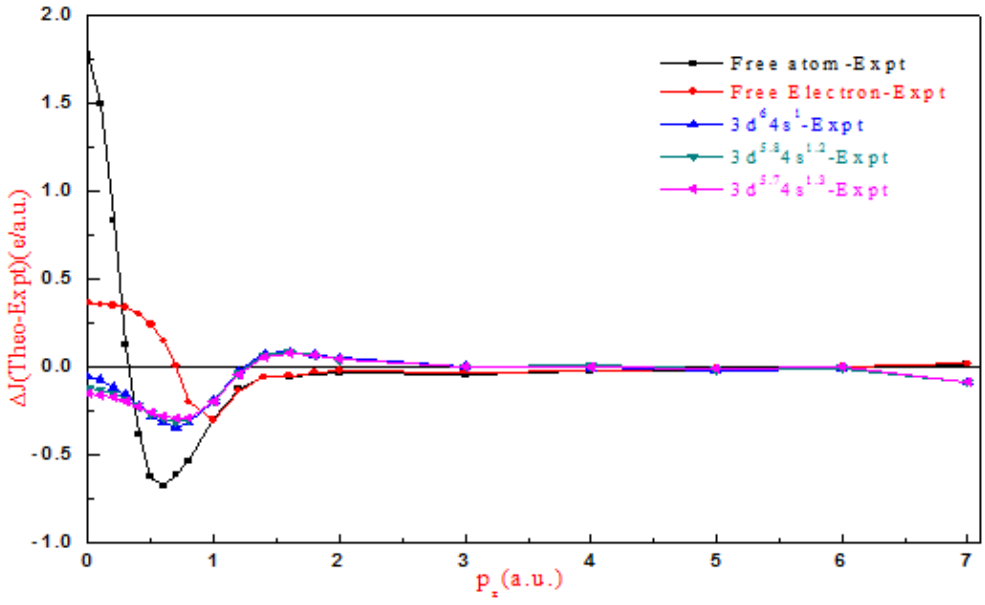


Fig (2). Difference between our theoretical and experimental [13] Compton profiles of β-Mn.

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