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Impulse Approximation in Compton Scattering

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Calculations of the core contributions to the Compton profiles for lithium ($1s^2$), carbon ($1s^2$), and aluminum ($2s^2 2p^6$) were made employing excited-state one-electron continuum wave functions evaluated in the ground-state Hartree-Fock-Slater potential of the nucleus and remaining electrons. Good agreement with the impulse approximation is obtained. Measurements of the Compton profiles of lithium metal, graphite, and aluminum metal were made with Mo $K\alpha$ radiation and the core contributions were separated from the valence electron contributions. The Compton profiles for the core electrons [$1s^2$ for lithium and carbon, $2s^2 2p^6$ for aluminum ($1s^2$ not excited)] were in fair agreement with the calculations. Total Compton cross sections (integrated over the Compton profile) were calculated in the impulse approximation and compared to the Waller-Hartree theory with and without the Bonham corrections. The agreement can be quite poor, differing at low angles by as much as a factor of 3.

I. INTRODUCTION

The recent interest in Compton profile measurements[1–4] makes it desirable to explore methods that enable one to separate the core electron contribution from that of the valence electeons. In solids where valence electrons undergo very pronounced changes in their momentum distributions from free atoms, the Compton profile provides a most critical test of band wave functions. On the other hand, the core electrons differ very little between free atom and solid so that a Hartree-Fock free-atom calculation for the core electrons should suffice to determine their contribution to the Compton profile in the solid. Calculations of the core contributions have been made for Hartree-Fock wave functions employing the so-called impulse approximation.[1] Eisenberger and Platzman[3] have studied the validity of the impulse approximation in the case of the hydrogenic atom. It is our purpose in Sec. II to extend the theoretical comparison to the many-electron atom as well as to compare theory with experiment for core electrons.

In addition, the theoretical total free-atom cross sections (integral over final photon frequencies) in the impulse approximation are compared in Sec. III with the total cross sections in the Waller-Hartree[5] approximation with and without the higher-order corrections suggested by Bonham.[6]

II. DIFFERENTIAL CROSS SECTION

Theory

To lowest order, the differential cross section for Compton scattering from an N-electron atom arising from the A^2 interaction (A = vector potential) of the electron with the electromagnetic field is

$$\frac{d^2\sigma}{d\Omega\, d\omega} = \left(\frac{e^2}{mc^2}\right)^2 K^2 \frac{\omega_2}{\omega_1} \int \left| \langle f| \sum_{i=1}^{N} e^{i\vec{S}\cdot\vec{r}_i} |0\rangle \right|^2 \times \delta(\epsilon_f - \epsilon_0 - \omega)\, d\vec{f} \;, \quad (1)$$

where ϵ_f and ϵ_0 are the final and initial energies of the system, ω_1 and ω_2 the initial and final photon energies, $\vec{S}$ and ω the momentum and energy transfers, K is the dot product of the incident and scattered polarization vectors,

$$|\vec{S}(\omega)| = 2\omega_1 \sin\theta \left[1 - \frac{\omega}{\omega_1} + \left(\frac{\omega}{\omega_1}\right)^2 \frac{1}{4\sin^2\theta}\right]^{1/2} ,$$

and θ is one-half the scattering angle. The δ function signifies conservation of energy. For conditions ordinarily met in practice, i.e., x-ray energies of 20 keV and elements up to $Z = 30$ the $\vec{p}\cdot\vec{A}$ term in the interaction is small since the x-ray energy is not near any absorption edge.

Let us take the initial state $|0\rangle$ as a single determinant of one-electron orbitals and the final states $|f\rangle$ as the same determinant with only one orbital, say, $|k\rangle$, being replaced by an excited state $|g\rangle$. The $|g\rangle$'s are one-electron eigenstates in the ground-state potential $V(\vec{r})$ of the nucleus and remaining electrons. Equation (1) then reduces to

$$\frac{d^2\sigma}{d\Omega\, d\omega}=\left(\frac{e^2}{mc^2}\right)^2 K^2\,\frac{\omega_2}{\omega_1}\sum_k\int\left|\langle g\,|e^{i\vec{S}\cdot\vec{r}}\,|k\rangle\right|^2 \times\delta(\epsilon_g-\epsilon_k-\omega)\,d\vec{g}\ , \tag{2}$$

where ϵ_g and ϵ_k are the orbital energies of $|g\rangle$ and $|k\rangle$, respectively. The replacement of $\epsilon_f-\epsilon_0$ by $\epsilon_g-\epsilon_k$ in the argument of the δ function is consistent with the one-electron picture presented here. For a spherical potential $V(r)$, Eq. (2) can be evaluated[7] using final-state continuum functions of the type

$$\Psi_g(\vec{r})=g_l(r,\epsilon)\,Y_l^m(\theta,\phi)\ , \tag{3}$$

where the $Y_l^m(\theta,\phi)$ are the spherical harmonics and the $g_l(r,\epsilon)$ are solutions of

$$\left(-\frac{1}{2}\,\frac{d^2}{dr^2}+V(r)+\frac{l(l+1)}{2r^2}-\epsilon\right)r\,g_l(r,\epsilon)=0\ .$$

Letting

$$\int d\vec{g}=\sum_{l\,m}\int\frac{dn}{d\epsilon}\,d\epsilon\ ,$$

the expression for the cross section becomes

$$\frac{d^2\sigma}{d\Omega\, d\omega}=\left(\frac{e^2}{mc^2}\right)^2 K^2\,\frac{\omega_2}{\omega_1}\sum_k\sum_{l,m}\frac{dn}{d\epsilon} \times\left|\int g_l(r,\epsilon)\,Y_l^m(\theta,\phi)\,e^{i\vec{S}\cdot\vec{r}}\,\Psi_k(\vec{r})\,d\vec{r}\right|^2\ , \tag{4}$$

where ϵ now stands for $\epsilon_k+\omega$ and $dn/d\epsilon$ is the density of (l,m) states. For light atoms the sum over k will involve mostly s and p states, so that for each k and l, selection rules permit only one or two terms from the partial expansion of $e^{i\vec{S}\cdot\vec{r}}$ to contribute. The density of states is determined by allowing g_l to vanish at the surface of a sphere of radius R. For r large,

$$g_l(r,\epsilon)\cong\frac{\sin(\sqrt{\epsilon}\,r+\gamma\ln\gamma+\eta_l)}{\sqrt{\epsilon}\,r}\ , \tag{5}$$

where η_l is a phase shift and the term in $\ln\gamma$ arises from the Coulomb tail of the potential. As $R\rightarrow\infty$ the density of states becomes

$$\frac{dn}{d\epsilon}\sim\frac{R}{2\pi\sqrt{\epsilon}}\ , \tag{6}$$

and from Eq. (5)

$$\int_0^R g_l^2(r,\epsilon)\,r^2\,dr=R/2\epsilon\ . \tag{7}$$

Finally for an electron initially in an s state with the radial wave function $\Psi_s(r)$ and with energy ϵ_s we have

$$\frac{d^2\sigma}{d\Omega\, d\omega}=\left(\frac{e^2}{mc^2}\right)^2 K^2\,\frac{\omega_2}{\omega_1}\,4(\epsilon_s+\omega)^{1/2}\sum_{l=0}^{\infty}\left|\int_0^{\infty} g_l(r;\epsilon_s+\omega)\,j_l(Sr)\,\Psi_s(r)\,r^2\,dr\right|^2\ , \tag{8}$$

excited-state approximation, while the contribution from a filled shell of six p electrons gives for the excited-state approximation

$$\frac{d^2\sigma}{d\Omega\, d\omega}=12\left(\frac{e^2}{mc^2}\right)^2 K^2\,\frac{\omega_2}{\omega_1}\,(\epsilon_p+\omega)^{1/2}\sum_{l=0}^{\infty}\left((l+1)\left|\int_0^{\infty} g_l(r;\epsilon_p+\omega)\,j_{l+1}(Sr)\,\Psi_p(r)\,r^2\,dr\right|^2 +l\left|\int_0^{\infty} g_l(r;\epsilon_p+\omega)\,j_{l-1}(Sr)\,\Psi_p(r)\,r^2\,dr\right|^2\right)\ . \tag{9}$$

The numerical calculations were performed to sufficiently large values of r so that the WKB approximation could then be used to obtain the normalization of Eq. (5). The normalized WKB approximation is

$$g_l(r,\epsilon)=\frac{1}{4(1-W/\epsilon)^{1/2}}\ \frac{\cos\{\int[\epsilon-W(r')]^{1/2}dr'+\eta\}}{\sqrt{\epsilon}\,r}\ , \tag{10}$$

where η is a phase shift and $W(r)=V(r)+l(l+1)/r^2$. On the other hand, for a closed shell the impulse approximation gives

$$\frac{d^2\sigma}{d\Omega\, d\omega}=\left(\frac{e^2}{mc^2}\right)^2 K^2\,\frac{\omega_2}{\omega_1}\,\frac{m}{|S|}\sum_i J_i(z)\ , \tag{11}$$

with

$$J_i(z)=2\pi\int_{|z|}^{\infty}|\chi_i(p)|^2\,p\,dp\ , \tag{12}$$

where the sum i runs over all occupied orbitals. The $\chi_i(p)$ are the one-electron ground-state momentum wave functions, normalized to

$$4\pi\int_0^{\infty}|\chi_i(p)|^2\,p^2\,dp=1\ . \tag{13}$$

The variable z is defined as

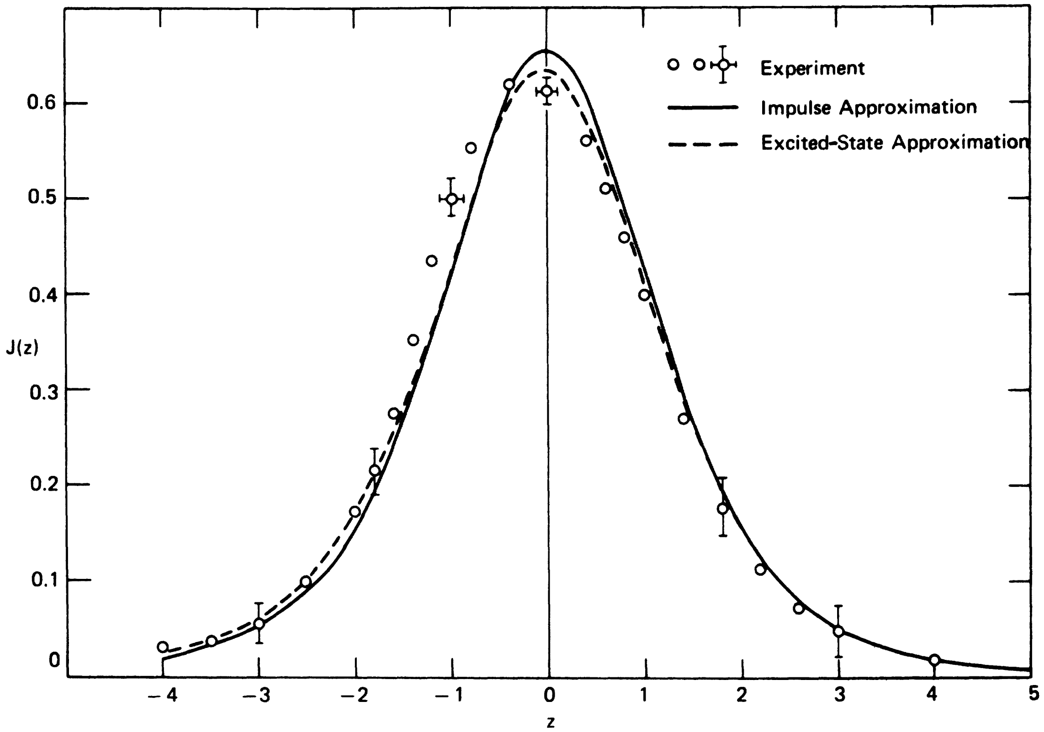

FIG. 1. The measured core electron ($1s^2$) Compton profile for lithium together with the theoretical profiles in the impulse [Eq. (12)] and the excited-state approximation [Eqs. (8) and (9)].

$$z(\omega) = m\omega/|S| - |S|/2 . \tag{14}$$

In a semiclassical picture z represents the component along $\vec{S}$ of the scattering electron's initial momentum.

The two calculations are compared in Figs. 1–3 for Li, C, and Al and are found to be in good mutual agreement. Of course the great advantage of the impulse approximation is that the cross section [Eq. (12)] explicitly involves only the ground-state wave functions.

Experiment

Compton profile measurements were made on lithium metal (polycrystal), aluminum metal (polycrystal), and on pyrolytic graphite employing Mo $K\alpha$ radiation scattered, respectively, 155°, 120°, and 160° from the three samples. After subtracting background and making a small K_β correction, the $\alpha_1 - \alpha_2$ separation was made by the Rachinger[8] method. Between 20 000 and 100 000 x rays were accumulated per point near the Compton peak at intervals of 0.02° 2θ employing a LiF (400) crystal as an analyzer. The technique is identical to that described earlier.[9]

The separated $K\alpha_1$ profiles were resolved into core and valence electron contributions by making use of the following guides:

(i) The valence electrons have a sufficiently small binding energy so that the impulse approximation is quite accurate. Hence, the valence electron profile is symmetric about the center of the line.

(ii) For the large values of $|\vec{S}|$ used, the area under the profiles can be normalized to (cf. Sec. II)

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 K^2 \left\{ N_v \left(\frac{\omega_2^0}{\omega_1}\right)^2 + \sum_{i=1}^{N_c} \left[\left(\frac{\omega_2^0}{\omega_1}\right)^2 - f_{ii}^2\right]\right\} , \tag{15}$$

where $N_v(N_c)$ is the number of valence (core) electrons per atom.

(iii) The core contribution is a very smoothly varying curve similar to that given by the impulse approximation while the valence contribution, in addition to being symmetric, is everywhere ≥ 0.

Using the above normalization conditions and the symmetry requirement for the valence profile, one can determine an experimental core profile $J_c(z)$ by a trial and error method taking the impulse approximation core profile as a starting point. Strictly speaking, such a profile is not unique. However, the requirement that the $J_c(z)$ be reasonably smooth and such that

$$J_T(z) > J_c(z) > 0 \quad [J_T(z) = \text{total profile}]$$

for all values of z, leaves indeed very little choice. Whatever uncertainty subsisted was added to the

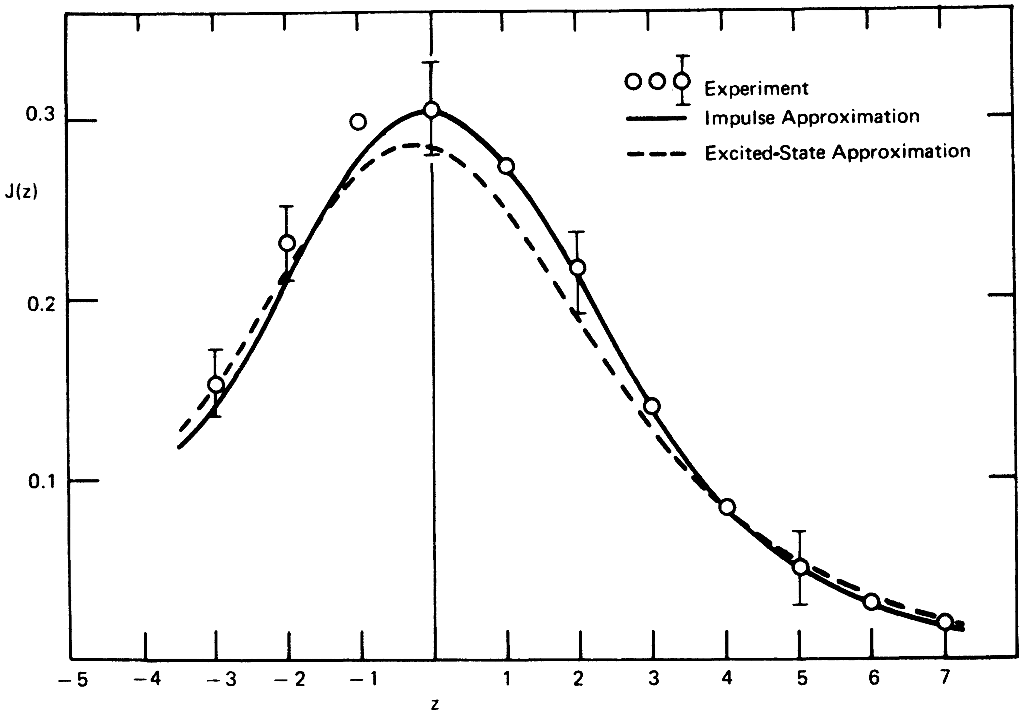

FIG. 2. The measured core electron ($1s^2$) Compton profile for carbon (graphite) together with the theoretical profiles in the impulse [Eq. (12)] and excited-state approximation [Eqs. (8) and (9)].

experimental uncertainties.

As it turned out, the departures from the impulse approximation were small. The separated experimental curves are given in Figs. 1–3 for Li, C, and Al, respectively, and show good agreement with theory. Only the lithium core shows a small departure from the impulse approximation. The lithium data were the most accurate of the three since the valence electron contribution was the smallest.

From the above results as well as from earlier work on Be,[7] it appears that depatures from the impulse approximation are small (even for core electrons) and that the use of free-atom wave functions in the impulse approximation yields a Compton profile for the core electrons accurate to a few percent. Compton profiles have been published for Hartree-Fock free-atom wave functions in the impulse approximation for elements Li through Ge.[10]

III. TOTAL CROSS SECTION

If we integrate Eq. (1) over final photon frequencies, we obtain the total cross section. The simplest and earliest known case considered is the scattering by a free electron. For an electron initially at rest in the laboratory frame we have the well-known Klein-Nishina formula, which for ordinary x rays reduces to

$$\frac{d\sigma}{d\Omega} \cong \left(\frac{e^2}{mc^2}\right)^2 K^2 \left(\frac{\omega_2^0}{\omega_1}\right)^2 , \tag{16}$$

where $(\omega_2^0/\omega_1)^2$ is commonly referred to as the Breit-Dirac factor. The superscript to the final photon energy ω_2^0 is employed to designate the special case of the electron initially at rest. For such a case the final photon energy is related to the initial energy by Compton's formula:

$$\omega_2^0 = \frac{\omega_1}{1 + 2\omega_1 \sin^2\theta / mc^2} . \tag{17}$$

Equation (16) is often written in terms of the wavelength change $\Delta\lambda$ upon scattering [$\Delta\lambda = (2h/mc) \times \sin^2\theta$]. Within the independent-particle model (cf. Sec. II), Eq. (16) can be extended to a Z-electron system yielding the well-known Waller-Hartree[5] expression

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 K^2 \left(\frac{\omega_2^0}{\omega_1}\right)^2 \left\{ Z - \sum_{i=1}^{z} f_{ii}^2 - \sum_{i \neq j}' f_{ij}^2 \right\}$$

$$\equiv \left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{WH}} , \tag{18}$$

where $\sum'$ extends over all couples of occupied orbitals of the same spin. The Breit-Dirac factor was inserted artificially into Eq. (18) in order to reach consistency with Eq. (16) in the free-elec-

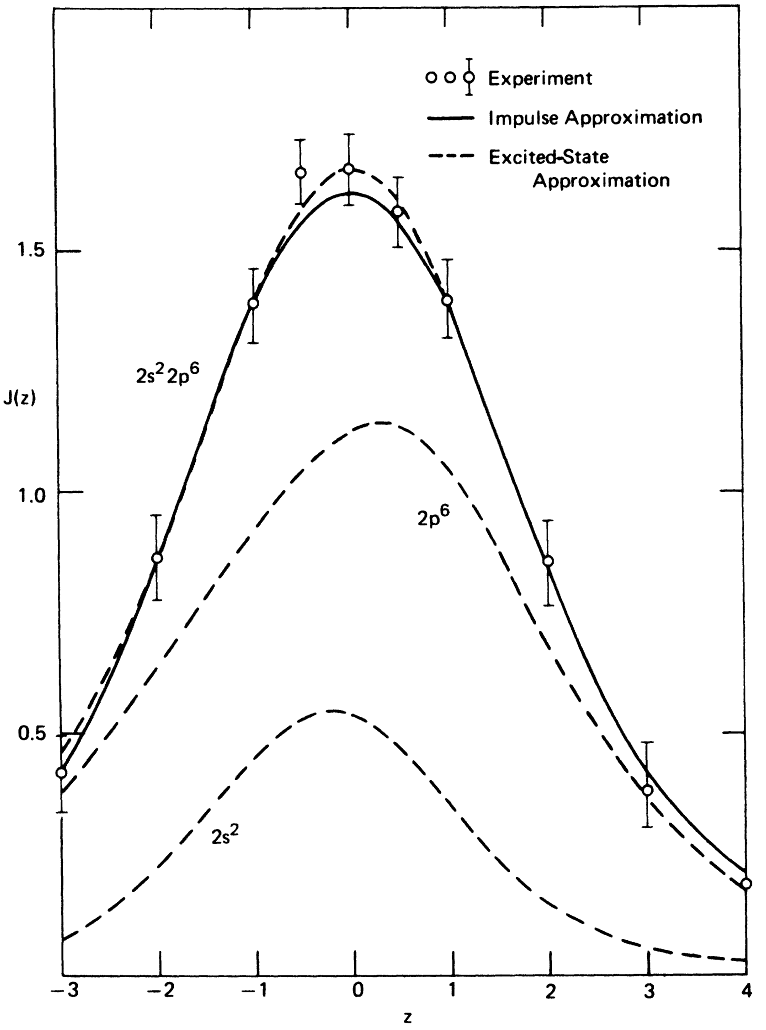

FIG. 3. The measured core electron ($2s^2 2p^6$) Compton profile for aluminum together with the theoretical profiles in the impulse [Eq. (12)] and excited-state approximation [Eqs. (8) and (9)]. Included are the $2s^2$ and $2p^6$ excited-state approximation profiles. The $1s^2$ are not excited over the range of interest of the profiles.

tron case. However, following Bonham's method[6] of time-dependent perturbation theory, first- and second-order correction terms to the Waller-Hartree expression have been calculated as

$$\frac{d\sigma}{d\Omega}=\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{WH}}+\delta_1+\delta_2 \quad , \tag{19}$$

where

$$\delta_1=-\left(\frac{e^2}{mc^2}\right)^2 K^2\,\frac{S^2(0)}{m\omega_1}\left\{\sum_{i=1}^{z} f_{ii}^2+\sum_{i\neq j}{}' f_{ij}^2\right\}, \tag{20}$$

$$\begin{aligned}\delta_2\cong\left(\frac{e^2}{mc^2}\right)^2 K^2\Bigg\{&\frac{2}{3}\,\frac{|E_0|}{mc^2}\,(1+4\sin^2\theta)\\&+3Z\left(\frac{S^2(0)}{2m\omega_1}\right)^2+\frac{ZS^2(0)}{mc^2\,2m}\Bigg\} \quad ,\end{aligned} \tag{21}$$

where E_0 is the total electronic energy of the system. The inclusion of δ_1 in Eq. (18) yields

$$\frac{d\sigma}{d\Omega}=\left(\frac{e^2}{mc^2}\right)^2 K^2\left\{Z\left(\frac{\omega_2^0}{\omega_1}\right)^2-\sum_{i=1}^{z} f_{ii}^2-\sum_{i\neq j}{}' f_{ij}^2\right\}, \tag{22}$$

so that the Breit-Dirac factor is now naturally included.

Finally it is also possible to express the total cross section in the impulse approximation as an integral over the Compton profile. For each one-electron orbital i we have

$$\left(\frac{d\sigma}{d\Omega}\right)_i=\left(\frac{e^2}{mc^2}\right)^2 K^2\int_{|\epsilon_i|}^{\omega_1}\frac{\omega_2}{\omega_1}\,\frac{m}{|S|}\;J_i(z)\,d\omega \ , \tag{23}$$

where Eq. (14) expresses the relationship between z and ω. The integral runs from the one-electron binding energy $|\epsilon_i|$ to an upper limit of the incident photon energy.

In Table I are listed some representative total cross sections in the Waller-Hartree theory and for the integrated profile in the impulse approximation together with the first- and second-order

TABLE I. The total Compton cross sections $d\sigma/d\Omega$ in electron units for Li and Ge in the Waller-Hartree theory, the Bonham corrections δ_1 [Eq. (20)] and δ_2 [Eq. (21)] to the Waller-Hartree theory (Mo $K\alpha$), and the total cross section obtained from an integration of the Compton profiles in the impulse approximation.

$\frac{\sin\theta}{\lambda}$	Li $\lvert E_0\rvert = 202$ eV		Ge $\lvert E_0\rvert = 56\,400$ eV			
	WH	Impulse	WH	δ_1	δ_2	Impulse
0	0	0	0	0	0.073	0
0.1	1.05	1.00	1.59	−0.02	0.075	5.72
0.2	1.44	1.47	4.73	−0.074	0.08	9.05
0.3	1.82	1.96	7.34	−0.15	0.08	11.2
0.4	2.16	2.32	9.91	−0.24	0.10	13.1
0.5	2.42	2.54	12.3	−0.34	0.12	14.7
0.6	2.60	2.66	14.4	−0.44	0.15	16.2
0.7	2.70	2.73	16.0	−0.53	0.19	17.5
0.9	2.76	2.76	18.5	−0.75	0.30	18.4
1.1	2.74	2.73	20.2	−0.98	0.46	20.4
		$\lvert\delta_1\rvert < 0.03$ $\lvert\delta_2\rvert < 0.02$				

Bonham corrections to the Waller-Hartree theory.

As can be seen in Table I, the difference between the Waller-Hartree theory even with the corrections by Bonham can be large [for example, at $(\sin\theta)/\lambda = 0.1$ Å^{-1} for Ge]. It is obviously desirable to have measurements to evaluate the two theories. A more complete set of tables of total Compton cross sections are available from one of the authors (R. J. W.).

IV. CONCLUSION

The most important point suggested by our results is that the impulse approximation for core electrons provides a convenient scheme for subtracting their contributions from the observed Compton profiles. Since the valence electron contribution is only sizable near the Compton peak, the error in separating the valence electron profile from the core can probably be kept to a few percent.

The total cross sections as calculated in the Waller-Hartree approximation and the impulse approximation can differ greatly but not at the large values of $(\sin\theta)/\lambda$ employed in Compton profile measurements. Thus, absolute normalization of the profiles can be done to within a few percent when conditions are chosen such that $(\sin\theta)/\lambda > 1.0$ Å^{-1}.

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Self-Consistent Electronic Structure of Titanium. II[†][*]

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The band structure, Fermi surface, and density of states from an augmented-plane-wave calculation of titanium are presented and compared with previously reported results. The present calculation differs from the earlier work in that the potential is a self-consistent muffin-tin form and further in that the coefficient of the Slater exchange term is $\frac{3}{4}$ instead of 1.

I. INTRODUCTION

The aims of the present calculation are (i) to obtain the self-consistent-potential energy bands and Fermi surface of titanium within the muffin-tin one-electron potential-energy form[1] for comparison with a previous "one-shot" calculation,[2] (ii) to determine the effect on the self-consistent energy eigenvalues at the Γ point for various choices of the parameter α in the $\chi\alpha$-method[3] exchange poten-