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Leading corrections to atomic impulse-approximation Compton profiles: A density-functional approach

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By use of a theory due to Gasser, Tavard, and others, the leading first-order corrections to the spherically averaged atomic Compton profiles (CP's) beyond the impulse approximation, for some inert closed-shell monatomic systems (He, Ne, Ar, and Kr), are obtained. This is accomplished by employing the Kohn-Sham technique of the density-functional theory, which leads to a self-consistent description, as opposed to the popular "effective-hydrogenic-potential" theories used in this context. The resulting corrections to the CP's thus obtained are seen to compare well with the earlier effective hydrogenic estimates. The orbital corrections are seen to conform to an empirical rule put forth ealier by Gasser and Tavard [Phys. Rev. A **27**, 117 (1983)].

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I. INTRODUCTION

Recently, some accurate theoretical [1–7] as well as experimental [8–11] investigations have been carried out, bringing out the deviations of the exact atomic Compton profiles (CP's) [12] from their corresponding impulse-approximation (IA) [13,14] theoretical estimates, an effect first predicted by F. Bloch [15] in 1934. Tavard and Bonham [16] and subsequently B. Bloch and Mendelsohn [17,18] developed theoretical framework for the calculation of atomic Compton profiles within the first Born approximation. Bloch and Mendelsohn employed the "exact" hydrogenic bound- and continuum-state wave functions for a one-electron atomic system, and, within the Born regime, obtained closed-form expressions for atomic, orbital CP's. Their studies clearly brought out the deviations of the exact CP's from the corresponding spherically averaged, symmetric, impulse-approximation CP's. A measure of asymmetry in the exact CP is the "Compton defect," which is the shift of $q_{\max}$, the value of the Compton parameter q corresponding to the peak value of the exact CP, from $q=0$, where the maximum in the corresponding IA-CP occurs. This phenomenon has been studied extensively by several workers [1–12,15–20].

In a systematic and accurate theoretical treatment developed to account for the aforementioned discrepancy, Gasser, Tavard, and others [1–3] obtained the exact atomic CP in the form of an infinite series as

$$J(q,\mathbf{k})=J^{(0)}(q)+J^{(1)}(q,\mathbf{k})+J^{(2)}(q,\mathbf{k})+\cdots\ , \qquad (1)$$

in which $J^{(0)}(q)$ is the IA profile and the subsequent terms represent the corrections to it. Here q, the Compton parameter is given by (Hartree atomic units are used throughout) $q=E/k-k/2$ with E the energy and $\mathbf{k}$ the momentum transferred by the projectile (such as a high-energy x-ray or γ-ray photon) to the electron, in the Compton process. Gasser and co-workers [1–3] showed that the terms in Eq. (1) are alternately symmetric and antisymmetric in q, and further that they bear a rather simple (and desirable) $k\equiv|\mathbf{k}|$ dependence: $J^{(n)}(q,\mathbf{k})\propto 1/k^n(n=0,1,2,\ldots)$; thus making the successive correction terms in Eq. (1) of diminishing significance, for large enough momentum transfers. Equation (1) is thus a power-series expansion of the exact CP, in powers of $1/k$, with their respective coefficients dependent on q and the direction of $\mathbf{k}$. For moderate momentum transfers ($k\sim 5Z$ a.u., Z being the nuclear charge) the only leading first-order (in $1/k$) term $J^{(1)}$ turns out to be a significant correction, as is borne out from the studies of Bell [4], Holm and Ribberfors [6], and Datta, Bera, and Talukdar [7]. This correction term precisely forms the theme of the preset work, aimed at obtaining decent estimates of $J^{(1)}(q,\mathbf{k})$ for a few closed-shell inert atomic systems viz. He, Ne, Ar, and Kr, using the atomic self-consistent-field (SCF) description offered by the Hohenberg-Kohn-Sham [21–23] density-functional approach. In short, we estimate the leading correction by means of a one-particle theory, viz. the Kohn-Sham (KS) [22–23] technique, applied to the many-electron, wave-function-based theory of Gasser and co-workers [1–3], which is accomplished through the method outlined below.

II. METHOD

Following Gasser and Tavard [2], we note that within the first Born approximation, the double-differential cross section for the scattering of an electron belonging to an N-electron scatterer may be expressed in terms of the exact compton profile J:

$$J(q,\mathbf{k})=(k/2\pi)\sum_{\mu=1}^{N}\int_{-\infty}^{\infty}dt\, e^{-ikqt}F_{\mu}(t,\mathbf{k})\ , \qquad (2)$$

with

$$F_\mu(t,\mathbf{k})=\langle\Phi_a|e^{it(X+C_\mu)}|\Phi_a\rangle \;, \tag{3}$$

and the operators X and C_μ stand for

$$X=(H-E_a) \;, \tag{4}$$

$$C_\mu=-i\mathbf{k}\cdot\nabla_\mu \;. \tag{5}$$

Here, H is the N-electron Hamiltonian with $\Phi_a(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\ldots,\mathbf{r}_N)\equiv\langle\mathbf{r}_1,\mathbf{r}_2,\ldots\mathbf{r}_N|\Phi_a\rangle$ the many-electron stationary state wave function prior to scattering (the "initial" state) with the associated energy eigenvalue E_a. By definition, $X|\Phi_a\rangle=0$. The operator $\exp[it(X+C_\mu)]$ has the decomposition [2,3]

$$e^{it(X+C_\mu)}=e^{itC_\mu}+ie^{itC_\mu}\int_0^t dt' e^{-it'C_\mu}Xe^{it'(x+C_\mu)} \;. \tag{6}$$

This operator identity may be established by preoperating throughout by $\exp(-itC_\mu)$ and then noting that both sides satisfy the same first-order differential equation in the time t, and further that they both reduce to the identity operator at $t=0$. Equation (6) may be iterated, engendering an infinite series in powers of $1/k$. In the inelastic scattering brought about, for example, by a high-energy photon imparting large enough energy and momentum transfers to the electron $[k\gg(E_b)^{1/2}$, where E_b is the binding energy of an electron in the N-electron system], E becomes overwhelmingly greater than $|E_a|$, making the time scales involved in the electronic transition from a bound electronic state to a continuum state extremely small [12–14], so that only the first term in Eq. (6) may be retained. This term, on substitution in Eq. (2) immediately gives a $\mathbf{k}$-independent, symmetric (in q) directional IA-CP. This CP, upon spherical averaging, is denoted herein by $J_{\mathrm{IA}}^{(0)}(q)$. It must be noted that the experimentally accessible CP, through scattering from gas phase atoms and molecules, also gives a spherically averaged information. The leading first-order correction to $J_{\mathrm{IA}}^{(0)}(q)$ is then obtained, invoking the approximation (valid for small time scales $\sim\hbar/E\ll1$ [12]) $\exp[it'(X+C_\mu)]\approx\exp(it'C_\mu)$ on the right-hand side of Eq. (6) and truncating the process there [2,3], yielding

$$J^{(1)}(q,\mathbf{k})=(k/2\pi)\sum_{\mu=1}^{N}\int_{-\infty}^{\infty}dt\, e^{-ikqt}F_\mu^{(1)}(t,\mathbf{k}) \;. \tag{7}$$

This explicit form of $F_\mu^{(1)}$ is

$$F_\mu^{(1)}(t,\mathbf{k})=i\langle\Phi_a|e^{itC_\mu}\int_0^t dt' e^{it'C_\mu}(H-E_a)e^{it'C_\mu}|\Phi_a\rangle \tag{8}$$

which, in terms of the many-body potential-energy operator $U(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)$, emerges as [2,3]

$$\begin{aligned}F_\mu^{(1)}(t,\mathbf{k})={}&(i/k)\int_0^R dR'\int d^3r_1d^3r_2\cdots d^3r_N\Phi_a^*(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)\\&\times[U(\mathbf{r}_1,\ldots,\mathbf{r}_\mu+\mathbf{R}',\ldots,\mathbf{r}_N)-U(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)]\Phi_a(\mathbf{r}_1,\ldots,\mathbf{r}_\mu+\mathbf{R},\ldots,\mathbf{r}_N) \;,\end{aligned} \tag{9}$$

where $\mathbf{R}=\mathbf{k}t$, $\mathbf{R}'=\mathbf{k}t'$. This Gasser-Tavard [2] equation is the key result used in the present work.

We employ Eq. (9) in conjunction with the density-functional formalism [21–23], in particular, the Kohn-Sham theory [22,23]. The merit of this approach is the simulation of an N-electron ground-state problem by one with N noninteracting electrons immersed in a common, one-body effective potential, viz. the Kohn-Sham [22,23] potential v_{KS}, leading to the prescription

$$U_{\mathrm{KS}}(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)=\sum_{i=1}^{N}v_{\mathrm{KS}}(\mathbf{r}_i) \;. \tag{10}$$

We take, for the wave function Φ_a,

$$\Phi_a(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)=A[\psi_1^{\mathrm{KS}}(\mathbf{r}_1)\psi_2^{\mathrm{KS}}(\mathbf{r}_2)\cdots\psi_N^{\mathrm{KS}}(\mathbf{r}_N)] \;, \tag{11}$$

where A is an antisymmetrizer, which in the present KS description gives a determinant, constructed *a posteriori*, out of the lowest lying, occupied KS orbitals ψ_i^{KS}, which themselves are the solutions of N single-particle KS equations:

$$[-\nabla^2/2+v_{\mathrm{KS}}(\mathbf{r})]\psi_i^{\mathrm{KS}}(\mathbf{r})=\varepsilon_i\psi_i^{\mathrm{KS}}(\mathbf{r}) \quad (i=1,2,\ldots,N) \;, \tag{12}$$

with

$$v_{\mathrm{KS}}(\mathbf{r})=-Z/r+\int d^3r'n(\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|+\delta E_{\mathrm{xc}}^{\mathrm{KS}}[n]/\delta n(\mathbf{r}) \;. \tag{13}$$

With the identification (10) of the many-body (effective) potential-energy operator U_{KS} in the KS picture, the KS equations (12) emerge out of a minimal property of $\langle\Phi_a|T+U_{\mathrm{KS}}|\Phi_a\rangle/\langle\Phi_a|\Phi_a\rangle$. The quantities ε_i in Eqs. (12) are the Lagrange multipliers that take into account the orthonormality of the KS orbitals. [T is the many-electron kinetic-energy operator $T\equiv\sum_{i=1}^{N}(-\nabla_i^2/2)$.] In Eq. (13), $n(\mathbf{r})=\sum_{i=1}^{N}|\psi_i^{\mathrm{KS}}(\mathbf{r})|^2$ gives the ground-state density of the system (that plays a central role in the density-functional theory [21–23]), of which the Kohn-Sham exchange-correlation energy $E_{\mathrm{xc}}^{\mathrm{KS}}$ is a universal (but hitherto explicitly unknown) functional.

It is desirable to use Eq. (9) in its given form (rather than evaluating the potential matrix elements separately), because Eq. (10), substituted in Eq. (9), results in cancellation of every other contribution to ΔU in Eq. (9), except that from $\mathbf{r}_\mu$. Thus the only term that survives inside the bracket in Eq. (9) is $v_{\mathrm{KS}}(\mathbf{r}_\mu+\mathbf{R}')-v_{\mathrm{KS}}(\mathbf{r}_\mu)$. This readily simplifies the expression for $F_\mu^{(1)}$, now expressible in terms of the μth orbital:

$$F_\mu^{(1)}(\mathbf{R},\mathbf{k})=(i/k)\int_0^R dR'\int d^3r\, \psi_\mu^{\mathrm{KS}*}(\mathbf{r})[v_{\mathrm{KS}}(\mathbf{r}+\mathbf{R}')-v_{\mathrm{KS}}(\mathbf{r})]\psi_\mu^{\mathrm{KS}}(\mathbf{r}+\mathbf{R})\ . \tag{14}$$

Note that $\mathbf{r}_\mu \equiv \mathbf{r}$ has now become a dummy integration variable, while the subscript μ on F as well as on ψ^{KS} denotes the *orbital state* index (such as 1*s*, 2*s*, etc.). The leading correction then emerges as a sum over the KS orbitals

$$J^{(1)}(q,\mathbf{k})=i(2\pi k)^{-1}\int_{-\infty}^{\infty} dR\, e^{-iqR}\int_0^R dR'\int d^3r \sum_{\mu=1}^{N} \psi_\mu^{\mathrm{KS}*}(\mathbf{r})[v_{\mathrm{KS}}(\mathbf{r}+\mathbf{R}')-v_{\mathrm{KS}}(\mathbf{r})]\psi_\mu^{\mathrm{KS}}(\mathbf{r}+\mathbf{R})\ , \tag{15}$$

$$J^{(1)}(q,\mathbf{k})=i(2\pi k)^{-1}\int_{-\infty}^{\infty} dR\, e^{-iqR}\int_0^R dR'\int d^3r[v_{\mathrm{KS}}(\mathbf{r}+\mathbf{R}')-v_{\mathrm{KS}}(\mathbf{r})]\Gamma_{\mathrm{KS}}^{(1)}(\mathbf{r}|\mathbf{r}+\mathbf{R})\ . \tag{16}$$

This last one expresses the correction in terms of the full, one-particle KS density matrix $\Gamma_{\mathrm{KS}}^{(1)}$. Equation (16) simplifies further into a sine transform,

$$J^{(1)}(q,\mathbf{k})=(1/\pi k)\int_0^{\infty} dR\, \sin(qR)\int_0^R dR'\int d^3r[v_{\mathrm{KS}}(\mathbf{r}+\mathbf{R}')-v_{\mathrm{KS}}(\mathbf{r})]\Gamma_{\mathrm{KS}}^{(1)}(\mathbf{r}|\mathbf{r}+\mathbf{R})\ , \tag{17}$$

which, evidently, is antisymmetric in q. The dependence of $J^{(1)}$ on the direction of the momentum transfer $\mathbf{k}$ comes through that on the direction of $\mathbf{R}=\mathbf{k}t$, which also is the support for $\mathbf{R}'$.

The one-body KS potential v_{KS} is the appropriate self-consistent, common effective potential, to be contrasted against the orbital-dependent "effective hydrogenic" $(-Z_{\mathrm{eff}}/r)$ potentials of Refs. [4–7]. Usage of a self-consistent theory is a very apt one, as has been recently emphasized by Issolah *et al.* [5], since it leads to a more realistic atomic potential than does the effective hydrogenic one.

For computation of the impulse profile, the spherically averaged IA-CP derived from the spherically averaged version of the electron momentum density $\gamma_{\mathrm{KS}}^{(0)}(\mathbf{p})=\sum_{i=1}^{N}|\chi_i^{\mathrm{KS}}(\mathbf{p})|^2$ is used herein, where $\chi_i^{\mathrm{KS}}(\mathbf{p})$ is the Fourier transform (FT) of $\psi_i^{\mathrm{KS}}(\mathbf{r})$. Of course, while the KS scheme yields, in principle, the exact coordinate-space ground-state electron density $n(\mathbf{r})$, it does not necessarily give the correct *momentum* density through the above prescription, as pointed out by Lam and Platzman [24]. This is due to the noninteracting model employed within the KS framework which subjects $\gamma_{\mathrm{KS}}^{(0)}(\mathbf{p})$ to an additional correction term [24]. It, however, turns out from the studies of Lam and Platzman [24] that the estimated corrections are only marginal (maximum relative correction, occurring at the peak of a CP, is typically less than 2%, as seen from the work of Harmalkar, Panat, and Kanhere [25]) and hence for consistency (in $J^{(0)}$ and $J^{(1)}$ computations) will not be taken into account here. Thus $J_{\mathrm{IA}}^{(0)}(q)=2\pi\int_{|q|}^{\infty}\tilde{\gamma}(p)p\,dp$, with $\tilde{\gamma}(p)=\int d\Omega_{\hat{\mathbf{p}}}\gamma_{\mathrm{KS}}^{(0)}(\mathbf{p})/(4\pi)$, the spherically averaged version of $\gamma_{\mathrm{KS}}^{(0)}(\mathbf{p})$. It must be noted that for the closed-shell atomic systems dealt with herein, the *total orbital* contribution to $\gamma_{\mathrm{KS}}^{(0)}(\mathbf{p})$ after summing over the magnetic substates will be spherically symmetric, and thus will not depend on m_l, the magnetic quantum number. Also, in the context of an atomic (or molecular) electron momentum density, what is experimentally tractable is essentially a spherically averaged information, as remarked above. The IA-CP thus obtained will be isotropic in q and monotone decreasing in $|q|$.

For the functional $E_{\mathrm{xc}}^{\mathrm{KS}}[n]$, we have taken a simple, local-density functional, viz. the Dirac-Slater X_α prescription $E_{\mathrm{xc}}^{\mathrm{KS}}[n]\approx-(9\alpha/8)(3/\pi)^{1/3}\int d^3r\, n^{4/3}(\mathbf{r})$ with $\alpha=\frac{2}{3}$ as recommended by Gáspár [26] and Kohn and Sham [22]. Of course, one may choose different kinds of exchange-correlation functionals that are replete in the literature [23], but the results presented herein with the above local form are not seen to alter qualitatively (and also appreciably quantitatively) after employing different approximate forms for $E_{\mathrm{xc}}^{\mathrm{KS}}[n]$. What is more crucial, as it turns out, is the use of an effective SCF potential that has the correct limiting $(r\to 0)\sim -Z/r$ and asymptotic $(r\to\infty)\sim -1/r$ behavior [5,14,27,28], satisfaction of which was made sure in this work by incorporating a correction due to Latter [27,28].

For numerical computations involved in Eq. (17), we have employed prolate spheroidal coordinates (ξ,η,φ) with the polar axis, which also is the axis of quantization for specifying the magnetic substates, being chosen along the momentum transfer $\mathbf{k}$, facilitating elimination of the azimuthal angle φ. Integrations over ξ (1 to ∞) and η (-1 to 1) were performed using an accurate Gaussian quadrature coupled with an accurate interpolator. Finally, the sine FT was carried out using the well-known FILON routine. In the following section, we summarize our numerical results on $J^{(1)}(q,\mathbf{k})$ and compare them with the corresponding IA-CP, $J_{\mathrm{KS}}^{(0)}(q)$, for He, Ne, Ar, and Kr. To further gauge the significance of the leading correction, we choose different momentum transfers for scattering from specific atomic orbitals. Also, whenever relevant (and possible), a comparison between the results obtained herein with those in the previous works will also be made.

III. RESULTS AND DISCUSSION

The results for the leading antisymmetric correction $J^{(1)}(q,\mathbf{k})$ are portrayed in Figs. 1–6. Figure 1 gives the percent relative deviation of total atomic $J^{(1)}$ from the respective $J_{\mathrm{IA}}^{(0)}$, i.e., $R\%=J^{(1)}(q,\mathbf{k})\times 100/J_{\mathrm{IA}}^{(0)}(q)$, versus q for the inert atoms He–Kr. For a direct comparison with Bell's [4] important work, we have chosen $k=5Z$ a.u. [We choose $J_{\mathrm{IA}}^{(0)}(q)$ here for comparison and not $J(q,\mathbf{k})\approx J_{\mathrm{IA}}^{(0)}(q)+J^{(1)}(q,\mathbf{k})$, in contrast with Bell, who has chosen $J(q,\mathbf{k})$ in the denominator.] A suitable summation over the orbital-magnetic substates for p and d or-

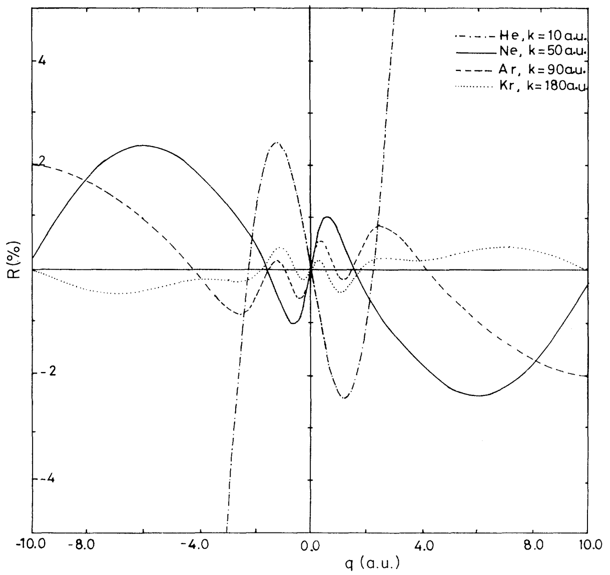

FIG. 1. Relative deviation $[R\ (\%)] = 100 J_k^{(1)}(q)/J_{\text{IA}}^{(0)}(q)$ for the atoms He, Ne, Ar, and Kr. The momentum transfers k are chosen to be $k=5Z$ a.u.

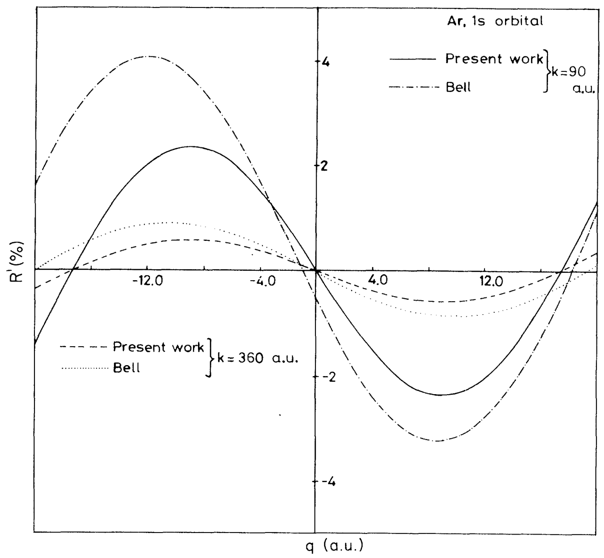

FIG. 2. Relative deviation $[R'\ (\%)] = 100 J_{k,1s}^{(1)}(q)/J_{\text{IA},1s}^{(0)}(q)$ for the Ar 1s orbital obtained in the present work compared against $[R'\ (\%)] = 100[J_{k,1s}^{\text{Born}}(q) - J_{\text{IA}}^{(0)}(q)]/J_{\text{IA}}^{(0)}(q)$ values and Bell [4], within the effective hydrogenic and Born regime (see text for further details).

bitals was carried out as $J_p^{(1)} = (\frac{1}{3})J_{p_0}^{(1)} + (\frac{2}{3})J_{p_{\pm 1}}^{(1)}$ and $J_d^{(1)} = (\frac{1}{5})J_{d_0}^{(1)} + (\frac{2}{5})[J_{d_{\pm 1}}^{(1)} + J_{d_{\pm 2}}^{(1)}]$. This makes the total first-order correction [denoted hereafter by $J_k^{(1)}(q)$] "isotropic" in **k**. The overall trends of R % for Ne and Ar are seen to match very well with their counterparts in Ref. [4] (Fig. 4, Ref. [4]), for $|q| < 5$ a.u., beyond which the asymmetry in R % in the plots of Bell is conspicuous. The present curves are, of course, manifestly antisymmetric as they represent only the leading first-order corrections (while Bell's work gives corrections, within the effective hydrogenic realm, to all orders in $1/k$). For He and Kr, the R % values are, respectively, rather large and small, a fact attributed to two causes: small $J_{\text{IA}}^{(0)}(q)$ and comparatively small k ($=10$ a.u.) for He, as opposed to their corresponding large values ($k=180$ a.u.) for Kr. For all the plots except He, the first maximum in R % is seen to occur for a positive (albeit small) q, which is the consequence of the dominant $p_0 \equiv p_z$ contribution (cf. Ref. [2], Figs. 3 and 4 therein). For a closer comparison with Bell's results, we choose the example of the Ar 1s orbital and compute the ratios $[R'\ (\%)] = 100 J_{k,1s}^{(1)}(q)/J_{\text{IA},1s}^{(0)}(q)$, obtained within the present regime, and compare with their corresponding values defined on a similar footing through the index $[R'\ (\%)] = 100[J_{k,1s}^{\text{Born}}(q) - J_{\text{IA}}^{(0)}(q)]/J_{\text{IA}}^{(0)}(q)$, obtained via a systematic recipe due to Bell [4], within the effective hydrogenic theory and the first Born approximation. The momentum transfers k are chosen to be $k=90$ and 360 a.u. with the q range -20 a.u. $\leq q \leq 20$ a.u. This comparison is made in Fig. 2, which brings out the similarity between the trends of R' under the two regimes and reveals that for the larger momentum transfer (360 a.u.), Bell's results for R' also tend to be fairly antisymmetric and the similarity in the trends of both the R' values is even closer. This reiterates that $J_k^{(1)}$, orbitalwise, gives a decent estimate of deviation from the impulse profile for wide range of q values.

Since the KS-SCF potential, by construction, gives a central field, the orbital angular momentum l and the related orbital magnetic quantum numbers m_l are good quantum numbers. This enables one to obtain different *orbitalwise* contributions to $J_k^{(1)}(q)$ and further compare it with the respective *orbital* IA-CP $J_{\text{IA}}^{(0)}(q)$. For representative cases, we have chosen the momentum transfers as

Orbital	k (a.u.)
He 1s	2
Ne $2p_0 \equiv 2p_z$	20
Ar $2p_{\pm 1}$	10
Kr $3d_{(m)}$, $m=0, \pm 1, \pm 2$	50

These results are presented in Figs. 3–6. For the orbital impulse approximation profiles presented in these Figures, we choose the normalization $\int^{\infty} J_{\text{IA}}^{(0)}(q)dq = \frac{1}{2}$. Interestingly enough, the $J^{(1)}$ plots for Ne $2p_0$ and Kr $3d_{\pm 1}$ orbitals are the only ones that have a global maximum for $q>0$, whereas all other cases exhibit a global minimum for $q>0$, conforming to the Gasser-Tavard [2] rule that for given l and m_l values (the leading contributions to $-$) the Compton defects have a positive sign for odd values of $l + |m_l|$ and a negative one for their even-valued combinations. Thus the orientation of orbitals as well as their mutual overlap both seem to play a significant role in deciding the sign of the defect. It is worth noting further that for krypton, the $3d_{\pm m}$ orbital $J_k^{(1)}(q)$ contributions

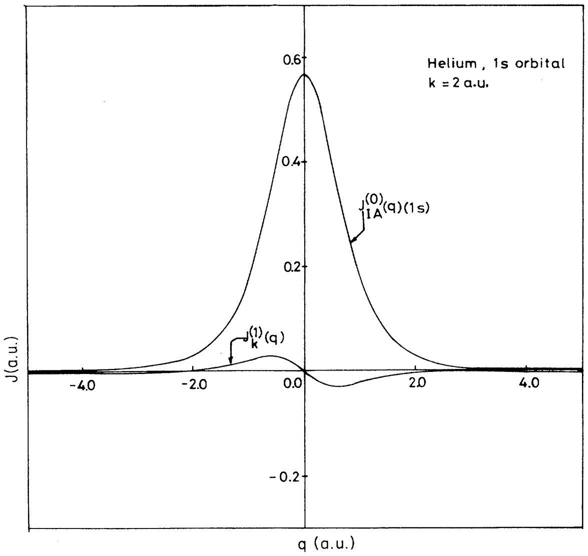

FIG. 3. Leading first-order correction $J_k^{(1)}(q)$ to the helium 1*s* orbital, compared with its corresponding zero-order, spherically averaged impulse Compton profile $J_{\rm IA}^{(0)}(q)$ (1*s*) (the latter is normalized to $\frac{1}{2}$ electrons). The momentum transfer $k=2$ a.u.

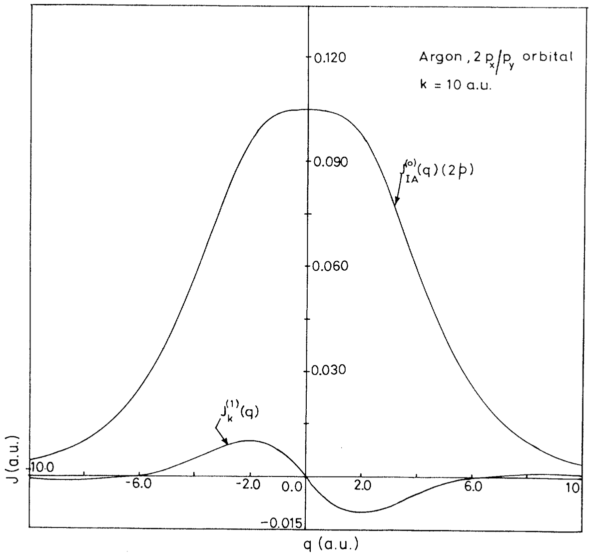

FIG. 5. $J_{\rm IA}^{(0)}(q)(2p)$ and $J_k^{(1)}(q)$ ($k=10$ a.u.) for the $2p_x/2p_y$ orbitals of argon.

emerge with decreasing significance with increasing $m=0,1,2$, an effect that may be traced back to the degree of polarization of an orbital along the direction of the momentum transfer.

Choosing different forms of gradient corrections, and further, addition of correlation to the X_α exchange functional is not seen to alter the above inferences qualitatively. For instance, our tests on the neon $J^{(0)}(q)$ and $J_k^{(1)}(q)$ results showed that they changed by less than three percent and one percent, respectively, after employing some sample gradient corrections and correlation functionals cited in Ref. [23]. As remarked above, the Latter correction [27,28], which mimics the real atomic potential asymptotically ($\sim -1/r$), overrides the exchange-correlation potential for large r values. In the present scheme, the construction of a Kohn-Sham "determinant" is essentially perforce, in the absence of any simple alternative prescription. On the affirmative side, we note that the KS theory gives a one-body self-consistent potential (duly rectified, self-consistently, for the correct asymptot-

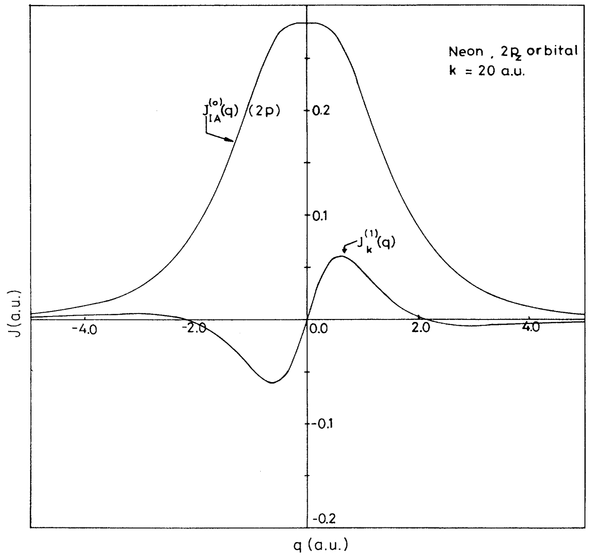

FIG. 4. $J_{\rm IA}^{(0)}(q)(2p)$, the spherically averaged orbital Compton profile for the 2*p* orbital and $J_k^{(1)}(q)$ ($k=20$ a.u.) for the $2p_z$ orbital of neon.

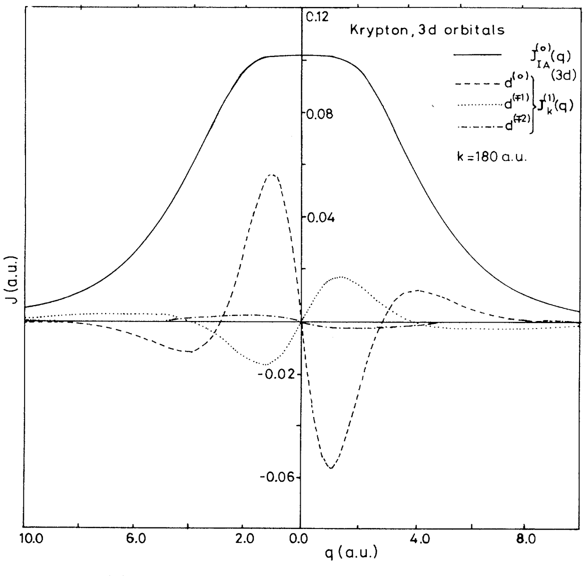

FIG. 6. $J_{\rm IA}^{(0)}(q)(3d)$ and $J_k^{(1)}(q)$ ($k=50$ a.u.) for the 3*d* orbitals of krypton, for different m_l values $=0,\pm1,\pm2$. Note that the signs of the peak corrections for positive q values are, respectively, negative, positive, and negative (see text for further details).

ic behavior), which takes into account the many-body effects of exchange and correlation, practically, albeit in an approximate way. The confluence of the Gasser-Tavard theory with the KS description is thus expected to lead to a better simulation of the actual one-electron atomic potential, in contradistinction with the effective hydrogenic picture.

We point out here that $J(q,\mathbf{k})$ can be calculated exactly (within the Born regime) with rather poor wave functions (scaled hydrogenic ones), whereas with better quality wave functions only the first correction, viz. $J^{(1)}$, is available, which estimates the correction to the impulse result only for moderately large momentum transfers to the electron within the Compton scattering process. For relatively small momentum transfers one must take into account the higher order corrections. However, computation of these becomes increasingly difficult. It would also be worthwhile to look at the effect of an incorporation of the self-interaction correction (SIC) [29,12] into the present framework, but then the SCF potential becomes orbital dependent, losing its simplicity. These two tasks will be pursued in the University of Poona's Physics Laboratory at Pune. For extremely large momentum transfers, one could make an asymptotic ($y \rightarrow \infty$) expansion of Bell's [4] functions $F_1(x,y)$, $F_2(x,y)$, and $F_3(x,y)$ (cf. Ref. [4]), in which case $F_1 \sim (\pi Z_{\rm eff})^{-1}(1+\pi n/y-2\pi^2 n^2/(3y^2)+\cdots)$, $F_2 \sim 1$, while F_3 is manifestly in the desired form of a power series in $1/y$. Here y and x are suitably scaled versions of k and q, respectively, and n is the principal quantum number of a given orbital. Thus an expansion of the Compton profile as a power series in $1/y$ (or $1/k$) is indeed obtainable; however, this being an effective hydrogenic scenario, a perfect antisymmetry/symmetry characteristic (as a function of x or q) of a given correction term is not guaranteed. Note, incidentally, that for extremely low momentum transfers, the Born approximation itself becomes questionable. It is thus indeed gratifying that the present simple and straightforward approach yields decent estimates for the leading part of the correction to the atomic impulse Compton profile for moderate ($k \sim 5Z$ a.u.) momentum transfers.

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