Compton-scattering studies on the helium atom beyond the impulse approximation

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(Received 14 December 1981)

A series expansion of the Born propagator allows one to represent successive corrections to the impulse approximation and exhibits their relative dependence in momentum transfer k and the Compton (target-structure) parameter q. The proposed treatment provides a physical interpretation of the observed Compton defects. It is shown here to be in close agreement with high-energy-electron-impact spectroscopy measurements obtained for helium atoms.

I. THEORY

Under Born assumptions, the following relationship

$$J(q,k)=kf(\Delta E,k)=k\left\langle a\left|\sum_{\mu=1}^{N}\delta(qk+i\vec{k}\cdot\vec{\nabla}_\mu-K+E_a)\right|a\right\rangle \tag{1}$$

has been shown[1-4] to represent the Compton profile (CP) obtained by x-ray, γ-ray, and the electron-impact experiments. The N electrons of the target system are in a given initial eigenstate $|a\rangle$ of the Hamiltonian K, for which $K|a\rangle=E_a|a\rangle$. k and ΔE, respectively, represent the momentum and the energy lost by the incident particle and hence transferred to the target $q=\Delta E/k-k/2$ (in a.u.) being the well-known Compton parameter. In this expression, indirect Compton scattering effects[1] are assumed negligible. For large momentum transfers Eq. (1) becomes independent of k and reduces to the impulse approximation (IA)

$$J^{\mathrm{IA}}(q,k)=J^{\mathrm{IA}}(q)=k\left\langle a\left|\sum_{\mu}\delta(qk+i\vec{k}\cdot\vec{\nabla}\mu)\right|a\right\rangle$$

$$=\left\langle a\left|\sum_{\mu}\delta\left[q+i\frac{\vec{k}}{k}\cdot\vec{\nabla}_\mu\right]\right|a\right\rangle=\left\langle a\left|\sum_{\mu}\delta(q+i\vec{u}\cdot\vec{\nabla}_\mu)\right|a\right\rangle, \tag{2}$$

where $\vec{u}$ is the unit vector along the $\vec{k}$ direction. Equation (2) shows clearly that $J^{\mathrm{IA}}(q)$ depends only on the physical properties of the scatterer, i.e., its momentum distribution $\rho(\vec{p})$:

$$J^{\mathrm{IA}}(q)=\int d\vec{p}\rho(\vec{p})\delta(\vec{u}\cdot\vec{p}-q)=\int\int_{p_z=q}dp_x dp_y\rho(\vec{p}), \tag{3}$$

where the integration is performed in a plane perpendicular to $\vec{u}$. But it has been found that a Doppler broadening effect of the Compton peak is not sufficient to explain the observed spectra at all momentum-transfer values, especially the typical asymmetry found in Compton profiles.[5-8] Equation (2) represents only the leading term of a perturbation expansion in inverse powers of momentum transfers

$$J(q,k)=\sum_{n=0}^{\infty}k^{-n}K_n(q). \tag{4}$$

In such an expansion, all coefficients $K_n(q)$ are characteristic of the target structure. For these terms, some general properties are derived in the Appendix. A leading contribution to the Compton defect[9] comes from the first antisymmetric correction $A(k,q)=K_1(q)/k$, here corresponding to

$$K_1(q)=\frac{1}{2\Pi}\int_{-\infty}^{\infty}dx\left\langle a\left|\sum_{\mu}\exp[ix(q-D_\mu)]U_1^\mu(x)\right|a\right\rangle \quad (5)$$

with

$$\begin{aligned}U_1^\mu(x)&=-i\int_0^x dx'\exp(ix'D_\mu)(K-E_a)\exp(-ix'D_\mu)\\&=-i\int_0^x dx'[K(x')-E_a]\end{aligned} \quad (6)$$

after use of the translation operator $D_\mu=-i\vec{u}\cdot\vec{\nabla}_\mu$. The transformation

$$[K(x')-E_a]\,|\,a\rangle=i\left[\int_0^{x'}dx''\exp(ix''D_\mu)[D_\mu,K]\exp(-ix''D_\mu)\right]|\,a\rangle \quad (7)$$

gives for Eq. (6) a slightly different expression:

$$\begin{aligned}U_1^\mu(x)&=\int_0^x dx'\int_0^{x'}dx''\exp(ix''D_\mu)[D_\mu,K]\exp(-ix''D_\mu)\\&=\int_0^x dx''(x-x'')\exp(ix''D_\mu)[D_\mu,K]\exp(-ix''D_\mu)\,.\end{aligned} \quad (8)$$

The commutator $[D_\mu,K]=[D_\mu,V]$ is applied to the total potential V of the target. It includes all electric fields due to the remaining (electrons and nuclei) particles of the scatterer and acts upon the ejected electrons. Since an exact calculation of $K_1(q)$ using Eqs. (5) and (8) is not easily possible, an approximation is here proposed. The integral occurring in Eq. (8) is carried out with the three-points integration method of Simpson, after some algebra, $K_1(q)$ exactly reduces to

$$\begin{aligned}K_1(q)&=\frac{1}{2\Pi}\int_{-\infty}^{\infty}dx\frac{x^2}{6}\left\langle a\left|\sum_{\mu}\exp[ix(q-D_\mu)][D_\mu,K]\right|a\right\rangle\\&=-\frac{1}{6}\frac{d^2}{dq^2}\left[\left\langle a\left|\sum_{\mu}\delta(q+i\vec{u}\cdot\vec{\nabla}_\mu)[-i\vec{u}\cdot\vec{\nabla}_\mu,V]\right|a\right\rangle\right]\,.\end{aligned} \quad (9)$$

Such a result can be shown to give exact analytical expressions for all finite odd sum rules relative to $J(q,k)$. It simply assumes a "continuous" behavior of the Hamiltonian $K(x)$ during the scattering process. For the antisymmetric correction

$$A(k,q)=-\frac{1}{6k}\frac{d^2}{dq^2}\left[\left\langle a\left|\sum_{\mu}\delta(q+i\vec{u}\cdot\vec{\nabla}_\mu)[-i\vec{u}\cdot\vec{\nabla}_\mu,V]\right|a\right\rangle\right] \quad (10)$$

explicit calculation requires generally a momentum representation for the ground-state wave function. Closure properties of plane waves $(2\Pi)^{-3/2}\exp(i\vec{p}\cdot\vec{r}_\mu)$ eigenstates of the translation operator $D_\mu=-i\vec{u}\cdot\vec{\nabla}_\mu$, allow such a representation. Equation (10) represents a successful simplification with respect to earlier studies[10,11] established for hydrogenic systems, for which exact first Born calculations are available.[2,12,13] It will be evaluated here for the helium atom without any further approximation and compared with the effective hydrogenic theory of Mendelsohn and Bloch[14] and some precise measurements[15] performed for several values of momentum transfer. A similar treatment can be developed to estimate the second-order (symmetric) correction $S(q,k)=K_2(q)/k^2$. It is found to generalize the results already established[10] for the specific case of hydrogenic systems.

II. COMPTON DEFECT FOR THE HELIUM ATOM

A preliminary investigation using Eq. (10) is reported here where the simple $|1S\overline{1S}|$ determinant has been used to represent the helium-atom ground-state wave function. In a first step, a Slater orbital (screening parameter $\zeta=\frac{27}{16}$) has been used to represent the $1S$ atomic orbital. Using various analytic Hartree-Fock atomic orbitals[16,17] changed the results typically 10%, and hence will not be discussed here.

The $A(q,k)$ antisymmetric correction is thus given by an analytic expression containing various contributions. Their relative importance will be discussed. Since V represents the total potential energy of the target atom, the commutator

$$[-i\vec{u}\cdot\vec{\nabla}_\mu, V]=\frac{i}{a_0}\left[\left[\vec{u}\cdot\vec{\nabla}_\mu,\frac{Z}{r_\mu}\right]-\sum_{\nu\neq\mu}\left[\vec{u}\cdot\vec{\nabla}_\mu,\frac{1}{r_{\mu\nu}}\right]\right] \tag{11}$$

involves electric field contributions from the nucleus and the remaining electrons ($a_0=1$, Bohr radius in a.u.). The nuclear contribution to the odd correction will be labeled A_Z, with $Z=2$ for the case of helium. Its explicit calculation, with the dimensionless parameter $Q=q/\zeta$, results in

$$A_Z(Q,k)=\frac{-32Z}{3\Pi\zeta(ka_0)}\frac{\arctan Q-3Q/4}{(1+Q^2)^3}\,, \tag{12}$$

and an exact A_E result now corresponds to the total potential V:

$$A_E(Q,k)=\frac{-32}{3\Pi\zeta(ka_0)}\frac{1}{(1+Q^2)^3}\left[(\arctan Q-3Q/4)+\left[\arctan(Q/3)-\frac{3Q^7+73Q^5+361Q^3+675Q}{2916[1+(Q/3)^2]^3}\right]\right], \tag{13}$$

which exhibits in its first term, the contribution A_1 due to a single proton ($Z=1$).

This odd A_E correction is represented in Fig. 1 for a number of ka_0 values together with the impulse profile given here by

$$J^{\mathrm{IA}}(Q)=\frac{16}{3\Pi\zeta}\frac{1}{(1+Q^2)^3}\,. \tag{14}$$

In fact, this first antisymmetric correction is universely proportional to k^{-1}, with a multiplicative function $K_1(q)$ given in Eq. (10) and mainly characteristic of the scattering system via its ground-state wave function $|a\rangle$.

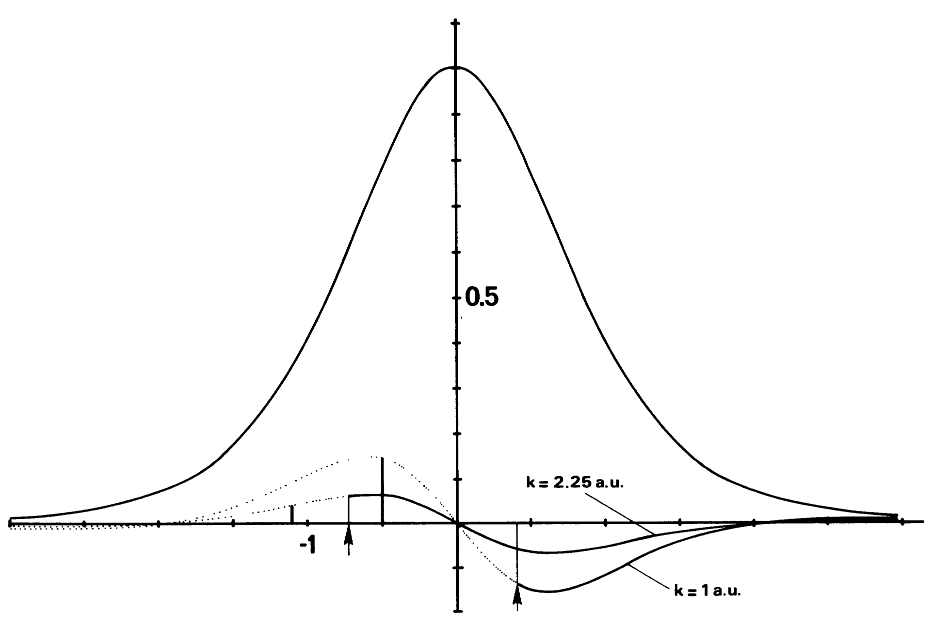

FIG. 1. Antisymmetric corrections for various momentum transfers k compared with the helium impulse profile $J^{\mathrm{IA}}(q)$.

III. OTHER CORRECTIONS

The first even correction $S(k,q)=K_2(q)/k^2$ to $J^{\mathrm{IA}}(q)$ is estimated here with the following expression:

$$S(k,Q)=\frac{16Z^2}{3\Pi\zeta(ka_0)^2}\frac{1}{[1+Q^2]^3}\left[\frac{11}{4}+\frac{1}{4Q^2}-\arctan Q\left[\frac{1}{4Q^3}+\frac{3}{Q}+Q\right]\right.$$
$$\left.+\frac{15}{8}\sum_{n=1}^{\infty}\frac{2^{2n}n!(n-1)!}{(2n+1)!(2n-1)!}\left[\frac{Q^2}{1+Q^2}\right]^n\right], \tag{15}$$

established previously[10] for 1s hydrogenic orbitals.

Indirect Compton scattering effects[1] are known to contribute to the inelastic scattering factor at small momentum transfers. Within the impulse assumptions, their calculation

$$J^{\mathrm{ind}}(q,k)\simeq\left\langle a\left|\sum_{\mu\neq\nu}\exp(i\vec{k}\cdot\vec{r}_{\mu\nu})\delta(q+i\vec{u}\cdot\vec{\nabla}_\mu)\right|a\right\rangle \tag{16}$$

gives explicitly, with the dimensionless variables $Q=q/\zeta$ and $\kappa=k/\zeta$,

$$J^{\mathrm{ind}}(Q,\kappa)=\frac{16}{\Pi\zeta\Delta^2}\frac{1}{(1+\kappa^2/4)^2}\left[\frac{1}{1+Q^2}+\frac{1}{1+Q^2+\Delta}-\frac{2}{\Delta}\ln\frac{1+Q^2+\Delta}{1+Q^2}\right]$$

$$=J^{\mathrm{ind}}(\Delta,\kappa)=\frac{16}{\Pi\zeta\Delta^2}\frac{1}{(1+\kappa^2/4)^2}\left[\frac{1}{1+\left[\frac{\kappa}{2}-\frac{\delta}{2\kappa}\right]^2}+\frac{1}{1+\left[\frac{\kappa}{2}+\frac{\Delta}{2\kappa}\right]^2}-\frac{2}{\Delta}\ln\frac{1+\left[\frac{\kappa}{2}+\frac{\Delta}{2\kappa}\right]^2}{1+\left[\frac{\kappa}{2}-\frac{\Delta}{2\kappa}\right]^2}\right] \tag{17}$$

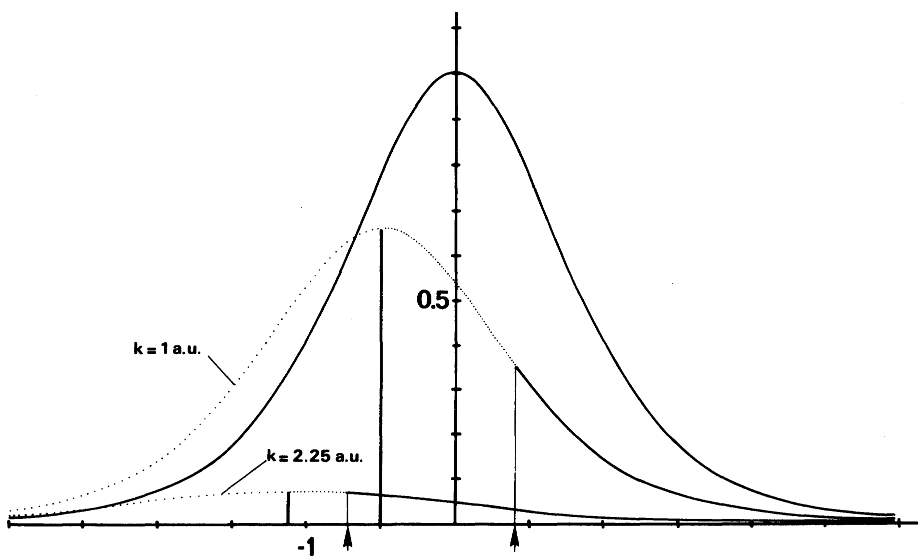

FIG. 2. Rough estimate of the indirect Compton scattering effect $J^{\mathrm{ind}}(q)$ for various momentum transfers k, compared with the helium impulse profile.

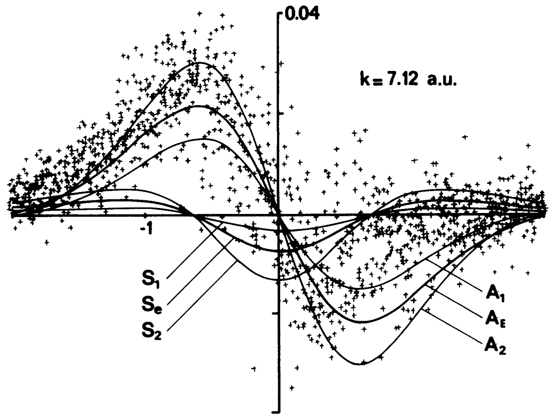

FIG. 3. Abscissas in $q=\Delta E/k-k/2$ (q-Compton parameter); A_1, A_2, A_E: antisymmetric corrections (respectively, for $Z=1$, $Z=2$, and exact potential V, approximately fitted by $Z=1.48$); S_1, S_2, S_e: symmetric corrections $Z=1$, $Z=2$, $Z=1.48$; $\Delta J=A_E+S_e$ estimated "defect"; + experimental results; Δ Mendelsohn's calculation; $J^{\mathrm{ind}}(q)$=rough estimate of indirect Compton-scattering effects.

and

$$\Delta=2\Delta E/\zeta^2=2\kappa Q+\kappa^2 \,. \tag{18}$$

Such a function generally satisfies

$$J^{\mathrm{ind}}(\Delta,\kappa)=J^{\mathrm{ind}}(-\Delta,\kappa)$$

and behaves symmetrically with respect to the elastic peak. As shown in Fig. 2, significant values are found only for small momentum transfers. Physically speaking, the impulse approximation does not represent a correct approach for the description of indirect Compton scattering but rather gives a rough estimate of the validity conditions for the binary-encounter approximation.

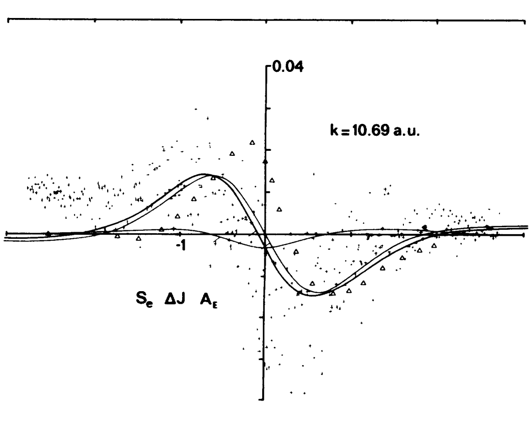

FIG. 4. See Fig. 3.

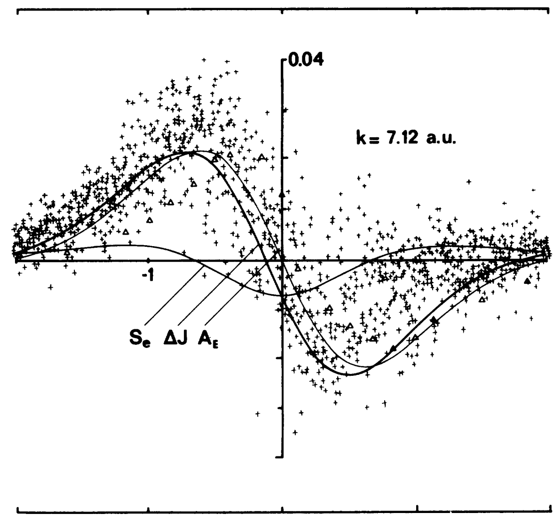

FIG. 5. See Fig. 3.

IV. THEORETICAL COMPTON DEFECT VERSUS EXPERIMENTAL RESULTS

For $ka_0=7.12$ a.u. all the different contributions discussed above are shown in Fig. 3. The exact A_E calculation [Eq. (13)] may thus be compared with A_1 (complete screening of the nucleus) and A_2 (total unscreening, $Z=2$) given by Eq. (12). A_2 and A_1 constitute approximate upper and lower bounds for A_E. The A_E slope at the origin ($Q=0$) is given by

$$\frac{-8}{3\Pi\zeta(ka_0)}\left[1+\frac{297}{729}\right]=\frac{-8}{3\Pi\zeta(ka_0)}1.407 \tag{19}$$

and corresponds to an effective $Z^*=1.407$.

A better agreement is found for $Z^*=1.48$ when A_Z is used to fit Eq. (13). In fact, Fig. 3 shows clearly both (nuclear and electronic) electric field

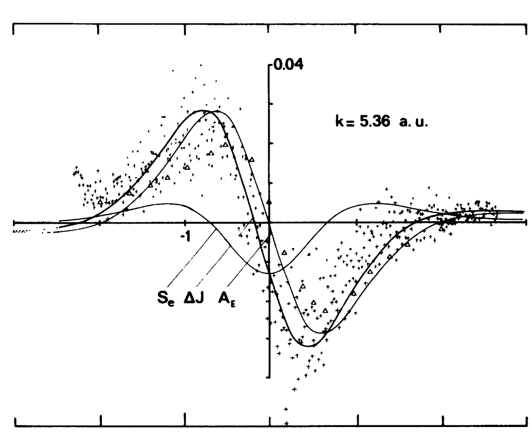

FIG. 6. See Fig. 3.

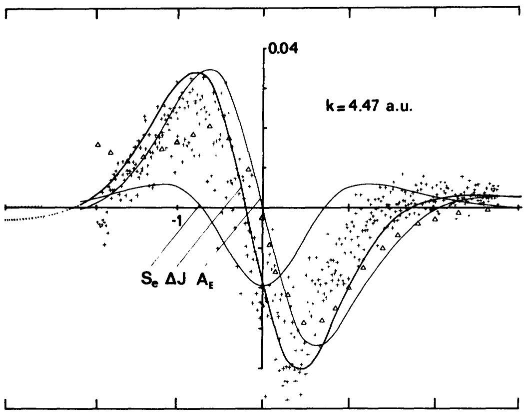

FIG. 7. See Fig. 3.

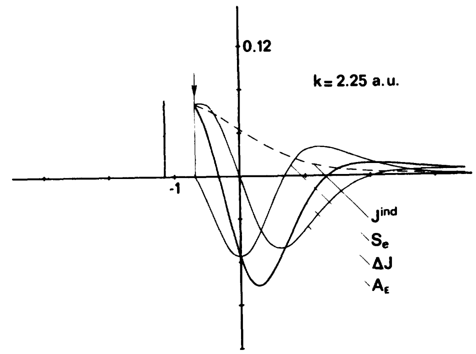

FIG. 8. See Fig. 3.

effects occurring in odd Compton profile corrections. The even correction has been approximated by Eq. (15). Use of $Z=2$, $Z=1$, and $Z^{*}=1.48$, respectively, gives S_2, S_1, and S_e, with corresponding curves shown in Fig. 3.

Figures 4 through 7 summarize the previous calculations for A_E, S_e, and their sum $\Delta J=A_E+S_e$, given here in reference to a number of experimental results obtained for $ka_0=10.69$, 7.12, 5.36, and 4.47, respectively. The crosses correspond to a complete set of independent measurements[15] from which an exact impulse Compton profile calculated by Benesch[18] has been subtracted. An excellent agreement is observed with Mendelsohn's theory and all experimental results, especially for cases where J^{ind} is weak.

For $ka_0=2.25$ (Fig. 8), the peak has its maximum near the ionization threshold and indirect Compton scattering effects are comparable to A_E and S_e, hence non-negligible differences with experimental results would be expected. However, the Compton defect remains negative, i.e., the shift of the maximum is expected to be towards smaller energy losses, and at low momentum transfers there is no theoretical evidence for a positive defect in the energy-loss spectra of helium. As a conclusion, refinements in the experimental measurements may make it possible to obtain separately the even and the odd contributions to the total Compton profile, these contributions independently being characteristic of the target structures.

ACKNOWLEDGMENTS

The authors wish to thank Professor M. Rouault and Dr. A. Lahmam-Bennani, for their helpful comments.

APPENDIX

In a time-dependent representation (or simply after a Fourier transform of the Dirac δ function), Eq. (1) can be rewritten as

$$J(q,k)=\frac{k}{2\pi}\int_{-\infty}^{\infty}dt\left\langle a\left|\sum_{\mu}\exp\left[itk\left[q+i\vec{\mu}\cdot\vec{\nabla}_{\mu}-\frac{K-E_a}{k}\right]\right]\right|a\right\rangle$$

$$=\frac{1}{2\pi}\int_{-\infty}^{\infty}dx\left\langle a\left|\sum_{\mu}\exp\left[ix\left[q+i\vec{u}\cdot\vec{\nabla}_{\mu}-\frac{K-E_a}{k}\right]\right]\right|a\right\rangle$$

$$=\frac{1}{2\pi}\int_{-\infty}^{\infty}dx\left\langle a\left|\sum_{\mu}\exp[ix(q+i\vec{u}\cdot\vec{\nabla}_{\mu})]\mathscr{U}_{\mu}(x,k)\right|a\right\rangle .$$

$D_{\mu}=-i\vec{u}\cdot\vec{\nabla}_{\mu}$ being the translation operator along the $\vec{u}$ direction, the unitary operator $\mathscr{U}_{\mu}(x,k)$ has the following expressions:

$$\mathscr{U}_\mu(x,k)=\exp(ixD_\mu)\exp\left[-ix\left[D_\mu+\frac{K-E_a}{k}\right]\right]$$

$$=1-\frac{i}{k}\int_0^x \exp(ix'D_\mu)(K-E_a)\exp(-ix'D_\mu)U_\mu(x',k)dx'$$

$$=\sum_{n=0}^{\infty}\mathscr{V}_n^\mu(x,k)=\sum_{n=0}^{\infty}\frac{1}{k^n}U_n^\mu(x)\ .$$

Since $U_0^\mu(x)=1$ and

$$U_{n+1}^\mu(x)=-i\int_0^x \exp(ix'D_\mu)(K-E_a)\exp(-ix'D_\mu)U_n^\mu(x')dx' \quad \text{for } n>0\ ,$$

all successive terms no longer depend on the momentum transfer k. Hence,

$$J(q,k)=\sum_{n=0}^{\infty}\frac{1}{k^n}\left[\frac{1}{2\pi}\int_{-\infty}^{\infty}dx\langle a \mid \sum_\mu \exp[ix(q+i\vec{u}\cdot\vec{\nabla}_\mu)]U_n^\mu(x)\right]|a\rangle$$

$$=\sum_{n=0}^{\infty}\frac{1}{k^n}K_n(q)\ . \tag{A1}$$

From the previous definitions, the properties

$$U_n^\mu(x)=(-1)^n U_n^\mu(-x)\ , \tag{A2}$$

$$K_n(q)=(-1)^n K_n(-q)\ ,$$

are derived easily. $K_0(q)$ simply represents the impulse Compton profile:

$$K_0(q)=J^{\mathrm{IA}}(q)=\left\langle a \left| \sum_\mu \delta(q+i\vec{u}\cdot\vec{\nabla}_\mu)\right| a\right\rangle\ .$$

The first-order correction $A(k,q)=K_1(q)/k$ is thus found to exhibit an odd behavior, the second-order correction $S(k,q)=K_2(q)/k^2$ being an even function of the Compton parameter q.

Of course, a truncated expansion of (A1) will not be sufficient to reproduce discrete excitations being observed. However, the first three terms are shown to be sufficient to reproduce exactly the four finite sum rules for $J(q,k)$ or, alternatively, for the corresponding generalized oscillator strengths.[19]

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