

³S. Klarsfeld, *Nuovo Cimento Lett.* **2**, 548 (1969); *Nuovo Cimento Lett.* **3**, 395 (1970). As a matter of fact, such results appear to be special cases of the so-called rules restricting the complexity of angular distributions in reactions which involve photons. See, for instance, C. N. Yang, *Phys. Rev.* **74**, 764 (1948); and M. Morita, A. Sugie, and S. Yoshido, *Prog. Theor. Phys.* **12**, 713 (1954).

⁴W. Zernik, *Phys. Rev.* **135**, A51 (1964).

⁵R. A. Fox, R. M. Kogan, and E. J. Robinson, *Phys. Rev. Lett.* **26**, 1416 (1971).

⁶G. Breit and H. A. Bethe, *Phys. Rev.* **93**, 888 (1954).

⁷See A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton U.P., Princeton, N. J., 1957); or D. M. Brink and G. R. Satchler, *Angular Momentum* (Oxford U.P., London, 1962).

⁸W. Zernik and R. W. Klopfenstein, *J. Math. Phys.* **6**, 262 (1965).

⁹W. Zernik, *Phys. Rev.* **176**, 420 (1968).

¹⁰In a subsequent experiment R. M. Kogan, R. A. Fox, G.

T. Burnham, and E. J. Robinson [*Bull. Am. Phys. Soc.* **16**, 1411 (1971)] also observed the two-photon ionization of atomic cesium by the second harmonic of the ruby laser.

¹¹S. Klarsfeld and A. Maquet, *Phys. Rev. Lett.* **29**, 79 (1972).

¹²For $N=2$, $N=3$, and an initial s state, the angular distribution and the maximum allowed ratio σ'/σ have been obtained in a somewhat different way by P. Lambropoulos [*Phys. Rev. Lett.* **28**, 585 (1972)].

¹³Cross sections for N -photon ionization of s states with circularly polarized light have been discussed very recently by P. Lambropoulos [*Phys. Rev. Lett.* **29**, 453 (1972)].

¹⁴H. B. Bebb, *Phys. Rev.* **149**, 25 (1966); *Phys. Rev.* **153**, 23 (1967).

¹⁵E. J. Robinson and S. Geltman, *Phys. Rev.* **153**, 4 (1967).

¹⁶L. P. Rapoport, B. A. Zon, and L. P. Manakov, *Zh. Eksp. Teor. Fiz.* **55**, 924 (1968) [*Sov. Phys.-JETP* **28**, 480 (1969)]; *Zh. Eksp. Teor. Fiz.* **56**, 400 (1969) [*Sov. Phys.-JETP* **29**, 220 (1969)].

Compton Scattering of High-Energy Electrons from Helium*†

H. F. Wellenstein and R. A. Bonham

Department of Chemistry, Indiana University, Bloomington, Indiana 47401

(Received 19 June 1972; revised manuscript received 5 September 1972)

The electron-impact energy-loss spectrum of He at a scattering angle of 7° has been measured with an energy resolution of 2.7 eV full width at half-maximum using 25-keV incident electrons. A binary-encounter approximation was used to obtain the electron Compton profile from the cross-section differential with respect to both the energy loss of the incident electron and solid angle of the scattered electron $d^2\sigma/dE d\Omega$. The electron Compton profile was corrected for interference scattering from pairs of target electrons and exchange. It was then compared to theoretical and x-ray experimental values for the Compton profile. The effects of background, multiple scattering, and energy resolution are discussed. The electron-impact and x-ray methods for measuring Compton profiles are compared.

I. INTRODUCTION

In 1923 Compton¹ reported that the spectral line of the inelastically scattered x rays was significantly broadened, and Du Mond² in 1929 derived a Doppler-broadening theory which pointed out that Compton scattering should be ideal for the measurements of momentum distributions of the electrons in molecules. But it has only been recently³⁻⁸ that such experiments were performed with a high enough accuracy to make meaningful comparisons between theory and experiment. An excellent review of the literature up to 1971 has been given by Cooper.⁹ All recent studies have been performed using x rays, although as Hughes and Mann¹⁰ showed in 1938 the energy spectrum obtained from electron scattering shows the same characteristic Compton profile. They did not, however, obtain good agreement with the theory^{11,12} which Duncanson¹³ ascribed to multiple scattering. This discouraged all further work using electron sources, which was unfortunate, and the authors will attempt to show,

using He as an example, that high-energy-electron spectroscopy is in most aspects superior to photon scattering.

II. EXPERIMENT

The high-energy-electron spectrometer used in these studies has been partially described in an earlier paper.¹⁴ The present apparatus is essentially identical to that given in Ref. 14 with the solid-state detector replaced by an electrostatic differential velocity analyzer of the Mollenstedt type¹⁵ which had an optimum resolution of at least 1.5×10^{-5} [0.4 eV full width at half-maximum (FWHM)/25 keV]. An incident electron beam intensity of 200 μA obtained from a telefocus electron gun¹⁶ with a diameter of 400- μ FWHM was allowed to impinge at a right angle on a gas jet with a nozzle diameter of 125 μ , and a flow rate of 1.6×10^{20} He atoms per second. The optimum vacuum of the scattering chamber, which had a pumping speed of about 15000 liter/sec, was 3×10^{-7} torr and increased to 4×10^{-5} torr during an experiment,

while the differentially pumped analyzer chamber remained at 8×10^{-7} torr. With the electron gun rotated to $+6.91^\circ$ and -7.21° , to test the symmetry of the scattering apparatus, an energy-loss spectrum was obtained over a 1-keV range at both angles. The results are shown in Fig. 1. To increase the count rate the energy resolution of the velocity analyzer was decreased to $\sim 10^{-4}$ (2.7-eV FWHM/25 keV about $\frac{1}{100}$ of the Compton profile FWHM) and to compensate for long-range fluctuations, data were collected in a signal averaging mode using 256 channels of a 512 multichannel analyzer (Northern Scientific, Model No. NS 600). The energy interval of one channel was about 4.7 V, and the dwelling time per channel was 0.600 sec, so that 16 000 counts were obtained at the Compton peak maximum after 30 sweeps taking a total of 80 min. The analyzer was calibrated by scanning the elastic peak across the signal averager (into the other 256 channels) using known bias voltages on the center electrode of the analyzer. The measured intensities had to be divided by the energy of the scattered electrons (not the energy loss), a small correction for the energy range used in this work. This correction was pointed out by Kollath¹⁷ for other spectrometers and will be discussed in more detail in a forthcoming paper together with the energy calibration procedure.¹⁵ By leaking gas into the chamber elsewhere, the background spectrum was found to be about 80 counts per channel and independent of energy. This was only slightly more than the contribution from the noise pulses of the silicon surface barrier detector.

The spectrum of Fig. 1 shows beside the elastic line at 0 eV and the Compton profile, for which the intensities were found to be proportional to the gas density, a feature between 20 and 50 eV, which was found to be proportional to the square

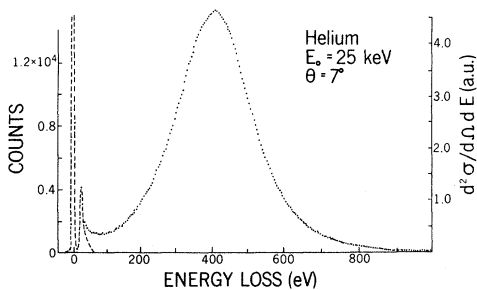


FIG. 1. Electron-impact energy-loss spectrum of helium, taken with 25-keV incident electron, at a scattering angle of 7° , with a resolution of 2.7-eV FWHM. The spectrum consists of the following features: The elastic line at an energy loss of 0 eV, the Compton profile with its peak at ~ 230 eV, and a structure between 20 and 50 eV due to multiple scattering. See text for a detailed discussion.

of the gas density. Furthermore, measurements at zero scattering angle showed an identical profile between 20 and 50 eV, indicating that this feature is a multiple scattering event involving a nearly zero-angle inelastic scattering and a 7° elastic scattering. It has been shown¹⁸ that the ratio of total inelastic to total elastic cross sections is about seven, also the inelastic differential cross section decreases by about two orders of magnitude in the angular range from 0 to 0.1° .¹⁹ Hence this observed multiple scattering is the only significant mechanism for multiple scattering corrections to the observed cross section. The ratio of double scattering to single elastic scattering at 7° was found to be 0.42%, producing a second Compton profile, 0.42% as intense as the single scattered profile, shifted by some 25 eV to the high-energy-loss side.

III. THEORY

An approximate theoretical description of the present experiment has been presented in detail elsewhere.²⁰ The starting point for obtaining this theoretical development was the first Born approximation for the total inelastic scattering including all exchange effects. The exact first Born expression was developed in an expansion in powers of \hbar where the zeroth-order term corresponded to a quantum-mechanical binary-encounter model. That is, a model in which each incident electron is scattered by at most two target electrons and conservation of energy and linear momentum are satisfied for the initial collision only. The corrections to the zeroth-order theory proportional to \hbar were investigated, and it was concluded that they should be negligible in the angular range between 10° and 70° at an incident electron energy of 25 keV.

The theory was simplified by introducing an approximate factorization which separated the interference and exchange corrections from the direct scattering. The factorization guaranteed that the resultant approximate theory yielded the correct results at both large and small scattering angles. The approximate form can be written

$$\frac{d^2\sigma}{d\Omega dE} = \frac{2k_s[1 - (E/2c^2)(1 - \beta^2)^{1/2}]}{k(1 - \beta^2)K^2(E) - (E^2/4c^2)^2} \times [J(q) + J_c(q)] E_{\text{ex}}(q), \quad (1)$$

where k_s is the magnitude of the scattered wave vector, k is the magnitude of the incident wave vector, and K is the magnitude of the inelastic momentum transfer on scattering ($\vec{K} = \vec{k} - \vec{k}_s$). Note that relativistic definitions of k , k_s , and K are used throughout and all expressions are given in Rydberg atomic units. The quantity $J(q)$ is the usual x-ray Compton profile defined as

$$J(q) = 2\pi \int_0^\infty dp p \rho(p), \quad (2)$$

with $q = (-E + K^2)/2K$, where E is the energy loss on scattering, p is the target electron momentum and $\rho(p)$ is the one-electron momentum density of the target. The third term in square brackets in Eq. (1) is given for atomic hydrogen and for a Hartree-Fock wave function for He as

$$J_c(q) = -2\pi f(K) \int_0^\infty dp p \rho[(p^2 + k^2 - k_s^2)^{1/2}, p], \quad (3)$$

where $f(K)$ is the one-electron x-ray coherent-scattering factor, with $f(0) = 1$, and $\rho(p', p)$ is the nondiagonal one-electron momentum density. The exchange and interference correction $F_{ex}(q)$ can be simplified in the limit of high-incident electron energy and written approximately

$$F_{ex}(q) \approx -\frac{K^2}{k_s^2} + \frac{K^4}{k_s^4} - \frac{2K^4 q^2}{k_s^6}, \quad (4)$$

where the first two terms on the right-hand side of the equal sign have been given previously by others^{21,22} for the first-order differential cross section and amount to about a 1½% correction in the present case.²⁰

It is useful to compare Eq. (1) with the corresponding x-ray result

$$\left(\frac{d^2\sigma}{dE d\Omega}\right) = \left(\frac{d\sigma}{d\Omega}\right)_{Th} \frac{\omega_1}{\omega_2} \frac{1}{K} [J(q) + J_c(q)], \quad (5)$$

where $(d\sigma/d\Omega)_{Th}$ is the Thompson cross section for electron-photon scattering, ω_1 is the energy of the scattered x-ray photon, and ω_2 is the energy of the incident x-ray photon. Note that the two expressions differ mainly in the expressions for the cross section for the single-particle scattering (i. e., electron-electron or Rutherford scattering and electron-photon or Thompson scattering) and in the presence of exchange and interference corrections in the electron scattering case.

IV. DATA ANALYSIS AND RESULTS

The experimentally determined background was subtracted directly from the raw data, also the elastic line was removed, and the double scattering feature was subtracted using the zero-angle profile. This produced a 20% uncertainty below 50 eV in the remaining spectrum. The area under the profile was normalized to the integral over E of Eq. (1) using Hartree-Fock values. The result was then corrected for $J_c(q)$ and $F_{ex}(q)$. The resulting experimental $J(q)$ function was then renormalized so that $\int_{-\infty}^{\infty} dq J(q) = 2$, the number of electrons in the He atom. To check on the effect of the systematic errors on large negative values of q , the low- and high-energy-loss sides were, after the above normalization, integrated separately [i. e., $\int_{-\infty}^0 J(q) dq$ and $\int_0^{+\infty} J(q) dq$] and each was found to add up to unity within the experimental accuracy.

Table I shows the $J(q)$ values, which are the average of the +6.91° and -7.21° measurements for the low- and high-energy-loss side, and compares them to theory and previous best x-ray and γ -ray data. The results appear to be, except for the low-energy-loss side, in extremely good agreement with theory.

V. DISCUSSION AND CONCLUSION

It might seem to be a strange procedure to first normalize the data to the differential cross section, which requires a knowledge of $J(q)$; but as shown in Table II these corrections are very small and any reasonably good wave function will be sufficient. As $J(q)$ values are renormalized to two, only the deviation in the percentage correction at different parts of the Compton profile will effect the $J(q)$ values. This relative correction is less than the uncertainties in the profile, and omitting them would not significantly change the final results. Similar arguments hold for the background correction, which if omitted would only affect $J(q)$ at large values of q , where the uncertainty is over 10%. The poor agreement with theory on the low-energy-loss side is due to the fact that the binary encounter theory does not include corrections for contributions from bound-state transitions or the threshold behavior in the energy-loss region of the first and second ionization potentials. According to theory $J(q)$ decreases monotonically from a maximum value at $q = 0$ to zero as $\pm q$ approaches infinity, but $q < -2.5$ corresponds to an energy loss of less than the first bound-state transition and hence $\partial^2\sigma/\partial\Omega \partial E$ has to vanish. To improve the low-energy-loss side, the scattering angle could be increased which would also shift the Compton profile away from the double scattering feature, but with a decreased count rate and an increased exchange correction.

Having demonstrated that Compton profiles can be measured to a high degree of accuracy by the use of electron-impact spectroscopy, it will be useful to compare the present technique with photon Compton scattering. The experiment by Eisenberger³ using x rays and Eisenberger and Reed's⁷ experiment using γ rays represent the present state of the art and have about the same statistical accuracy as the present work and hence will serve well for comparison. Table III lists the relevant points for comparison.

Time Taken

This is the most striking difference, the electron experiment was taken in less than $\frac{1}{100}$ of the time. Hence it would be feasible to increase the statistical accuracy in the electron case to the 0.1% level, which would require a 2-yr experiment with photon scattering.

TABLE I. Comparison between Compton profiles for He, obtained from electron scattering, x-ray scattering, and theory.

q	Electron scattering		x-ray scattering Eisenberger ^a	γ -ray scattering Eisenberger and Reed ^b	Theory Henneker ^c
	Low-energy- loss side	High-energy- loss side			
0	1.070	1.070 \pm 1.8%	1.066 \pm 0.7%	1.071 \pm 1.5%	1.068
0.1	1.049	1.050	1.052	1.058	1.055
0.2	1.018	1.014	1.012	1.019	1.015
0.3	0.960	0.949	0.954	0.958	0.954
0.4	0.890	0.873	0.876	0.881	0.876
0.5	0.815	0.785	0.789	0.795	0.788
0.6	0.721	0.703	0.700	0.705	0.698
0.7	0.626	0.616 \pm 1%	0.612	0.616	0.609
0.8	0.559	0.530	0.527 \pm 1%	0.533 \pm 2.3%	0.525
0.9	0.471	0.454	0.448	0.456	0.449
1.0	0.407	0.388	0.382	0.388	0.381
1.2	0.298	0.280	0.275	0.274	0.271
1.4	0.218	0.197	0.195	0.188	0.191
1.6	0.163	0.142	0.137	0.129	0.135
1.8	0.118	0.102	0.098	0.093	0.096
2.0	0.089	0.071	0.067	0.069	0.069
2.5		0.033 \pm 5%	0.027 \pm 10%	0.030 \pm 15%	0.031
3.0		0.014	0.008	0.013	0.015
3.5		0.007 \pm 10%			
4.0		0.002			

^aReference 3.^bReference 7.^cMulticonfiguration-self-consistent yield calculations; see Ref. 23.

Experimental Parameters

These values are listed only for reference, and it will be difficult to increase any of those significantly to improve the experiment. (Stronger γ sources are available but difficult to handle.⁷)

Beam Energy

The γ beam and electron beam are essentially monoenergetic; the $PbK\alpha$, $K\beta$ contributions in the γ case result from the lead casing and seem to lie below the Compton profile. The x-ray beam is a doublet with a Bremsstrahlen background, both of which can be corrected for, but any procedure of this kind will increase the uncertainty. The main advantage of electrons over the photons in the present case is provided by the difference in scattering power for the two particles as given by the ratio of Rutherford to Thompson cross section which is about 10^4 . If the electron experiment is performed at larger angles, this advantage will decrease as $\sin^4 \frac{1}{2}\theta$, where θ is the scattering angle.

Background

While the background correction was one of the major difficulties in the x-ray experiment, it represents no problem in the γ -ray and electron experiments. The extremely low background in the electron experiment (most of it is detector noise) is due to the small scattering volume of approxi-

mately 1 mm^3 (the analyzer entrance window was 1 m away from the scattering center) and the use of a double-aperture system which limited the angular acceptance region to the scattering volume.

Resolution

Both photon experiments had rather poor energy resolution, and corrections to the experimental data had to be made to remove the effect of finite energy resolution. No correction was necessary

TABLE II. The theoretical percentage corrections from the interference scattering from pair of electrons $J_c(q)$ and the exchange factor $F_{\text{ex}}(q)$.

q	$J(q)$	$J_c(q)/J(q) \times 100$	$(F_{\text{ex}} - 1)100$	$\Delta J(q)/J(q)100^a$
-2	0.089	-2.6		-1.5
-1.5	0.185	-1.2	-1	-0.4
-1.0	0.407	-0.6	-1	0
-0.5	0.815	-0.3	-1	0
0.0	1.070	-0.2	-1.5	0
0.5	0.785	-0.1	-1	0.2
1.0	0.388	-0.2	-1	0.4
1.5	0.169	-0.3	-1	0.5
2.0	0.071	-0.3	-1	0.6
2.5	0.033	-0.5		0.8
3.0	0.014	-0.7		0.6
3.5	0.007	-1.5		0.5
4.0	0.002			1.0

^aThis correction is defined as $\Delta J(q)/J(q) = \{ [J_c(q)/J(q)] + [F_{\text{ex}}(q) - 1] - [J_c(0)/J(0)] - [F_{\text{ex}}(0) - 1] \} \times 100$, because of the renormalization of the profile.

TABLE III. Comparison of x rays, γ rays, and electron experiments.

	x rays	γ rays	Electrons
Time taken for experiment (hours)	≈ 150	≈ 150	1.3
Experimental parameters:			
Source strength, particles/sec	5×10^{14}	4×10^{10}	3×10^{15}
Beam energy (keV)	17	160	25
Energy spread of beam	$K_{\alpha 1}, K_{\alpha 2}$	monochromatic	0.40 eV FWHM
Beam structure	K_{β} , Bremsstrahlen	Pb $K_{\alpha}, K_{\beta 2}$	none
Gas pressure	450 lb/in. ²	525 lb/in.	≈ 0.3 torr
Solid angle of analyzer	$\pm 3^{\circ}, \pm 0.1^{\circ}$	$\pm 4^{\circ}, \pm 4^{\circ}$	$\pm 0.04^{\circ}, \pm 0.0025^{\circ}$
Cross section (cm ²)	10^{-25}	10^{-25}	10^{-24}
Background	guessed	measured 600 counts/channel	measured 80 counts/channel
Relative resolution: Analyzer (FWHM)/Compton profile (FWHM)	50/250=0.20	470/2100=0.22	2.7/270=0.01

in the electron experiment, as the error is approximately proportional to the square of the ratio of the FWHM of the energy resolution to the FWHM of the Compton profile, i. e., 0.01%. If the FWHM of the energy resolution is increased to the level of the photon experiment, the count rate would increase by a factor of 400; as the Möllenstedt analyzer efficiency increases with the square of the resolution. This, in effect, means if the resolution of the photon experiments is used, that the same experimental data could be taken in 12 sec, and that only 2.0 mmole of sample per valence electron would be required. Note, however, that the amount of sample required is strongly dependent on the size of the largest atom in the target. If the correction given in Eq. (3) is to be maintained at the 1% level or less, then for the first two rows of the Periodic Table ($Z < 18$) the amount of sample needed relative to hydrogen is roughly proportional to $N^{2.5}$, where N is the number of electrons in the largest atom in the target.

These comparisons were made using the He ex-

periments, but they will apply for other atoms and molecules as well. As the atomic number Z increases, both experiments, although for different reasons, become more difficult, in spite of the fact that the Compton-scattering cross section increases proportional to Z . In the photon case, the photoelectric effect attenuates the beam approximately proportional to Z^4/E^3 , where Z is the atomic number and E is the incident energy before it reaches the observed scattering volume. Hence, the experiment gets rapidly more difficult. The γ -ray experiment gains a factor of $\approx 10^3$ advantage over the x-ray experiment because of its higher energy.⁷ In the electron case, the angle where the $J_c(q)$ correction is negligible becomes larger and the Compton cross section decreases by $\sin^4 \frac{1}{2} \theta$.

From this work it should appear that the electron-scattering experiment is, at least for light atoms, more suitable for Compton-profile measurements on single atoms or molecules than the x-ray and γ -ray experiments. Measurements on other atoms and molecules are in progress.

*Work supported under AFOSR Grant No. AFOSR-70-1900.

¹Publication Number 2158 from the Chemical Laboratories of Indiana University.

²A. H. Compton, Phys. Rev. **22**, 412 (1923).

³Du Mond, Phys. Rev. **33**, 643, 1929.

⁴P. Eisenberger, Phys. Rev. A **2**, 1678 (1970).

⁵R. J. Weiss, J. Chem. Phys. **52**, 2237 (1970).

⁶P. Eisenberger, Phys. Rev. A **5**, 628 (1970).

⁷P. Eisenberger and W. C. Mara, Phys. Rev. Lett. **27**, 1412 (1971).

⁸P. Eisenberger and W. A. Reed, Phys. Rev. A **5**, 2085 (1972).

⁹P. Eisenberger *et al.*, J. Chem. Phys. **56**, 1207 (1972).

¹⁰M. Cooper, Adv. Phys. **20**, 453 (1971).

¹¹A. L. Hughes and M. M. Mann, Phys. Rev. **53**, 50 (1938).

¹²A. L. Hughes and M. A. Starr, Phys. Rev. **54**, 189 (1938); Phys. Rev. **55**, 343 (1939).

¹³A. L. Hughes and T. Enns, Phys. Rev. **60**, 345 (1941).

¹⁴W. E. Duncanson, Proc. Camb. Philos. Soc. **39**, 180 (1943).

¹⁵H. Schmoranzler, R. C. Ulsh, R. A. Bonham, and J. Ely, J. Chem. Phys. (to be published).

¹⁶A. J. F. Metherell and M. J. Whelan, J. Appl. Phys. **37**, 1737 (1966); H. F. Wellenstein, J. Appl. Phys. (to be published).

¹⁷H. H. Steigerwald, Optik (Stuttg.) **5**, 469 (1949); H. F. Wellenstein (unpublished).

¹⁸R. Kollath, Ann. Phys. (Leipz.) **27**, 721 (1936).

¹⁹R. A. Bonham and E. W. Ng, Chem. Phys. Lett. **6**, 403 (1971).

²⁰J. Geiger, Z. Phys. **175**, 530 (1963).

²¹R. A. Bonham and C. Tavard, J. Chem. Phys. (to be published).

²²N. F. Mott, Proc. R. Soc. A **126**, 259 (1930).

²³C. Möller, Ann. Phys. (Leipz.) **14**, 531 (1932).

²⁴B. Henneker, see Ref. 3.