High-energy electron scattering from helium

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(Received 21 September 1981)

Total (elastic and inelastic) differential scattering cross sections for 40-keV electrons incident on helium have been measured with a precision of 0.1%. Theoretical scattering intensities as given by an explicitly correlated wave function are compared with experimental results. It is shown that corrections to the Morse expression for kinematic effects, at small angles, are adequate to relate scattering intensities at constant angle to incoherent x-ray scattering factors.

I. INTRODUCTION

Helium is one of the simplest atoms and consequently the subject of much theoretical attention. High-quality configuration-interaction (CI) wave functions are available for helium. In the past, scattering calculations based on these wave functions have served as a reference for experimentalists to calibrate their techniques.[1] Critical comparison of theoretical predictions for He with electron scattering experiments has been hampered by the relatively large uncertainties in the experimental results.[2] Recently new questions were raised about the quality of the wave function at very large atomic distances and the scattering process with very small momentum transfer.[3] The aim of this work has been to make high-precision (0.1%) measurements of the scattered intensity for 40-keV electrons scattering from He and to critically compare these results with theoretical predictions.

II. THEORY

The description of the scattering of high-energy electrons is given, in the first Born approximation, in terms of the x-ray form factor $F(K)$ and the incoherent x-ray scattering factor $S(K)$. These are given by

$$F(K)=\int_0^\infty D(r)j_0(Kr)dr \ , \qquad (1)$$

$$S(K)=N+2\int_0^\infty P(r)j_0(Kr)dr-[F(K)]^2 \qquad (2)$$

with

$$K=\frac{4\pi}{\lambda}\sin\tfrac{1}{2}\theta \ ,$$

where λ is the wavelength of the incident electron, θ is the scattering angle $j_0(x)$ is the spherical Bessel function of order zero, N is the number of electrons in the target atom and $D(r)$ and $P(r)$ are the electron-nuclei and the electron-electron distribution functions, respectively.

For free-atom scattering the elastic ($I^{\rm el}$) scattered intensity is given, in atomic units, in the Born approximation as

$$I^{\rm el}(K)=4K^{-4}[N-F(K)]^2 \qquad (3)$$

and the inelastic scattering in the Morse approximation[4] as

$$I^{\rm inel}(K)=4K^{-4}S(K) \ . \qquad (4)$$

The elastic exchange scattering amplitude is given by $f_{\rm ex}=(K^2/k^2)F(K)$. The direct and the exchange amplitudes add coherently to give the total elastic scattering intensity as

$$I^{\rm el}(K)=4K^{-4}\left\{[N-F(K)]^2+2K^2[N-F(K)]\frac{F(K)}{k^2}+\frac{K^4}{k^4}[F(K)]^2\right\} . \qquad (5)$$

The equivalent treatment of the inelastic process leads to[5]

$$I^{\rm inel}(K)=4K^{-4}\left[1-\frac{K^2}{k^2}+\frac{K^4}{k^4}\right]S(K) \ , \qquad (6)$$

where k^2 is the nonrelativistic energy of the incident electron.

This simple picture of the Morse approximation has been called into question by experimental results.[6,7] The problem is that at the same scattering angle the momentum transfer for inelastic scattering is not the same as that for elastic scattering. For elastic scattering the center of mass and the laboratory system lie at the nucleus. However, for an ionizing collision the center-of-mass system for two electrons is different from the laboratory system in which θ is determined. So, one obtains

$$I^{\mathrm{inel}}(K)=4K_B^{-4}\left[1-\frac{K_B^2}{k^2}+\frac{K_B^4}{k^4}\right]S(K_B)\cos\theta\ , \tag{7}$$

where $K_B=(2\pi/\lambda)\sin\theta$ rather than the elastic value $K=(4\pi/\lambda)\sin(\theta/2)$ (note that the scattering angle in the center-of-mass system is twice the laboratory angle for two free identical particles). K_B is the maximum in the inelastic intensity distribution as given by the classical description of the Compton effect. The difference between K_B and K is 0.4% at $\theta=10°$ and 1.5% at $\theta=20°$.

Another approach is to write the inelastic intensity in terms of the generalized oscillator strength (GOS) $(df/dE)(K,E)$ as[8,9]

$$I^{\mathrm{inel}}(K)=\sum_{n=1}^{k^2}\left[\frac{k_nK^2}{kE_n}\right]\frac{4}{K^4}\left[1-\frac{K^2}{k^2}+\frac{K^4}{k^4}\right]\times\frac{df}{dE}(K,E)\ , \tag{8}$$

where $k_n^2=k^2-E$, with E the energy transferred to the target by the incident electron. The symbol $\sum$ denotes a sum over the bound states and an integration over continuum states accessible to excitation by the incident energy k^2.

Following the reasoning given in Ref. 9 one can represent the GOS, with sufficient accuracy, by the first two terms in the series expansion about the maximum point in the inelastic spectrum as

$$\frac{df}{dE}(K,E)=\frac{df}{dE}(\bar{K},E)+(K^2-\bar{K}^2)\left[\frac{d}{dK^2}\left[\frac{df}{dE}(K,E)\right]\right]_{K=\bar{K}}, \tag{9}$$

where $\bar{K}$ is given exactly by

$$\bar{K}^2=2k^2-\bar{E}-2k^2\left[1-\frac{\bar{E}}{k^2}\right]^{1/2}\cos\theta \tag{10a}$$

which reduces to

$$\bar{K}^2\cong K^2\left[1-\frac{\bar{E}}{2k^2}\right]+\frac{\bar{E}^2}{4k^2}\cos\theta+O\left[\frac{\bar{E}^3}{k^4}\right] \tag{10b}$$

by expansion in powers of $\bar{E}/k^2$, where $\bar{E}$ is an appropriately chosen average energy loss which is a function of the scattering angle and K is the elastic momentum transfer $2k\sin(\theta/2)$. Since K is a function of both E and θ, data collected at a fixed scattering angle cannot be associated with a single value of $\bar{K}$. At best the appropriate value of K would be given by an average over all possible energy-loss values weighted by the spectral distribution $(df/dE)(K,E)$. Because the spectral distribution was not measured in this experiment the energy loss was replaced by an average value selected to fit the experimental data.

As $\theta\to0$, the momentum transfer for the elastic scattering, $K\to0$ whereas for the inelastic scattering $\bar{K}$ approaches $\bar{E}/2k$. The fact that $\bar{K}$ depends upon the energy loss necessitates certain corrections to be made when using data at constant scattering angle. The effect of this is to modify Eq. (6) to

$$I^{\mathrm{inel}}(K)=\frac{4}{\bar{K}^4}\left[1-\frac{\bar{K}^2}{k^2}+\frac{\bar{K}^4}{k^4}\right]\left[S(\bar{K})-\frac{3N\bar{K}^2}{4k^2}\right]. \tag{11}$$

Consequently, one can write

$$I^{\mathrm{tot}}(K)=I^{\mathrm{el}}(K)+I^{\mathrm{inel}}(K)\ ,$$

$$I^{\mathrm{tot}}(k)=\frac{4}{K^4}\left[[N-F(K)]^2+2K^2\frac{[N-F(K)]}{k^2}F(K)+\frac{K^4}{k^4}[F(K)]^2\right]+\frac{4}{\bar{K}^4}\left[1-\frac{\bar{K}^2}{k^2}+\frac{\bar{K}^4}{k^4}\right]\left[S(\bar{K})-\frac{3N\bar{K}^2}{4k^2}\right]. \tag{12}$$

The first-order term $-3N\bar{K}^2/4k^2$ makes a larger correction as $K\to0$ and is important only for small scattering angle.

III. EXPERIMENTAL

The experimental setup used is described in detail elsewhere.[10] A 40-keV electron beam with a current of about 10 μA was crossed with the atomic gas jet at 90°. The resultant scattered electron intensity was measured as a function of angle. The uncertainty in the scattering angle was ±1 arcsec and the scattered intensities were recorded with a precision of 0.1%. The electron wavelength was calculated by calibrating the accelerating voltage against diffraction patterns of gaseous CO_2 [r_a(CO)$=1.1646$ Å].[11] The resulting data for 40 keV electrons incident on helium are of sufficient accuracy, for the first time, to make critical comparisons with theoretical predictions.

Eight measurements were made on helium in the K range 0.32—7.57 a.u. The data are documented in PAPS.[12] These different sets of data were scaled to each other by matching $K^4 I(K)$. The resulting combined data were then put on an absolute scale by matching them to theoretical results, at large values of momentum transfer.

IV. RESULTS

Our scaled experimental intensities are compared to theoretical ones by plotting $\Delta\sigma(K)$, where

$$\Delta\sigma(K)=K^4(I_{\rm expt}^{\rm tot}-I_{\rm theory}^{\rm tot}) .$$

Figure 1 shows the plot of $\Delta\sigma(K)$ where the theoretical intensities are calculated from a Hartree-Fock wave function of Clementi.[13] The $\Delta\sigma(K)$ curve based on such a wave function shows a deep minimum around $K=2.0$ a.u. This is to be expected since the Hartree-Fock wave function does not take into account the effects of correlation which are significant, especially for the inelastic channel. From the difference function $\Delta\sigma(K)$ the correlation energy can be calculated using Tavard's theorem[5]:

$$E_c=-(2\pi)^{-1}\int_0^\infty \Delta\sigma(K)dK .$$

The value of the correlation energy from this study is 0.04 a.u. which compares well with the spectroscopic value of 0.042 a.u. (Ref. 13).

There are numerous CI wave functions available for helium. We use an explicitly correlated wave function of Thakkar and Smith for which $F(K)$ and $S(K)$ values have been computed.[14,15] This is the best choice since both one- and two-electron CI effects are properly treated. These $F(K)$ and $S(K)$ values were converted to total scattering intensities, in the Morse approximation, according to Eqs. (5) and (6). Figure 2 shows a plot of $\Delta\sigma(K)$ for such a correlated wave function. This $\Delta\sigma(K)$ curve shows a slight dip below $K=2.0$ a.u. This small dip suggests that the Morse approximation is not valid, especially for small angles. Theoretical total scattering intensities were also calculated within the binary-encounter theory. However, even for this case the $\Delta\sigma(K)$ curve was essentially the same as the one shown in Fig. 2. The use of the binary-encounter theory is hardly any improvement on the Morse approximation (as it should not be for small to intermediate K values).

Both the theoretical intensities considered so far deviate from the experimental intensities at small angles. The correction to the inelastic scattering intensity as given in Eq. (11) is a small angle correction. So it seems reasonable to expect that the use of the first-order correction will remove the descrepancy between theoretical scattering intensities and experimental ones. However there is a certain ambiguity in using Eq. (11). The average momentum transfer $\bar{K}$ is related to an "appropriate" average energy loss $\bar{E}$ according to Eq. (10). In a total (elastic and inelastic) scattering experiment, like ours, the value of $\bar{E}$ does not come out of the experiment. However, one can, by trial and error, find a value of $\bar{E}$ which results in good agreement between experimental and theoretical intensities.

Figure 3 shows a plot of $\Delta\sigma(K)$ for theoretical intensities calculated according to Eq. (12) for three

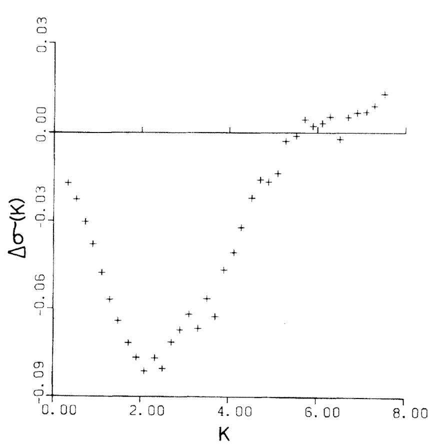

FIG. 1. $\Delta\sigma(K)$ curve based on a Hartree-Fock wave function. Atomic units are used. Error bars at small K values are smaller than the size of the points. Experiment matched to theory in the range $K=5$ to 7.5 a.u.

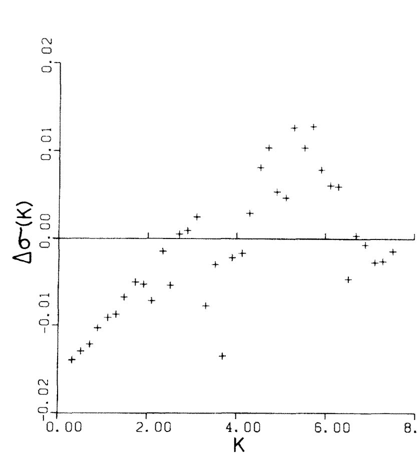

FIG. 2. $\Delta\sigma(K)$ curve based on the explicitly correlated wave function of Ref. 14 in the Morse approximation. Atomic units are used. Error in the data is given by the scatter in the points.

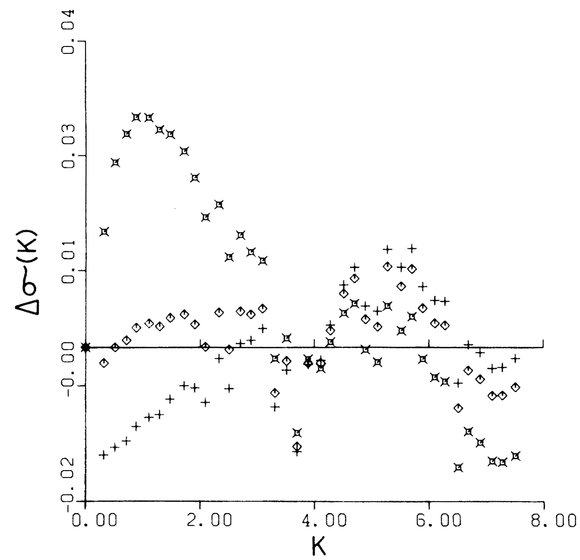

FIG. 3. $\Delta\sigma(K)$ curve based on the explicitly correlated wave function, with first-order corrections applied for dynamic effects. Curves are $+$ $\bar{E}=0$ eV, ◊ $\bar{E}=55$ eV, ✧ $\bar{E}=100$ eV. Atomic units are used. Error in the data is given by the scatter in the points. At $K=0$ $K^4 I^{tot}_{expt} = K^4 I^{tot}_{theor} = 0$.

different values of $\bar{E}$, one with $\bar{E}=0$ eV (the Morse case), one with $\bar{E}=55$ eV and one with $\bar{E}=100$ eV. As can be seen from this figure a value of $\bar{E}=55$ eV results in reasonably good agreement between the theoretical intensities and experimental ones, within the experimental error bars. A value of 55 eV for the average energy seems like a reasonable one since the appropriate value for He for $\theta=0°$ is 31.77 eV and this value should increase with increasing scattering angle.[16] Although the $\Delta\sigma(K)$ curve is not perfectly flat it is difficult to say if the slight deviations are due to the second-order correction term which was neglected.

In conclusion, our experimental results show that both the Morse and the binary-encounter theories are inadequate in relating experimental inelastic intensities at constant angle to the incoherent x-ray scattering factors. A first-order correction, which takes into account the fact that K is not constant over the energy loss, explains the experimental results. An ambiguity over the choice of an average energy loss still exists.

The results presented here show that studies in high-precision measurement of the differential inelastic scattering factors are needed to evaluate the second-order density-matrix elements of the CI wave functions. The high-precision elastic data will provide the base for a comparison with the first-order density-matrix element.

ACKNOWLEDGMENTS

The authors wish to express their gratitude to the entire counting electron diffraction group at the University of Texas for their help in collecting the data. This work was supported by the National Science Foundation and the Joint Services Electronics Program, under U. S. Air Force Office of Scientific Research Contract No. F49-620-77-C-0101, and National Science Foundation Grant No. CHE-8000253 made to the Indiana University Foundation, Bloomington, Indiana.

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[12]See AIP document No. PAPS-PLRAA-27-806 for 4 pages of the experimental scattering intensities for helium. Order by PAPS number and journal reference from American Institute of Physics, Physics Auxilliary Publication Service, 335 East 45th Street, New York, N.Y. 10017. The price is $1.50 for each microfiche (98 pages), or $5 for photocopies of up to 30 pages with $0.15 for each additional page over 30 pages. Airmail additional. Make checks payable to the American Institute of Physics.

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