# Tests of Born Approximations: Differential and Total $2^3S$, $2^1P$, and $2^1S$ Cross Sections for Excitation of He by 100- to 400-eV Electrons*

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(Received 14 July 1967)

The angular dependence of $2^3S$, $2^1P$, and $2^1S$ excitation of He for incident electron energies from 100 to 225 eV ($2^3S$) and 400 eV ($2^1P$ and $2^1S$) has been measured. Apparent generalized oscillator strengths $f(K)$ and differential cross sections for the transition $1^1S \rightarrow 2^1P$ are obtained by normalizing to Schiff and Pekeris's optical oscillator strength. From the experimental intensity ratios $2^3S/2^1P$ and $2^1S/2^1P$ we then calculate differential $1^1S \rightarrow 2^3S$ and $1^1S \rightarrow 2^1S$ cross sections. The differential cross sections are integrated to get total cross sections. The $f(K)$ found here for $2^1P$ and $2^1S$ excitation decrease faster with increasing momentum transfer $K\hbar$ than in earlier studies. Departures from the Born approximation appear only below 200 eV for $2^1P$ excitation, but occur at higher energy and are larger (especially for large momentum transfers) for $2^1S$ excitation. The angular dependence found for $2^3S$ excitation disagrees strongly with the Ochkur (Bonham) approximation. Our total $2^3S$ cross sections are much lower than all other existing theoretical and experimental data.

## I. INTRODUCTION

THIS paper deals with two major subjects: a study of the breakdown of the Born approximation for $2^1P$ and $2^1S$ excitation in He by 100 to 400 eV electrons and a study of $2^3S$ excitation in He between 100 and 225 eV.

Not much information on the range of validity of the Born approximation[1] is available. Only recent optical measurements by Moustafa, de Heer, and Schutten[2] show that the Born approximation breaks down at much higher incident energies for $n^1S$ than for $n^1P$ and $n^1D$ excitation and at higher energies for $n^1D$ than for $n^1P$ excitation. However, these optical measurements are of total cross sections and give no information about differential cross sections. With an electron spectrometer we measured the angular dependence of the $2^1P$, $2^1S$, and $2^3S$ scattered intensity for various incident electron energies. By fitting our relative $2^1P$ data to suitable analytical expressions and normalizing on the $1^1S \rightarrow 2^1P$ optical oscillator strength of Schiff and Pekeris,[3] we obtained absolute differential and total $2^1P$ excitation cross sections. The ratio of intensities of $2^1S$ and $2^3S$ to $2^1P$ were used to derive cross sections for the $S$ states.

The cross sections for the $2^1P$ and $2^1S$ states so obtained were tested against the predictions of the Born approximation. For $2^3S$ excitation, which can only occur via electron exchange, comparison was made with the Ochkur (Bonham),[4] Born-Oppenheimer,[5] and first-order exchange[5] approximations.

## II. THE EXPERIMENTAL PROCEDURE

To obtain absolute cross sections directly it would be necessary to know the absolute number density of the gas, the absolute current collection efficiency of detectors measuring incident and scattered currents, the absolute dispersion of the analyzer, and a considerable number of geometrical parameters involving input beam profiles and intensity distributions. Moreover, these parameters must be continuously monitored or assumed to remain constant over the time that the experiment takes to run. The beam parameters and collection efficiencies must, moreover, be known for all beam energies and energy losses of interest.

This laboratory has embarked on a long term effort to achieve this end but at the present time the state of the art of high-resolution scattering experiments permits only partial realization of this goal. We believe that within stated limits of accuracy we can determine the angular dependence of the inelastic scattered intensity and the intensity ratio between different inelastic events at any given primary energy. Hence our techniques, as those of past workers, depend on measurements of these relationships and normalization to an absolute standard.

To achieve a response independent of energy loss necessitated careful analyzer design. In our case the analyzer acceptance is set by apertures in field-free space before any electron optics and hence is achromatic. The dispersing element and final collector are operated at fixed voltage independent of energy loss and hence at fixed dispersion and collection efficiency. The energy scan is achieved by an "energy add" lens which need operate over only a voltage ratio change of less than 5% in the worst case. Such narrow ranges lie well within dependable electron optical data and aberration terms are negligible. Extensive tests of the system were unable

* This research was supported by the Advanced Research Projects Agency of the Department of Defense under Contract No. 703.

† Guest worker from the University of Utrecht, The Netherlands, supported by NATO.

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to detect a change of $\pm 2\%$ in over-all analyzer efficiency over a 20% ratio change. Confidence in the apparatus is gained by the fact that these ratios reproduce from week to week and remain invariant as the resolution, and hence, focusing conditions of the analyzer are altered.

Dependability of angular distributions is also difficult to achieve. In this case after removing the gross distortion by canceling the earth's magnetic field one is left with residual distortion caused by instrumental effects. These distortions show up as small asymmetries between angular measurements made at equal angles on either side of the apparent zero of angle. They arise from two principal sources: The incident beam is not perfectly collimated and has finite size; the incident beam does not precisely intersect the axis of rotation of the analyzer and its geometric axis. The first of these sources of asymmetries is energy-dependent and increases as the energy decreases and the collimation becomes less perfect. Even with careful adjustment these cannot be eliminated completely. The effect of these residual asymmetries on our results are discussed in Sec. VII.

The electron spectrometer used in this work has been described by Kuyatt and Simpson[6] (see Fig. 7 of Ref. 6) and previous measurements with this apparatus are reported by Simpson, Menendez, and Mielczarek.[7] The angular resolution of the device is about 0.75° and for the present measurements we used energy resolutions of about 0.09 eV for 100-eV electrons to 0.13 eV for 400-eV electrons. These resolutions are sufficient to separate the $2^3S$, $2^1S$, and $2^1P$ levels and still give enough intensity to measure $2^3S$ excitation up to 225 eV and 15°.

For scattering angles larger than about 5°, the effective path length of the incident beam as seen by the entrance slits of the analyzer is simply proportional to $(\sin\theta)^{-1}$. Therefore we restricted ourselves to scattering angles equal to or larger than 5°. For large scattering angles we may expect trouble with double (elastic + inelastic) scattering. The importance of double scattering has been underestimated until recently[8] in measuring the angular dependence of inelastic cross sections in gases. Double scattering is important because elastic cross sections decrease only slowly with increasing momentum transfer (angle) while inelastic cross sections decrease much faster. For instance for the transitions $1s \rightarrow 1s$, $2s$, and $2p$ in atomic hydrogen, the Born-approximation cross sections per unit solid angle become proportional to $K^{-4}$, $K^{-12}$, and $K^{-14}$, respectively, for large momentum transfer $K\hbar$. For He the behavior of the cross sections is qualitatively similar[9] to that of H. Because of this difference in $K$ dependence, the combination of large angle elastic and small angle inelastic scattering becomes increasingly significant for larger $K$. These considerations limit our maximum angle to about 20° for 100 eV and about 10° for 400-eV electrons.

During the present measurements the pressure in the scattering chamber was about $10^{-2}$ Torr. Reducing the pressure by a factor of two produced no difference in the measured angular dependence of the $2^1P$ cross section for 400 eV up to 10° so that double scattering was considered to be negligible.

Within each series of measurements we first adjusted the incident energy and aligned the apparatus on the elastic 0° peak. Next[10] we measured for one angle the $2^3S$, $2^1S$, and $2^1P$ peak intensities; subsequently we advanced to the next angle and again measured the $2^3S$, $2^1S$, and $2^1P$ peak intensities and so on. For each angle we measured and corrected for background intensities.

We found no difference in peak width for the $2^3S$, $2^1S$, and $2^1P$ scattered currents and we found no difference in peak width for different scattering angles (within the experimental accuracy, which is about 5% for $2^3S$ and about 2% for $2^1S$ and $2^1P$). We may assume therefore that the peak intensities are proportional to the differential cross sections times $(\sin\theta)^{-1}$, which corrects for the effective path length. To determine the constant of proportionality the $2^1P$ relative cross sections at each energy are transformed to apparent generalized oscillator strength, extrapolated to zero momentum transfer and normalized to an accepted theoretical value of optical oscillator strength. Once the $2^1P$ cross section is thus determined, all other cross sections are obtained by direct comparison.

This normalization procedure, of course, must be justified and is considered in the next sections.

## III. THE BORN APPROXIMATION

In the first Born approximation,[1] which applies at sufficiently high incident electron energy $T$, the collision cross section $\sigma_{E,K}$ per unit momentum between $Ka_0$ and $Ka_0+d(Ka_0)$ is

$$\sigma_{E,K}d(Ka_0)=\frac{8\pi a_0^2R^2}{TE}f(K)\frac{d(Ka_0)}{Ka_0}, \qquad (1)$$

where $E$ is the excitation energy, $R$ the rydberg energy, $a_0$ the radius of the first Bohr orbit of hydrogen, and $f(K)$ the generalized oscillator strength defined by[1]

$$f(K)=(E/R)(Ka_0)^{-2}|\textstyle\sum_s(\psi_f|\exp(i\mathbf{K}\cdot\mathbf{r}_s)|\psi_i)|^2, \qquad (2)$$

where $\psi_i$ and $\psi_f$ are the initial- and final-state wave functions. The sum in Eq. (2) is over all atomic electrons with coordinates $\mathbf{r}_s$. In differential-cross-section mea-

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[7] J. Arol Simpson, M. G. Menendez, and S. R. Mielczarek, Phys. Rev. **150**, 76 (1966).
[8] G. E. Chamberlain, J. A. Simpson, S. R. Mielczarek, and C. E. Kuyatt, J. Chem. Phys. (to be published). This paper contains preliminary results and a discussion of the multiple scattering effect. A more detailed study of this effect for different sorts of transitions will be given later.
[9] L. Vriens, Phys. Rev. **160**, 100 (1967).
[10] For details see the discussion in Sec. VII.

surements we do not measure $\sigma_{E,K}$, but the cross section per unit solid angle $\sigma(\Omega)$ times the effective path length of the electrons in the scattering chamber, with

$$\sigma(\Omega)d\Omega=\frac{4a_0^2R}{E}\left(\frac{T-E}{T}\right)^{1/2}\frac{f(K)}{(Ka_0)^2}d\Omega, \quad (3)$$

where $d\Omega=\sin\theta d\theta d\varphi$. The total cross section for excitation to a level is

$$\sigma=\frac{8\pi a_0^2R^2}{TE}\int_{(Ka_0)\min}^{(Ka_0)\max}f(K)\frac{d(Ka_0)}{Ka_0}. \quad (4)$$

In this paper we do not calculate generalized oscillator strengths $f(K)$ via Eq. (2), but obtain apparent generalized oscillator strengths $f(K)$ from the experimental $\sigma(\Omega)$ via Eq. (3). For sufficiently large $T$, the apparent $f(K)$ should be independent of $T$; conversely any observed variation of the apparent $f(K)$ for small $T$ serves as an index of the breakdown of the Born approximation.

## IV. THE $1^1S\rightarrow 2^1P$ TRANSITION

Several series of measurements on the angular dependence of the $2^1P$ scattered-electron intensities $I$ were made. Averaged intensities relative to the 5° intensities and error margins are given in column 4 of Table I (see for a discussion Sec. VII). The incident electron energy $T$, the scattering angle $\theta$, and the square of the momentum transfer $K$ in units $a_0^{-1}$ are given in columns 1, 2, and 3.

Since, as mentioned in Sec. II, we could only measure in a limited range of scattering angles, we use an analytical extrapolation to extend our results. Vriens[9] found that the $f(K)$ can be represented with the series expansion

$$f(K)=\frac{f(0)}{(1+x)^6}\left\{1+\sum_{\nu=1}^{\infty}c_\nu\left(\frac{x}{1+x}\right)^\nu\right\}, \quad (5)$$

where the optical oscillator strength $f(0)=0.27616$ (see Ref. 3), $x=(Ka_0/\alpha)^2$ with $\alpha^2=3.391$, and the $c_\nu$ are unknown coefficients. Here $\alpha$ is related to the ionization energy $Q$ and excitation energy $E$ by $\alpha=(Q/R)^{1/2}+[(Q-E)/R]^{1/2}$. The $K$ dependence of $f(K)$ and thus the angular dependence of $\sigma(\Omega)$ is determined by the coefficients $c_\nu$, $f(0)$ being just a multiplication factor which determines the absolute values of $f(K)$. With $c_1=0.86$ and $c_\nu=0$ for $\nu\geqslant 2$, Eq. (5) reproduces[9] the theoretical $f(K)/f(0)$ from Lassettre and Jones[11] and Silverman and Lassettre[12] within 1 to 2%. With $c_\nu=0$ for $\nu\geqslant 2$ and the $c_1$ (further indicated by $c$) values of column 7 of Table I, we reproduce the experimentally found angular dependence of $\sigma(\Omega)$ with a largest deviation of 4% and average deviations of about 1%. This is illustrated in column 5 of Table I where we list the experimental scattered intensities $I$ times the inverse of the effective path length $\sin\theta$ and divided by the $\sigma(\Omega)$ calculated via Eqs. (3) and (5) and the $c_1$ (further indicated by $c$) values of column 7. These calculated $\sigma(\Omega)$ are given in column 6 of Table I. The known $f(0)$ value provides us with an absolute normalization of the $f(K)$ and $\sigma(\Omega)$. The $f(K)$ calculated with Eq. (5) and with the $c$ values of Table I are given in Fig. 1 by solid lines and the normalized experimental results by circles, squares, and triangles. In the graph we can hardly distinguish between the curves with $c=0.1$ and $c=-0.1$. We thus find that the momentum-transfer dependence of the apparent $f(K)$ does not change appreciably above $T=100$ eV. In the optically measured total $n^1P$ cross sections of Moustafa *et al.*[2] for $n=2$ to 6 no departures from the Born approximation above about 100 to 150 eV are found. Hence no measurable breakdown of the Born approximation is detected above 200 eV in either experiment and the Born approximation may be assumed to be valid above 200 eV. Consequently, our normalization on $f(0)$ for $T\geqslant 200$ eV is also justified. The analytical extrapolation of the apparent $f(K)$ for $T\geqslant 200$ eV towards the smaller $K$ values which are not mapped by experiment (see Fig. 1) is believed to be highly accurate since the higher-order terms in the expansion of Eq. (5) are negligible for small $K$ and small errors in

TABLE I. Differential cross sections for $2^1P$ excitation of helium. The "theoretical" cross sections of column 6 have been calculated with Eqs. (3) and (5) and with the $c$ values of column 7.

| $T$ (eV) | $\theta$ (deg) | $(Ka_0)^2$ | $I$ | $I\sin\theta/\sigma(\Omega)a_0^{-2}$ | $\sigma_{2^1P}(\Omega)/a_0^2$ | $c$ |
|---|---|---|---|---|---|---|
| 400 | 5 | 0.239 | 10 000 | 452 | 1.928 | 0.1 |
| | 7.5 | 0.511 | 1985±20 | 440 | 0.589 | |
| | 10 | 0.891 | 502±20 | 447 | 0.195 | |
| 300 | 5 | 0.190 | 10 000 | 336 | 2.597 | 0.1 |
| | 7.5 | 0.392 | 2400±40 | 344 | 0.911 | |
| | 10 | 0.674 | 672±20 | 337 | 0.346 | |
| 225 | 5 | 0.158 | 10 000 | 268 | 3.249 | 0.1 |
| | 7.5 | 0.308 | 2750±80 | 274 | 1.310 | |
| | 10 | 0.517 | 877±25 | 270 | 0.564 | |
| | 15 | 1.111 | 119±10 | 272 | 0.113 | |
| 200 | 5 | 0.149 | 10 000 | 251 | 3.472 | 0.1 |
| | 10 | 0.466 | 977±30 | 253 | 0.671 | |
| | 15 | 0.991 | 146±10 | 255 | 0.148 | |
| 175 | 5 | 0.142 | 10 000 | 240 | 3.635 | −0.1 |
| | 10 | 0.417 | 1065±30 | 235 | 0.786 | |
| | 15 | 0.872 | 168±10 | 230 | 0.189 | |
| 150 | 5 | 0.137 | 10 000 | 232 | 3.755 | −0.1 |
| | 10 | 0.370 | 1250±50 | 230 | 0.944 | |
| | 15 | 0.756 | 230±15 | 234 | 0.255 | |
| 100 | 5 | 0.142 | 10 000 | 254 | 3.431 | −0.1 |
| | 10 | 0.291 | 1910±40 | 254 | 1.307 | |
| | 15 | 0.537 | 465±30 | 252 | 0.477 | |
| | 20 | 0.880 | 131±30 | 256 | 0.175 | |

[11] E. N. Lassettre and E. A. Jones, J. Chem. Phys. **40**, 1218 (1964).

[12] S. M. Silverman and E. N. Lassettre, J. Chem. Phys. **40**, 1265 (1964).

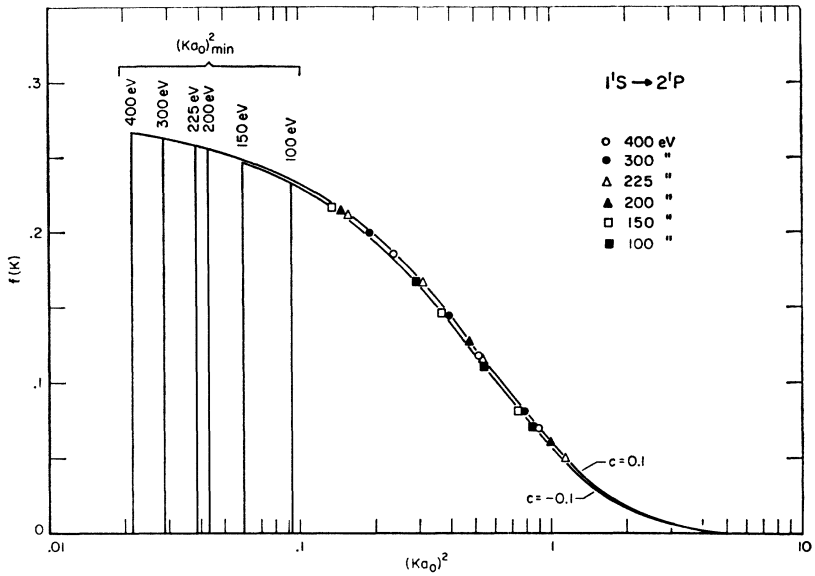


FIG. 1. Apparent generalized oscillator strengths for the transition $1^1S \rightarrow 2^1P$ in helium. The solid lines are obtained from Eq. (5) with $c_\nu = 0$ for $\nu \geq 2$. The circles, triangles, and squares are obtained from the experimental data.

$c_1$ will have very small effect on $f(K)$. The analytical extrapolation of the $f(K)$ for $T \geq 200$ eV towards large $K$ is not as good because all $c_\nu$ coefficients in Eq. (5) become equally important in the limit of $K \rightarrow \infty$. However, $f(K)$ is very small for large $K$. The way of plotting our results in Fig. 1 is similar to that first used by Miller and Platzman[13] and is highly convenient because the areas under the curves are directly proportional to the total excitation cross section. The apparent $f(K)$ seem to decrease a little faster with increasing $K$ for $T < 200$ eV than for $T \geq 200$ eV, indicating small departures from the Born approximation. In the region of $T$ values in which the Born approximation is not valid, there are no *a priori* reasons to believe that Eq. (5) is still useful as an analytic representation of the apparent $f(K)$. However, from column 5 of Table I and from Fig. 1 it follows that Eq. (5) with only one adjustable constant ($c_1$) still works very well for small $T$. It also follows that, for instance for $T = 100$ eV, nearly the complete region of $K$ values of interest is mapped by experiment. The only question which remains is whether the normalization on $f(0)$ is still valid for small $T$.

TABLE II. Total cross sections for $2^1P$ excitation in helium.

| $T$ (eV) | $100\sigma_{2^1P}/\pi a_0^2$ Present[a] | MHS[b] | Vriens[c] | OB[d] | Miller[e] | Present/MHS | Present/Miller |
|---|---|---|---|---|---|---|---|
| 100 | 14.0[f] | 9.89 | (15.3) | 13 | 13.9 | 1.42[f] | 1.01[f] |
| 150 | 11.9 | 9.21 | (12.8) | 11 | 12.8 | 1.29 | 0.93 |
| 175 | 11.0 | ... | (11.8) | ... | ... | ... | ... |
| 200 | 10.4 | 8.07 | 10.9 | 9.9 | 11.2 | 1.29 | 0.93 |
| 225 | 9.74 | ... | 10.2 | ... | ... | ... | ... |
| 300 | 8.22 | 6.58 | 8.6 | 7.9 | 8.8 | 1.25 | 0.93 |
| 400 | 6.85 | 5.73 | 7.1 | 6.6 | 7.3 | 1.20 | 0.94 |
| 600 | 5.21 | 4.40 | 5.4 | ... | 5.5 | 1.18 | 0.95 |
| 800 | 4.25 | 3.83 | 4.4 | ... | 4.5 | 1.11 | 0.94 |
| 1000 | 3.62 | 3.29 | 3.7 | ... | 3.8 | 1.10 | 0.95 |
| 1500 | 2.67 | 2.39 | 2.7 | ... | ... | 1.12 | ... |
| 2000 | 2.14 | 1.92 | 2.2 | ... | ... | 1.11 | ... |

[a] Calculated with Eqs. (4) and (5) and the $c$ values of Table I.
[b] Moustafa *et al.* (Ref. 2).
[c] Vriens (Ref. 9).
[d] Ochkur and Brattsev (Ref. 4).
[e] Miller (Ref. 16).
[f] These values for $T = 100$ eV may be slightly too high (e.g., 4 to 8%) because of our normalization on $f(0)$.

Comparison of our results with those of other investigators shows that, apart from the normalization on $f(0)$, our apparent $f(K)$ for large $T$ decrease faster with increasing $K$ than found by Altshuler[14] (theory), Lassettre and Jones[11] (theory), Silverman and Lassettre[12] (theory and experiment), Lassettre, Krasnow, and Silverman[15] (experiment), and Miller and Platzman.[13] The data of Refs. 11–15 all lead to a $c$ value in Eq. (5) of about 0.86. For $(Ka_0)^2 = 0.5$ our $f(K)$ is lower by about 10% and for $(Ka_0)^2 = 1$ by about 17%. For small $T$ no previous apparent $f(K)$ have been reported.

Total $2^1P$ cross sections calculated with Eqs. (4) and (5) and the $c$ values of Table I are given in Table II. The values for $T > 400$ eV are calculated with $c = 0.1$. Since the Born approximation seems to be valid above 200 eV, we should not expect any variation in $c$ for $T > 400$ eV. The present data are compared with optically measured cross sections of Moustafa *et al.*,[2] cross sections calculated[9] with Eqs. (4) and (5) and $c = 0.86$ as obtained from Lassettre's[11,12] data, theoretically calculated cross sections of Ochkur and Brattsev,[4] and cross sections extrapolated by Miller[16] from all experimental (optical) and theoretical cross sections known in 1956. Ratios of the present and optical cross sections are also given in Table II. Moustafa *et al.*[2]

[13] W. F. Miller and R. L. Platzman, Proc. Phys. Soc. (London) **70**, 299 (1957).

[14] S. Altshuler, Phys. Rev. **87**, 992 (1952); **89**, 1093 (1953).
[15] E. N. Lassettre, M. E. Krasnow, and S. M. Silverman, J. Chem. Phys. **40**, 1242 (1964).
[16] W. F. Miller, Ph.D. thesis, Purdue University, 1956 (unpublished).

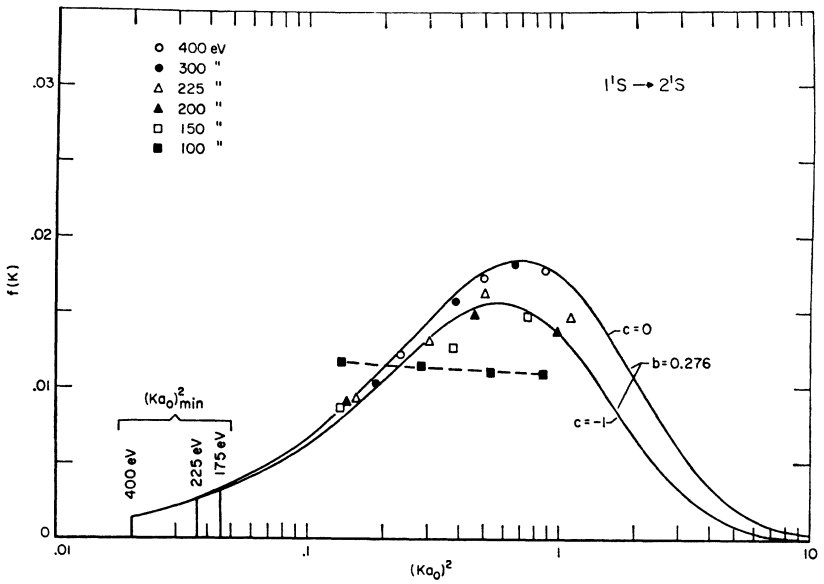


FIG. 2. Apparent generalized oscillator strengths for the transition $1^1S \rightarrow 2^1S$ in helium. The solid lines are obtained from Eq. (7). The circles, triangles, and squares are obtained from the experimental data, via Eq. (6). The dashed curve for 100 eV is drawn through the experimental points.

measured absolute $n^1P$ cross sections for $n \geq 3$, but measured relative $2^1P$ cross sections. They normalized their relative data via the optical oscillator strength. In a plot of $TE\sigma_{2^1P}/4\pi a_0^2R^2$ against $\ln(T/R)$ a straight line was obtained[2] above 100 eV; the slope of this straight line must be equal to $f(0)$. That the ratio of our cross sections and those of Ref. 2 decreases with increasing $T$ between 100 and 200 eV may partly be due to the normalization of our apparent relative $f(K)$ on $f(0)$ and partly to possible energy-dependent errors in the cross sections of Ref. 2. Such possible energy-dependent errors may be responsible for the decrease in the ratio for $T > 200$ eV.

## V. THE $1^1S \rightarrow 2^1S$ TRANSITION

Values of $I_{2^1S}/I_{2^1P}$ and their estimated uncertainties are listed in column 4 of Table III. Absolute $2^1S$ cross sections calculated with

$$\sigma_{2^1S}(\Omega) = (I_{2^1S}/I_{2^1P})\sigma_{2^1P}(\Omega) \qquad (6)$$

and with the $2^1P$ cross sections of Table I are given in column 5 of Table III.

Since $\sigma_{2^1S}(\Omega)$ decreases more slowly with increasing $\theta$ than does $\sigma_{2^1P}(\Omega)$, the problem with double scattering is less severe for $2^1S$ than for $2^1P$ excitation. In the normalization of $\sigma_{2^1S}(\Omega)$ via $\sigma_{2^1P}(\Omega)$ [Eq. (6)], the effect of double scattering on $2^1P$ cancels out, since if $\sigma_{2^1P}(\Omega)$ would be too high due to double scattering, then $I_{2^1P}$ is also too high by the same amount.

For an analytical extrapolation of our results, we use the formula (first given by Lassettre,[17] see also Ref. 9)

$$f(K) = \frac{bx}{(1+x)^6}\left\{1+\frac{cx}{1+x}\right\}, \qquad (7)$$

with $x = (Ka_0/\alpha)^2$ and $\alpha^2 = 3.551$. By fitting the coefficients $b$ and $c$ to his experimental data Lassettre[17] obtained $b = 0.319$ and $c = 0.132$ (Lassettre gave an $\alpha^2$ value of 3.572). With an improved apparatus Skerbele and Lassettre[18] later found $b = 0.352$ with $c = 0$. In the region of $T$ values in which the Born approximation holds, the ratio of the "experimental" $\sigma_{2^1S}(\Omega)$ obtained via Eq. (6) and given in column 5 of Table III and the "theoretical" $\sigma_{2^1S}(\Omega)$ obtained via Eqs. (3) and (7) must be equal to 1 and independent of $\theta$. For large $T$ we obtain this result with the $b$ and $c$ values of Table III as illustrated in column 6 of this table. The $\sigma_{ex}(\Omega)$ are

TABLE III. Ratios of experimental $2^1S$ and $2^1P$ scattered intensities and $2^1S$ cross sections for helium.

| $T$ (eV) | $\theta$ (deg) | $(Ka_0)^2$ | $10^4 I_{2^1S}/I_{2^1P}$ | $10^3\sigma_{2^1S}(\Omega)/a_0^2$ | $\sigma_{expt}(\Omega)/\sigma_{theoret}(\Omega)$ | $b$ | $c$ |
|---|---|---|---|---|---|---|---|
| 400 | 5 | 0.238 | 684± 25 | 132 | 0.99 | 0.271 | 0 |
| | 7.5 | 0.510 | 1490± 30 | 87.8 | 1.00 | | |
| | 10 | 0.890 | 2650± 30 | 51.7 | 1.01 | | |
| 300 | 5 | 0.189 | 567± 25 | 147 | 1.01 | 0.282 | −0.3 |
| | 7.5 | 0.391 | 1130± 40 | 103 | 0.98 | | |
| | 10 | 0.674 | 1990± 50 | 68.9 | 1.01 | | |
| 225 | 5 | 0.156 | 455± 15 | 148 | 1.01 | 0.276 | −0.6 |
| | 7.5 | 0.306 | 840± 20 | 110 | 0.97 | | |
| | 10 | 0.515 | 1420± 30 | 80.1 | 1.00 | | |
| | 15 | 1.111 | 2950± 80 | 33.3 | 1.02 | | |
| 200 | 5 | 0.147 | 427± 15 | 148 | 1.01 | 0.275 | −1.0 |
| | 10 | 0.464 | 1200± 30 | 80.5 | 0.98 | | |
| | 15 | 0.990 | 2350± 90 | 34.8 | 1.00 | | |
| 175 | 5 | 0.139 | 425± 10 | 154 | 1.01 | 0.286 | −1.1 |
| | 10 | 0.415 | 1083± 30 | 85.1 | 0.94 | | |
| | 15 | 0.871 | 2330±100 | 44.0 | 1.05 | | |
| 150 | 5 | 0.134 | 420± 10 | 158 | 1.05 | 0.289 | −1.5 |
| | 10 | 0.367 | 910± 30 | 85.9 | 0.91 | | |
| | 15 | 0.754 | 1900± 80 | 48.5 | 1.05 | | |
| 100 | 5 | 0.137 | 596± 5 | 204 | 1.21 | 0.363 | −3.1 |
| | 10 | 0.286 | 730± 10 | 95.4 | 0.82 | | |
| | 15 | 0.534 | 1020± 80 | 48.7 | 0.79 | | |
| | 20 | 0.877 | 1675± 40 | 29.3 | 1.18 | | |

[17] E. N. Lassettre, J. Chem. Phys. 43, 4479 (1965).

[18] A. Skerbele and E. N. Lassettre, J. Chem. Phys. 45, 1077 (1966).

taken from column 5 and the $\sigma_{th}(\Omega)$ are calculated via Eqs. (3) and (7) with the $b$ and $c$ values of columns 7 and 8. By comparing Eqs. (5) and (7) and the $c$ values of Tables I and III, we see that the relative shape of $f(K)$ as a function of momentum transfer changes much more and already at higher $T$ for $2^1S$ than for $2^1P$ excitation. Hence the Born approximation breaks down at higher $T$ for $2^1S$ than for $2^1P$ excitation. This behavior is further illustrated in Fig. 2. The apparent $f(K)$ calculated via Eq. (6) are given by circles, squares, and triangles. The analytical $f(K)$ calculated via Eq. (7) with $b=0.276$ (average of 400-, 300-, 225-, and 200-eV data) and some of the $c$ values of Table III are given by solid lines. Figure 2 also shows that the Born approximation breaks down in a very different way for different $K$, and for $T>150$ eV more for large than for small $K$. Our observations are consistent with the results of Moustafa *et al.*[2] for total $n^1S$ and $n^1P$ excitation cross sections.

Total $2^1S$ cross sections calculated with Eqs. (4) and (7), with $b=0.276$ and with the $c$ values of Table III, are given in Table IV. No cross sections for $T<200$ eV are given, since Eq. (7) does not seem to be satisfactory for $T<200$ eV (see column 6 of Table III) and because the range of $K$ values of interest is not sufficiently mapped by experiment. Further, no extrapolated data for $T>400$ eV are given since it is very doubtful (see also Ref. 2) whether the Born limit has been reached at $T=400$ eV. The present $2^1S$ cross sections are compared with those of Lassettre[17] (experiment plus a similar analysis as made here) and with cross sections extrapolated by Miller[16] from theoretical calculations of Altshuler[14] and from experiment. The increasing disagreement between our and Lassettre's cross sections for smaller $T$ comes from the fact that Lassettre's[17] calculations are based on 500-eV data (see also Ref. 18), so that the variation of $c$ with $T$ was not taken into account.

## VI. THE $1^1S \rightarrow 2^3S$ TRANSITION

Values and estimated uncertainties of the ratio $I_{2^3S}/I_{2^1P}$ of the experimental $2^3S$ and $2^1P$ scattered-electron currents as a function of $T$ and $\theta$ are given in column 4 of Table V and the $2^3S$ cross sections calculated with

$$\sigma_{2^3S}(\Omega)=(I_{2^3S}/I_{2^1P})\sigma_{2^1P}(\Omega) \tag{8}$$

are given in column 5 of Table V.

TABLE IV. Total cross sections for $2^1S$ excitation in helium.

| $T$ (eV) | $1000\sigma_{2^1S}/\pi a_0^2$ Present[a] | Miller[b] | Lassettre[c] |
|---|---|---|---|
| 400 | 4.82 | 3.92 | 5.74 |
| 300 | 6.04 | 5.17 | 7.59 |
| 225 | 7.50 | 6.82 | 9.99 |
| 200 | 7.72 | 7.54 | 11.17 |

[a] Calculated with Eqs. (4) and (7) and with the $c$ values of Table III.
[b] Reference 16.
[c] Reference 17.

TABLE V. Ratios of experimental $2^3S$ and $2^1P$ scattered intensities and differential $2^3S$ cross section for helium.

| $T$ (eV) | $\theta$ (deg) | $(Ka_0)^2$ | $10^5 I_{2^3S}/I_{2^1P}$ | $10^5\sigma_{2^3S}(\Omega)/a_0^2$ | $\eta$ | $d$ |
|---|---|---|---|---|---|---|
| 225 | 5 | 0.154 | 20± 2 | 65 | 3.62 | 3.7 |
| | 10 | 0.513 | 77± 10 | 43 | 3.43 | |
| | 15 | 1.110 | 230± 50 | 26 | 3.52 | |
| 200 | 5 | 0.144 | 27± 2 | 94 | 3.67 | 3.7 |
| | 10 | 0.462 | 110± 10 | 74 | 3.97 | |
| | 15 | 1.000 | 300± 50 | 44 | 3.80 | |
| 175 | 5 | 0.136 | 40± 3 | 145 | 3.79 | 3.7 |
| | 10 | 0.412 | 110± 10 | 86 | 2.96 | |
| | 15 | 0.869 | 380± 50 | 72 | 3.78 | |
| 150 | 5 | 0.130 | 59± 4 | 221 | 3.77 | 3.0 |
| | 10 | 0.364 | 150± 15 | 142 | 3.23 | |
| | 15 | 0.752 | 420± 50 | 107 | 3.76 | |
| 100 | 5 | 0.130 | 174± 10 | 597 | 3.24 | 2.5 |
| | 10 | 0.280 | 367± 25 | 480 | 3.26 | |
| | 15 | 0.529 | 782± 50 | 373 | 3.56 | |
| | 20 | 0.874 | 1020±100 | 179 | 2.63 | |

Because $\sigma_{2^3S}(\Omega)$ decreases slowly with increasing $\theta$, double scattering is negligible for $2^3S$ excitation.

The relationship between the excitation of singlet and triplet states is most easily seen if we express the excitation cross sections in terms of the direct and exchange scattering amplitudes $f$ and $g$. Then the cross section for $2^1S$ excitation is proportional to $|f-g|^2$ and the cross section for $2^3S$ excitation is proportional to $3|g|^2$. The space wave functions for $2^3S$ and $2^1S$ states are both $1s2s$, but are different (see for instance Marriott[19]) due to different spins and consequently $g(2^3S)\neq g(2^1S)$. For large $T$ and not too small $K$, the Ochkur approximation[4] and the Born-Oppenheimer approximation[20] are nearly the same (see also Rudge[21]) and the absolute magnitude of the exchange scattering amplitude is primarily determined by the overlap of the initial- and final-state space wave functions for small $r$. This overlap is larger for $2^3S$ than for $2^1S$ excitation. As a crude estimate we take $|g(2^3S)|^2=2|g(2^1S)|^2$. For large $T$ (e.g., >150 eV) we may further assume that $|g(2^1S)|\ll|f(2^1S)|$. Thus

$$\sigma_{2^3S}(\Omega)/\sigma_{2^1S}(\Omega)\approx 3|g(2^3S)|^2/|f(2^1S)|^2. \tag{9}$$

According to the Ochkur[4] approximation,

$$|g|=(Ka_0)^2(R/T)|f|$$

for scattering of electrons by atomic hydrogen. For scattering of electrons by helium the relationship between $|g|$ and $|f|$ is different and we find (see also Ref. 9) that $|g(2^1S)|=\frac{1}{2}(Ka_0)^2(R/T)|f(2^1S)|$. Equation (2) of Ochkur and Brattsev[4] is incorrect on this point for

[19] R. Marriott, in *Proceedings of the Third International Conference on Electronic and Atomic Collisions*, edited by M. R. C. McDowell (North-Holland Publishing Co., Amsterdam, 1964), p. 114.
[20] J. R. Oppenheimer, Phys. Rev. **32**, 361 (1928).
[21] M. R. H. Rudge, Proc. Phys. Soc. (London) **85**, 607 (1965).

TABLE VI. Ratios of experimental $2^3S$ and $2^1S$ cross sections and the corresponding theoretical ratios as expected from the Ochkur approximation.

| $T$ (eV) | $\theta$ (deg) | $(Ka_0)^2$ | $10^4\sigma_{2^3S}(\Omega)/\sigma_{2^1S}(\Omega)$ (experiment) | $10^4\sigma_{2^3S}(\Omega)/\sigma_{2^1S}(\Omega)$ [Ochkur, Eq. (10)] |
|---|---|---|---|---|
| 225 | 5 | 0.155 | 44 | 1.3 |
| | 10 | 0.514 | 54 | 14 |
| | 15 | 1.110 | 78 | 67 |
| 200 | 5 | 0.146 | 63 | 1.4 |
| | 10 | 0.463 | 92 | 15 |
| | 15 | 0.995 | 128 | 68 |
| 175 | 5 | 0.138 | 94 | 1.7 |
| | 10 | 0.414 | 102 | 16 |
| | 15 | 0.870 | 163 | 69 |
| 150 | 5 | 0.132 | 140 | 2.2 |
| | 10 | 0.366 | 165 | 17 |
| | 15 | 0.753 | 221 | 70 |
| 100 | 5 | 0.134 | 292 | 5 |
| | 10 | 0.283 | 503 | 22 |
| | 15 | 0.532 | 767 | 79 |
| | 20 | 0.876 | 609 | 213 |

singlet excitation of helium. From the above relations it follows that

$$\sigma_{2^3S}(\Omega)/\sigma_{2^1S}(\Omega)\approx\tfrac{3}{2}(Ka_0)^4(R/T)^2. \quad (10)$$

The experimental and "theoretical" [Eq. (10)] ratios $\sigma_{2^3S}(\Omega)/\sigma_{2^1S}(\Omega)$ are given in columns 4 and 5 of Table VI. Since the $(Ka_0)^2$ values for given $T$ and $\theta$ are slightly different for $2^3S$ and $2^1S$ excitation, we used in the calculation the average values listed in column 3 of Table VI. From Table VI it follows that the Ochkur approximation predicts an incorrect angular dependence of the $1^1S\rightarrow 2^3S$ cross section and much too small cross sections for small $\theta$. The $2^3S$ cross sections given in Table V cannot be compared with other theoretical or experimental differential cross sections since none exist.

We therefore try to get estimates of total $2^3S$ cross sections from our differential cross sections. In a previous paper[9] and in the previous sections it is shown that knowledge of $\sigma_{2^1P}(\Omega)$ or $\sigma_{2^1S}(\Omega)$ for only one or two suitable angles is sufficient to make accurate estimates of $\sigma_{2^1P}(\Omega)$ or $\sigma_{2^1S}(\Omega)$ for other angles and thus to calculate the total $\sigma_{2^1P}$ or $\sigma_{2^1S}$. In the case of $2^3S$ excitation, however, we know much less about the nature of the differential cross sections. In an empirical way we found that the analytical formula

$$\sigma_{2^3S}(\Omega)=\eta a_0^2\left(\frac{R}{T}\right)^3\left(\frac{T-E}{T}\right)^{1/2}\left[1+\frac{(Ka_0)^2}{d}\right]^{-4} \quad (11)$$

fits the experimental data when $\eta$ and $d$ take the values of Table V. Estimates of total cross sections are obtained by replacing $\eta$ by $\bar{\eta}$ (Table VII) and subsequently integrating Eq. (11) over all scattering angles. This gives

$$\sigma_{2^3S}/\pi a_0^2\approx\bar{\eta}(R/T)^4(\tfrac{1}{3}d). \quad (12)$$

Note that $\bar{\eta}$ and $d$ may be $T$-dependent. Since $\sigma_{2^3S}$ is expected to decrease with $T^{-3}$ with increasing $T$ for sufficiently high $T$, we also calculated $(T/R)^3\sigma_{2^3S}/\pi a_0^2$ using the above values of $\bar{\eta}$ and $d$. The results are shown in Table VII and in Fig. 3 together with theoretical results of Refs. 4 and 5. Within the "experimental" accuracy (see Sec. VII) our data are consistent with the expectation that $\sigma_{2^3S}$ decreases with $T^{-3}$ and we find $(T/R)^3\sigma_{2^3S}/\pi a_0^2\approx 0.32$ for $T$ between 100 and 225 eV. The theories[4,5] suggest that $(T/R)^3\sigma_{2^3S}/\pi a_0^2\approx 1.48$ for $T>200$ eV, a factor 4.6 higher than our result. Concerning our choice of Eq. (11), we note that $\sigma_{2^3S}(\Omega)$ according to Eq. (11) decreases with $K^{-8}$ for large $K$ as we expect to be correct on basis of the Born-Oppenheimer and Ochkur approximations. This large $K$ behavior follows from general considerations, such as given in Ref. 9 for singlet excitation, plus Eq. (10) of this paper. The zero angle cross sections according to Eq. (11) decrease more slowly with increasing $T$ than the Ochkur zero-angle cross sections.

## VII. DISCUSSION

### A. Present Experiment

Estimates of random and systematic errors were obtained by repeating the measurements several times over a period of three months in which time the apparatus had been disassembled twice. The angular dependence of the $2^1P$ and $2^1S$ scattered current was reproducible within a few percent if the electrical alignment of the electron spectrometer (monochromator) was not changed. The scattered current for $2^3S$ excitation is much smaller and hence the random errors in the $I_{2^3S}/I_{2^1P}$, and thus in $\sigma_{2^3S}(\Omega)$, are larger.

Because of residual asymmetries, the angular dependences for positive and negative angles were often found to be different and, as expected, to change with different electrical alignments. In a typical measurement for $2^1P$ excitation we found $c=0.3$ and $-0.1$ for positive and negative angles for $T=300$ eV, and $c=-0.9$ and 0.7 for positive and negative angles for $T=100$ eV. A large part of the systematic error due to asymmetries was removed by averaging over positive and negative angles. The average angular dependences and intensity ratios repro-

TABLE VII. Average $\eta$ values, total $2^3S$ cross sections, and total $2^3S$ cross sections times $(T/R)^3$.

| $T$ (eV) | $\bar{\eta}$ | $10^4\sigma_{2^3S}/\pi a_0^2$ | $(T/R)^3\sigma_{2^3S}/\pi a_0^2$ |
|---|---|---|---|
| 225 | 3.52 | 0.58 | 0.26 |
| 200 | 3.81 | 1.01 | 0.32 |
| 175 | 3.51 | 1.58 | 0.33 |
| 150 | 3.59 | 2.43 | 0.33 |
| 100 | 3.17 | 9.05[a] | 0.36[a] |

[a] These values may be too high by about 4 to 8% due to our normalization on $f(0)$.

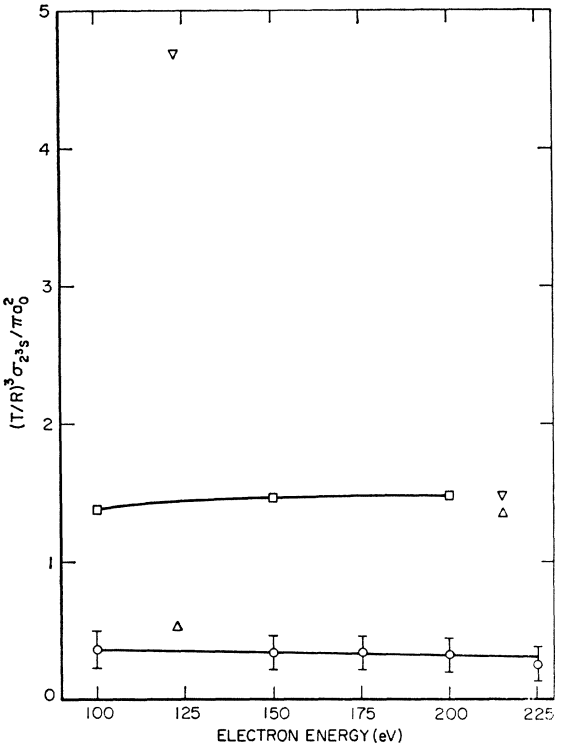


FIG. 3. Total $2^3S$ cross sections times $(T/R)^3$ versus $T$. The open circles are our present results obtained via Eq. (12). The triangles $\triangle\triangle$ are obtained with the first-order exchange approximation (Ref. 5), the triangles $\nabla\nabla$ with the Born-Oppenheimer approximation (Ref. 5), and the squares with the Ochkur approximation by Ochkur and Brattsev (Ref. 4).

duced much better (see error margins in Tables I, III, and V) than did those for either the positive or negative angles. However, since we can only rotate the analyzer around one axis, we could thus only correct for a part of the asymmetry in the plane perpendicular to the axis of rotation. We cannot correct for asymmetries in the plane through the geometrical axis and the axis of rotation. Further, the finite angular width of the electron beam entering the scattering chamber may lead to an apparent too slow decrease of $\sigma(\Omega)$ with $\theta$ and thus to slightly too large $c$. An additional source of small systematic errors may be the finite acceptance angle of the analyzer.

Our best estimates of the uncertainties in the $c$ values of Table I are $\pm 0.2$ for $T \geqslant 200$ eV, $\pm 0.25$ for $T = 150$ and 175 eV, and $\pm 0.3$ for $T = 100$ eV. The uncertainties in the $c$ values of Table III will be about the same.

The systematic errors due to our normalization of the relative $f(K)$ on $f(0) = 0.27616$ are difficult to estimate. The presently available data (last two columns of Table II) suggest that the normalization on $f(0)$ for $T = 100$ eV leads to cross sections which are about 4 to 8% too high. Reasonable estimates of the systematic errors for larger $T$ seem to be 3% for 150 eV, 2% for 175 eV, 1% for 200 eV, and less than 1% for 225 eV. For small $T$, for instance 100 eV, the normalization of our relative data on $f(0)$ may thus be a "large" source of error for all transitions. A better normalization for small $T$ may be obtained either on basis of absolute differential-cross-section measurements, which have not yet been carried out, or on basis of more accurate total cross sections which may be obtained via optical measurements.

If we finally assume that use of Eq. (11) can lead to errors of about 30% in $\sigma_{2^3S}$, we get an uncertainty of the total $2^3S$ cross sections of about 40%.

### B. Lassettre's Experiment

The data of Lassettre and co-workers[11,12,15] for the transition $1^1S \rightarrow 2^1P$, lead via our analyzing procedure[9] to $c = 0.86$ for large $T$ contrary to our value $c = 0.1 \pm 0.2$. In an attempt to explain this difference we made a new analysis of their data. Lassettre and co-workers normalized their (relative) experimental $f(K)_{ex}$ on their theoretical $f(K)_{th}$. However, for $0.4 < (Ka_0)^2 < 1.0$ their $f(K)_{ex}$ decrease faster with increasing $K$ than their $f(K)_{th}$ (see Fig. 1 of Ref. 12). Applying our analyzing technique to these $f(K)_{ex}$ gave $c = 0.2$ instead of $c = 0.86$. This $c$ value of 0.2 may further be slightly too high due to convergence or divergence of the electron beam and finite angular resolution of the analyzer. The apparatus used in Refs. 12 and 15 was not yet as sophisticated as the newer electron spectrometers used in the present work[6] and used in more recent work of Lassettre and co-workers.[18,22,23] The $c = 0.2$ value obtained from the experimental data of Refs. 12 and 15 agrees with our value within the experimental accuracy. The apparent decrease in $f(K)_{ex}/f(K)_{th}$ in Fig. 1 of Ref. 12 for $(Ka_0)^2 < 0.3$ may be due to finite angular resolution of the apparatus. We find a similar effect for smaller $K$. The apparent increase in $f(K)_{ex}/f(K)_{th}$ in Fig. 1 of Ref. 12 for $(Ka_0)^2 > 1.1$ may well be due to double scattering.

### C. Optical Excitation Functions

While the differential-cross-section measurements of Refs. 12 and 15 suggest a slightly larger $c$ value than found here, and thus larger total cross sections, the total optical cross sections of Moustafa *et al.*[2] are smaller than our total $2^1P$ cross sections. Since our normalization on $f(0)$ is believed to be very good for $T \geqslant 200$ eV, this discrepancy can, as mentioned before, perhaps be explained by possible energy-dependent errors in the relative optical excitation function of Moustafa *et al.*[2] Otherwise our $c$ value(s) should be much too large which seems unlikely to be the case. Energy-dependent errors in the optical $2^1P$ cross sections may be caused[2] by polarization effects.

[22] V. D. Meyer, A. Skerbele, and E. N. Lassettre, J. Chem. Phys. 43, 805 (1965).
[23] E. N. Lassettre, A. Skerbele, and V. D. Meyer, J. Chem. Phys. 45, 3214 (1966).

Excitation of the $2^3S$ and $2^1S$ states cannot be measured optically and no direct comparison of optical data and our data is possible.

### D. Theoretical Cross Sections

The wave functions used in the theoretical calculations for $2^1P$ excitation[11,12,14] are not believed to be highly accurate so that no direct conclusions can be drawn from agreement or disagreement of our data and theoretical cross sections for large $T$.

Part of the discrepancy between our results and the theoretical $2^3S$ cross sections may be due to the approximate nature of the wave functions used in the theoretical calculations.[4,5]

## VIII. CONCLUSIONS

The differential cross sections and generalized oscillator strengths for $2^1P$ and $2^1S$ excitation seem to decrease faster with increasing $K$ than previously found by Silverman and Lassettre.[12] Since Lassettre and coworkers normalized their $f(K)$ values and differential cross sections for other transitions and for other atoms and molecules on their $f(K)$ for the transition $1^1S \rightarrow 2^1P$ in He, their other data might also be too high for large $K$. Consequently, if total cross sections are calculated on the basis of Lassettre's results, these total cross sections may also be too high. One of us,[9] for instance, used the $f(K)$ of Ref. 15 for the transition $1^1S \rightarrow 3^1P$ in He to calculate total $3^1P$ cross sections and obtained higher total $3^1P$ cross sections than found in the optical measurements of Ref. 2. A large part of this discrepancy may thus be due to the possibility that the $f(K)$ of Ref. 15 are too high for large $K$. For $2^1P$ excitation very small departures from the Born approximation are found below 200 eV. Departures from the Born approximation appear to be more serious for $2^1S$ excitation and already occur for this transition at higher incident electron energies. Our observation that the apparent $f(K)$ for the transition $1^1S \rightarrow 2^1S$ are larger than the Born $f(K)$ for small $K$ and small $T$ (100 eV), is consistent with a previous analysis of zero-angle cross sections for 30 eV$<T<$80 eV by Heideman and Vriens.[24] The apparent $f(K)$ for the transition $1^1S \rightarrow 2^1S$ derived from these zero-angle cross sections were also much larger than the Born $f(K)$ (see Fig. 3 of Ref. 24).

The angular dependence found here for $2^3S$ excitation entirely disagrees with the angular dependence predicted by the Ochkur approximation. However, our results certainly do not exclude the possibility that the Ochkur approximation becomes applicable for instance for $T>400$ eV and $(Ka_0)^2>1$. That the present total $2^3S$ cross sections are much lower than the theoretical ones, may be due to failure of the theoretical approximations or use of not sufficiently accurate wave functions or both.

## ACKNOWLEDGMENTS

We are grateful to U. Fano, J. W. Cooper, C. E. Kuyatt, M. Inokuti, and G. E. Chamberlain for stimulating interest and valuable discussions concerning this work, and to F. J. de Heer and H. R. Moustafa for the permission to use their optical $2^1P$ cross sections prior to publication. One of us (L. V.) wishes to especially thank J. Arol Simpson and K. G. Kessler for enabling him to work as a guest worker in the Atomic Physics Division.

[24] H. G. M. Heideman and L. Vriens, J. Chem. Phys. **46**, 2911 (1967).