Analysis of Low-Energy Electron Scattering by Helium, Neon, and Argon*

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A least-squares method has been used to fit Ramsauer and Kollath's data on the scattering of electrons from helium, neon, and argon with a set of four phase shifts. All the known experimental points between 1.0 eV and an energy 0.6 eV below the threshold for inelastic scattering in each gas were analyzed. The effect of spin-orbit splitting on the description of the scattering has been neglected, as usual. The angular distributions and the total cross sections computed from the four phase shifts at each energy agree well with the original data, except possibly for argon, where more than four phase shifts are probably required. A graphical comparison has been made between the variation with energy of the phase shifts so determined and various recent theoretical predictions, as well as Andrick and Ehrhardt's recent direct measurements.

INTRODUCTION

IN recent years, there has been a resurgence of interest in the scattering of low-energy electrons from rare-gas atoms. Of importance in this connection are the resonances which have recently been discovered[1–6] in electron scattering from helium, neon, and argon at energies slightly below the threshold for inelastic scattering. It has been suggested[7] that in neon and argon these resonances may lead to spin polarization of the scattered electron beam. An important parameter in the analysis of the polarization is the so-called potential-scattering phase shift, that is, the nonresonant part of the phase shift associated with the partial wave involved in the resonance.

The work to be described here grew out of an attempt to derive reliable values for the potential-scattering phase shifts in neon and argon from a least-squares analysis of existing experimental data. Our aim was to find phase shifts which would reproduce the experimental angular distribution of the elastic scattering as a function of energy, and then to extrapolate the phase shifts into the resonance region. Except for Westin's calculations with an analog computer,[8] a systematic analysis of this kind, using modern computer techniques, does not seem to have been carried out previously. As the work progressed, we realized that the computed variation of phase shifts with energy would be of more general interest than in connection with polarization effects alone. For this reason, we have also extended our analysis to helium.

In every case, we have restricted ourselves to an energy region extending from a lower limit of about 1.0 eV to an upper limit of about 0.6 eV below the threshold for inelastic scattering. The lower limit was chosen arbitrarily because at 1.0 eV and lower electron energies, the experimental data do not lend themselves very readily to a least-squares analysis, as may be seen from the rather irregular variation of the very small phase shifts computed for $l \gtrsim 1$ as 1.0 eV is approached through decreasing energy values. The upper limit is set by the occurrence of the scattering resonances mentioned previously.

To obtain the scattering phase shifts from the experimental angular distributions, a least-squares method was employed. In such a computation, absolute values of the differential cross sections are needed. It is remarkable that the only extensive absolute measurements in the energy region of interest here were made by Ramsauer and Kollath in 1932.[9] Our analysis is based entirely on these data.

We have attempted to compare the computed dependence of phase shifts on energy with theoretical predictions, and with the experimental phase shifts in the resonance region recently determined by Andrick and Ehrhardt[6] from the resonance parameters in helium and neon.

METHOD OF COMPUTATION

With the exception of the rare isotopes of helium and neon, electron scattering from the lighter noble gases can be described as a scattering of spin-$\frac{1}{2}$ by spin-0 particles. Under these circumstances in general, there are two phase shifts, $\delta_{l,l-1/2}$ and $\delta_{l,l+1/2}$, associated with every partial wave of angular momentum lh. The differential scattering cross section of initially unpolarized electrons will then be given by

$$\sigma(\theta)=|g(\theta)|^2+|h(\theta)|^2, \quad (1)$$

where

$$g(\theta)=(2ik)^{-1}\{\exp(2i\delta_0)-1+\sum_{j=l-1/2}^{l+1/2}\sum_{l=1}^{\infty}(j+\tfrac{1}{2}) \times[\exp(2i\delta_{lj})-1]P_l(\cos\theta)\}, \quad (2)$$

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TABLE I. Elastic scattering phase shifts in degrees of arc for helium, as derived from the experimental data of Ramsauer and Kollath (Ref. 9). The symbol E refers to the electron energy in eV; σ is the total cross section computed from the four listed phase shifts in units of 10^{-16} cm^2; σ_{exp} refers to the total cross section measured directly by Ramsauer and Kollath (Ref. 9), in the same units. The numbers in square brackets refer to Ehrhardt's recent measurements (Ref. 6).

E	δ_0	δ_1	δ_2	δ_3	σ	σ_{exp}
1.8	332.5	181.57	−0.32	−0.30	5.75	5.91
2.9	322.5	181.61	−0.95	−0.64	6.21	6.19
4.2	314.6	182.38	−1.20	−0.81	5.88	5.88
5.3	309.7	182.65	−1.54	−0.95	5.46	5.46
6.7	303.4	182.81	−2.01	−1.16	5.11	5.06
8.2	298.0	181.32	−3.01	−1.61	4.68	4.67
10.7	287.8	181.12	−4.43	−2.93	4.28	4.21
13.8	294.3	197.00	+5.60	+1.80	3.97	3.70
15.8	293.7	195.94	+6.48	+2.51	3.46	3.45
19.2	289.1	197.26	+6.62	+2.48	3.09	3.14
[19.3]	[289±2]	[200±5]	[0.5]			

$$h(\theta)=(2ik)^{-1}\sum_{l=1}^{\infty}[\exp(2i\delta_{l,l+1/2})-\exp(2i\delta_{l,l-1/2})]\times\sin\theta P_l'(\cos\theta). \quad (3)$$

Here kh is the (nonrelativistic) electron momentum, and θ is the scattering angle in either the laboratory or the center-of-mass system. (The distinction between these systems can be ignored here.)

For a given value of $l\geqslant 1$, the phase shifts $\delta_{l,l-1/2}$ and $\delta_{l,l+1/2}$ would be expected to pass through identical values at slightly different energies, because of the splitting produced by the spin-orbit interaction. In the scattering of p electrons from neon at an energy of approximately 16 eV, the spin-orbit splitting is known to be about 0.1 eV, whereas for 11-eV p electrons scattered by argon, the spin-orbit splitting is approximately 0.17 eV, as inferred from the separation of the resonances which correspond to $P_{1/2}$ and $P_{3/2}$ configurations of the compound atom. In helium, the spin-orbit splitting should be smaller than in neon. From these data and the computed variation of p-wave phase shifts with energy, it is possible to estimate that $\delta_{1,1/2}$ and $\delta_{1,3/2}$ cannot differ by more than 0.5° of arc for any of the gases studied, except in the vicinity of the scattering resonances in neon and argon. Since we have not included the resonance region in our analysis, we can safely ignore the distinction between $\delta_{l,l-1/2}$ and $\delta_{l,l+1/2}$. In that case, Eqs. (1)–(3) reduce to the expression for the differential cross section which applies to the scattering of spin zero by spin-zero particles:

$$\sigma(\theta)=\frac{1}{4k^2}\left|\sum_{l=0}^{\infty}(2l+1)[\exp(2i\delta_l)-1]P_l(\cos\theta)\right|^2. \quad (4)$$

A phase-shift analysis based on this equation was carried out by Westin,[8] with the aid of an analog computer which produced a display of the modulus of the scattering amplitude. This display was compared visually with a plot of the experimental angular distribution. The phase shifts were then varied by trial and error until the two curves appeared to coincide approximately. As many as eight phase shifts could be accommodated on Westin's device. From Westin's results, the largest values of l for which $\delta_l\neq 0$ in the energy range 0–13.6 eV are $l=6$ for helium, $l=3$ for neon, and $l=5$ for argon.

We have restricted our analysis to $l\leqslant 3$. Our procedure for determining the four phase shifts δ_0, δ_1, δ_2, and δ_3 for each gas at the energy values for which Ramsauer and Kollath's data are tabulated[9] is an iterative one based on an approximate linearization of nonlinear equations; as such, it requires an initial approximation to start the iterations. In most cases, we have used as our initial approximations the values interpolated from the tables of phase shifts given by Westin. In a few instances, the initial approximations used were the ones obtained from a fit to the data using only three phase shifts, instead of the four which we have generally used.

If we assume that only the first four phase shifts just mentioned will be significantly different from zero, we can replace the upper limit in Eq. (4) by the number 3, and thus obtain the following explicit expression for the differential cross section:

$$\sigma(\theta)=\sum_{n=0}^{6}F_n(\delta_0,\delta_1,\delta_2,\delta_3)\cos^n\theta. \quad (5)$$

The F_n are nonlinear trigonometric functions of the four phase shifts, and also depend on k.

Since the true values of δ_0, δ_1, δ_2, and δ_3 are not known, we expand F_n in a Taylor series about a set of assumed initial values Δ_0, Δ_1, Δ_2, Δ_3:

$$F_n(\delta_0,\delta_1,\delta_2,\delta_3)\cong F_n(\Delta_0,\Delta_1,\Delta_2,\Delta_3)+\sum_{l=0}^{3}\left[\frac{\partial F_n(\delta_0,\delta_1,\delta_2,\delta_3)}{\partial\delta_l}\right]_{\Delta}\eta_l. \quad (6)$$

TABLE II. Elastic scattering phase shifts in degrees of arc for neon, as derived from the data of Ramsauer and Kollath (Ref. 9). Symbols and units are explained in the caption of Table I. Parentheses enclose digits that are not significant; the values in square brackets are Ehrhardt's recent measurements (Ref. 6).

E	δ_0	δ_1	δ_2	δ_3	σ	σ_{exp}
0.99	350.2	180.10	0.25	−0.05	1.42	1.4(7)
1.17	348.5	180.15	0.41	−0.07	1.64	1.6(4)
1.35	347.4	180.10	0.30	0.07	1.70	1.7(0)
1.67	345.7	179.79	0.30	0.05	1.76	1.7(5)
1.70	345.5	179.78	0.43	0.06	1.78	1.7(5)
1.80	344.9	179.53	0.36	0.13	1.83	1.8(1)
2.20	342.6	179.31	0.27	0.18	1.97	1.9(5)
2.60	340.6	178.73	0.46	0.19	2.06	2.0(4)
2.8	339.6	178.92	0.15	0.22	2.10	2.0(9)
3.6	335.9	177.64	0.32	0.07	2.29	2.2(6)
4.9	330.3	176.52	0.45	0.17	2.51	2.5(2)
6.4	324.3	175.52	−0.37	−0.23	2.69	2.6(9)
7.9	318.3	174.17	0.37	0.85	2.88	2.86
10.4	312.1	169.50	3.41	−1.11	3.09	3.11
13.1	301.3	167.28	3.77	−1.03	3.29	3.25
15.9	290.0	164.26	3.39	−1.18	3.39	3.39
[16.1]	[317±20°]	[155±3]	[10±2]			

TABLE III. Elastic-scattering phase shifts in degrees of arc for argon, as computed from Ramsauer and Kollath's data (Ref. 9). Symbols and units are explained in the caption of Table I. Parentheses enclose digits that are not significant.

E	δ_0	δ_1	δ_2	δ_3	σ	σ_{exp}
1.1	349.0	179.92	1.19	0.16	1.68	1.7(0)
1.5	345.2	178.63	1.96	0.51	2.35	2.3(5)
2.0	338.4	178.42	1.74	0.64	3.44	3.39
2.3	334.9	177.87	1.78	0.98	3.98	3.93
2.4	333.3	177.58	1.75	0.80	4.27	4.24
2.8	332.7	173.54	4.49	−0.42	4.79	4.92
3.2	327.8	172.58	5.25	−0.44	5.63	5.71
3.3	327.0	171.79	4.87	−0.87	5.74	5.80
3.6	323.2	171.46	5.25	−1.26	6.25	6.36
4.0	318.2	170.74	5.73	−1.01	6.88	7.18
5.0	300.3	167.58	6.73	−1.32	9.17	9.11
5.4	294.3	165.72	7.08	−0.83	9.68	9.84
6.7	287.7	165.74	20.33	5.56	12.57	12.7(3)
8.0	287.6	169.33	30.91	8.62	14.91	15.4(1)
9.0	283.9	158.06	34.11	10.72	16.92	17.8(2)
10.3	276.1	141.39	37.51	11.26	19.91	20.4(2)

The subscript Δ indicates that the partial derivative is to be evaluated at $\delta_0=\Delta_0$, $\delta_1=\Delta_1$, $\delta_2=\Delta_2$, and $\delta_3=\Delta_3$. Also, $\delta_l=\Delta_l+\eta_l$, where η_l is the small correction to be found. The corrected value of δ_l, namely, $\Delta_l+\eta_l$, is then used as a new initial value, and the procedure is iterated. For economy of notation, we shall write $F_n(\delta)$ for $F_n(\delta_0,\delta_1,\delta_2,\delta_3)$, and also define

$$f_{n,l}=\left[\frac{\partial F_n(\delta)}{\partial\delta_l}\right]_\Delta.$$

In this notation, Eq. (5) becomes

$$\sigma(\theta)=\sum_{n=0}^{6}[F_n(\Delta)+\sum_{l=0}^{3}f_{n,l}\eta_l]\cos^n\theta. \tag{7}$$

With the knowledge of the experimental differential cross sections $\sigma_E(\theta_s)$ for a number of scattering angles θ_s ($s=1, 2, \cdots, m$), our object is to find those values of the phase shifts which minimize

$$\epsilon=\sum_{s=1}^{m}[\sigma(\theta_s)-\sigma_E(\theta_s)]^2, \tag{8}$$

or

$$\epsilon=\sum_{s=1}^{m}\{\sum_{n=0}^{6}[F_n(\Delta)+\sum_{l=0}^{3}f_{n,l}\eta_l]\cos^n\theta_s-\sigma_E(\theta_s)\}^2. \tag{9}$$

Note that

$$\frac{\partial}{\partial\eta_r}[F_n(\Delta)+\sum_{l=0}^{3}f_{n,l}\eta_l]=f_{n,r}. \tag{10}$$

The minimum condition $\partial\epsilon/\partial\eta_r=0$ ($r=0, 1, 2, 3$) on the free parameters η_l leads to

$$\sum_{s=1}^{m}\{\sum_{n=0}^{6}[F_n(\Delta)+\sum_{l=0}^{3}f_{n,l}\eta_l]\cos^n\theta_s-\sigma_E(\theta_s)\}\times\{\sum_{q=0}^{6}f_{q,r}\cos^q\theta_s\}=0,\quad r=0, 1, 2, 3. \tag{11}$$

This may be considered to be a matrix equation of the form

$$\mathbf{C}\cdot\mathbf{D}=\mathbf{R}, \tag{12}$$

where

$$C_{rl}=\sum_{s=1}^{m}[\sum_{n=0}^{6}f_{n,l}\cos^n\theta_s][\sum_{q=0}^{6}f_{q,r}\cos^q\theta_s], \tag{13}$$

$$D_l=\eta_l,\quad r=0, 1, 2, 3 \tag{14}$$

$$R_r=\sum_{s=1}^{m}[\sigma_E(\theta_s)-\sum_{n=0}^{6}F_n(\Delta)\cos^n\theta_s]\times[\sum_{q=0}^{6}f_{q,r}\cos^q\theta_s]. \tag{15}$$

Given $\sigma_E(\theta_s)$ and the initial values Δ_l, $\mathbf{D}$ can be found as the solution vector to the simultaneous linear equations (12), whereupon the new approximations to the phase shifts δ_l are given by $\Delta_l(\text{new})=\Delta_l(\text{old})+\eta_l$. The procedure is iterated until η_l becomes very small.

The iterative procedure just outlined may converge on a local minimum of the error ϵ instead of the absolute

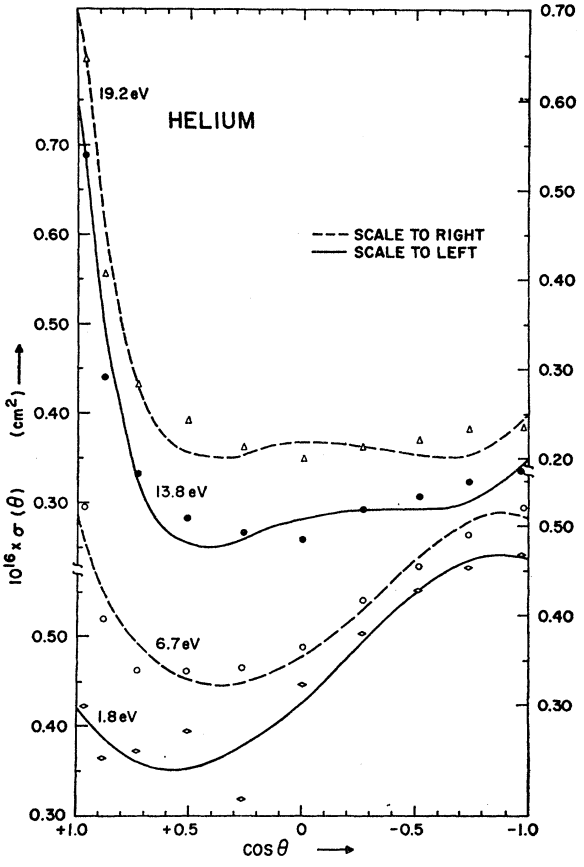

FIG. 1. Comparison between differential scattering cross section for helium computed from the four least-squares phase shifts listed in Table I (dashed and solid lines) and the experimental points of Ramsauer and Kollath on which the analysis is based (discrete points), at electron energies of 1.8, 6.7, 13.8, and 19.2 eV. (Note that the vertical scales for the different energies are displaced with respect to each other.)

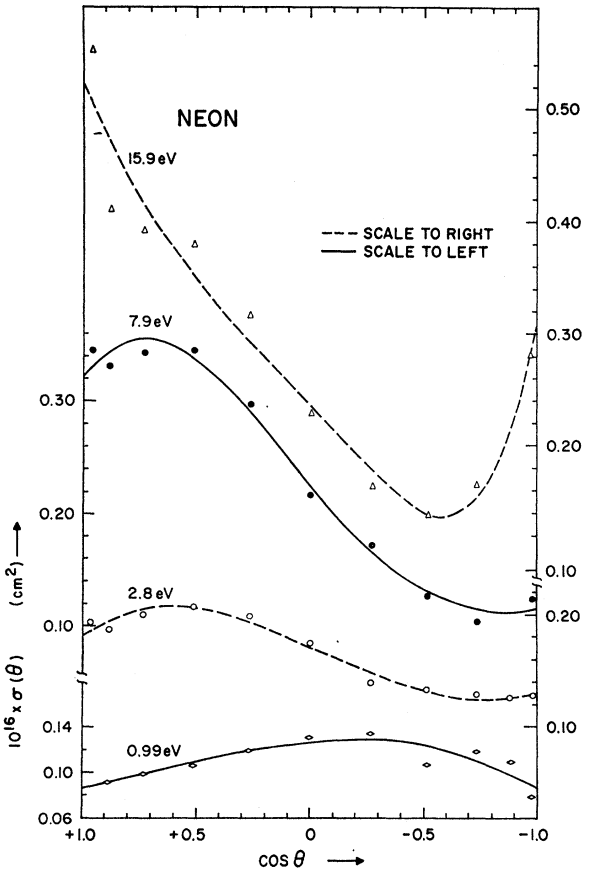

FIG. 2. Differential scattering cross section for neon derived from the four least-squares phase shifts of Table II (dashed and solid lines) compared with the experimental points of Ramsauer and Kollath, at electron energies of 0.99, 2.8, 7.9, and 15.9 eV. (The relative vertical displacement of the different vertical scales should again be noted.)

minimum. It was found in one case that the choice of a new set of initial approximate values Δ_l led to a set of final phase shifts δ_l that reproduced the experimental angular distributions considerably better than an earlier and different set of initial values Δ_l. In most cases, however, the same final δ_l were obtained independent of the initial values Δ_l, although no systematic study of this point was made.

From a detailed consideration of the analytical forms of the functions F_n, it is apparent that the phase shifts are defined only modulo 180° by this procedure. We have been unable to find rigorous conclusions regarding correct absolute zero-energy phase shifts in published discussions of this subject.[10,11] Therefore we have arbitrarily used 360° as the zero-energy limit of δ_0, 180° as the limit of δ_1, and 0° as the limit for all other phase shifts. The total and differential scattering cross sections computed from the phase shifts are unaffected by this arbitrary choice of the zero-energy limits.

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RESULTS OF ANALYSIS

A FORTRAN program was written to apply the procedure outlined above to the experimental data of Ramsauer and Kollath[9] in the energy range from 1 eV to just below the energies corresponding to the scattering resonances in each of the three gases, using Westin's phase shifts[8] as initial approximations, as discussed above. The actual computations were carried out on an IBM 360 computer of the Boston University Computing Center, with the results listed in Tables I–III. The tables also list the total cross sections at each energy, calculated from the least-squares phase shifts, as well as the total cross sections, measured directly by Ramsauer and Kollath.[9]

It will be seen that in the case of argon, the phase shift δ_3, corresponding to the partial wave of the largest angular momentum considered in the analysis, becomes so large for energies greater than about 6 eV that partial waves of still higher angular momentum are probably no longer negligible and should be included

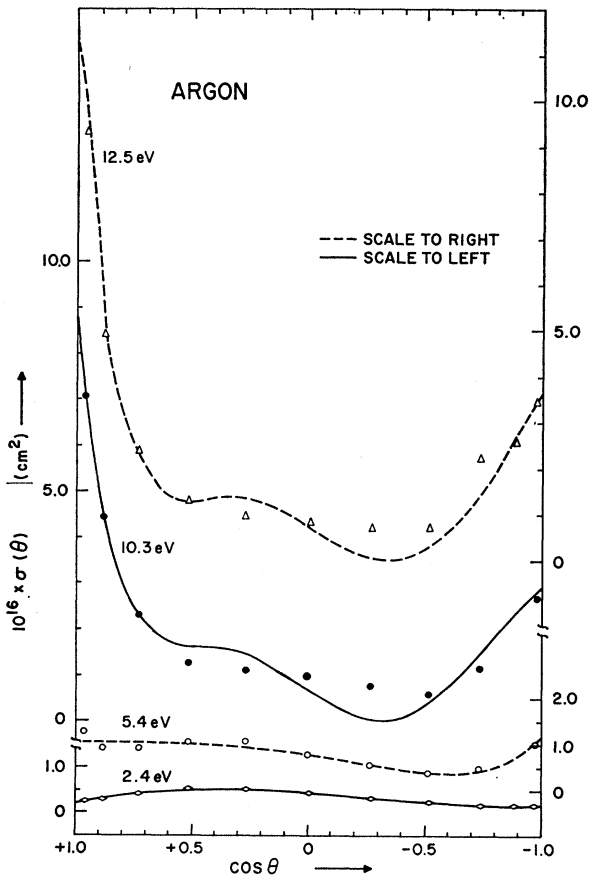

FIG. 3. Differential scattering cross section for argon calculated from the four least-squares phase shifts tabulated in Table III (dashed and solid lines) compared with the experimental points of Ramsauer and Kollath, at electron energies of 2.4, 5.4, 10.3, and 12.5 eV. (The ordinates are again displaced for each energy. Note that 12.5 eV is above the inelastic scattering threshold; the angular distribution at this energy is shown for illustration only.)

in a more careful analysis. In the other two gases, the convergence of the partial-wave expansion with the first four phase shifts cannot be questioned. The irregular small fluctuations at low energies of δ_2 and δ_3 for helium, and of δ_3 for argon, are not likely to be physical, but the result of uncertainties in the original data.

In Figs. 1–3, the experimental points of Ramsauer and Kollath are compared with differential cross sections calculated from the computed phase shifts at a number of energies for each of the three gases. (Note that for argon, the angular distribution at 12.5 eV corresponds to an energy above the inelastic scattering threshold. This curve is shown for purposes of illustration only.)

In Figs. 4–6, the computed principal phase shifts are displayed as functions of energy. We have not plotted δ_2 and δ_3 for helium, nor δ_3 for neon, since their values did not seem to be determined with sufficient reliability to justify a plot. Also shown are the experimental points of Andrick and Ehrhardt.[6] All open symbols on the three graphs correspond to published theoretical predictions, as discussed below.

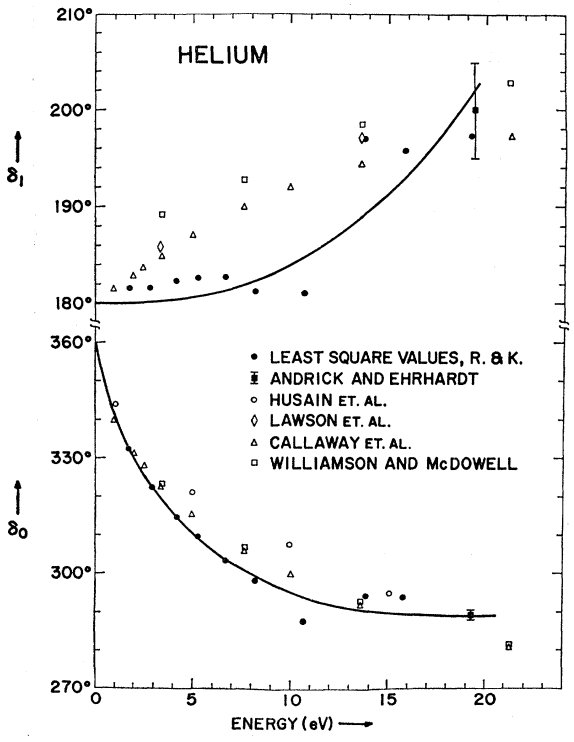

FIG. 4. Variation with energy of the first two elastic scattering phase shifts in helium. The solid circles are the least-squares values computed from the data of Ramsauer and Kollath, and listed in Table I. (The continuous curve has been drawn in order to fit these points approximately.) The solid squares and vertical bars are the phase shifts derived by Andrick and Ehrhardt (Ref. 6) from the parameters of the scattering resonance at 19.3 eV. All other symbols refer to theoretical values, as follows: (a) open circles, Husain *et al.* (Ref. 19); (b) open triangles, Callaway *et al.* (Ref. 16); (c) open squares, Williamson and McDowell (Ref. 17); (d) open diamonds, Lawson *et al.* (Ref. 18). [The *s*-wave phase shifts agree closely with (b) and (c), and have not been plotted.]

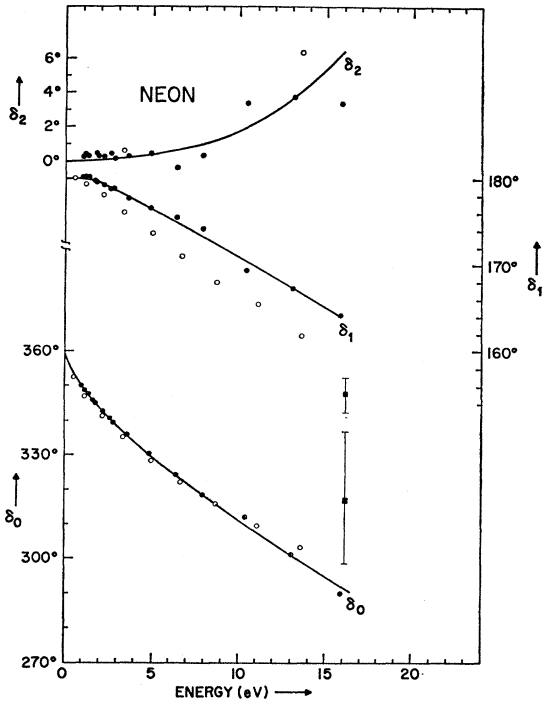

FIG. 5. Variation with energy of the first three elastic scattering phase shifts in neon. The solid circles represent the least-squares phase shifts computed from Ramsauer and Kollath's data, as listed in Table II. (The continuous curve has again been drawn in order to fit these points approximately.) The solid squares and vertical bars represent the measurements of Andrick and Ehrhardt (Ref. 6), who derived phase shifts from the parameters of the scattering resonance at 16.1 eV. Andrick and Ehrhardt also derived a value of $10°\pm2°$ for δ_2 (not shown). The open circles are Thompson's theoretical values (Ref. 25).

DISCUSSION

An extensive literature exists on the theory of the scattering of electrons by helium.[12–20] We shall confine ourselves here to a comparison of the theoretical predictions made in a selected group of the most recent papers on this subject with our least-squares analysis of the Ramsauer and Kollath data. Only those calculations will be considered in which both exchange and electric polarization effects have been taken into account.

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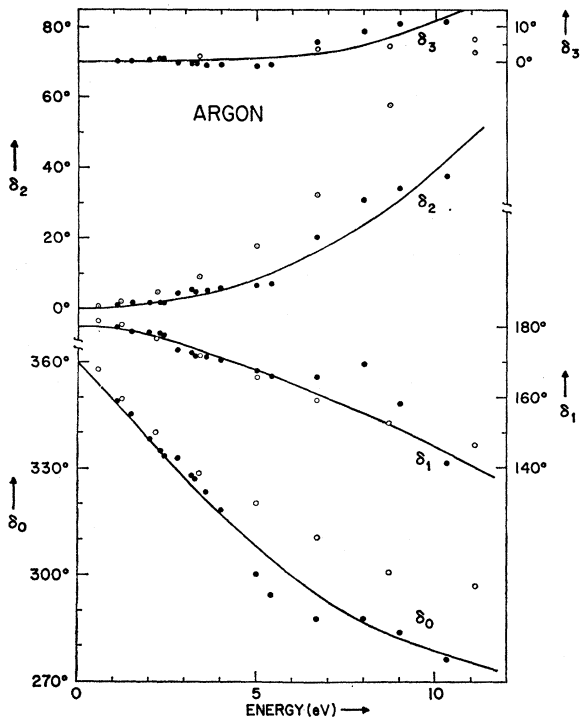

FIG. 6. Variation with energy of the first four elastic scattering phase shifts in argon. The solid circles represent the least-squares values listed in Table III, and are based on our analysis of Ramsauer and Kollath's data. (The continuous curve fits these points approximately.) The open circles are Thompson's theoretical values (Ref. 25).

Callaway *et al.*[16] have applied a form of the polarized orbital approximation to the solution of the scattering problem, using a Hartree-Fock ground-state wave function for the helium atom, and describing the distortion of the atom caused by the scattered electron by an extended polarization potential which includes dynamic effects. The phase shifts predicted by these authors are indicated by open triangles in Fig. 4.

The paper just cited is the latest of a series of articles that Callaway and La Bahn have published in this field. References to earlier publications will be found in Ref. 16.

Williamson and McDowell[17] solved a similar problem, using a ground-state wave function suggested by Shull and Löwdin,[21] and a type of polarization potential first suggested by Bethe,[22] but with a static polarizability differing from that of Callaway *et al.*[16] The resulting phase shifts are indicated by open squares in Fig. 4.

Lawson *et al.*[18] have made a careful review of these and other methods of approximation employed in the solution of the scattering equation. They then applied the exchange-adiabatic approximation, determining the polarization potential as in the method of Temkin and Lamkin,[23] and again using a Hartree-Fock approximation for the ground-state wave function. The p-wave phase shifts computed by these authors are indicated by open diamonds in Fig. 4. Their s-wave phase shifts have not been plotted because they agree closely with those calculated by Callaway *et al.*[16] and by Williamson and McDowell.[17]

Husain *et al.*[19] applied the adiabatic approximation with still another form of the ground-state wave function, using different forms of a polarization potential suggested by Reeh.[24] The s-wave phase shifts computed by these authors with the first form of potential have been plotted as open circles in Fig. 4.

The only recent theoretical treatment of electron scattering by neon and argon that we can compare with our analysis was published by Thompson.[25] Thompson's method of approximation is similar to that employed by Lawson *et al.* in the solution of the helium problem, using Hartree-Fock ground-state atomic wave functions for both gases and following Temkin's treatment of polarization.[23] Polarization of the inner electron shells is regarded as negligible and is therefore ignored.

Thompson also made an estimate, with the aid of Born approximation, of the contribution to the scattering cross section of higher partial waves neglected in the analysis, with the following conclusion. For zero scattering angle, the higher partial waves are important, but for finite angles, only the first three or four partial waves need be considered in the energy range of interest here. Comparison of the results for an exchange-only with an exchange-plus-polarization approximation shows that the inclusion of polarization is necessary to explain known experimental results.

The phase shifts computed by Thompson for neon are plotted as open circles (or open circles with dots) in Fig. 5 for comparison with the least-squares phase shifts computed here. A similar comparison for argon is plotted in Fig. 6.

It is clear that more careful experimental work with modern techniques is required to make a meaningful comparison between theory and experiment in this field.

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