all such calculations that have been carried out for this problem in the past have proved to be very unrewarding: at the most recent attempt[8] the lower bounds came out about two or three orders of magnitude farther away from the apparent convergence point than did the upper bounds. This situation may be understood qualitatively as follows. The convergence rate of the upper bound is determined by the ability of the given trial functions to represent the effects of the singularities in the Hamiltonian H. The lower-bound calculations involve the average value of H^2, and here the singularities are made more severe; thus the convergence rate will be much slower.

We have not computed any properties of the helium ground state other than the energy, since it appears that the work already published by Pekeris is suffi-

ciently accurate to cope with all practical needs of the present and near future. The work reported here was undertaken merely to investigate whether the insertion of half-powers in the Hylleraas series did achieve the vast improvement hoped for. We are presently applying this technique to a calculation of the fine structure of the lowest ^3P state in helium, since we have already found that the conventional basis does not there converge rapidly enough to allow a determination of the fine-structure constant to the new anticipated accuracy of $1/10^6$.

ACKNOWLEDGMENTS

[8] C. L. Pekeris, Phys. Rev. 126, 1470 (1962).

Measurement of Total Cross Sections for the Scattering of Low-Energy Electrons by Lithium, Sodium, and Potassium*†

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The atomic beam recoil technique has been used to measure the total cross sections for the scattering of electrons by lithium in the energy range of about 1 to 10 eV. In this method, the atom beam is crossfired by a modulated electron beam. Recoil results in a scattering out of the atom beam at the modulation frequency. A phase-sensitive lock-in amplifier is used to detect the atom-scattering signal, which is obtained here from a continuously oxygenated surface ionization detector. The angular resolution of the apparatus at 1 eV for lithium and potassium is approximately 9° and 14°, respectively, in the electron polar scattering angle.

The lithium data were normalized to potassium at each energy at which measurements were made, by the use of two simultaneously operating ovens. As a check on the method, total cross-section measurements in the same energy range were made for sodium. These are in good agreement with the results of Brode, as are some absolute determinations made on sodium, potassium, and cesium at several energies. The general shape of the lithium curve is quite similar to that of the potassium curve over the ranges studied. The lithium cross section values vary from about 50 to 70% of the potassium values in the 1- to 10-eV region. There is the appearance of a resonance at about the same energy (1.5 eV) as those observed in the other alkalis.

I. INTRODUCTION

$\mathbf{B}$Y means of a modified Ramsauer technique, Brode performed a series of experiments in which he measured the total cross sections for the scattering of low-energy electrons by all of the alkali metal vapors except lithium, as well as some other metal vapors and gases.[1] His results revealed, first, that the absolute

values of the alkali cross sections were extremely large (over 10^{-14} cm^2 in the entire low-energy range), and second, that the relative shapes of all the alkali curves were similar, particularly with regard to a strong resonance at energies of a few electron volts. Lithium was not studied because of the experimental difficulties associated with the high temperature necessary to obtain sufficient lithium vapor pressure, and the particularly serious corrosive action of hot lithium vapor.[2]

Early calculations of Allis and Morse predicted a very flat elastic cross section vs energy curve for lithium as well as quite small ($\sim 10^{-15}$ cm^2) absolute values, but they did not take either polarization or exchange into account.[3] Furthermore, the resonance

[1] R. B. Brode, Phys. Rev. 25, 636 (1925); 34, 673 (1929); 35, 504 (1930); 37, 570 (1931).
[2] R. B. Brode (private communication).
[3] W. P. Allis and P. M. Morse, Z. Physik 70, 567 (1931).

near the first excitation energy observed by Brode has been attributed[4] to the strong coupling between the ground state ($^2S_{1/2}$) and the first excited states ($^2P_{1/2,3/2}$). The combined oscillator strengths for transitions between these states is about 0.98 for all the alkalis except lithium. Since the corresponding oscillator strength for lithium is 0.75, one would expect a similar though less pronounced resonance at about 2 eV in the lithium cross-section curve. The magnitudes of the total cross section for lithium at very low electron energies should be comparable to those of the other alkalis. This is so since the ground state polarizability of lithium is about the same as that of sodium.[5] Furthermore, exchange effects should be about the same as for the other alkalis, and exchange accounts for a sizable fraction of the elastic cross section.[6,7]

Rubin, Perel, and Bederson used a recoil beam technique in 1959 to perform certain electron-potassium studies, including the obtaining of upper and lower bounds on the exchange cross section in the energy range of 0.5 to 4 eV, a study of the relative differential cross sections at several energies between 0.6 and 9 eV, and qualitative determinations of total cross sections in the 1- to 7-eV range. In this method, an atomic beam is crossfired by an electron beam, and total scattering is observed by measuring the decrease in atomic beam intensity in the forward direction, caused by momentum transfer during the scattering process. The method possesses certain advantages over the more conventional crossed-beam experiment, in which observation is made on the scattered electron beam. Since one is not concerned with the scattered electron trajectories in the recoil technique, it is not necessary to employ a highly collimated electron beam. This allows for the use of a considerably simplified electron gun design, as well as an axial magnetic field to confine the electron beam. Also, the problems associated with stray electric fields at the low energies of this experiment are diminished since the trajectory of the neutral scattered atom is unaffected by these fields. The principal limitation of the recoil method is the relatively poor detection efficiency for most neutral atomic systems. However, the alkalis, including lithium, can be detected with exceptionally high efficiency using surface ionization. Because of the relatively high ionization potential of lithium, it is necessary to raise the work function of the hot wire by continuous oxygenation of the surface. As a result of the oxygenation and also because of the high operating temperature necessary for fast response time, considerable noise is introduced when detecting lithium. It is this noise, in fact, which limited the accuracy of the present experiment.

[4] A. Salmona and M. J. Seaton, Proc. Phys. Soc. (London) **77**, 617 (1961).

[5] A. Salop, E. Pollack, and B. Bederson, Phys. Rev. **124**, 1431 (1961).

[6] K. Rubin, J. Perel, and B. Bederson, Phys. Rev. **117**, 151 (1960).

[7] H. G. Dehmelt, Phys. Rev. **109**, 381 (1958).

II. EXPERIMENTAL METHOD

An atomic beam is crossfired by an electron beam and the decrease of the atom beam intensity in the forward direction is measured. The scattering intensity is measured by two independent methods. The first (dc method) is a direct measurement using an electrometer of the difference in atomic beam intensity with and without scattering. Using this method absolute cross-section determinations can be made. In the second (ac method) the electron beam is modulated at 30 cps and the ac component of the atomic beam is detected by a phase-sensitive lock-in amplifier. This method is useful primarily in obtaining relative cross-section values because of the uncertainties involved in determining the over-all gain of the detection system. In order to obtain absolute values using the ac method, scattering signals must be normalized to a known cross section. This was accomplished by operating two ovens simultaneously, one containing lithium and the other the reference element (potassium). The oven platform was designed so that the ovens could be alternately placed in the beam position to perform rapid measurements of cross-section ratios at a given electron energy. The lithium cross section was obtained in terms of the potassium cross section by use of the expression (see Appendix)

$$Q_{\text{Li}}=\frac{\bar{v}_{\text{Li}}}{\bar{v}_{\text{K}}}\frac{k_{\text{K}}}{k_{\text{Li}}}\frac{S_{\text{Li}}/I_{\text{Li}}}{S_{\text{K}}/I_{\text{K}}}Q_{\text{K}}, \tag{1}$$

where k is the atomic beam detection efficiency and S/I is the ratio of the measured scattering signal to the total beam intensity. The quantity $\bar{v}_{\text{Li}}/\bar{v}_{\text{K}}$ is the ratio of lithium to potassium average beam speeds, given by

$$\frac{\bar{v}_{\text{Li}}}{\bar{v}_{\text{K}}}=\left(\frac{T_{\text{Li}}M_{\text{K}}}{T_{\text{K}}M_{\text{Li}}}\right)^{1/2}, \tag{2}$$

where T is the absolute oven temperature and M the atomic mass. As a check on the method, relative measurements were also made using sodium, again with potassium as the reference element.

III. APPARATUS

A description of the apparatus is contained in reference 6. Typical operating pressures were about 5×10^{-7} Torr and 8×10^{-7} Torr in the oven and main sections, respectively. Conventional alkali ovens were used.[8,9] They were fabricated out of molybdenum for lithium, and iron for the other alkalis. The lithium oven was enclosed in a tantalum radiation shield. When the oven temperatures were 850 and 520°K for lithium and potassium, respectively, both beam currents at the detector corresponded to about 10^9 particles per second.

[8] N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956).

[9] P. Kusch and V. W. Hughes, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1959), Vol. 37.
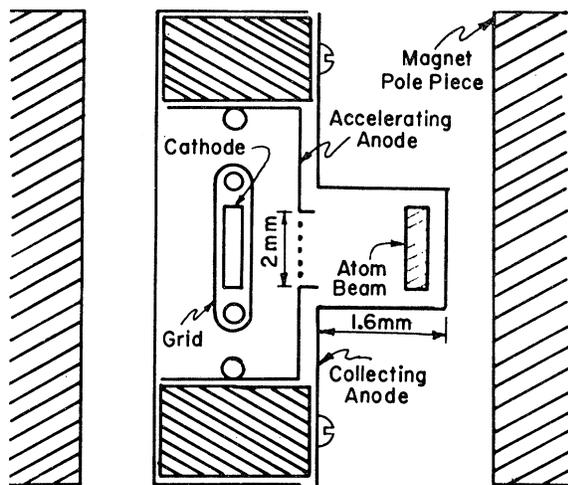
FIG. 1. Cross-sectional diagram of electron gun.

A cross section of the electron gun is shown in Fig. 1. The cathode, heater filament, grid, and mica spacer in the structure were 6L6 tube assembly parts. The accelerating anode is insulated from the collecting anode, although they were kept at the same potential during a run. A screen was welded across the opening in the accelerator anode in order to reduce the leakage of field lines into the scattering region. A magnetic field of several hundred gauss was used to confine the electron beam. Under normal operating conditions, with the grid held slightly positive in order to obtain anode currents of between 0.1 and 0.2 mA, the energy resolution was about 20% in the 3- to 12-eV range. At 1 eV, the energy resolution was 40%. The ac modulation was obtained by applying a 30-cycle square-wave negative signal to the grid.

The detection wire is the alloy 479 (92% platinum and 8% tungsten) made by Sigmund Cohn, Mt. Vernon, New York. Guard rings surrounded the wire in order to collect positive ions from the portion of the hot wire not directly exposed to the beam. The dc noise was found to decrease by a factor of more than 10 when the guard rings were employed. Furthermore, they aided in reducing the detector capacitance, which was useful in the ac detection method. A hypodermic needle was used to direct the oxygen flow onto the hot-wire surface in order to improve the detection efficiency for lithium.

A block diagram of the electronics circuits is shown in Fig. 2. The dc electrometer is a battery-operated Keithley model 600. The first stage of the ac circuit is a General Electric GL-5674 electrometer with a 10^9-Ω input impedence. After passing through a Tektronix type 122 preamplifier and Ballantine voltmeter model 300, the ac scattering signal was rectified by a narrow-band phase sensitive amplifier whose reference signal was obtained from the electron gun modulation circuit. A printing integrator was generally employed when

taking data. Under favorable operating conditions, scattering signals representing somewhat less than 10^4 atoms per second could be distinguished using this detection system.

IV. MEASUREMENTS OF ION ADSORPTION TIMES

Because of the phase sensitivity of the detection system, it is necessary to ensure that surface adsorption time does not introduce a phase shift in the detection signal. It was, in fact, observed that the scattering signals were affected by the hot-wire temperature, indicating adsorption times that were comparable to the modulation period. A study was therefore made of the adsorption time as a function of hot-wire temperature for potassium, lithium, and sodium. The rate at which ions leave the hot-wire surface is γN, where γ is the reciprocal of the mean ion lifetime on the wire's surface and N is the number of ions present on the surface. It is then easy to show that if the incident atom beam current is $S_0 \cos\omega t$, the ion beam current S' is

$$S' = \frac{S_0}{(1+\omega^2/\gamma^2)^{1/2}} \cos(\omega t - \delta),$$

where $\delta = \tan^{-1}(\omega/\gamma)$. In general, γ is dependent upon the element being detected, the nature of the hot wire and the temperature. The output of the lock-in amplifier (i.e., the signal) is then proportional to $S_0 \times \cos(\delta+\phi)$, where ϕ is the phase difference between the incident atom beam current and the reference voltage.

This behavior is shown in the curves of Fig. 3, which were used to obtain values of γ for lithium, sodium, and potassium as a function of temperature for the Pt-W 479 alloy used in these experiments. The curves are arranged in the order of increasing temperature towards the right, with potassium, sodium, and lithium in the first, second, and third rows, respectively. The last curve on the right is the lock-in response to the reference signal itself. The abscissa of each curve is the phase difference between the reference and the scattering signal using a nonlinear scale. The ordinate is the ac scattering intensity in arbitrary units. The hot-wire current is shown on each curve, but the wire temperature is given only when it was possible to make meas-
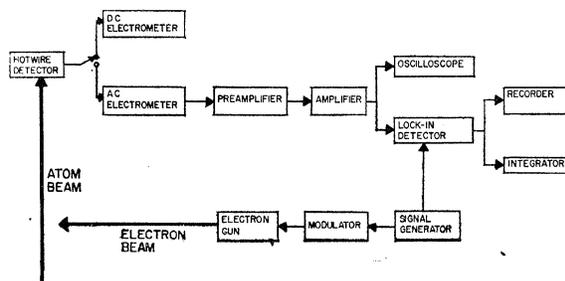

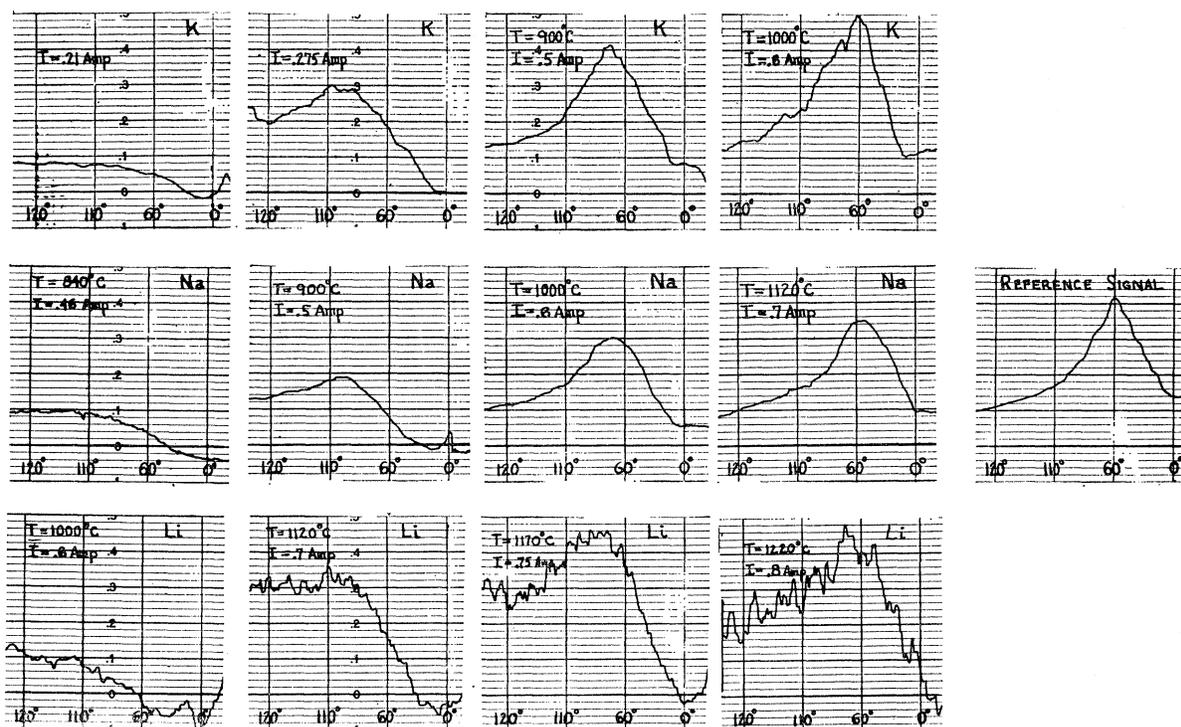FIG. 2. Block diagram of the measurement system.

FIG. 3. Response of lock-in amplifier as a function of reference phase setting for potassium, sodium, and lithium at various values of the hot-wire current I, or hot-wire temperature T. The "ideal" response, due to the reference signal itself, is also shown, and has a maximum at 60°.

urements with the optical pyrometer. The responses shown in the fourth column are similar to the ideal response obtained with the reference signal. Thus temperatures of 1000°C for potassium, 1120°C for sodium, and 1220°C for lithium were necessary to insure adequate response of the hot-wire detector. Note that the high hot-wire temperature and the oxygen spray employed in the detection of lithium results in a disproportionately high noise level for lithium. The adsorption time as a function of reciprocal hot-wire temperature for lithium and sodium is shown in Fig. 4, along with a similar curve obtained by Hughes and Levinstein[10] for rubidium on tungsten.

V. ANALYSIS OF DATA AND DISCUSSION

1. Potassium and Sodium Studies Using ac Method

Extensive studies were first made using potassium and sodium in order to compare our results with those of Brode, and to test the over-all reliability of the method.[11] In general, we obtained agreement with Brode over the energy range studied ($\sim$1 to 13 eV). It

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[10] F. L. Hughes and H. Levinstein, Phys. Rev. **113**, 1029 (1959).
[11] It appears as though no quantitative low-energy electron total cross-section measurements for the alkalis exist, other than Brode's, despite the importance of these quantities in current alkali research. We have been informed that several laboratories are at this time undertaking improved Ramsauer-type experiments to repeat and extend Brode's work.

was necessary, of course, to modify Brode's curves to take into account the larger energy spread in our electron beam. Figure 5 shows a plot of the combined data of several runs using the ac method in the energy range of 0.4 to 4.5 eV. It should be emphasized that at the low end of this range the energy spread approaches half the nominal energy. The error flags represent the standard deviations of all measurements made at the given energy. A single measurement generally consisted of the difference between the integrator print-out with electron beam off and on for a period of 1 to 2 min each. Our relative curve, normalized to unit atom and electron beam intensities, is further normalized to Brode's curve at 2 eV. Both curves exhibit the resonance peak at about 1.3 eV.

It is seen that the general shapes are in fair agreement, although there is a somewhat less rapid falloff with energy in the present work. This may possibly be attributed to the difference in the energy dependence of the angular resolving power θ_0 of the recoil and Ramsauer methods. When the beam width in a recoil experiment is somewhat larger than the detector width, as is the case here, an approximate formula for θ_0 is obtained by assuming that the minimum observable atom deflection in the detector plane is equal to the beam half-width. Transforming to the electron polar scattering angle, it is readily shown that

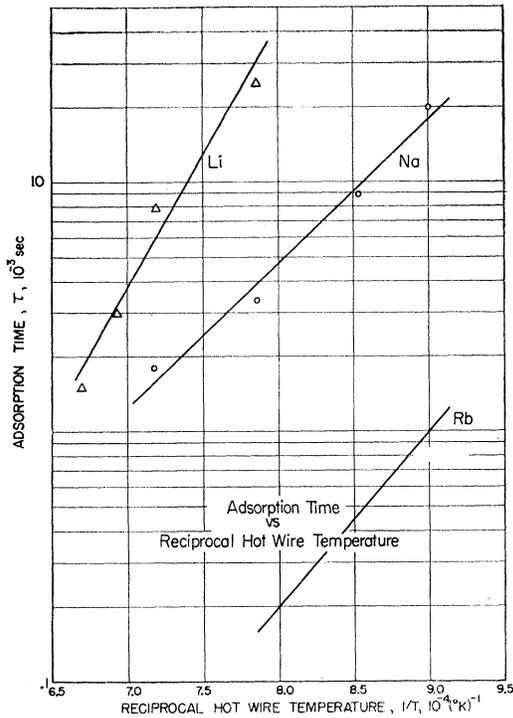$$\theta_0 \sim (32w^2MkT/\pi L^2 m^2 v^2)^{1/4}, \tag{3}$$

FIG. 4. Plot of adsorption time of Pt-W alloy 479 as a function of reciprocal temperature for lithium and sodium. A similar curve for rubidium on pure tungsten (reference 10) is shown for comparison.

where w is the beam half-width, L is the distance from the scattering region to the detector, v is the electron velocity, and m and M are the electron and atomic masses, respectively. It is here assumed that $(mv/MV)\ll1$, and that the entire atom beam possesses the mean oven speed $\bar{v}=(8kT/\pi M)^{1/2}$ (see Appendix).[12] In the present work, $w=0.020$ cm and $L=34$ cm, while the detector width was 0.010 cm. Thus, θ_0 is approximately 9° and 14° for lithium and potassium, respectively, at 1 eV. It is more difficult to estimate θ_0 for a Ramsauer-type experiment, since scattering occurs along the entire path from source to detector. Furthermore, the electron beam is considerably broader than the widths of the collimating slits placed along its path length. This causes scattering back into the detector. A crude analysis of Brode's apparatus indicates an "effective" θ_0 to be of the order of several degrees, and to be essentially independent of the electron energy.

Because of the favorable signal-to-noise ratios obtainable with potassium, it was possible to obtain complete cross-section curves directly on the recorder, by maintaining constant collecting anode current while continuously sweeping the anode voltage. One such

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[12] As a result of the beam velocity distribution, the angular resolution becomes less well defined. Equation (3) must actually be averaged over the modified (v^3) Maxwellian distribution. The resulting expression for $\theta_0(av)$ is essentially the same as Eq. (3). A small error in the total cross section then results, due to the poorer-than-average resolution of the faster beam particles.

curve, taken in a total time of 8 min, is shown in Fig. 6. At the low-anode voltage end of this curve, it was necessary to run the grid several volts positive, resulting in a poorer-than-average electron energy spread and a further diminution of the resonance peak.

Having established the reliability of the ac method for potassium, measurements were then made on sodium using potassium as a reference, and normalizing to Brode's potassium curve (including energy spread corrections) at each energy. For these measurements the two ovens containing sodium and potassium, respectively, were alternately rotated into the beam position to take data. The surface ionization detector was maintained at a temperature of about 1300°K, and was sprayed with oxygen occasionally to maintain the level of the sodium detection efficiency. The potassium-sodium velocity ratios were calculated using Eq. 2, and Q_{Na} then calculated from Eq. 1, in terms of $Q_K(B)$, the absolute potassium cross section taken from Brode at the same energy. The values of Q_{Na} so obtained are plotted in Fig. 7, along with Brode's sodium curve.

2. Lithium Total Cross Sections Relative to Potassium

The method for obtaining lithium cross sections, relative to potassium, was the same as that employed in the sodium work. For the lithium measurements, however, it was necessary to operate the surface ionizer at a somewhat higher temperature and to oxygenate it continuously. The ac noise greatly increased, relative to the noise level in the sodium and potassium experiments, resulting in a somewhat greater experimental error for the lithium results. A hot-wire temperature of about 1400°K was used for lithium detection. The adsorption time of about 8×10^{-3} sec decreased the detection efficiency to about 0.5. Below this temperature, the efficiency dropped sharply, whereas above it the hot-wire noise increased rapidly (see Fig. 4). It was thus necessary to apply phase and amplitude cor-
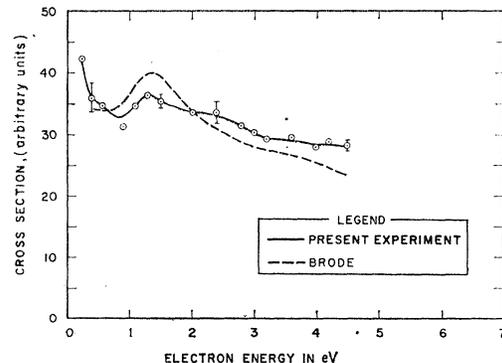

FIG. 5. Plot of total cross-section measurements for potassium, up to 4.5 eV. Comparison is made to Brode curve, which is here modified to include the same energy spread in the electron beam as present experiment. The curves are made to coincide at 2 eV.
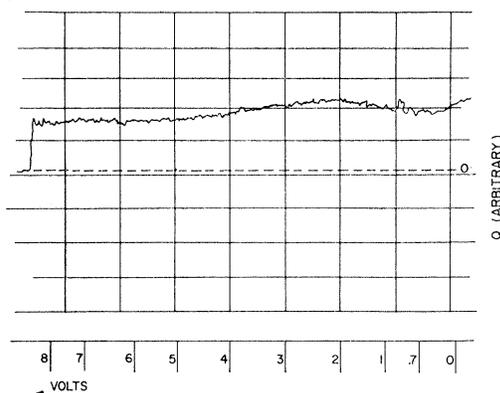
FIG. 6. A potassium total cross-section curve plotted directly on the recorder. The grid voltage was continuously adjusted so as to maintain constant anode current as the voltage was varied. Voltages as shown are uncorrected for contact potentials.
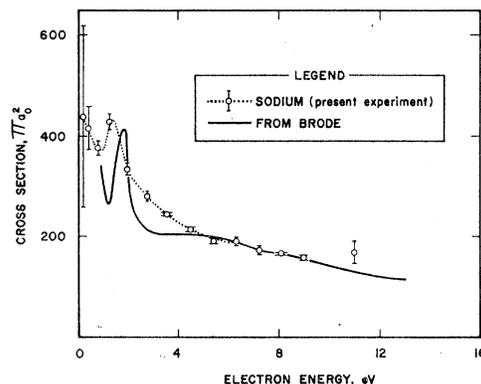

FIG. 7. Plot of total cross-section measurements for sodium, up to 11 eV. Comparison is made to unmodified Brode curve.

rections to the scattering signals. Again, the total cross section Q_{Li} is obtained using Eq. (1), with the velocity ratio $\bar{v}_{Li}/\bar{v}_K$ obtained using Eq. (2). Normal operating oven temperatures for lithium and potassium were about 830 and 490°K, respectively, corresponding to $\bar{v}_{Li}/\bar{v}_K$ of approximately 3.08.

The combined data of seven runs are presented in Table I, which lists the energies at which measurements were taken, the average calculate ratios Q_{Li}/Q_K, the absolute values $Q_K(B)$ for potassium taken from Brode's curve in units of πa_0^2, and the standard deviations in the same units. Q_{Li} is plotted, along with the standard deviations, in Fig. 8. The general shape and magnitude of the lithium cross-section curve are indeed similar to those of the other alkalis, as expected. The somewhat less pronounced peak at about 2 eV is consistent with Seaton's conclusion that all of the alkali peaks are due to strong s-p coupling, as discussed in Sec. I.

3. Absolute Cross-Section Determinations (dc Method)

Absolute determinations of atomic cross sections (i.e., without normalization to a known cross section) can often contain large systematic errors, arising from several sources. Among these are the determinations of vapor density and effective path length in beam attenuation experiments (e.g., the Ramsauer technique), and velocity-distribution determinations and precise knowledge of the beam geometries in atomic beam experiments. There is, in addition, the possibility that inadequate angular resolution results in a serious underestimation of the total cross section, where small-angle scattering plays a significant role. It was therefore of interest to compare absolute determinations made during the present work with those of Brode.

For the recoil method, the total cross section Q is given by (see Appendix)

$$Q = he\bar{v}S/Ii, \qquad (4)$$

where h is the atom-beam height in the scattering region, equal to 0.2 cm in the present work, i/e is the

TABLE I. Combined data for lithium total cross sections, normalized to potassium at each energy.

Electron energy (eV)	Number of readings	Q_{Li}/Q_K	$Q_K(B)$[a] (πa_0^2)	Q_{Li} (πa_0^2)
0.25	5	0.553	455	252±81
0.8	4	0.534	460	246±52
1.3	8	0.592	525	311±55
1.7	2	0.594	515	306±54
2.0	11	0.630	450	284±40
2.8	13	0.582	388	226±34
3.6	9	0.624	355	222±35
4.5	14	0.622	306	190±20
5.4	8	0.642	280	180±23
6.3	13	0.602	277	167±18
7.2	7	0.648	250	162±23
8.1	10	0.621	238	145±21
9.0	7	0.668	221	148±17
10.0	5	0.580	210	122±34

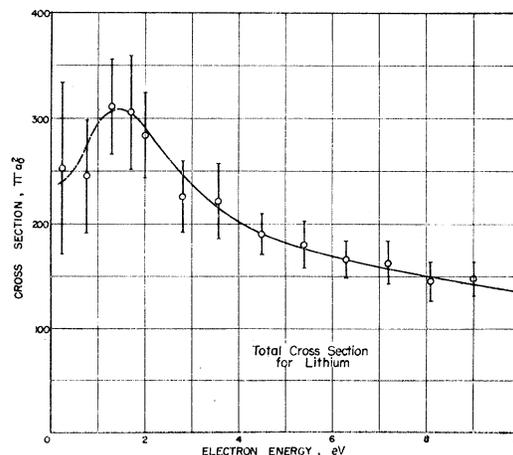[a] $Q_K(B)$ is Brode's value of the potassium cross section.

FIG. 8. Plot of total cross-section measurements for lithium.

TABLE II. Four individual determinations of the absolute total cross section at 13 eV for potassium, sodium, and cesium.

Alkali	i (10^{-3} A)	T (°K)	v (10^4 cm/sec)	S (10^{-12} A)	I (10^{-10} A)	Q (10^{-14} cm^2)	$\frac{Q}{\pi a_0^2}$	$\frac{Q(B)^a}{\pi a_0^2}$
K	1.70	510	5.24	1.40	0.94	1.47	165	182
Cs	1.70	470	2.73	6.00	1.25	2.47	277	263
K	0.85	520	5.30	1.50	1.50	1.99	223	182
Na	0.85	620	7.63	0.75	1.60	1.35	151	130

[a] $Q(B)$ is the value of the cross sections as taken from Brode's curves at 13 V.

electron current in electrons/sec, and $\bar{v}$ is the mean atomic speed in the oven.[13] I is the total dc beam intensity, and S is the scattered beam intensity, equal to the difference in dc beam intensities with and without the electron beam operating. Note that the detector efficiency need not be known, since the same detection and amplifying system is used to measure both I and S. In general, somewhat less than 1% of the atom beam was scattered.

As an illustration of the dc method, measured and calculated quantities necessary for an absolute determination are presented in Table II, for four individual measurements on potassium, sodium, and cesium at 13 eV. The quantities i, T, $\bar{v}$, S and I are listed, along with Q calculated using Eq. (4). The Brode values $Q(B)$ at 13 eV are also listed for comparison.

A total of 61 individual determinations were made for potassium at 12 eV. These yielded a combined average of 209, compared to the Brode value of 185, in units of πa_0^2. A total of 22 determinations for potassium at 10 eV yielded a combined average of 224, compared to the Brode value of 210, in units of πa_0^2. Finally, 22 determinations were made for lithium at 10 eV, yielding a combined average of 160, in units of πa_0^2. The ratio of our absolute cross sections Q_{Li}/Q_K at 10 eV is 0.71, while the same ratio for the relative determination is 0.64. The estimated probable error in the absolute measurements is 25%. In view of the large uncertainties, the agreement with Brode is seen to be quite good.

ACKNOWLEDGMENTS

[13] Assuming a modified (v^3) Maxwellian velocity distribution in the beam, the total number of beam particles in the scattering region is $N = Ia/\bar{v}$, where I is the beam current, a is the length of the scattering region, and $\bar{v} = (8kT/\pi M)^{1/2}$.

APPENDIX. RELATIVE CROSS-SECTION DETERMINATION

The scattering region geometry is shown in Fig. 9. The number of atoms S' scattered per second is given by

$$S' = QiI/hev,$$

where Q is the total cross section, (i/e) is the electron current in electrons per second, I is the total atom
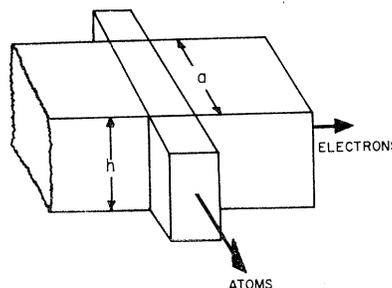

FIG. 9. Geometry of crossed beam interaction region.

beam current in atoms per second, h is the electron (and atom) beam height, and v is the atomic velocity. The scattering signal S is then

$$S = ckS',$$

where k is the neutral beam detection efficiency and c is a constant representing the over-all gain of the amplification system. Thus, the scattering cross section for lithium is given by

$$Q_{Li} = \frac{hev_{Li}\,S_{Li}}{ick_{Li}\,I_{Li}}.$$

The cross section for potassium is given by an equation of the same form, with, of course, all the measurable quantities referring to potassium. When measurements on potassium and lithium are made successively using two ovens, both i and c are maintained essentially constant and

$$Q_{Li} = \frac{v_{Li}}{v_K}\frac{k_K}{k_{Li}}\frac{S_{Li}/I_{Li}}{S_K/I_K}Q_K.$$