

Excitation of resonance levels of alkali metal atoms by electron impact

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Detailed and reliable experimental investigations are carried out on the effective cross sections for excitation of resonance levels of alkali metal atoms by electrons with energies ranging from the process threshold up to 300 eV. The effective cross sections at the excitation peak range from 4.9×10^{-15} cm² for Li to 14.2×10^{-15} cm² for Cs. For electron energies exceeding about 20 threshold units a satisfactory agreement between the first Born approximation calculations and the experimental data is observed with calculations by the strong-coupling method.

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A group of papers in the Sixties^[1-6] was devoted to investigations of the excitation of alkali atoms by low-energy electrons (up to 30–40 keV). In the experiments, the electron-atom collisions were simulated by the method of one beam.^[1] This method is relatively simple, especially if a spectroscopic procedure is used to register the excited atoms. These studies yielded considerable information on the excitation functions and on the effective cross sections of the process for a large number of S, P, D (and also F in the case of cesium) levels of all the atoms. As to the resonant levels, however, the data on the effective cross sections of their electronic excitation^[1,2,4-6] are less reliable. The reason lies, first, in the indirect method used to determine the excitation cross sections of the resonance lines,^[1,4] and second in a number of difficulties and experimental inaccuracies which were not overcome at that time.^[1-4,6]

On the other hand, a number of reviews^[7-9] and original calculations^[10-18] were published somewhat later, and there the theoretical research was focused mainly on the character of the excitation of the resonant levels of alkali elements by slow electrons. An analysis of these papers shows that at present one cannot speak of good correlation between the results obtained by various methods and by different workers. Furthermore, within the framework of one approximation, there is no agreement with experiment for different alkali atoms.

In light of the foregoing it is clear that a real need arose for organizing new and more perfect experiments aimed at a detailed investigation of the effective excitation cross sections of the resonant levels of the atoms and of their energy dependence not only in the range of some dozen threshold units, but also in a more extensive region. A realization of such a program, in particular, will afford one of the rare possibilities of comparing the results of experiments on the excitation of energy levels of atoms, on the one hand, and the Born approximation, on the other, in that range of interacting-particle velocities where the Born theory is sufficiently well founded.^[2]

The purpose of the present paper was the following: 1) to obtain as reliable data as possible on the effective excitation cross sections of the resonant levels of alkali elements in the electron energy range $E = 0 - 300$ eV, and 2) to compare the experimental results with Born-approximation calculations and to assess their agree-

ment with the available theoretical calculations at low energies.

REMARKS ON THE EXPERIMENTAL PROCEDURE

To produce the purest conditions for simulation of the excitation of alkali atoms by electron impact, we used the method where an electron beam crosses an atomic beam. The corresponding apparatus was described in detail earlier,^[20] and we therefore indicate here only the main parameters that characterize the beams.

Both beams were formed and crossed at right angles in a metallic vacuum chamber, in which the pressure of the residual gas did not exceed 5×10^{-7} Torr under the working conditions. The atom beam was produced by the oven method with a divergence angle 8° . The concentration of the atoms in the region of intersection with the electron beam ranged from 10^{10} to 10^{12} atoms/cm³. The electrons emitted from an oxide cathodes were shaped into a beam by flat diaphragms and a 127° electrostatic selector.^[21] The electron-beam intensity ranged from 0.05 to 1 μ A, and the energy scatter was 0.1–0.5 eV.

The number of excitations of atoms to a given energy state was measured spectroscopically; it was equal to the number of photons emitted per unit time. The latter makes it possible to obtain the total effective cross section for the excitation of any energy level (other than metastable levels), by using the expression^[22]

$$I_{km}(v)/h\nu_{km} = n_0 N v \left[Q_k(v) + \sum_{i=k+1}^{\infty} Q_{ik}(v) \right] A_{km} / \sum_{m=0}^{k-1} A_{km}, \quad (1)$$

(where n_0 is the concentration of the atoms in the ground state and N and v are the concentration and velocity of the electrons; ν_{km} , A_{km} and I_{km} are the frequency, spontaneous-emission probability, and intensity of the spectral line corresponding to the transition $k \rightarrow m$; Q_k and Q_{ik} are the cross sections for the excitation of the level and the spectral line of the corresponding cascade transition to this level).

In the case of the cross section for the excitation of a resonant level of an alkali atom, formula (1) simplifies to

$$Q_k(v) = Q_{k0}(v) - \sum_{i=k+1}^{\infty} Q_{ik}(v); \quad (2)$$

here $Q_{k0}(v) = I_{k0}(v)/n_0(i/e)h\nu_{k0}$ is the cross section

for the excitation of the resonant line, i is the density of the electron-beam current, and e is the electron charge ($i/e = Nv$).

The radiation produced by collision of the electrons with the atoms was observed in a narrow solid angle in a direction perpendicular to both beams. The spectral lines were separated by a large-aperture monochromator (MDR-2) and registered with photomultipliers (FÉU-106, 18A) with subsequent amplification of the output current.

The procedure for investigating the cross sections for the excitation of the resonant levels consisted of three stages. We first investigated in detail the energy dependences of the excitation cross sections (i.e., the excitation functions) of the required spectral lines. In the second stage we determined the cross sections for the excitation of the latter by measuring the absolute intensities by the known method of comparing with a calibrated radiation source. Finally, in the third stage, we determined the effective excitation cross sections of the directly resonant levels by eliminating from the absolute cross section of the resonance-line excitation the fraction introduced by the summary cascade contribution from the higher levels (in accord with Eq. (2)).

To obtain sufficiently reliable data on the cross sections of the excitation of the energy levels of the atoms it is necessary to satisfy a number of experimental requirements concerning the electron beam, the atom beam, and the detected photon flux.

Electron beam. The form of the excitation functions of the spectral lines can be affected by the following factors: the appreciable energy inhomogeneity of the electron beam, the entry of slow reflected (and secondary) electrons from the receiver into the collision zone, and also refocusing of the beam at various energies. In the present experiments we undertook primarily measures aimed at effective elimination of all these factors.

Atom beam. The absolute cross sections for the excitation of the spectral lines depends strongly on the reliability of the method used to determine the atom concentration in the collision zone. In each individual experiment, the atom concentration was determined by us simultaneously by two different methods: radiotechnical^[20] or from the total ion current (using data on the ionization cross sections of the investigated atoms^[20, 23]). The results obtained independently by the two methods agreed each time within 8%.

However, in either case the accuracy with which the atom concentration is measured depends in turn on correct allowance for the geometry of the intersection of the electron and atom beams, and also on the degree of condensation of the atoms by the cold surface of the neutral-particle detector. In our experiments, the geometric factor was taken into account by the method described in^[20], and the coefficient of condensation on the surface of a detector kept at liquid-nitrogen temperature reached unity for practically all the alkali atoms.^[24]

For even greater assurance of the reliability of the results, the absolute excitation cross sections of a number of lines of the subordinate series (which did not experience absorption by their own atoms) were measured in additional experiments by the "cell"

method,^[25] (i.e., a greatly improved variant of the one-beam method). The atom concentration was determined in this case by the surface-ionization method.^[26] By way of example, Table I lists the results for certain lines obtained both in a "cell" and in intersecting beams at $F = 15$ eV. We see that the cross sections of these lines measured by three different methods agree within 10%. This good agreement assures us of the correctness of our procedure of determining the atom concentration and the geometric factor.

Photon flux. In the course of the measurements of the absolute intensities of the spectral lines (meaning also their excitation cross sections), substantial errors may result from failure to allow for such factors as the polarization of the radiation of the electron-excited atoms, and reabsorption of the resonance lines.

As shown by experiments specially performed by us with potassium, rubidium, and cesium, and also by the data of^[27] on lithium and sodium, the degree of polarization of the resonant radiation is negligible ($\lesssim 10\%$). Therefore the error introduced by this effect in the determination of the absolute excitation cross sections of the resonance lines will not exceed 3%^[17] and can be neglected in these experiments. As to reabsorption of the resonant radiation, it was previously assumed^[2, 6] that it is negligible in the method of intersecting beams (in view of the small thickness of the radiating layer). A detailed investigation of this phenomenon in the present study has shown, however, that even at atomic-beam concentrations 10^{11} atoms/cm³ the reabsorption is still appreciable and reaches 50% at the chosen geometry of the beam-intersection zone.

For all the resonance lines, we performed careful measurements of the total absorption by the "one-mirror" method^[28] and introduced the corresponding corrections into the absolute cross sections for the excitation of these lines. In addition, we succeeded in measuring the cross sections of the resonant potassium and rubidium lines which are most convenient from this point of view (more accurately, of their strong components) at so low a concentration (10^9 atoms/cm³) that the reabsorption was indeed negligibly small. In both cases we obtained comparable (within 6–8%) values of the absolute excitation cross sections.

RESULTS AND DISCUSSION

The net results of our investigations were detailed and repeated studies of the excitation function of the resonance lines and their absolute cross sections for all alkali atoms in the energy range from the thres-

TABLE I. Cross sections for the excitation of the lines of the subordinate series of potassium and cesium (in units of 10^{-18} cm²), measured by various methods at $E = 15$ eV.

Element	Spectral transitions	λ , nm	Method of transporting the atoms and method* of determining their concentration in the excitation zone		
			Intersecting beams		Cell
			I	II	III
K	$4^2P_{1/2} - 8^2S_{1/2}$	532.3	4.8	4.35	4.81
	$4^2P_{3/2} - 6^2D_{3/2}$	536.0	17.6	16.8	18.0
Cs	$6^2P_{1/2} - 11^2S_{1/2}$	574.6	7.2	6.6	7.0
	$6^2P_{3/2} - 9^2D_{3/2}$	584.5	14.7	13.6	13.4

*I—radiotechnical, II—from the total ion current, III—from surface ionization.

hold of the process to 300 eV. In addition we measured again, in the same energy interval, the excitation cross sections of a large number of lines (in the spectral region 240–900 nm) needed for a reliable estimate of the contribution of the cascade population of the resonance levels in electron-atom collisions. The error in the relative measurements of the excitation functions is 2%, and the rms error in the absolute cross sections of the resonance lines is 8%.

The excitation functions of the resonance lines are shown in Fig. 1, while Table II lists the corresponding spectral transitions and the excitation-maximum threshold energies, obtained both in our experiment and earlier.

As seen from the figure, a weak structure is observed on the ascending parts of the curves up to the maximum. Although this structure is comparable with the experimental error in the case of lithium, sodium, and potassium, it had appeared repeatedly from measurement to measurement. We note that in the region of low energies a reasonable interpretation of

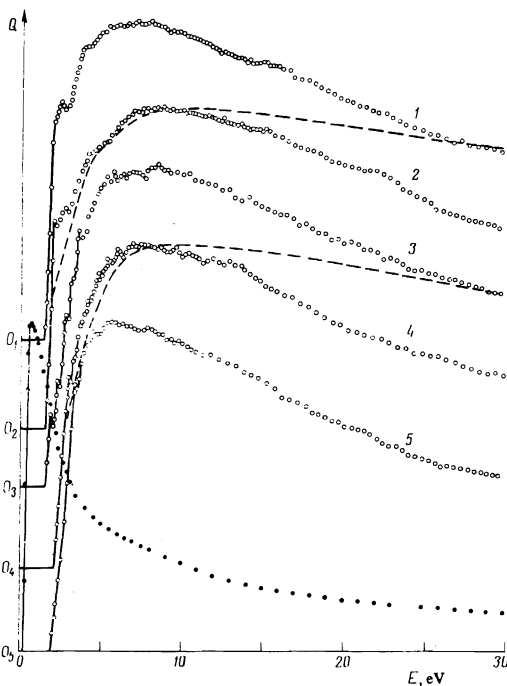


FIG. 1. Excitation functions of resonance lines: 1— λ 8521 Å Cs, 2— λ 7800 Å Rb, 3— λ 7665 Å K, 4— λ 5890/96 Å Na, 5— λ 6708 Å Li (dark circles—curve 5 in 0–300 eV scale). The dashed curves are the excitation functions of the sodium and rubidium resonance lines from [4], normalized to our experiment at the maximum.

TABLE II. Maximum cross sections for the excitation of resonance lines (in units of 10^{-15} cm²)

Element	λ , nm	Transition	E_{thr} , eV	E_{max} , eV	Our experiment	Earlier results
Li	670.8	$2^2S_{1/2} - 2^2P_{1/2,3/2}$	1.85	6	5.10	4.5[3]; 2.48[9]
Na	589.0/9.5	$3^2S_{1/2} - 3^2P_{1/2,3/2}$	2.10	7	5.90	2.95 [29]; 2.18 [4]
K	769.9	$4^2S_{1/2} - 4^2P_{1/2}$	1.61	8	3.00	4.3 [1]; 2.3 [4]
	766.5	$4^2S_{1/2} - 4^2P_{3/2}$	1.62	8	6.10	8.5 [1]; 4.5 [4]
Rb	794.7	$5^2S_{1/2} - 5^2P_{1/2}$	1.56	8	3.03	4.56 [4]
	780.0	$5^2S_{1/2} - 5^2P_{3/2}$	1.59	8	5.85	3.30 [4]
Cs	894.3	$6^2S_{1/2} - 6^2P_{1/2}$	1.39	8	4.96	3.27 [4]
	852.1	$6^2S_{1/2} - 6^2P_{3/2}$	1.45	8	9.50	6.80 [4]

this structure may be connected with the formation of autoionization states of the negative ion.^[30] The noticeable deviations of the excitation functions past the maximum from those previously measured are obviously due to the distorting action of the secondary electrons, the appearance of which in the collision region was not effectively predicted in the earlier studies.^[1–6]

It is seen from Table II that the resonance-line excitation cross sections measured in our experiments are as a rule larger by 1.5–2 times than the previously obtained ones. This is obviously due to the fact that reabsorption was not taken into account in the earlier work, nor was the appearance of secondary electrons. In addition, in much earlier investigations by the volume method^[1,3,4] the determination of the atom concentration from the temperature of a stub with metal was also far from faultless.

The cascade contribution to the population of the resonance levels of the investigated atoms is due to transitions from the nS and nD levels of the subordinate series. For both series of each atom, we measured anew the excitation cross sections of the spectral transitions from 5–8 lower levels; this enabled us to estimate quite fully (taking into account the rapid fall-off of the excitation cross sections with increasing principal quantum number^[5,31]) the role of the cascade transitions in accordance with formula (2). The fraction of the cascade contribution turned out to be relatively small, from 6% for cesium to 12% for sodium. The effective cross sections for the excitation of the resonance levels were determined by graphically subtracting the summary cascade contribution from the absolute excitation functions of the resonance lines. The results on the level excitation cross sections are shown in Figs. 2–6 and in Table III.

Let us analyze now the resultant data on the effective resonance-level excitation cross sections in light of the available theoretical models and concrete calculations. We consider first the behavior of the curves at high energies, i.e., in the so-called Born region. Analysis shows that starting with $20E_{thr}$ the character

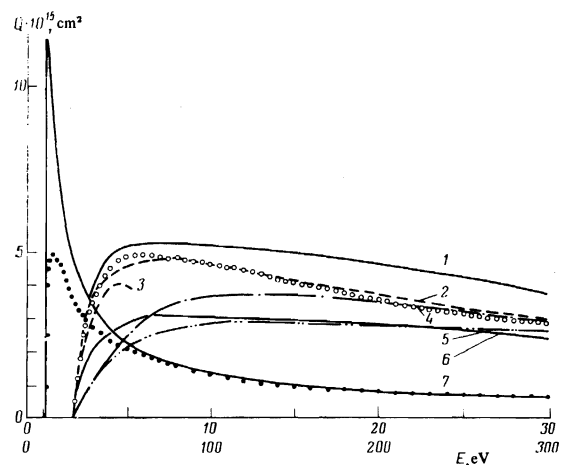


FIG. 2. Effective cross sections for the excitation of the resonance level of lithium. Points—our experiment: ●—scale 0–300 eV, ○—scale 0–30 eV; curves—calculation: 1—by the Seaton method [35], 2—by the strong-coupling method [11], 3—by the strong-coupling method, [34] 4—in the Glauber approximation, [15] 5—in the correlation approximation, [16] 6—using the Vainshtein model, [33] and 7—in the Born approximation.

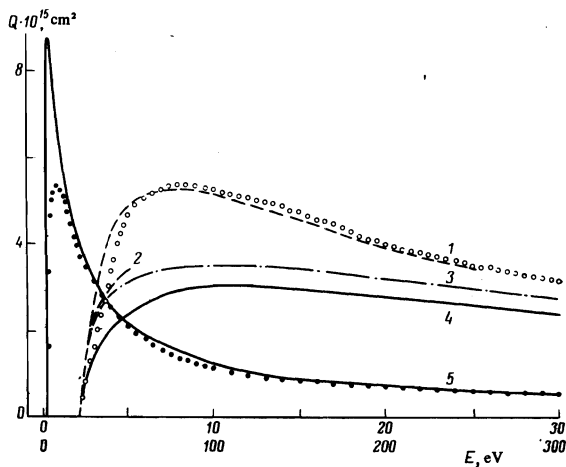


FIG. 3. Effective cross sections for the excitation of the resonance level of sodium. Points—experiment: ●—scale 0–300 eV, ○—scale 0–30 eV; the curves were calculated: 1—by the strong-coupling method, [10], 2—by the strong-coupling method, [34] 3—in the Glauber approximation, [15] 4—by the Vainshtein model, [33] and 5—in the Born approximation.

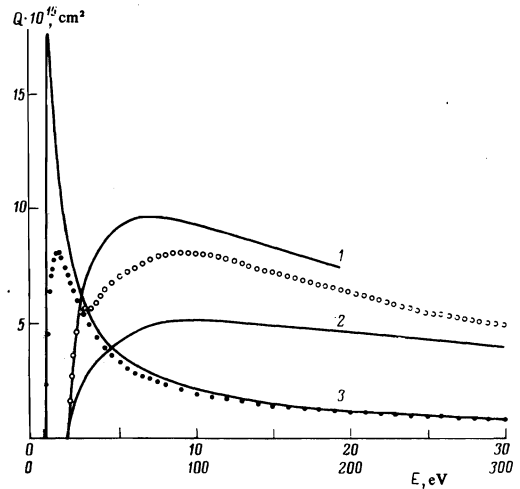


FIG. 5. Effective cross sections for the excitation of the resonance level of rubidium. Points—present experiment: ●—scale 0–300 eV, ○—scale 0–30 eV. Curves—calculation: 1—in the Born approximation with allowance for strong coupling, [18], 2—by the Vainshtein model, [33] 3—in the Born approximation.

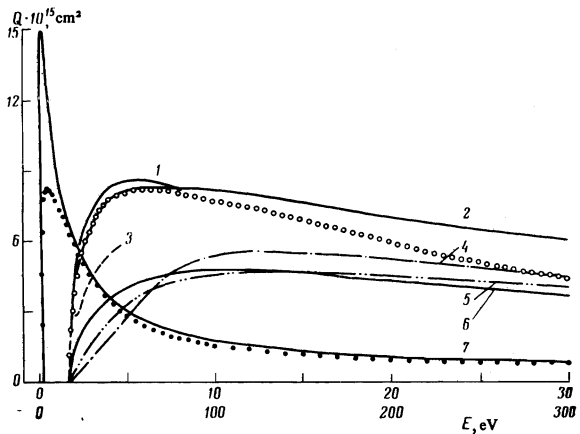


FIG. 4. Effective cross sections for the excitation of the resonance level of potassium. Points—experimental data: ●—scale 0–300 eV, ○—scale 0–30 eV; curves—calculation: 1—by the Born method with allowance for strong coupling, [18] 2—by the Seaton method, [35] 3—by the strong-coupling method, [34], 4—by the Glauber method, [15] 5—in the correlation approximation, [16] 6—by the Vainshtein method, [33] 7—in the Born approximation.

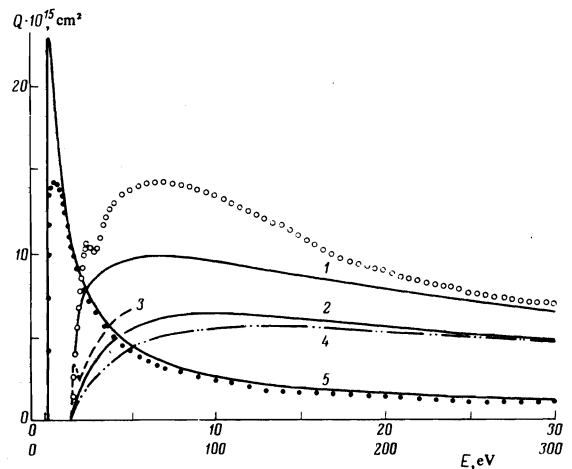


FIG. 6. Effective cross sections for the excitation of the resonance level of cesium. Points—our experiment: ●—scale 0–300 eV, ○—scale 0–30 eV; curves—calculation: 1—by Seaton's method, [35] 2—by the Vainshtein model, [33], 3—by the strong-coupling method, [34] 4—in the correlation approximation, [16] 5—in the Born approximation.

of the fall-off of the excitation functions of the resonance levels of all alkali atoms is fundamentally the same. Since we do not know at present of any concrete theoretical calculations in this energy region for alkali atoms, we have proceeded in the following manner.

It is known^[32] that in the Born approximation the excitation cross sections are well approximated, in the case of optically allowed transition, by the two-parameter formula

$$\sigma = \pi a_0^2 \left(\frac{Ry}{\Delta E} \right)^2 \left(\frac{E_1}{E_0} \right)^{1/2} \frac{Q_{\text{min}}}{2l_0 + 1} \Phi(u), \quad (3)$$

where

$$\Phi(u) = C \left(\frac{u}{u+1} \right)^{1/2} \ln(16+u), \quad (4)$$

u is the energy in threshold units; Ry is the Rydberg energy unit; E_0 and E_1 are the initial and final energy levels; ΔE is the threshold excitation energy; C and φ

TABLE III. Effective cross sections at the maximum of the alkali-metal resonance-level excitation

Element	Level	Our data	$Q \cdot 10^{15}, \text{cm}^2$				
			Theoretical calculations**				
			I	II	III	IV	V
Li	2^2P	4.90	4.82 ^[11] 4.29 ^[12] 4.00 ^[34]	3.10	5.3 ^[35] 11.5 ^[17] 5.7 ^[16]	4.2 ^[15] 4.52 ^[14]	2.92
Na	3^2P	5.40 (4.62*)	5.8 ^[10] 3.6 ^[13] 3.35* ^[34]	3.00	5.3 ^[35] 5.07 ^[18]	3.52 ^[15] 3.5 ^[14]	2.80
K	4^2P	8.20 (7.95*)	5.82* ^[34]	4.82	8.28 ^[35] 11.5 ^[17] 8.54 ^[18]	6.35 ^[15]	4.72
Rb	5^2P	8.15	—	5.22	9.7 ^[35]	—	4.30
Cs	6^2P	14.2 (13.8*)	6.5* ^[34]	6.34	11.2 ^[35] 10.4 ^[36]	—	5.60

*Cross sections at $E = 5$ eV.

**I—strong coupling method, II—Vainshtein model, [33] III—modified Born approximation, IV—Glauber method, V—correlation approximation. [16]

are tabulated constants; l_0 is the orbital quantum number of the optical electron of the initial state; $Q_{\kappa \min}$ is a factor that depends on the angular-momentum quantum numbers ($\kappa = |l_0 - l_1|, \dots, l_0 + l_1$). Using this formula, we calculated the resonance-level excitation functions up to 300 eV, and then normalized them at $E = 30E_{\text{thr}}$ to the corresponding cross sections calculated in the Born approximation.^[33, 3] As seen from Figs. 2–6, the agreement between the experimental and theoretical data is quite satisfactory in a wide energy interval. The best agreement is observed for lithium; in the interval from $20E_{\text{thr}}$ to 300 eV, the difference between theory and experiment is $\sim 1\%$. With increasing serial number Z of the atom, this difference increases somewhat and reaches 4% for cesium, but still remains within the limits of the experimental error.

As to the low-energy region (several threshold units), the picture here is much more complicated. An analysis of a large number of calculations by various authors in different approximations and assumptions shows that none describes quite fully the character of the resonance-level excitation of alkali atoms in this energy region. It is therefore reasonable to distinguish here between the near-threshold region and the region of the maximum, and to carry out the analysis of the various theoretical methods by starting (a) from the convergence of the calculations performed by various methods and different authors, and (b) from the effect of various assumptions made within the framework of one method.

As seen from Fig. 2, the best agreement for Li in the region from E_{thr} to 30 eV was obtained in the calculations of Burke and Taylor,^[11] performed by the strong-coupling method with allowance for exchange. As to the calculations performed by others and by different methods, almost all undervalue the results in the maximum and in the threshold regions. The energy dependence of the cross sections in the near-threshold region was investigated by the strong-coupling method by Karule and Peterkop.^[34] A comparison of our data with these calculations shows that they yield cross sections smaller than the experimental ones, and furthermore this difference increases with increasing Z . It follows therefore (see also Table III and Figs. 2–4 and 6) that calculations performed by the strong-coupling method but by different workers^[34, 10, 11, 13] differ from one another by up to 20%, which is frequently more than their deviations from experiment. As to the sodium atom (Fig. 3), the picture here is similar to that for lithium: experiment agrees with calculations^[10] performed by the strong-coupling method.

Using the potassium atom as an example, let us see which of the simplest calculation methods (modifications of the Born method) give the best agreement with experiment. As seen from Fig. 4, at the excitation maximum the experiment agrees with the calculations of Damburg and Kravchenko.^[35] With increasing energy, however, the agreement becomes much worse (at 30 eV the theoretical curve lies 20% above the experimental one). We note that a similar picture is typical also of other atoms. With respect to the latest theoretical calculations performed in the Born approximation with allowance for close coupling, it can be stated that the best agreement with experiment is provided by the calculations of McCavert and Rudge.^[18] The greatest disparity between experiment and the available theoretical calculations takes place for cesium and rubidium

atoms. Unfortunately, the theoretical calculations for these atoms are much scantier than for the first three alkali elements.

It follows thus from the experiments and from the analysis that the new experimental data agree best with calculations in the Born approximation at $E \gtrsim 20E_{\text{thr}}$ and with calculations in the strong-coupling approximation at low energies.

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¹⁾With the exception of experiments on the excitation of the lithium atom [^{2,6}], in which the method of intersecting beams was used.

²⁾In this respect, we know of only one experimental study [¹⁹] in which it is demonstrated that the results on the excitation of the 2P level of He coincide with calculations by the Born method within 1% at an electron energy $50E_{\text{thr}}$ (threshold units).

³⁾The calculations in [³³] were performed only up to $30E_{\text{thr}}$.

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