ENERGY SPECTRA OF ELECTRONS RELEASED IN COLLISIONS BETWEEN FAST ALKALI METAL ATOMS AND He AND Ne ATOMS

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Submitted June 3, 1969

Zh. Eksp. Teor. Fiz. 57, 1908–1916 (December, 1969)

The energy spectra of electrons ejected in collisions between fast Na, K, Rb, and Cs atoms with He and Ne atoms are studied. The energies T of the Na atoms varied between 200 and 3000 eV, those of the Rb atoms between 500 and 3000 eV, of the K atoms between 260 and 3000 eV, and the Cs atoms between 1500 and 2950 eV. The measurements were performed by the retarding potential technique and electrons moving perpendicular to the primary beam of neutral atoms were recorded. It is shown that along with the group of "slow" electrons whose behavior in general can be predicted by the Demkov-Komarov theory, faster electrons are also produced. For certain combinations of the partners intense discrete electron groups were observed. Their appearance is interpreted as being the result of excitation of auto-ionized states of the alkali metal atom.

INTRODUCTION

THE energy spectra of electrons appearing upon ionization as a result of collision of two neutral atoms, have been investigated very little. Prior to our earlier paper[1], this question was investigated experimentally only by Berry[2], who studied the energy spectra of electrons emitted upon ionization as a result of collision between two atoms of inert gases (for the pair He-He at incoming-particle energies T from 510 to 2500 eV, and at energies from 300 to 3,000 eV for the pair Ar-Ar).

Demkov and Komarov[3] calculated the energy spectrum of the electrons emitted upon ionization of the neutral atom with a low ionization potential, as a result of its slow collision with a neutral atom having a high ionization potential. (A case of this kind occurs upon collision between an alkali-metal atom and an inert-gas atom.) For the integral of the distribution function $\rho(\epsilon)$ of the electrons with respect to the energy

$$\omega(E) = \int_E^\infty \rho(\varepsilon)\, d\varepsilon$$

they obtained the following formula:

$$\omega(E) = \omega_0 \exp\left[-\frac{2L}{\hbar v}(E_0 + E)\right], \qquad (1)$$

where ω_0 is the probability that a system of two colliding atoms reaches the term E_0 when the atoms come close together, v is the rate of approach of the nuclei at the point of the pseudointersection of the terms (at an internuclear distance $R = R_0$), and L is a constant that cannot be determined within the framework of the Demkov-Komarov theory. The energy spectrum of the electrons emitted upon collision of two neutral atoms was considered also by Smirnov[4], who obtained a formula similar to (1).

In our paper[1] we presented preliminary results of the investigation of the energy spectrum of the electrons for the pairs K-He and K-Ne at K-atom energies from T ~ 500 eV to T ~ 2,350 eV. It was established there that for the K-He pair, besides a group of relatively slow electrons (with energies up to 5 eV), the behavior of which in general corresponds to the predictions of the theory of Demkov and Komarov, when the kinetic energy T of the fast atom increases there appears also a group of relatively fast electrons with energies E close to 15 eV. The appearance of this group was ascribed to excitation of auto-ionization space of the K atom during the collision process.

In the present investigation our purpose was to study systematically the energy spectra of the electrons resulting from ionization of alkali-metal atoms colliding with inert-gas atoms. The alkali-metal atoms are of interest as objects forming a homogeneous group with identical structure of the outer part of the electron shell, and having low ionization potentials. The partners in the collisions were chosen to be He and Ne, the ionization potentials of which are much higher than the ionization potentials of the atoms of the alkali metals. One could expect at sufficiently low energies T the ionization process of the atoms of the alkali metal to predominate. We had in mind to obtain information concerning the general character of the energy spectra of the electrons, to ascertain the possibility of appearance of intense discrete groups of fast electrons similar to those observed by us in[1] for the K-He, and to establish their contribution to the total number of emitted electrons in those cases when they are excited. We chose for the investigation the retarded-potential method, since it makes it possible to obtain these data directly.

EXPERIMENTAL SETUP AND MEASUREMENT METHOD

An overall view of the experimental setup is shown in Fig. 1. The method of obtaining beams of neutral atoms of alkali metals in the ground state was described in detail earlier[5]. A beam of positive ions of the investigated alkali metal, obtained from a thermo-

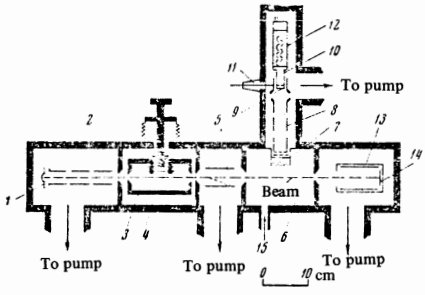

FIG. 1

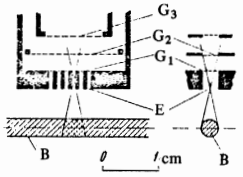

FIG. 2

FIG. 1. Diagram of experimental setup: 1 – thermo-ionic source, 2 – focusing system, 3 – charge-exchange chamber, 4 – cell with alkali metal, 5 – deflecting capacitor, 6 – collision chamber, 7 – system of electrodes for energy analysis of the electrons, 8 – system for the acceleration and focusing of the electrons, 9 – scintillator, 10 – light pipe, 11 – high-voltage lead, 12 – photomultiplier, 13 – receiver for beam of fast neutral atoms, 14 – target, 15 – tube for admission of gas.

FIG. 2. Magnified picture of system of electrodes for the energy analysis of the electrons emitted at an angle 90° to the direction of the fast-atom beam: B – beam; E – electrode with rectangular apertures 0.6 × 3 × 3 mm, $G_1 - G_3$ – grids.

ionic source[1)] was accelerated, focused, and entered the chamber 3, where charge exchange of these ions was effected in vapor of the same alkali metal. The beam of the produced neutral atoms was rid of the positive ions that did not experience charge exchange in a transverse electric field, which was produced between plates of capacitor 5. The fast atoms of the alkali metal entered the collision chamber 6 filled with the investigated gas. To monitor the intensity of the stream of fast neutral atoms, we measured the current of the secondary electrons produced when the atoms struck the target 14.

The electrode system 7 was intended for the analysis of the energies of the electrons emitted perpendicular to the direction of the beam of neutral atoms. In choosing the geometrical dimensions of the electrodes, their distance from the beam axis, and the other factors determining the geometrical resolving power of the instrument, it was necessary to satisfy a number of mutually contradictory requirements. On the one hand, it was necessary to have the resolving power sufficiently high, so as to ensure a distinct separation of the expected discrete group of electrons. On the other hand, in the registration of low-energy electrons (on the order of 1 eV and less), it was necessary to take into account the influence of the electron velocity component along the beam of the neutral atoms. This component is equal to the velocity of the fast atoms and increases with increasing energy T of the latter; the efficiency of registration of the electrons with these energies is hence lower. We have therefore employed an experimental geometry designed to ensure a resolution 1 : 15 and an effective registration of electrons with energies exceeding 1 eV.

[1)]The absence of a magnetic mass analyzer, due to the use of a thermoionic source, has made it possible to avoid the difficulties of screening or compensation of the stray magnetic field. It was ascertained in [1] that the earth's magnetic field does not influence the experimental results, within the limits of experimental errors of our measurements.

Figure 2 shows, in a magnified scale, the electrode system 7 and indicates the factors determining the geometrical resolution. (To decrease the influence of the contact potential differences, all the electrodes were made of one material—red copper). The rectangular apertures in the electrode E were used to separate electrons emitted perpendicular to the direction of the neutral-atom beam. An analysis of the energy of the electrons passing through these apertures was carried out in a homogeneous retarding electric field between the grids G_1 and G_2. (We used copper electrolytic grids with a transparency greater than 90% and a spacing of eight lines/mm.) The electrons passing through the retarding potential difference were accelerated to an energy 50 eV, and then to 11 keV, and were subsequently registered by a scintillation electron detector. (The construction of this detector is described by us in greater detail in[6].) By measuring the dependence of the electron current registered by the detector on the potential of the grid G_2 we could obtain a delay curve, which characterizes the number of electrons with energy larger than a specified value (the same quantity is given also by formula (1)). We plotted automatically the delay curves at a time-constant flux of fast atoms, varying the grid potential linearly in time. (A check has shown that the deviation of the sweeping voltage from linearity did not exceed 5%). The sweep period was 90 sec. Under these conditions, the intensity of the flux of fast neutral atoms and the pressure of the gas in the collision chamber turned out to be practically constant.

By way of an example, Fig. 3 shows the automatically plotted delay curve for the Cs-Ne pair. The small wiggles on the curve are connected with the statistical scatter of the number of electrons registered by the counting-rate meter at an integrating-network time constant of 0.1 sec. In the data reduction, a smooth curve was drawn through the curves of this type.

The instrument was evacuated to a pressure of 2×10^{-6} mm Hg. Measurements of the delay curves were carried out at pressures p of the investigated gas not exceeding 5×10^{-4} mm Hg. The linearity of the investigated effect in p, observed in the working region of

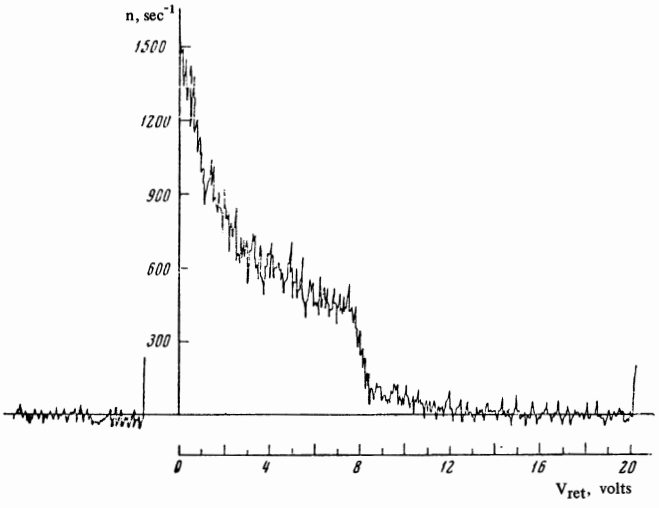

FIG. 3. Automatically plotted delay curve for the Cs-Ne pair (T = 3,000 eV); n – number of pulses (per second) registered by the electron detector, V_{ret} – retarding potential.

p, indicated that single collisions were produced. The extremely unfavorable ratio of the cross sections for the alkali atom—inert-gas atom pair and alkali atom—residual-gas molecule pair made it necessary occasionally to take into account the background effect. We therefore measured the delay curve in the residual gas (at the same intensity of the neutral-atom beam) and subtracted it from the delay curve corresponding to the investigated inert gas. Even in the most unfavorable cases, however, the fraction of the background did not exceed 25% of the investigated effect. The figures of the next section show the delay curves with the background subtracted.

As indicated above, it was our intent to investigate the energy spectra of the electrons resulting from ionization of fast atoms of alkali metals. Since the partners in the collision were chosen to be inert-gas atoms, we expected at sufficiently low values of T, that the process of interest to us will predominate. For an experimental verification of this circumstance, a special cylindrical capacitor (see[7,8]) the same as used in[1] was placed in the collision chamber in place of the electrode system 7 and the electron detector. By connecting an electrometric amplifier to the collector of this capacitor, we could measure, as a function of the retarding potential, both the electron current produced as a result of the collision and the current of the positive ions which would appear if the inert-gas atoms were also to be ionized. Such a verification was carried out for the pairs K-He, K-Ne, and Na-He. It turned out that in these cases the positive current that could be attributed to the current of positive ions resulting upon ionization of the gas never exceeded several per cent of the total number of emitted electrons.

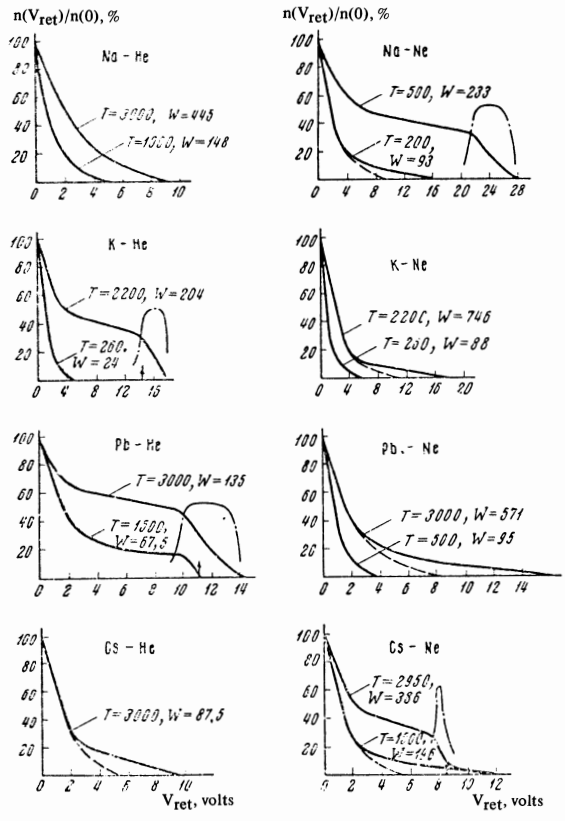

FIG. 4. Integral energy spectra of electrons (delay curves) in collisions of fast Na, K, Rb, and Cs atoms with He and Ne atoms. The dash-dot curves show the result of differentiation of the delay curve in the region of the steep descent. The values of the energies T and W are given in electron volts.

MEASUREMENT RESULTS AND DISCUSSION

The measurements were carried out at energies T from 1500 to 2950 eV for Cs atoms, from ~500 to ~3000 eV for Rb atoms, from 260 to 2200 eV for K atoms, and from ~200 to 3000 eV for Na atoms. The lower limit of the energy interval was determined by the magnitude of the effective cross section of the ionization process, by the possibility of obtaining a sufficiently intense beam of neutral atoms, and by the sensitivity of our measuring system. The upper limit was determined by the fact that we attempted to confine ourselves to cases in which the ionization process of the alkali-metal atoms was predominant.

The main results of the measurements are shown in Fig. 4, which contains the delay curves of the electrons emitted upon collision between fast alkali-metal atoms (Cs, Rb, K, Na) with He and Ne atoms (the figures show for clarity only some of the obtained delay curves; the curves are also marked with the kinetic energy of the relative of the colliding particles $W = mT/(m + M)$). As seen from these figures, the delay curves have different forms for different atom-atom pairs, but a number of common features can also be observed.

Let us stop first to discuss those cases when the delay curve of the electrons decreases smoothly towards the abscissa axis. As stated in the introduction, on the basis of the theory of Demkov and Komarov we could expect the delay curves of the electrons produced upon ionization as a result of slow collision of two neutral atoms to decrease monotonically with increasing delaying potential, in accordance with formula (1). The delay curves pertaining to the lowest energies T for the pairs Na-He, K-He, K-Ne, and Rb-Ne (Fig. 4) correspond, within the limits of the experimental accuracy, to the $\omega(E)$ dependence given by formula (1).

For the pairs Na-Ne (P = 200 eV), Cs-He (T = 3000 eV), and Cs-Ne (T = 1500 eV), and also for K-Ne and Rb-Ne at increased energy T, the delay curves also decrease smoothly towards the abscissa axis, but starting with certain values $E = eV_{ret}$ the number of fast electrons decreases with increasing E more slowly than in accordance with formula (1). To illustrate this circumstance, the dashed line on the corresponding figures shows the curves obtained by extrapolating the initial part of the delay curve in accordance with formula (1).

With increasing energy T, the delay curves for the pairs Na-Ne, K-He, Rb-He, and Cs-Ne, at certain values of V_{ret}, reveal a noticeable kink. For example, for the pair Cs-Ne (T = 2950 eV) one can see clearly the region of a sharp decrease of the delay curve towards the abscissa axis in the region $V_{ret} = 8$ V, thus evidencing the presence of a discrete group of electrons in this region. In the case of the Cs-He pair at T = 4000 eV (the corresponding curve is not shown in the figure) there is also a noticeable indication of the existence of a discrete group of electrons with E = 8 eV. Discrete groups of electrons appear also in the case of the pairs Na-Ne, and Rb-He. As an illustration, the dash-dot curves in

Table I. Experimentally observed thresholds* for the appearance of discrete groups of electrons (eV)

Alkali-metal atoms	Inert-gas atom	
	He	Ne
Na	At $T<3000$, $W<444$ not observed	$T_0 \lesssim 230$, $W_0 \lesssim 107$
K	$T_0 \lesssim 900$, $W_0 \lesssim 84$	At $T<3000$, $W<1017$ not observed
Rb	$T_0<1500$, $W_0<67$	At $T<3000$, $W<571$ not observed
Cs	$T_0<4000$, $W_0<116$	$T_0 \lesssim 2600$, $W_0 \lesssim 340$

*T_0 and W_0 – threshold values of the energy (eV).

Table II. Transitions corresponding to the excitation of the lowest auto-ionization levels of alkali-metal atoms, and the energy of the electrons released upon auto-ionization of these states

Atom	Transition	E, eV
Cs	$(5p)^6 6s^1 S_0 \rightarrow (5p)^5 6s^2\, ^2P_{3/2}$	8,4 [11, 14]
Rb	$(4p)^6 5s^1 S_0 \rightarrow (4p)^5 5s^2\, ^2P_{3/2}$	11,1 [10, 14]
K	$(3p)^6 4s^1 S_0 \rightarrow (3p)^5 4s^2\, ^2P_{3/2}$	14,4 [9, 12, 14]

the corresponding figures show the result of differentiating the delay curves in the region of their steep descent.

By plotting the delay curves for different energies T, it is possible to estimate the position of the experimental thresholds T_0 for the appearance of the indicated groups of electrons. The data obtained by us relative to the position of the experimental thresholds of the appearance of the discrete electrons groups are summarized in Table I. On the basis of the data given in Fig. 4 and Table I, we can draw the following conclusions.

1. In the interval of our measurements, the indicated groups of electrons are not always excited with sufficient intensity, and only at a definite combination of partners.

2. The positions of the experimental threshold, for those cases when these groups are observed, also depend on the combination of the partners (compare the case of Cs-Ne and other cases).

3. In some cases the contribution of the three groups of electrons to the total number of emitted electrons can be appreciable (40% for Rb-He at an energy T = 3 keV)[2].

4. The energy interval for a given discrete group, in those cases when the group is excited, depends on the nature of the alkali-metal atom, decreasing with increasing atomic number of the alkali metal.

As stated earlier, in our earlier paper[1] we interpreted the appearance of the electron group for the K-He pair as the consequence of excitation of auto-ionization states of the atoms K with their subsequent decay. The results obtained in the present investigation agree fully with this point of view. From investigations of the absorption of light beyond the ionization threshold of alkali-metal atoms[9-12] it is known that there are a number of auto-ionization states of the atoms Cs, Rb, and K in the spectral region corresponding to the emitted-electron kinetic energies $E = h\nu - U_i$ of interest to us (U_i is the ionization potential of the alkali-metal atom). The kinks observed in our investigations on the delay curves, corresponding to the lower energies of the discrete groups of electrons, can be set in correspondence with the excitation energies established in[9-12] for the electron from the internal shell of the alkali-metal atom to the lowest of three states. In Table II are shown data pertaining to the transitions into the lowest of these states for the atoms Cs, Rb, and K. The energies E indicated in Table II are designated by arrows on the abscissa axes of the corresponding plots in Fig. 4. We see that they are close to the energies of the kinks on the delay curves for the corresponding cases. (We know of no other investigations of photoabsorption in Na vapor in the spectral region corresponding to E = 20–27 eV).

In the case of the Cs-Ne pair, under our conditions, only one discrete group of electrons with energy E = 8.4 eV is apparently excited with sufficient intensity[3]. The auto-ionization level closest in energy, $(5p)^5 6s^2\, ^2P_{1/2}$, would correspond, according to[11], to E = 9.6 eV. On the other hand, for the pairs Rb-He and K-He, and particularly for Na-Ne, the widths of the observed discrete groups of electrons amount to several electron volts.

It is therefore natural to assume that several auto-ionization levels are excited in these cases. (An investigation of these groups with the aid of an analyzer having a higher resolving power should answer this question.) Indeed, from studies of the photoabsorption in alkali-metal vapors, particularly from the recently published data[12], and also from investigations of electron-atom collision processes[13,14], it follows that, in the corresponding wavelength region (or electron energy region), there appear in these cases, to a greater or lesser degree, also other auto-ionization states with the different lifetimes, corresponding to the excitation of an electron from the external filled shell of the alkali-metal atom.

We can thus conclude the following:

1. In a number of the aforementioned cases, at low values of T (and correspondingly at low kinetic energy of the relative motion W), the formula of the Demkov and Komarov theory, based on an analysis of the transition of the discrete term of the quasimolecule to the continuous spectrum, generally provides a correct description of the energy distribution of the emitted electrons.

[2] The numerical values of the relative fraction of the indicated groups in the total amount of emitted electrons may be somewhat distorted, since under our experimental conditions electrons with energies on the order of tenths of an electron volt or less may not be entirely registered.

[3] Since the observed width of this discrete group consists of the "true" width of the line and of the smearing connected with the limiting resolving power of our analyzer, we can estimate the resolving power of the analyzer from the width of the "experimental" peak at half height (neglecting the "true" width of the electron). It turned out to be ~ 1:15.

2. With increasing T (and W), a deviation from the predictions of this theory may be observed, in that the number of "faster" electrons is larger than expected from formula (1).

3. In a number of the cases under consideration, starting with a certain "threshold" energy, there appear processes of electron excitation from the outer filled shell of the alkali-metal atoms; these processes may play a noticeable role even at relatively low energies of relative motion of the colliding particles.

4. Excitation of the corresponding discrete groups of electrons in the case of certain combinations of partners and their absence in the case of others cannot be explained if account is taken only of the kinetic energy of the relative motion or of the relative velocity of the colliding particles. It is probable that the properties of the quasi-molecule made up of the colliding particles come into play here.

In conclusion, the authors are deeply grateful to Professor V. M. Dukel'skiĭ for interest in the work.

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Translated by J. G. Adashko
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