EXPERIMENTAL INVESTIGATION OF THE EXCITATION OF Ar II *AND* Kr II *IN ELECTRON-ION COLLISIONS*

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A brief description is given of an intersecting-beam apparatus for investigating the excitation of ions in electron–ion collisions. The beams in this apparatus are of relatively high density and a monochromator is used for spectral separation of the observed radiation. Special features of the apparatus and of the measurement method are considered in detail. A discussion is given of the results obtained in the excitation of the spectral lines of Ar II from the levels $4p^2P^0_{1/2}$, $4p^2P^0_{3/2}$, $4p^4P_{5/2}$, $4p^2D^0_{3/2}$, $4p^2D^0_{5/2}$, and of the spectral lines of Kr II from the levels $5p^4P^0_{3/2}$, $5p^4P^0_{5/2}$, $5p^4D^0_{5/2}$, $5p'^2D^0_{5/2}$, $5p'^2F^0_{7/2}$. An interpretation is given of the most important features of the excitation of ions by electron impact.

THE inelastic processes of ionization and dissociation in electron–ion collisions have already been studied. However, the excitation of atomic ions is dealt with in just three experimental papers, each of which describes a study of the excitation of a single level or a spectral line (the metastable level of He II[1] and the resonance lines of Ba II[2] and Ca II[3]). In view of this, there is an urgent need to develop an apparatus and a measurement method which would be sufficiently universal and suitable for the investigation of the electron-impact excitation of various singly and multiply charged ions.

We spent many years in developing such apparatus. First of all, it was necessary to use a method for the generation and formation of ion beams which would ensure a considerable density in the region of intersection with an electron beam and this density would have to be much higher than the density in ion beams employed in earlier experiments.[1-3] The present paper describes the apparatus and measurement method as well as the results obtained in an investigation of the excitation of ten spectral lines of inert-gas ions Ar II and Kr II by electron–ion collisions.[1)]

APPARATUS

The apparatus consisted of the following principal units: an ion source, a mass spectrometer, a special-vacuum chamber, a power supply of the ion source and mass-spectrometer magnet, a system for modulation of the electron and ion beams, a system for recording the ion current and the emitted radiation, and a suitably branched vacuum pumping system.

A schematic diagram of the apparatus is given in Fig. 1. Ions were formed in an arc-discharge source 1 operating in a longitudinal magnetic field. The extraction, focusing, and acceleration of the ions were performed along the lines of force of the magnetic field of a solenoid 2 with the aid of an ion-optical system. In the energy range 5–40 keV the ion source produced an intense and well-focused beam which passed through the vacuum chamber 5 of a mass spectrometer which separated the ions in accordance with their specific charge. We used a 180° mass spectrometer with an inhomogeneous magnetic field up to 3000 Oe intensity and an equilibrium ion-trajectory radius of 0.5 m. The dispersion of our spectrometer was 5 mm per 1% change in the mass. The spectrometer foci were outside the magnet gap: the ion source was located at one focus and the other focus was close to the point of intersection of the electron and ion beams. The arc current of the ion source, the acceleration voltage, and the power supply of the electromagnet 7 of the mass spectrometer were stabilized so that the long-term stability of the ion current at the point of intersection of the beams was ± 2%.

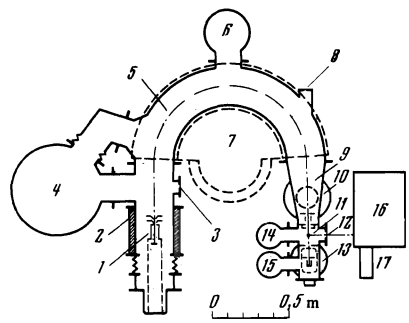

FIG. 1. Schematic diagram of the apparatus. 1–ion source 2–solenoid; 3, 8–viewing windows 4, 6, 10, 13, 14, 15–high-vacuum pumping units; 5–mass-spectrometer vacuum chamber; 7–mass-spectrometer magnet; 9–special high-vacuum chamber; 11–collision chamber 12–window for extraction of radiation; 16–spectroscopic instrument; 17–radiation (photon) detector.

After the mass spectrometer the ions entered a special high-vacuum chamber shown schematically in Fig. 2. This chamber contained an ion-beam modulator, collimating slits, an electron gun, as well as ion and electron detectors. The chamber was sealed with copper spacers and this made it possible to heat the chamber to 300–400°C. The chamber was divided into three sections, which were pumped separately. The electron gun was located in the middle section (II). The two beams intersected at right-angles. A window was provided for the extraction of the radiation generated at the point of intersection. The third section contained the

[1)] Some of the results of Ar II were published in a brief note. [4]

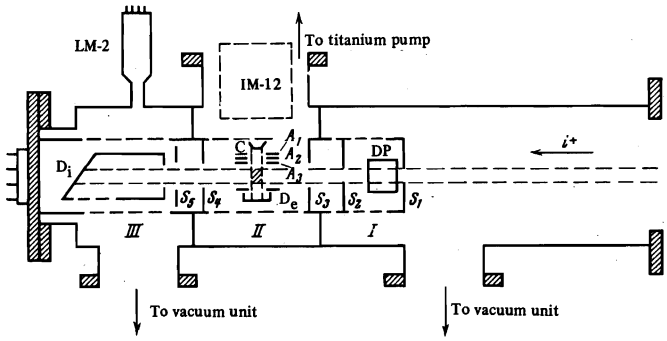

FIG. 2. Schematic diagram of the special high-vacuum chamber: I–III– sections of the vacuum chambers; S_1–S_4 are slits; DP are deflecting plates; C is the electron gun cathode A_1–A_3 are control electrodes in the electron gun; D_e and D_i are electron and ion detectors; S_5 is an electrode for retardation of secondary electrons; IM-12 are LM-2 vacuum gauges.

ion detector which was a deep Faraday cylinder with an electrode for retarding secondary electrons.

The use of a gas-discharge ion source and the need to inject an intense ion beam into the collision chamber demanded a high-power vacuum system capable of high pumping rates. Therefore, the section containing the ion source was pumped with a VA-5-4 pumping unit, the mass spectrometer chamber and the section where the ion beam was modulated were pumped with two VA-0.5-4 units, the collision chamber was pumped by a single heated unit with a special nitrogen trap and a titanium pump, and the ion detector section was pumped with a VA-0.5-4 unit. In this way we were able to achieve very low pressures ($\sim 5 \times 10^{-8}$ torr) in the collision chamber.

SPECIAL FEATURES OF THE APPARATUS AND MEASUREMENT METHOD

The radiation emitted by ions covers a wide range of wavelengths from visible to vacuum ultraviolet and can extend even to the x-ray region for multiply charged ions. The first stage of our investigation was restricted to a study of the radiation emitted by singly charged ions in the visible and ultraviolet parts of the spectrum.[2)] The investigated process excites the ions from the ground state and follows the reaction

$$A^+ + e \to A^{+*}(np) + e \to h\nu + A^{+*}(ns) + e, \qquad (1)$$

where A^+ are the ions in the ground state; A^{+*} are the excited ions; e are the electrons; ν is the frequency of the observed radiation. However, it is very difficult to isolate the signal due to the process (1) because of the presence of a background originating from many other processes. We shall now consider the main processes responsible for the background. The first process is the excitation of ions colliding with the atoms and molecules of the residual gases and terminating in the same energy states as in Eq. (1), i.e.,

$$A^+ + M \to A^{+*}(np) + M \to h\nu + A^{+*}(ns) + M, \qquad (2)$$

where M represents the molecules or atoms of the residual gases. The second process is due to the unavoidable penetration (for various reasons) of the neutral atoms of the investigated ions into the collision region. The concentration of these atoms may be considerably higher than the concentration of the atoms of the residual gases or of the ions in the primary beam. Consequently, the background at the same wavelength as before increases due to the following reactions

$$\begin{aligned} &\bar{A}^+ + A \to \bar{A}^{+*}(np) + A \to h\nu + \bar{A}^{+*}(ns) + A, \\ &\bar{A}^+ + A \to \bar{A}^+ + A^{+*}(np) + e \to \bar{A}^+ + h\nu + A^{+*}(ns) + e, \\ &\bar{A}^+ + A \to \bar{A} + A^{+*}(np) \to \bar{A} + h\nu + A^{+*}(ns), \end{aligned} \qquad (3)$$

and

$$e + A \to A^{+*}(np) + 2e \to h\nu + A^{+*}(ns) + 2e, \qquad (4)$$

where A are the neutral atoms of the investigated ions.

Thus, in the experiments involving the electron excitation of ions in intersecting beams, the useful signal is normally much smaller than the total background. Therefore, the useful signal can be detected only if the ion and electron beams are modulated. In our experiments the beams were modulated by rectangular pulses with an off-duty factor equal to unity and a phase shift amounting to a quarter of a period. The modulation frequency was made higher than the frequency of any systematic variations in the pressure of the residual gases.

The spectral line was selected with a monochromator and recorded with a photomultiplier which counted separate photoelectrons. The pulses produced by the photomultiplier were amplified by a preamplifier and a wide-band amplifier. The pulses generated in this way were locked to synchronization pulses provided by the modulation unit and applied to two PP-12 counters. The ion beam reached the collision zone during the first and second quarters of the modulation period but not during the third and fourth quarters. The electron beam was extracted by positive pulses in the second and third quarters but was blocked during the first and fourth quarters. In this modulation system the radiation was detected in two channels, one of which received signals during the first and third quarters of the period, and the other during the second and fourth quarters. In both channels the signals were summed during a specified time interval and the subsequent subtraction enabled us to identify the useful signal in its pure form.[5]

At low electron energies (up to 30 eV) some additional difficulties were encountered in the correct recording to the useful signal by simultaneous modulation of both beams and these difficulties were particularly due to the mutual influence of the electron and ion beams. However, in this range of energies one could use a method in which only the electron beam was modulated. In this case, the recorded signal was switched between two counting channels of the recording system in synchronism with the electron beam modulation. Subtraction of the counts in the two channels gave the sum of two signals, one representing the background due to the interaction of the electron beam with the atoms of the investigated ions in the residual gases and the other being the useful signal. This method made it possible to extract the useful signal in its pure form in the range of electron energies from the threshold of excitation of the investigated transitions from the ground state of the ion [reaction (1)] to the threshold of the excitation of the

[2)] A suitable spectral and detection with is being constructed in our laboratory for the extension of the investigation of the excitation of ions to the vacuum ultraviolet and soft x-ray regions.

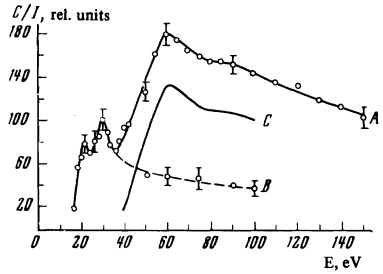

FIG. 3. Experimental excitation curves for the Ar II λ = 4880Å line: the continuous curve A (with points) is the excitation curve obtained by modulating just the electron beam; the dashed curve B (with points) is the excitation curve obtained by simultaneous modulation of both beams the continuous curve C is the difference between the other two curves, which is identical with the excitation function of the corresponding Ar II line in accordance with the reaction (4). [7]

same transitions from the ground state of the neutral atom [reaction (4)]. The two variants of our measurement method are illustrated in Fig. 3 by experimental curves obtained in the excitation of the λ = 4880 Å line of the argon ions.

The measurement method described above gave only the excitation function of the spectral line of a given ion. The absolute value of the cross section for the electron excitation of ions could be determined only after allowance for the motion of the excited ions in the direction perpendicular to the observation axis. Consequently, the number of ions emitting a given line in the viewing zone of the phonon detector was only a fraction of the total number of ions excited in this zone and emitting the same wavelength. We used a correction coefficient which gave the ratio of these two numbers under specified experimental conditions (this coefficient depended on the lifetime of the excited state of the ions, their velocity, and the slit width of the spectroscopic instrument).

The total excitation cross section Q_1 of a spectral line excited by electrons from the ground state was calculated from the following expression:

$$Q_1 = \frac{4\pi C_1 K}{S(\lambda)t(\lambda)N_i N_{e1}(v_{e1}^2 + v_i^2)^{1/2} V\Omega}, \quad (5)$$

where C_1 and $S(\lambda)$ are, respectively, the counting rate and the sensitivity of the recording apparatus; K is the correction coefficient allowing for the motion of the excited ions; $t(\lambda)$ is the transmission coefficient of the spectroscopic instrument at the wavelength λ; N_i and N_{ei} are, respectively, the concentrations of the ions and electrons in the collision zone; V is the volume of the collision zone from which the radiation is collected; Ω is the solid angle from which radiation is collected by the photon detector; v_{e1} is the velocity of the incident electrons; v_i is the velocity of the ions. For beams consisting of 10—15 keV ions the relative velocity $(v_{e1}^2 + v_i^2)^{1/2}$ was very nearly (with the exception of ions of the lightest elements) equal to the electron velocity v_{e1} in the laboratory system of coordinates.

The expression (5) for the determination of the excitation cross section could be used directly only when the various parameters characterizing the photon detector were available. Therefore, in many cases, it was convenient to calibrate the detector sensitivity by comparing the cross section Q_1 with the known cross section Q_2 of the reaction (4). The latter cross section could be represented in the form similar to Eq. (5),

$$Q_2 = \frac{4\pi C_2}{S(\lambda)t(\lambda)N_0 N_{e2} v_{e2} V\Omega}, \quad (6)$$

where C_2 is the counting rate of the recording apparatus; N_0 is the concentration of atoms of the investigated ionic species; N_{e2} and v_{e2} are, respectively, the concentration and velocity of electrons corresponding to the energy at which Q_2 is being determined. For a fixed configuration of the intersecting beams we find that Eqs. (5) and (6) yield the following working formula:

$$Q_1 = KQ_2 \frac{C_1 I_{e2} N_0}{C_2 I_{e1} N_i}, \quad (7)$$

where I_{e1} and I_{e2} are the total electron currents at the energies at which the cross sections are being compared.

EXPERIMENTAL RESULTS AND DISCUSSION

In the course of our first experiments on the excitation by electron—ion collisions, we determined the excitation functions and found the excitation cross sections of five spectral lines of Ar II originating from the levels $4p^2P^0_{1/2}$, $4p^2P^0_{3/2}$, $4p^4P_{5/2}$, $4p^2D^0_{3/2}$, $4p^2D^0_{5/2}$, and of five lines of Kr II originating from the levels $5p^4P^0_{3/2}$, $5p^4P^0_{5/2}$, $5p^4D^0_{5/2}$, $5p'^2D^0_{5/2}$, $5p'^2F^0_{7/2}$, which are of great interest particularly in connection with the mechanism of the coherent emission from inert-gas ion lasers.[6] The measurements were carried out in the electron energy range limited from above by the threshold values of the excitation energies of the same lines from the ground state of the neutral atoms, i.e., ~ 35 eV for Ar II and 30—32 eV for Kr II. The concentrations of the 10—15 keV argon and krypton ions in the beam-intersection zone was $(1—2) \times 10^8$ cm^{-3} and the density of the current of 10—100 eV electrons was $(3—5) \times 10^{-3}$ A/cm^2. The working voltage in the collision chamber was $(2—3) \times 10^{-7}$ torr. The energy inhomogeneity of the electrons at mid-amplitude of the distribution curve was 2—2.5 eV. An MDR-2 high-luminosity diffraction monochromator was used for the spectral separation of the radiation emitted by the excited ions.

The radiation was detected by modulating the electron beam with rectangular pulses and recording synchronously the pulses generated by an FÉU-64 photomultiplier in two counting channels. The absolute values of the excitation cross sections were found by calibrating the recording system with the aid of the known cross sections for the excitation of the investigated spectral lines from the ground state of the neutral atoms,[7,9] i.e., using Eq. (4). The error in the relative measurements of the excitation functions was ± 15% and the rms error in the determination of the cross sections was ± 40%.

The energy dependences of the excitation cross sections of the investigated Ar II lines are plotted in Figs. 4 and 5 and the corresponding dependences for the Kr II lines are given in Figs. 6 and 7. The 90% confidence interval in the relative measurements of the excitation functions is indicated in these figures and dashed vertical lines are used to denote the excitation thresholds of the upper levels of the relevant transitions. The meas-

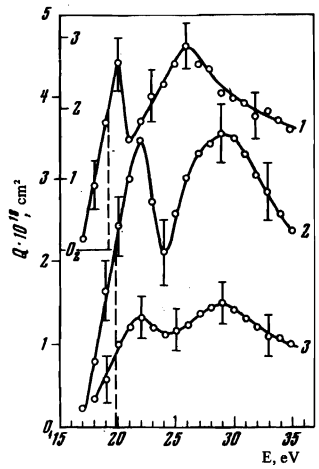

FIG. 4. Energy dependences of the excitation cross sections of Ar II lines 1–λ = 4806Å ($4s^2P_{3/2}$–$4p^2D^0_{5/2}$); 2–λ = 4658Å ($4s^2P^0_{3/2}$–$4p^2P^0_{1/2}$); 3–λ = 4545Å ($4s^2P_{3/2}$–$4p^2P^0_{3/2}$).

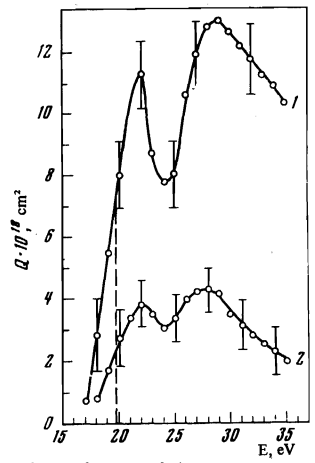

FIG. 5. Energy dependences of the excitation cross sections of Ar II lines: 1–λ = 4880Å ($4s^2P_{3/2}$–$4p^2D^0_{5/2}$); 2–λ = 4965Å ($4s^2P_{1/2}$–$4p^2D^0_{3/2}$).

ured cross sections indicated that the efficiency of the excitation of the argon and krypton ion lines by the impact of low-energy electrons was more than an order of magnitude higher than the maximum efficiency of the excitation in the electron–atom collisions.

It is evident from these figures that the emission was observed at about 2 eV below the excitation threshold of the upper levels. The energy of the electrons in the intersecting beams was calibrated reliably with the aid of the excitation threshold of the λ = 4200 Å line of the neutral argon. Therefore, the only reason for this shift was the energy scatter in the electron beam. Reduction of this scatter should shift the subthreshold maxima in the curves right up to the excitation thresholds of the investigated levels and should increase their amplitudes.

A characteristic feature of the excitation functions of all the investigated lines was the presence of two clear maxima, one located near the threshold and the other at electron energies of 28–29 eV in the case of Ar II and 23–26 eV in the case of Kr II. The nature of this structure was deduced by comparing one of our λ = 4880 Å curves for Ar II with the excitation functions for He II and Ca II obtained by Dance, Harrison, and Smith[1] and

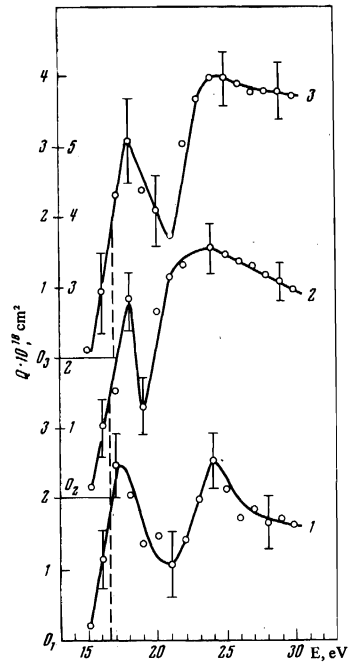

FIG. 6. Energy dependences of the excitation cross sections of Kr II lines: 1–λ = 4659Å ($5s^4P_{3/2}$–$5p^4P^0_{5/2}$) 2–λ = 4739Å ($5s^4P_{5/2}$–$5p^4P^0_{5/2}$); 3–λ = 4765Å ($5s^4P_{3/2}$–$5p^4D^0_{5/2}$).

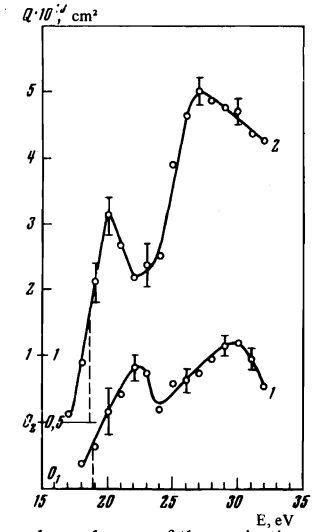

FIG. 7. Energy dependences of the excitation cross sections of Kr II lines 1–λ = 4088Å ($5s'^2D_{5/2}$–$5P'^2D^0_{5/2}$); 2–λ = 4577Å ($5s'^2D_{5/2}$–$5p'^2F^0_{7/2}$).

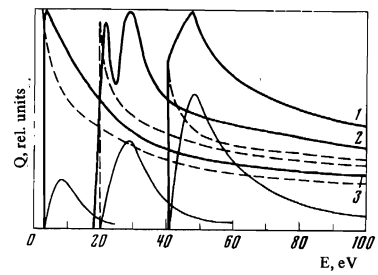

FIG. 8. Various electron-impact excitation functions of ions. The thick continuous curves are the experimental results: 1–He II; [1] 2–Ar II (our results) 3–Ca II. [3] The dashed curves are the results of calculations in the Coulomb-Born approximation based on a formula given in [10]. The thin continuous curves are the results of calculations by the Gryzinski method based on a formula given in [11].

Table I

Ion	Transitions	λ, Å	V_{exc}, eV	Position of maximum, eV	Q_{max}, cm² (experiment)	Q_{max}^{exch}, cm² (Gryzinski calc.)
HeII	$1s^2S_{1/2} \rightarrow 2s^2S_{1/2}$	—	40,8	48,0	$1.5 \cdot 10^{-18}$	$1 \cdot 10^{-18}$
ArII	$4p^2D^0_{5/2} \rightarrow 4s^2P_{3/2}$	4880	19,68	22; 29	$1.3 \cdot 10^{-17}$	$5 \cdot 10^{-18}$
CaII	$4p^2P^0_{3/2} \rightarrow 4s^2S_{1/2}$	3933	3,15	5	$2.7 \cdot 10^{-15}$	$5 \cdot 10^{-17}$

by Taylor and Dunn[3] (Fig. 8). We used a formula allowing for the Coulomb interaction (Coulomb-Born approximation[10]) to calculate the excitation cross sections for Ar II, He II, and Ca II (to within the oscillator strength). Next, we calculated the cross sections due to the exchange processes between the incident electrons and the electrons in the shells of the ions; this was done employing the classical Gryzinski formula.[11] All these curves are plotted on an arbitrary scale in Fig. 8. The excitation potentials of the levels of the ions, the maximum values of the excitation cross sections obtained in various experiments, and the values calculated using the Gryzinski formula collected in Table I.

An analysis of the curves in Fig. 8 and of the data in Table I shows that the contributions of the Coulomb interaction and the exchange process to the total excitation cross section depend quite differently on the atomic number of the ion and the excitation potential of the upper level. This is why the excitation functions behave differently in the subthreshold region: the sole maximum observed near the threshold of Ca II (and Ba II) is almost entirely due to the Coulomb interaction; the one maximum obtained in the case of He II is due to the exchange process and the Coulomb interaction is responsible only for the kink near the threshold; the two clear maxima observed for Ar II (and Kr II) are a consequence of approximately the same maximum efficiency of the two processes.

Thus, allowance for only these two interaction processes in the electron–ion collisions makes it possible to interpret in a unified manner the most important features of the excitation of the levels of all the atomic ions investigated experimentally so far.

It follows from this hypothesis that the individual nature of the energy levels is manifested less clearly in the electron excitation of ions than in the corresponding excitation of neutral atoms.

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