Experimental investigations of radiative decay of autoionizing states of alkali and alkaline earth elements

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An investigation was made of the vacuum ultraviolet emission spectra due to radiative decay of autoionizing states excited by collisions of electrons with K, Rb, Cs, Ca, Sr, and Ba atoms. The energy dependences of the excitation cross sections were determined for the strongest spectral lines corresponding to such decay, and the lines were identified. It was established that the autoionizing states whose radiative decay was investigated were subject also to considerable electron decay. The absolute values of the effective excitation cross sections were determined for some of the spectral lines of alkali atoms.

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1. INTRODUCTION

Autoionizing states of atomic systems have become the object of close attention in recent years. The term "autoionizing" is used for discrete energy states embedded in a continuum. They can appear as a result of excitation of an electron from an inner shell or of excitation of several electrons from an outer (valence) shell of an atom. The interaction of these states with the continuum gives rise to nonradiative decay, which is the dominant decay mechanism for these states. This is why many experimental investigations of autoionizing states have been carried out by the method of electron spectroscopy.

In principle, radiative decay of autoionizing states is also possible. Some experimental investigations of the excitation of alkali and alkaline earth atoms by electron impact have provided an indirect but convincing proof of radiative decay.[1,2] The ratio of the radiative and nonradiative decay probabilities is the prime factor which determines how realistic it is to expect radiative decay of a given autoionizing state. Since the probability of the Coulomb autoionization is 3–5 orders of magnitude higher than the probability of radiative transitions, it is clear that radiation can be expected only for those autoionizing states whose Coulomb autoionization is for some reason forbidden (in other words, we can expect radiative decay only in the case of levels which are metastable with respect to autoionization).

The existence of significant radiative decay of some autoionizing states of Li atoms is indicated by the results of beam–film experiments.[3] The first direct observations of radiative decay of such states of alkali and alkaline earth atoms excited by electrons were made in our laboratory. The first results of these observations were reported earlier.[4-6] The use of the term "autoionizing states" in our case is quite arbitrary precisely because of the observation of the radiative decay of these states. However, we shall show later that in no way this excludes the possibility of decay of these states accompanied by the release of an electron. Moreover, it is also known that autoionizing states metastable in respect of the autoionization and radiative decay exist in the case of alkali and alkaline earth elements.[7,8]

The present paper reports and analyzes the results of an investigation of radiative decay of autoionizing states of alkali and alkaline earth atoms excited by electron collisions with atoms.

2. EXPERIMENTAL METHOD

We used intersecting electron and atomic beams in an arrangement shown schematically in Fig. 1a. The electron beam was generated by a Pierce gun, whereas the atomic beam was produced by an effusion chamber with two shaping slits. Typical parameters of the beams were as follows: the current density in the electron beam was $\sim 2\times 10^{-3}$ A/cm^2 with the electron energy scatter ~1.5 eV; the concentration of atoms in the beam intersection region did not exceed $\sim 10^{12}$ cm^{-3}.

Vacuum ultraviolet (VUV) radiation which appeared in the beam intersection region was analyzed spectroscopically with a vacuum monochromator based on the optical Seya–Namioka scheme. This monochromator

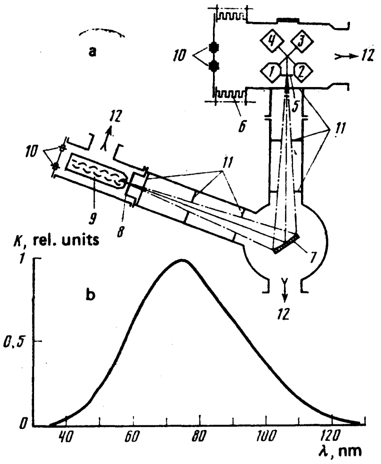

FIG. 1. a) Schematic diagram of the apparatus: *1*) electron beam source; *2*) atomic beam source; *3*) electron detector; *4*) collector of atoms; *5*) entry slit; *6*) adjustment bellows; *7*) diffraction grating; *8*) exit slit; *9*) photomultiplier; *10*) electrical terminals for vacuum system; *11*) slits; *12*) to vacuum pump. b) Relative spectral sensitivity of the monochromator—multiplier system.

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included a diffraction grating replica with a radius of curvature 0.5 m and 1200 lines/mm; the replica was coated by an aluminum film. Radiation was detected with an open photomultiplier operated as a photon counter.

The monochromator–photomultiplier system made it possible to investigate the spectral range 40–120 nm. The relative spectral sensitivity of the system was determined (Fig. 1b) using synchrotron radiation from the VEPP-2M storage ring at the Insititute of Nuclear Physics, Siberian Branch of the USSR Academy of Sciences, Novosibirsk.

3. EXPERIMENTAL RESULTS AND DISCUSSION

An investigation of the VUV spectra of alkali and alkaline earth atoms excited by electron–atom collisions established that the strongest (in this wavelength range) were the resonance lines of singly charged ions in the case of alkali atoms and of doubly charged ions in the case of alkaline earth atoms.[9-12] Moreover, there were several spectral lines not observed earlier. These lines appeared in the spectra when the bombarding electron energy was insufficient to excite the resonance lines just mentioned. The wavelengths and excitations thresholds of these lines are listed for each element in Table I with an indication of an experimental error. We included also in Table I the maximum intensities (in percent), relative to the maximum intensities of the corresponding resonance lines obtained allowing for the spectral sensitivity of the apparatus. The following notation is used in Table I: λ is the emission wavelength (nm), E_{exc} is the excitation energy of a given spectral line (eV), E_{max} is the energy of the incident electrons (eV) at which the excitation function of the spectral line has its maximum, I_{max} is the intensity of the spectral line at the maximum of its excitation function [in the case of alkali atoms the intensities are given in percent relative to the intensities of the resonance lines of singly charged ions corresponding to the transitions $np^6\,^1S_0 - np^5(^2P^0_{3/2})(n+1)s[\frac{3}{2}]_1^0$, whereas in the case of alkaline earth atoms the corresponding intensities are given for the lines due to the $np^6\,^1S_0 - np^5(^2P^0_{3/2})md[\frac{1}{2}]_1^0$ transitions in doubly charged ions, where $n=3$, 4, and 5 refers to K and Ca, Rb and Sr, Cs and Ba, respectively], Q_{max} is the excitation cross section of the spectral line (in units of 10^{-17} cm^2) corresponding to the maximum of its excitation function.

The excitation functions were determined for the strongest of these lines. The relative error in the determination of the excitation function ordinates never exceeded 10% (confidence level 0.9). The energies at which the excitation functions had their maxima are also listed in Table I. The excitation functions were generally similar for each of the investigated elements. Figures 2 and 3 show the excitation functions for the high-intensity lines (one line for each element). It should be pointed out that the excitation functions of the newly discovered lines of alkali and alkaline earth elements differ strongly from the excitation functions of the remaining lines present in the spectra (mainly the resonance lines of the singly and doubly charged ions whose typical excitation functions[9-12] are included in Figs. 2 and 3 for comparison). A characteristic feature of the excitation function of the new lines is the presence of a steep near-threshold rise and the attainment of a maximum near the threshold. The excitation of these lines occurs effectively in a relatively narrow range of energies of the bombarding electrons. The exception to this rule is strontium (curve 3 in Fig. 3), because in this case the excitation function not only has a steep near-threshold maximum but also a second rise with a maximum at ~100 eV.

A careful analysis of the experimental conditions showed that all the lines listed in Table I belong to atoms or ions of the investigated elements. The experimental values of the excitation thresholds of these lines (Table I) led to a firm conclusion that they are due to the excitation of autoionizng states of atoms or ions formed with the participation of the outer p electrons. The attribution of each of the lines to an appropriate atom or ion is given in Table I. We can see that in the case of alkali elements all the lines belong to neutral atoms, whereas in the case of alkaline earths this is true only of strontium. The lines of other alkaline earths represent singly charged ions.

TABLE I.

	Experiment		Calculation		Identification	E_{max}	I_{max}	Q_{max}
	λ	E_{exc}	λ	E_{exc}				
K I	67.5±0.2	20±2	67.3	20.021	$3p^64p\,^2P^0_{3/2} - 3p^54s4p\,^4S_{3/2}$ [21]	–	–	–
	72.3±0.2	19.8±0.5	72.1	19.865	$3p^63d\,^2D_{5/2}$ — $3p^54s3d\,^4P^0_{5/2}$ [21]	28	9.5	0.29
	75.5±0.2	19.8±0.5	75.3	19.865	$3p^64d\,^2D_{5/2}$ —	28	4	0.13
	77.0±0.2	20±2	76.9	19.865	$3p^65d\,^2D_{5/2}$ —	–	–	–
Rb I	82.4±0.2	17.4±1	82.4	16.633	$4p^65p\,^2P^0_{3/2} - 4p^55s5p\,^4S_{3/2}$?	22	4	0.28
	85.0±0.2	17±2	85.1	16.966	$4p^64d\,^2D_{5/2} - 4p^55s4d\,^4P^0_{5/2}$ [22]	–	–	–
Cs I	96.0±0.2	14.3±1.5	95.9	12.927	$5p^66s\,^2S_{1/2} - 5p^56s5d\,^4P^0_{3/2}$ [23] (?)	16	11	1.1
	104.8±0.3	14±2	–	–	—	–	–	–
	108.5±0.3	14.1±1.5	108.8	13.201	$5p^65d\,^2D_{5/2} - 5p^56s5d\,^4P^0_{5/2}$ [23]	14.5	60	5.8
	111.1±0.3	14±2	111.4	12.927	$5p^65d\,^2D_{3/2} - 5p^56s5d\,^4P^0_{3/2}$ [23]	–	–	–
	119.6±0.4	14±2	–	–	—	–	–	–
Ca II	51.1±0.2	32.1±0.5	51.1	32.047	$3p^63d\,^2D_{5/2} - 3p^54s3d\,^4P_{7/2}$ [21]	39	18	–
	53.4±0.2	31.0±0.5	53.3	31.072	$3p^63d\,^2D_{5/2} - 3p^54s3d\,^4P_{5/2}$ [21]	40	71	–
Sr I	58.4±0.2	24.8±1.5	58.5	23.45	$4p^64d\,^3D_3$ — 23.45 эВ [18]	28; 100	5.0	–
	62.4±0.2	22.1±1.5	62.4	23.45	$4p^64s\,^3S_1$ — 23.45 эВ [18]	28; 130	2.4	–
Ba II	71.2±0.2	23.3±0.5	–	–	—	27	48	–

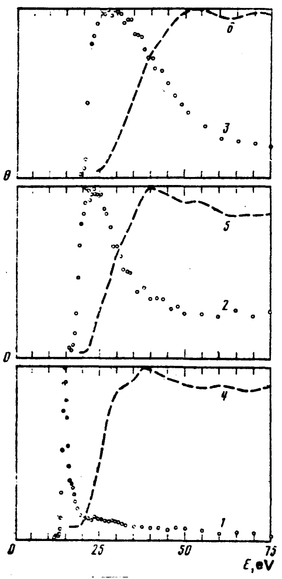

FIG. 2. Excitation functions of the spectral lines of alkali elements (relative units) corresponding to transitions from photoionizing states (points): *1*) 108.5 nm (Cs); *2*) 82.4 nm (Rb); *3*) 72.3 nm (K). The dashed curves represent the excitation functions of the resonance lines of singly charged ions of the same elements corresponding to the $np^6\,{}^1S_0$–$np^5({}^2P^0_{3/2})(n+1)s[\frac{3}{2}]^0_1$ transitions[9]: *4*) 92.7 nm (Cs); *5*) 74.1 nm (Rb); *6*) 61.3 nm (K).

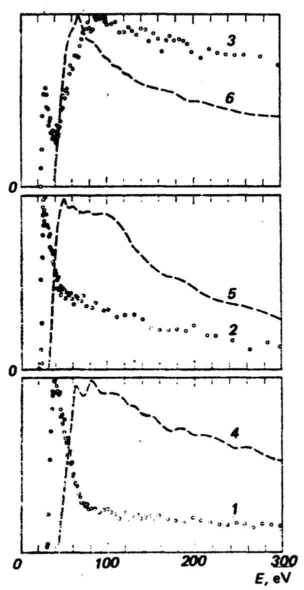

FIG. 3. Excitation functions of spectral lines of alkaline earth elements (relative units) corresponding to transitions from autoionizing states (points): *1*) 53.4 nm (Ca); *2*) 71.2 nm (Ba); *3*) 58.4 nm (Sr). The dashed curves represent the excitation functions of the resonance lines of doubly charged ions of the same elements corresponding to the $np^6\,{}^1S_0$–$np^5({}^2P^0_{3/2})nd[\frac{1}{2}]^0_1$ transitions[10–12]: *4*) 49.1 nm (Ca); *5*) 74.3 nm (Ba); *6*) 56.3 nm (Sr).

In our earlier communications[4–6] we made the first attempt to identify the lines due to autoinizing states of alkalis and alkaline earths. In the present paper we shall review the earlier identifications and also identify other lines.

We have been able to identify the spectral lines corresponding to the decay of autoionizing states by utilizing the results of work on the electron spectra of the elements in question.[13–22] The electron spectra of potassium and calcium[13,16,17,21] have been investigated and identified more thoroughly than the others. We used these spectra to identify the majority of the autoionizing levels of K I and Ca II belonging to the lower configurations. In particular, almost all the levels of the configuration $3p^54s3d$ were identified. Since the K I and Ca II lines identified by us from the measured excitation thresholds also lie in the region of autoionizing states of the lowest configurations, we used the results of Ref. 21 to identify these lines. The identification procedure consisted of a selection of the upper (among the autoionizing levels) and lower (among the ordinary levels) states in such a way that the calculated and measured wavelengths agreed within the limits of the experimental error and that the excitation thresholds obtained from the electron spectra agreed with those determined in our study. Naturally, we had to satisfy the selection rules for the dipole electric transitions, or at least the most general of these rules relating to the parity $\Delta\pi = \pm 1$ and the total momentum $\Delta J = 0, \pm 1$. Moreover, in the selection of the upper state we gave preference to the optically forbidden autoionizing levels (relative to the ground state of the atom), since all the optically allowed levels experienced such fast autoionization that the radiative decay could not compete with it.

This procedure allowed us to establish reliably that all the new potassium and calcium lines were due to radiative transitions from the same autoionizing states whose decay was observed in the electron spectra. Table I lists the wavelengths and excitation thresholds of these lines calculated from the electron specta. It also includes the results of our identification procedure. We can see that in the case of potassium the 72.1, 75.3, and 76.9 nm lines are members of a series with the common upper level $3p^54s3d^4P^0_{3/2}$, whereas the 67.3 nm line originates from the level $3p^54s4p^4S_{3/2}$. In the case of calcium the identification procedure showed that both observed lines originate from the levels with the same configuration $3p^54s3d^4F^0_{3/2}$ and $3p^54s3d^4P^0_{5/2}$.

As far as the other alkalis are concerned, an analysis of the electron spectra of rubidium[14,22] made it possible to identify the majority of the low autoionizing levels of Rb I, whereas as in the case of cesium[15,23] only the ground-state doublet $5p^56s^2\,{}^2P_{1/2,3/2}$ was identified together with five lebels of the $5p^56s5d$ configuration. As in the case of potassium and calcium, it was possible to identify tentatively some of the new rubidium and cesium lines (Table I).

The identification of the Rb I line at 82.4 nm as due to the $4p^65p^2P^0_{3/2} - 4p^55s5p^4S_{3/2}$ transition was based on the analogy between potassium and rubidium. If this analogy is justified, the line in question should correspond to a line with an excitation energy of 16.683 eV in the electron spectrum. However, such a line was not mentioned in Ref. 14, although it might have been present in the spectrum at low energies of the exciting electron beam: it could be masked by a strong line at 16.683 eV (see the spectrum in Ref. 14). However, it should be pointed out that, according to the calculations of Ref. 22, the state $4p^5 5s5p^4 S_{3/2}$ was located considerably lower at 15.853 eV.

In spite of the good agreement between the experimental and calculated wavelengths and excitation thresholds, the identification of the cesium line at 96.0 nm as due to a combination of the $5p^5 6s5d\,^4P^0_{3/2}$ level with the ground state of the cesium atom was very doubtful because the photoabsorption spectra of cesium vapor showed no such line. The cesium lines at 104.8 and 119.6 nm could not be attributed to any lines in the electron spectra. The origin of some of the unidentified cesium lines could be due (as in the case of potassium) to the excitation of the $^4S_{3/2}$ state with the p^5sp configuration.

In the case of alkaline earth elements (strontium and barium), the situation was more complex. Although the electronic spectra were available for these elements, there have been hardly any identifications.

In an analysis of the electron spectrum of strontium[18,19] we were also able to find autoionizing levels which we could, in principle, attribute it to new Sr I lines. It was found that both observed lines (58.4 and 62.4 nm) had the same upper level. In Table I we gave only the excitation energy of this level, because the question of its identification is still open. The corresponding line in the electron spectrum of strontium had the energy 17.76 eV and corresponding to decay of the autoionizing state of the Sr atom producing the ground state of the Sr^+ ion. In Ref. 18, this line was not assigned definitely to either Sr I or Sr II, although it was suggested that it could be due to decay of the autoionizing state of Sr I $4p^5 5s^2 4d\,^1P_1$ producing the excited state $4p^6 4d\,^2D$ of the Sr II ion. In Ref. 19 the energy of this line was given as 17.81 eV and it was attributed definitely to the autoionizing transition in question. Clearly, the reliability of our selection of the autoionizing level in the identification of the new strontium lines depended on the state in which the Sr^+ ion appeared after electron decay of the autoionizing state corresponding to the line at 17.76 eV (or 17.81 eV). Resolution of the latter question should make it possible to determine accurately the threshold of appearance of this line in the electron spectrum.

In the case of barium we were unable to select from the levels in Ref. 20 the autoionizing state appropriate for the identification of the Ba II line at 71.2 nm.

The electron lines of the alkali (excluding cesium) and alkaline earth elements corresponding to our new optical lines had a low intensity.

A good verification of the proposed identification was provided by a comparison of the energy dependences of the electron and optical lines brought into one-to-one correspondence. Unfortunately, the published information on the electronic spectra of alkalis and alkaline earths enabled us to make this comparison only in the case of cesium. Figure 4 shows the excitation functions of the spectral line of cesium at 108.5 nm identified by us as a transition from the $5p^5 6s5d^4P^0_{5/2}$ level and the excitation function[24] of the electron line of cesium of 12.930 eV energy[15] corresponding to decay of the autoionizing state $5p^5 6s5d^4P^0$ (Ref. 23) (this comparison was meaningful because we were dealing with levels of the same multiplet differing only in respect to the total angular momentum, and characterized by similar excitation functions). The excellent agreement between these excitation functions provides a convincing proof of the correctness of the identification of the 108.5 nm line.

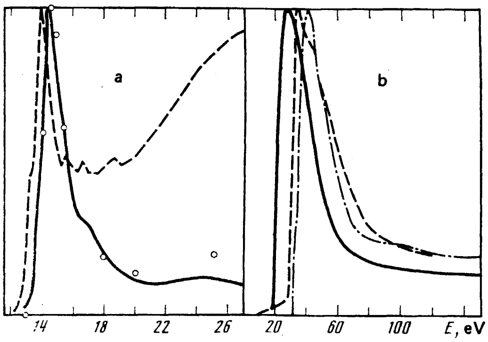

FIG. 4. a) Comparison of the excitation functions of the autoionizing states of the cesium atom: the continuous curve represents our results for the 108.5 nm line and the dashed curve gives the results of Feldman and Novick,[7] and the points the results of Pejčev and Ross.[24] b) Comparison of the excitation functions of the autoionizing states of potassium and calcium: the continuous curve represents our data for the 72.3 line of K, the dashed curve gives our results for the 53.4 nm line of Ca, and the chain curve represents the results of Kupriyanov.[8]

We determined the relative intensities of the observed spectral lines for each element (Table I). In the case of the alkali atoms the strongest (relative to the resonance lines of the corresponding singly charged ions) were the lines of cesium. In the case of alkaline earths the highest relative intensity was recorded for the calcium lines. In the case of cesium, calcium, and barium these lines were comparable in intensity with the resonance lines of the ions. The lowest relative intensities were those of the new lines of rubidium and strontium (almost an order of magnitude less than in the case of cesium and calcium). The generally anomalous behavior of strontium compared with the other alkaline earths has already been pointed out above. There must be some common factor responsible for all these anomalies of strontium, but it is at present difficult to see what this factor is.

The last column of Table I gives the maximum absolute values of the excitation cross sections of the strongest of the new lines of the alkali atoms. These cross sections were determined from the ratio of the intensities of these lines to those of the resonance lines of singly charged ions, and from the absolute excitation cross sections of the latter found by normalization of their excitation functions to the theoretical ionization cross sections of the outer p electrons[25] at the energy of 300 eV. Our estimates indicated that the absolute cross sections found in this way were accurate to within a factor of 2–3. One should mention particularly the high values of the excitation cross sections ($>10^{-17}$ cm^2) of the spectral lines of cesium at 108.5 and 96.0 nm.

Unfortunately, the same method could not be used to determine the absolute excitation cross sections of the new lines of the alkaline earth elements, because there were no data whatever on the absolute excitation cross sections of the resonance lines of their doubly charged ions formed as a result of collisions between electrons and atoms.

It was interesting to compare also the excitation cross sections of the autoionizing states of various types (particularly the difference between the nature of the decay):

1) the states decaying mainly by the release of an electron;

2) the states with a significant radiative decay, i.e., those investigated in the present study;

3) the states metastable in respect of electron and radiative decay. The maximum excitation cross sections of the states of the last (third) type of alkali atoms determined by Feldman and Novick[7] were very small: ~10^{19}–10^{20} cm^2. It is clear from Table I that the excitation cross sections of the autoionizing states of the same atoms, deduced by us from radiative decay, i.e., the excitation cross sections of states of the second type were 1–2 orders of magnitude higher than the corresponding values for the states of the third type. In fact, this difference should be even greater because the absolute excitation cross sections were determined by considering only the radiative decay channel and ignoring the electron decay.

Knowing the excitation cross sections of a state obtained by investigating each of the decay channels separately, we could have determined the ratio of the probabilities of the electron and radiative decay of the state, which would have been of considerable interest for the physics of autoionizing states. Unfortunately, no experimental determinations have yet been made of the absolute excitation cross sections of the autoionizing states on the basis of the electron decay.

One could also compare our excitation cross sections of the $np^5(n+1)snd\,^4P_{5/2}$ states of the alkali atoms ($n=3, 4, 5$, respectively, for K, Rb, and Cs) with those calculated in Ref. 26. This comparison yielded an agreement (within a factor of 3) between the theoretical cross sections (calculated in the approximation of distorted waves allowing for the exchange) and the experimental cross sections at electron energies 1.2–2 times higher than the excitation thresholds. However, calculations carried out by the Born–Oppenheimer method[26] in the case of K and Rb gave cross sections an order of magnitude higher than those found experimentally. The energy dependences of the excitation cross sections in the case of K and Rb calculated by the Born–Oppenheimer method agree better with the experimental values than the cross sections found by the method of distorted waves allowing for exchange although in the case of Cs both theoretical methods gave results very different from those found experimentally.

We shall now consider the excitation functions of the new spectral lines. We pointed out earlier that the excitation of these lines occurred effectively in a relatively narrow range of energies (with the exception of strontium). In the case of alkali elements we found (Fig. 2) that the lowest "width" (~3 eV at midamplitude) was exhibited by the cesium excitation functions.[1)] The excitation functions of potassium and rubidium were considerably wider (~30 and ~15 eV, respectively). The narrow excitation functions of the new cesium lines were typical of the exchange excitation processes accompanied by a change in multiplicity. This provided an additional proof that the initial states of the 96.0 and 108.5 nm cesium lines were quartets (Table I) because the ground state of the cesium atom was a doublet. In the case of potassium and rubidium the initial states of the new lines were also of the quartet type (Table I) but the excitation functions of these lines were very different from those of the quartet (Fig. 2). Naturally, one could assume that this was due to the possible mixing of the initial quartet states with the rapidly autoionizing doublets, which would account for the broadening of the excitation functions of the corresponding lines. Thus, if the difference in the behavior of the excitation functions were indeed due to the mixing of doublets and quartets, this mixing should be minimal in the case of cesium and it should increase on reduction in the atomic number. It follows from the published results[21] that the mixing effects are greater in the case of heavier atoms.

Feldman and Novick[7] determined the excitation functions for electron impact of metastable (in respect of autoionizing and radiative decay) autoionizing states of alkali atoms (the excitation functions of the corresponding states of singly charged calcium and strontium ions formed by collisions of atoms with electrons were investigated by Kupriyanov[8]). A comparison revealed an unexpectedly good agreement between these functions for K, Rb, and Cs and the excitation functions of the new spectral lines of the same elements, particularly in the threshold region. The agreement improved on increase in the atomic weight of the atom. Figure 4a presents such a comparison for cesium. We can see that the excitation functions agree practically completely in the region of the threshold maximum (the rise beginning from ~15 eV in the case of Cs is attributed by Feldman and Novick[7] to the experimental inaccuracy). Clearly, this similarity of the excitation functions of the different states can be explained by the common nature of the excitation mechanism (exchange accompanied by a change in the multiplicity), because the metastable levels of the alkali atoms investigated by Feldman and Novick[7] are also quartets.

In the case of the excitation functions of the new lines of alkaline earths, a comparison of the same kind as for alkalis can be made only for calcium (Fig. 4b). We can see that the excitation functions of calcium being compared are very similar. This similarity is not accidental and it gives the key to the understanding of the mechanism for the excitation of the corresponding ionic autoionizing states of calcium and barium, if we bear in mind the following circumstance.

Photoelectron spectroscopy of barium atoms[27] revealed the effective formation of Ba^{2+} ions by a two-stage autoionization process: first the atomic autoionizing states of the $5p^56s^25d$, configurations were excited and these decayed producing autoionizing states $5p^56s5d$ of the Ba^+ ion, decaying finally to the ground state of Ba^{2+}. This process is possible because of a certain characteristic of the energy structure of barium; some of the autoionizing states of the Ba^+ ion are located below the atomic autoionizing states on the energy scale. The same feature is exhibited also by calcium,[28] but not by strontium. Therefore, it is quite likely that the excitation of the autoionizing states of Ca^+ and Ba^+ observed by us is not a direct process but occurs via atomic autoionizing states of different multiplicity than the ground state of the atom (which is a triplet).

In conclusion, the authors express their deep gratitude to the Staff at the Institute of Nuclear Physics of the Siberian Branch of the USSR Academy of Sciences for an opportunity to use synchrotron radiation from the VEPP-2M storage ring and the to late S. E. Kupriyanov for valauble discussions.

[1)] In the case of cesium the "width" of the excitation function was close to the energy scatter of the electron beam. Therefore, one could expect a further improvement in the monokineticity of the electron beam and reduce further the "width" of these excitation functions.

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