Electron spectroscopy of autoionizing states of ytterbium

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(Submitted 30 June 1982)
Zh. Eksp. Teor. Fiz. **84**, 502–512 (February 1983)

The spectra of the electrons emitted by ytterbium atoms and ions in the decay of autoionizing states were found to contain 76 lines, more than double the previously known number. The low-energy part of the spectrum, observed for the first time ever, is caused by the decay of the states of the $4f^{14}$ and $6s^2$ subshells of the atom.

PACS numbers: 32.80.Dz

The investigation of autoionizing states (AIS) of ytterbium atoms and ions is of considerable interest not only from the point of view of atomic-structure theory, but also in connection with the practical needs of quantum electronics[1] and of methods of laser isotope separation, which are being developed in recent years.[2]

Atomic AIS were investigated by the classical photoabsorption method in Refs. 3 and 4. An appreciable number of these states lies near the first ionization limit ($E_i = 6.25$ eV) and is due to excitation of the $4f^{14}$ and $6s^2$ subshells of the atom.[3] The high-energy part of the spectrum is due to excitation of the $5p^6$ subshell and is characterized by the presence of a giant $5p^5(^2P_{1/2})6s^25d\,[3/2]_1$ resonance, whereas the intensity of the other transitions is low.[4] Naturally, this method can be used to observe only AIS that combine with the ground $5p^64f^{14}6s^2\,{}^1S_0$ state of the atom.

Qualitatively new results were obtained following the advent of the method of multistep laser ionization of atoms.[5] This method, with the use of various intermediate lasers, has made it possible to record atomic transitions that are optically forbidden in the usual sense of this word. Experiment revealed exceedingly narrow ytterbium AIS produced by twice exciting the valence $6s^2$ subshell. The theoretical calculations are in good agreement with experiment and predict the existence of new states.[5,6]

The decay of the atomic and ionic AIS of Yb was investigated by the method of electron spectroscopy practically simultaneously in two independent studies.[7,8] The authors of these references first arrived at the conclusion that the AIS undergo multichannel decay, a conclusion that constituted an undisputed success of the method of electron spectroscopy in the study of autoionizing phenomena. The authors of Refs. 7 and 8 noted also a strong continuum background against which discrete lines are observed and undergo a substantial broadening that exceeds the width of the instrumental function.

Notwithstanding the interesting results obtained in Refs. 7 and 8, we note that the investigations were carried out at exciting-electron-beam energies exceeding 70 eV, much above the thresholds of the observable processes, at an insufficient energy resolution, and with a sharp decrease of the analyzer transmission at $E_{em} < 6$ eV. The last circumstances in conjunction with the small contrast of the lines gave the impression that the spectra are relatively simple. We have succeeded, with a better energy resolution, to drop to the threshold energy of the exciting beam and investigate the decay of states adjacent to the single-ionization limit; this allowed us to observe a large number of lines hitherto unobserved in the electron spectra.

APPARATUS

We used the electron spectrometer described in Ref. 9 and revamped to permit high-temperature investigations. Just as in Ref. 9, we recorded the spectra of the electrons emitted by the atoms at an angle 90° to the propagation direction of the primary electron beam. The high sensitivity and stability of the spectrometer parameters made it possible

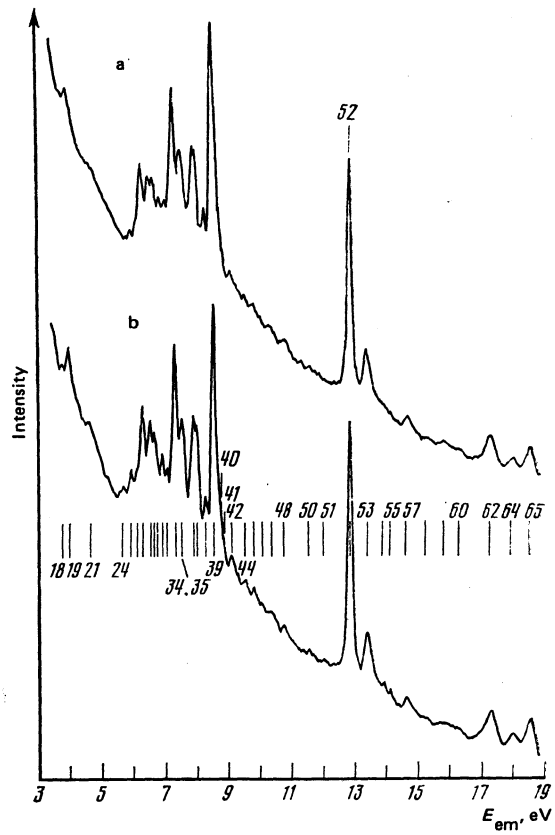

FIG. 1. Spectra of emitted electrons for beam energies $E = 400$ (a) and 200 eV (b).

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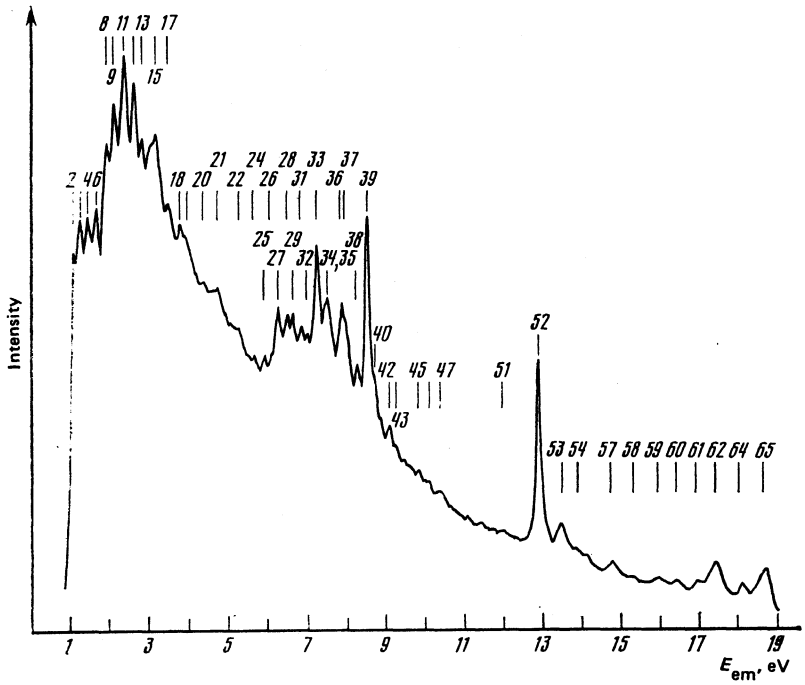

FIG. 2. Spectrum of emitted electrons at an energy $E = 73$ eV.

to record reliably the spectra in the primary-beam energy interval $E = 8$–500 eV and in the emitted-electron energy interval $E_{em} = 0.9$–30 eV. The primary-beam current at maximum energy was $\sim 10^{-4}$ A, and the pulse counting rate of the emitted-electron detector (a channel electron multiplier) was 10^5 sec^{-1}. The experiments were performed with the metallic ytterbium reservoir at a temperature 400 °C. The spectra were plotted at a constant analysis energy (1–1.5 eV) and they were scanned by linear variation of the potential of the analyzer entrance diaphragm.

RESULTS

Typical spectra of the emitted electrons, recorded at various energies of the bombarding beam, are shown in Figs. 1–5. The time required to record one spectrum was about 5 min. The absolute energy scale was calibrated against the intense line 52 corresponding to the Auger transition $5p^5 4f^{14} 6s^2\ ^2P_{3/2} \rightarrow 5p^6 4f^{14}\ ^1S_0$. The energy position of this line was determined with sufficient reliability by the authors of Refs. 7 and 8 and corresponds to the transition energy 12.91 eV known from spectroscopic data.[10] It was this that prompted us to choose it as the reference.

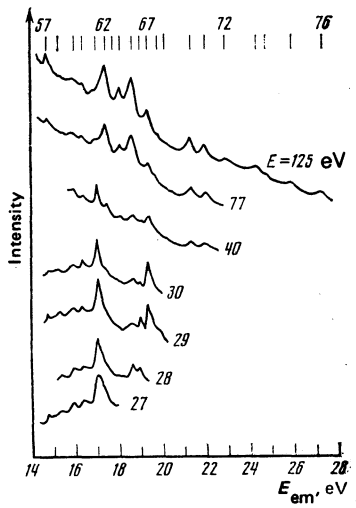

FIG. 3. High-energy part of the spectrum for different beam energies.

The energy resolution of the analyzer was determined from the width of the narrowest line in the spectrum, measured at half maximum. Such a line was line 6, whose half-width at 15 eV amounted to 0.04 eV. We note that the real resolution can be even better.

The energies of the lines observed by us are gathered in Table I, which lists also experimental results obtained by others and known to us. The values in the table are averages of not less than over 20 spectra, and the accuracy with which the energy was determined for most lines is estimated at $\pm$ (0.02–0.03) eV.

We subdivide arbitrarily the entire spectral interval into three regions: up to 5 eV, 5–15 eV, and 15–30 eV. We consider first the energy region 5–15 eV, which contains the most intense lines. With respect to their character and the relative line intensities our spectra are close to the previously observed ones,[7,8] and when the energy is varied from 500 to 70 eV (see Figs. 1 and 2) no noticeable changes take place in the structure of the spectrum, a fact already noted in Ref. 7. The improved energy resolution, however, enabled us to observe a large number of additional lines that were not resolved by others. Many known lines have a fine structure, and new lines previously masked by the background appear. Most lines in this region are the result of the multichannel decay of the state YbII$5p^5 4f^{14} 6s^2\ ^2P_{3/2}$ to the ground and excited states of YbIII. We attribute to them also the newly recorded lines.

The ability to work with low-energy-electron beams al-

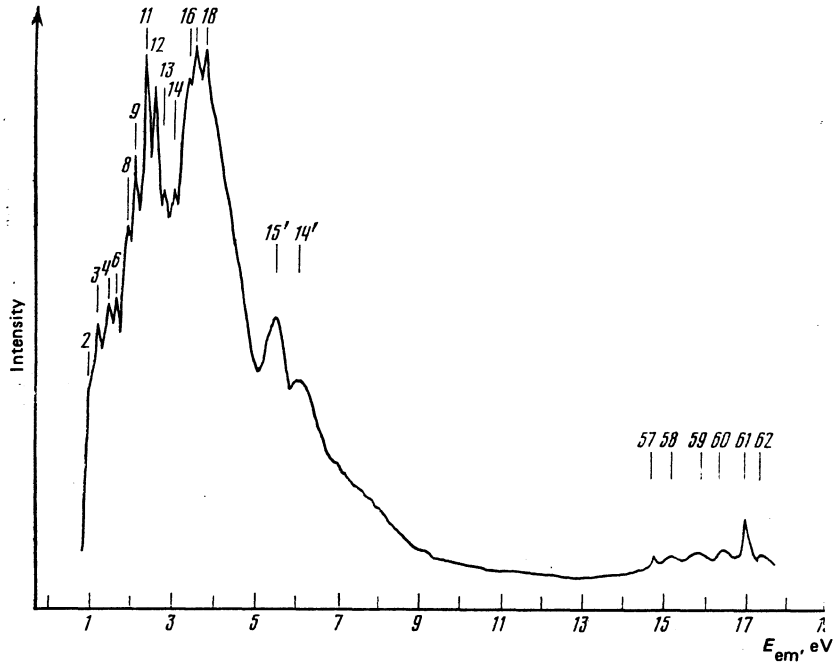

FIG. 4. Spectrum of electrons emitted at an energy 31.3 eV.

lowed us to determine experimentally the thresholds for the appearance of the lines and by the same token check their assumed origin. When the energy of the primary beam was lowered to the threshold of the $^2P_{3/2}$ state of YbII (31, 35 eV), the lines 21–56 drop out completely from the spectrum (see Fig. 4). This indicates that these lines are indeed the result of Auger decay of the indicated state. Figure 4 shows also why we divided the entire energy interval of the emitted electrons into three parts.

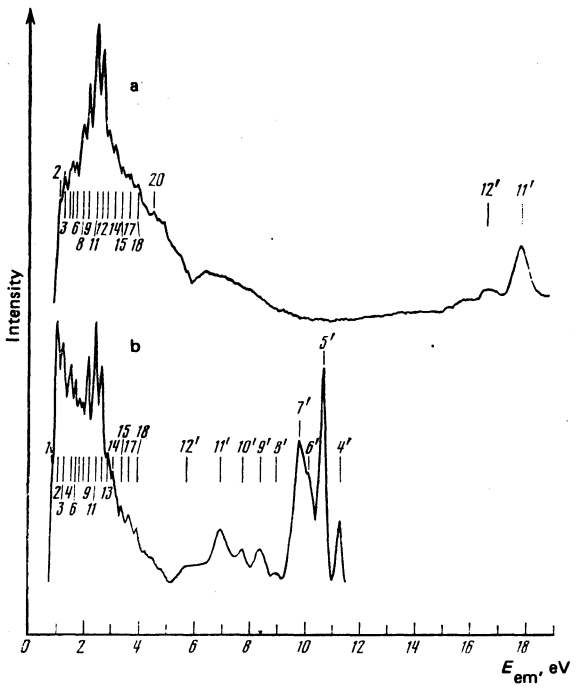

FIG. 5. The same as Fig. 4, but for energies 16 (a) and 15.5 eV (b).

We measured the energy dependences of the intensities of the most typical lines (see Fig. 6). It can be seen that practically all lines have indeed a threshold energy ~31 eV. We note that line 53, classified by the authors of Ref. 8 as decay of the state YBII($^2P_{1/2}$) (E_{em} = 37.52 eV) has in the proposed threshold still a sufficiently high intensity; we are therefore inclined to retain for it the classification of Aleksakhin *et al.*[7] (see Table I).

It might be assumed that the decay of the $^2P_{1/2}$ level of YbII into various final states also contributes to the formation of the structure of the spectra in this region. Thus, lines 43, 46, and 55 can be treated as transitions into the states $4f^{13}(^2F_{3/2})6p_{3/2}(J=2,5)$; $4f^{13}(^2F_{7/2})6p_{1/2}(J=3)$; $4f^{13}(^2F_{7/2})5d_{5/2}(J=1,2)$. However, the transition of YbII($^2P_{1/2}$) into the ground state of YbIII (E_{em} = 18.44 eV) should have been accompanied in this case by emission of an electron with energy 19.08 eV, and this line would apparently be the most intense in this series of transitions, but no such line was observed by us. The line 66, which is closest in energy (E_{em} = 19.02 eV) was reliably recorded by us at energies substantially lower than the excitation threshold of the $^2P_{1/2}$ state. It can therefore be concluded, as indicated also by others,[8] that the autoionization decay of the YbII($^2P_{1/2}$) ion is most ineffective.

The high-energy region of the spectrum is due to the decay of the AIS of the $5p^6$ subshell (see Table I). The variation of the spectrum in this region with changing energy is demonstrated in Fig. 3. When the electron-beam energy tends to the AIS thresholds one can observe the appearance of new lines and an increase in their intensity. We assume this to be due, on the one hand, to excitation of optically forbidden transition, and on the other to population of AIS near the threshold via intermediate states of the short-lived negative YB^- ions. Since we were able to work with beams

TABLE I.

Line No.	E_{em}, eV	Multistep ionization[5]	Photoabsorption[3]
1	0.89	—	
2	1.05	1.08 $5p^64f^{14}7s6p^3P_0$	1.10 $4f^{13}6s6p^2\,(^3/_2, {}^1/_2)_1$
3	1.23	1.24 $5p^64f^{14}7s6p^3P_1$	1.18 $4f^{13}6s6p^2\,(^5/_2, {}^3/_2)_1$
4	1.47	1.50 $5p^64f^{14}7s6p^3P_2$	1.47 $4f^{14}5d5f^1P_1$; $4f^{13}5d6s6d\,[^3/_2]_1$
5	1.53	—	1.55 $4f^{14}5d7p^3D_1$
6	1.66	1.64 $5p^64f^{14}7s6p^1P_1$	1.63 $4f^{14}5d7p^3P_1$; $4f^{13}5d6s6d\,[^1/_2]_1$
7	1.81	—	1.80 $4f^{13}6s^26d\,[^1/_2]_1$
8	1.92	—	1.93 $4f^{13}5d6s7s\,[^3/_2]_1$; $4f^{13}6s6p^2\,(^5/_2, {}^3/_2)$
9	2.12	—	2.11 $4f^{14}5d8p^3D_1$; $4f^{13}5d6s6d\,[^3/_2]_1$
10	2.28	—	2.27 $4f^{13}\,5d6s6d\,[^3/_2]_1$; $4f^{13}5d^26s\,[^3/_2]_1$
11	2.40	—	2.41 $4f^{13}5d6s6d\,[^3/_2]_1$; $4f^{13}5d^26s\,[^3/_2]_1$
12	2.60		2.61 $4f^{13}5d^26s\,[^3/_2]_1$; $4f^{13}5d6s7d\,[^3/_2]_1$
13	2.83	Electron spectroscopy[8]	2.83 $4f^{14}6p8d^3P_1$; $4f^{13}6s^27d\,[^3/_2]_1$
14	3.02	—	3.02 $4f^{13}5d6s10d\,[^1/_2]_1$; $4f^{13}5d6s6d\,[^3/_2]_1$
15	3.27	3.26 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{7/2})\,6p_{3/2}$ $(J = 2,5)$	3.22 $4f^{14}6p15s^3P_1$; $4f^{13}5d6s7s\,[^3/_2]_1$
16	3.37	—	3.38 $4f^{14}6p9d^1P_1$; $4f^{13}5d6s13s\,[^3/_2]_1$
17	3.54	—	3.54 $4f^{13}5d6s9d\,[^3/_2]_1$
18	3.82	—	3.83 $4f^{13}5d6s7s\,[^1/_2]_1$
19	4.00	4.03 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{7/2})\,6p_{1/2}$ $(J = 3)$	3.98 $4f^{13}5d6s6d\,[^3/_2]_1$
20	4.43	—	4.47
21	4.74	4.79	—
22	5.32	—	5.33
23	5.54	—	—
24	5.70	—	—
25	5.96	5.90	—
26	6.17	—	—
27	6.30	6.30 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{5/2})\,5d_{5/2,\,3/2}$ $(J = 4,1)$	—
28	6.54	6.57 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{5/2})\,5d_{5/2}$ $(J = 2)$	—
29	6.67	—	—
30	6.76	—	
31	6.90	—	
32	7.06	—	Electron spectroscopy[7]
33	7.33	7.33 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{5/2})\,6s_{1/2}$ $(J = 2,3)$	7.41 *
34	7.51	7.56 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{7/2})\,5d_{5/2}$ $(J = 3,4,5)$	7.64 *
35	7.59		
36	7.95	7.97 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{7/2})\,5d_{5/2}$ $(J = 1,2,6)$	8.17 *
37	8.03	—	—
38	8.31	—	—
39	8.58	8.57 Yb II $(^2P_{3/2}) \rightarrow 4f^{13}\,(^2F_{7/2})\,5d_{5/2}$ $(J = 3,4)$	8.64 Yb II $(^2P_{3/2}) \rightarrow 5p^64f^{13}6s$
40	8.78	—	—
41	8.90	—	—
42	9.15	—	—
43	9.30	9.26	
44	9.57	—	—
45	9.82	—	—
46	10.09	—	10.14 $5p^5\,(^2P_{3/2})\,5d^2\,(^3P, {}^3F) \rightarrow 5p^64f^{14}\,{}^1S_0$
47	10.41	—	—
48	10.82	10.8	10.86 $5p^5\,(^2P_{3/2})\,5d^2\,(^3P, {}^3F) \rightarrow 5p^64f^{14}\,{}^1S_0$
49	11.42	—	11.23 $5p^5\,(^2P_{3/2})\,5d\,(^3P)\,6s\,(^2P) \rightarrow 5p^64f^{14}\,{}^1S_0$
50	11.64	—	—
51	12.02	—	—
52	12.91	12.92 Yb II $(^2P_{3/2}) \rightarrow 5p^64f^{14}\,{}^1S_0$	12.91 Yb II $(^3P_{3/2}) \rightarrow 5p^64f^{14}\,{}^1S_0$
53	13.47	13.47 Yb II $(^2P_{1/2}) \rightarrow 4f^{13}\,(^2F_{7/2})\,5d_{5/2}$ $(J = 2,3)$	13.56 $5p^5\,(^2P_{3/2})\,5d\,(^3D)\,6s\,(^2D) \rightarrow 5p^64f^{14}\,{}^1S_0$
54	13.92	14.00	—

TABLE I. *(Continued)*

Line No.	E_{em}, eV	Electron spectroscopy[8]	Electron spectroscopy[7]
55	14.14	—	—
56	14.37	—	—
57	14.75	14.73 $5p^5\,(^2P_{3/2})\,6s^27d\,[1/2]_1 \rightarrow 5p^67d^2D_{3/2,\,5/2}$	
58	15.22	—	—
59	15.98	15.98 $5p^5\,(^2P_{3/2})\,6s^26d\,[1/2]_1 \rightarrow 5p^67d^2D_{3/2,\,5/2}$	
60	16.35	16.34	16.24 $5p^5\,(^2P_{3/2})\,5d^2\,[nd+(n+1)\,s]_1 \rightarrow 5p^64f^{14}5d\,(^2D_{3/2})$
61	17.02	—	—
62	17.41	17.39 $5p^5\,(^2P_{3/2})\,6s^25d\,[1/2]_1 \rightarrow 5p^64f^{13}6s^2\,(^2F_{7/2})$	17.35 $5p^5\,(^2P_{3/2})\,5d^2\,[nd+(n+1)\,s]_1 \rightarrow 5p^64f^{14}6p\,(^2P_{3/2})$
63	17.76	—	—
64	18.08	18.09 $5p^5\,(^2P_{3/2})\,6s^25d\,[1/2]_1 \rightarrow 5p^64f^{13}6s^2\,(^2F_{5/2})$	18.06 $5p^5\,(^2P_{3/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^64f^{13}6s^2\,(^2F_{5/2})$
65	18.65	18.61 $5p^5\,(^2P_{3/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^66p\,(^2P_{1/2})$	18.53 $5p^5\,(^2P_{3/2})\,6s^25d\,[1/2]_1 \rightarrow 5p^64f^{14}5d\,(^2D_{3/2})$
66	19.02	—	—
67	19.34	19.32 $5p^5\,(^2P_{3/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^64f^{13}6s^2(^2F_{7/2})$	19.32 $5p^5\,(^2P_{3/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^64f^{13}6s^2\,(^2F_{7/2})$
68	19.80	—	—
69	20.11	—	—
70	21.34	21.30 $5p^5\,(^2P_{3/2})\,6s^25d\,[1/2]_1 \rightarrow 5p^66s^2S_{1/2}$	21.38 $5p^5\,(^2P_{3/2})\,6s^25d\,[1/2]_1 \rightarrow 5p^66s^2S_{1/2}$
71	21.99	21.96 $5p^5\,(^2P_{3/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^66s^2S_{1/2}$	21.98 $5p^5\,(^2P_{3/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^66s^2S_{1/2}$
72	22.88	22.97	—
73	24.28	24.26 $5p^5\,(^2P_{3/2})\,6s^27d\,[1/2]_1 \rightarrow 5p^66s^2S_{1/2}$	—
74	24.73	—	—
75	25.92	—	—
76	27.32	27.33 $5p^5\,(^2P_{1/2})\,6s^25d\,[3/2]_1 \rightarrow 5p^66s^2S_{1/2}$	—

*The energies of these lines are missing from the table of Ref. 7 and were determined from the spectrum given in that reference.

having threshold energies, an attempt could be made to record the post-collisional interaction of the emitted and scattered electrons.[11] We found no clearly pronounced shift of the lines with decreasing energy beam, at least one reason being the absence of a reliable reference at $E < 31$ eV, since the line 52 previously chosen as the reference dropped out of the spectrum at this energy. Nevertheless, an intensity increase and a broadening were observed for line 61 near the threshold, possibly as a result of the interference between the resonant population of the AIS and the post-collisional interaction.[11]

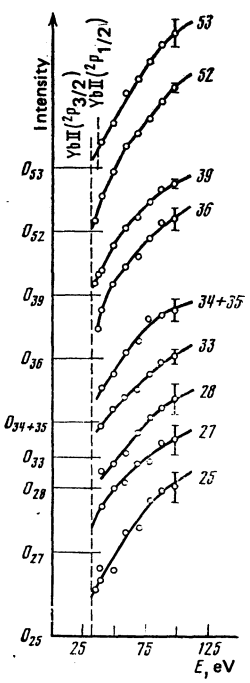

FIG. 6. Energy dependence of the intensities of certain spectral lines. The number at the curve corresponds to the number of the line in Table I.

We proceed now to discuss the low-energy part of the spectrum. It is of interest for several reasons. First, as indicated in Ref. 8, the multichannel decay of the states *pf* of the $5p6$ subshell

$$\mathrm{Yb}^*(5p^5n_1l_1n_2l_2n_3l_3) \rightarrow \mathrm{Yb}^{+*}(5p^5n_1l_1n_2l_2) + e_{em}$$

should be accompanied by formation of electron lines of low energy but having sufficiently high thresholds. The low-energy part of the spectrum was not investigated in Refs. 7 and 8 because of the drop in the transmission of the energy analyzer. We succeeded in observing here a large number of lines, moreover of high intensity. Their origin, however, cannot be attributed to multichannel decay of the AIS of the $5p^6$ subshell into various final states of YbII, since they were recorded in practice up to the beam energies $E = E_{em} + E_i$. We therefore attribute them to decay of the AIS that arise upon excitation of the $4f^{14}$ and $6s^2$ subshells of the atom to the ground state of the ion YbII($5p^54f^{14}6s\ ^2S_{1/2}$).

We find it difficult to approach rigorously the question of classification of these states, since the number of only optically allowed transitions is here tremendous,[3] and contributions can be made here to the structure of the spectrum also by forbidden transitions.

One can note in this region an enhancement of the intensity of certain lines when the beam energy tends to the AIS thresholds. For example, the line 4 more than doubles in intensity near the threshold, despite the simultaneous decrease of the primary-beam current. In other words, a contribution of resonance effects to the population of the AIS

near the threshold is observed in this energy region, too.

Second, interest in this energy region is due to the presence, in the ytterbium photoabsorption spectrum, of a "giant" resonance ($E_a = 33.29$ eV, $\Delta E_{1/2} = 1.4$ eV),[4] located between the thresholds of the $^2P_{3/2}$ and $^2P_{1/2}$ levels of YbII. As stated above, the spectrum of the emitted electrons contains practically no lines corresponding to the decay of the $^2P_{1/2}$ state and, on the contrary, lines due to the decay of the $^2P_{3/2}$ state predominate. This is attributed[8] to the intense nonradiative transition

$$\mathrm{YbI}5p^5(^2P_{1/2})6s^25d[^3/_2]_1 \rightarrow \mathrm{YbII}(^2P_{3/2}). \quad (1)$$

Since no decay of the giant resonance to the ground state of YbII was observed, decay via the indicated channel should be dominant. The transition

$$\mathrm{YbI}5p^5(^2P_{1/2})6s^25d[^3/_2]_1 \rightarrow \mathrm{YbII}5p^66s^2S_{1/2} \quad (2)$$

presupposes emission of an electron with $E_{em} = 27.04$ eV. Although the authors of Ref. 8 treat the line with $E_{em} = 27.33$ eV (see Table I) as a possible decay in accord with the scheme (2), and attribute the energy defect to the large width of the decaying state, it seems to us that their result is quite incorrect. Not being in possession of spectra in the low-energy region, the authors of Ref. 8 indicate that one should expect here a very intense giant autoionization line with $E_{em} = 1.94$ eV, corresponding to the transition (1). Our spectra contains line 8 with energy $E_{em} = 1.92$ eV, but this line is preserved in the spectrum down to very low collision energies, ~8 eV (see Figs. 4 and 5), and therefore cannot be a manifestation of giant autoionization. Nor does the curvilinear background near 2 confirm this, for the same reason.

At $E > 150$ eV the low-energy region is represented by a small number of lines among which one can note lines with $E_{em} = 1.9$ and 3.2 eV and of width ~0.2 eV, but their intensity is many times lower than that of the Auger-transition lines and they can hardly result from giant autoionization.

We investigated in addition the spectra of inelastically scattered electrons, whose lines carry information on the excitation energies of the atomic levels. In the figures presented they are marked by primed numbers. Table II lists the level excitation energy data obtained from the analysis of the spectra of the scattered electrons. An appreciable part of the lines is due to excitation of autoionizing states. Attention is called to a state whose energy exceeds by only 0.3 eV the threshold of single ionization of the atom. One can also note that the same states were observed by us both in the spectra of the emitted and of the scattered electrons (see Table II). In view of the large line widths in the scattered-electron spectra, the accuracy with which the energy was determined amounts for the different states to ± (0.05 to 0.10) eV. It is interesting that we have not observed here any line corresponding to excitation of a giant resonance.

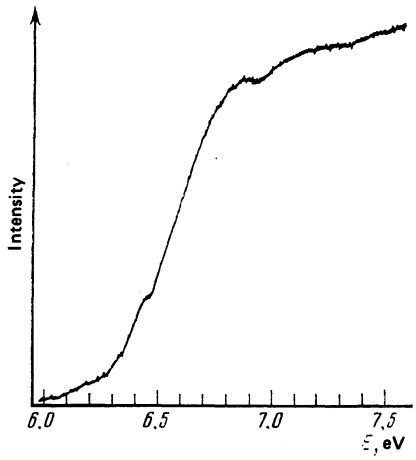

FIG. 7. Energy dependence of the apparent cross section for ytterbium ionization near the threshold.

One other method of investigating autoionizing states is the study of the energy dependences of the atom-ionization cross sections. Using the same spectrometer, we measured the apparent differential cross section for the ionization of the ytterbium atoms in the energy interval from the threshold of the process to 60 eV. A weakly pronounced structure was observed in the cross section only near the single-ionization threshold (see Fig. 7). It is due to the decay of the low-lying AIS. At higher energies, including also the ionization threshold of the $5p^6$ subshell, at the given energy resolution and structural features of the instrument, we were unable to observe a structure in the ionization cross section.

CONCLUSION

The electron spectra of the AIS of ytterbium are characterized by a large number of lines corresponding to the decay of the states of the $6s^2$, $4f^{14}$, and $5p^6$ subshells, as well as of

TABLE II.

N'	E_a	$E_a - E_i$	N	N'	E_a	$E_a - E_i$	N
1	2.26	–	–	10	7.77	1.52	4.5
2	3.11	–	–	11	8.48	2.23	10
3	3.45	–	–	12	9.62	3.37	16
4	4.29	–	–	13	10.25	4.00	19
5	5.02	–	–	14	25.12	18.90	66
6	5.54	–	–	15	25.85	19.60	–
7	5.90	–	–	16	26.69	20.44	–
8	6.58	0.33	–	17	27.60	21.35	70
9	7.11	0.86	1	18	28.20	21.95	71

Note: N' is the line number in the scattered-electron spectra, N is the line number in the emitted-electron spectra.

the state YbII($^2P_{3/2}$). Investigations at an observation angle 90° offer no confirmation whatever of the proposed effective population of the $^2P_{3/2}$ level of the ion on account of radiative decay of a giant resonance.

Despite the sufficiently good agreement between the structures of the spectra obtained by different authors, the nomenclature of the transitions remains in many respects contradictory and calls for further analysis.

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Translated by J. G. Adashko