



International Atomic Energy Agency

INDC(CCP)-219/GA

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INTERNATIONAL NUCLEAR DATA COMMITTEE

PROGRESS IN RESEARCH ON ATOMIC AND IONIC SPECTRA

AND THEIR DEGREE OF INVESTIGATION

UP TO THE PRESENT TIME

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Translated by the IAEA

March 1984

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

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Reproduced by the IAEA in Austria
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84-02805

Key words: Atomic spectroscopy, atomic and ionic spectra, degree of ionization, classification of spectra, energy levels.

ABSTRACT

The paper briefly summarizes the main stages in the investigation of atomic and ionic spectra, the fundamental task of atomic spectroscopy. Detailed data are given on the current state of research and on the degree of investigation of the atomic and ionic spectra for all elements of the periodic system. The highest degree of investigation of the spectra has been achieved in the range from hydrogen to molybdenum as a result of the considerable progress made in research on high-temperature plasmas, where spectra for fifty-fold ionization have been obtained. Analysis of the state of investigation of the spectra shows that 1063 spectra had been studied up to December 1981, which is 19.5% of the total number for the first 104 elements. The data presented here on the state of investigation of spectra will enable the research worker to select those required for plasma diagnostics or to stake out a programme for new spectroscopic studies.

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INTRODUCTION

The fundamental task of atomic spectroscopy is the investigation of atomic and ionic spectra. On the basis of regularities discovered in these spectra it has been possible to construct a model of the atom, to establish energy levels for many atoms and ions and to solve important scientific and practical problems with the help of atomic spectroscopy. Atomic spectroscopy as the science of atomic and ionic spectra is a field which is still far from having been covered exhaustively.

Systematic research on the spectra of the chemical elements began in 1859, when Kirchhoff and Bunsen formulated the well-known law of the uniqueness of the emission-line spectrum for each element. This stimulated the study of atomic spectra and soon led to the discovery of fourteen new elements in Mendeleev's table (Cs, Rb, Tl, In, He, Ga, Ge, Pr, Nd, Sm, Ho, Tm, Yb, Lu). This law served as a starting point for the development of qualitative spectral analysis and, later on, of quantitative analysis based on the dependence of the spectral line intensity on the concentration of a particular element in the sample under examination.

The first attempts to interpret atomic spectra were undertaken by Rydberg. In 1889, he showed that the wave numbers of the spectral lines of some chemical elements could be represented in the form of a difference between two numerical quantities which he called "spectral terms". On the basis of the varying structure of the spectral lines he divided these terms into "sharp", "principal" and "diffuse". The first letters of their English names subsequently came to be used as symbols for the electron shells of atoms as well as for the terms. The physical interpretation of these symbols and a definite notion of atomic energy levels were provided by Bohr in 1913 when he propounded his theory of the atom on the basis of spectroscopic data. He showed that atoms may be in various atomic states with different energies. The spectral lines were due to radiation emitted during the transition of an atom from one state to the next, the frequency of the radiation being proportional to the difference in energy between the two atomic levels. Since that time scientific spectroscopy of atoms and ions, including the classification of spectra and the determination of energy levels, has been further developed.

1. SPECTRAL RESEARCH SINCE THE THEORY OF THE ATOM WAS FORMULATED

It is interesting to see how the study of atomic and ionic spectra progressed after 1913 and what has been achieved by spectroscopic science up to the present day. Our task is made easier by the fact that the eminent American scientist William Meggers published a paper in 1951 containing exhaustive information about the investigation and classification of atomic and ionic spectra in the period 1913-51 [1]. The paper showed that 231 spectra of 69 chemical elements had been studied by 1932, when the energy levels obtained from spectral analysis were published in a well-known book by Bacher and Coudsmit [2]. At this time, four of the 92 elements between H and U in Mendeleev's table remained to be discovered (Tc, Pm, At, Fr). Of the known 88 elements, the spectra of 19 had not yet been studied. Since each chemical element can produce as many different spectra as it has electrons in its neutral atom shell, the number of possible spectra at that time was 4002. The degree of investigation of the atomic and ionic spectra may be characterized by the ratio of the number of spectra studied to the total number of elements known. Thus, in 1932 this ratio was $231/4002 = 5.8\%$.

By 1951 a further 273 spectra had been studied. The total number then was 504 for 84 elements. The atom and ion energy levels were collected in a three-volume work by Moore [3]. At this time 98 chemical elements were known, including californium. However, the spectra of 14 elements had still not been examined (Pm, Ho, Er, Po, At, Fr, Ac, Pa, Np, Pu, Am, Cm, Bk, Cf). The number of possible spectra of the 98 elements was 4851. Hence the ratio in 1951 was $504/4851 = 10.4\%$.

During the next 30 years (1951 to December 1981), scientists in many countries all over the world made great efforts to study atomic and ionic spectra, assisted by major advances in the development of spectroscopy techniques which were achieved because problems arose in the solution of which atomic spectroscopy was found to be valuable. Such advances included spectral elementary and isotopic analysis of products and materials used in atomic technology; investigation of the complex spectra of the actinide and rare-earth (lanthanide) elements and of their hyperfine and isotopic structure; and use of spectroscopic diagnostic methods to determine the main parameters of low- and high-temperature plasmas.

Consequently, a situation arose in which new development possibilities opened up for atomic spectroscopy itself. With the help of nuclear reactors, new chemical elements were discovered and their quantities by weight determined. The study of the optical spectra of these elements, their classification and the determination of the energy levels of the neutral atoms and of the first ions was undertaken. In devices for the investigation of high-temperature plasmas spectra were obtained for multiply charged ions.

From 1967 onwards much effort was devoted to studying the spectra of highly ionized atoms. A substantial contribution came from Soviet spectroscopists. At the Spectroscopy Institute of the USSR Academy of Sciences, under the leadership of Eh.Ya. Kononov, 140 ion spectra of 26 elements between aluminium and tin have been studied during the last 15 years. The wavelengths of the spectral lines have been measured with high accuracy and the lines have been classified and the energy levels determined. Considerable research has been carried out at the P.N. Lebedev Physical Institute of the USSR Academy of Sciences under the leadership of V.A. Bojko: experiments have been performed to study the spectra of hydrogen-like, helium-like and lithium-like ions over the range of elements from sodium to caesium, as well as ion spectra of other elements. Altogether about 50 spectra have been studied in the X-ray range from 20 to 2 Å.

The degree of investigation of the atomic and ionic spectra for the last 30 years we evaluated on the basis of articles published in journals and collected in the bibliographical handbooks of the United States National Bureau of Standards [4-7]. In addition, we used generalized data from selected tables on the atomic spectra of certain elements [8], new books on the classified spectral lines of atoms and ions [9-13], on the energy levels of the rare-earth elements [14] and on Grotrian diagrams [15], reference data on the same topics [16-32], calculated data on the spectra and levels of hydrogen-like ions [33,34] and collections of papers on ionization potentials [35,36]. Two review papers published in 1981 should also be included. One of these discusses the classification of highly ionized atomic spectra over the last 7 years [37], presenting the results of observations of the sun's radiation spectra from space and also spectra obtained on various types of high-temperature plasma device. The other paper is concerned with the development of highly ionized atom spectroscopy and its use for plasma diagnostics [38].

Besides these publications our list of references also cites some papers [39-105] on atomic and ionic spectrum research which were published later than July 1979 and are therefore not included in the NBS bibliographic handbooks. These papers enabled us to take our review of the state of investigation up to December 1981. According to rough estimates some 3000 original papers have been published during the last 30 years on the investigation of atomic and ionic spectra and energy levels.

2. STATE OF INVESTIGATION OF SPECTRA AT THE PRESENT TIME

The situation as regards our present knowledge of the spectra of atoms and ions is presented in Table 1. The first column successively gives the symbols of the 104 elements from hydrogen to kurchatovium. Each row (line) of the table indicates the degree of experimental investigation of all spectra of the element which have been studied, beginning with the neutral atom and continuing with the ions in order of increasing degree of ionization. Meggers's scale [1] of degrees of investigation was used for the sake of comparability. The letter A indicates thoroughly studied spectra with a reasonably complete system of energy levels and with an established link between levels of different multiplicity. The letter B indicates that the spectrum has been fairly thoroughly studied and that many levels of different multiplicity have been found. The letter C indicates an intermediate degree of investigation: up to 20 levels found, but the connection between levels of different multiplicity not established. The letter D indicates that up to 10 levels have been found, and E means that preliminary studies have been undertaken, the wavelength of the lines being measured, the degree of ionization established, and one or several transitions given. Lower-case a, b, c, d, e indicate the degree of investigation for 1951. This means that these spectra have not been studied more recently.

Table 1 shows that the spectra of the first 20 elements from hydrogen to calcium have been studied in some depth for all degrees of ionization. Further down, gaps appear. The hydrogen-like spectra of Sc, V, Mn, Co, Ni and the subsequent elements, except Mo, have not so far been studied experimentally. Calculated data are available for the spectra of these single-electron ions [34]. From copper to molybdenum many spectra have not been studied at all or only to a slight degree (E). Beyond molybdenum

the degree of investigation declines sharply. In this region only the spectra of the neutral atoms and of some ions have been studied. Those spectra for higher degrees of ionization which have been studied can be seen in Table 1 forming isoelectronic series clustering round the sloping lines for Li I, Na I, K I, Cu I, Rb I, Ag I, Er I and Au I. The studied spectra of multiply charged ions neighbouring on the isoelectronic series are arranged near these lines. The highest degrees of ionization (45-52) are obtained for heavy elements in the region Hf-Au for the case of nickel-, copper- and zinc-like ions.

Table 2 presents data characterizing the degree of investigation of spectra for seven groups of elements and for all the elements together. The table shows that as the atomic number Z of the elements increases, the degree of investigation of the spectra diminishes because the ion spectra have been insufficiently studied. This is due to the experimental difficulty of exciting and observing multiply charged ions, as the resonance lines of ions with degrees of ionization exceeding 10 and ionization potentials of more than 1 keV fall in the X-ray region. The high degree of investigation of the first three groups of elements is due to the spreading of research on high-temperature plasmas, which has made it possible to study the spectra of multiply charged ions with Z between 10 and 42. The degrees of investigation ratio for the first three groups are 100, 94.9 and 55.5% respectively. For the further groups this ratio falls to 19.6, 11.0 and 8.1%, and for the radioactive elements (Po-Ku) it is only 1.3%. In the latter region only 53 of the total of 1974 spectra have been studied. The last row of Table 2 shows that all the currently known chemical elements together have 5460 spectra, of which 1063 have been studied. Thus, the degree of investigation of the spectra for all elements is 19.5%. Only half of these spectra have been studied in depth ($A + B = 47\%$), the rest require further investigation. One fifth are in group E (17%), i.e. research on these is only just beginning.

This paper on the degree of investigation of atomic and ionic spectra was prepared mostly in mid-1981. In December we became aware of the paper by Cowan [38], whose tables agreed with ours in almost every respect. Only a few evaluations of the degree of investigation of spectra differ, apparently because of some difference in the criteria used, or perhaps even as a result of subjective evaluation. According to Ref. [38], 1019 spectra

had been studied by March 1981. The present paper covers the published literature up to December 1981 inclusive.

CONCLUSION

Using the data of Ref. [38] and the degree of investigation of 1063 spectra established for December 1981, we can trace in greater detail the development of spectral research from 1913 to the present time. Table 3 shows the change in time of the degree of investigation of the spectra. The first column indicates the year of the evaluation, the second the aggregate number studied up to that year, and the third and fourth columns give the number of spectra studied during the period in question and the number studied annually during that period.

No doubt the first increase in spectral research during 1922-39 was due to the development of Bohr's theory of complex atoms, whereas the second peak was a result of progress in the investigation of multiply charged ion spectra (1968-81). The low point corresponds to the war and post-war years. The slight increase from 1946 to 1951 is due to the investigation of spectra of the newly discovered actinides and other radioactive elements.

In conclusion, we would like to stress that considerable attention is currently being paid to research on atomic and ionic spectra and that significant progress has been made in the last decade. By 1982 the degree of investigation ratio for all elements was 19.5% as against 10.4% in 1951. If theoretical calculations for the hydrogen-like atoms are included [34], then the spectra of the first 28 elements (H-Ni) have been studied for all degrees of ionization. The degree of investigation of the next group of elements from Cu to Mo still remains inadequate at 60.8%. Here 195 of a total of 487 spectra have not been studied, and for 68 spectra only a few classified lines have been established. No doubt these spectra will be of great importance in the investigation of high-temperature plasmas. For this reason the efforts of spectroscopists studying ion spectra will in the near future be directed towards solving this problem.

TABLE 1

Degree of investigation of spectra and energy levels
up to December 1981

- Note: A = Spectrum classified and full system of levels given;
B = Up to 30 levels of different multiplicity found;
C = Up to 20 levels found, connections of different multiplicity
not established;
D = Up to 10 levels found;
E = one or more transitions established.

Upper-case letters: degree of study up to January 1982; lower-case = up to 1951.

TABLE 2

Degree of investigation of spectra for various groups of elements

Range of elements	Number of elements	Total spectra	For 1951-81		Total studied up to December 1981	Degree of investigation			Number of spectra not studied	Relative degree of investigation, %
			Newly studied	Degree of investigation improved		A and B	C and D	E		
H-Ar	18	171	30	77	171	138	33	-	None	100
K-Zn	12	294	137	117	279	144	130	5	15	94.9
Ga-Mo	12	438	161	43	243	82	99	62	195	55.5
Tc-Ba	14	693	70	22	136	41	51	44	557	19.6
La-Lu	15	960	78	23	106	42	28	36	854	11.0
Hf-Bi	12	930	42	20	75	38	17	20	855	8.1
Po-Ku	21	1974	44	5	53	12	26	15	1921	1.3
For all spectra	104	5460	562	307	1063	497	387	182	4397	19.5

TABLE 3

Degree of investigation of spectra for various time periods

Years	Total number of spectra studied	Number studied during period	Number of spectra studied annually
1913	0	0	0
1922	38	38	4.2
1932	231	193	19.3
1939	400	169	24.1
1946	445	45	6.4
1951	504	59	11.8
1959	511	7	0.9
1969	648	137	13.7
1981	1063	415	34.6

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