Isotopic Constitution of Tellurium, Silicon, Tungsten, Molybdenum, and Bromine*

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(Received November 20, 1945)

The relative abundances of the isotopes of tellurium, silicon, tungsten, molybdenum, and bromine have been re-determined by means of a mass spectrograph of the Nier type. The results are as follows: *Tellurium.* Samples in the form of tellurium hexafluoride were employed and the ions TeF_5^+ and TeF_3^+ were used in measuring relative abundances.

Isotope	130	128	126	125	124	123	122	120
Relative abundance	100	92.3	54.2	20.2	13.3	2.47	7.05	0.26.

The existence of isotope 120 was confirmed. With the assumption of a packing fraction of -4, the above values for relative abundance lead to a value of 127.64 ± 0.01 for the chemical atomic weight. *Silicon.* Samples in the form of silicon tetrafluoride were employed and the ions SiF_4^+ and SiF_3^+ were used. The results are as follows:

Isotope	28	29	30
Isotopic abundance	100	5.07	3.31.

With Aston's values for the isotopic weights, the above values for isotopic abundances give 28.086 for the chemical atomic weight. *Tungsten.* The isotopic constitution of tungsten has been re-investigated with a view to obtaining more exact data on relative abundance. Relative isotopic abundances were measured for the W^+ ion obtained by electronic bombardment of tungsten hexacarbonyl vapor. The existence of isotope 180 has been confirmed. The estimates of isotopic abundances obtained by electro-metrical methods are:

Isotopic	186	184	183	182	180
Relative abundance	92.7	100	47.0	86.2	0.44.

On the basis of a packing fraction of $+2$, these estimates lead to a value of 183.88 for the chemical atomic weight. *Molybdenum.* The isotopic constitution of molybdenum has been re-investigated. The values for isotopic abundance as determined electro-metrically from Mo^+ and Mo^{++} ions produced by electronic bombardment of molybdenum hexacarbonyl are:

Mass number	100	98	97	96	95	94	92
Relative abundance	40.5	100.0	39.8	69.5	66.1	38.4	66.8.

These values are in closer agreement with values reported by Valley than with values obtained by Aston or Mattauch. *Bromine.* Values for the relative abundance of the bromine isotopes have been obtained from measurements on Br_2^+, Br^+, and Br^{++} ions formed by electronic bombardment of bromine vapor. The relative abundances are in essential agreement with the values reported by Blewett:

Mass number	79	81
Relative abundance (present study)	100	97.9 ± 0.4
Relative abundance (Blewett)	100	97.5 ± 2.5.

No new isotopes of bromine were observed.

THE development of nuclear physics has made necessary a more exact knowledge of the isotopic constitution of the elements. Most of the early work in the field of mass spectroscopy was concerned primarily with the detection of isotopes and the accurate measurement of masses and only incidentally with the accurate determination of isotopic abundances.[1] With the exception of the apparatus described by Dempster[2] the instruments used employed photographic methods of detection. Relative abundances were based on photometric measurements.

During recent years several instruments have been designed with a view to obtaining more accurate information on relative abundance. These spectrometers employ electrical methods of detection and measurement.[3-5] With these instruments the accuracy of measurement of normal ratios of abundance is approximately one percent, and data on numerous elements have been reported by Nier, Bleakney, Valley, and others.[6]

The work reported in the present papér is concerned chiefly with data on the relative abundance of the isotopes of several elements not previously investigated by methods utilizing electrical methods of measurement.

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* This article has been prepared by the Editor from four reports which were submitted by the authors after clearance by the War Department.

[1] F. W. Aston, *Mass Spectra and Isotopes* (Edward Arnold and Company, London, 1942), Part II.

[2] A. J. Dempster, Phys. Rev. **20**, 631 (1922).

[3] M. B. Sampson and W. Bleakney, Phys. Rev. **50**, 456 (1936).

[4] A. O. Nier, Phys. Rev. **52**, 933 (1937).

[5] A. O. Nier, Rev. Sci. Inst. **11**, 212 (1940).

[6] F. W. Aston, Summary of Work in *Mass Spectra and Isotopes* (Edward Arnold and Company, London, 1942), Part III.

APPARATUS AND PROCEDURE

The spectrometer used in the present study was designed by Nier and was similar in most essentials to the one mentioned above.[5] The general principles on which the operation of this type of instrument is based have been discussed by Barber[7] and Stephens and Hughes.[8] These authors have shown that if a slightly divergent beam of ions having the same momentum enters a homogeneous magnetic field between two V-shaped pole pieces in a direction normal to one boundary of the field and after deflection leaves the field in a direction normal to the other boundary of the field, re-focusing of the ion paths will occur. The nature of the re-focusing is such that the source of the ions, the apex of the V, and the point of re-focusing lie on a straight line. In the present spectrometer the beam of ions entering the magnetic field from the entrance slit is mono-energetic; hence, the ions of a given mass can be focused at the exit slit by giving the appropriate energy to the ions entering the field, provided the proper geometry connecting entrance slit, the apex of the V, and the exit slit is realized. The radius of curvature of the collected ion beam and the mass of the ion are given by the following familiar relationship:

$$m/e = 4.82 \times 10^{-5} r^2 H^2 / V, \tag{1}$$

where m/e is in atomic mass units/electronic charge, r is the radius in cm, H is the magnetic field intensity in gauss, and V is the energy of the ions in electron volts. In operating the instrument, H is held constant at some appropriate value and the various isotopes are brought into focus by adjusting V.

The potentials used for accelerating the ions were obtained from a potentiometer connected to a 2000-volt electronically regulated power supply. In most of the present work the energies of the ions were between 1000 and 2000 electron volts.

The magnet used in the spectrometer was a large electromagnet with wedge-shaped 60° pole pieces. The separation of the pole pieces was $\frac{9}{16}''$. The current for the coils of the magnet was supplied from a constant-current power supply with

electronic regulation. The radius of the circle described by the collected ion beam in the magnetic field was 15 cm.

The collector for the ion current was located behind the exit slit of the spectrometer. A small negative potential applied to a diaphragm near the collector served to minimize the effects of secondary electron emission. The collector was connected to the grid of an FP-54 electrometer tube in a conventional DuBridge-Brown Circuit employing a grid resistance of 40,000 megohms. A high sensitivity galvanometer equipped with an Ayrton shunt was used with the amplifier. With a scale distance of two meters the amplifier-galvanometer system had a maximum sensitivity of 3×10^{-16} amp./mm. When the amplifier tube housing was evacuated with a forepump, the stability of the amplifier used at maximum sensitivity was usually adequate. Potentials from a Leeds and Northrup potentiometer could be applied in the grid circuit of the amplifier for purposes of calibration.

A two-stage mercury diffusion pump and a Welch Duo-Seal forepump were used in evacuating the spectrometer tube. With liquid nitrogen as a trap refrigerant, this combination was capable of producing a vacuum of 10^{-8} mm of mercury as measured by a Distillation Products ion gauge. Under normal operating conditions the pressure was not allowed to rise above 2×10^{-7} mm of mercury.

During operation a volatile compound of the element being investigated was admitted to the spectrometer source through a variable leak from a separate system. Ions were produced by electronic bombardment of the gas essentially in the manner described by Nier[5] except that the required voltages were obtained from a vacuum tube power supply.

In making relative abundance measurements on two isotopes A and B, galvanometer deflections produced by the two ion beams were measured. Deflections in the order AB, BA, AB, etc., were observed. After completion of these observations, the observed galvanometer deflections were reproduced by the application of known voltages from the potentiometer. These voltages were then compared to measure isotopic abundance. The chief advantage of this method of measuring abundance over the null method,

[7] N. F. Barber, Proc. Leeds Phil. Soc. 2, 427 (1933).
[8] W. E. Stephens and A. L. Hughes, Phys. Rev. 45, 123 (1934); W. E. Stephens, Phys. Rev. 45, 513 (1934).
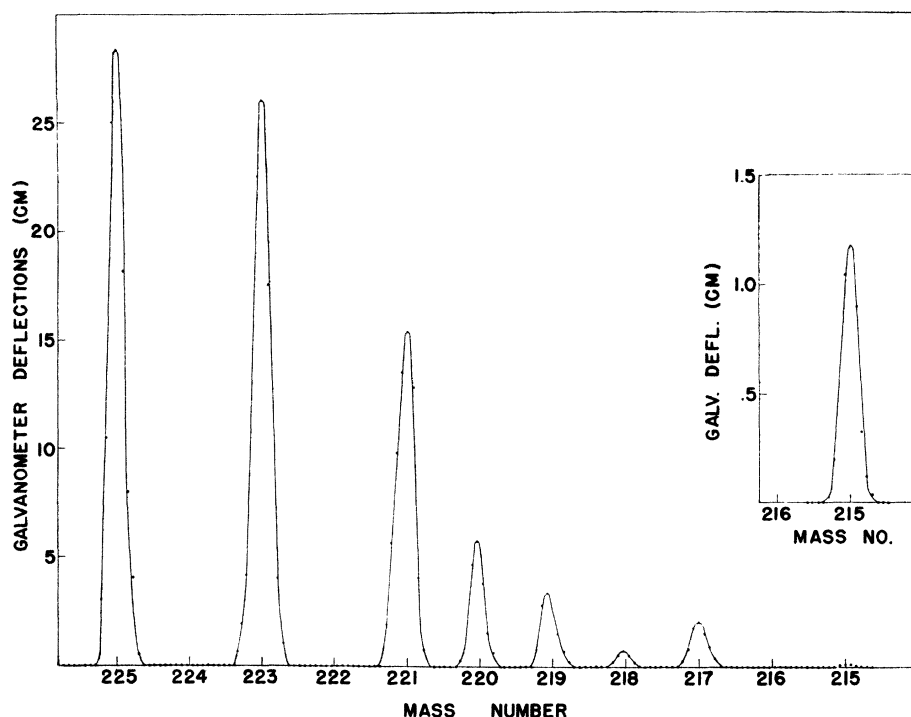
FIG. 1. Mass spectrum in the vicinity of the TeF_5^+ ion. The peak at mass position 225 corresponds to a beam current of 1.5×10^{-11} ampere.

in which the voltage produced in the grid resistor by the collected ion beam is balanced out by applied voltages from a potentiometer, lies in the fact that in the shorter time required for making observations the ion currents do not change as much as in the longer observation times required for null-method measurements. The chief disadvantage lies in the possibility of variations in grid resistance produced by large ion currents. Tests show that for ion currents of the magnitude actually used the grid resistance remains constant to within less than one percent. Experience with the present instrument seems to indicate that the deflection method of measurement when combined with immediate calibration gives more reliable and more reproducible results than the null method.

TELLURIUM

The isotopic constitution of tellurium has long been of interest on account of the anomalous position of its atomic weight in reference to that of iodine in the periodic table. Previous work by

Aston,[9] Bainbridge,[10] Dempster,[11] and Bainbridge and Jordan[12] led to values of the abundance ratio of the isotopes presented in the last column of Table III.

The material used in the present work was tellurium hexafluoride. Samples were prepared in a vacuum system by a method involving a reaction between powdered tellurium and anhydrous cobaltic fluoride. Owing to the simple isotopic constitution of fluorine, results obtained with any of the ions formed are immediately

TABLE I. Relative abundance of ions formed by bombardment of tellurium hexafluoride with 100-volt electrons.

Ion	Relative abundance	Ion	Relative abundance
TeF_6^+	weak	TeF_2^+	6.3
TeF_5^+	100.0	TeF^+	9.1
TeF_4^+	5.5	Te^+	12.7
TeF_3^+	9.3		

[9] F. W. Aston, Phil. Mag. 42, 241 (1921); 45, 942 (1923); 49, 1197 (1925); Proc. Roy. Soc. A132, 487 (1931).
[10] K. T. Bainbridge, Phys. Rev. 39, 1021 (1932).
[11] A. J. Dempster, Phys. Rev. 50, 186 (1936).
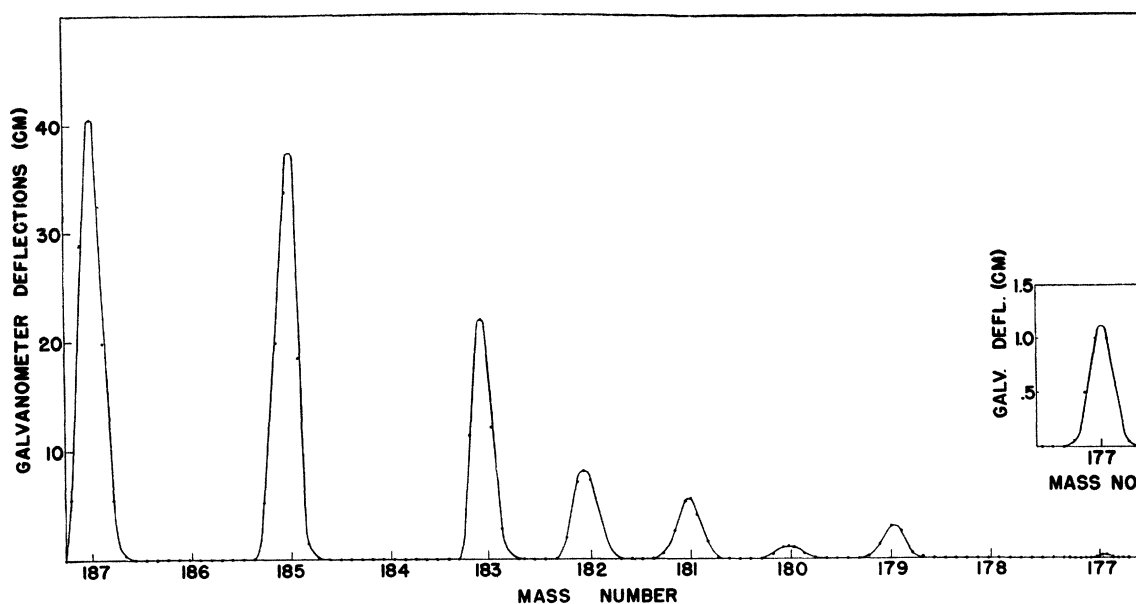[12] K. T. Bainbridge and E. B. Jordon, Phys. Rev. 50, 282 (1936).

FIG. 2. Mass spectrum in the vicinity of the TeF_3^+ ion. The peak at mass position 187 corresponds to a beam current of 1.5×10^{-12} ampere.

interpretable. The relative abundances of the ions produced by electronic bombardment are given in Table I. The estimates given in the table are probably accurate to ten percent.

Owing to the large relative abundance of the TeF_5^+ and TeF_3^+ ions formed, these ions were chosen for study in measuring isotopic abundances. The mass spectrum observed in the vicinity of the TeF_5^+ ion is shown in Fig. 1. In obtaining the data represented in this figure the exit slit was made just wide enough to receive

TABLE II. Measured isotopic abundance of tellurium isotopes. Relative abundance measured from indicated ion (all measurements in terms of 130 except as indicated).

Isotope	TeF_5^+ I	TeF_5^+ II	TeF_3^+	Average
130	100	100	100	100
128	92.5	92.5	92.0	92.3
126	54.5	54.2	53.8	54.2
125	20.5	20.15	19.8	20.15
124	13.6	13.4	13.0	13.3
123	2.52	2.51	2.38	2.47
122	7.25	7.14	6.77	7.05
120	0.261	—	—	0.261
124*	13.3	13.4	13.2	13.3
123*	2.47	2.51	2.42	2.47
122*	7.12	7.14	6.89	7.05
120*	0.256	—	—	0.256

* Using 125/130 =20.15 and taking ratios to 125, the above abundances were obtained.

the total ion beam for the peaks shown. The indicated resolution obtained is adequate for making abundance measurements. The inset shows the peak observed at mass number 215 where the amplifier was operated at highest sensitivity. No peaks were observed at 226, 224, 222, or 216.

In Fig. 2 are shown the results obtained for the TeF_3^+ ion. Here again the resolution seems to be adequate for making dependable abundance estimates. The inset in this figure shows the 177 peak as observed with the amplifier operating at highest sensitivity.

In Table II are given the relative abundances of the tellurium isotopes as measured from the peaks observed for the ions TeF_5^+ and TeF_3^+. Each value in the table represents the average of at least four ratios. Two complete sets of values are given for TeF_5^+; these two sets were obtained from data taken under different conditions of pressure, collector slit width, etc. The values obtained from these measurements on TeF_5^+ are in fair agreement with each other and with the measurements on TeF_3^+. As indicated in the table, most of the figures on abundance were obtained by direct comparison of each peak with the peak produced by the most abundant isotope

130. However, in order to obtain an additional check on the rarer isotopes 124, 123, 122, and 120, their abundances were re-measured in terms of 125. The results of these measurements are given in the lower part of Table II. The agreement between these values and the ones obtained by direct comparison with 130 is satisfactory. A search for other isotopes was made. The upper limit for abundances of 131, 129, 127, and 121 relative to 130 can be set at 1/20,000.

The final estimates of abundance based on the measurements described above are summarized in Table III for comparison with the values obtained by Bainbridge from a comparison of his own measurements on 124, 123, and 122 with Aston's earlier estimates on the more abundant isotopes. The uncertainties indicated for the values obtained in the present study are based on variations in the individual determinations and do not include systematic errors arising from changes in the grid resistance of the electrometer tube circuit, which may be as large as one percent.

The values in the column headed "Atomic percentage" imply an accuracy greater than justified by the data and were used only in computing the atomic weights. In this computation a packing fraction of -4 was assumed for all isotopes. The resulting value for the chemical atomic weight is 127.64 ± 0.01. The uncertainty indicated includes only the uncertainties in the abundance measurements and not uncertainties in packing fraction. The value obtained from the present work is slightly larger than the accepted value for the International Atomic Weight 127.61.

TABLE III. Relative abundance of the isotopes of tellurium.

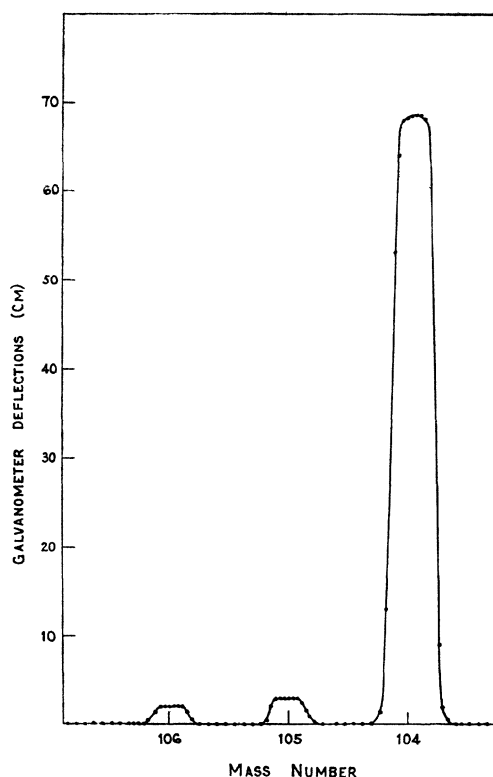| Isotope	Present study		Aston and Bainbridge values
	Relative abundance	Atomic percentage	Atomic percentage
130	100.00	34.51%	33.1
128	92.3±0.3	31.85	32.8
126	54.2±0.2	18.70	19.0
125	20.2±0.2	6.98	6.0
124	13.3±0.4	4.59	4.5
123	2.47±0.05	0.85	1.6
122	7.05±0.18	2.43	2.9
120	0.256±0.017	0.09	W

FIG. 3. Mass spectrum in the vicinity of the SiF_4^+ ion. The peak at mass position 104 corresponds to a beam current of 5×10^{-12} ampere.

SILICON*

Aston[13] was the first to study the isotopic composition of silicon. Much valuable information on the isotopic constitution of silicon has also been obtained from studies of the band spectrum of silicon nitride. Mulliken[14] confirmed the existence of isotope 30. Later McKellar[15] made an estimate of relative abundances of silicon isotopes from photometric measurements of the violet bands of silicon nitride. The atomic percentages obtained from these measurements are given in the last column of Table V.

13 F. W. Aston, Phil. Mag. 40, 628 (1920); 49, 1198 (1925); Proc. Roy. Soc. A132, 491 (1931); Nature 137, 613 (1936); 138, 1094 (1936).
14 R. S. Mulliken, Nature 113, 423, 489 (1924).
15 A. McKellar, Phys. Rev. 45, 761 (1934).

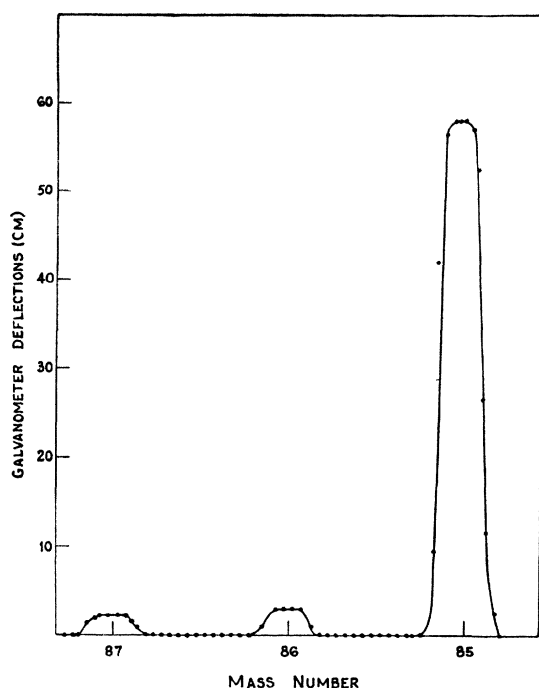F IG. 4. Mass spectrum in the vicinity of the $SiF_3{}^+$ ion. The peak at mass position 85 corresponds to a beam current of 7.5×10^{-12} ampere.

The material used in the present study was silicon tetrafluoride and was prepared under vacuum by a reaction between elementary silicon and anhydrous cobaltic fluoride. When the silicon tetrafluoride vapor was subjected to electronic bombardment, peaks corresponding to ions of the types $SiF_4{}^+$, $SiF_3{}^+$, $SiF_2{}^+$, SiF^+, and Si^+

TABLE IV. Measured isotopic abundance of silicon isotopes.

	Relative abundance measured from indicated ion			
Isotope	$SiF_4{}^+$	$SiF_3{}^+$ I	$SiF_3{}^+$ II	Average
28	100	100	100	100
29	5.10	5.07	5.05	5.07
30	3.32	3.30	3.31	3.31

TABLE V. Relative abundance of silicon isotopes.

	Present study		McKellar's value
Isotope	Relative abundance	Atomic percentage	Atomic percentage
28	100	92.27	89.6
29	5.07±0.02	4.68	6.2
20	3.31±0.01	3.05	4.2

were observed. However, most of the actual measurements were made on $SiF_4{}^+$ and $SiF_3{}^+$. These ions were the most abundant, and the resolution obtained for them was satisfactory.

In Fig. 3 is shown the mass spectrum obtained in the vicinity of $SiF_4{}^+$ ion. In taking the data represented in this figure, the collector slit was wide enough to receive the total ion current for each peak. The peaks are well resolved. Examination of this region before and after the sample was studied showed no evidence of background at any place in the region.

Figure 4 shows the mass spectrum obtained in the vicinity of the $SiF_3{}^+$ ion. Here, again, resolution is adequate and there are no evidences of impurities. The $SiF_3{}^+$ ion was the most abundant ion formed under the conditions under which the source was operated.

In Table IV are given the results obtained on isotopic abundance in three independent sets of measurements. The first set of measurements was made on the $SiF_4{}^+$ ion and the other two were made on the $SiF_3{}^+$ ion under different conditions of pressure and source "tuning." The agreement between the values obtained is fair. A search for other isotopes was also made; none was found. The upper limits of abundances of 27 and 31 relative to 28 can be set at 1/20,000.

In Table V the values for isotopic abundances obtained in this study are summarized. The indicated uncertainties for relative abundances are based on the actual spread in the values obtained from the present measurements. There is also a possibility of systematic errors amounting to as much as one percent of the values listed. The source of this possible error has been discussed above. In the last column of Table V McKellar's values for atomic percentage are listed for comparison with the present values. Agreement between the two sets is poor.

In calculating the chemical atomic weight from the present data, Aston's values of 27.9869 and 28.9864 were used for the two lighter isotopes. A value of 29.9862 was assumed for the third isotope. With these values for the masses, the present values for abundance give a value of 28.086 for the chemical atomic weight. This value is in fair agreement with the international value of 28.06.
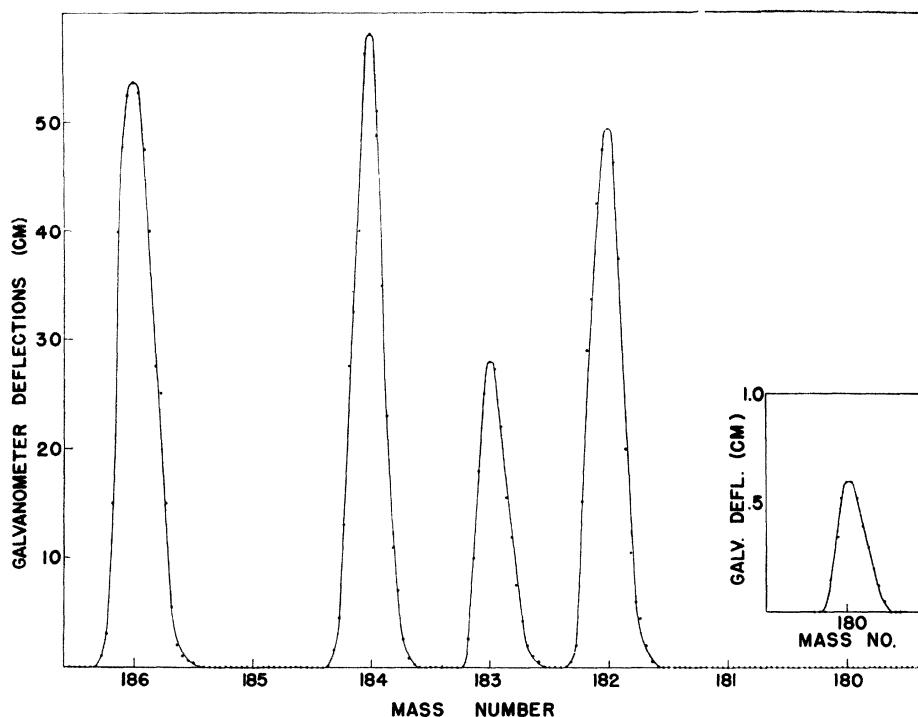
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Fig. 5. Mass spectrum in the vicinity of the W^+ ion. The peak at mass position 184 corresponds to a beam current of 7.5×10^{-12} ampere.

TUNGSTEN

Early measurements of the abundance ratio of the isotopes of tungsten were by Aston[16] and by Dempster.[17] The values they obtained are shown in the last column of Table VI. The material used was tungsten hexacarbonyl, which in spite of its low vapor pressure gave satisfactory results.

Tungsten hexacarbonyl undergoes thermal decomposition into metallic tungsten and carbon monoxide at approximately 400°C. Hence, it was not surprising that the most abundant ion formed during electronic bombardment at 100 volts was the metallic ion. Rough estimates indicate that more than half of the positive ions formed were singly charged tungsten ions. However, other species of ions representing all stages of dissociation from $W(CO)_6^+$ to W^+ were observed. As there was evidence that the CO groups themselves decomposed, it was impossible to obtain interpretable data on any singly

charged ions except W^+ because of overlapping. It was also impossible to obtain interpretable data on W^{++} because these peaks were overlapped by peaks produced by multiply charged ions of other species.

The mass spectrum observed in the vicinity of the singly charged tungsten ion is shown in Fig. 5. The peaks shown in this figure are sufficiently well resolved to permit accurate determination of isotopic abundances. There is no indication of background arising from impurities. The inset at mass position 180 shows this peak as observed with the amplifier operating at highest sensitivity. No minor peaks of com-

[16] F. W. Aston, Proc. Roy. Soc. A115, 507 (1927); A132, 491 (1931).
[17] A. J. Dempster, Phys. Rev. 52, 1074 (1937).

TABLE VI. Relative abundance of tungsten isotopes as determined from tungsten caronyl.

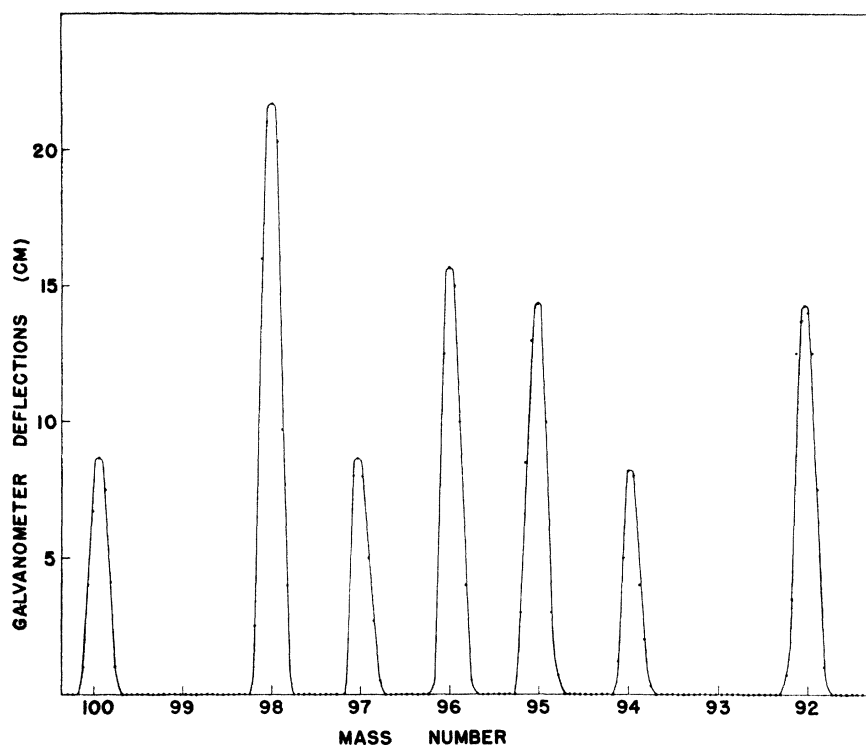| Isotope | Measured abundance from W^+ ion under different conditions of pressure and source "tuning" | | | Atomic percentage | Aston-Dempster values |
	W^+ I	W^+ II	Average		
186	92.9	92.6	92.7	28.41	29.8
184	100.0	100.0	100.0	30.64	30.1
183	47.0	47.1	47.0	14.40	17.3
182	86.2	86.2	86.2	26.41	22.6
180	0.438	0.443	0.440	0.13	0.2

FIG. 6. Mass spectrum in the vicinity of the Mo⁺ ion. The peak at mass position 98 corresponds to a beam current of 1×10^{-11} ampere.

parable magnitude were observed at other mass positions.

The results obtained on isotopic abundance are given in Table VI. Data are shown for two different conditions of pressure and spectrometer "tuning." Agreement between the two sets of data is fair. However, it would have been desirable to obtain data on another ion for purposes of comparison. It is believed that the values for isotopic abundance are accurate to ±2 percent. In the last column are shown the atomic percentages of the various isotopes as determined by Aston and Dempster. Agreement is good for isotopes 186 and 184 and rather poor for 183 and 182. In the case of the rare isotope 180, there is excellent agreement between the present results and Dempster's original estimate of its abundance as one percent of that of 183.

A search was made for isotopes of mass 179, 181, 185, and 187. No peaks were observed at these mass positions, although peaks of 1/10,000 that at mass position 184 could have been detected.

The packing fraction for tungsten has been measured by Aston,[18] who obtained a value of +1.7, and more recently by Dempster,[19] who reported a packing fraction of approximately +2. On the basis of a packing fraction of +2, the data obtained in the present study lead to a value of 183.88 for the chemical atomic weight. This value is in fair agreement with the currently accepted value of 183.92.

MOLYBDENUM

Previous measurements on the relative abundance of the molybdenum isotopes were made by Aston,[20] de Gier and Zeeman,[21] Mattauch and Lichtblau,[22] and Valley.[23] Their values will be found in Table VIII.

[18] F. W. Aston, Proc. Roy. Soc. A132, 491 (1931).
[19] A. J. Dempster, Phys. Rev. 53, 64 (1938); 53, 869 (1938).
[20] F. W. Aston, Proc. Roy. Soc. A130, 308 (1931).
[21] J. de Gier and P. Zeeman, Proc. Roy. Acad. Amst. 39, 327 (1936).
[22] J. Mattauch and H. Lichtblau, Zeits. f. physik. Chemie 42, 288 (1939).
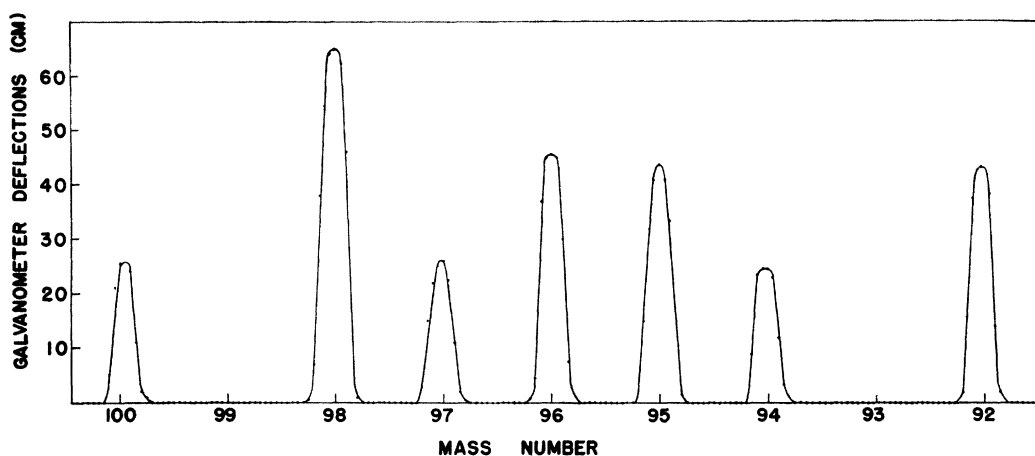[23] G. E. Valley, Phys. Rev. 57, 1058 (1940).

FIG. 7. Mass spectrum in the vicinity of the Mo^{++} ion. The peak at mass position 98 corresponds to a beam current of 2.5×10^{-12} ampere.

The material used in the present work was molybdenum hexacarbonyl vapor. As in the case of tungsten hexacarbonyl, it was found that bombardment of the vapor by 100-volt electrons produced the singly charged metallic ion (Mo^+) in greater abundance than other singly charged positive ions. The quantity of doubly charged mercury ions produced in vapor from the diffusion pump was not sufficient to introduce difficulties in measuring the Mo^+ peaks. In fact, with liquid nitrogen as a trap refrigerant, even the magnitude of the peaks due to singly charged mercury ions was small compared to the molybdenum peaks. However, in order to be certain that no spurious effects were caused by doubly charged mercury ions, abundance measurements on molybdenum were repeated for the Mo^{++} ions.

The mass spectrum observed in the vicinity of the singly charged molybdenum ion is shown in Fig. 6. Resolution seems to be adequate for abundance measurements. No background is visible at mass position 99 which can be attributed to $(Hg^{198})^{++}$ nor was a peak observed at mass position 101 where $(Hg^{202})^{++}$ ions would appear if present in measurable quantity. The mass spectrum in the vicinity of the doubly charged molybdenum ion is shown in Fig. 7. Here, again, the resolution is satisfactory and no spurious background effects are apparent.

The data obtained for isotopic abundances are given in Table VII. Each value shown in the table represents the average of at least four

careful determinations. Agreement between the data obtained from measurements on Mo^+ and Mo^{++} is fair. The final values for isotopic abundance are shown in Table VIII for purposes of comparison with the results of other investigations. It is believed that the abundances obtained in this study are accurate to ± 1 percent. If isotopes other than those listed in the table exist, they are present to less than 1/10,000 of the abundance of isotope 98.

It will be noted from Table VIII that the values for relative abundance as determined in

TABLE VII. Relative abundance of molybdenum isotopes as determined from molybdenum carbonyl.

	Measured abundance from indicated ion		
Isotope	Mo^+	Mo^{++}	Average
100	40.4	40.5	40.5
98	100.0	100.0	100.0
97	40.0	39.7	39.8
96	69.8	69.2	69.5
95	66.5	65.7	66.1
94	38.4	37.6	38.0
92	66.8	66.4	66.6

TABLE VIII. Relative abundances of molybdenum isotopes.

	Aston	Mattauch	Valley	Present study
100	42.6	33.8	38.4	40.5
98	100	100	100	100
97	41.7	34.2	40.0	39.8
96	77.4	66.1	68.9	69.5
95	67.4	64.2	66.8	66.1
94	43.5	34.3	39.0	38.0
92	61.7	61.0	61.8	66.6

the present study are in fair agreement (within the stated limits of error) with Valley's results for all isotopes except 100 and 92. In the case of isotope 100, the difference amounts to approximately 5 percent; for isotope 92 the difference is 7 percent. These differences are well beyond the limits of experimental error and could be accounted for by the presence of impurities. However, the close agreement between the results obtained for Mo+ and Mo++ ions in the present work would seem to preclude the possibility of large amounts of impurities. The large differences between the present results and Valley's results for isotopes 100 and 92 indicate that further work on molybdenum would be desirable.

The packing fraction for molybdenum has been measured by Dempster[24] and by Graves.[25] The value obtained by Graves is −5.5. Use of this value for the packing fraction in connection with the abundances obtained in the present study gives a value of 95.90 for the chemical atomic weight. The International Atomic Weight is 95.95.

BROMINE

Early experiments[26] indicated that bromine consists of the two isotopes 79 and 81 in practically equal proportions. Even careful photometric measurements[27] failed to reveal any differences in the intensity of the two observed lines. However, Blewett[28] by methods involving electrical measurements determined the ratio of 81 to 79 as 0.975 ± 0.025. Blewett's measurements were made on the peaks produced by Br+ and Br− ions, and measurements on Br++ and Br−− ions served as a check.

The present study was undertaken with the purpose of obtaining more accurate data on relative abundance. Since the two bromine isotopes are so nearly equal in abundance, instrumental errors produced by small variations in the grid resistor with ion current are negligible and high precision is readily obtainable. As the resolving power of the spectrometer used in this work was higher than that of the instrument used by Blewett, it was possible to make measure-
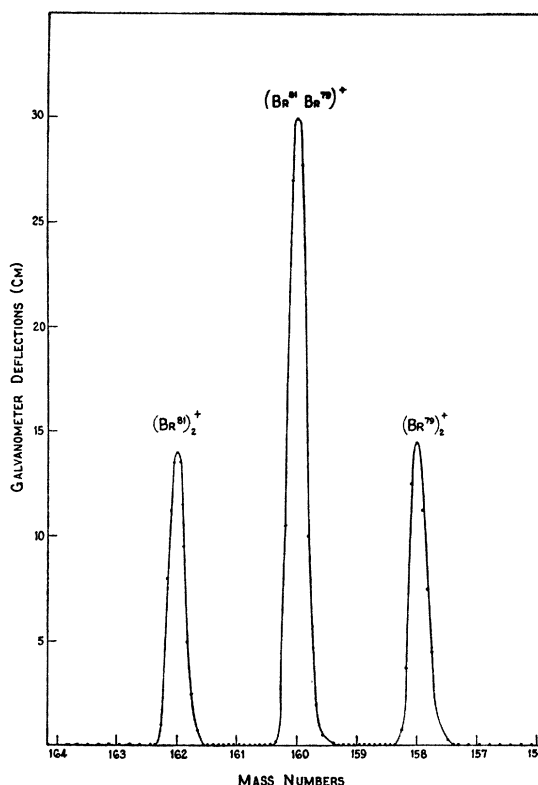
[24] A. J. Dempster, Phys. Rev. **53**, 64 (1938).
[25] A. C. Graves, Phys. Rev. **55**, 863 (1939).
[26] F. W. Aston, Phil. Mag. **40**, 631 (1920).
[27] F. W. Aston, Proc. Roy. Soc. **A132**, 489 (1931).
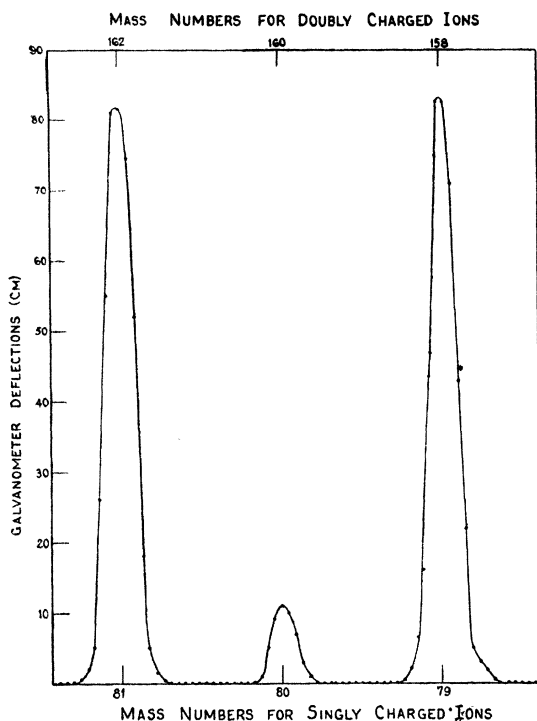[28] J. P. Blewett, Phys. Rev. **49**, 900 (1935).

FIG. 8. Mass spectrum in the vicinity of the Br_2^+ ion. The peak at mass position 160 corresponds to a beam current of 1.5×10^{-11} ampere.

ments on the Br_2^+ peaks as well as on the Br+ and Br++ peaks.

There are obvious advantages in determining isotopic abundance from measurements of the Br_2^+ peaks. If N_{79} and N_{81} are the numbers of atoms of Br^{79} and Br^{81} present in a given sample, the numbers of molecules of type $(Br^{79})_2$, $(Br^{79}Br^{81})$, and $(Br^{81})_2$ are

$$\frac{N_{79}^2}{2(N_{79}+N_{81})}, \quad \frac{N_{79}N_{81}}{N_{79}+N_{81}}, \quad \text{and} \quad \frac{N_{81}^2}{2(N_{79}+N_{81})},$$

respectively. Hence, the ratio of atomic abundance will be equal to the square root of the measured ratio of $(Br^{79})_2^+$ and $(Br^{81})_2^+$ peaks. The effects of random errors of measurement are reduced accordingly. Abundances determined in this manner can be checked by comparisons of $(Br^{79})_2^+$ and $(Br^{81})_2^+$ peaks with the $(Br^{79}Br^{81})^+$ peak. With the spectrometer used, these measurements could all be performed satisfactorily.

Two samples of bromine were used in the

MASS NUMBERS FOR DOUBLY CHARGED IONS

MASS NUMBERS FOR SINGLY CHARGED IONS

FIG. 9. Mass spectrum in the vicinity of the Br^+ and Br_2^{++} ions. The peak at mass position 79 corresponds to a beam current at 1×10^{-11} ampere.

present work. One of these was prepared under vacuum by a reaction between anhydrous cobaltic fluoride and potassium bromide. The other was a Baker's product, which was subjected to prolonged pumping at dry ice temperatures in order to remove possible traces of HBr. No peaks attributable to HBr were observed in ions from either sample.

The mass spectrum obtained in the vicinity of the Br_2^+ ions is shown in Fig. 8. The spectrum has the general appearance to be expected on the basis of previous results. The peaks are sufficiently well resolved for satisfactory abundance measurements, and no background effects could be detected.

In Fig. 9 is shown the spectrum obtained in the vicinity of the Br^+ ion. Large peaks were observed at mass positions 79 and 81, while a small peak was observed at mass position 80. Since ions of the type Br_2^{++} have the same m/e as the Br^+ ions, it was suspected that the peak at mass position 80 was actually due to ions of the type $(Br^{81}Br^{79})^{++}$. Accordingly, the observed magnitudes of the $(Br^{79})^+$ and $(Br^{81})^+$ peaks were
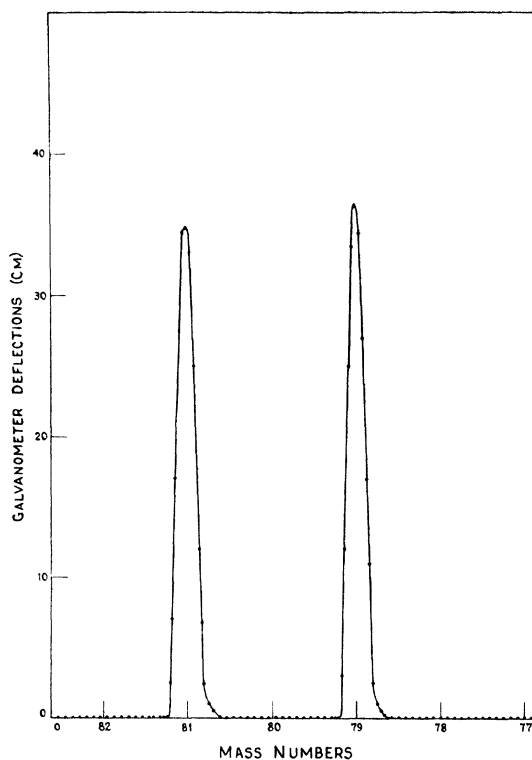

MASS NUMBERS

FIG. 10. Mass spectrum in the vicinity of the Br^{++} ion. The peak at mass position 79 corresponds to a beam current of 1.5×10^{-12} ampere.

subjected to appropriate corrections for $(Br^{79})_2^{++}$ and $(Br^{81})_2^{++}$ before abundances were calculated.

In order to check the above explanation of the small peak shown in Fig. 9 as well as to obtain further data on abundance, the spectrum in the region of the Br^{++} ion was carefully investigated. As the diatomic ion of equal m/e is Br_2^{++++} and as the probability of the formation of such an ion is slight, only two peaks were to be expected in this region. In Fig. 10 the observed mass spectrum is shown. The only peaks observed corresponded to ions of the type $(Br^{79})^{++}$ and $(Br^{81})^{++}$.

TABLE IX. Determinations of relative abundance of bromine isotopes. Weighted mean value for ratio 81/79 $= 0.979 \pm 0.004$.

Type of ion	Peaks compared	Number of determinations	Ratio 81/79
Br_2^+	162, 158	80	0.980 ± 0.002
Br_2^+	162, 160	28	0.974 ± 0.002
Br_2^+	160, 158	28	0.982 ± 0.004
Br^+	81, 79	12	0.978 ± 0.002
Br^{++}	81, 79	16	0.974 ± 0.002

In Table IX is a summary of the results obtained from measurements on various ionic species. Values obtained from the data on different types of ions are in fair agreement. The final value for the abundance of Br[81] relative to Br[79] is 0.979±0.004. No isotopes except 79 and 81 were detected. If isotopes 78, 80, or 82 exist, their abundance is less than 1/10,000 that of isotope 79.

The most recent figure[29] for the packing frac-

tion of bromine is −7.4. In connection with this value the data obtained in this study give a value of 79.908 for the chemical atomic weight. The accepted international value is 79.916.

ACKNOWLEDGMENT

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Note on the Lifetime of Metastable States*

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The available data on approximately forty γ-emitting metastable states of isomeric nuclei has been compared with the present theories. Nearly all of the points lie near two straight lines when $\log \lambda$ (sec.$^{-1}$) is plotted against $\log E$ (Mev). These lines fall close to the theoretical curves of Bethe calculated for electric 2^4 and 2^5 pole radiation.

INTRODUCTION

SINCE the original proposal of the existence of metastable states by von Weizsacker,[1] several theories have been developed which relate the lifetime of a metastable state against γ-emission to the energy of this state. Included in these are the calculations on the liquid droplet model by Bethe,[2] Lowen,[3] Flügge,[4] Fierz,[5] and Koyenuma.[6] Calculations of the lifetime energy relationship have also been worked out on the basis of an alpha-particle moving in a potential well by Hebb and Uhlenbeck[7] and by Segrè.[8] Several of these theories have recently been dis-

cussed by Berthelot.[9] In addition, a correction of the earlier formulae has been proposed by Hebb and Uhlenbeck to take into account the additive decay probability brought about by internal conversion. This correction factor $[1+(N_e/N_\gamma)]$, where N_e/N_γ is the ratio of the number of conversion electrons emitted to the number of γ-rays emitted, becomes extremely important at low energies and approaches unity for high energies. It should be noted, however, that the lifetime relationships are meant merely as approximations and should, therefore, be expected to give only the order of magnitude of the lifetime as a function of the energy. In addition to this, the important ratio N_e/N_γ has been calculated only for low atomic numbers.

EXPERIMENTAL DATA

In Fig. 1 we have plotted the log of the decay constant vs. log of the energy of the metastable state for the states thus far studied. This includes nuclei in which the ground state is stable as well

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