Deepest Terms in Ions of the Isoelectronic Sequences Ni I to Kr IX and Pd I to Xe IX

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Radiations corresponding to the energy differences between the terms $3d^{10}\ {}^1S_0$ and $3d^9\,4p\ {}^3P_1^0$, ${}^1P_1^0\ {}^3D_1^\circ$ in Ge V, As VI, Se VII and Br VIII have been found. Also, the corresponding radiations, $4d^{10}\ {}^1S_0-4d^9\,5p\ {}^3P_1^0$, ${}^1P_1^0$, ${}^3D_1^0$ in Sb VI, Te VII and I VIII have been observed. A new relation involving displaced frequencies has been used to predict the wave-length of the lines observed and the corresponding lines of Kr IX and Xe IX as yet not observed. A study of the ionization potentials in the sequences shows that it is possible to calculate the ionization potentials of atoms more highly ionized than those reported.

INTRODUCTION

VARIOUS investigators[1] have studied the spectra of elements of the isoelectronic sequence Ni I, Cu II, etc. Except in the case of Ni I the deepest term is a $^1S_0$ due to $3d^{10}$ electrons. Above this term are the $3d^9\,4s\ {}^1D\ {}^3D$ terms and still higher the $3d^9\,4p\ {}^{1,3}(P^0D^0F^0)$ terms are found. Between these terms and the ground state only three combinations are possible. They are $3d^{10}\ {}^1S_0-3d^9\,4p\ {}^3P_1^0$, ${}^1P_1^0$, ${}^3D_1^0$. Lines corresponding to these transitions had been found in Ni I, Cu II, Zn III, and Ga IV, but were not known previously in Ge V, As VI, Se VII and Br VIII.

In the isoelectronic sequence Pd I, Ag II, etc., the $4d^{10}\ {}^1S_0$ terms[2] were known for Pd I, Ag II, Cd III, In IV, and Sn V, but were not known previously for Sb VI, Te VII, or I VIII. The present paper presents data which establish the $3d^{10}\ {}^1S_0$ terms of Ge V, As VI, Se VII, Br VIII and the $4d^{10}\ {}^1S_0$ terms of Sb VI, Te VII and I VIII.

[1] Ni I. Russell, Phys. Rev. **34**, 821 (1929).
Cu II. Shenstone, Phys. Rev. **28**, 382 (1927); Lang, Phys. Rev. **31**, 773 (1928); Menzies, Proc. Roy. Soc. **A119**, 249 (1928); Kruger, Phys. Rev. **34**, 1122 (1929).
Zn III. Laporte and Lang, Phys. Rev. **30**, 378 (1927).
Ga IV. Mack, Laporte and Lang, Phys. Rev. **31**, 748 (1928).
Ge V. Mack, Laporte and Lang, Phys. Rev. **31**, 748 (1928).
As VI. Pattabhiramiah and Rao, Zeits. f. Physik **53**, 587 (1929); Mack, Phys. Rev. **37**, 470 (1931).

[2] Pd I. Shenstone, Phys. Rev. **36**, 670 (1930).
Ag II. Shenstone, Phys. Rev. **31**, 321 (1928); McLennan and Smith, Proc. Roy. Soc. Canada **20**, 110 (1926).
Cd III. McLennan, McLay and Crawford, Trans. Roy. Soc. Canada **22**, 45 (1928).
Cd III In IV. Gibbs and White, Phys. Rev. **31**, 776 (1928).
Sn V. Gibbs and White, Proc. Nat. Acad. Sci. **14**, 345 (1928).

EXPERIMENTAL

All spectrograms were taken with a 21 ft. grazing incidence (87°) vacuum spectrograph which was built by the Mann Instrument Company. The arrangement of the apparatus was essentially the same as that previously described[3] except for the spark chamber and electrode holders.

In previous work the temperature of the spectrograph did not remain constant during an exposure of 2–3 hours despite careful thermostating of the spectrograph assembly and the room. This temperature change was caused by heat conducted to the spectrograph from the spark chamber during operation.

A new spark chamber was constructed of two thin concentric copper spheres between which water was circulated. Adjustable water-cooled electrode holders were also constructed, as shown in Fig. 1. A one inch extension of these was permitted by 2 inches of sylphon tubing *D* placed in the outer wall. A running sleeve *C* was used to eliminate any side play in the electrode holders since this would cause the electrodes to get out of line when an adjustment of the electrodes was made during an exposure. Such maladjustment would cause the electrodes to become badly angled during a run, a condition which shielded the slit from the light of the spark. The running sleeve was machined carefully to 0.001 inch and a thin silver coating was applied to the surfaces to insure free sliding *C*.

With the water-cooled spark chamber and electrode holders it was possible to keep the

[3] Kruger, Rev. Sci. Inst. **4**, 128 (1933).

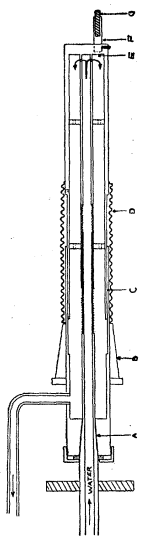

FIG. 1. Assembly of parts for water-cooled electrode holder.

temperature variation of the entire spectrograph to less than $\pm 0.1°C$.

The electrodes were made of 0.25 inch diameter rod F and were inserted into the ends of the electrode holders. The water-cooling stream E then comes within 0.1 inch of the electrode proper.

For best excitation the elements to be excited were made into copper compounds; for example, Cu_2Te, Cu_5As_2, Cu_2Se. These compounds were made by heating in vacuum with an oxyhydrogen flame a mixture of the proper proportions of the elements placed in a quartz tube. Reaction readily takes place leaving sticks of the proper diameter to be used. These sticks G were 0.125 inch in diameter and were pressed into a copper shell and used only in the positive terminal of the spark. Metallic germanium and antimony not compounded with copper were used. Powdered Cu_2Br_2 and Cu_2I_2 were each packed into a copper shell for the excitation of bromine and iodine. The negative terminal electrode was always a pure copper rod.

The length of the vacuum spark varied from 1–2 mm for the various elements. An air-cooled air gap 3 inches long was placed in series with this vacuum gap. The current was supplied by a 4 kenotron bridge set at an output potential of 100 kv. The capacity used was 0.04 mf. The condensers were automatically charged and discharged twice per second, the average spark duration being 0.05 sec.

DATA

Table I gives the intensity, wave-length, wave number and classification of lines observed in Ge V, As VI, Se VII and Br VIII. The wave-lengths were determined by using secondary standard lines[4] of copper. Calculations were made over a range of 5A or less, so that the corrections to be applied to the interpolated wave-lengths rarely exceeded 0.010A.

Ge V $^1{}^3D_1{}^0-{}^1P_1$ terms have been reported to have a $\Delta\nu$ separation of 1299 cm^{-1} by Mack, Laporte and Lang. The lines involving these terms, as reported in Table I, have a separation of 1298 cm^{-1} which is a satisfactory check for the identification of the lines.

As VI[1] has been reported by Pattabhiramiah and Rao but the present data do not fit their $^3P_1{}^0$, $^1P_1{}^0$, $^3D_1{}^0$ terms, a situation which throws considerable doubt on their classification. Borg and Mack[5] discussed the As VI spectrum at a meeting of the American Physical Society. Their terms[6] with respect to $3d^9\,4s\,{}^3D_3=0$ have the following values.

TABLE I. *Lines from transitions into the deepest terms of Ge V, As VI, Se VII and Br VIII.*

Element	Int.	λ	cm^{-1}	Combinations $3d^{10}-3d^9 4p$
Ge V	5	304.967A	327,904	$^1S_0-{}^3P_1{}^0$
	50	295.636	338,254	$^1S_0-{}^1P_1{}^0$
	30	294.506	339,552	$^1S_0-{}^3D_1{}^0$
As VI	5	232.217	430,632	
	5	226.836	440,847	$^1S_0-{}^3P_1{}^0$
	50	221.045	452,397	$^1S_0-{}^1P_1{}^0$
	40	219.982	454,583	$^1S_0-{}^3D_1{}^0$
Se VII	4	180.220	554,877	
	2	176.781	565,672	$^1S_0-{}^3P_1{}^0$
	35	172.920	578,302	$^1S_0-{}^1P_1{}^0$
	25	171.965	581,514	$^1S_0-{}^3D_1{}^0$
Br VIII	15	139.792	715,349	$^1S_0-{}^1P_1{}^0$
	15	138.927	719,802	$^1S_0-{}^3D_1{}^0$

[4] Kruger and Cooper, Phys. Rev. **44**, 826 (1933).
[5] Borg and Mack, Phys. Rev. **37**, 470 (1931).
[6] As communicated by letter from Professor Mack to the authors.

$3d^9\,4p\,{}^3D_1{}^0 = 123{,}390\ \text{cm}^{-1}$,

$3d^9\,4p\,{}^1P_1{}^0 = 121{,}220$,

$3d^9\,4p\,{}^3P_1{}^0 = 109{,}710$.

Table II compares the differences between the term values of $3d^9\,4p\,{}^3D_1{}^0 - {}^1P_1{}^0 - {}^3P_1{}^0$ as previously published and the values given by the authors. For Ge V the agreement is good. In the case of As VI the authors' data agree with Borg and Mack's data. However, on account of the greater accuracy of measurement in the region of the spectrum where Mack and his collaborators have taken their data the ${}^3D_1{}^0$, ${}^1P_1{}^0$, and ${}^3P_1{}^0$ term values for Ge V and As VI in Table III correspond to their values.

No data were available on Se VII or Br VIII, so that in these cases it was necessary to rely on intensity relations and predicted $\Delta\nu$ separation from the curves shown in Fig. 2, for the identification of the lines. However, there can be no mistaken identity since the lines given in Table I for Se VII and Br VIII are the only ones which were found on the plates in the predicted region, the intensities are the expected intensities, and the classified lines are the only ones which fit the curves of Fig. 2. It is interesting to note that the ${}^1S_0 - {}^3D_1{}^0$ transition is almost as strong as the ${}^1S_0 - {}^1P_1{}^0$ transition and much stronger than ${}^1S_0 - {}^3P_1{}^0$, a condition which exists throughout

TABLE II. *Comparison of frequency differences between terms.*

$\Delta\nu$ cm^{-1}	Ge V			As VI			
	Mack, Laporte and Lang	Kruger and Shoupp	Difference in $\Delta\nu$ cm^{-1}	Pattabhiramiah and Rao	Borg and Mack	Kruger and Shoupp	Difference in $\Delta\nu$ cm^{-1}
$3d^9\,4p\,{}^3D_1{}^0 - 3d^9\,4p\,{}^1P_1{}^0$	1299	1298	1	1304	2170	2186	16
$3d^9\,4p\,{}^1P_1{}^0 - 3d^9\,4p\,{}^3P_1{}^0$	10,400	10,350	50	11,729	11,510	11,550	40
	Te VII				Sb VI		
	Schoepfle published	Schoepfle unpublished	Kruger and Shoupp	Difference $\Delta\nu$ cm^{-1}	Schoepfle published	Schoepfle unpublished	Kruger and Shoupp	Difference $\Delta\nu$ cm^{-1}
$4d^9\,5p\,{}^3D_1{}^0 - 4d^9\,5p\,{}^1P_1{}^0$	7247	8556	8530	26	5948	6412	6440	28
$4d^9\,5p\,{}^1P_1{}^0 - 4d^9\,5p\,{}^3P_1{}^0$	8842	7533	7502	31	8948	8484	8537	47

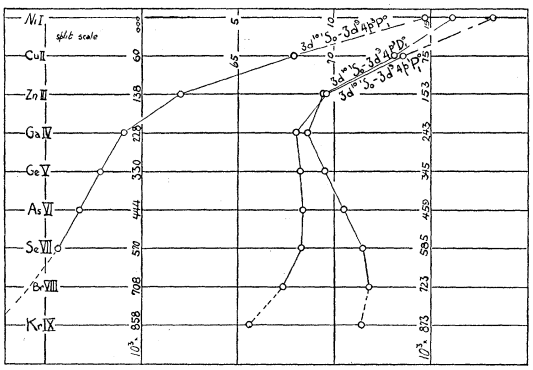

FIG. 2. Displacement of radiated frequencies.

the sequence. This means that the coupling is largely *jj* and not L.S.

TABLE III. *Term values.*

Element	$3d^{10}\,{}^1S_0$	$3d^9\,4p\,{}^3P_1{}^0$	$3d^9\,4p\,{}^1P_1{}^0$	$3d^9\,4p\,{}^3D_1{}^0$
Ge V	753,770	425,915	415,515	414,216
As VI	1,028,800	587,913	576,403	574,233
Se VII	1,341,900	776,228	763,598	760,386
Br VIII	1,692,400	(990,200)	977,051	972,598
Kr IX	(2,080,400)	(1,229,960)	(1,216,790)	(1,211,000)

Table III gives the term values of $3d^{10}\,{}^1S_0$ and $3d^9\,4p\,{}^3P_1{}^0$, ${}^1P_1{}^0$, ${}^3D_1{}^0$. The ${}^1P_1{}^0$ term values were calculated by assuming $(\nu/R)^{\frac{1}{2}}$ to be a linear function of the atomic number, beyond Ga IV. All other term values have been based on the ${}^1P_1{}^0$ terms and have been calculated from them by using the data in Table I except for Ge and As as described above.

Fig. 3, a Moseley diagram, shows the relationship between $(\nu/R)^{\frac{1}{2}}$ and atomic number for the sequences.

Table IV gives the new lines of Sb VI, Te VII and I VIII, with their classifications. These wave-lengths were determined in the same way as those in Table I.

Schoepfle[7] has reported the $4d^9\,5p\,{}^3P_1{}^0$, ${}^1P_1{}^0$, ${}^3D_1{}^0$ terms for Sb VI and Te VII, but his published data cannot be fully substantiated by the data in this paper. Schoepfle's ${}^1P_1{}^0$ terms are incorrectly designated. These data can be made to agree by the classification of the Sb VI line 952.16A as $4d^9\,5s\,{}^3D_2 - 4d^9\,5p\,{}^1P_1{}^0$ and the Te

[7] Sb VI Te VII. Schoepfle, Phys. Rev. **43**, 742 (1933).

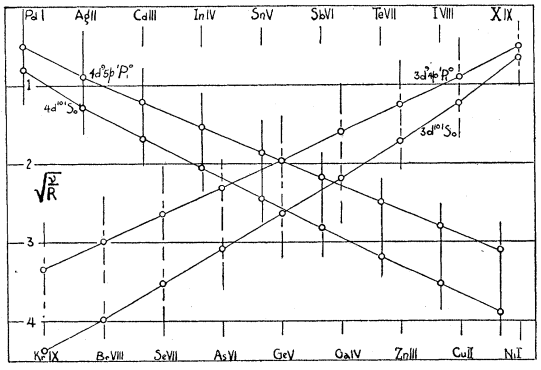

FIG. 3. Moseley diagram.

VII line 845.06A as $4d^9\ 5s\ ^3D_2 - 4d^9\ 5p\ ^1P_1^0$.[8] This gives the Sb VI $^1P_1^0$ term the value 107,795 and the Te VII $^1P_1^0$ term the value 121,425 on Schoepfle's term scale with $4d^9\ 5s\ ^3D_3 = 0$. The term difference for these two elements are compared in Table II and again the agreement is satisfactory for the unpublished terms of Schoepfle. As in the case of Ge V and As VI the $^3D_1^0$, $^1P_1^0$, $^3P_1^0$ terms of Sb VI and Te VII listed in Table V correspond with Schoepfle's data.

No data were previously available on I VIII, but again in this case the observed lines have the proper intensity and occur at a wave-length which makes them fit the curves in Fig. 4. The only other iodine line on the plate in this region was at 201.694A which could not be considered since it did not fit the curves in Fig. 4. The relative intensities of the lines in this sequence are analogous to those in the sequence Ni I, Cu II, etc., and check the assigned classification in all cases.

TABLE IV. *Lines from transitions into the deepest terms of Sb VI, Te VII and I VIII.*

Element	Int.	λ	ν cm^{-1}	Combinations $4d^{10} - 4d^9\ 5p$
Sb VI	15	292.405A	341,991	
	5	291.991	342,476	$^1S_0 - ^3P_1^0$
	75	284.890	351,013	$^1S_0 - ^1P_1^0$
	60	279.757	357,453	$^1S_0 - ^3D_1^0$
Te VII	5	243.880	410,038	
	5	242.248	412,800	
	2	237.541	420,980	
	2	236.460	422,905	$^1S_0 - ^3P_1^0$
	40	232.338	430,407	$^1S_0 - ^1P_1^0$
	35	227.823	438,937	$^1S_0 - ^3D_1^0$
I VIII	5	201.694	495,801	
	1	196.553	508,769	$^1S_0 - ^3P_1^0$
	50	194.152	515,060	$^1S_0 - ^1P_1^0$
	40	190.158	525,878	$^1S_0 - ^3D_1^0$

[8] From Dr. Schoepfle's data as communicated to the authors by him.

TABLE V. *Term values.*

Element	$4d^{10}$ $^1S_0$	$4d^9\ 5p$ $^3P_1^0$	$^1P_1^0$	$^3D_1^0$
Sb VI	868,140	525,611	517,127	510,715
Te VII	1,106,860	683,986	676,453	667,897
I VIII	1,370,370	861,601	855,310	844,492
Xe IX	(1,659,250)	(1,059,225)	(1,054,440)	(1,041,140)

Table V gives the term values of $4d^{10}\ ^1S_0$ and $4d^9\ 5p\ ^3P_1^0$, $^1P_1^0$, $^3D_1^0$. These values were calculated in the same way as those of Table III.

Figs. 2 and 4 show a new relationship between displaced frequencies in an isoelectronic sequence. Gibbs and his collaborators have shown that there is a linear displacement toward larger values in the frequency of multiplets from terms due to $3d^9\ 4s$ and $3d^9\ 4p$ and similar electron configurations. In the present case, where radiation which involves terms due to $3d^{10}$ and $3d^9\ 4p$ and similar electron configurations, the displacement is not linear. Table VI gives the wave numbers of lines represented by $3d^{10}\ ^1S_0$ $3d^9\ 4p\ ^1P_1$ in the isoelectronic sequence Ni I, Cu II, etc., and for $4d^{10}\ ^1S_0 - 4d^9\ 5p\ ^1P_1^0$ in the sequence Pd I, Ag II, etc., together with the first and second differences between these values. It is observed that the second differences are

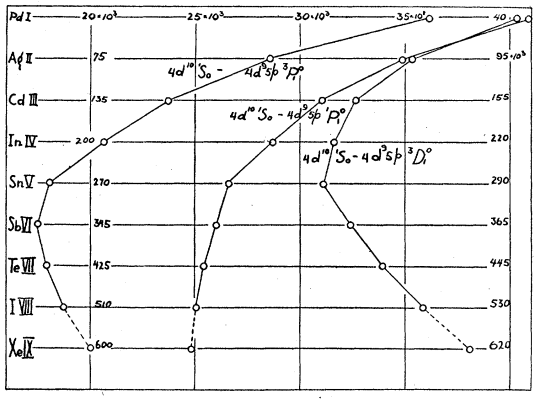

FIG. 4. Displacement of radiated frequencies.

TABLE VI. *Radiated frequencies with first and second differences.*

Elements of sequence	$3d^{10}\,{}^1S_0 - 3d^9\,4p\,{}^1P_1$ cm^{-1}	First difference cm^{-1}	Second difference cm^{-1}	Elements of sequence	$4d^{10}\,{}^1S_0 - 4d^9\,5p\,{}^1P_1$ cm^{-1}	First difference cm^{-1}	Second difference cm^{-1}
Ni I	18,254			Pd I	40,839		
		55,339				49,052	
Cu II	73,593		18,659	Ag II	89,891		7142
		73,998				56,194	
Zn III	147,591		14,488	Cd III	146,085		6423
		88,486				62,617	
Ga IV	236,077		13,691	In IV	208,702		5271
		102,177				67,888	
Ge V	338,254		11,966	Sn V	276,590		6519
		114,143				74,407	
As VI	452,397		11,762	Sb VI	351,013		4987
		125,905				79,394	
Se VII	578,302		11,142	Te VII	430,407		5259
		137,047				84,653	
Br VIII	715,349		(11,200)	I VIII	515,060		(5100)
		(148,247)				(89,753)	
Kr IX	(863,600)			Xe IX	(604,813)		

essentially constant and particularly so after the fifth or sixth stage of ionization has been reached.

From these data the curves in Figs. 2 and 4 were constructed. In Fig. 2, for example, 78,000 cm^{-1} are added to 60,000 cm^{-1}, the value of a point on the scale for Cu II, to get 138,000 cm^{-1} the value of the corresponding point for Zn III. Then 12,000 cm^{-1}, the approximate value of the second differences in Table VI for this sequence, is added to 78,000 cm^{-1} to give 90,000 cm^{-1}, the value added to 138,000 cm^{-1} to get the corresponding point on the graph for Ga IV. Next 12,000 cm^{-1} are added to 90,000 cm^{-1} which gives 102,000 cm^{-1}, the number to be added to 228,000 cm^{-1} in order to obtain the corresponding value 330,000 cm^{-1} for the point on the graph for Ge V. In this way the scale in Fig. 2 is obtained. A plot of the radiated frequencies gives the curves as shown. From them it is seen that, for ionization higher than Ga IV, the second differences are constant but have slightly different values for ${}^1S_0 - {}^3P_1{}^0$, ${}^1S_0 - {}^1P_1{}^0$ and ${}^1S_0 - {}^3D_1{}^0$. The scale for Ni I on this graph has been shifted 6000 cm^{-1} to smaller values for convenience in plotting the Ni I points.

In Fig. 4 the same procedure has been followed. There the constant second difference has been chosen as 5000 cm^{-1} so that after adding 55,000 cm^{-1} to 20,000 cm^{-1} (the point on the Pd I axis) to get 75,000 cm^{-1} for the point on the Ag II axis, 5000 cm^{-1} are added to 55,000 cm^{-1} to give 60,000 cm^{-1} the numbers to be added to 75,000 cm^{-1} to get the value 135,000 cm^{-1} for Cd III, etc. In this case the actual second differences are not as constant as those for Ni I, Cu II, etc., but are varying so slowly that it is possible to extrapolate the curves accurately to predict the corresponding radiation in Xe IX. Such an extrapolation has been made for both Xe IX and Kr IX and these values are included in all tables and curves. A study of the data in Table VI indicates that the extrapolated values should not be in error by more than ± 400 cm^{-1}, a variation which is in agreement with the check obtained between predicted and observed values during the process of following the observed lines through the sequences.

Tables VII and VIII give the ionization potentials for the isoelectronic sequence Ni I, Cu II, etc., and Pd I, Ag II, etc., and the first and second differences between the ionization potentials. Again it has been found that the second differences are essentially constant. From these data it is possible to derive an empirical formula from which the ionization potentials can be calculated. It is

$$I_n = I_1 + (n-1)(D_1 + [(n-2)/2]D_2),$$

where I_n is the ionization potential and n is stage of excitation which takes on the values one for Ni I or Pd I, two for Cu II or Ag II, etc.

TABLE VII. *Ionization potentials in the isoelectronic sequence Ni I, Cu II, etc.*

Element	$3d^{10}\,{}^1S_0$ cm^{-1}	Ionization potential in volts	First difference in volts	Second difference in volts	Ionization potential as calc. fr. formula
Ni I	46,850	5.8			4.4 v.
			14.4		
Cu II	163,634	20.2		4.9	19.5
			19.3		
Zn III	320,000	39.5		5.0	39.3
			24.3		
Ga IV	517,000	63.8		4.9	63.8
			29.2		
Ge V	753,770	93.0		4.7	93.0
			33.9		
As VI	1,028,800	126.9		4.7	126.9
			38.6		
Se VII	1,341,900	165.5		4.7	165.5
			43.3		
Br VIII	1,692,400	208.8		(4.5)	208.8
			(47.8)		
Kr IX	(2,080,400)	(256.6)			256.8

$$I_n = I_1 + (n-1)(D_1 + [(n-2)/2]D_2)$$
$$I_1 = 4.4;\quad D_1 = 15.1;\quad D_2 = 4.7$$

TABLE VIII. *Ionization potentials in the isoelectronic sequence Pd I, Ag II, etc.*

Element	$4d^{10}\,{}^1S_0$ cm^{-1}	Ionization potential in volts	First difference in volts	Second difference in volts	Ionization potential as calc. fr. formula
Pd I	67,236	8.3			8.1 v.
			13.6		
Ag II	177,164	21.9		3.5	21.5
			16.1		
Cd III	308,318	38.0		3.7	38.1
			19.8		
In IV	468,214	57.8		3.1	57.9
			22.9		
Sn V	654,527	80.7		3.5	80.9
			26.4		
Sb VI	868,140	107.1		3.0	107.1
			29.4		
Te VII	1,106,860	136.5		3.2	136.5
			32.6		
I VIII	1,370,370	169.1		3.0	169.1
			35.6		
Xe IX	(1,659,250)	(204.7)			204.9

$$I_n = I_1 + (n-1)(D_1 + [(n-2)/2]D_2)$$
$$I_1 = 8.1;\quad D_1 = 13.4;\quad D_2 = 3.2$$

I_1 corresponds roughly to the ionization potential of the first member of the sequence and has a value of 4.4 e.v. for the Ni I sequence and 8.1 e.v. for the Pd I sequence.

D_1 is approximately the first, first difference. It has a value of 15.1 for the Ni I sequence and 13.4 for the Pd I sequence.

D_2 is the value of the constant second difference and has been chosen as 4.7 for the Ni I sequence and 3.2 for the Pd I sequence.

The last columns of Tables VII and VIII give the values of ionization potentials as calculated from the above formula. The agreement with the experimental values is surprisingly good and is best for high stages of ionization. For this reason it seems that the calculation of ionization potentials for ions from Kr IX to Ag XX and Xe IX to Tb XX should not be in error by more than a few percent.

It is hoped that the extrapolation of the formula can be checked by the completion of the work now in progress on Rb X, Sr XI, Cs X and Ba XI.

It is to be regretted that the lack of a vacuum spectrograph which covers the range 600A to 2000A makes it impossible for the authors to obtain data involving the $d^9p - d^9s$ terms in these sequences. It is hoped that another laboratory having the proper facilities will undertake that study.

Note added in proof:

The following is a list of known As VI levels from Borg and Mack's data which they have asked the authors to add to this paper. The last digit of the term values has been rounded to the nearest zero.

$3d^9\,4s$	$^3D_3$	0 cm^{-1}	$3d^9\,4p$	$^3F_2$	112,620 cm^{-1}	?
	$^3D_2$	2,050		$^3P_0$	112,880	
	$^3D_1$	5,690		$^3D_2$	118,280	
	$^1D_2$	9,050		$^3D_3$	118,290	
$3d^9\,4p$	$^3P_2$	104,250		$^1F_3$	120,620	
	$^3F_3$	108,730		$^1P_1$	121,220	
	$^3P_1$	109,710		$^3D_1$	123,390	
	$^3F_4$	111,890		$^1D_2$	124,380	