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ABSTRACT

The rhenium KLL, KLM, KLN, and KLO Auger lines have been observed in experiments on the decay of Os^{182} and Os^{183}. The measured energies are compared with those given by semi-empirical theories due to Bergström and Hill and to Asaad and Burhop; satisfactory agreement is found. A comparison of the measured intensities with those given by the non-relativistic theory of Asaad and Burhop shows less satisfactory agreement.

Conversion lines attributed to a previously reported M3 isomer in Os^{189} have been observed and the transition energy found to be 30.81 ± 0.03 kev. L and M subshell ratios are reported. The transition appears to be hindered by a factor of 5×10^4 relative to the single particle estimate.

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1. INTRODUCTION

In two previous papers hereafter referred to as (I)[1] and (II)[2] investigations of the decay spectra of the isotopes Os^{182} and Os^{183} were reported. During the course of this work information was also obtained on the KLL and KLM Auger transitions in rhenium. Rather little experimental information has been available up to the present on Auger transitions in any element and the work reported here is the most extensive so far apart from the recent work on tungsten by Gallagher, Strominger, and Unik.[3] Information has also been obtained on the isomeric state of Os^{189} which was reported by Scharff-Goldhaber, Alburger, Harbottle, and McKeown.[4]

The experimental method was fully described in paper (I)[1] to which the reader is referred.

2. THE RHENIUM AUGER TRANSITIONS

2.1 Experimental methods.

In the measurements reported in (I) two instruments were used to observe the low energy electrons. These were a permanent magnet type of 180° spectrograph with photographic recording and a double-focussing spectrometer. Only the first of these was of use in investigations of the Auger spectrum, since the higher resolution was necessary both in order to resolve the various lines of the Auger spectrum from one another and also to resolve them from other lines present in the same energy region. Most of these measurements were done with a 50-gauss spectrograph. As explained in (I) the energy measurements in this region are expected to have an accuracy of the order of ±0.05%. The intensities were obtained by scanning the lines with a recording densitometer and using the method of Mladjenovic and Slätis.[5] The errors on the intensities using this method are usually taken to be ±20%.

However with lines in a small energy region such as that in which the Auger lines lie the accuracy may well be higher. In fact comparison of the results for various plates suggests that the standard deviation is about ±10%.

2.2 Theoretical

When a vacancy is produced in the K shell of an atom the vacancy is in general filled by one of two processes. In the first of these the vacancy is filled by an electron from a higher shell, an X ray being emitted to conserve energy. In the second process the vacancy is again filled by an electron from a higher shell X but instead of an X ray being emitted the excess energy is lost in the emission of another electron from a higher shell Y. This last process is known as the Auger process and the emitted electron a KXY Auger electron. Clearly several Auger electrons may be emitted after the production of a single K shell vacancy and the atom may become multiply ionized.

The fraction ω_K of K shell vacancies which are filled with subsequent emission of a KX ray is known as the K shell fluorescent yield. According to Burhop[6] ω_K depends on the nuclear charge Z as follows:

$$\left\{\frac{\omega_K}{1-\omega_K}\right\}^{1/4} = -A + BZ - CZ^3 \qquad (1)$$

A, B, and C are constants and the values obtained for them by Hagedorn and Wapstra[7] to fit the most recent data are $A = 6.4 \times 10^{-2}$, $B = 3.40 \times 10^{-2}$, and $C = 1.03 \times 10^{-6}$.

If the atomic electrons obey the jj coupling rules, which are in fact fairly good for heavy nuclei, the energy of a KXY Auger electron is given by the relation

$$E_Z(KXY) = E_Z(K) - E_Z(X) - E_Z^X(Y) = E_Z(K) - E_Z^Y(X) - E_Z(Y) \qquad (2)$$

where $E_Z^X(Y)$ is the binding energy of an electron in the Y shell of an atom with nuclear charge Z and with an X shell vacancy. This quantity will be

expected to lie between the values of $E_Z(Y)$ and $E_{Z+1}(Y)$ corresponding to no screening and complete screening of the electron in the Y shell by the electron in the X shell. As suggested by Bergström and Hill[8] we may therefore write

$$E_Z^X(Y) = E_{Z+\Delta Z}(Y)$$

where $E_{Z+\Delta Z}(Y)$ is the binding energy in a hypothetical neutral atom with charge $Z+\Delta Z$. Of course ΔZ will vary for different X and Y but we might hope that it will not vary much with Z. So far there is not a great deal of experimental evidence on this subject but what there is suggests that for the KLL Auger electrons values of ΔZ of 0.54 for atoms with L_1 or L_2 shell vacancies and of 0.76 with L_3 shell vacancies fit the data to an accuracy of about one or two parts in a thousand.[3,9,10,11]

The above is only an empirical approach to the problem. Most theoretical treatments of the Auger spectra have been made in the limits of jj coupling, for heavy elements, and of LS coupling for light elements. The agreement with experiment is not good. Recently Asaad and Burhop[12] have made non-relativistic calculations in intermediate coupling. The results are in better, but still not very good, agreement with experiment than those from previous calculations. However neglect of relativity in the heavier elements is not justified and relativistic calculations are to be made. According to Asaad and Burhop it is possible to make a semi-empirical correction to the energies for relativistic effects. They deduced the constants in this correction formula from the results of Mladjenovic and Slätis[13] for Z = 83 and it is claimed that the results should be applicable to other elements. More lines are predicted in intermediate coupling than in the jj and LS limits but the new lines are very weak and so far have not been seen.

2.3 Experimental Results and comparison with theory.

In Table 1 the experimental results are shown. They are compared with some other data for charge numbers 74, 79, 80, and 83. As can be seen our experimental energies are fitted rather well by the method of Bergström and Hill[8] when the following values for ΔZ are taken: for the KLL Auger lines and L_1 or L_2 vacancies $\Delta Z = 0.48 \pm .05$ and for L_3 vacancies $\Delta Z = 0.71 \pm .05$;

for the KLX Auger lines $\Delta Z = 0.75 \pm 0.2$ for all L vacancies. The errors are given in order to give an idea of the spread in ΔZ allowed by the experimental results; they are not intended to be limits of error or standard deviations in a strict sense. These values of ΔZ for the KLL Augers are in satisfactory agreement with those of 0.55 for the L_1 and L_2 shells and 0.76 for the L_3 shell found by Bergström and Hill[8] for Z = 80 and with corresponding values of 0.52 and 0.76 found by Mladjenovic and Slätis[9] for Z = 83.

The KLL Auger energies are also compared in Table 1 with those calculated from the semi-empirical recipe of Asaad and Burhop.[12] The agreement is very good except that the theoretical values seem to be systematically lower than the experimental values by about 0.04 kev.

There is moderately good agreement between the experimental intensities for charge numbers 74, 75, 79, 80, and 83 considering the errors of measurement. The theoretical values (for Z = 80) calculated on the intermediate coupling theory of Asaad and Burhop[12] are clearly not in very good agreement with the experimental results. This disagreement may be due to not taking into account relativity.

From the intensity results here, the ratio of the sum of the intensities of the KLX lines to that of the KLL lines can be calculated. A value of 0.57 is obtained. It is in reasonable agreement with other experimental data on this quantity.[15]

The fluorescent yield of the rhenium K shell was estimated in the following way. The total K Auger intensity was measured relative to the K line of the 114.44 kev transition in Os^{183}; this line has an energy of 42.80 kev which is quite close to those of the Auger lines. The multipolarity of this transition is known accurately from the L subshell intensity ratios (see paper II). It is therefore reasonable to suppose that the K conversion coefficient of this transition can be well estimated from the theoretical conversion coefficients of Rose.[16] With this value and the measured relative intensities of the 114.44 kev gamma ray and the K X rays (see paper I) the intensity of the Auger electrons relative to that of the K X rays can be obtained. The experimental value for the ratio of the intensity of the K Auger lines to that of the K line of the 114.44 kev transitions is 0.145. Taking the conversion coefficient as 3.0 and the ratio of K X ray intensity to

Table I. Observed and Calculated Auger Spectra

The column exp. gives the measured energies from this work. The column calc B. gives the energies calculated by the method of Bergström and Hill.[8] For the KLL Augers we have taken Z = 0.48 with L_1 and L_2 vacancies and 0.71 with L_3 vacancies. For the KLX Augers we have taken Z = 0.75 for all L vacancies. The energies in column calc A. were calculated from the recipe given by Asaad and Burhop.[12] The experimental intensity data for Z = 79 are those of Mihelich,[14] those for Z = 80 are the data of Bergström and Hill,[8] those for Z = 83 are the data of Mladjenovic and Slätis,[13] those for Z = 74 are the data of Gallagher, Strominger, and Unik[3] and those for Z = 75 are taken from the present work.

Transition	Energy in kev			Intensities					
	exp.	calc. B	calc. A	Z=79	Z=80	Z=83	Z=74	Z=75	calc.
KL_1L_1	46.40	46.40	46.36	1.0	1.0	1.0	1.0	1.0	1.00
KL_1L_2	47.01	46.97	46.95	1.7	1.2	1.8	2.1	1.4	1.20
KL_1L_3	48.37	48.37	48.33	1.20	0.7	1.1	0.7	1.1	2.27
KL_2L_2	47.51	47.55	47.45	0.3	0.2	0.2		0.1±.05	0.15
KL_2L_3	48.91	48.93	48.89	1.4	1.4	1.6	1.9	2.0	4.32
KL_3L_3	50.37	50.36	50.33	0.8	0.6	0.8	0.8	0.95	2.40
KL_1M_1	56.19	56.12				0.4	0.46	0.44	0.31
KL_1M_2	56.39	56.38				0.5	0.40	0.30	0.19
KL_1M_3	56.68	56.71				0.8	0.46	0.62	0.37
KL_2M_1		56.68							0.16
KL_1M_4		57.13							0.019
KL_1M_5		57.20							0.032
KL_2M_2	56.91	56.94						V.weak	0.07
KL_2M_3	57.30	57.27				0.7	0.23	0.38	0.93
KL_2M_4	57.71	57.69				0.1		0.32	0.055
KL_2M_5		57.76							0.27
KL_3M_1	58.11	58.11				0.21	0.38	0.32	0.31
KL_3M_2	58.35	58.37				0.7	0.54	0.59	0.80
KL_3M_3	58.65	58.70				1.0	0.69	0.60	0.85
KL_1N		58.61							
KL_3M_4	59.15	59.13					0.38	0.32	0.33
KL_2N		59.17							
KL_3M_5		59.19							0.20
KL_3N	60.66	60.60					0.15	0.16±.08	
KL_1O		59.06							
KL_2O	59.60	59.62							
KL_3O	61.08	61.05							

114.44 kev gamma ray intensity as 7.15, a value of 0.061 is obtained for the ratio of K Auger intensity to K X ray intensity; the error is likely to be about ±20%. Thus the fluorescent yield ω_K is equal to 0.94 ± .01. This is in satisfactory agreement with the value of 0.947 calculated from the theoretical formula mentioned in section 2.2.

3. THE ISOMERIC TRANSITION IN Os^{189}

Activities with half lives of 6 or 7 hours which might be attributed to the decay of an isomeric state in Os^{189} were first reported by Chu[17] and by Greenlees and Kuo.[18] More recently Scharff-Goldhaber, Alburger, Harbottle, and McKeown[4] reported an isomeric transition in Os^{189} with an energy of 30.0 kev and a half life of 5.7 hours. Since the energy of this transition is below the K binding energy in osmium, any K X rays which are seen must arise from states fed by the 30 kev transition. Scharff-Goldhaber et al. claim that there are no such K X rays; thus, the activity seen by Greenlees and Kuo who observed K X rays cannot be connected with the 30 kev decay in Os^{189}. Scharff-Goldhaber et al. found that the ratio of the intensities of the sum of the L_1 and L_2 conversion lines to that of the L_3 line was less than 0.2 and that the conversion coefficient was about 5×10^3.

In this experiment conversion lines attributable to a transition of energy 30.81 kev in osmium were seen. The transition was observed to have a half life of five or six hours from the decay of the lines on the photographic plates of the 50 gauss spectrograph. No attempt was however made to obtain an accurate value for the half life or to decide to which isotope of osmium this transition should be attributed. All that can be said regarding the latter point is that the activity could not have been formed from an $(\alpha,4n)$ reaction on tungsten. From Table 1 of paper (I) it can be seen that the activity could therefore be from any of the osmium isotopes having mass numbers betweem 183 and 189. Owing to the similarity in half life, energy and subshell ratios (see Table II) between this transition and that observed by Scharff-Goldhaber et al. it seems plausible to assume that they are in fact the same. It must be remarked however that there is an appreciable discrepancy in energy between our value of 30.81 ± 0.03 kev and that of 30.0 kev reported by Scharff-Goldhaber et al., and this assumption could be incorrect.

Table II. Energies and Intensities of the Conversion Lines

E_γ is the transition energy calculated from the appropriate binding energy. I(exp) is the experimental intensity and I(MX) etc. is the theoretical intensity for an MX transition.[16] The values in brackets are for the intensities of the M lines relative to that of the M_3 line.

Transition	E_e kev	E_γ kev	I(exp)	I(M2)	I(M3)	I(M4)
L_1	17.89	30.86	0.3	1.72	0.18	0.056
L_2	18.47	30.85	0.02	0.10	0.012	0.004
L_3	19.95	30.81	1.0	1.0	1.0	1.0
M_1	27.74	30.79	0.07	(1.7)	(0.21)	(0.07)
M_3	28.37	30.82	0.33	(1.0)	(1.0)	(1.0)
N_1	30.14	30.79	0.03			
N_3	30.31	30.77	0.12			
O_3	30.76	30.80	0.03			

The intensities which were obtained by the method of Mladjenovic and Slätis[5] from a densitometer trace are also given in Table II. The curve of efficiency for detection against gamma ray energy is very steep in this region, so that these results are subject to greater errors than in the higher energy region. Nevertheless they should not be too bad for close lying lines; the ratio M_1/M_3 might be expected to be more accurate than that for L_1/L_3. Comparison between the observed and theoretical[16] L subshell ratios for M2, M3, and M4 transitions clearly establishes the transition as M3. If this is so, the theoretical L shell conversion coefficient should be 2.5×10^5. Allowing a factor of 0.3 for the M, N, etc. shells this gives a total conversion coefficient of $3.3.10^5$. Thus if we take the half life of the transition to be 5.7 hours, as given by Scharff-Goldhaber et al.,[4] the gamma ray half life is 6.8×10^9 seconds. The half life calculated from the single particle formula is 1.4×10^5 sec so that the transition appears to be hindered by a factor of 5×10^4.

The ground state of Os^{189} is known to have spin 3/2[4] and this spin could be expected rather naturally from the Nilsson Scheme of levels in a deformed nuclear potential;[19] the state would be the 3/2- [512] using the notation of paper II. The isomeric state would then be, equally naturally, the state 9/2- [505]. The M3 transition between these states is allowed according to the selection rules in the asymptotic quantum numbers. However, Os^{189} is getting rather far removed from the region of highly deformed nuclei where the asymptotic quantum numbers might be expected to be fairly good quantum numbers. The 3/2- state arises originally from the $f_{5/2}$ spherical state and the 9/2- state arises from the $h_{9/2}$ spherical state. An M3 transition between spherical states with these orbital angular momenta would be forbidden.

An M3 transition which is probably the same as that in Os^{189} but inverted occurs in Os^{191}.[4] This transition has a similar large hindrance factor of 2×10^4.

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