# Oscillator strength determination for heavy elements

# ABSTRACT

Some important developments during the years 1980-1989 concerning the determination of atomic oscillator strengths for the heavy elements (Z > 28) are presented. More particularly, we focus attention here on the progress concerning f value determination in the copper to krypton sequences and also concerning lifetime measurements for neutral and singly ionized refractory elements. Astrophysical implications are also mentioned.

# INTRODUCTION

Calculations of atomic structure and radiative transition rates for heavy atoms and ions (Z > 28) have become more tractable during recent years mostly due to the advent of large supercomputers which make the simultaneous consideration of correlation and relativistic effects more feasible and more realistic. The calculation of transition probabilities have also benefited from the increase in experimental data concerning the atomic structure of moderately or highly ionized elements (laser produced plasmas -Tokamak devices - beam-foil spectroscopy) which often makes the use of semi-empirical methods for calculating oscillator strengths more reliable.

During the past decade, a significant change of orientation in the experimental determination of oscillator strengths for heavy elements has consisted in the fact that the activities in this field, which were previously concentrated upon the use of absorption and emission techniques, have progressively moved to radiative lifetime measurements and to branching ratio determinations. This change is related directly to the intensive and general use of pulsed dye lasers for selective excitation and to the development of powerful beam-laser techniques.

It is not the purpose of this limited review to discuss all the aspects of the progress realized in the whole field but we want to focus here on two specific subfields. In the first section of this paper, we will concentrate upon the oscillator strength determination along the copper to krypton sequences and spotlight some specific characteristics of these results. The second section will present a summary of lifetime and branching ratio measurements for heavy refractory elements (neutral or singly ionized) which remain of major interest in astrophysics for the determination of the chemical composition of the sun.

THE COPPER TO KRYPTON SEQUENCES : STATE OF THE ART

The knowledge of the atomic structure of moderately or strongly ionized elements along the Cu to Kr sequences has progressed substantially in the recent past. The state of the analysis of atomic spectra for the elements Cu to W is illustrated in Table 2. This table updates a previous compilation of Cowan (1981a). These developments have given an important impulse to the determination of transition probabilities (or related quantities), although the available data remain very fragmentary. The main references containing f values (or related quantities) are summarized in Table 1. They concern allowed (E1) or forbidden (M1, E2) transitions.

Table 1 - Recent progress in f value or lifetime determinations along the Cu to Kr sequences (period 1980-1989).

lons	Type	Reference		
		Cu Sequence		
Cu	E1	Bezuglov et al. (1982)		
Cu to M	o E1	Curtis (1981)		
Cu	E1	Hannaford and Lowe (1983a)		
Cu	E1	Osherovich et al. (1981)		
Cu	E1	Kono and Hattori (1982)		
Cu	E1	Cederquist et al. (1984)		
Cu	E1	Carlsson et al. (1987)		
Cu	E1	Carlsson (1988)		
Cu	E1	Carlsson et al. (1987)		
Cu	E1	Zettl et al. (1984)		
Ga	E1	Lindgård et al. (1982)		
Ga,Ge	E1	Ryabtsev and Wyart (1987)		
Ge	E1	Pinnington et al. (1981a)		

As Ge Scu Cu Kr Mo Sr I Xer Tm Cu	to to to to to to	Br Mo In Kr Ag Nd Pt Bi U	E1 E1 E1 E1 E1 E1 E1 E1 E1 E1 E1 E1 E1 E	Pinnington et al. (1981b) Ryabtsev et al. (1984) Wyart et al. (1984) Bahr et al. (1982) Victor and Taylor (1987) Lindgård et al. (1980) Pinnington et al. (1982) Livingston et al. (1982) Livingston et al. (1981) Denne and Poulsen (1981) Biémont (1988) Johnson et al. (1985) Breton et al. (1988) Finkenthal et al. (1986) Mandelbaum et al. (1983) Curtis (1989) Curtis and Theodosiou (1989) Zn Sequence
				Ell'ocquerice
Zn			E1	Afaneseva (1982)
Zn			E1	Bruneau (1984)
Zn	to	UE	E1,M1,	Anderson and Anderson (1983)
			E2	100 S. 100 M.22 MA 200
Zn			E2	Beck (1981)
Zn			E1	Ueda et al. (1981)
Zn	to	Мо	E1	Victor and Taylor (1987)
Mo,	Ag	3	E1	Trabert (1989)
Mo		-	E1	Finkenthal et al. (1981)
Zn	to	Kr	E1	Pinnington et al. (1982)
Zn	to	W	E1	Biémont and Godefroid (1980)
As			E1	Pinnington et al. (1981b)
Kr			E1	Pinnington et al. (1984)
KD	to	xe	E1	Biémont et al. (1989a)
xe D-			E1	Breton et al. (1988)
PT		w	E1	Finkenthal et al. (1986)
RD Tm	10	M DL	C1	Blemont (1989)
1.10	LU	ΓL	C I	Mandelbaum et al. (1983)
				Ga Sequence
Ga	to	Kr	E1	Aashamar et al. (1983)
Ga			E1	Brage et al. (1987)
Ga			E1	Tursunov and Eshkobilov (1984)
Ga			E1	Lindoard et al. (1982)
Ge			E1	Miller and Bengtson (1980)
As			E1	Pinnington et al. (1981b)
Ga	to	Se	E1	Migdalek (1983)
Br	to	In	E1	Biémont and Quinet (1989)
Мо			E1	Finkenthal et al. (1981)
Xe			E1	Breton et al. (1988)
Ga	to	Xe	M1,E2	Biémont and Hansen (1987)
				Ge Sequence
Ge	to	Ag	M1,E2	Biémont and Hansen (1986b)

ьe	to	Ag	MI,EZ	Blemont and Hansen (1986D)
Kr			E1	Fawcett and Bromage (1980)
RЬ			E1	O'Sullivan (1989)
Sr			E1	O'Sullivan and Maher (1989)
Zr			E1	Chaghtai et al. (1980)
Ru	to	Pd	E1	O'Sullivan et al. (1988a)

E1 Xe Breton et al. (1988) As Sequence As to Ag M1,E2 Biémont and Hansen (1986b) E1 Lotrian et al. (1980) As Kr E1 Fawcett and Bromage (1980) O'Sullivan and Maher (1989) Sr E1 Zr E1 Khan et al. (1980) Br to Mo E1 O'Sullivan (1989) Xe E1 Breton et al. (1988) Ru,Rh E1 O'Sullivan and Kane (1989) Se Sequence Se to Ag M1,E2 Biémont and Hansen (1986a) Coetzer et al. (1982) Breton et al. (1988) Kr E1 Xe E1 Ru, Rh, Pd E1 O'Sullivan et al. (1988b) Br Sequence Br to Xe M1,E2 Biémont et al. (1988) Кr E1 Blagoev (1981) Kr E1 Brandt et al. (1982) Kr E2 Fonseca and Campos (1982) Kr E1 Ward et al. (1985b) Kr E1 Schade et al. (1989) Costello and O'Sullivan (1984) Cd E1 Xe E1 Breton et al. (1988) Kr Sequence E1 Kr Brandt et al. (1982) Kr E1 Fonseca and Campos (1980) Kr to Mo E1 Sureau et al. (1984) Rh E1 Ceyzeriat et al. (1980) Cd E1 Costello and O'Sullivan (1984) Xe E1 Breton et al. (1988) Pr,Dy E1 Finkenthal et al. (1986) A) The experimental results Although the experimental determination of oscillator strengths mostly concerns the first ionization degrees (up to 8), some results have been obtained by beam-foil spectroscopy for Mo XIV (Denne and Poulsen,

results have been obtained by beam-foil spectroscopy for Mo XIV (Denne and Poulsen, 1981) and for I XXV (Johnson et al., 1985) in the copper sequence. In fact, the Cu, Zn and Ga sequences have been investigated mostly by beam-foil spectroscopy up to Kr VIII (Pinnington et al., 1981a,b; 1982, 1984; Bahr et al., 1982; Cederquist et al., 1984; Livingston et al., 1980). Some laser excitation results have been reported by Hannaford and Lowe (1983a) (Cu I) and by Ward et al. (1985b) (Kr II). Results obtained by different "classical" techniques (hook method, wall-stabilized arcs, shock tube, <u>Key</u>: A : Analysis essentially complete; B : the lowest configurations and portions of some higher ones known; C : two dozens or so levels known; D : about a dozen levels known; E : very fragmentary analysis, 2 to 5 levels known. Due to limited space available, the references used for preparing this table are not quoted here.





delayed coincidences,...) have been reported for Cu I, Cu II, Zn I, Ge II, Kr I, Kr II, As I (see e.g. Miller and Bengston, 1980; Lotrian et al., 1980; Ueda et al., 1981; Kono and Hattori, 1982, Fonseca and Campos, 1980, 1982; Blaqoev, 1981; Zettl et al., 1984).

### B) The theoretical results

The choice of a theoretical method for performing calculations of atomic structure in heavy elements is essentially determined by a compromise between the wanted accuracy and the computer time needed.

Large MCDF codes (e.g. Desclaux, 1975; Grant et al., 1980) are available and allow a "fully" relativistic treatment of the heavy atoms or ions but their use is, in practice, limited by the "complexity" and the number of configurations to be introduced in the calculations. Nevertheless, extensive results have been obtained recently along the zinc sequence (Biémont, 1989) and more limited data along the Ge, As and Se sequences (O'Sullivan et al., 1988a,b; O'Sullivan, 1989; O'Sullivan and Maher, 1989; O'Sullivan and Kane, 1989).

The model potential (PM) techniques look also promising and their potential has been discussed recently by Hibbert (1989). Among the recent results obtained by such techniques, let us quote here the data published by Migdalek (1983), by Aashamar et al. (1983) (Ga sequence) and by Victor and Taylor (1987) (Cu and Zn sequences).

parametric potential The method of Klapisch et al. (1977) in its relativistic version (RPPT) has been used frequently as support for experiments concerning moderately or highly ionized elements (see e.g. Mandelbaum et al., 1983; Wyart et al., 1984; Finkenthal et al., 1986; Ryabtsev and Wyart, 1987).

The relativistic Hartree-Fock (HXR, HFR) method of Cowan (1981b) is among the techniques widely used both for allowed (see e.g. Biémont, 1988; Biémont and Quinet, 1989; Biémont et al., 1989a) and for forbidden transitions (see e.g. Biémont and Hansen, 1986a,b; 1987; 1988) while the non relativis-tic MCHF technique has provided results for zinc-like (Biémont and Godefroid, 1980) and gallium-like atoms (Brage et al., 1987).

A limited comparison between the HFR, MCDF, and PM results obtained along the zinc isoelectronic sequence is presented in Table 3.

Table 3 - Comparison between recently reported f values along the zinc sequence; HFR : Biémont et al. (1989a); PM : Victor and (1987); MCDF-EAL : this work Taylor (Babushkin Gauge).

452	<sup>1</sup> s	-	4s4p	1 <sub>P</sub> 0
45	3	_	434P	

Rb	VIII	Y	X	Мо	XIII	Sn	XXI	Xe	XXV
				101100000	and the second se	and the second second			and the second sec

HFR	1.718	1.629	1.510	1.282	1.205
PM	1.722	1.718	1.489	-	-
MCDF	1.692	1.615	1.509	1.291	1.217

$$4s4p P^{0} - 4p^{2} S$$

0.211 0.199 0.185 0.164 0.155 HFR 0.213 0.207 0.185 PM 0.210 0.199 0.186 0.165 0.160 MCDF

$$4s4p P^{0} - 4p^{2} D$$

HFR 0.169 0.171 0.166 0.136 0.115 PM 0.262 0.255 0.316 0.169 0.168 0.162 0.131 0.115 MCDF

HFR 1.659 1.527 1.361 1.061 0.975 PM 1.683 1.630 1.384 -1.656 1.540 1.386 1.095 1.006 MCDF

$$4s4p P^{0} - 4s4d D^{3}$$

HFR	1.038	0.974	0.884	0.708	0.639
PM	1.010	0.936	0.880	-	
MCDF	1.020	0.963	0.878	0.705	0.648

$$4s4p P^{0} - 4p^{2} P^{0}$$

HFR	0.465	0.432	0.339	0.344	0.315
PM	0.577	0.576	0.502	-	-
MCDF	0.343	0.344	0.332	0.299	0.287

The agreement between MCDF and HFR data is excellent for the ions Rb VIII to Xe XXV. Larger differences are observed between MCDF (or HFR) and PM data particularly for the

4s4p <sup>1</sup>P<sup>0</sup> - 4p<sup>2</sup> <sup>1</sup>D transition. We shall now briefly spotlight some general characteristics which are encountered when calculations of atomic structure and intensities are carried out along isoelectronic sequences. These general considerations will be illustrated by some specific examples chosen among recent HXR and MCDF results reported along the zinc sequence for the ions Rb VIII to W XLV (Biémont, 1989; Biémont et al.. 1989a) :

- change of the percentage compositions (and the corresponding difficulties for the designation of the levels) when Z increases along the sequence. The HXR composition is illustrated for three specific cases on



Fig. 1. Percentage composition (jj coupling) of the 4p4f(1/2, 7/2), levels along the zinc isoelectronic sequence.



level. 4f2 3F is written for 4f<sup>2</sup> <sup>3</sup>F.

Figures 1 to 3 which show respectively the strong interaction between  $4p4f(1/2, 7/2)_3$  and  $4p4f(1/2, 5/2)_3$  at around  $q = Z-N+1 \ \ 15$  (Fig. 1), the smooth change of composition for the  $4f^2 \ ^3F_4$  level (Fig. 2) (more correctly designated as  $4f^2 \ ^1G_4$  at  $q \ \ 30$ ) and the sudden change of composition for  $4p4f(3/2, 5/2)_4$  (which is better designated as  $4d^2 \ ^3F_4$  at  $q \ \ 16$  (Fig. 3);



Fig. 3. Same as Fig. 1 for the 4p4f(3/2,  $5/2)_4$  or  $(4d^2 \ ^3F_4)$  level. 4d2 3F is written for  $4d^2 \ ^3F$ .

- the increasing importance of relativistic effects when Z and the ionization degree increase along the sequence and, consequently, the need for a "fully" relativistic approach for very high electric charges. This is illustrated on Figures 4 and 5 which show the differences (in %) between MCDF-EAL and HXR ab initio values for the even levels belonging to the configurations  $4s^2 + 4p^2 +$  $4d^2 + 4f^2 + 4s4d + 4p4f$  in In XX and Tm XL respectively. It appears (Figure 5) that the differences tend to become systematically positive for Tm XL due to the fact that the relativistic effects are considered in a more accurate way in the MCDF approach than in the HXR method;

- the appearance of level crossings. This is illustrated on Fig. 6 in the case of the  $4d^2$  + 4p4f configurations for the ions Rb



Fig. 4. Comparison between MCDF-EAL and HXR ab initio values of the even parity energy levels of In XX (configurations  $4s^2 + 4p^2 + 4d^2 + 4f^2 + 4s4d + 4p4f$ ).



Fig. 5. Same as Fig. 4 for Tm XL.

VIII to Cs XXVI. The level crossings are frequently responsible for noticeable irregularities appearing in the curves showing the behaviour of f values as a function of Z. Some examples have been illustrated recently by Biémont and Quinet (1989) in the case of the gallium sequence.



Fig. 6. Zinc isoelectronic sequence : theoretical energy level values divided by q = Z = N + 1 as a function of q.

RADIATIVE LIFETIMES AND OSCILLATOR STRENGTHS FOR HEAVY REFRACTORY ELEMENTS (Z > 39)

It has appeared, in the beginning of the eighties, that our knowledge of the radiative transition probabilities for the heavy refractory elements was still very incomplete. This situation was essentially related to the fact that it is very difficult by conventional thermal means to produce a vapour pressure sufficient for the measurement of radiative lifetimes and, on the other hand, that the calculation of accurate theoretical data for these neutral and singly elements requires ionized a detailed consideration of correlation and of relativistic effects. The use of selective laser excitation

by different groups (Clayton, Berlin, Kiel, Wisconsin, Aarhus, ...) combined with branching ratio measurements (mostly by Fourier transform spectroscopy) has given a definitive boost to the determination of accurate transition probabilities for these elements.

We have summarized in Table 4 the different lifetime measurements concerning neutral or singly ionized elements present in the solar photospheric spectrum obtained with techniques based on laser excitation. These elements have been classified as refractory or siderophile according to the classification adopted for the meteorites. This table updates a review published by Richter (1984) a few years ago.

Table 4 - Heavy refractory (and siderophile) elements : lifetime and branching ratio determinations (period 1980-1989). In the last column, are given the solar abundances as derived from the lifetime measurements (starred reference) and the difference ( $\Delta$ ) between photospheric and meteoritic abundances (in the usual logarithmic scale where the hydrogen abundance is = 12.00).

# A) Refractory

Reference	Solar abundance
Y I-YII *Hannaford et al.(1982) Hannaford and Lowe (1982) Rudolph and Helbig (1982b) Hannaford and Lowe (1983a) Gorshkov and Komarovskii (1	*2.18 <u>+</u> 0.12 (Υ Ι) *2.25 <u>+</u> 0.03 (Υ ΙΙ) Δ = 0.03 986)
Wännström et al. (1988)	
Zr I-Zr II	[
* Biémont et al. (1981)	*2.57 <u>+</u> 0.07 (Zr I)
Hannaford and Lowe (1981)	*2.56+0.05 (Zr II)
Poulsen et al. (1981)	$\Delta = 0.04$
Duquette et al. (1982a)	
Poulsen et al. (1982)	
Rudolph and Helbig (1982a)	
Hannaford and Lowe (1983a)	
Nb I-Nb II	
Duquette and Lawler (1982)	*1.42+0.06 (Nb II)
Kwiatkowski et al. (1982a)	$\Delta = 0.02$
Rudolph and Helbig (1982c)	
*Hannaford et al. (1985)	
Ma I Ma I	T
$\frac{MO}{1-MO}$	¥1 92,0 05 (Mo I)
Kwistkowski et al. (1981)	Λ = 0.04
Rudalah and Halbig (1992a)	<b>G</b> = 0.04
*Different at al (1993)	
Happenford and Lowe (1993a)	
Hannaford and Lowe (1983b)	
Schoebage et al. (1983)	
Whaling at al (1984)	
Plaklatking and Varalainen	(1985)
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Whaling et al. (1986) Whaling and Brault (1988) Ru I \*Biémont et al. (1984) \*1.84+0.07 (Ru I) Salih and Lawler (1985)  $\Delta = 0.02$ Rh I \*Kwiatkowski et al.(1982a) \*1.12+0.12 (Rh I) ∆ = 0.03 Salih et al. (1983) Duquette and Lawler (1985) Ba II Gaillard et al. (1982) Nd I - Nd II Marek and Stahnke (1980) \*1.47+0.07 (Nd II) \*Ward et al. (1984) Δ= 0.00 Ward et al. (1985a) Sm I - Sm II \*1.01+0.06 (Sm II) Brand et al. (1980) Vogel et al. (1988) **△** = 0.04 \*Biémont et al. (1989b) Eu I – Eu II Klimkin and Prokofiev(1980) \*0.51+0.08(Eu II) Meyer et al. (1981)  $\Delta = 0.03$ \*Biémont et al. (1982b) Karner et al. (1982) Arnesen et al. (1983) Gd I - Gd II Marek and Stahnke (1980) \*1.12+0.04 (Gd II) \*Bergström et al. (1988) **∆** = 0.05 Er I - Er II Marek and Stahnke (1980) \*0.93+0.06 (Er II) Gorshkov and Komarovskii (1981) 🛆 = 0.02 Bentzen et al. (1982) \*Biémont and Youssef (1984) Lu I Kwiatkowski et al. (1980) Hf I Duquette et al. (1982b) W I - W II\*1.11+0.15 (W I) Duquette et al. (1981a) Kwiatkowski et al. (1982b)  $\Delta = 0.43$ \*Holweger and Werner (1982) Obbarius and Kock (1982) Kwiatkowski et al. (1984a) Pleklotkina and Verolainen (1985) Den Hartog et al. (1987) Os I \*Kwiatkowski et al.(1984b) \*1.45+0.10 (Os I) A= 0.07 \*Gough et al. (1983) Ir I \*1.38+0.05 (Ir I) Hannaford and Lowe (1983a) Δ= 0.01 \*Youssef and Khalil (1988) \*Gough et al. (1982) \*1.74+0.05 (Pt I) Hannaford and Lowe (1983a)  $\Delta = 0.06$ \*Youssef and Khalil (1987) B) Siderophile Pd I \*1.69+0.04 (Pd I) \*Biémont et al. (1982a)  $\Delta = 0.30$ 

Au I

\*Hannaford et al. (1981) \*1.3+0.12 (Au I) Bezuglov et al. (1982) Δ= 0.30 Hannaford and Lowe (1983a)

From the numerous comparisons performed with the different sets of lifetimes, it has appeared that :

- the results obtained with laser excitation are much more accurate than those published previously (i.e. basically before 1980) and they differ sometimes considerably from them;

- the different sets of lifetimes published after 1980 are generally consistent within a few percent.

These two conclusions are illustrated on Figures 7 and 8 where we have plotted the ratio between different sets of lifetimes for a number of selected elements. As seen on Fig. 7, this ratio is close to unity when comparing two sets of measurements obtained



Fig. 7. Ratio of lifetime measurements for different refractory elements. We have plotted  $\tau$  (HL)/ $\tau$ (DL) for Zr I,  $\tau$ (KZ)/ $\tau$ (DS) for Nb I,  $\tau$ (KM)/ $\tau$ (DU) for W I and  $\tau$ (BG)/ $\tau$ (VE) for Sm II.

The symbols have the following meaning : KZ : Kwiatkowski et al. (1982a); DS : Duquette and Lawler (1982); KM : Kwiatkowski et al. (1982b):DL : Duquette et al. (1982a); BG : Biémont et al. (1989b); VE : Vogel et al. (1988); HL : Hannaford and Lowe (1981); DU : Duquette et al. (1981a). from laser experiments while the dispersion of the results is much greater (Fig. 8) when comparing these lifetimes with those obtained by different techniques.

We give in the last column of Table 4 the solar photospheric abundances as they have been deduced on the basis of the lifetime measurements quoted in the same table (starred references) and also the difference ( $\Delta$ , on the logarithmic scale) between photospheric and meteoritic abundances, the meteoritic results being taken from a recent compilation by Anders and Grevesse (1989).

As appears from Table 4, there is now, on the basis of the new atomic data, agreement within the respective error bars between the carbonaceous chondrites and the photosphere for all the heavy refractory elements if we except W and Au. Though the large difference observed for W could be real according to the careful solar analysis carried out by Holweger and Werner (1982), it is more probable that W is only a small contributor to the two weak absorption lines used for the solar analysis. Similar conclusions are valid for Au.





KM : Kwiatkowski et al. (1981); BL : Baumann et al. (1978); RA : Ramanujam and Andersen (1978); GH : Gough et al. (1983); BG : Biémont et al. (1989b); B : Blagoev (1978).

The overall agreement sun-meteorites. which is now observed for all these elements. was hidden until recently because large uncertainties were affecting the atomic data (mostly the oscillator strengths) used for the solar analyses.

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