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Relativistic K shell decay rates and fluorescence yields for Zn, Cd and Hg

C. Casteleiro¹, F. Parente³, P. Indelicato², and J. P. Marques¹

¹ Centro de Física Atómica e Departamento Física, Faculdade de Ciências, Universidade de Lisboa, Campo Grande, Ed. C8, 1749-016 Lisboa, Portugal, e-mail: jmmarques@fc.ul.pt

² Laboratoire Kastler Brossel, École Normale Supérieure; CNRS; Université P. et M. Curie - Paris 6 Case 74; 4, place Jussieu, 75252 Paris CEDEX 05, France, e-mail: paul.indelicato@spectro.jussieu.fr

³ Centro de Física Atómica da Universidade de Lisboa e Departamento Física da Faculdade de Ciências e Tecnologia da Universidade Nova de Lisboa, Monte da Caparica, 2825-114 Caparica, Portugal, e-mail: facp@fct.unl.pt

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Abstract. In this work we use the multiconfiguration Dirac-Fock method to calculate the transition probabilities for all possible decay channels, radiative and radiationless, of a K shell vacancy in Zn, Cd and Hg atoms. The obtained transition probabilities are then used to calculate the corresponding fluorescence yields which are compared to existing theoretical, semi-empirical and experimental results.

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1 Introduction

The fluorescence yield of an atomic shell or subshell is defined as the probability that a vacancy in that shell or subshell is filled through a radiative transition. An atom with a vacancy is in an excited one-hole state. The fluorescence yield of an atomic shell or subshell is given by

$$\omega_i = \frac{\Gamma_{R_i}}{\Gamma_{R_i} + \Gamma_{NR_i}} \quad (1)$$

where Γ_{R_i} is the radiative width of an one-hole in that shell or subshell state and Γ_{NR_i} is the radiationless width of the same state.

A good knowledge of K X-ray fluorescence yields is important for the interpretation of a large variety of measurements in fields such as nuclear, atomic, molecular and plasma physics as well as in medical physics, engineering and astrophysics [1].

Fluorescence yields have been object of intense research for several decades, both experimentally and theoretically, and a large number of articles have been published on this subject, describing different methods of measurement and calculation. In 1972, Bambynek *et al.* [2] published a review article with a selection of the most reliable experimental ω_K values for 25 elements with $13 \leq Z \leq 92$. In 1979, Krause [3] published a compilation of ω_K values for all elements in the range $5 \leq Z \leq 110$, using for that both experimental and theoretical data. More recently (1994), Hubbell *et al.* [4] compiled experimental

and semi-empirical K shell fluorescence yields for elements with $11 \leq Z \leq 99$. Theoretical values of ω_K for elements with $4 \leq Z \leq 54$ were calculated with the Hartree-Fock-Slater method by McGuire in 1969 and 1970 [5,6] and Walthers and Balla in 1971 [7]. In 1980 Chen *et al.* [8] performed a relativistic Dirac-Hartree-Slater calculation of ω_K for selected elements with $18 \leq Z \leq 96$. Although these data are already several decades old, they are still widely used for comparison with experimental results.

In the last few years there has been a considerable increase in the number of experimental high precision measurements of the K shell fluorescence yields for many elements [9,10,11,12,13,14,15,16,17]. The results show several discrepancies when compared with theoretical and semi-empirical data. For Cd, for example, four experimental results have been published in this decade [11,13,14,17]. Yet, no relativistic calculation has been performed so far for this element. So, we decided to perform this calculation with state of art methods. We extended this calculation to Zn and Hg, which have electronic configurations similar to Cd. For Zn and Hg, the old relativistic results of Chen [8], using a simpler Dirac-Slater approach, can be used for comparison, and four experimental results have been published in the Zn case [11,13,15,16], and one for Hg [10], in the last eight years.

In this work we use the Multi-Configuration Dirac-Fock code of Desclaux and Indelicato [18,19], which includes QED corrections, to calculate the K-shell fluorescence yields for the elements Zn, Cd and Hg. This is, to our knowledge, the first full relativistic calculation of this quantity for these elements. We note, that for Zn and

Hg, Chen *et al.* presented relativistic values, but they were obtained using a simpler relativistic Dirac-Slater approach [8].

2 Radiative and Radiationless Transitions

A general relativistic program developed by Desclaux and Indelicato [19] was used to compute the energies and wavefunctions, as well as radiative and radiationless transition probabilities concerning the K fluorescence yields for Zn, Cd and Hg. Details on the Hamiltonian and the processes used to build the wavefunctions can be found elsewhere [18, 20, 21, 22].

For the radiative transition probabilities the code implements exact relativistic formulas [23, 24, 25, 18, 26]. For these transitions, the so-called Optimized Levels (OL) method was used to determine the wavefunction and energy for each state involved. This method allows for a full relaxation of both initial and final states, providing much better energies and wavefunctions. However, spin-orbitals in the initial and final states are not orthogonal, since they have been optimized separately. To account for the wavefunctions nonorthogonality, the code uses the formalism described by Löwdin [27].

In what concerns radiationless transitions, we assume that the creation of the inner-shell hole is independent of the decay process. In this way the primary ejected electron in the inner-shell ionization process does not interact with the Auger electron and radiationless transitions probabilities are calculated from perturbation theory. The continuum electron wavefunctions were obtained by solving the DF equations with the same atomic potential of the initial state. With this treatment, the continuum wavefunctions are made orthogonal to the initial bound state wavefunctions and the orthogonality of the wavefunctions is assured. No orbital relaxation is included in this calculation. The interaction potential between the core electron and the Auger electron include both Coulomb and magnetic parts. The Auger interaction two-electron operator is chosen to be the Breit operator [28].

3 Results and Discussion

In Table 1 we list the radiative transition probabilities for all electron transitions from higher subshells that could fill a vacancy in the K shell for Zn, Cd and Hg. In the first column the final state vacancy subshell is identified. Our results for the total radiative transition probabilities agree closely with the ones obtained by Scofield [29], which are shown in the same table.

In Table 2 we present K shell radiationless transition probabilities from an initial state with a vacancy in the K shell to a final state with two vacancies for Zn, Cd and Hg. In the first column, for example, L1-LMN means a final state with a first vacancy in the $L1$ subshell and the second vacancy in either the L , M , or N shells. The disagreement between our results and the ones from Chen *et al.* [30] for

Table 1. K shell radiative transition probabilities for Zn, Cd and Hg (in a.u.). In the first column we identify the final state subshell vacancy.

	Zn	Cd	Hg
L1	2.59E-08	3.23E-06	7.852E-04
L2	8.77E-03	6.47E-02	5.577E-01
L3	1.71E-02	1.22E-01	9.462E-01
M1	5.24E-09	8.73E-07	2.506E-04
M2	1.19E-03	1.14E-02	1.077E-01
M3	2.33E-03	2.21E-02	2.084E-01
M4	1.59E-11	8.10E-05	2.766E-03
M5	2.17E-06	1.13E-04	3.396E-03
N1	3.50E-10	1.79E-07	7.002E-05
N2		2.01E-03	2.623E-02
N3		3.88E-03	5.165E-02
N4		1.15E-05	7.557E-04
N5		1.58E-05	9.235E-04
N6			1.021E-06
N7			1.152E-06
O1		1.70E-08	1.440E-05
O2			4.831E-03
O3			9.090E-03
O4			9.090E-03
O5			1.035E-04
O6			1.703E-06
Total	2.94×10^{-02}	2.26×10^{-01}	$1.93 \times 10^{+00}$
Scofield [29]	2.90×10^{-02}	2.26×10^{-01}	$1.94 \times 10^{+00}$

Zn and Hg, presented in the same table, result, naturally, from the different methods employed, namely a full Dirac-Fock calculation and an approximate Dirac-Hartree-Slater calculation in Chen's work.

In Table 3 we compare the fluorescence yields obtained in this work for Zn, Cd and Hg with other theoretical and experimental data. We note that the present relativistic results are in general lower than existing non-relativistic theoretical data and are close to Chen *et al.* [8] relativistic values. In the same table we also list the semi-empirical values of Bambynek *et al.* [2], Krause [3], and Hubbell *et al.* [4]. For Zn, these values lie systematically below most theoretical as well as experimental data.

In Figure 1 existing theoretical and experimental fluorescence yield values for Zn, Cd and Hg are graphically compared with the results obtained in this work. In what concerns experimental data, our results agree, within the error bars, with the more recent experimental values except for the Gudennavar *et al.* Zn value. We note that the pre-1960 experimental results for Hg present much smaller error bars than the more recent ones.

We can conclude that the precision of existing experimental data is not good enough to distinguish between the different theoretical values, thus calling for more precise experiments, in order to stimulate new calculations.

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Table 2. K shell radiationless transition probabilities for Zn, Cd and Hg (in a.u.). For example, L1-LMN stands for a final state with one vacancy in the L1 subshell and other vacancy in either of the L, M, and N shells. The initial state has one vacancy in the K shell.

	Zn		Cd		Hg
L1-LMN	1.10×10^{-02}	L1-LMNO	1.63×10^{-02}	L1-LMNOP	3.74×10^{-02}
L2-LMN	1.53×10^{-02}	L2-LMNO	1.53×10^{-02}	L2-LMNOP	1.78×10^{-02}
L3-LMN	4.78×10^{-03}	L3-LMNO	4.15×10^{-02}	L3-LMNOP	8.83×10^{-03}
M1-MN	1.47×10^{-04}	M1-MNO	4.34×10^{-04}	M1-MNOP	1.50×10^{-03}
M2-MN	2.06×10^{-04}	M2-MNO	3.87×10^{-04}	M2-MNOP	7.30×10^{-04}
M3-MN	2.57×10^{-05}	M3-MNO	2.43×10^{-04}	M3-MNOP	6.08×10^{-04}
M4-MN	8.91×10^{-07}	M4-MNO	2.60×10^{-04}	M4-MNOP	3.34×10^{-05}
M5-MN	2.15×10^{-07}	M5-MNO	7.01×10^{-06}	M5-MNOP	2.93×10^{-05}
N1-N	1.48×10^{-07}	N1-NO	1.09×10^{-05}	N1-NOP	7.82×10^{-05}
		N2-NO	2.23×10^{-06}	N2-NOP	3.47×10^{-05}
		N3-NO	1.34×10^{-06}	N3-NOP	2.60×10^{-05}
		N4-NO	8.28×10^{-08}	N4-NOP	1.85×10^{-06}
		N5-NO	2.12×10^{-08}	N5-NOP	2.85×10^{-05}
		O1-O	2.37×10^{-08}	N6-NOP	1.12×10^{-08}
				N7-NOP	2.23×10^{-08}
				O1-OP	2.29×10^{-06}
				O2-OP	7.91×10^{-07}
				O3-OP	3.91×10^{-07}
				O4-OP	8.22×10^{-09}
				O5-OP	2.83×10^{-09}
				P1-P	9.01×10^{-09}
Total	3.14×10^{-02}		7.44×10^{-02}		6.71×10^{-02}
Chen <i>et al.</i> [30]	3.04×10^{-02}				7.48×10^{-02}

Table 3. K shell fluorescence yields for Zn, Cd and Hg.

	Zn	Cd	Hg
Theoretical			
This work	0.485	0.842	0.966
McGuire [6]	0.499		
Kostroun [31]	0.482	0.855	
Walters <i>et al.</i> [7]	0.501	0.871	
Chen [8]	0.488		0.962
Semi empirical			
Hubbel [4]	0.469	0.843	0.966
Bambynek [2]	0.479	0.841	0.965
Krause [3]	0.474	0.836	0.980
Experimental			
Broyles <i>et al.</i> [40]			0.946 ± 0.008
Roos [32]	0.446 ± 0.012		
Patronis <i>et al.</i> [33]	0.44 ± 0.02		
Nall <i>et al.</i> [41]			0.952 ± 0.003
Arora <i>et al.</i> [34]	0.49 ± 0.02		
Bhan <i>et al.</i> [37]		0.853 ± 0.075	
Al-Nasr <i>et al.</i> [38]		0.874 ± 0.048	
Sidhu <i>et al.</i> [42]			0.980 ± 0.009
Piuos <i>et al.</i> [35]	0.471 ± 0.025		
Balakrishna <i>et al.</i> [39]		0.853 ± 0.041	
Durak <i>et al.</i> [36]			0.971 ± 0.036
Simşek <i>et al.</i> [16]	0.482 ± 0.022		
Durak <i>et al.</i> [15]	0.482 ± 0.032		
Simşek <i>et al.</i> [17]		0.860 ± 0.045	
Özdemir <i>et al.</i> [14]		0.852 ± 0.042	
Gudennavar <i>et al.</i> [13]	0.464 ± 0.010	0.855 ± 0.017	
Yashoda <i>et al.</i> [11]	0.471 ± 0.018	0.837 ± 0.029	
Apaydin <i>et al.</i> [10]			0.984 ± 0.086

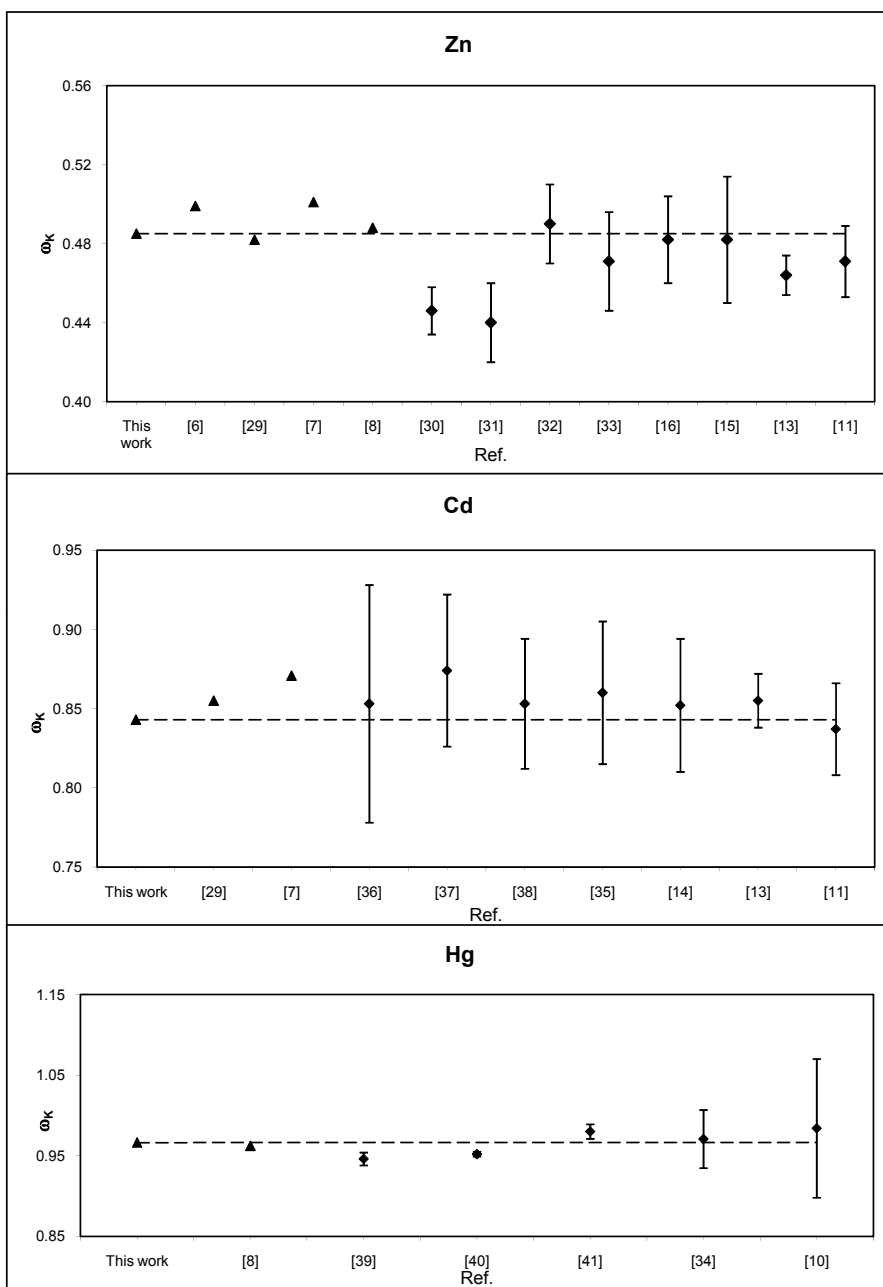


Fig. 1. Comparison between experimental (\blacktriangle) and calculated (\blacklozenge) K shell fluorescence yields for Zn, Cd and Hg.