Published in "Physica Scripta 2011(T144): 014021, 2011"
which should be cited to refer to this work.

Lifetimes of doubly *K*-shell ionized states

M Polasik[1], K Słabkowska[1], K Kozioł[1], J Starosta[1], J-Cl Dousse[2], J Hoszowska[2] and J Rzadkiewicz[3,4]

[1] Faculty of Chemistry, Nicholas Copernicus University, 87-100 Toruń, Poland
[2] Institute of Physics, Jan Kochanowski University, 25-406 Kielce, Poland
[3] The Andrzej Soltan Institute for Nuclear Studies, 05-400b Otwock-Świerk, Poland
[4] Institute of Plasma Physics and Laser Microfusion, Hery 23, 01-497 Warsaw, Poland

E-mail: mpolasik@uni.torun.pl

Abstract
The present work provides a reliable interpretation of the $K^h\alpha_1/K^h\alpha_2$ intensity ratios and an explanation of the lifetime values for K-shell hollow atoms based on an advanced theoretical analysis (using extensive multiconfiguration Dirac–Fock calculations with the inclusion of the transverse Breit interaction and quantum electrodynamics corrections). It was found that, as a result of closing the $K^h\alpha_1$ de-excitation channel in the pure LS coupling scheme, the $K^h\alpha_1/K^h\alpha_2$ intensity ratio changes with the atomic number from small values (for the LS coupling limit at low Z) to about 1.5–1.6 (for the j–j coupling limit at high Z). However, closing the $K^h\alpha_1$ de-excitation channel (due to the domination of the pure LS coupling for the low-Z atoms) does not enlarge the lifetimes of hollow atoms.

PACS numbers: 32.30.Rj, 32.70.Cs, 32.80.Fb, 32.70.Jz

1. Introduction

In recent decades, the hypersatellite lines in x-ray spectra were measured by using various experimental techniques. High-resolution measurements of such spectra [1–4] give direct access to information on the natural width of the hypersatellite lines and the lifetimes of the K-shell hollow (i.e. doubly K-shell ionized) atoms.

Recently, the $K^h\alpha_{1,2}$ hypersatellite lines in x-ray spectra of Zr, Nb, Mo and Pd targets bombarded with 250 MeV carbon and 360 MeV oxygen ions have been measured with high-resolution diffraction spectrometry by Rzadkiewicz *et al* [2]. The de-excitation of the studied doubly K-shell ionized state via the spin flip in the $K^h\alpha_1$ transition is forbidden in the pure LS coupling scheme ($^1S_0 \rightarrow {}^3P_1$) but allowed in j–j coupling (see figure 1). As a result, the $K^h\alpha_1/K^h\alpha_2$ intensity ratio changes with the atomic number (see figure 2) from small values (for the LS coupling limit at low Z) to about 1.5–1.6 (for the j–j coupling limit at high Z).

In a work by Rzadkiewicz *et al* [2], the experimental widths of $K^h\alpha_{1,2}$ lines have been compared with two formulae: the first one—statistical (proposed by Mossé *et al* [7])—is based on the assumption $\Gamma_{KK} \approx 2\Gamma_K$ and the second one (phenomenological—proposed in [2]) takes into account the possibility of enlarging the lifetime of the K^{-2} hole state (and correspondingly decreasing the natural width of the hypersatellite line) as a result of closing the $K^h\alpha_1$ de-excitation channel in the pure LS coupling scheme. However, it was found that the experimental natural widths of the $K^h\alpha_1$ and $K^h\alpha_2$ lines significantly exceed the values obtained from the formula proposed by Rzadkiewicz *et al* [2], but are in good agreement with the simple statistical formula. Moreover, the experimental lifetimes of the double K-hole states (deduced from their measured natural widths) are close to those predicted by the statistical formula, but significantly shorter than the values determined by means of the formula proposed by Rzadkiewicz *et al* [2].

The present work provides a reliable interpretation of the $K^h\alpha_1/K^h\alpha_2$ intensity ratios and an explanation of the lifetime values for the K-shell hollow atoms based on the advanced theoretical analysis (using extensive multiconfiguration Dirac–Fock (MCDF) calculations with the inclusion of the transverse Breit interaction and quantum electrodynamics (QED) corrections [8]).

2. Results and discussion

The proper description of the atomic state requires applying an intermediate coupling (i.e. mixing of configuration state

Table 1. Coefficients for $(1s^{-1}2p^{-1})\,{}^1P_1$ and $(1s^{-1}2p^{-1})\,{}^3P_1$ states in the upper and lower final $(1s^{-1}2p^{-1})$ states (see equations (1) and (2)), $(1s^{-1}2p^{-1})\,{}^1P_1$ contributions for upper and lower final states, and $K^h\alpha_1/K^h\alpha_2$ intensity ratios for selected atoms.

Atoms	Z	Coefficients for the final $(1s^{-1}2p^{-1})$ states		${}^1P_1$ state contribution for the upper state	${}^1P_1$ state contribution for the lower state	${}^1P_1$ state contributions ratio	$K^h\alpha_1/K^h\alpha_2$ intensity ratio
		a	b				
–	0	1.0	0.0	1.0	0.0	0.0	–
Ne	10	0.999 936	0.011 309	0.999 872	0.000 128	0.000 128	–
Ar	18	0.9925	0.1220	0.9851	0.0149	0.015	0.014
Zn	30	0.8509	0.5254	0.7240	0.2760	0.381	0.377
Kr	36	0.7500	0.6614	0.5626	0.4374	0.778	0.793
Zr	40	0.7034	0.7108	0.4948	0.5052	1.021	1.041
Mo	42	0.6856	0.7279	0.4701	0.5299	1.127	1.145
Pd	46	0.6584	0.7527	0.4334	0.5666	1.307	1.313
Rn	86	0.5867	0.8098	0.3442	0.6558	1.906	1.610
No	102	0.5819	0.8132	0.3386	0.6614	1.953	1.490
–	∞	0.5773	0.8165	0.333	0.6667	2.0	–

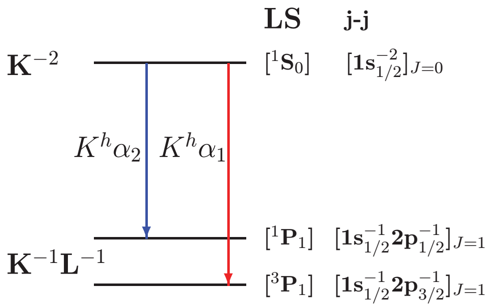

Figure 1. $K^h\alpha_{1,2}$ hypersatellite x-ray transitions.

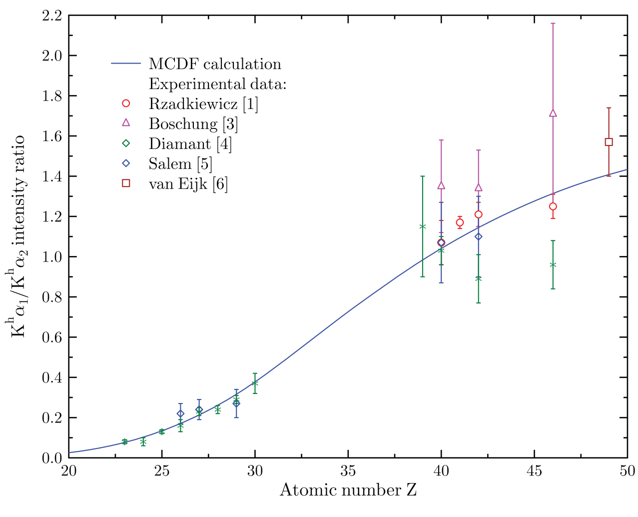

Figure 2. Theoretical predictions for $K^h\alpha_1/K^h\alpha_2$ intensity ratios compared with some experimental data [1, 3–6].

functions of a certain J). Therefore, to describe the $K^{-1}L^{-1}$ hole states (see figure 1) for all the studied atoms, we can apply the intermediate coupling for j–j or LS basis. For the j–j basis (typical in MCDF calculations) the final upper and the final lower states are given as a combination of $[1s^{-1}_{1/2}2p^{-1}_{1/2}]_{J=1}$ and $[1s^{-1}_{1/2}2p^{-1}_{3/2}]_{J=1}$ states. In this case, transitions from $[1s^{-2}_{1/2}]_{J=0}$ to both states ($[1s^{-1}_{1/2}2p^{-1}_{1/2}]_{J=1}$ and $[1s^{-1}_{1/2}2p^{-1}_{3/2}]_{J=1}$) are allowed. For heavy atoms the intermediate coupling is well enough approximated by pure j–j coupling. In this coupling, the $K^h\alpha_1$ transition should be two times stronger than the $K^h\alpha_2$ transition (two times more $2p_{3/2}$ electrons than $2p_{1/2}$). However, the energy of the $K^h\alpha_1$ transition is significantly higher than that of the $K^h\alpha_2$ transition, and moreover, $2p_{3/2}$ and $2p_{1/2}$ orbitals differ significantly enough. Therefore, for heavy atoms the $K^h\alpha_1/K^h\alpha_2$ intensity ratio does not amount to 2, but is a bit smaller, i.e. it amounts to about 1.5–1.6 (see column 8 in table 1). For the LS basis in the intermediate coupling, the final upper state (commonly labeled as ${}^1P_1$, see figure 1) is given as

$$a\cdot|(1s^{-1}2p^{-1})\,{}^1P_1\rangle + b\cdot|(1s^{-1}2p^{-1})\,{}^3P_1\rangle \tag{1}$$

and the final lower state (commonly labeled as ${}^3P_1$) is given as

$$b\cdot|(1s^{-1}2p^{-1})\,{}^1P_1\rangle - a\cdot|(1s^{-1}2p^{-1})\,{}^3P_1\rangle. \tag{2}$$

The coefficients a and b for selected atoms are presented in columns 3 and 4 in table 1. As can be seen, for light atoms the intermediate coupling is well approximated by the LS coupling (i.e. the mixing coefficients differ from each other, e.g. for Ar). However, in the LS coupling only $(1s_{1/2})^{-2}\,{}^1S_0 \rightarrow (1s^{-1}2p^{-1})\,{}^1P_1$ transition is allowed, but $(1s_{1/2})^{-2}\,{}^1S_0 \rightarrow (1s^{-1}2p^{-1})\,{}^3P_1$ is forbidden. Thanks to the intermediate coupling in the LS basis, the $K^h\alpha_1/K^h\alpha_2$ intensity ratio can be estimated as a contributions ratio of the pure $(1s^{-1}2p^{-1})\,{}^1P_1$ state in the final lower state (responsible for $K^h\alpha_1$ transition) and in the final upper state (responsible for $K^h\alpha_2$ transition)—see columns 7 and 8 in table 1.

From table 1, one can also see that going from heavy to light atoms closing the $K^h\alpha_1$ channel is accompanied by strengthening of the $K^h\alpha_2$ channel (with fully seizing the contribution of the closed $K^h\alpha_1$ channel). It is a fact that the sum of the pure $(1s^{-1}2p^{-1})\,{}^1P_1$ state contributions for both final states is constant and amounts to 1.0 (see the sums of values in columns 5 and 6 of table 1).

Based on the results of MCDF calculations [10, 11], the lifetimes (and radiative and total natural widths) of K^{-2} and K^{-1} hole states and widths of the $K^h\alpha_1$ and $K^h\alpha_2$ hypersatellite lines have been evaluated (see table 2). It was found that with a change of the atomic number Z only the relative roles of both channels are changed and closing the $K^h\alpha_1$ channel for lighter atoms does not lead to increasing

Table 2. The K^{-2} and K^{-1} hole state lifetimes, the K^{-2}/K^{-1} natural width ratios and the natural linewidths for $K^{\mathrm{h}}\alpha$ lines for selected atoms.

Atom	Z	Lifetimes (s)		K^{-2}/K^{-1} natural width ratio		Linewidth for $K^{\mathrm{h}}\alpha$ lines (eV)	
		K^{-2}	K^{-1}	Radiative	Total	$K^{\mathrm{h}}\alpha_1$	$K^{\mathrm{h}}\alpha_2$
Ne	10	4.491×10^{-14}	1.725×10^{-13}	3.336	2.04	0.89	0.89
Ar	18	4.258×10^{-16}	1.010×10^{-15}	2.67	2.37	2.32	2.33
Zn	30	1.835×10^{-16}	4.105×10^{-16}	2.31	2.22	5.88	6.26
Kr	36	1.102×10^{-16}	2.477×10^{-16}	2.27	2.25	9.82	9.88
Zr	40	7.871×10^{-17}	1.743×10^{-16}	2.21	(2.2)[a]	13.65	13.77
Mo	42	6.684×10^{-17}	1.472×10^{-16}	2.20	(2.2)[a]	16.01	16.15
Pd	46	4.882×10^{-17}	1.076×10^{-16}	2.20	(2.2)[a]	21.65	21.83
Rn	86	4.425×10^{-18}	9.353×10^{-18}	2.11	(2.1)[a]	226.0	225.9
No	102	2.291×10^{-18}	4.817×10^{-18}	2.10	(2.1)[a]	–	–

[a] Possible changes in the fluorescence yields have been considered by Chen [9] and found to be relatively small.

lifetimes (and decreasing the natural width) of K^{-2} states (see the values in columns 3–6 of table 2). Moreover, from columns 3 and 4 of table 2, one can see that the lifetimes of the K^{-2} state are always more than two times shorter than the lifetimes of the K^{-1} state.

3. Summary and conclusions

As a result of closing the $K^{\mathrm{h}}\alpha_1$ de-excitation channel in the pure LS coupling scheme, the $K^{\mathrm{h}}\alpha_1/K^{\mathrm{h}}\alpha_2$ intensity ratio changes with atomic number from small values (for the LS coupling limit at low Z) to about 1.5–1.6 (for the j–j coupling limit at high Z). However, it was found that closing the $K^{\mathrm{h}}\alpha_1$ de-excitation channel (due to the domination of the pure LS coupling for the low-Z atoms) does not enlarge the lifetimes of hollow atoms. The lifetimes of doubly K-shell ionized states are a bit more than two times shorter than for singly K-shell ionized ones for all of the atomic number range. The obtained theoretical results are in agreement with measured $K^{\mathrm{h}}\alpha_1/K^{\mathrm{h}}\alpha_2$ intensity ratios and the lifetimes of the doubly K-shell ionized states.

Acknowledgments

This work was supported by the Polish Ministry of Science and Higher Education through grant no. N N202 1465 33 and by the Swiss National Science Foundation.

References

[1] Rzadkiewicz J *et al* 1999 *Phys. Lett.* A **264** 186
[2] Rzadkiewicz J *et al* 2005 *Nucl. Instrum. Methods* B **235** 110
[3] Boschung B *et al* 1995 *Phys. Rev.* A **51** 3650
[4] Diamant R *et al* 2009 *Phys. Rev.* A **79** 062511–1
Diamant R *et al* 2009 *Phys. Rev.* A **79** 062512–1
[5] Salem S I *et al* 1984 *Phys. Rev.* A **29** 2634
Salem S I *et al* 1982 *Phys. Rev. Lett.* **49** 1240
[6] van Eijk C W E, Wijnhorst J and Popelier M A 1981 *Phys. Rev.* A **24** 854
[7] Mossé J P, Chevallier P and Briand J P 1985 *Z. Phys.* A **322** 207
[8] Grant I P 1984 *Int. J. Quantum Chem.* **25** 23
[9] Chen M H 1991 *Phys. Rev.* A **44** 239
[10] Dyall K G, Grant I P, Johnson C T, Parpia F A and Plummer E P 1989 *Comput. Phys. Commun.* **55** 425
[11] Polasik M 1995 *Phys. Rev.* A **52** 227