



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

A Large-scale Relativistic Configuration-Interaction Calculation for the 4s-4p Transition Energies of Copperlike Heavy Ions

K. T. Cheng, M. H. Chen

July 5, 2005

X05, 20th International Conference on X-ray and Inner-shell
Processes
Melbourne, Australia
July 4, 2005 through July 8, 2005

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

A large-scale relativistic configuration- interaction calculation for the $4s - 4p$ transition energies of copperlike heavy ions [★]

K. T. Cheng and M. H. Chen

Lawrence Livermore National Laboratory, Livermore, CA 94550 USA

Abstract

The $4s - 4p$ transition energies for high- Z copperlike ions are calculated using the relativistic configuration-interaction (RCI) method. These calculations are based on the relativistic no-pair Hamiltonian which includes Coulomb and frequency-dependent, retarded Breit interactions and use B-spline orbitals as basis functions. Mass polarization and quantum electrodynamic (QED) corrections are also calculated. The present RCI energies agree very well with results from the relativistic many-body perturbation theory. With QED corrections included, our total transition energies are in very good agreement with recent high-precision measurements.

Key words: Atomic spectroscopy, QED corrections

PACS: 31.30.Jv, 32.30.Rj, 31.25.-v, 31.15.Ar

The spectra of high- Z Cu-like ions feature prominent lines from the $4s - 4p$ transitions. These resonance lines have been subjected to several theoretical and experimental investigations in the past two decades. The first such measurements were carried out in the mid 80's with laser produced plasmas by Seely *et al.* using the OMEGA laser [1]. Theoretical calculations at the time were mostly based on the multiconfiguration Dirac-Fock (MCDF) method [1–3] and were not very accurate. While comparisons between theory and experiment did show the importance of finite nu-

clear size and quantum electrodynamic (QED) corrections [3], precision tests of these effects were nevertheless hampered by the lack of correlation corrections in these early MCDF calculations.

The situation has changed with the development of relativistic correlation calculations such as the relativistic many-body perturbation theory (RMBPT) [4] and the relativistic configuration-interaction (RCI) method [5]. Atomic theory can now match the accuracy of experiment, enabling rigorous tests of many-electron QED theory in the presence of strong external fields. Recent high-precision measurements [6–8] of the $4s - 4p$ transition energies in Cu-like heavy ions have exceeded the accuracy, and revealed the deficiency, of early laser-plasma experiments [1,2]. To compare with these new results, we have carried out large-scale RCI and QED calculations for these transitions.

[★] This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

Email addresses: ktcheng@llnl.gov (K. T. Cheng), chen7@llnl.gov (M. H. Chen).

Details of our RCI method can be found in Ref. [5]. Briefly, our RCI calculations are based on the relativistic no-pair Hamiltonian [9,10] which includes the Coulomb and frequency-dependent, retarded Breit interactions. Positive-energy projection operators are used in the no-pair Hamiltonian to prevent spurious interactions with virtual electron-positron pairs. Many-electron configuration-state functions (CSF) are constructed from one-electron B-spline (basis spline) functions which are radial Dirac orbits of an electron moving in a screened nuclear potential confined to a finite cavity [11]. B-spline orbitals form a complete, finite basis set and separate cleanly into positive- and negative-energy states. The no-pair requirement is implicitly satisfied by using only positive-energy B-spline orbitals in our RCI calculations.

In this work, we use 29-electron Dirac-Kohn-Sham (DKS) potentials with Fermi nuclear charge distributions as our model potentials to generate one-electron B-spline basis functions in a cavity about 3 a.u. in radius. Parameters for the Fermi charge distribution of the nucleus are from Johnson and Soff [12], except for thorium and uranium which are from Zumbro *et al.* [13,14]. For each angular momentum state, 40 positive- and 40 negative-energy B-spline orbitals are generated and the lowest 25 positive-energy B-spline orbitals are used in our RCI calculations. Contributions from the remaining higher energy basis functions are small and can be ignored.

Starting from the reference configuration of a $4l_v$ valence electron outside a closed $3d^{10}$ Ni-like core, all single- and double-excitations from core-valence (CV) and core-core (CC) interactions are systematically included for well converged RCI energies. For CV excitations, CSFs include configurations from single excitations of the valence electron $(3d^{10})nl_v$ and those from excitations of the valence electron plus one core electron

$$3d^{-1}nl_n'l', 3p^{-1}nl_n'l', 3s^{-1}nl_n'l', \\ 2p^{-1}nl_n'l', 2s^{-1}nl_n'l', 1s^{-1}nl_n'l',$$

where $0 \leq l, l' \leq 5$ and a^{-n} denotes n electrons removed from the core state a . Single excitations of a core electron are part of the above configurations with $nl = 4l_v$. For CC excitations, their contribu-

Table 1
Contributions to the $4s - 4p$ transition energies (eV) of Cu-like uranium. Here, CV and CC are core-valence and core-core contributions to the Coulomb energies, while $\omega = 0$ and $\omega \neq 0$ are frequency-independent and frequency-dependent contributions to the Breit energies.

Contribution		$4s - 4p_{1/2}$	$4s - 4p_{3/2}$	
RCI	Coulomb	146.72	473.14	
	CV	0.04	0.01	
	Sum	146.76	473.15	
	Breit	$\omega = 0$	2.62	-0.31
	$\omega \neq 0$	0.08	-0.54	
	Sum	2.70	-0.85	
Total		149.46(2)	472.30(2)	
Mass Polarization		0.00	0.00	
QED	Self-energy	-4.24	-4.33	
	Uehling	1.02	1.25	
	Wichmann-Kroll	-0.05	-0.06	
	Core Relaxation	-0.02	0.03	
	2-loop Lamb Shift	0.02	0.02	
	Total		-3.27(5)	-3.09(5)
Theory		146.19(5)	469.21(5)	

tions are small and are calculated for Coulomb energies only. To limit the size of these RCI expansions which can easily get out of hand, CSFs are limited to excitations from the $n = 3$ subshells and to $0 \leq l, l' \leq 3$. They are given by

$$3d^{-2}4l_vnl_n'l', 3p^{-1}3d^{-1}4l_vnl_n'l', \\ 3p^{-2}4l_vnl_n'l', 3s^{-1}3d^{-1}4l_vnl_n'l', \\ 3s^{-2}4l_vnl_n'l', 3s^{-1}3p^{-1}4l_vnl_n'l'.$$

Resulting large-scale RCI expansions reach over 300,000 configurations and Davidson's method [15,16] is used to solve for the first few eigenstates of these big RCI matrices. Mass polarization (MP) corrections are then calculated as expectation values of the operator $H_{MP} = (1/M)\sum_{i>j} \mathbf{p}_i \cdot \mathbf{p}_j$, where M is the nuclear mass, using RCI eigenwave functions.

Our QED corrections are evaluated from the one-loop self-energy and vacuum polarization diagrams with methods similar to those used in

Table 2
The $4s - 4p$ transition energies (eV) of Cu-like ions.

Transition	Contribution	Reference	W ⁴⁵⁺	Au ⁵⁰⁺	Pb ⁵³⁺	Th ⁶¹⁺	U ⁶³⁺	
$4s - 4p_{1/2}$	Coulomb	This work	97.85	110.49	118.43	140.85	146.76	
		RMBPT [4]	97.83	110.47	118.40	140.83	146.74	
	Breit	This work	1.05	1.40	1.64	2.46	2.70	
		RMBPT [4]	1.06	1.40	1.65	2.47	2.72	
	QED	This work	-1.36	-1.78	-2.07	-3.00	-3.27	
		Blundell [23]	-1.34		-2.05	-2.98	-3.25	
		Kim <i>et al.</i> [24]	-1.33	-1.74	-2.02	-2.92	-3.18	
	Total	This work	97.54	110.11	118.00	140.31	146.19	
		Dielec. recomb. [6]			118.010(1)			
		Laser-plasma [2]	97.63(1)	110.22(1)	118.15(2)		146.39(37)	
	$4s - 4p_{3/2}$	Coulomb	This work	200.25	255.25	294.78	430.73	473.15
			RMBPT [4]	200.25	255.22	294.76	430.72	473.13
Breit		This work	-0.13	-0.24	-0.33	-0.71	-0.85	
		RMBPT [4]	-0.14	-0.24	-0.33	-0.72	-0.86	
QED		This work	-1.23	-1.63	-1.91	-2.81	-3.09	
		Blundell [23]	-1.22		-1.88	-2.78	-3.05	
		Kim <i>et al.</i> [24]	-1.21	-1.59	-1.86	-2.73	-2.99	
Total		This work	198.89	253.38	292.54	427.21	469.21	
		EBIT [7]	198.90(1)	253.40(1)	292.60(4)	427.20(4)	469.06(3)	
		EBIT [8]				427.21(1)	469.22(3)	
		Laser-plasma [2]	198.99(5)	253.40(8)	292.65(10)	427.68(22)	469.53(25)	

Ref. [17]. Specifically, leading vacuum polarization corrections are calculated as expectation values of the Uehling potential. Electron self-energy (SE) and Wichmann-Kroll (WK) corrections are calculated non-perturbatively to all orders of $Z\alpha$ with partial wave expansions of bound-state Green's functions. Details of our SE and WK calculations can be found in Refs. [18] and [19], respectively. To account for screening and relaxation corrections, one-loop QED energies are evaluated in DKS potentials specific to the valence configurations of the initial and final states. This procedure has yielded very accurate QED corrections for high- Z Li-like and Be-like ions [20] as well as for Na-like

to Si-like uranium [17]. It should work just as well for heavy Cu-like ions here. Finally, higher-order two-loop Lamb shift contributions are small and are estimated from the H-like $1s$ results [21,22] using the $1/n^3$ scaling rule.

Table 1 shows various contributions to the $4s - 4p$ transition energies, using Cu-like uranium as an example. Here, core-core contributions to the Coulomb energies are seen to be very small and their contributions to the Breit energies should thus be quite negligible. Also, while frequency-dependent Breit ($\omega \neq 0$) energies are significant and can even be larger than frequency-independent Breit ($\omega = 0$) energies, mass polar-

ization corrections are found to be completely negligible. Theoretical uncertainties quoted here are estimates only. For RCI energies, they are due mainly to the neglect of triple and quadruple excitations. For QED corrections, they come from the approximated treatment of screening and relaxation corrections and, to a lesser extent, the neglect of negative-energy contributions to electron correlation energies.

Tables 2 shows $4s - 4p$ transition energies for high- Z Cu-like ions. It can be seen that our RCI Coulomb and Breit energies agree with RMBPT results of Johnson *et al.* [4], while our QED energies agree with Blundell's *ab initio* results [23] but deviate from those obtained with the Welton method by Kim *et al.* [24]. Also, our $4s - 4p_{1/2}$ transition energy in Pb^{53+} is in excellent agreement with the dielectronic recombination measurement [6], while our $4s - 4p_{3/2}$ transition energies are in excellent agreement with recent EBIT measurements [7,8]. It should be noted that the EBIT energy of 469.06(3) eV for the $4s - 4p_{3/2}$ transition in Cu-like uranium as reported in Ref. [7] appears to be way off. The new EBIT value of 469.22(3) eV as report in Ref. [8] are in much better agreement with theory and are more consistent with the systematic trend along the isoelectronic sequence. From this table, it is also clear that early laser-plasma measurements of Seely *et al.* [2] are consistently too high in energy when compare with results of recent high-precision experiments.

In conclusion, we have calculated the $4s - 4p$ transition energies for copperlike ions with $Z = 74, 79, 82, 90$ and 92 . Our results show that large-scale RCI calculations including core-valence and core-core excitations can get very accurate relativistic correlation energies for Cu-like ions, and that screening and relaxation corrections to the QED energies can be well approximated by evaluating self-energy and vacuum polarization contributions in Dirac-Kohn-Sham model potentials.

References

- [1] J. F. Seely, J. O. Ekberg, C. M. Brown, U. Feldman, W. E. Behring, J. Reader, and M. C. Richardson, Phys. Rev. Lett. **57**, 2924 (1986).
- [2] J. F. Seely, C. M. Brown, and U. Feldman, At. Data Nucl. Data tables **43**, 145 (1989).
- [3] K. T. Cheng and R. A. Wagner, Phys. Rev. A **36**, 5435 (1987).
- [4] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **42**, 1087 (1990).
- [5] M. H. Chen, K. T. Cheng, W. R. Johnson, and J. Sapirstein, Phys. Rev. A **52**, 266 (1995).
- [6] E. Lindroth, H. Danared, P. Glans, Z. Pešić, M. Tokman, G. Viktor, and R. Schuch, Phys. Rev. Lett. **86**, 5027 (2001).
- [7] S. B. Utter, P. Beiersdorfer, E. Träbert, and E. J. Clothiaux, Phys. Rev. A **67**, 032502 (2003).
- [8] E. Träbert, P. Beiersdorfer, and H. Chen, Phys. Rev. A **70**, 032506 (2004).
- [9] G. E. Brown and D. G. Ravenhall, Proc. R. Soc. London, Ser. A **208**, 552 (1951).
- [10] M. H. Mittleman, Phys. Rev. A **4**, 893 (1971); **5**, 2395 (1972); **24**, 1167 (1981).
- [11] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **37**, 307 (1988).
- [12] W. R. Johnson and G. Soff, At. Data Nucl. Data Tables, **33**, 405 (1985).
- [13] J. D. Zumbro, R. A. Naumann, M. V. Hoehn, E. B. Shera, C. E. Bemis, Jr., and Y. Tanaka, Phys. Lett. **167B**, 383 (1986).
- [14] J. D. Zumbro, E. B. Shera, Y. Tanaka, C. E. Bemis, Jr., R. A. Naumann, M. V. Hoehn, W. Reuter, and R. M. Steffen, Phys. Rev. Lett. **53**, 1888 (1984).
- [15] E. R. Davidson, J. Comp. Phys. **17**, 87 (1975).
- [16] A. Stathopoulos and C. Froese Fischer, Comput. Phys. Commun. **79**, 1 (1994).
- [17] M. H. Chen, K. T. Cheng, P. Beiersdorfer, and J. Sapirstein, Phys. Rev. A **68**, 022507 (2003).
- [18] K. T. Cheng, W. R. Johnson, and J. Sapirstein, Phys. Rev. Lett. **66**, 2960 (1991); Phys. Rev. A **47**, 1817 (1993).
- [19] J. Sapirstein and K. T. Cheng, Phys. Rev. A **68**, 042111 (2003).
- [20] K. T. Cheng, M. H. Chen, and J. Sapirstein, Phys. Rev. A **62**, 054501 (2000).
- [21] V. A. Yerokhin, P. Indelicato, and V. M. Shabaev, Phys. Rev. Lett. **91**, 073001 (2003).
- [22] T. Beier, P. J. Mohr, H. Persson, G. Plunien, M. Greiner, and G. Soff, Phys. Lett. A **236**, 329 (1997).
- [23] S. A. Blundell, Phys. Rev. A **47**, 1790 (1993).
- [24] Y.-K. Kim, D. H. Baik, P. Indelicato, and J. P. Desclaux, Phys. Rev. A **44**, 148 (1991).