Lifetimes of $3s3p^2$ $J=\frac{1}{2},\frac{5}{2}$ levels in Au^{66+} and Br^{22+}

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Relativistic configuration-interaction length and velocity lifetimes have been obtained for $3s3p^2$ $J=1/2$, 5/2 levels in Au^{66+} and Br^{22+}. Results from the two gauges agree well, and we have moderately improved earlier multiconfigurational Dirac-Fock results, as compared to experiment. However, there remains a discrepancy for the Au^{66+} $J=5/2$ lifetime, which may be due to satellite spectra associated with $5g$ and other spectator electrons.

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I. INTRODUCTION

Most level lifetimes in highly ionized high Z ions, which decay by an electric dipole process, are too short to be currently measured. Intercombination lifetimes, such as $3s3p^2$ ''^4P'' $\rightarrow 3s^2\,3p\,^2P$, differ; their lifetimes can be a few tens of ps [Au^{66+}] or longer, thus opening up an isoelectronic sequence for study [1]. Au is an important plasma material, and M-shell x-ray transitions have been found to be prominent in laser-produced plasmas, with nickel-, copper-, zinc-, and gallium-like ions being most likely [2]. In the next section, we report results for the Ni-like $3d^{10}\rightarrow 3d^9\,4f$ transition in Au^{51+}, of interest to the plasma fusion community and which we use to help benchmark our method.

Our main concern here, though, is to try to account for the discrepancies between theory and experiment for the $3s3p^2$ $J=5/2$ lifetime in Au^{66+}, and the $3s3p^2$ $J=1/2$ lifetimes in Au^{66+} and Br^{22+} as reported by Träbert *et al.* [1,3]. The earlier theoretical results [4] were obtained using the length gauge, using multiconfigurational Dirac-Fock (MCDF) wave functions generated from the complex (all configurations with three $n=3$ electrons and a neon core), and MCDF energy differences. For Au^{66+}, it was found [1] that the $J=1/2$ energy difference was about 1% in error, and the $J=5/2$ energy difference about 4% in error ($\sim$6.8 eV). This amount of energy ($\sim$6.8 eV) would seem difficult to account for by means of ''beyond the complex'' correlation effects. It should be noted that the MCDF results [4] included the effects of the Breit operator (magnetic and retardation), but not radiative effects. These last effects were found [5] to be important for $3s^2\,3p$ fine structure, and are even more likely to be important when the $3s$ occupation is changing, as it is here.

For Au^{66+}, the experimental paper [1] recomputed the MCDF [4] lifetimes using the experimental energies, with the result that the $J=1/2$ lifetime was 16.4% too low, and the $J=5/2$ lifetime was 18.8% too high. Since lifetimes depend on the third power of the energy difference, a 4% change in energy makes a significant difference. However, there is one problem, and one potential problem, with what was done [1]. The problem is that the MCDF $J=1/2$ lifetime was misquoted: rather than 17.8 ps, it should be 19.98 ps (MCDF dE), which drops the discrepancy (experimental dE) to about 6%.

The potential problem is concerned with whether it is appropriate to use the experimental energy difference. If it is correct, as we have argued in the past [6], then we should use it, as our main effort is concentrated in getting the transition moment calculated correctly. But for highly ionized species, satellite spectra have also been observed associated with spectator electrons, and these spectators may introduce shifts of a few eV [2]. Satellites can also affect lifetime determinations [2,7], for example, shortening lifetimes by $\sim$20%. In the next section, we will present results which decrease the theoretical-experimental discrepancy to an acceptable level, except in the case of the Au^{66+} $J=5/2$ lifetime and energy difference. Based on the agreement in the two gauges, and the known potential difficulties in measuring lifetimes of highly ionized atoms [7], it may be that the remaining discrepancy is of a more experimental than theoretical origin.

II. METHODOLOGY AND RESULTS

The $1s\ldots 3p$ radial functions are generated by solving the Dirac-Coulomb equations for a single manifold ($3s3p^2$ or $3s^2\,3p$) using Desclaux's algorithm [8]. All other radial functions, both virtual and the $3d$'s, were represented by relativistic screened hydrogenic functions, and their screening constants (Z^*) were determined using the energy variational principle. Magnetic, retardation, and radiative effects were added to energy differences, using the newer version of Desclaux's algorithm [9]. Magnetic effects were introduced into the Au^{66+} $J=5/2$ wave function, but had little impact ($\sim$2% for the length gauge).

Correlation effects are introduced by making single and double excitations from the $3s3p^2$ and $3s^2\,3p$ manifolds. Schematically, these are as follows: $3s\rightarrow s+d$; $3p\rightarrow p+f$; $3p^2\rightarrow s^2+p^2+d^2+f^2+sd+pf$; $3s3p\rightarrow sp+pd+df$; $3s^2\rightarrow s^2+p^2+d^2+f^2$. Some $n=3$ triple excitations were also included, but these had little effect. The largest energy and f-value contributions were from within the ''complex,'' viz., $3s\rightarrow 3d$ and $3s^2\rightarrow 3p^2$ for $3s^2\,3p$, and $3s\rightarrow 3d$ and $3p^2\rightarrow 3s3d$ for $3s3p^2$.

For more accurate results, particularly for the velocity operator, excitations from the $n=2$ subshells should be allowed. For example, the $n=3$ Au^{66+} $J=1/2$ velocity result changes 14% when $n=2$ excitations are allowed. Some guidance as to which configurations make significant contributions can be gotten from the first order theory of oscillator strengths (FOTOS), which is presented in Ref. [10]. Here, FOTOS includes the excitations $2p\rightarrow s+d$ and $2s\rightarrow p$ from

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TABLE I. Lifetimes of $3s3p^2J$ levels.

Species	dE (a.u.)			τ	
	RCI	Expt.	MCDF	RCI [a]	Expt.
Au^{66+} $J=1/2$	6.894	6.904±.02[b]	20.7 ps[c]	21.2 ps (L) 21.9 ps (V)	22±4 ps[b]
Au^{66+} $J=5/2$	6.358	6.268±.008	57.5 ps[c]	57.2 ps (L) 56.8 ps (V)	50.5±2 ps[b]
Br^{22+} $J=1/2$ [d]	1.774	1.783±.002[e]	1.71 ns[c]	1.87 ns (L) 1.79 ns (V)	1.9±.2 ns[e]
Br^{22+} $J=5/2$	1.747	1.752±.002[e]	2.05 ns[c]	1.949 ns (L) 1.894 ns (V)	2.05±.10 ns[e]

[a]This work. L = length gauge, V = velocity gauge. For $\tau_{1/2}$, the experimental dE is used.
[b]Reference [1].
[c]Reference [4]. For τ the experimental dE is used and τ is corrected [Au^{66+}] to the original Huang [4] value.
[d]Includes the two decay branches, viz., $3s3p^2$ $J=1/2\rightarrow 3s^2\,3pJ'$; $J'=1/2$, 3/2. dE is given for the $J=1/2\rightarrow J=1/2$ branch.
[e]Ref. [3].

the Dirac-Fock (DF) manifolds. Thus, for example, $2s^2\,2p^5d3s3p^2$ is included in the $3s^2\,3p$ wave function (this is $2p3s\rightarrow d3p$ from $3s^2\,3p$). For the $J=1/2$ lifetimes, this brings the two gauge results very close to each other (see Table I), and in particularly good agreement with experiment for Au^{66+}. It may be noted that the MCDF [4] and relativistic configuration interaction (RCI) length results are in good agreement for all four results.

However, at this stage the Au^{66+} $J=5/2$ dE and lifetime were not in good agreement with experiment [1], so a much more extensive set of $n=2$ excitations were included. For this transition, all $2l$, $3l'$ pair excitations, $2s^2$, $2s2p$, $2p^2$ exclusion effects (excitation into $3s$ and/or $3p$ subshells), and $2s$, $2p$ single excitations were included, with the results given in Table I. Excitations from $n=2$ required substantially different virtuals than $n=3$ excitations (Z^* were two to four times larger). While the two gauges are in excellent agreement, the RCI dE is 2.45 eV higher than experiment, and the lifetime is ~13% higher than the experimental value [1]. These are both rather large discrepancies, taken in the context of the other results. The Br^{22+} $J=5/2$ lifetime calculated at the MCDF level in the length gauge was already in good agreement with experiment (see Table I), but the velocity value was improved with the inclusion of the $n=2$ excitations.

The large contribution of radiative effects (~5.9 eV) and the magnetic operator (~5.3 eV) may leave open the possibility that higher order QED or relativistic effects, when they become feasible for ''complicated'' states, might be significant, at least for dE. However, there seem to exist two other possibilities, on the experimental side, the first involving satellite spectra and spectator electrons [2,7], for which additional calculations are feasible, and may be of value.

Specifically, we have chosen to add an nlj electron to both $3s^2\,3p$ $J=3/2$ and $3s3p^2$ $J=5/2$. This has been done by keeping the core coupled as for Au^{66+}, and varying nl (j was fixed at $j=l+1/2$) to see what the change of energy was. The largest shift obtained at the DF level [8] for a single spectator was for a $5g_{9/2}$ attachment ($3d$–$6d$, $4f$, and $6h$ were also tried). If the final states are coupled to $J=5$ (upper) and $J=6$ (lower), the shift is 1.48 eV (recall that the RCI-experiment dE discrepancy was 2.45 eV). Since the $3s3p^25g$ state has alternative decay modes (e.g., $5g\rightarrow 4f$ has a ''lifetime'' of 0.015 ps for a $5g_{9/2}$ $J=5\rightarrow 4f_{7/2}$ $J'=4$ decay), if this state were being observed, it would have a shorter lifetime, i.e., in the same direction as the observed lifetime. Use of two $5g$ spectators gave an additional 0.3 eV shift ($J=21/2\rightarrow J'=19/2$).

The second possibility, is that the Au^{66+} $J=5/2$ decay was only followed for 80 ps [1], which is a little more than the decay time (~50 ps), so the experimental uncertainty might be greater than the quoted [1] 4%. However, this possibility does not seem to address the discrepancy in dE.

We also did a limited RCI calculation for the $3d^{10}$ $J=0\rightarrow 3d^9\,4f$ $J=1$ transition in Au^{51+}. Using FOTOS [10] predictions, we included $3d^8$ $(4f^2+4p4f)$ in the ground state, and $3d^94p$ in the excited state. The RCI length result for the f value was 1.9195 and the velocity result was 1.9172, a spread of only 0.12%. There was a 5.1% spread in the DF gauges, as contrasted to ~10% spread reported earlier [2].

[1] E. Träbert, U. Staude, P. Bosselmann, K. H. Schartner, P. H. Molder, and X. Tordoir, Eur. Phys. J. D **2**, 117 (1998).
[2] K. Honda, K. Mima, and F. Koike, Phys. Rev. E **55**, 4594 (1997).
[3] E. Träbert, J. Suleiman, S. Cheng, H. G. Berry, R. W. Dunford, E. P. Kanter, C. Kurtz, A. E. Livingston, K. W. Kukla, F. G. Serpa, and L. J. Curtis, Phys. Rev. A **47**, 3805 (1993).
[4] K.-N. Huang, At. Data Nucl. Data Tables **27**, 539 (1982).
[5] Y.-K. Kim, J. Sugar, V. Kaufman, and M. A. Ali, J. Opt. Soc. Am. B **5**, 2225 (1988).

[6] D. R. Beck and O. Sinanoglu, Phys. Rev. Lett. **28**, 945 (1972).
[7] Y. Zou, R. Hutton, S. Huldt, I. Martinson, K. Ando, T. Kambara, H. Oyama, and Y. Awaya, Phys. Rev. A **60**, 982 (1999).
[8] J. P. Desclaux, Comput. Phys. Commun. **9**, 31 (1975).
[9] J. P. Desclaux, in *Methods and Techniques in Computational Chemistry*, edited by E. Clementi (Stef. Cagliara, Italy, 1994), Vol. A.
[10] C. A. Nicolaides and D. R. Beck, Chem. Phys. Lett. **36**, 79 (1975).