PHYSICAL REVIEW A VOLUME 45, NUMBER 3 1 FEBRUARY 1992

Relativistic excitation energies and oscillator strengths for the $6s^2\,{}^1S_0 \rightarrow 6s6p\ {}^1P_1, {}^3P_1$ transitions in Hg-like ions

Hsiang-Shun Chou* and Keh-Ning Huang
Institute of Atomic and Molecular Sciences, Academia Sinica, P.O. Box 23-166, Taipei, Taiwan 10764, Republic of China
and Department of Physics, National Taiwan University, Taipei, Taiwan 10764, Republic of China

(Received 18 September 1991)

Excitation energies and oscillator strengths from the ${}^1S_0$ ground state to the first ${}^3P_1$ and ${}^1P_1$ excited states of Hg-like ions are calculated by using the multiconfiguration relativistic random-phase approximation. Results are compared with other theories and with experiment. The discrepancy between the theoretical and experimental results is discussed.

PACS number(s): 31.20.−d, 32.70.Cs

I. INTRODUCTION

The spectrum of Hg-like ions has aroused considerable interest in recent years. Systematic studies of oscillator strengths in Hg-like ions have been performed by measuring the lifetimes of excited atomic states using the beam-foil technique [1–5]. A nonrelativistic Hartree-Fock calculation [6] has yielded an oscillator strength for the resonance transition $6s^2\,{}^1S_0 \rightarrow 6s6p\ {}^1P_1$ in Hg that is nearly a factor of 3 greater than the experimental result [1]. Desclaux and Kim [6] accounted for half the discrepancy with a relativistic Dirac-Fock calculation. A multiconfiguration Dirac-Fock (MCDF) calculation further reduced the discrepancy, but a difference of 30% between the experimental and theoretical results remained. Hafner and Schwarz [7] performed a relativistic pseudopotential calculation with Hameed's [8] core-polarization correction to account for core-valence correlations. Core-polarization effects reduced the frozen-core oscillator strength by approximately 40%, but there is still a significant discrepancy. Migdalek, Baylis, and Bojara [9–12] carried out both MCDF and configuration-interaction (CI) calculations with a semiempirical core-polarization (CP) potential to account for valence-core correlations. A discrepancy of 17–92 % exists between the theoretical and the experimental oscillator strengths. A reliable atomic calculation which takes into account both relativistic and correlation effects is required in order to resolve the discrepancy.

The relativistic random-phase approximation (RRPA) [13] and MCDF theories have played two of the major roles in attacking the correlation problem in relativistic calculations and have produced extensive data on a variety of atoms and ions. The MCDF approach has the merit of being applicable to arbitrary systems and being capable of treating certain correlations not included in the RRPA approach. The RRPA can be applied only to closed-shell systems, and only a single configuration is allowed to describe the reference state of the system. Nevertheless, the RRPA approach does have several advantages: First, the results are gauge independent, second, both discrete and continuum correlations can be dealt with, and finally, core polarization can be readily treated.

The RRPA can be improved upon by using a multiconfiguration wave function as the reference state. The electron-electron correlation effects due to the presence of "real" doubly excited configurations in the initial state are thereby included. This approach is called the multiconfiguration relativistic random-phase-approximation theory (MCRRPA). In essence, it has certain features of the MCDF, while preserving all of the advantages, especially the gauge invariance, of the RRPA. Applications of the MCRRPA to photoexcitations of Be-, Mg, and Pb-like ions [14–16] and to photoionization of Be, Mg, Zn, and Sr atoms [17–21] have been carried out and are in excellent agreement with experiment. In this paper we apply the MCRRPA to the resonance transition $6s^2\,{}^1S_0 \rightarrow 6s6p\ {}^1P_1$ and the intercombination transition $6s^2\,{}^1S_0 \rightarrow 6s6p\ {}^3P_1$ in Hg-like ions.

II. THEORY AND RESULTS

The MCRRPA theory is based on an approximate relativistic Hamiltonian

$$H(t)=\mathcal{H}+V(t)\ , \tag{2.1}$$

where

$$\mathcal{H}=\sum_{n=1}^{N} h_n+\sum_{n\,(<m)}^{N} v_{nm}\ . \tag{2.2}$$

Here h_n is a single-electron Dirac Hamiltonian, and v_{nm} are mutual Coulomb interactions between electrons,

$$v_{nm}=\frac{e^2}{r_{nm}}\ . \tag{2.3}$$

In Eq. (2.1), $V(t)$ is an external potential which induces transitions between atomic states,

$$V(t)=v_+e^{-i\omega t}+v_-e^{i\omega t}\ , \tag{2.4}$$

with

$$v_\pm=\sum_{n=1}^{N} v_{n\pm}\ . \tag{2.5}$$

The Breit interaction is not considered here but may be easily included in the calculation if required.

Our point of departure is the time-dependent variational principle [22]. We describe the N-electron system as a superposition of configuration wave functions with time-dependent weights. Since the external perturbation may have components with nonvanishing angular momentum and odd parity, the atomic wave function contains terms of mixed angular momentum and parity. Applying the variational principle we derive time-dependent multiconfiguration Dirac-Fock equations describing the response of the atom to the external field. Terms independent of the external field lead to the usual MCDF description of an atomic state. Those terms proportional to the external field lead to equations describing the linear response of the atomic state to the external field; in the sequel we refer to these linear response equations as the MCRRPA equations. If we start from a single-configuration reference state, the MCRRPA equations reduce to the usual RRPA equations. The MCRRPA equations may alternatively be derived from an equation-of-motion point of view [23]. In the MCRRPA formulation, the reference state of Hg-like ions is given by a multiconfiguration wave function as

$$\Psi = C_1(6s_{1/2}^2)+C_2(6p_{1/2}^2)+C_3(6p_{3/2}^2)\ , \tag{2.6}$$

where the symbol $(6l_j^2)$ designates a Slater determinant constructed from the $6l_j$ valence orbitals and 21 core orbitals. The parameters C_a $(a=1,2,3)$ in Eq. (2.6) are configuration weight coefficients. The core and valence orbitals and the configuration weights are determined by solving the MCDF equations numerically. Since the MCRRPA equations include all one-particle excitations of the ground state, they automatically include, in addition to the usual RRPA correlation effects, the important final-state correlations between an excited electron and excited $6p_{1/2}$ or $6p_{3/2}$ ionic cores. These two-particle–two-hole final-state correlations were omitted in previous RRPA studies. As a further simplification, we adopt a truncated version of the MCRRPA in which only excitations of the valence electrons are considered. Seven coupled channels are considered in the electric dipole approximation:

$$\begin{aligned} &6s_{1/2} \rightarrow np_{1/2}, np_{3/2}\ , \\ &6p_{1/2} \rightarrow ns_{1/2}, nd_{3/2}\ , \\ &6p_{3/2} \rightarrow ns_{1/2}, nd_{3/2}, nd_{5/2}\ . \end{aligned} \tag{2.7}$$

The neglect of the core-polarization effects leads to a difference between the length and the velocity oscillator

TABLE II. Excitation energies (in cm^{-1}) for the intercombination transition $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^3P_1$ in Hg-like ions.

	MCDF-CP[a]	CI-CP[b]	MCRRPA[c]	Expt.[d]
Hg	39 045	34 928	33 461	39 417
Tl^{1+}	51 786	48 657	46 703	52 399
Pb^{2+}	63 683	60 387	58 905	64 398
Bi^{3+}	75 174	71 761	70 615	75 935
Po^{4+}	86 489	83 013	82 038	
At^{5+}	97 591	94 099	93 279	
Rn^{6+}	108 610	105 114	104 388	
Fr^{7+}			115 422	
Ra^{8+}			126 410	
Ac^{9+}			137 376	
Th^{10+}			148 334	
Pa^{11+}			159 311	
U^{12+}			170 301	
Np^{13+}			181 342	

[a] Reference [10].
[b] Reference [12].
[c] Present results.
[d] Reference [24].

TABLE I. Excitation energies (in cm^{-1}) for the resonance transition $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1$ in Hg-like ions.

	MCDF-CP[a]	CI-CP[b]	MCRRPA[c]	Expt.[d]
Tl^{1+}	77 445	71 948	74 670	75 669
Pb^{2+}	97 970	91 983	95 537	95 351
Bi^{3+}	117 916	111 588	115 615	114 615
Po^{4+}	137 842	131 303	135 607	
At^{5+}	158 097	151 431	155 847	
Rn^{6+}	178 804	172 056	176 522	
Fr^{7+}			197 791	
Ra^{8+}			219 757	
Ac^{9+}			242 515	
Th^{10+}			266 144	
Pa^{11+}			290 729	
U^{12+}			316 337	
Np^{13+}			343 061	

[a] Reference [10].
[b] Reference [12].
[c] Present results.
[d] Reference [24].

TABLE III. Oscillator strengths for the resonance transition $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1$ in Hg-like ions.

	MCDF-CP[a]	CI-CP[b]	MCRRPA[c]	Expt.[d]
Tl^{1+}	1.55	1.31	2.307	1.21±0.20
Pb^{2+}	1.64	1.43	2.455	1.01±0.20
Bi^{3+}	1.69	1.51	2.515	0.88±0.15
Po^{4+}	1.72	1.57	2.543	
At^{5+}	1.75	1.62	2.556	
Rn^{6+}	1.78	1.66	2.562	
Fr^{7+}			2.564	
Ra^{8+}			2.564	
Ac^{9+}			2.563	
Th^{10+}			2.563	
Pa^{11+}			2.564	
U^{12+}			2.565	
Np^{13+}			2.567	

[a] Reference [10].
[b] Reference [12].
[c] Present results.
[d] Reference [1].

TABLE IV. Oscillator strengths for the intercombination transition $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^3P_1$ in Hg-like ions. Numbers in square brackets indicate the exponent.

	MCDF-CP[a]	CI-CP[b]	MCRRPA[c]	Expt.
Hg	1.98[−2]	2.86[−2]	1.604[−2]	2.41±0.02[−2][d] 2.37±0.08[−2][e] 2.49±0.1[−2][f] 2.47±0.04[−2][g]
Tl^{1+}	3.66[−2]	3.82[−2]	3.739[−2]	
Pb^{2+}	5.44[−2]	5.52[−2]	6.151[−2]	
Bi^{3+}	7.25[−2]	7.36[−2]	8.666[−2]	
Po^{4+}	9.05[−2]	9.21[−2]	1.117[−1]	
At^{5+}	1.08[−1]	1.10[−2]	1.359[−1]	
Rn^{6+}	1.25[−1]	1.27[−2]	1.589[−1]	
Fr^{7+}			1.805[−1]	
Ra^{8+}			2.004[−1]	
Ac^{9+}			2.188[−1]	
Th^{10+}			2.357[−1]	
Pa^{11+}			2.510[−1]	
U^{12+}			2.650[−1]	
Np^{13+}			2.776[−1]	

[a] Reference [10].
[b] Reference [12].
[c] Present results.
[d] Reference [2].
[e] Reference [3].
[f] Reference [4].
[g] Reference [5].

strengths. Nevertheless, the calculations are expected to be more accurate with increasing nuclear charges because the core-polarization effects are less important for higher ionization stages. In Tables I and II, excitation energies of the Hg-like ions from the MCRRPA theory are compared with those from other theories and from experiment. The MCRRPA excitation energies for the resonance transition agree within 1.3% with experiment, whereas differences of 7–15 % exist for the intercombination transition. In Tables III and IV, oscillator strengths of the Hg-like ions from the MCRRPA theory are compared with those from other theories and from experiment. Despite the neglect of the core-polarization effects, the length and velocity oscillator strengths agree within 0.2% for the intercombination transition and within 5% for the resonance transition. Only the length values are presented in Tables III and IV. For the intercombination transition in Hg, a significant discrepancy exists between the MCRRPA and experimental results. The discrepancy may be due to the core-polarization and further correlation effects which are neglected in the present calculations. For the resonance transition, the MCRRPA oscillator strengths are nearly 2–3 times greater than the experimental values. The frozen-core MCRRPA calculations seem to overestimate the oscillator strengths. Inclusion of the core-polarization effects will reduce the oscillator strengths. Nevertheless, the increasing discrepancy between theory and experiment with increasing nuclear charges suggests that the discrepancy is not primarily due to the neglect of the core-polarization effects. A similar discrepancy has been observed in the resonance transition of the Zn-like ions [25–28] which are homologous to the Hg-like ions. The discrepancy in the Zn-like ions was then resolved by Andersen *et al.* [29] with a proper account of the cascade effects. Younger and Wiese [30] have shown that the cascade effects are most serious for highly charged ions and become more severe as one moves upward along a homologous family of atoms. Thus, it seems that a substantial part of the discrepancy between the theoretical and experimental oscillator strengths for the resonance transition of Hg-like ions may arise from an improper account of the cascade effects in beam-foil lifetime measurements.

ACKNOWLEDGMENT

This work was supported in part by the National Science Council of the Republic of China.

---

*Present address: Division of General Education, National Taiwan Ocean University, Keelung, Taiwan 20224, Republic of China.

[1] T. Andersen, A. K. Nielsen, and G. Sorenson, Phys. Scr. **6**, 122 (1972).
[2] G. C. King and A. Adams, J. Phys. B **7**, 1712 (1974).
[3] J. A. Halstead and R. R. Reeves, J. Quant. Spectrosc. Radiat. Transfer **28**, 289 (1982).
[4] K. A. Mohamed, J. Quant. Spectrosc. Radiat. Transfer **30**, 225 (1983).
[5] J. S. Deech and W. E. Baylis, Can. J. Phys. **40**, 90 (1970).
[6] J. P. Desclaux and Y.-K. Kim, J. Phys. B **8**, 1177 (1975).

[7] P. Hafner and W. H. E. Schwarz, J. Phys. B **11**, 2975 (1978).
[8] S. Hameed, A. Herzenberg, and M. G. James, J. Phys. B **1**, 822 (1968).
[9] J. Migdalek and W. E. Baylis, J. Phys. B **17**, L459 (1984).
[10] J. Migdalek and W. E. Baylis, J. Phys. B **18**, 1533 (1985).
[11] J. Migdalek and A. Bojara, J. Phys. B **20**, L1 (1987).
[12] J. Migdalek and A. Bojara, J. Phys. B **21**, 2221 (1988).
[13] W. R. Johnson and C. D. Lin, Phys. Rev. A **20**, 964 (1979).
[14] W. R. Johnson and K.-N. Huang, Phys. Rev. Lett. **48**, 315 (1982).
[15] K.-N. Huang and W. R. Johnson, Nucl. Instrum. Methods B **9**, 502 (1985).
[16] H.-S. Chou, K.-N. Huang, and W. R. Johnson, Phys. Rev. A **44**, 2769 (1991).
[17] H.-C. Chi, K.-N. Huang, and K.-T. Cheng, Phys. Rev. A **43**, 2542 (1991).
[18] H.-C. Chi and K.-N. Huang, Phys. Rev. A **43**, 4742 (1991).
[19] H.-C. Chi and K.-N. Huang (unpublished).
[20] C.-Y. Hwang, H.-C. Chi, and K.-N. Huang, Phys. Rev. A **44**, 7189 (1991).
[21] C.-M. Wu, H.-C. Chi, and K.-N. Huang (unpublished).
[22] P. W. Langhoff, S. T. Epstein, and M. Karplus, Rev. Mod. Phys. **44**, 602 (1972).
[23] K.-N. Huang, Phys. Rev. A **26**, 734 (1982).
[24] C. E. Moore, *Atomic Energy Levels,* Natl. Bur. Stand. (U.S.) National Standard Reference Data Series-35 (U.S. GPO, Washington, DC, 1971).
[25] P. Shorer and A. Dalgarno, Phys. Rev. A **16**, 1502 (1977).
[26] P. Shorer, Phys. Rev. A **18**, 1060 (1978).
[27] C. Froese Fischer and J. Hansen, Phys. Rev. A **17**, 1956 (1978).
[28] T.-C. Cheng and K.-N. Huang, Phys. Rev. A (to be published).
[29] T. Andersen, P. Eriksen, O. Poulsen, and P. S. Ramanujam, Phys. Rev. A **20**, 2621 (1979).
[30] S. M. Younger and W. L. Wiese, Phys. Rev. A **17**, 1944 (1978).