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2013 Phys. Scr. 2013 014037

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Radiative and collisional spectroscopy of hyperfine lines of the Li-like heavy ions and Tl atom in an atmosphere of inert gases

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Received 20 August 2012

Accepted for publication 4 November 2012

Published 28 March 2013

Online at stacks.iop.org/PhysScr/T153/014037

Abstract

The combined relativistic energy approach and relativistic many-body perturbation theory with an optimized zeroth one-particle approximation are used for the calculation of the Li-like ions ($Z = 11–42, 69, 70$) and thallium atom spectral parameters of radiative transitions from the ground state to the excited state. A key feature of the presented approach is an implementation of the optimized relativistic model potential method in the framework of the energy approach for further studying the spectral and collisional characteristics of the multi-electron atoms. In addition, the method combined with the spectral line kinetic theory is used to study the hyperfine structure collision shift for the Tl atom in an atmosphere of inert gases.

PACS numbers: 32.30.–r, 32.70.Cs, 34.50.Fa

1. Introduction

Theoretical radiative–collisional spectroscopy of heavy neutral and ionized atoms is of fundamental importance in many fields of atomic physics (spectroscopy, spectral lines theory), astrophysics, plasma physics, laser physics and so on. It should be mentioned that correct data about radiative decay widths, probabilities and oscillator strengths of atomic transitions are needed in astrophysics and the laboratory, in thermonuclear plasma diagnostics and in fusion research. In light of this, there is special interest in the study of the spectral characteristics of heavy atoms such as thallium, lanthanides and actinides atoms, and corresponding high- Z multicharged ions such as He-, Li-like ions, etc (see, e.g., [1–34]). Martin and Wiese [1, 2] have performed a critical evaluation and compilation of the spectral parameters for Li-like ions ($Z = 3–28$). The results of high-precision non-relativistic calculations of the energies and oscillator strengths of $1s22s–1s22p$ for Li-like systems up to $Z = 50$ are presented in [9–14]. The Hylleraas-type variational method and the $1/Z$ expansion method have been used. Chen Chao and Wang Zhi-Wen [14] have listed the non-relativistic dipole-length, -velocity and -acceleration absorption oscillator strengths

for the $1s22s–1s22p$ transitions of the LiI isoelectronic sequence on the basis of calculation within a full core plus correlation method using multiconfiguration interaction wave functions. Fully variational non-relativistic Hartree–Fock (RHF) wave functions have been used by Bièmont [16] in the calculation of the $1s2n2L$ ($n < 8 = s, p, d$ or f ; $3 < Z < 22$) states of the LiI isoelectronic sequence. There have been reports of calculations and compilation of energies and transition probabilities for thallium (see, e.g., [30–34]). Known methods such as the RHF and Dirac–Fock (DF) methods, time-dependent relativistic perturbation theory (PT) with the Hartree–Fock (HF) zeroth approximation (TDRHF), relativistic many-body PT (RMBPT) with the DF zeroth approximation, etc have been used. Nevertheless, the great interest in studying the spectral and radiative characteristics of heavy atoms stimulates further development of the theory and obtainment of new spectroscopic data for heavy atoms, including the thallium atom and others. It should be noted that the Tl atom is a very interesting system from the point of view of studying the electroweak $e–N$ interaction and the parity violation effect.

Another topic of our paper is the study of the collisional shifts and broadening of the hyperfine structure lines for

heavy elements (alkali, alkali-earth, lanthanides, actinides and others) in an atmosphere of inert gases which is of great importance to modern quantum chemistry, optics and atomic, molecular spectroscopy, plasma physics, etc [35–45]. The calculation of the hyperfine structure line shift and broadening allows one to check the quality of wave functions and to study the contribution of relativistic effects in two-center (multi-center) atomic systems. From the applied science point of view, the mentioned physical effects form the basis for creating an atomic quantum measure of frequency [42]. For a long time, the corresponding phenomenon for the thallium atom attracted special attention because of the possibility to create the thallium quantum frequency measure. Alexandrov *et al* [42] have realized the optical pumping of the thallium atoms on the line of 21 GHz, which corresponds to a transition between the components of hyperfine structure for the ground state, and have measured the collisional shift of this line due to a buffer (bath) gas. The inert buffer gases (He, Ar, etc) are usually used. The detailed non-relativistic theory of the collisional shift and broadening of the hyperfine structure lines for simple elements (light alkali elements, etc) has been developed by many authors (see discussions in [35–44]). However, consideration of heavy elements faces serious difficulties related to accounting for the relativistic and correlation corrections.

In this paper, the combined relativistic energy approach [23–26] and RMBPT with the optimized zeroth-order one-particle representation and accounting for the exchange-correlation effects as the RMBPT second and higher orders corrections [21, 22, 27–29] are used for the calculation of the high-Z Li-like ions and the thallium atom energies and radiative transition characteristics from the ground state to the excited state. In addition, the method combined with the kinetic theory of spectral lines [37–40] is applied to calculate the hyperfine structure collision shift for the thallium atom in an atmosphere of inert gases. A comparison with the available data for the collisional shifts of the Tl atom is made.

2. The theoretical method

As the relativistic energy approach and RMBPT with an optimized model potential or DF zeroth approximation are described in detail in [4, 5, 21–29], we are limited only by the key aspects. Within the energy approach the imaginary part of the electron energy shift of a multielectron atom is directly connected with a radiation decay possibility (transition probability), which is determined in the PT lowest order as follows [4]:

$$\text{Im}\Delta E(B) = -\frac{e^2}{4\pi} \sum_{\substack{\alpha>n>f \\ [\alpha<n\leq f]}} V_{\alpha n \alpha n}^{|\omega_{\alpha n}|}, \quad (1)$$

where $\sum_{\alpha>n>f}$ for the electron and $\sum_{\alpha<n\leq f}$ for the vacancy (in the atomic core). The matrix element is determined as follows:

$$V_{ijkl}^{|\omega|} = \int \int dr_1 dr_2 \psi_i^*(r_1) \psi_j^*(r_2) \frac{\sin |\omega| r_{12}}{r_{12}} \times (1 - \alpha_1 \alpha_2) \psi_k^*(r_2) \psi_l^*(r_1). \quad (2)$$

The separated terms of the sum in (2) represent the contributions of different channels and a probability of dipole transition is

$$\Gamma_{\alpha_n} = \frac{1}{4\pi} V_{\alpha_n \alpha_n}^{|\omega_{\alpha n}|}. \quad (3)$$

In order to take into account the correlation effects (polarization of the atomic core or screening corrections, etc), one should add the effective polarization (correlation) functional to the potential in the matrix element (3). There are a set of effective exchange-polarization potentials. Below, we use the relativistic polarization potential from [29], which has the following form:

$$V_{\text{pol}}^d(r_1 r_2) = X \left(\int dr' \rho_c^{(0)1/3}(r') \theta(r') / |r_1 - r'| |r' - r_2| - \left(\int dr' \rho_c^{(0)1/3}(r') \theta(r') / |r_1 - r'| \int dr'' \rho_c^{(0)1/3}(r'') \theta(r'') / |r'' - r_2| \right) / \int dr \rho_c^{(0)1/3}(r) \theta(r) \right), \quad (4)$$

$$\theta(r) = \{1 + [3\pi^2 \rho_c^{(0)}(r)]^{2/3} / c^2\}^{1/2},$$

where X is the numerical coefficient, c is the speed of light and $\rho_c^{(0)}$ is the electron core density.

Under calculating the matrix elements (3) one should use the angle symmetry of the task and write the expansion for potential $\sin|\omega|r_{12}/r_{12}$ on the spherical functions as follows [4]:

$$\frac{\sin |\omega| r_{12}}{r_{12}} = \frac{\pi}{2\sqrt{r_1 r_2}} \sum_{\lambda=0}^{\infty} (\lambda) J_{\lambda+1/2}(|\omega| r_1) \times J_{\lambda+1/2}(|\omega| r_2) P_{\lambda}(\cos r_{12}), \quad (5)$$

where $J_{\lambda+1/2}$ is the Bessel function of first kind and $(\lambda) = 2\lambda + 1$. This expansion corresponds to the usual multipole expansion for the probability of radiative decay. Substitution of the expansion (5) into the matrix element of interaction gives

$$V_{1234}^{\omega} = [(j_1)(j_2)(j_3)(j_4)]^{1/2} \sum_{\lambda\mu} (-1)^{\mu} \begin{pmatrix} j_1 j_3 & \lambda \\ m_1 - m_3 & \mu \end{pmatrix} \times \text{Im} Q_{\lambda}(1234), \quad Q_{\lambda} = Q_{\lambda}^{\text{Qu}} + Q_{\lambda}^{\text{Br}}, \quad (6)$$

where j_i is the total single-electron momentum, m_i are the projections, Q_{λ}^{Qu} is the Coulomb part of the interaction and Q_{λ}^{Br} is the Breit part. Both the parts of the Q_{λ}^{Qu} , Q_{λ}^{Br} are expressed in terms of radial integrals R_{λ} and angular coefficients S_{λ} . A detailed description of the corresponding integrals and coefficients is given in [4, 5, 21–25].

The thallium atom can be strictly treated as the three-quasiparticle system, i.e. the system with three valence electrons ($6n_{lj}$, $n \geq 6$) above the relatively rigid core. The relativistic quasiparticle wave functions are calculated by solution of the Dirac equation with the potential, which includes the ‘outer electron-ionic core’ potential and polarization and screening potentials [4, 5, 23, 29]. In order to describe the interaction of the outer electron with the core,

Table 1. Oscillator strengths of the $2s_{1/2}-2p_{1/2,3/2}$ transitions in Li-like ions.

Method	DF [6]	DF [6]	[2]	[2]	This work	This work
Ion	$2s_{1/2}-2p_{1/2}$	$2s_{1/2}-2p_{3/2}$	$2s_{1/2}-2p_{1/2}$	$2s_{1/2}-2p_{3/2}$	$2s_{1/2}-2p_{1/2}$	$2s_{1/2}-2p_{3/2}$
Ca ¹⁷⁺	0.0234	0.0542	0.024	0.054	0.0236	0.0541
Fe ²³⁺	0.0177	0.0482	0.018	0.048	0.0179	0.0481
Zn ²⁷⁺	0.0153	0.0477	–	–	0.0156	0.0475
Mo ³⁹⁺	–	–	0.011	0.056	0.0107	0.0556
Sn ⁴⁷⁺	0.0092	0.0686	–	–	0.0095	0.0684
Tm ⁶⁶⁺	–	–	–	–	0.0071	0.1140
Yb ⁶⁷⁺	0.0067	0.1170	–	–	0.0069	0.1167

Table 2. Oscillator strengths of the $2s_{1/2}-np_{1/2}$ transitions in the Ca¹⁷⁺ ion.

Transition	QDA [6]	DF [6]	[2]	This work
$2s_{1/2}-3p_{1/2}$	–	–	0.123	0.122
$2s_{1/2}-3p_{3/2}$	–	–	0.241	0.243
$2s_{1/2}-4p_{1/2}$	–	–	–	0.029
$2s_{1/2}-8p_{1/2}$	2.54 ^a	2.53 ^a	–	2.55 ^a
$2s_{1/2}-10p_{1/2}$	1.24 ^a	1.24 ^a	–	1.25 ^a

Note: ^a $10^{-3}gf$.

the advanced Ivanova–Ivanov-type model potential [4, 28] is used. Calibration of the single-model potential parameter is performed on the basis of the special *ab initio* procedure in the relativistic energy approach [24]. In [24], the lowest order multielectron effects, in particular, the gauge-dependent radiative contributions $\text{Im} \Delta E_{\text{niv}}$ for a certain class of photon propagator calibration are treated. This value is considered to be representative of the electron correlation effects, whose minimization is a reasonable criterion in the search for the optimal one-electron basis of the RMBPT. The minimization of the density functional $\text{Im} \Delta E_{\text{niv}}$ leads to the integral–differential equation that can be solved using one of the standard numerical codes. Thus, it provides the construction of the optimized one-particle representation. The formalism of the RMBPT with an optimized model DF-like zeroth approximation allows us to take into account exchange-correlation effects as the RMBPT second and higher orders corrections [21–24]. All calculations are performed by using the code Superatom-ISAN (version 93).

3. Hyperfine line shift for the ‘heavy atom-inert gas’ system

To calculate the collision shift of a hyperfine structure spectral line, one could use the following expression known from kinetic theory of the spectral line form (see [35, 40]):

$$f_p = \frac{\Delta}{p} = \frac{4\pi N_b}{p} \int_0^\infty e^{-U(R)/kT} [1 + g(R)] \delta\omega(R) R^2 dR, \quad (7)$$

where $U(R)$ is the effective potential of the interatomic interaction, which has a central symmetry in the case of the systems A–B (in our case, e.g., B = He; A = Tl); T is the temperature, p is the pressure; $\delta\omega(R) = Dw(R)/w_0$ is the relative local shift of the hyperfine structure lines, which arises due to the disposition of the active atoms (say, atoms of thallium Tl and helium He) on a distance R ; w_0 is a frequency of the hyperfine structure transition in the isolated

active atom; $\{1 + g(R)\}$ is a temperature factor. To calculate an effective potential of the interatomic interaction, a method of the exchange PT (the modified version EL-HAV) [35] has been used. Within exactness to second-order terms on the potential of Coulomb interaction of the valence electrons and atomic cores, one can write

$$\delta\omega(R) = \frac{S_0}{1 - S_0} + \Omega_1 + \Omega_2 - \frac{C_6}{R^6} \left(\frac{2}{E_a} + \frac{1}{E_a + E_b} \right), \quad (8)$$

where C_6 is the van der Waals constant for the interaction A–B (e.g., the pair of Tl–He; look below); $I, E_{1a,b}$ are the ionization potential and excitation energy for atoms A and B correspondingly; S_0 is the overlapping integral. The values Ω_1, Ω_2 in the expression (2) are the non-exchange and exchange non-perturbation sums of the first order correspondingly, which are defined as follows:

$$\Omega_1 = \frac{2}{N(1 - S_0)\rho_0} \sum_k \frac{\langle \Phi'_0(1) | H'_{CT} | \Phi'_k(1) \rangle V_{k0}}{E_0 - E_k},$$

$$\Omega_2 = \frac{2}{N(1 - S_0)\rho_0} \sum_k \frac{\langle \Phi'_0(1) | H'_{CT} | \Phi'_k(1) \rangle U_{k0}}{E_0 - E_k}, \quad (9)$$

where H'_{CT} is the operator of hyperfine interaction, N is the total number of electrons taken into account in calculation; $E_k, \Phi'_k(1) = F'_{k_a}(1)\phi_{k_b}(2 \dots N)$ are the energy and non-symmetrized wave function of state $k = \{k_a, k_b\}$ for isolated atoms A and B.

The non-exchange matrix element of the Coulomb interatomic interaction is as follows:

$$V_{k0} = \langle \Phi'_k(1) | V(1) | \Phi'_0(1) \rangle.$$

Correspondingly, the exchange matrix element is as follows:

$$U_{k0} = \sum_{i=2}^N \langle \Phi'_k(1) | V(i) | \Phi'_0(i) \rangle.$$

The operator $V(i)$ in the case of the system Tl–He is as follows:

$$V(i) = U_{\text{SCF}}(r_{a3}) + U_{\text{SCF}}(r_{a4}) - 2U_{\text{SCF}}(R) + \frac{1}{r_{bi}}, \quad (10)$$

where $U_{\text{SCF}}(r)$ is the self-consistent field, created by the thallium atomic core.

Usually (see [2]), the non-RHF sets of the wave functions are used. A more sophisticated approach is based on using the relativistic DF wave functions (the first variant) [40, 45].

Table 3. Energies of levels (in cm^{-1}) for the thallium atom Tl: the experimental data [34] and theoretical results obtained by different theoretical methods (see the text).

State	RHF	TDRHF	RMBPT-DF	This work	Experiment [34]
$7s_{1/2}$	-21 100	-22 952	-22 818	-22 799	-22 786
$6p_{1/2}$	-43 909	-50 654	-49 266	-49 295	-49 264
$7p_{1/2}$	-14 282	-15 203	-	-15 142	-15 104
$6p_{3/2}$	-36 670	-42 704	-41 432	-41 497	-41 471
$7p_{3/2}$	-13 359	-14 224	-	-14 135	-14 103
$6d_{3/2}$	-12 218	-13 130	-13 175	-13 167	-13 146
$6d_{5/2}$	-12 167	-13 042	-	-13 083	-13 064

Table 4. The shift f_p (Hz/Torr) of the Tl hyperfine line for the pairs: Tl–He, Kr, Xe ($T = 700$ K).

System	Tl–He	Tl–Ar	Tl–Xe
Experiment	130 ± 30	-490 ± 20	-1000 ± 80
Theory [41]	155.0	-850.0	-1420.0
Theory [39]	137.2	-504	-1052
Our theory	135.4	-501	-1044

Another variant is using the relativistic wave functions as the solutions of the Dirac equations with different density functionals (the Kohn–Sham density-functional theory) and effective potentials [37–41, 43]. In this paper, we have used the set of relativistic functions, generated by the Dirac equation with the optimized model potential [28, 39]. The detailed approbation of this model potential in studying spectra and radiative characteristics of the ytterbium and thallium atoms is given in [37, 38], too. In a number of papers, it has been strictly shown that using the optimized sets for calculating the atomic electron density-dependent properties has a very important role (see discussions in [21, 22, 24]). In this work, we will not discuss this question in detail.

4. Results

We applied the above-described approach to calculating the energies and oscillator strengths of transitions in spectra of the Li-like ions ($Z = 11–42, 69, 70$). There are studied the radiative transitions from the ground state to the low-excited and Rydberg states, especially, $2s_{1/2}–np_{1/2,3/2}$, $np_{1/2,3/2}–nd_{3/2,5/2}$ ($n = 2–12$). To test the obtained results, we compare our calculation data on the oscillator strengths values for some Li-like ions with the known theoretical and compiled results [1, 2, 6, 8].

As an example, in table 1, we present the oscillator strengths for the $2s_{1/2}–2p_{1/2,3/2}$ transitions in Li-like ions S^{13+} , Ca^{17+} , Fe^{23+} , Zn^{27+} , Zr^{37+} , Mo^{39+} , Sn^{47+} , Tm^{66+} , Yb^{67+} . The DF calculation data of Zilitis [6] and the ‘best’ compiled (experimental) data [1, 2] for the low- Z Li-like ions are listed in table 1 for comparison, too. It should be remembered that experimental data on the oscillator strengths for many (especially high- Z) Li-like ions are absent hitherto. On the whole, there is physically reasonable agreement between the listed data. The important features of the approach applied are using the optimized one-particle representation and accounting for the polarization (exchange-correlation) effects. It should be noted that the estimate of the gauge-non-invariant contributions (the difference between the oscillator strengths calculated using the transition operator in the form of ‘length’

and ‘velocity’) is about 0.3%. The results obtained using different photon propagator gauges (Coulomb, Babushkon and Landau) are practically equal. In table 2, we present the oscillator strengths values for the $2s_{1/2}–np_j$ ($n = 3–12, j = 1/2, 3/2$) transitions in the spectrum of the Ca^{17+} ion.

The quantum defect approximation (QDA) [6, 30] and the DF oscillator strengths calculation results obtained by Zilitis [6] and some compiled (experimental) data obtained by Martin–Weiss [1, 2] are also listed in table 2 for comparison. It is self-understood that the QDA oscillator strengths data become more exact with the growth of the principal quantum number. At the same time the accuracy of the DF data may be decreased. The agreement between the Martin–Weiss data and our results for the transitions between low-lying terms is sufficiently good. In table 3, we list the energy level values for low-excited states of the thallium atom, obtained on the basis of calculation within different theoretical methods: RHF, TDRHF, RMBPT-DF [30–34, 46] and our approach. The experimental data from [34] are presented too. The general conclusion is that accurately accounting for the exchange-correlation and relativistic effects allows us to obtain the results that are in physically reasonable agreement with the experimental data.

In table 4, we present our theoretical results for the shift f_p (Hz/Torr) for the Tl–He, Ar, Xe pairs. The observed value of the line shift ($T = 700$ K) and other theoretical results with using DF and optimized DF methods are also listed in table 4 [39, 41].

An important feature of our scheme is correctly accounting for the correlation and polarization effects using special effective functionals. The difference between our data and other theoretical data is explained using different sets of relativistic wave functions and different PT schemes.

Acknowledgment

The authors thank the anonymous referee for useful comments and recommendations.

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