Radiative Lifetimes of Rydberg States in Neutral Gallium

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Radiative lifetimes were calculated using weakest bound electron potential model theory for $4s^2\ ns\ ^2S_{1/2}$ $(n \geq 7)$, $4s^2\ np\ ^2P^0_{1/2}$ $(n \geq 5)$, $4s^2\ np\ ^2P^0_{3/2}$ $(n \geq 6)$, $4s^2\ nd\ ^2D^0_{3/2}$ $(n \geq 6)$, $4s^2\ nd\ ^2D^0_{5/2}$ $(n \geq 6)$ series in neutral gallium. The use of the quantum defect theory and Martin's expressions allowed us to supply lifetime values along by means of the series above. The results obtained in this work presented good agreement with theoretical and experimental values. Moreover, some lifetime values not existing in the literature for highly excited Rydberg states in gallium atom were obtained using this method.

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1. Introduction

With the advent of the better laser systems and other optical devices, spectroscopic parameters such as transition probabilities and atomic lifetimes can be studied in highly excited atomic and ionic systems particularly on the high-lying Rydberg states. The determination of spectroscopic data for neutral and ionized atomic systems has been an active research area in astrophysics, plasma physics, laser physics and thermonuclear fusion research. It is important to know the lifetime of highly excited Rydberg atoms, which is a parameter that can be compared to existing theories in the literature. During the last two decades, the most of researchers have studied the transition probabilities, energy levels, oscillator strengths and lifetimes of excited states belonging to gallium being in group-three elements having a ground state configuration of $4s^2$ of $4p$ [1–5]. Especially, Zheng et al. have calculated energy values using fit procedure in gallium atom. They have considered four coefficients in Martin's expression [5]. Particularly, lifetimes research in high Rydberg states is important, because the exact lifetime measure are used in astrophysics, laser physics, physical chemistry [6, 7]. The most of investigations are limited to few low excitation radiative lifetimes ($n \leq 8$–12). Many of the modern experimental techniques still encounter difficulties in the exact measurement of spectroscopic parameters such as oscillator strengths, transition probabilities and lifetimes. The choices of theoretical methods for calculation of the spectroscopic data in many electron atoms and heavy ions are not obvious and not so easy. The problem can be solved more or less exactly for systems having few electrons such as helium. More general multi-electron systems cannot be treated with such precision. It is impossible yet to solve many electron systems without imposing severe approximations. Many theoretical methods exist for calculation of spectroscopic parameters for atomic or ionic systems such as the Hartree–Fock approximations, configuration interaction methods, semi-empirical methods and many-body perturbation theories. The calculation of some physical parameters corresponding to individual energy levels, especially highly lying levels and the Rydberg states are always difficult problems in theoretical studies, because of indistinguishability of equivalent electrons and the necessity of taking into account many configurations or orbital basis-set functions [8]. Therefore, semi-empirical methods where one or more parameters are needed to be adjusted according to the existing experimental data can be useful [9–11].

In the present paper, we have studied the lifetimes of some high Rydberg levels in neutral gallium using the weakest bound electron potential model theory (WBEPMT). The results were listed here for atoms having the principal quantum number up to $n = 50$. Our theoretical procedure is described in Sect. 2, we present new spectroscopic results. Conclusive remarks are given in Sect. 3.

2. The method

The WBEPMT has been developed [12, 13] and applied for determination of some physical parameters in many-electron atomic and ionic systems by Zheng et al. [14–19]. Zheng suggested a new model potential to describe the electronic motion in multi-electron systems and separated electrons within a given system into two groups, the weakest bound electron (WBE) and non-weakest bound electrons (NWBE). The WBE in many-

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-electron systems is also the electron which can most easily be excited or ionized. According to the WBEPM theory, the WBE moves in the potential field produced by the nucleus and the non-weakest bound electron. This potential field can be divided into two parts, one of which is the Coulomb potential. The second part is the dipole potential. This dipole moment affects the behavior of the WBE. It would make the principal quantum number n and the angular momentum quantum number l of the WBE to be the effective principal quantum number n^* and effective angular momentum quantum number l^*. The introduction of d effectively modifies the integer quantum numbers n and l into non-integers n^* and l^*. These terms differ from the usual core polarization potential which behaves as $1/r^4$ asymptotically, in that the effective dipole moment of the core is used as a parameter rather than being derived from the polarizability of the core in the electric field of the WBE. Some atomic or ionic properties in multi-electron systems such as transition, excitation and ionization may be referred to weakest bound electron's behavior. The treatment of WBE gives some accurate information about these properties [12–19].

In the WBEPM theory, the Schrödinger equation of the weakest bound electron under non-relativistic approximation is given as [5, 17, 18]:

$$\left[-\frac{1}{2}\nabla^2 + V(r_i)\right]\psi_i = \varepsilon_i\psi_i, \tag{1}$$

$$V(r_i) = -\frac{Z^*}{r_i} + \frac{d(d+1)+2dl}{2r_i^2}. \tag{2}$$

In Eq. (2), $V(r_i)$ is potential function produced by the non-weakest bound electrons and nucleus. Moreover, Z^* is the effective nuclear charge, r_i is the distance between the weakest bound electron i and nucleus. In this theory, electronic radial wave functions are presented as a function of the Laguerre polynomial in terms of some parameters [5, 17, 18]:

$$R_{nl}(r) = \left(\frac{2Z^*}{n^*}\right)^{l^*+3/2}\left[\frac{2n^*\Gamma(n^*+l^*+1)}{(n-l-1)!}\right]^{-1/2} \times \exp\left(-\frac{Z^*r}{n^*}\right) r^{l^*} L_{n-l-1}^{2l^*+1}\left(\frac{2Z^*r}{n^*}\right). \tag{3}$$

n^* and l^* parameters have been given to be

$$n^* = n + d \quad \text{and} \quad l^* = l + d. \tag{4}$$

The energy expression of weakest bound electron is given as

$$\varepsilon = -\frac{Z^{*2}}{2n^{*2}}, \tag{5}$$

where both Z^* and d are unknown parameters. This complication could be removed making Eq. (5) similar to quantum defect theory. The energy expression in the quantum defect theory is [5, 20]:

$$\varepsilon = -\frac{Z_{\text{net}}^2}{2n^{*2}}, \tag{6}$$

where $n^* = n - \delta$ and δ is quantum defect. δ is approximately constant for a given fixed orbital quantum number. Z_{net} is the ion core charge. For a neutral atom Z_{net} is 1 and for a singly charged ion Z_{net} is 2 etc. For neutral atoms, energy eigenvalue can be given as

$$\varepsilon(n) = -\frac{1}{2(n-\delta)^2}, \tag{7}$$

where n is the principal quantum number. Also, δ quantity in Eq. (7) can be determined by Martin's expression [5, 21]:

$$\delta = a + b(n-\delta_0)^{-2} + c(n-\delta_0)^{-4} + d(n-\delta_0)^{-6} + e(n-\delta_0)^{-8}, \tag{8}$$

where δ_0 is quantum defect of lowest energy state in the given Rydberg series and it is a constant. Coefficients a, b, c, d, e can be determined from the first four experimental values by solving Eq. (7) and Eq. (8). The energy levels of high Rydberg states can be obtained by extrapolating with these values. Martin has been applied to sodium atom for analysis of spectra and the results have been obtained in high precision.

The lifetime of excited levels for many electron atomic systems can be calculated using the following formula [5, 22]:

$$\tau = \tau_0(n^*)^\alpha, \tag{9}$$

where the parameters τ_0 and α are coefficients in relevant series. According to the WBEPM theory, these coefficients will be determined using the experimental values of energy and lifetime.

3. Results and conclusions

The coefficients a, b, c, d, e in Eq. (8) and the values of δ_0 belonging to different series are fitted to experimental energy values [23] in the gallium atom and these are given in Table I and Table II. In this paper, we have considered five coefficients by adding one coefficient to the coefficients considered by Zheng et al. [5] in Martin's expression to determine energy values. τ_0 and α coefficients have been calculated using the WBEPM theory. We have calculated the values of energy levels in the gallium atom and compared them with the experimental values taken from NIST. These are listed in Tables III–VII. For some levels, where no documented values exist, our calculated values have not been compared. Even though in some cases the deviations are 5–6 cm^{-1}, it can be seen that most of the deviations are less than 1 cm^{-1} in considering all data. Radiative lifetime values of six Rydberg series $4s^2\ ns\ ^2S_{1/2}$ ($n \geq 7$), $4s^2\ np\ ^2P^0_{1/2}$ ($n \geq 5$), $4s^2\ np\ ^2P^0_{3/2}$ ($n \geq 6$), $4s^2\ nd\ ^2D^0_{3/2}$ ($n \geq 6$), $4s^2\ nd\ ^2D^0_{5/2}$ ($n \geq 6$) have been calculated in Ga(I). Available experimental and theoretical data are quite limited for high Rydberg states. Therefore, our lifetime results obtained from the WBEPM theory have been compared with relativistic many-body perturbation theory (RMBPT) results given by Safronova et al. [24] and the results of Carlsson et al. including both experimental and theoretical data [25].

TABLE I

Spectral coefficients of energy level series for Ga(I).

Series	a	b	c	d	e	δ_0
$4s^2\ ns\ ^2S_{1/2}\ (n \geq 7)$	2.75309	6.70038	−328.8181	6812.0098	−49791.45	2.8026804
$4s^2\ np\ ^2P^0_{1/2}\ (n \geq 5)$	2.26856	4.12798	−103.6777	1053.8857	−3592.08	2.3256794
$4s^2\ np\ ^2P^0_{3/2}\ (n \geq 6)$	2.12244	18.82179	−752.8694	12578.377	−73587.37	2.2893749
$4s^2\ nd\ ^2D^0_{3/2}\ (n \geq 6)$	1.12970	14.7326	−830.4935	20096.877	−175367.05	1.22481943
$4s^2\ nd\ ^2D^0_{5/2}\ (n \geq 6)$	1.16642	8.982337	−523.2097	12995.02111	−115674.76	1.22217292

TABLE II

The coefficients of lifetime for Ga(I).

Series	τ_0	α
$4s^2\ ns\ ^2S_{1/2}\ (n \geq 7)$	1.031116130	2.647711554
$4s^2\ np\ ^2P^0_{1/2}\ (n \geq 5)$	2.769519725	3.126649554
$4s^2\ np\ ^2P^0_{3/2}\ (n \geq 6)$	2.648067267	3.151399018
$4s^2\ nd\ ^2D^0_{3/2}\ (n \geq 6)$	0.2218400714	3.716096661
$4s^2\ nd\ ^2D^0_{5/2}\ (n \geq 6)$	0.2207353275	3.717972243

TABLE III

Energy values and radiative lifetimes for Ga(I): $4s^2\ ns\ ^2S_{1/2}\ (n \geq 7)$ series.

n	$E_{\rm exp}$ [cm^{-1}] (Ref. [23])	$E_{\rm cal}$ [cm^{-1}]	This work $\tau_{\rm cal}$ [ns]	RMBPT results (Ref. [24])	Exp. results (Ref. [25])	MCHF results (Ref. [25])	Corrected for black-body radiation from Ref. [25]
7	42158	42158	46	44.1	49	40.3	49
8	44332	44325	81	94.5	80	70.3	80
9	45536	45530	129		135	111	136
10	46274	46269	192		200	169	204
11	46758	46754	270		250	247	257
12	47093	47090	367			346	
13	47334	47332	482			476	
14	47514	47512	618				
15	47651	47650	775				
16	47758	47757	955				
17	47844	47843	1.16×10^3				
18	47913	47912	1.39×10^3				
19	47970	47969	1.64×10^3				
20	48017	48016	1.92×10^3				
30		48239	6.48×10^3				
40		48308	1.48×10^4				
50		48338	2.80×10^4				

TABLE IV

Energy values and radiative lifetimes for Ga(I): $4s^2\ np\ ^2P^0_{1/2}$ ($n \geq 5$) series.

n	$E_{\rm exp}$ [cm^{-1}] (Ref. [23])	$E_{\rm cal}$ [cm^{-1}]	This work $\tau_{\rm cal}$ [ns]	RMBPT results (Ref. [24])	Exp. results (Ref. [24]) $\tau_{\rm exp}$ [ns]	MCHF results (Ref. [25])	Corrected for black-body radiation from Ref. [25]
5	33044	33044	60	51.3	55	48	55
6	40376	40259	162	162	172	142	172
7	43442	43365	344	326		316	384
8		44979	630	624		592	690
9		45924	1047			1010	1010
10		46524	1620			1580	1700
11		46929	2376			2340	2500
12		47215	3342			3300	
13		47424	4.54×10^3				
14		47582	6.01×10^3				
15		47704	$7.77.\times10^3$				
16		47757	9.86×10^3				
17		47878	1.23×10^4				
18		47940	1.51×10^4				
19		47992	1.83×10^4				
20	48038	48036	2.20×10^4				
30		48244	8.94×10^4				
40		48310	2.34×10^5				
50	48339	48339	4.90×10^5				

TABLE V

Energy values and radiative lifetimes for Ga(I): $4s^2\ np\ ^2P^0_{3/2}$ ($n \geq 6$) series.

n	$E_{\rm exp}$ [cm^{-1}] (Ref. [23])	$E_{\rm cal}$ [cm^{-1}]	This work $\tau_{\rm cal}$ [ns]	RMBPT results (Ref. [24])	Exp. results (Ref. [25])	MCHF results (Ref. [25])	Corrected for black-body radiation from Ref. [25]
6	40417	40417	165	156	167	142	167
7	43462	43442	350	314		316	384
8		45022	642	526		592	690
9		45950	1067			1010	1010
10		46541	1653			1580	1700
11		46941	2428			2340	2500
12		47223	3420			3300	
13		47431	4.65×10^3				
14		47587	6.17×10^3				
15		47708	7.99×10^3				
16		47803	1.01×10^4				
17		47880	1.26×10^4				
18		47943	1.55×10^4				
19		47994	1.89×10^4				
20	48038	48037	2.27×10^4				
30		48244	9.31×10^4				
40		48310	2.46×10^5				
50	48339	48339	5.16×10^5				

TABLE VI

Energy values and radiative lifetimes for Ga(I): $4s^2\ nd\ ^2D^0_{3/2}$ ($n \geq 6$) series.

n	$E_{\rm exp}$ [cm^{-1}] (Ref. [23])	$E_{\rm cal}$ [cm^{-1}]	This work $\tau_{\rm cal}$ [ns]	RMBPT results (Ref. [24])	Exp. results (Ref. [25]) $\tau_{\rm exp}$ [ns]	MCHF results (Ref. [25])	Corrected for black-body radiation from Ref. [25]
6	43575	43575	74	41.9	69	74	69
7	45071	45097	150	72.9	150	169	151
8	45969	45997	271		270	282	275
9	46547	46572	453		460	400	475
10	46943	46962	710			525	
11	47222	47239	1060			652	
12	47428	47442	1.52×10^3				
13	47585	47596	2.12×10^3				
14	47706	47715	2.86×10^3				
15	47802	47809	3.79×10^3				
16	47878	47884	4.92×10^3				
17	47941	47946	6.28×10^3				
18	47993	47997	7.88×10^3				
19	48036	48040	9.78×10^3				
20	48072	48076	1.20×10^4				
30	48255	48255	5.85×10^4				
40		48314	1.77×10^5				
50		48341	4.16×10^5				

TABLE VII

Energy values and radiative lifetimes for Ga(I): $4s^2\ nd\ ^2D^0_{5/2}$ ($n \geq 6$) series.

n	$E_{\rm exp}$ [cm^{-1}] (Ref. [23])	$E_{\rm cal}$ [cm^{-1}]	This work $\tau_{\rm cal}$ [ns]	RMBPT results (Ref. [24])	Exp. results (Ref. [25]) $\tau_{\rm exp}$ [ns]	MCHF results (Ref. [25])	Corrected for black-body radiation from Ref. [25]
6	43580	43580	74	41.5	69	74	69
7	45075	45100	150	81.8	150	169	151
8	45972	45998	271		270	282	275
9	46549	46573	453		460	400	475
10	46943	46963	710			525	
11	47222	47239	1060			652	
12	47428	47442	1.52×10^3				
13	47585	47596	2.12×10^3				
14	47707	47715	2.87×10^3				
15	47802	47809	3.79×10^3				
16	47879	47885	4.92×10^3				
17	47941	47946	6.28×10^3				
18	47993	47997	7.89×10^3				
19	48036	48040	9.79×10^3				
20	48073	48076	1.20×10^4				
30	48254	48255	5.86×10^4				
40		48314	1.78×10^5				
50		48341	4.17×10^5				

Carlsson et al. employed experimental laser spectroscopic techniques. It is known that the measuring of radiative lifetimes is normally performed at room temperature. In this case, there will be a background field due to present black-body radiation in the measuring volume [25]. This field is very weak at optical frequencies, but as one goes into the infrared region it will increase in strength. Since the frequencies for transitions between different high-lying Rydberg states are often in the infrared region, transitions will be induced by this black--body background field. The measured values will be shorter than the values measured without the black-body induced transitions. Therefore, some estimates and corrections were done for this effect by Carlsson et al. These values are presented in our tables. At the same paper, Carlsson et al. used the multi-configurational Hartree–Fock (MCHF) wave functions to calculate lifetime values in a frozen-core approximation [25]. Their theoretical results are given as a comparable data in Tables III–VII.

While the calculation procedure for the systems with a few electrons can be carried out easily, the calculations have become more difficult and complex in the case of increasing number of electrons. Especially, for the excited states and Rydberg states of many-electron systems, more configurations must be considered. Therefore, calculations become more complicated. Because of the difficulties mentioned above, the theoretical and experimental studies generally considers low lying states rather than highly excited states. It can be seen from tables that our results are very close to the corresponding theoretical and experimental results. These results prove that Martin's expression is convenient for the Rydberg series of Ga(I). The WBEPM theory has a simple calculation procedure. It can be used to calculate the lifetimes for both highly excited states and low lying states without any increase in complexity in calculation process. Previously, we have employed the WBEPM theory for several physical parameters such as transition probabilities, oscillator strengths and ionization potentials in many-electron atoms. We have obtained very satisfactory results [26–30]. In this study, by courtesy of this method, we have calculated higher lifetimes than published in the literature in Ga(I).

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