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RELATIVISTIC CALCULATION OF THE RADIATIVE TRANSITION PROBABILITIES AND LIFETIMES OF EXCITED STATES FOR THE RUBIDIUM ATOM IN A BLACK-BODY RADIATION FIELD

We present the results of relativistic calculation of the radiative transition probabilities and excited states lifetimes for a heavy Rydberg atomic systems in a black-body (thermal) radiation field on example of the rubidium. As theoretical approach we apply the combined generalized relativistic energy approach and relativistic many-body perturbation theory with ab initio Dirac zeroth approximation. There are obtained the calculational data for the radiative transition probabilities and excited states lifetimes, in particular, the rubidium atom in the Rydberg states with principal quantum number $n=10-100$. It is carried out the comparison of obtained theoretical data on the effective lifetime for the group of Rydberg nS states of the rubidium atom at a temperature of $T = 300K$ with experimental data as well as data of alternative theoretical calculation based on the improved quasiclassical model. It is shown that the accuracy of the theoretical data on the radiative transition probabilities and excited states lifetimes is provided by a correctness of the corresponding relativistic wave functions and accounting for the exchange-correlation effects.

1. Introduction

In the last two decades, a new field of research has been very actively developed in modern optics and spectroscopy, namely quantum optics and spectroscopy of Rydberg atoms (RA) and ions. These are atomic systems that are in highly excited states with large values of the principal quantum number n . By the way, it is known that a number of astrophysical processes in interstellar gas, which interacts with fragments of supernova explosions, collisions of interstellar clouds, and supersonic gas flows during star formation lead to PA with very large n (~ 1000). The reasons for the significant interest in RA are well known and are connected, first, with the properties extraordinary for ordinary atoms, namely: for the lifetime: $n^3 a_0 \hbar / e^2 Z$ ($\tau \sim n^3$ for $l \sim 1$, $\tau \sim n^5$ for $n \sim l$; l --orbital quantum number), geometric dimensions $\sim n^2$ ($n^2 a_0 / Z$, $a_0 = \hbar^2 / me^2 = 0,5291773 \text{ \AA}$), geometric cross sections $\pi n^4 a_0^2 / Z^2$, Rydberg electron binding energies $Z^2 R_\infty / n^2$ ($R_\infty = 13.6058 \text{ eV}$), polarizabilities $\sim n^2$, dipole moments of radiation transitions $\sim n^2$, shifts due to external fields (Stark shift $\sim n^7 l^5$, Zeeman effect $\sim n^2$). Moreover, the idea of

the existence of the fifth state of matter, namely, the Rydberg substance, is well-founded. As a result, in recent years it has stimulated intensive research in the field of standard fundamental spectroscopy of RA, related to the calculation of energy and spectroscopic parameters of these atoms, important for the general development of relativistic (quantum-electrodynamic, QED) theory of atomic spectra, and applied research in fields of quantum optics, computer science, cryptography, quantum computing (e.g. [1-28]).

The vast majority of existing papers on the description of Rydberg atoms in the thermal radiation field (c.g. [1-32]) are based on the Coulomb approximation (CA), model potential (MP) approach, , different versions of the quantum defect method, classical and quasiclassical model approaches. The authors of the papers [3-10] applied the Coulomb approximation, quantum defect formalism, different versions of the model and pseudo-potential method etc. It should be noted separately the cycles of theoretical and experimental works by Nishimoto et al and especially Ryabtsev-Beterov et al [2,3], as well as theoretical works of Dyachkov-Pankratov and others (c.g.[1-10]), in which the ad-

vanced versions of a quasi-classical approach to the calculation of radiation amplitudes, oscillator strengths, and cross-sections for the Rydberg atoms in the BBR radiation field were actually developed.

In the papers [1-3,7-10] the authors present the calculational data on the ionization rates for Rydberg atoms of alkali elements (lithium, sodium, potassium, caesium) by a BBR radiation field. The calculations were carried out for the nS, nP, and nD states in the wide range of principal quantum numbers and temperatures. The above theoretical works and relevant models were substantially based on non-relativistic approximation. At the same time one should note that for heavy Rydberg atoms (both in the free state and in an external electromagnetic field) it is fundamentally important to accurately account for both relativistic and exchange-correlation effects.

In this paper the results of relativistic calculation of the radiative transition probabilities and excited states lifetimes for a heavy Rydberg atomic systems in a black-body (thermal) radiation field are presented on example of the rubidium atom. As theoretical approach we apply the combined generalized relativistic energy approach and relativistic many-body perturbation theory with ab initio Dirac zeroth approximation. There are obtained the calculational data for the radiative transition probabilities and excited states lifetimes, in particular, the rubidium atom in the Rydberg states with principal quantum number $n=10-100$. It is carried out the comparison of obtained theoretical data on the effective lifetime for the group of Rydberg nS states of the rubidium atom at a temperature of $T = 300\text{K}$ with experimental data as well as data of alternative theoretical calculation based on the improved quasiclassical model. .

2. Relativistic theory of multielectron atom in a Black-body radiation field: Some theoretical aspects

From the physical viewpoint, a qualitative picture of the BBR Rydberg atoms ionization

is easily understandable. Even for temperatures of order $T=10^4$ K, the frequency of a greater part of the BBR photons ω does not exceed 0.1 a.u. Usually, it is enough to use a single- electron approximation for calculating the ionization cross section $\sigma_{nl}(\omega)$.

The latter appears in a product with the Planck's distribution for the thermal photon number density:

$$\rho(\omega, T) = \frac{\omega^2}{\pi^2 c^3 [\exp(\omega / kT) - 1]}, \quad (1)$$

where $k=3.1668 \times 10^{-6}$ a.u., K^{-1} is the Boltzmann constant, $c = 137.036$ a.u. is the speed of light. Ionization rate of a bound state nl results in the integral over the Blackbody radiation frequencies:

$$P_{nl}(T) = c \int_{|E_{nl}|}^{\infty} \sigma_{nl}(\omega) \rho(\omega, T) d\omega. \quad (2)$$

The ionization cross-section from a bound state with a principal quantum number n and orbital quantum number l by photons with frequency ω is as follows:

$$\sigma_{nl}(\omega) = \frac{4\pi^2 \omega}{3c(2l+1)} [LM_{nl \rightarrow El-1}^2 + (l+1)M_{nl \rightarrow El+1}^2], \quad (3)$$

where the radial matrix element of the ionization transition from the bound state with the radial wave function $R_{nl}(r)$ to continuum state with the wave function $R_{El}(r)$ normalized to the delta function of energy. The corresponding radial matrix elements are written by the standard way. Other details can be found in Refs. [9-16].

We apply a generalized energy approach [9-20] and relativistic perturbation theory with the zeroth approximation [21-32] to computing the Rydberg atoms ionization parameters. According to Ref. [11,22], the RMBPT zeroth order Hamiltonian of the Rydberg atomic system is as follows:

$$H_0 = \sum_i \{ \alpha c p_i - \beta m c^2 + [-Z / r_i + U_{MF}(r_i | b) + V_{XC}(r_i)] \} \quad (4)$$

where c is the velocity of light, α_i, α_j – the Dirac matrices, ω_{ij} – the transition frequency, Z is a charge of atomic nucleus. The general potential in (4) includes self-consistent Coulomb-like mean-field potential $U_{MF}(r_i | b)$, arbitrary one-particle exchange-correlation (relativistic generalized exchange Kohn-Sham potential plus generalized correlation Lundqvist-Gunnarsson potential) $V_{XC}(r_i | b)$ with the gauge calibrated parameter b (it is determined within special relativistic procedure on the basis of relativistic energy approach; c.g. [21-32]).

The perturbation operator is as follows:

$$H^{PT} = \sum_{i>j} \exp(i\omega_{ij}r_{ij}) \cdot \frac{(1 - \alpha_i \alpha_j)}{r_{ij}} - \sum_i [U_{MF}(r_i) + V_{XC}(r_i | b)] \quad (5)$$

The multielectron interelectron exchange-correlation effects (the core polarization and screening effects, continuum pressure etc) are taken into consideration as the RMBPT second and higher orders contributions. The details of calculation of the corresponding matrix elements of the polarization and screening interelectron interaction potentials are described in Refs. [9,22,33-38].

In relativistic theory radiation decay probability (ionization cross-section etc) is connected with the imaginary part of electron energy shift. The total energy shift of the state is usually presented in the form: $\Delta E = \text{Re}\Delta E + i \Gamma/2$, where Γ is interpreted as the level width, and a decay probability $P = \Gamma$. The imaginary part of electron energy shift is defined in the PT lowest order as:

$$\text{Im}\Delta E(B) = -\frac{e^2}{4\pi} \sum_{\substack{\alpha>n>f \\ [\alpha<n\leq f]}} V_{\alpha n \alpha n}^{|\omega_{\alpha n}|} \quad (6)$$

where ($\alpha>n>f$) for electron and ($\alpha<n<f$) for vacancy.

The matrix element is determined as follows:

$$V_{ijkl}^{|\omega|} = \iint dr_1 dr_2 \Psi_i^*(r_1) \Psi_j^*(r_2) \frac{\sin|\omega|r_{12}}{r_{12}} (1 - \alpha_1 \alpha_2) \Psi_k^*(r_2) \Psi_l^*(r_1) \quad (7)$$

Their detailed description of the matrix elements and procedure for their computing is presented in Refs. [16-20]. The relativistic wave functions are calculated by solution of the Dirac equation with the potential, which includes the Dirac-Fock consistent field potential and additionally polarization potential [22].

The total ionization rate of the Rydberg atomic system in the BBR radiation field is usually determined as the sum of direct BBR ionization rate of the initially excited state, the ionization (field ionization) rate of highly excited states, which are populated from the initial Rydberg state via absorption of the BBR photons, the rate of direct BBR-induced ionization of atoms from the neighbouring Rydberg states and the rate of field ionization of high-lying Rydberg states (with populating through so called two-step process via the BBR photons absorption).

The total width of the Rydberg state (naturally isolated from all external electromagnetic fields except BBR one) consists, apparently, of natural, spontaneous radiation width Γ_{nl}^{sp} and BBR-induced (thermal) width Γ_{nl}^{BBR} :

$$\Gamma_{nl}^{tot} = \Gamma_{nl}^{sp} + \Gamma_{nl}^{BBR}(T). \quad (8)$$

Accordingly, the effective lifetime of the Rydberg state is inversely proportional to the total decay rate as a result of spontaneous transitions and transitions induced by the BBR radiation:

$$\frac{1}{\tau_{eff}} = \Gamma_0 + \Gamma_{BBR} = \frac{1}{\tau_0} + \frac{1}{\tau_{BBR}} \quad (9)$$

The detailed procedures of calculation of the radial and angular integrals (amplitudes) in

the matrix elements are described in the Refs. [16-44]. All calculations are performed on the basis of the code Superatom.

3. Results and conclusions

In Table 1 we present our theoretical data (Our) values of the lifetimes (in ns) of the group of some excited states of the rubidium atom, as well as experimental data and alternative theoretical results obtained on the basis of different approaches: the Coulomb

Table 1.

The theoretical and experimental lifetimes (ns) of the group of the Rb some excited states (see text)

level	QA-MP	PTDF ^{SD}	PT-DF ^{SDc}
6s _{1/2}	-	45.4	45.4
7s _{1/2}	-	88.3	88.3
8s _{1/2}	-	161.8	161.9
9s _{1/2}	266.36	-	271.7
10s _{1/2}	417.84	-	426
6p _{1/2}	-	123	122.5
6p _{3/2}	-	113	112.4
7p _{1/2}	-	280	277.8
7p _{3/2}	-	258	255.2
8p _{1/2}	-	508	501.0
8p _{3/2}	-	471	464.2
7d _{3/2}	331.08	-	339.5
7d _{5/2}	319.57	-	327.0
level	MP-EA	Our	Exp.
6s _{1/2}	45.5	45.5	45.57(17)
7s _{1/2}	88.1	88.2	88.07(40)
8s _{1/2}	161.4	161.5	161(3)
9s _{1/2}	262.1	262.1	253(14)
10s _{1/2}	421.3	426.4	430(20)
6p _{1/2}	124.1	124.1	125(4)
6p _{3/2}	112.1	112.6	112(3)
7p _{1/2}	274.3	274.1	272(15)
7p _{3/2}	249.0	248.8	246(10)
8p _{1/2}	497.4	496.9	
8p _{3/2}	456.2	455.1	400(80)
9p _{1/2}	796.4	795.6	
9p _{3/2}	743.6	742.7	665(40)
10p _{1/2}	964.2	963.8	
10p _{3/2}	921.0	920.5	
7d _{3/2}	336.2	340.5	345(9)

7d _{5/2}	324.8	328.1	325(22)
8d _{3/2}	488.3	490.4	515(30)
8d _{5/2}	431.1	432.8	421(25)

approximation (QA) and model potential (MP), and PTDFSD (many-body perturbation theory with Dirac-Fock SD zero approximation) plus the same data of this method with compilation contribution; method of model potential with the relativistic energy approach (REA) - MP-REA - (data from Refs. [1-5]).

Analysis of the data in Table 1 shows that the error in calculating the lifetime of different levels in the rubidium atom obviously depends on the accuracy, consistency and correctness of the main relativistic and correlation corrections, and above all, exchange-polarization.

Next, we present the results of calculating the effective lifetime of the Rydberg states of the rubidium atom depending on the principal quantum number at the fixed temperature T. In Figure 1, we present theoretical data (dashed line) on the dependence of the values of the effective lifetime for the group of Rydberg (a) nS states of the rubidium atom at a temperature of T = 300K, in dependence upon the principal quantum number as well as experimental data – black circles [1] and data of alternative theoretical calculation based on the improved quasiclassical model by Beterov et al [2]. - continuous line.

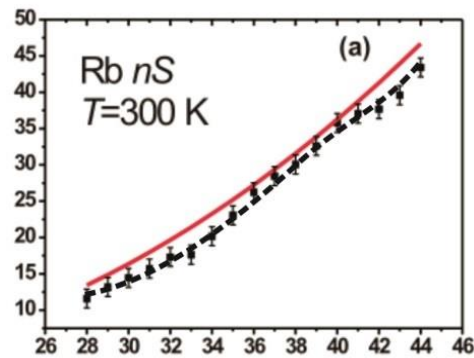


Figure 1. Our theoretical data (dashed line) on the dependence of the values of the effective lifetime for the group of Rydberg (a) nS states of the Rb atom at T = 300K, as well as experimental data – black circles [1] and data of alternative theoretical calculation based on the improved quasiclassical model by Beterov et al [2]. - continuous line

The corresponding values of the lifetimes of the levels are obtained for the values of the principal quantum number n up to 100. In general, the results of our non-empirical relativistic theory are in physically realistic agreement with the experimental data, at least much better than some other alternative theoretical approaches. The same applies to the effect of thermal radiation reduces the value of a lifetime.

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Key words: Rydberg heavy atoms, relativistic theory, black-body radiation field.

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РЕЛЯТИВИСТСКИЙ РАСЧЕТ ВЕРОЯТНОСТЕЙ РАДИАЦИОННЫХ ПЕРЕХОДОВ И ВРЕМЕНИ ЖИЗНИ ВОЗБУЖДЕННЫХ СОСТОЯНИЙ ДЛЯ АТОМА РУБИДИЯ В ПОЛЕ ИЗЛУЧЕНИЯ ЧЕРНОГО ТЕЛА

Резюме. Представлены результаты релятивистского расчета вероятностей радиационных переходов и времен жизни возбужденных состояний для тяжелой ридберговской атомной системы в поле чернотельного (теплового) излучения на примере рубидия. В качестве теоретического подхода мы применяем комбинированный релятивистский энергетический подход и релятивистскую многочастичную теорию возмущений с оптимизированным дираковским нулевым приближением. Получены расчетные данные для вероятностей радиационных переходов и времен жизни возбужденных состояний, в частности атома рубидия в ридберговских состояниях с главным квантовым числом $n = 10-100$. Проведено сравнение полученных теоретических данных по эффективным временам жизни для группы ридберговских nS состояний атома рубидия при температуре $T = 300$ К с экспериментальными данными, а также данными альтернативного теоретического расчета на основе усовершенствованной квазиклассической модели. Показано, что точность теоретических данных обеспечивается корректностью вычисления соответствующих релятивистских волновых функций и полнотой учета обменно-корреляционных эффектов.

Ключевые слова: ридберговские тяжелые атомы, релятивистская теория, тепловое излучение.

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РЕЛЯТИВІСТСЬКИЙ РОЗРАХУНОК ЙМОВІРНОСТЕЙ РАДІАЦІЙНИХ ПЕРЕХОДІВ І ЧАСУ ЖИТТЯ ЗБУДЖЕНИХ СТАНІВ ДЛЯ АТОМУ РУБІДІЮ В ПОЛІ ВИПРОМІНЮВАННЯ ЧОРНОГО ТІЛА

Резюме. Представлені результати релятивістського розрахунку ймовірностей радіаційних переходів і часів життя збуджених станів для важкої рідбергівської атомної системи в поле чорнотільного (теплового) випромінювання на прикладі рубідію. В якості теоретичного підходу ми застосовуємо комбінований релятивістський енергетичний підхід і релятивістську багаточастинкову теорію збурень з оптимізованим діраківським нульовим наближенням. Отримано розрахункові дані для ймовірностей радіаційних переходів і часів життя збуджених станів, зокрема атома рубідію в рідбергівських станах з головним квантовим числом $n = 10-100$. Проведено порівняння отриманих теоретичних даних щодо ефективних часів життя для групи рідбергівських nS станів атома рубідію при температурі $T = 300$ К з експериментальними даними, а також даними альтернативного теоретичного розрахунку на основі удосконаленої квазікласичної моделі. Показано, що точність теоретичних даних забезпечується коректністю обчислення відповідних релятивістських хвильових функцій і повнотою обліку обмінно-кореляційних ефектів.

Ключові слова: рідбергівські важкі атоми, релятивістська теорія, теплове випромінювання.