

# ОПТИЧНІ, ОПТОЕЛЕКТРОННІ І РАДІАЦІЙНІ СЕНСОРИ

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## OPTICAL AND OPTOELECTRONIC AND RADIATION SENSORS

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### RELATIVISTIC APPROACH TO CALCULATION OF IONIZATION CHARACTERISTICS FOR RYDBERG ALKALI ATOM IN A BLACK-BODY RADIATION FIELD

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**Abstract.** The combined relativistic energy approach and relativistic many-body perturbation theory with the zeroth Dirac-Fock-Sham approximation are used for computing the thermal Blackbody radiation ionization characteristics of the alkali Rydberg atoms, in particular, the sodium in Rydberg states with principal quantum number  $n=10-100$ . The detailed analysis of the data of computing ionization rates for the Rydberg sodium atom demonstrates physically reasonable agreement between the theoretical and experimental data. The accuracy of the theoretical data is provided by a correctness of the corresponding relativistic wave functions and accounting for the exchange-correlation effects.

**Keywords:** Rydberg alkali atoms, relativistic theory, Blackbody radiation field

## РЕЛЯТИВИСТСЬКИЙ ПІДХІД ДО РОЗРАХУНКУ ІОНІЗАЦІЙНИХ ХАРАКТЕРИСТИК РІДБЕРГІВСЬКИХ ЛУЖНИХ АТОМІВ У ПОЛІ ТЕПЛОГО ВИПРОМІНЮВАННЯ

*О. В. Глушков, В. Б. Терновський, В. В. Буяджи, А. В. Цудік, П. А. Заічко*

**Анотація.** Комбінований релятивістський енергетичний підхід і релятивістська теорія збурень багатьох тіл з з оптимізованим дірак-кон-шемівським нульовим наближенням використовуються для обчислення іонізаційних характеристик лужних рідбергівських атомів в полі теплового випромінювання абсолютно чорного тіла, зокрема, атома натрію в рідбергівських станах з головним квантовим числом  $n = 10-100$ . Аналіз даних обчислення швидкості іонізації атома натрію у рідбергівських станах демонструє фізично розумну згоду між теоретичними і експериментальними даними. Точність теоретичних даних забезпечується коректністю обчислення відповідних релятивістських хвильових функцій і повнотою урахування обмінно-кореляційних ефектів.

**Ключові слова:** рідбергівські лужні атоми, релятивістська теорія, теплове випромінювання

## РЕЛЯТИВИСТСКОЙ ПОДХОД К РАСЧЕТУ ИОНИЗАЦИОННЫХ ХАРАКТЕРИСТИК РИДБЕРГОВСКИХ ЩЕЛОЧНЫХ АТОМОВ В ПОЛЕ ТЕПЛОГО ИЗЛУЧЕНИЯ

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**Аннотация.** Комбинированный релятивистский энергетический подход и релятивистская теория возмущений многих тел с оптимизированными дирак-кон-шесмоским нулевым приближением используются для вычисления ионизационных характеристик щелочных ридберговских атомов в поле теплового излучения черного тела, в частности, атома натрия в ридберговских состояниях с главным квантовым числом  $n=10-100$ . Анализ данных вычисления скоростей ионизации атома натрия в ридберговских состояниях демонстрирует физически разумное согласие между теоретическими и экспериментальными данными. Точность теоретических данных обеспечивается корректностью вычисления соответствующих релятивистских волновых функций и полнотой учета обменно-корреляционных эффектов.

**Ключевые слова:** ридберговские щелочные атомы, релятивистская теория, тепловое излучение

### Introduction

A significant progress in experimental laser physics, appearance of the tunable lasers allow to study unique properties of atoms and ions in the Rydberg states. The experiments with Rydberg atoms had very soon resulted in the discovery of an important ionization mechanism, provided by unique features of the Rydberg atoms. Relatively

new topic of the modern theory is connected with consistent study of ionization of Rydberg atoms by blackbody radiation [1-5]. An account for the AC Stark shift, fast redistribution of the levels' population and photoionization provided by the environmental black-body radiation (BBR) field became of a great importance for successfully handling atoms in their Rydberg states [1-14]. Spectroscopic data on the excita-

tion and ionization processes of Rydberg atoms by black-body radiation are very important not only for the above numerous applications, including spectroscopy of ultracold plasma, but also for the physics of the Rydberg atoms in resonators, Rydberg masers, and microwave detection techniques quantum level of sensitivity, optical and atomic standards of frequency, atomic clocks, which in turn opens new ways of constructing quantum standards for measuring fundamental constants (including the elucidation of drift value of the fine structure constant) testing physical postulates of inertial navigation, magnetometry and more. Finally, it is important that many of the effects of low-energy quantum optics and electrodynamics (e.g., indiscriminate Rabi oscillation damping in a resonator quantum field or Cummings collapse, Dicke radiation, processes in single-atom masers, two-photon masers, two-photon masers, , the subtle effects associated with the quantum properties of the photonic field in the microwave range) can be quantitatively adequately studied on the basis of physical systems with RA.

The standard methods for computing the spectroscopic parameters of the Rydberg atoms in the BBR field are based on the different versions of the model potential (MP), quasiclassical methods (see e.g., [5-24]). It should be mentioned the simple and quite effective quasiclassical approach to compute the thermal ionization rate for Rydberg atoms [1,4,5]. Naturally, the standard methods of the theoretical atomic physics, including the Hartree-Fock and Dirac-Fock ones, should be used in order to determine a thermal ionization characteristics of neutral and Rydberg atoms. The correct calculation of spectroscopic parameters for the heavy Rydberg atoms in a black-body radiation field requires using strictly relativistic models and an accurate accounting for the exchange-polarization effects.

In this paper an energy approach and relativistic perturbation theory (PT) with the model density functional Dirac-Kohn-Sham zeroth approximation [25-33] are used to compute the spectroscopic parameters (rate of the BBR-induced decay, radiative lifetimes) of the Rydberg alkali atom atom in a black-body radiation field.

## 2. The Rydberg atom in a Blackbody radiation field: theoretical approach

The physical aspects of interaction of the Rydberg atom with black-body electromagnetic radiation field are considered many times from different points of view. One could remind that the frequency of a greater part of the black-body radiation field photons  $\omega$  does not exceed 0.08 atomic units including temperatures of the order thousand K. The standard approach supposes using ( in a case of alkali atom) the known one-particle model approximation for calculating the energy and spectral parameters such as (excitation and ionization probabilities, ionization cross section etc). Usually one should start from a product with the Planck's distribution for the thermal photon number density [1,9]:

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

where  $c$  is the speed of light and  $k$  is the Boltzmann constant, Ionization probability (rate) of the bound state  $nl$  results in the integral over the Blackbody radiation frequencies:

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

The total expression for the magnitude of the full rate of the Rydberg atom ionization can be written as the sum

$$W_{BBR}^{tot} = W_{BBR} + W_{SFI} + W_{BBR}^{mix} + W_{SFI}^{mix} \quad (3)$$

where the first term in (3) describes the direct photoionization rate from the initially excited state  $nL$ , the second term, second term is the ionization rate of the atoms in the high-lying states inhabited by thermal radiation, an electric field; the third term in (3) is the full rate of direct ionization of the Rydberg atom in the surrounding to the initially excited states; lastly, the latter is the ionization of high-lying states inhabited by the known two-stage process.

The total width of an experimentally observable Rydberg state of an atom or ion (naturally

isolated from all external fields except BBR) consists obviously of natural, spontaneous radiation width and BBR-induced (thermal) width:

$$\Gamma_{nl}^{tot} = \Gamma_{nl}^{sp} + \Gamma_{nl}^{BBR}(T) \quad (4)$$

It should be mentioned that the above formulas for determining probabilities, transition rates, lifetimes are written in a non-relativistic approximation and are actually used in the vast majority of modern theories and model representations about the processes of excitation and ionization of Rydberg atom in the field of thermal radiation (see, e.g., [1]). It should be emphasized that, in fact, the process of excitation and ionization of RA by thermal radiation is not a simple process, as it may seem at first glance; moreover, I emphasize the theoretical and experimental complexities of the description of Rydberg atom.

From a theoretical point of view, the sequential consideration of the process should include direct photoionization from the initially excited state nL, ionization by pulling electric impulses of high-lying Rydberg states inhabited by thermal radiation, direct photoionization of the adjacent rarefied states, thermal radiation, at the end of the ionization by electrical impulses above the located states, populated by two-many-step processes. In our view, in any case, the most correct and consistent quantum theory of excitation and ionization of Rydberg atom in the field of thermal radiation is preferably based on the principles of QED, or the corresponding relativistic approximation.

We apply a generalized energy approach [25-29] to compute the Rydberg atoms spectroscopic characteristics (rate of decay or ionization, radiative lifetimes etc). The radiation decay probability is connected with the imaginary part of electron energy shift. The latter is presented as:  $\Delta E = \text{Re}\Delta E + i \Gamma/2$ , where  $\Gamma$  is a level width, and decay probability  $P=\Gamma$ . The imaginary part of a shift  $\Delta E$  is defined in the PT second order as (in atomic units):

$$\text{Im} \Delta E = (1/4\pi) \sum_{\alpha>n>f} V_{\alpha n \alpha n}^{|\alpha_{lm}|}, \quad (5)$$

where  $(\alpha>n>f)$  for electron and  $(\alpha<n<f)$  for vacancy. The matrix element is determined as follows:

$$V_{ijkl}^{|\omega|} = \iint d r_1 d r_2 \Psi_i^*(r_1) \Psi_j^*(r_2) \frac{\sin|\omega| r_{12}}{r_{12}} (1 - \alpha_1 \alpha_2) \Psi_k^*(r_2) \Psi_l^*(r_1) \quad (6)$$

The effective lifetime  $\tau_{eff}$  of the Rydberg atom state is naturally inversely proportional to the full decay rate of the excited state as a result of spontaneous transitions  $\Gamma_0(\tau_0)$  and transitions induced by thermal radiation  $\Gamma_{BBR}(\tau_{BBR})$ , that is, it can be written:

$$\frac{1}{\tau_{eff}} = \Gamma_0 + \Gamma_{BBR} = \frac{1}{\tau_0} + \frac{1}{\tau_{BBR}} \quad (7)$$

The detailed description of the matrix elements and procedure for their computing are presented in Refs. [2,28-33]. The relativistic wave functions are calculated by solution of the relativistic Dirac equation with the model Dirac-Kohn-Sham zeroth approximation plus correlation potential [29-32]. All calculations (the numeral code Superatom-ISAN, version 93 is used) are performed with an accurate accounting for the exchange-correlation effects (including polarization, screening effects, continuum pressure and others) as the effects of the PT second and higher orders.

### 3. The results and conclusions

In Table 1, we present our theoretical data as well as experimental Gallakher-Cooke data (Virgindia group) and Gounand estimates for the effective lifetime of the Rydberg states 17p, 18p in the spectrum of the Na atom [4,5,9].

In 1 we present experimental and theoretical data on the total rate of BBR-induced ionization for the Rydberg nS states of the sodium atom (T = 300K): Experiment (circles and squares) [3]; Theory: an improved quasiclassical model of Beterov etal (continuous line) [5] and our theory (dashed line).

In the case of states with n below 15, the experimental data were obtained using a fluorescence technique at a temperature of 400K. The lifetime of states with n more than 15 were measured using the field ionization method. Comparison of our data with the experimental results shows a physically reasonable good agreement of these data, and for nS and nD states the agreement is better than for nP states.

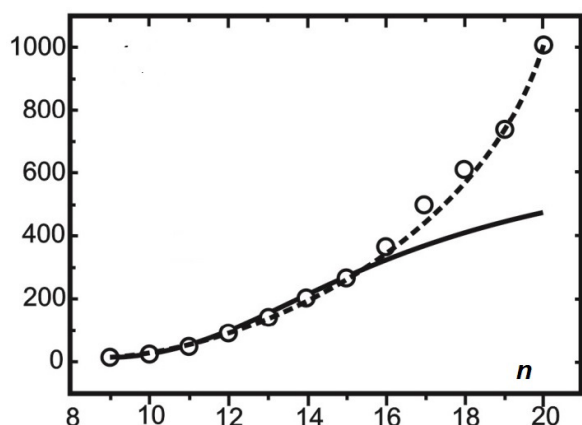


Figure 1. The total rate of BBR-induced ionization for the Rydberg  $nS$  states of the sodium atom ( $T = 300K$ ): Experiment (circles and squares); Theory: an improved quasiclassical model of Beterov et al (continuous line) and our theory (dashed line).

for these states are quite large, while for the  $nD$  states of the sodium atom the pattern is fundamentally different. In our theory, the effects sought are taken into account quite carefully and correctly, and therefore the theory is in a physically reasonable agreement with the experiment.

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Table 1.

**Our theoretical data, Gounand estimates and experimental Gallakher-Cookt data (Virdg-  
inia group) for the effective lifetime ( $\mu s$ ) of the Rydberg states 17p, 18p in the spectrum of  
the Na atom ( $T = 300K$ )**

State	Exp. data Gallakher- Cooke	Data by Gounand	Theory Gallakher- Cooke	This work
17p	$11.4 \pm \begin{matrix} +5.6 \\ -1.4 \end{matrix}$	48.4	15.5	16.8
18p	$13.9 \pm \begin{matrix} +8.8 \\ -2.9 \end{matrix}$	58.4	17.9	18.5

Analysis of the data shown in Fig. 1 shows that, first, both theories describe the experimental data for the  $nS$  states and the  $nD$  states of the sodium atom from  $n$  to 15 fairly well, but a significant deviation of the quasiclassical calculation data from the experiment is further observed. The most adequate explanation of this important fact was provided by Beterov et al in the known papers [1,5], attributing the deviation of quasiclassical theory from experiment to the overriding importance of exchange-correlation corrections, in particular, for  $nS$  states. In fact, for  $nS$  states there is a certain anomaly, because the orbital for the desired states penetrates deep enough into the atomic core, which causes quite strong interaction with it. An additional factor is the known fact, namely, the quantum defects

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### Summary

Spectroscopic data on the excitation and ionization processes of Rydberg atoms by black-body radiation are very important for numerous applications, including spectroscopy of ultracold plasma, physics of the Rydberg atoms in resonators, Rydberg masers, and microwave detection techniques on quantum level of sensitivity, optical and atomic standards of frequency, atomic clocks, magnetometry and more. In this paper the combined relativistic energy approach and relativistic many-body perturbation theory with the zeroth Dirac-Fock-Sham approximation are used for computing the thermal Blackbody radiation ionization characteristics of the alkali Rydberg atoms, in particular, the sodium in Rydberg states with principal quantum number  $n=10-100$ . The detailed analysis of the data of computing ionization rates for the Rydberg sodium atom demonstrates physically reasonable agreement between the theoretical and experimental data. The deviation of the data of quasiclassical calculations from the experiment is due to the neglect of quantitatively important exchange-correlation effects, especially in the case of  $nS$  states. The accuracy of our theoretical data is provided by a correctness of the corresponding relativistic wave functions and accounting for the exchange-correlation effects.

**Keywords:** Rydberg alkali atoms, relativistic theory, Blackbody radiation field



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### Реферат

Спектроскопічні дані про процеси збудження та іонізації атомів рідбергівських атомів у полі випромінювання чорного тіла є дуже важливими для численних застосувань, включаючи спектроскопію ультрахолодної плазми, фізику рідбергівських атомів в резонаторах, рідбергівських мазерів, розвиток нових методів мікрохвильового детектування на квантовому рівні чутливості, оптичні атомні стандарти частоти, атомні годинники, магнітометрія тощо. Комбінований релятивістський енергетичний підхід і релятивістська теорія збурень багатьох тіл з оптимізованим дірак-кон-шемівським нульовим наближенням використовуються для обчислення іонізаційних характеристик лужних рідбергівських атомів в полі теплового випромінювання абсолютно чорного тіла, зокрема, атома натрію в рідбергівських станах з головним квантовим числом  $n = 10-100$ . Аналіз даних обчислення швидкості іонізації атома натрію у рідбергівських станах демонструє фізично розумну згоду між теоретичними і експериментальними даними. Відхилення даних квазікласичних розрахунків від експерименту пов'язано із неврахуванням кількісно важливих обмінно-кореляційних ефектів, особливо у випадку  $nS$ -станів. Точність наших теоретичних даних забезпечується коректністю обчислення відповідних релятивістських хвильових функцій і повнотою врахування обмінно-кореляційних ефектів.

**Ключові слова:** рідбергівські лужні атоми, релятивістська теорія, теплове випромінювання