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RELATIVISTIC SPECTROSCOPY OF HEAVY RYDBERG ATOMIC SYSTEMS IN A BLACK-BODY RADIATION FIELD

We present the results of studying the spectroscopic characteristics of heavy Rydberg atomic systems in a black-body (thermal) radiation field. 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 presented the calculational data for the thermal black-body radiation ionization characteristics of the alkali Rydberg atoms, in particular, the sodium in Rydberg states with principal quantum number n=10-100 and ytterbium ion. Application of theory to computing the spectral parameters of studied atomic systems have demonstrated 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.

1. Introduction

At the present time, the study of Rydberg atoms (molecules) is definitely one of the most popular and very interesting directions of modern quantum physics and chemistry, atomic optics and spectroscopy. The huge relevance of the investigation of the energy and spectral properties of the Rydberg atoms (molecules) is, of course, due to the standard requirements for spectroscopic information of a number of applications and related physical disciplines, which include physics and chemistry of laboratory, astrophysical plasma, astrophysics and radioastronomy, atomic and molecular optics and spectroscopy, laser physics and quantum electronics and many others [1-94]). From the other side, 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 treating the Rydberg atoms in a field of the Blackbody radiation (BBR). It should be noted that the BBR is one of the essential factors affecting the Rydberg states in atoms [1]. The account for the ac Stark shift, fast redistribution of the levels' population and photoionization provided by the environmental BBR became of a great importance for successfully handling atoms in their Rydberg states. 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 hydrogen-like approximation, different versions of the quantum defect method, classical and quasiclassical model approaches, the model and pseudo – potential methods. The authors of the papers [3-10] applied the Coulomb approximation, quantum defect formalism, different versions of the model and pseudo-potential method etc (as a rule, the non-relativistic versions are used) to determine the spectral and radiative properties of different Rydberg atoms and ions.

It should be noted separately the cycles of theoretical and experimental works by Ryabtsev-Beterov et al [2,3], as well as theoretical works of Dyachkov-Pankratov and others (c.g.[1-10]), in which the advanced 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.

The quality and consistency of accounting for these effects also determine the accuracy of description of the energy and spectroscopic parameters of the heavy Rydberg atoms, including these atoms in a thermal radiation field.

Naturally, the standard methods of the theoretical atomic physics, including the Hartree-Fock and Dirac-Fock approximations should be used in order to determine the thermal ionization characteristics of neutral and Rydberg atoms [2].

One could note that the correct treating of the heavy Rydberg atoms parameters in an external electromagnetic field, including the BBR field, requires using strictly relativistic models. In a case of multielectron atomic systems it is necessary to account for thee exchange-correlation corrections.

Among the fundamentally important exchange-correlation effects for essentially many-electron systems, one should single out such effects as polarization interaction and screening, continuum pressure, the non-Coulomb grouping of levels in the heavy Rydberg atoms spectra etc. It should be noted that these effects are not correctly considered, for example, within simplified Coulomb approximation or quantum defect models (c.g.[11-20]). Their account requires using very consistent methods.

We present the results of studying the spectroscopic characteristics of heavy Rydberg atomic systems in a black-body (thermal) radiation field.

As theoretical approach we apply the combined generalized relativistic energy approach and relativistic many-body perturbation theory with ab anitio Dirac zeroth approximation.

2. Atom in a Black-body radiation field: 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:

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

where k=3.1668×10^{-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:

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

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:

$$\sigma_{nl}(\omega)=\frac{4\pi^2\omega}{3c(2l+1)}[M^2_{nl\to E,l-1}+ \\ +(l+1)M^2_{nl\to E,l+1}], \qquad (3)$$

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].

3. Relativistic perturbation theory and energy approach

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:

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

where c is the velocity of light, a_i, a_j – the Dirac matrices, w_{ij} –the transition frequency, Z is a charge of atomic nucleus. The general potential in (4) includes self-consistent Coulomb-like mean-field potential $U_{MF}(r_i \mid b)$, ab ibitio one-particle exchange-correlation (relativistic generalized exchange Kohn-Sham potential plus generalized correlation Lundqvist-Gunnarsson potential) $V_{XC}(r_i \mid b)$ 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:

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

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: DE = ReDE + i G/2, where G is interpreted as the level width, and a decay probability P = G. The imaginary part of electron energy shift is defined in the PT lowest order as:

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

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

$$V^{|\omega|}_{ijkl} = \iint dr_1 dr_2 \Psi^*_i(r_1) \Psi^*_j(r_2) \frac{\sin|\omega| r_{12}}{r_{12}} (1 - \alpha_1 \alpha_2) \Psi^*_k(r_2) \Psi^*_l(r_1) \quad (7)$$

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 $\tilde{A}^{sp}_{nl}$ and BBR-induced (thermal) width $\tilde{A}^{BBR}_{nl}$:

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

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:

$$\frac{1}{\tau_{eff}} = \tilde{A}_0 + \tilde{A}_{BBR} = \frac{1}{\tau_0} + \frac{1}{\tau_{BBR}} \quad (9)$$

The detailed procedures of calculation of the radial and angular integrals (amplitudes) in the matrix elements are described in Refs. [9-20,22,38-41]. All calculations are performed on the basis of the numeral code Superatom-ISAN (version 93).

4. Results and conclusions

In Table 1 we present our theoretical data on the effective lifetime of the sodium nP, nD Rydberg states and for comparison some theoretical data by Beterov et al [2,3] for temperatures T=300, 600K. In Table 2 we present our theoretical data on the effective lifetime of the sodium nP, nD Rydberg states for temperatures T=300

and 600K. Obviously, the accuracy of the theoretical data is provided by a correctness of the corresponding relativistic wave functions and accounting for the exchange-correlation effects.

Table 1.

Effective lifetime (μs) of the nP Rydberg states in the sodium spectrum for the temperature T = 300: [2]- theory by Beterov et al and this work.

n	T= 300 K $P_{1/2}$ $P_{3/2}$ Ref. [2]	T= 300 K $P_{1/2}$ $P_{3/2}$ This work
10	4.80 4.76	4.84 4.81
20	20.99 20.89	21.06 20.96
30	48.71 48.56	48.84 48.70

Table 2.

Effective lifetime (μs) of the nP, nD Rydberg states in the sodium spectrum for the temperatures T = 300, 600K (this work).

n	T= 600 K $P_{1/2}$ $P_{3/2}$ This work	T= 300 K $D_{3/2}$ $D_{5/2}$ This work	T= 600 K $D_{3/2}$ $D_{5/2}$ This work
10	2.84 2.83	0.913 0.914	0.837 0.838
20	11.42 11.38	6.263 6.266	5.164 5.167
30	26.03 25.97	18.602 18.609	14.281 14.285

In conclusion we also present our result of computing the relative blackbody radiative shift (in 10^{-14}) for singly ionized Yb: β=--0.097. The similar β values are obtained using third-order relativistic many-body calculations [4] β=−0.0983 and *ab initio* method [5]: β=−0.094. In these calculations different methods are used to compute matrix elements and different orbital basises are used. The details of this problem will be presented in a separate paper.

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Summary. We present the results of studying the spectroscopic characteristics of heavy Rydberg atomic systems in a black-body (thermal) radiation field. 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 presented the calculational data for the thermal black-body radiation ionization characteristics of the alkali Rydberg atoms, in particular, the sodium in Rydberg states with principal quantum number n=10-100 and ytterbium ion. Application of theory to computing the spectral parameters of studied atomic systems have demonstrated 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.

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

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РЕЛЯТИВИСТСКАЯ СПЕКТРОСКОПИЯ ТЯЖЕЛЫХ РИДБЕРГОВСКИХ АТОМНЫХ СИСТЕМ В ПОЛЕ ИЗЛУЧЕНИЯ ЧЕРНОГО ТЕЛА

Резюме. Представлены результаты изучения спектроскопических характеристик тяжелых ридберговских атомных систем в поле чернотел(теплового) излучения. В качестве теоретического подхода мы применяем комбинированный релятивистский энергетический подход и релятивистскую многочастичную теорию возмущений с оптимизированныи дираковским нулевым приближением. Представлены результаты расчета спектроскопических характеристик щелочных ридберговских атомов в поле теплового излучения черного тела, в частности, натрия в ридберговских состояниях с главным квантовым числом n=20-100 и иона иттербия. Применение теории к вычислению спектральных параметров исследуемых атомных систем продемонстрировало 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 тяжелые атомы, релятивистская теория, тепловое излучение.

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РЕЛЯТИВІСТСЬКА СПЕКТРОСКОПІЯ ВАЖКИХ РІДБЕРГІВСЬКИХ АТОМНИХ СИСТЕМ В ПОЛІ ВИПРОМІНЮВАННЯ ЧОРНОГО ТІЛА

Резюме. Представлені результати вивчення спектроскопічних характеристик важких рідбергівських атомних систем в полі чорнотільного (теплового) випромінювання. В якості теоретичного підходу ми застосовуємо комбінований релятивістський енергетичний підхід і ре-

лятивістську багаточастинкову теорію збурень з оптимізованим дираківським нульовим наближенням. Представлені результати розрахунку спектроскопічних характеристик лужних рідбергівських атомів в полі теплового випромінювання, зокрема, натрію в рідбергівських станах з головним квантовим числом n = 20-100 та іону ітербію. Застосування теорії до обчислення спектральних параметрів досліджуваних атомних систем продемонструвало фізично розумну згоду між теоретичними і експериментальними даними. 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.