ФІЗИЧНІ, ХІМІЧНІ ТА ІНШІ ЯВИЩА, НА ОСНОВІ ЯКИХ МОЖУТЬ БУТИ СТВОРЕНІ СЕНСОРИ

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THERMAL PHOTOIONIZATION OF THE RYDBERG ATOMS BY THE BLACKBODY RADIATION: NEW RELATIVISTIC APPROACH

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Abstract. Within the gauge-invariant quantum electrodynamics perturbation theory it is presented a new relativistic approach to definition of the thermal photoionization characteristics for the Rydberg atoms, in particular, the Rydberg atoms, ionized by the Blackbody radiation. Theory of corresponding phenomena in the Rydberg systems is a basis for creation of new class of the atomic sensors.

Keywords: sensing Rydberg atoms, thermal photoionization, Blackbody radiation, new relativistic approach

ТЕРМІЧНА ФОТОІОНІЗАЦІЯ РІДБЕРГІВСЬКИХ АТОМІВ ВИПРОМІНЮВАННЯМ ЧОРНОГО ТІЛА: НОВА ТЕОРЕТИЧНА СХЕМА

Т. М. Зеленцова, Т. Б. Перелигіна

Анотація. Запропонований новий релятивістський підхід в межах калібровочно-інваріантної КЕД теорії збурень до визначення характеристик термічної фотоіонізації рідбергівських атомів, зокрема, атомів, що іонізуються чорнотільним випромінюван-ням. Теорія відповідних явищ у рідбергівських системах може бути основою для створення відповідних атомних сенсорів нового класу.

Ключові слова: детектування рідбергівських атомів, термічна фотоіонізація, випромінювання чорного тіла, нова релятивістська схема

ТЕРМИЧЕСКАЯ ФОТОИОНИЗАЦИЯ РИДБЕРГОВСКИХ АТОМОВ ИЗЛУЧЕНИЕМ ЧЕРНОГО ТЕЛА: НОВАЯ ТЕОРЕТИЧЕСКАЯ СХЕМА

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Аннотация. Предложен новый релятивистский подход на основе калибровочно-инвариантной КЭД теории возмущений к определению характеристик термической фотоионизации ридберговских атомов, в частности, ридберговских атомов, ионизируемых чернотельным излучением. Теория соответствующих явлений в ридберговских системах является основой для создания атомных сенсоров нового класса.

Ключевые слова: детектирование ридберговских атомов, термическая фотоионизации, излучение черного тела, новая релятивистская схема

1. Introduction

The modern quantum theory of atomic systems in an electromagnetic field can be considered as a fundamental basis for treating a wide cycle of phenomena, including the excitation, photoionization, radiation and autoionization decay etc [1-8]. In the last years a great success has been achieved in the applied atomic physics, laser physics, atomic sensors electronics. One could mention very perspective papers devoting to engineering so called Rydberg atoms with pulsed electric fields [4]. New schemes of the different atomic sensor devices are proposed. Naturally, for more than 80 years, the theory of atomic photo effect and correspondingly atomic photoionization was developing and considering mainly the ground states and lowest excited states in usual neutral atoms, beginning from the hydrogen one. But a great progress in experimental laser physics and appearance of the so called tunable lasers allow to get the highly excited Rydberg states of atoms. In fact this is a beginning of a new epoch in the atomic physics with external electromagnetic field. It has stimulated a great number of papers on the ad and dc Stark effect [5-16]. An especial interest attracts a case of the strong external field. 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. One of the interesting phenomena is the so-called blackbody radiation-induced (or thermal) ionization [17-19]. The blackbody radiation-induced ac Stark shift, population redistribution and photoionization should also be taken into account , as unremovable detuning and destructive factors, in designing the high-precision time–frequency standards based on such a finely tuned quantum system as the neutral atoms in an optical lattice of a Stark-compensating 'magic' wavelength [19]. Naturally a range of the physical phenomena in the Rydberg systems which can be a basis for creation of the atomic sensors new class, is significantly wider [1-8]. From the theoretical point of view, the Blackbody radiation could essentially affect on the Rydberg states in atoms [9]. Its role was estimated theoretically and observed in the tunable lasers experiments (look for example, [17-19]). The account for the ac Stark shift, fast redistribution of the atomic levels' population and photoionization caused by the blackbody radiation is of a great importance for successfully handling atoms in their Rydberg states. In the last years there is appearing a sufficiently great number of papers devoted to the spectroscopy of the Rydberg atoms in the black-body radiation (look review in refs. [17,19]). Usually the standard methods of atomic physics, including the Hartree-Fock, different model potential schemes , the quantum defect method etc [1-3,17-26] were used in order to define the thermal ionization characteristics of neutral and even Rydberg atoms. The significant advantage of the simple model potential and quantum defect approached (non-relativistic schemes) in comparison with other methods and, particularly, other model potentials is the possibility of presenting analytically, in terms of the hypergeometric functions, the quantitative characteristics for arbitrarily high orders, related to both bound–bound and bound–free transitions. From the other side, the heavy Rydberg atomic systems in a external field, including the Blackbody radiation, should be considered within strictly relativistic theory. Here we present the gauge-invariant relativistic perturbation theory approach to definition of the thermal photoionization characteristics of the Rydberg atoms, in particular, the Rydberg atoms in the Blackbody radiation. The important feature of new theory is using gauge-invariant procedure to calculating the corresponding electron relativistic wave functions that, as it was underlined in many papers (look, for example, [2,3, 18,19-26]), has a decisive value in correct definition of the corresponding properties.

2. The photoionization of the Rydberg atoms in the Blackbody radiation

Here we consider a non-relativistic scheme for definition of the thermal photoionization of the atom systems [2,17,19]. First of all, it should be mentioned that even for temperatures of order $T=10^4$ K, the frequency of a greater part of the Blackbody radiation photons ω does not exceed 0.1 atomic units (a.u.). It is indeed lying far below the excitation energy and hence far below the ionization potential of many atoms and ions. Under these conditions, usually used a single- electron approximation [1] is quite appropriate for calculating the photoionization cross section $\sigma_{nl}(\omega)$. The latter appears in a product with Planck's distribution for the thermal photon number density:

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

where $k=3.1668\times10^{-6}$ a.u., K^{-1} is the Boltzmann constant, $c = 137.036$ a.u. is the speed of light. Further the photoionization rate of a bound state *nl* results in the integral over the Blackbody radiation frequencies:

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

In formular (2), the cross section of photoionization from a bound state with a principal quantum number *n* and orbital quantum number *l* by photons with frequency ω,

$$\sigma_{nl}(\omega)=\frac{4\pi^2\omega}{3c(2l+1)}[lM^2_{nl\to El-1}+(l+1)M^2_{nl\to El+1}]. \quad (3)$$

In formular (3) the radial matrix element of the ionization transition from the bound state with the radial wavefunction $R_{nl}(r)$ to the state of continuum with the wavefunction $R_{El}(r)$ normalized to the delta function of energy is defined as follows:

$$M_{nl\to El'}=\int\limits_0^{\infty}R_{El'}(r)r^3R_{nl}(r)dr. \quad (4)$$

With the use of expression (3) for the cross section in combination with Planck's distribution over frequencies for the Blackbody radiation photon number density (1), the rate of the Blackbody radiation- induced ionization at a fixed temperature T is written as:

$$P_{nl}=\frac{4}{3c^3}\int\limits_{|E_{nl}|}^{\infty}[\frac{l}{2l+1}M^2_{nl\to El-1}+\frac{l+1}{2l+1}M^2_{nl\to El+1}]\cdot \cdot\frac{\omega^2}{\pi^2c^3[\exp(\omega/kT)-1]}. \quad (5)$$

To calculate the corresponding parameters, one should use the non-relativistic expressions for wave functions. As it was indicated, the heavy Rydberg atomic systems in a external field, including the Blackbody radiation, should be considered within strictly relativistic theory.

3. Relativistic approach to determination of Rydberg atom photoionization parameters

Here we consider our theoretical relativistic scheme. Naturally, for heavy atoms it is necessary to start from the relativistic Hamiltonian. The wavefunction can be presented in a standard form as follows:

$$\Psi(\Gamma PJM)=\sum_r^{NCF}c_r\Phi(\gamma_r PJM) \quad (6)$$

which should be received from the self-consistent solutions of the Dirac–Fock type equations.

Configuration mixing coefficients c_r are usually obtained through diagonalization of the Dirac-Coulomb Hamiltonian, which is chosen by us in the following form [3]:

$$H_{DC}=\Sigma_i[c\alpha_i p_i+(\beta_i-1)c^2-V(r|nlj)]+ + \Sigma_{i>j}\exp(i\omega r_{ij})(1-\alpha_1\alpha_2)/r_{ij}, \quad (7)$$

where α_i, β are the Dirac matrices. The potential in H_{DC} contains the electrical potential of a nucleus, the electron self-consistent potential and the potential of exchange inter-electron interaction. As an example, further we consider the atomic system with single electron above the core of closed electron shells. Naturally, the approach is generalized on the multi-electron system. Within the QED perturbation theory formalism [3] a probability of ionization (radiation transition) is directly connected with imaginary part of electron energy of the system as follows:

$$\mathrm{Im}\Delta E(B)=-\frac{e^2}{4\pi}\sum_{\substack{\alpha>n>f\\ [\alpha<n\le f]}}V^{|\omega_{\alpha n}|}_{\alpha n\alpha n}, \quad (8)$$

where $\sum$ – for electron and $\sum_{\alpha<n\le f}$ – for vacancy. The potential V is as follows:

$$V_{ijkl}^{|\omega|}=\iint dr_1dr_2\Psi_i^*(r_1)\Psi_j^*(r_2)\frac{\sin|\omega|r_{12}}{r_{12}}\times \\ \times(1-\alpha_1\alpha_2)\Psi_k^*(r_2)\Psi_l^*(r_1). \quad (9)$$

The separated terms of the sum in (8) represent the contributions of different channels and a probability of the single-electron approximation dipole transition is:

$$\Gamma_{\sigma_n}=\frac{1}{4p}\cdot V_{\sigma_n\alpha_n}^{|u_{\sigma_n}|}. \quad (10)$$

The bound-bound transition oscillator strength : $gf=\lambda_g^2\cdot\Gamma_{\alpha_n}/6.67\cdot10^{15}$, where g is the degeneracy degree, λ is a wavelength in angstrems (Å). To calculate the matrix elements (9) the angle symmetry of the atomic task is usually. The corresponding expansion for potential $\sin|\omega|r_{12}/r_{12}$ on spherical functions as follows [3]:

$$\frac{\sin|\omega|r_{12}}{r_{12}}=\frac{\pi}{2\sqrt{r_1r_2}}\times \\ \times\sum_{\lambda=0}^{\infty}(\lambda)J_{\lambda+1/2}(|\omega|r_1)J_{\lambda+1/2}(|\omega|r_2)P_\lambda(\cos\mathbf{r}_1\mathbf{r}_2), \quad (11)$$

where J –is the Bessell function of first kind and $(\lambda)=2\lambda+1$. It should be underlined that the expansion (11) is corresponding to usual multipole one for probability of radiative decay. Substitution of the expansion (11) to matrix element of interaction gives as follows:

$$V_{1234}^{\omega}=\left[(j_1)(j_2)(j_3)(j_4)\right]^{1/2}\times \\ \times\sum_{\lambda\mu}(-1)^{\mu}\begin{pmatrix} j_1j_3 & \lambda\\ m_1-m_3 & \mu\end{pmatrix}\times\operatorname{Im}Q_\lambda(1234); \\ Q_\lambda=Q_\lambda^{\mathrm{Qul}}+Q_\lambda^{\mathrm{Br}}, \quad (9)$$

where j_i are the entire single electron momentums, m_i — their projections; Q_λ^{Qul} is the Coulomb part of interaction, Q_λ^{Br} - the Breit part. The Coulomb part Q_λ^{Qul} is expressed in terms of radial integrals R_λ (the analog of the (4) matrix elements), angular coefficients S_λ [3]:

$$Q_\lambda^{\mathrm{Qul}}=\frac{1}{Z}\mathrm{Re}\{R_l(1243)S_\lambda(1243)+ \\ +R_\lambda(\tilde{1}24\tilde{3})S_\lambda(\tilde{1}24\tilde{3})+R_\lambda(1\tilde{2}4\tilde{3})S_\lambda(1\tilde{2}4\tilde{3}) \quad (10) \\ +R_\lambda(\tilde{1}\tilde{2}4\tilde{3})S_\lambda(\tilde{1}\tilde{2}4\tilde{3})\}$$

$$R_\lambda(1243)=\iint dr_1r_1^2r_2^2f_1(r_1)f_3(r_1)\times \\ \times f_2(r_2)f_4(r_2)Z_\lambda^{(1)}(r_<)Z_\lambda^{(1)}(r_>). \quad (11)$$

where f is the large component of radial part of single electron state Dirac function and function Z is [3]:

$$Z_\lambda^{(1)}=\left[\frac{2}{|\omega_{13}|\alpha Z}\right]^{\lambda+1/2}\frac{J_{\lambda+1/2}(\alpha|\omega_{13}|r)}{r^\lambda\Gamma(\lambda+3/2)}. \quad (12)$$

The other items in (11) include small components of the Dirac functions; the sign "~" means that in (3) the large radial component f_i is to be changed by the small g_i one and the moment l_i is to be changed by $\tilde{l}_i=l_i-1$ for Dirac number $ж_1>0$ and l_i+1 for $ж_i<0$. The definition of the Q Breit part is in details given in [3].

4. Gauge-invariant approach to definition of the electron wave functions

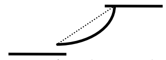
In calculating the matrix elements M in equation (5) it is of a great importance using the correct basises of the electron wave functions. Though in the Rydberg atoms these functions in are similar in many aspects to the hydrogen-like functions, however a role of the multi-0electron effects can be very significant. We propose to use the gauge invariant QED algorithm [26] in the scheme of definition the thermal photoionization parameters for Rydberg atoms, ionized by the Blackbody radiation,. Usually in the zeroth QED perturbation theory approximation it is introduced the one electron bare potential

$$V_N(r)+V_C(r), \quad (13)$$

with $V_N(r)$ describing the electric potential of the nucleus, $V_C(r)$, imitating the interaction of the single electron (quasiparticle) with the core. The self-consistent potential $V_C(r)$ is related to the electron density $\rho_C(r)$ in a standard way. The latter fully defines the single-electron representation.
The perturbation in terms of the second quantization representation reads:

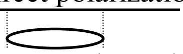
$$-V_C(r)\,\psi^+(r)\psi(r)-j_\mu(x)\,A^\mu(x), \quad (14)$$

where all notations are standard. In ref. [26] the lowest order multielectron effects, in particular, the gauge dependent radiative contribution $\mathrm{Im}\,\delta E_{\mathrm{ninv}}$ for the certain class of the photon propagator calibration is treated. The key fundamental idea is a minimization of the density functional $\mathrm{Im}\,\delta E_{\mathrm{ninv}}$.

In the lowest second order of the EDPT for the δE there is the only one-particle Feynman diagram A (A = ), that has a non-zeroth contribution to the imaginary part of electron energy Im δE (the radiation decay width). In the fourth order of the QED PT there are diagrams, whose contribution into the ImδE accounts for the core polarization effects. It is on the electromagnetic potentials gauge (the gauge non-invariant contribution).

The imaginary part of the diagram A contribution in the case of the Lorentz calibration has been presented previously as a sum of the partial contributions of α-s transitions from the initial state α to the final state s [26]:

$$\mathrm{Im}\delta E_\alpha\,(\mathrm{a}) = \sum_s \ \mathrm{Im}\ \delta E\,(\alpha\text{-}s;\ \mathrm{a}).$$

The most important diagram of the EDPT fourth order is direct polarization diagrams B (B= ). Its contribution into ImδE_α is gauge-dependent, though the results of the exact calculation of any physical quantity must be gauge-independent. All the non-invariant terms are multielectron by their nature (the particular case — non-coincidence of the oscillator strengths values, obtained in calculations with the "length" and "velocity" transition operator forms). The A diagram contribution into the Im δE related to the α -s transition is defined as follows:

$$-\frac{e^2}{8\pi}\iint \mathrm{d}r_1\,\mathrm{d}r_2\,\psi_\alpha^{+}(r_1)\ \psi_s^{+}(r_2)\times$$
$$\times\ \frac{1-\alpha_1\alpha_2}{r_{12}}\sin\,(\omega_{\alpha s}r_{12})\ \psi_\alpha\,(r_2)\ \psi_s\,(r_1),$$

for transverse part of the photon propagator and

$$-\frac{e^2}{8\pi}\iint \mathrm{d}r_1\,\mathrm{d}r_2\,\psi_\alpha^{+}(r_1)\ \psi_s^{+}(r_2)\ \{[(1-\alpha_1 n_{12}\alpha_2 n_{12})/$$
$$/r_{12}]\ \sin\,(\omega_{\alpha s}r_{12}) + \omega_{\alpha s}(1+\alpha_1 n_{12}\alpha_2 n_{12})\times$$
$$\times\cos(\omega_{\alpha s}r_{12})\}\ \psi_\alpha\,(r_2)\ \psi_s\,(r_1),$$

for longitudinal part where $\omega_{\alpha s}$ is the α -s transition energy. According to ref.[3], the $D_{\mu\nu,\mathrm{L}}$ contribution vanishes, if the one-quasi-particle functions ψ_α, ψ_s satisfy the same Dirac equation. The contribution of the B diagram to the Im δE_{ninv} is as follows [26]:

$$\mathrm{Im}\ \delta E_{\mathrm{ninv}}\,(\alpha\text{-}s;\ \mathrm{B}) = -\,C\iiiint \mathrm{d}r_1\,\mathrm{d}r_2\,\mathrm{d}r_3\,\mathrm{d}r_4\times$$
$$\times\sum_{\substack{n>f\\ m\le f}}\left(\frac{1}{\omega_{mn}+\omega_{\alpha s}}+\frac{1}{\omega_{mn}-\omega_{\alpha s}}\right)\psi_\alpha^{+}(r_1)\times$$
$$\times\ \psi_m^{+}(r_2)\ \psi_s^{+}(r_4)\ \psi_n^{+}(r_3)\ \frac{1-\alpha_1\alpha_2}{r_{12}}\ \times$$
$$\times\{[\,(\alpha_3\alpha_4-\alpha_3 n_{34}\alpha_4 n_{34})\,/\,r_{14}]\ \sin\,[\,\omega_{\alpha n}(r_{12}+r_{34})] +$$
$$+\ \omega_{\alpha n}\cos\,[\,\omega_{\alpha n}(r_{12}+r_{34})]\ (1+\alpha_3 n_{34}\alpha_4 n_{34})\}\times$$
$$\times\ \psi_m\,(r_3)\ \psi_\alpha\,(r_4)\ \psi_n\,(r_2)\ \psi_s\,(r_1). \qquad (15)$$

Here, f is the boundary of the closed shells; $n \ge f$ indicating the unoccupied bound and the upper continuum electron states; $m \le f$ indicates the finite number of states in the core and the states of the negative continuum (the latter can be omitted for the Rydberg atoms according the known reasons). In result, the minimization of the density functional Im δE_{ninv} leads to the integral differential equation for the ψ. Finally, one could get the optimal basis of the relativistic wave functions, which further should be used in calculating the matrix elements of the (5) type and corresponding ionization probabilities.

5. Conclusions

We have presented a new gauge-invariant relativistic approach to definition of the thermal photoionization characteristics for the Rydberg atoms, in particular, the Rydberg atoms, ionized by the Blackbody radiation, within the QED perturbation theory [3]. The concrete numerical application of approach now is in a progress. At the sane time, the multiple atomic calculations [1-3,11,20-26] show that a gauge invariant relativistic scheme is significantly more advantagable in comparison with the standard Hartree-Fock (HF) approximation and even Dirac-Fock method and its improved versions [1-3]. It should be very illustrative to present some data for the valence shell photoionization cross-section σ of alkali atoms (for example, K) by photons of the energies ω=0.4,0.8 Ry [1]: σ^{HF}(ω=0.4 Ry; r-form)=0.045 Mb, σ^{HF}(ω=0.4 Ry; ∇-form)=0.018 Mb, experiment σ^{exp}(ω=0.4 Ry)=0.13Mb, the random-phase approximation with exchange (RPAE) σ^{RPAE}(ω=0.4 Ry)=0.17Mb and σ^{HF}(ω=0.8Ry; r-form)=0.105Mb, σ^{HF}(ω=0.8Ry; ∇-form)=0.065Mb, σ^{exp}(ω=0.8Ry)= 0.41Mb, σ^{RPAE}(ω=0.8 Ry)=0.38Mb. The non-coincidence degree of the corresponding transition matrix elements in the “r” and “∇” forms (in fact, different gauges of photon propagator) is evidence of the violation of the gauge invariance and correctness of accounting for important exchange-correlation effects. The presented data show that the gauge invariance violation in the HF approach is in very large (~60%) difference between the “r” and “∇” values of σ. This confirms importance of accounting for the multi-electron correlation effects. Really, more correct RPAE approximation provides significantly better results, but some discrepancy remains [1,21]. Naturally one could get practically the entire coincidence of the "r" and "∇" values of σ ~2% [1] or corresponding oscillator strengths 0.3% [3] in the gauge invariant scheme. From physical point of view, the same situation can take qualitatively a place in our case, but more correct conclusion can be done only after the corresponding numerical calculation. In any case, it is obvious that the heavy Rydberg systems can be correctly treated only within relativistic scheme [1-3]. At last, the quantitative accuracy of describing corresponding phenomena in the Rydberg systems can be very critical as for the correct treating cited phenomena, including the thermal atomic photoionization, as their using (look the introduction) as basis for creation of new classes of the atomic sensors [3,4,16].

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