

Interaction of blackbody radiation with rubidium and caesium atoms in small-angular-momentum Rydberg states

I.L. Glukhov, A.A. Kamenski, V.D. Ovsiannikov

Abstract. We examine the possibilities of refining an asymptotic description and quantitative calculations of the effects induced by thermal blackbody radiation (BBR) of the environment on the Rydberg states of atoms. Numerical values are calculated and asymptotic expressions are proposed for simplified estimates of natural lifetimes and threshold photoionisation cross sections for Rydberg states of rubidium and caesium atoms with large values of the principal quantum number, $n \geq 20$, and small orbital momenta, $l = 0, 1, 2, 3$. Based on analytical expressions, we present numerical estimates for the contributions of photoionisation probabilities to the BBR-induced broadening of the Rydberg energy level, as well as the contributions of continuum integrals to thermally induced shifts in the Rydberg-state energy levels.

Keywords: alkali atoms, Rydberg states, radiative transitions, blackbody radiation, ionisation cross section, quantum defect method.

1. Introduction

Ultracold atoms in optical traps have been attracting much attention in modern research aimed at developing the state-of-the-art methods for precision measurements of frequency and time, as well as at developing quantum devices and information processing algorithms [1–4]. Atoms in highly excited Rydberg states are extremely sensitive to external fields and can be used for high-precision measurement of temperature of thermal radiation of the environment [so-called blackbody radiation BBR] in a given region of space [5, 6], for measuring weak fields [7], and also for metrological measurements of the characteristics of radiofrequency radiation [8]. Well-developed methods of laser excitation of high-energy states make Rydberg states with arbitrary values of the principal n and orbital l quantum numbers of the valence electron experimentally accessible [9–12].

Alkali metal atoms are promising objects of research not only due to the availability of pure substances and the ease of cooling and manipulation of individual particles, but also due to the simplicity of the one-electron optical spectrum, which makes it possible to use the one-electron approximation in calculating the characteristics of interaction with external electromagnetic fields. Rubidium and caesium atoms are the

most convenient ones for deep cooling and trapping in optical traps, because they have the largest mass among alkali atoms, providing the lowest recoil energy and the most convenient ways to trap and control the movement of atoms in optical traps.

The structure of the excitation energy spectrum of the outer (valence) electron of an alkali atom is described with good accuracy by the Rydberg formula, in which the energies of one-electron states is written using the formulae of a hydrogen-like atom with the introduction of the concept of a quantum defect. A semi-empirical method of the Fues model potential (FMP) and the quantum defect method (QDM) have been developed on the basis of the Rydberg formula, and are widely used in numerical calculations, in which single-electron wave functions are represented as modified wave functions of a hydrogen-like atom, making it possible to obtain analytical expressions for the amplitudes of electromagnetic transitions between bound and free states of many-electron atoms and ions. The numerical values of quantum defects are determined from the most reliable tables of energy levels [13, 14]. For levels with large principal quantum numbers, the energies of which are not available in the literature, the definition of quantum defects is based on asymptotic expressions similar to those given, for example, in [15, 16].

In this work, we study the interaction of thermal radiation with rubidium and caesium atoms in small-angular-momentum Rydberg states. The amplitudes of bound–bound transitions are calculated in the QDM approximation and their results are compared with the results obtained in the framework of the FMP method. The data of numerical calculations of the lifetimes of the nS , nP , nD , and nF Rydberg states of rubidium and caesium atoms are discussed in Section 2. Tables are given of numerical values of the natural lifetimes of excited states with $n \leq 10$, calculated within the framework of the QDM and FMP approximations, in comparison with well-known literature data.

In Section 3, we consider the possibility of determining the dependence of the shift and broadening of the Rydberg states in the BBR field on the principal and orbital quantum numbers. General formulae for the cubic term of the Farley–Wing function [17] are presented and their possible use to obtain corrections to the asymptotic formulae of works [18, 19] in analytical form [20–23] is discussed.

In Section 4, we consider the contribution of continuum states to the broadening and shift of Rydberg states with small orbital momenta. Unlike states with large orbital momenta, $l > n/2$, for which the amplitudes of radiative transitions are exponentially small, the contribution of transitions to continuum states to the shift and width of Rydberg states with principal quantum numbers $n > 20$ and small orbital

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momenta can reach significant values (up to 10% and higher in the range of BBR temperatures over 300 K). Therefore, taking into account the ionisation contribution to the broadening and the contribution of the integral over the continuous spectrum to the shift of energy levels with $l < 5$ becomes relevant. For numerical estimates of the corresponding quantities, use is made of semi-classical expressions for the threshold ionisation cross sections. Within the framework of the QDM, numerical values of the cross sections for a series of nS , nP , nD , and nF Rydberg states with principal quantum numbers $n = 20-300$ are obtained. For fast estimates of cross sections, a polynomial asymptotic formula is given with polynomial coefficients determined by the standard method of polynomial approximation of the numerical values of a function for given arguments. All formulae are presented in the atomic system of units.

2. Natural width of small-orbital-momentum Rydberg states

The natural width of the excited state of an atom $|nl\rangle$ is determined by the total probability of spontaneous radiative decays into states $|n'l'\rangle = l \pm 1$ with lower energy:

$$\Gamma_{nl}^{\text{sp}} = \frac{4}{3c^3(2l+1)} \sum_{\substack{l_s \omega_{n'n}^3 |\langle n'l'|r|nl\rangle|^2 \\ (E_{n'l'} < E_{nl})}} l_s \omega_{n'n}^3 |\langle n'l'|r|nl\rangle|^2, \quad (1)$$

where $l_s = (l+l'+|l-l'|)/2$ is the larger of the two values, l and l' ; and $\omega_{n'n}$ is the transition energy. The main quantities that determine probability (1) are the radial matrix elements of dipole transitions $\langle n'l'|r|nl\rangle$. The analytical expression for the radial wave function of the bound state in the FMP method is written in terms of the associated Laguerre polynomial [21],

$$R_{nl}^{\text{FMP}}(r) \propto \exp(-x/2) x^\lambda L_{n_r}^{2\lambda+1}(x),$$

with the argument $x = 2Zr/v$ proportional to the radial variable r , with a noninteger effective principal quantum number determined by the energy of the state $v = Z/\sqrt{-2E_{nl}}$, where Z is the charge of the residual ion in whose field the Rydberg electron moves; and $n_r = 0, 1, 2, \dots$ is the radial quantum number and λ is the effective orbital quantum number ($\lambda = v - n_r - 1$). The polynomial of the wave function of the QDM [23]

$$\begin{aligned} R_{nl}^{\text{QDM}}(r) &\propto \frac{W_{v,l+1/2}(x)}{x} \\ &= \exp(-x/2) x^{v-1} {}_2\tilde{F}_0(l+1-v, -l-v; -1/x) \end{aligned}$$

is determined from the truncated asymptotic expansion of the Whittaker function $W_{v,l+1/2}(x)$ and consists of the products of the factor x^{v-1} bounded at $r \rightarrow 0$ and the first terms of the series of the hypergeometric function ${}_2\tilde{F}_0(l+1-v, -l-v; -1/x)$ with argument $-1/x = -v/(2Zr)$. The matrix element $\langle n'l'|r|nl\rangle$ both in the FMP method and in the QDM can be written as a hypergeometric type polynomial in two arguments [16, 23].

Despite the identical fundamentals of the FMP and quantum defect methods based on the semi-empirical hydrogen-like form of one-electron wave functions, there is a significant difference between the results of numerical calculations of the

amplitudes of radiative transitions within the framework of these methods. In particular, the difference (especially for $nD_{5/2}$ states) can be seen for the calculated natural lifetimes τ_{nl}^{sp} of excited states with small orbital momenta of Rb and Cs atoms presented in Tables 1 and 2 in comparison with the most reliable literature data [14].

Table 1. Lifetimes of the excited nS , $nP_{3/2}$, $nD_{5/2}$, and $nF_{7/2}$ states with the maximum total angular momentum $J = l + 1/2$ in Rb atoms, calculated in the QDM approximation, in comparison with the data obtained by the FMP method and with the most reliable data compiled in the information system [14].

State	Lifetime/ns		
	QDM	FMP	[14]
6S	52.6	77.4	50
7S	102.3	134.8	95
8S	191.6	235.6	160
9S	329.7	386.7	290
10S	527.2	597.5	450
5P _{3/2}	26.05	25.1	27
6P _{3/2}	91.3	98.4	120
7P _{3/2}	198.1	210.2	240
8P _{3/2}	354.2	371.2	440
9P _{3/2}	570.2	595.5	730
10P _{3/2}	855.3	895.5	1100
4D _{5/2}	85.2	91.8	80
5D _{5/2}	214.1	753.2	238.5
6D _{5/2}	207.8	1811.2	238
7D _{5/2}	257.4	2549.5	331
8D _{5/2}	339.6	3348.4	434
9D _{5/2}	450.4	4368.8	596
10D _{5/2}	590.6	5659.5	806
4F _{7/2}	55.03	42.8	55
5F _{7/2}	101.3	80.9	100
6F _{7/2}	168.9	136.3	165
7F _{7/2}	262.2	212.5	250
8F _{7/2}	385.4	313.1	365

As can be seen from Tables 1 and 2, calculations within the framework of the QDM yield numerical values of τ_{nl}^{sp} , which are mostly closer to the literature data than the numerical values obtained using the FMP method. This result is explained, in particular, by the fact that the main contribution to the probability of spontaneous decay of the states of the nl series is made by the amplitudes of dipole-allowed radiative transitions to the lowest states of the series with $n'l' = l \pm 1$. In the region of localisation of wave functions of low states $|n'l'\rangle$, QDM wave functions containing terms from the asymptotic polynomial for the Whittaker function, proportional to $r^{(v)-1}$, yield more accurate values of radial integrals than the rapidly decreasing FMP functions (proportional to r^{v-1}). Here $\{\{v\} = v - [v]$ is the fractional part of the number v , and $[v]$ is the integer. The results of numerical calculations performed in the QDM approximation for the lifetimes $\tau_{nl}^{\text{sp}} = 1/\Gamma_{nl}^{\text{sp}}$ of Rydberg states with principal quantum numbers $n = 20-350$ using formula (1) for the widths are approximated by expressions of the form

$$\tau_{nl}^{\text{sp}} = \tau_l^{(0)} n^3 \left(1 + \frac{\tau_l^{(1)}}{n} + \frac{\tau_l^{(2)}}{n^2} \right) \quad (2)$$

Table 2. Lifetimes of the excited nS , $nP_{3/2}$, $nD_{5/2}$, and $nF_{7/2}$ states in Cs atoms, calculated in the QDM approximation, in comparison with the results obtained by the FMP method and with the most reliable data compiled in the information system [14].

State	Lifetime/ns		
	QDM	FMP	[14]
7S	58.1	89.6	55
8S	109.5	150.9	100
9S	199.9	258.2	170
10S	337.3	417.9	280
11S	537.1	643.1	450
6P _{3/2}	30.0	28.9	30
7P _{3/2}	105.8	82.3	135
8P _{3/2}	234.2	161.0	320
9P _{3/2}	419.6	274.8	600
10P _{3/2}	671.3	433.7	1200
5D _{5/2}	1344.7	1026.9	1150
6D _{5/2}	60.6	622.3	60
7D _{5/2}	83.6	621.0	95
8D _{5/2}	122.8	780.6	150
9D _{5/2}	183.8	1123	240
10D _{5/2}	260.7	1542	350
4F _{7/2}	53.2	30.1	50
5F _{7/2}	95.7	55.7	85
6F _{7/2}	157.9	93.2	140
7F _{7/2}	245.0	145.3	220
8F _{7/2}	360.3	214.1	320

taking into account the asymptotic dependence on the principal quantum number, $\tau_{nl}^{sp} \propto n^3$.

The numerical values of the coefficients $\tau_i^{(j)}$ ($i = 0, 1, 2$) of the polynomial in (2) listed in Table 3 in powers of the reciprocal principal quantum number $1/n$ are determined by the standard method of polynomial interpolation of the values of the function $\tau_{nl}^{sp}(n)$ at $n = 20, 60, 120$ (see, for example, [16]). Comparison with the results obtained using formula (1) shows that the relative error of the approximation values (2) does not exceed 1% in the region $20 < n < 120$. In the regions below ($15 < n < 20$) and above ($120 < n < 350$) of the interpolation area, the error does not exceed 2%–3%.

Table 3. Coefficients of the quadratic polynomial of the asymptotic approximation (2) for the natural lifetimes of the nS , $nP_{3/2}$, $nD_{5/2}$, and $nF_{7/2}$ Rydberg states in rubidium and caesium atoms.

Atom	Series	$\tau_i^{(0)}/ns$	$\tau_i^{(1)}$	$\tau_i^{(2)}$
Rb	nS	1.4854	−1.5379	−93.390
	$nP_{3/2}$	1.5251	−2.8479	−26.835
	$nD_{5/2}$	0.41793	10.585	−101.88
	$nF_{7/2}$	0.70945	0.063801	3.3892
Cs	nS	1.6263	−8.5518	−18.887
	$nP_{3/2}$	1.3394	−1.5699	−51.421
	$nD_{5/2}$	0.34250	1.0137	−54.749
	$nF_{7/2}$	0.67003	0.36313	−3.3485

Numerical calculations show that the use of a cubic or higher (fourth, fifth, and so on) order of the approximating polynomial in (2) does not lead to a decrease in the approximation error. Therefore, we restrict ourselves to the most efficient quadratic approximation.

3. Interaction energy of BBR with the Rydberg atom

Obviously, when measuring the lifetime of an excited state, the atom is influenced by the ubiquitous thermal radiation (BBR) of the environment, produced by objects that have a non-zero temperature T . Under the influence of the BBR field, the energy levels experience a shift and broadening due to the dynamic Stark effect. The effects of shift and broadening of the bound energy level can be quantitatively determined using the general expression for the energy of interaction with the BBR [17, 20–23]

$$E_{nl}^{BBR}(T) = -\frac{2(k_B T)^3}{3\pi c^3} \left\{ \sum_{n'l'm'} \left[|\langle n'l'm' | r_\mu | nlm \rangle|^2 F\left(\frac{\omega_{n'n}}{k_B T}\right) \right] + \sum_{l'm'\mu} \int_0^\infty |\langle \epsilon l' m' | r_\mu | nlm \rangle|^2 F\left(\frac{\epsilon + |E_{nl}|}{k_B T}\right) d\epsilon \right\}, \quad (3)$$

where m and m' are magnetic quantum numbers; μ is the projection of the electric dipole moment operator; ϵ is the state energy in the continuous spectrum; and

$$F(y) = -2y \int_0^\infty \frac{x^3 dx}{(x^2 - y^2)(\exp x - 1)} \quad (4)$$

is a function introduced by Farley and Wing [17], which takes into account the integration of the thermally induced dynamic Stark shift over the BBR frequency spectrum described by the Planck distribution.

The asymptotic expression for the energy (3) of an atom in the Rydberg state $|nl\rangle$ with a large value of the principal quantum number n and an arbitrary orbital angular momentum l can be written as [17–19]

$$E_{nl}^{BBR}(T) \approx \epsilon_0(T) - i \frac{\Gamma_0(T)}{2} = \frac{\pi(k_B T)^2}{3c^3} - i \frac{2Z^2 k_B T}{3c^3 n^2} \\ = 2.41665 \left(\frac{T}{300}\right)^2 - i 1619.40 \frac{Z^2 T}{n^2 300}, \quad (5)$$

where E_{nl}^{BBR} is expressed in kHz. The real part of this expression, obtained from (3) in the limit $Z^2/(n^2 k_B T) \rightarrow 0$, determines the thermally induced shift $\epsilon_0(T) = \pi(k_B T)^2/(3c^3)$, and the imaginary part is the broadening of the energy level $\Gamma_0(T) = 4Z^2 k_B T/(3c^3 n^2)$. Note that the shift $\epsilon_0(T)$ is the same for all Rydberg states satisfying the condition $Z^2/n^2 \ll k_B T$ and does not depend on either the principal n or the orbital quantum numbers l . The imaginary part [broadening $\Gamma_0(T)$] depends only on n and does not depend on the orbital quantum number. Dependences on n and l appear only in corrections to the asymptotic expression (5), which are represented as an expansion in powers of a small parameter $\eta = Z^2/(n^3 k_B T)$ [21].

The calculation of corrections to the asymptotic expression (5) for atoms in Rydberg states with small orbital momenta ($l \leq 3$) must be started with an investigation of the analytic properties of function (4). The singularity of the integrand indicates the complexity of this function, which corresponds to the complexity of energy (3). To isolate the real and imaginary parts, it suffices to use the Sokhotski–Plemelj theorem written using generalised functions in the form

$$\frac{f(x)}{x - (y + i\varepsilon)} \Big|_{\varepsilon \rightarrow 0} = \text{P} \frac{f(x)}{x - y} + i\pi f(x) \delta(x - y), \quad (6)$$

where P is the principal value of the Cauchy integral and $\delta(x - y)$ is the Dirac delta function. Let us isolate the real and imaginary parts of function (4), first transforming it to the form

$$F(y) = -\frac{\pi^2}{3}y - 2y^3\Phi(y), \quad (7)$$

where

$$\Phi(y) = \int_0^\infty \frac{x dx}{(x^2 - y^2)(\exp x - 1)}.$$

Then

$$\text{Re}[F(y)] = -\frac{\pi^2}{3}y - 2y^3 \text{P} \int_0^\infty \frac{x dx}{(x^2 - y^2)(\exp x - 1)}, \quad (8)$$

$$\text{Im}[F(y)] = -2y^3 \text{Im}[\Phi(y)] = \frac{\pi |y|^3}{\exp |y| - 1}, \quad (9)$$

By summing over the magnetic quantum numbers m' and over the projections μ of the electric dipole moment operator in (3), we can obtain the following expressions for the BBR-induced shift $\varepsilon_{nl}^{\text{BBR}}(T) = \text{Re}[E_{nl}^{\text{BBR}}(T)]$ and the broadening $\text{Im}[E_{nl}^{\text{BBR}}(T)] = -\Gamma_{nl}^{\text{BBR}}(T)/2$:

$$\begin{aligned} \varepsilon_{nl}^{\text{BBR}}(T) &= \frac{2(k_{\text{B}}T)^3}{\pi c^3(2l+1)} \sum_{l'} l' | \langle n'l' | r | nl \rangle |^2 \\ &\times \left\{ \frac{\pi^2 \omega_{n'n}}{3k_{\text{B}}T} + 2 \left(\frac{\omega_{n'n}}{k_{\text{B}}T} \right)^3 \text{Re} \left[\Phi \left(\frac{\omega_{n'n}}{k_{\text{B}}T} \right) \right] \right\}, \quad (10) \end{aligned}$$

$$\Gamma_{nl}^{\text{BBR}}(T) = \frac{4}{3c^3(2l+1)} \sum_{l'} l' \frac{|\omega_{n'n}|^3 | \langle n'l' | r | nl \rangle |^2}{\exp[\omega_{n'n}/(k_{\text{B}}T)] - 1}. \quad (11)$$

The summation over the set of bound states with principal quantum numbers n' in these expressions also implies integration over the states of the continuous spectrum with positive energy $\varepsilon > 0$, which is explicitly shown in formula (3). The first term in curly brackets in expression (10) for the shift allows one to use the rule of summation of oscillator strengths and obtain the value of the real part $\varepsilon_0(T)$ of the asymptotic formula (5).

However, the linear term in (8) ensures the accuracy of at least four decimal places of the real part $\text{Re}[F(y)]$ only for $|y| < 0.05$. The cubic term can be taken into account using, for example, the integral representation of the logarithmic derivative of the gamma function $\psi(x) = d\{\ln[\Gamma(x)]\}/dx$ [23], which can be written as a series [24, 25]:

$$\begin{aligned} 2\text{Re}[\Phi(y)] &= \ln \xi - \text{Re}[\psi(i\xi)] \\ &= \ln \xi + \gamma - \xi^2 \sum_{k=1}^{\infty} \frac{1}{k(k^2 + \xi^2)}. \quad (12) \end{aligned}$$

Here $\gamma = 0.5772156649$ is Euler's constant. The convergence of the series in this expression strongly depends on the numer-

ical value of the parameter $\xi = |y|/2\pi$, so that for $\xi > 1$ the complexity of determining the numerical value of the series exceeds the complexity of direct calculation of the integral in (7). However, if we single out the first terms by limiting the summation index to an integer $k_0 \gg |\xi|$,

$$S = \sum_{k=1}^{\infty} \frac{1}{k(k^2 + \xi^2)} = S_0 + S', \quad (13)$$

where

$$S_0 = \sum_{k=1}^{k_0} \frac{1}{k(k^2 + \xi^2)},$$

$$S' = \sum_{k=k_0+1}^{\infty} \frac{1}{k(k^2 + \xi^2)},$$

then the remaining part of the series, represented by the second term S' , converges no worse than the series

$$\sum_{k=k_0+1}^{\infty} \frac{1}{k^3}.$$

To estimate this term, summation can be replaced by integration:

$$S' \approx \int_{k_0+1}^{\infty} \frac{dx}{x(x^2 + \xi^2)} = \frac{1}{2\xi^2} \ln \left[1 + \left(\frac{\xi}{k_0+1} \right)^2 \right]. \quad (14)$$

Substituting expressions (12)–(14) into (8), we obtain

$$\begin{aligned} \text{Re}[F(y)] &= -\frac{2\pi^3}{3}\xi \\ &\times \left\{ 1 + 12\xi^2 \left[\frac{1}{2} \ln \left(\frac{\xi^2}{1 + \xi^2/(k_0+1)^2} \right) + \gamma - \xi^2 S_0(\xi) \right] \right\}. \quad (15) \end{aligned}$$

Numerical calculation shows that the accuracy of this expression decreases as the parameter ξ increases. Nevertheless, despite a significant decrease in the number of significant digits of the terms inside the square bracket (more than five digits), then in the curly bracket (five more digits), for $y \leq 25$, expression (15) yields a numerical value of the function $\text{Re}[F(y)]$ with an error of less than 1%. In practical calculations of the real part of energy (10), to determine the numerical values of function (8), it is advisable to use expression (15) only in the range of numerical values of the argument $|y| < 10$, where it can be accurate to at least five decimal places. In this case, the numerical value of the parameter ξ is limited by the relation $\xi < 2$, and it is sufficient to limit the values of the parameter k_0 in formulae (13)–(15) by the condition $k_0 < 10$. In the region $|y| < 40$, the accuracy of the fifth decimal place of the function $\text{Re}[F(y)]$ can only be obtained by numerical integration in the second term of expression (8). For $|y| > 40$, the accuracy of five decimal places is provided by the first four terms of the asymptotic expansion in a series in odd powers of the inverse argument $1/y$ [20]:

$$\text{Re}[F(y)] = \sum_{p=0}^{\infty} (-1)^{p+1} \frac{B_{2p+4}(2\pi)^{2p+4}}{(2p+4)y^{2p+1}} =$$

$$= \frac{2\pi^4}{15y} \left[1 + \frac{10}{21} \left(\frac{2\pi}{y} \right)^2 + \frac{1}{2} \left(\frac{2\pi}{y} \right)^4 + \frac{10}{11} \left(\frac{2\pi}{y} \right)^6 + \dots \right], \quad (16)$$

where B_{2p+4} are the Bernoulli numbers [25].

To calculate the BBR-induced broadening of energy levels, expression (11) can be conveniently represented as

$$\Gamma_{nl}^{\text{BBR}}(T) = \frac{4k_B T}{3c^3(2l+1)} \times \sum_{n', l' = l \pm 1} l_{>} \omega_{n'n}^2 |\langle n'l'|r|nl \rangle|^2 \frac{y}{\exp y - 1}, \quad (17)$$

where $y = |\omega_{n'n}|/(k_B T)$. Using the expansion in a power series with Bernoulli coefficients B_k for the function $f(y) = y/(\exp y - 1)$ [25]

$$f(y) = \sum_{k=0}^{\infty} B_k \frac{y^k}{k!} = 1 - \frac{y}{2} + \frac{y^2}{12} - \frac{y^4}{720} + \frac{y^6}{720 \times 42} - \frac{y^8}{720 \times 42 \times 40} + \dots, \quad (18)$$

we obtain an explicit expression for the thermally induced width, taking into account the corrections to the asymptotic value (5) depending on the quantum numbers n , l , and temperature T :

$$\Gamma_{nl}^{\text{BBR}}(T) = \frac{4k_B T}{3c^3(2l+1)} \sum_{n', l' = l \pm 1} l_{>} |\langle n'l'|r|nl \rangle|^2 \times \left[\omega_{n'n}^2 - \frac{|\omega_{n'n}|^3}{2k_B T} + \sum_{k=1}^{\infty} \frac{B_{2k}}{(2k)!} \frac{\omega_{n'n}^{2k+2}}{(k_B T)^{2k}} \right]. \quad (19)$$

Here, the first term in square brackets corresponds to the first term from expansion (18), which, after summing over the entire set of atomic states, including the integral over the states of the continuum, yields an asymptotic expression for the imaginary part from formula (5):

$$\Gamma_{nl}^{\text{BBR}}(T) \approx \Gamma_0(T) = \frac{4Z^2 k_B T}{3c^3 n^2}.$$

The second term in (19), which corresponds to the second term in expansion (18), yields a temperature-independent expression, which exactly coincides with expression (1) for the spontaneous width with a negative sign, $-\Gamma_{nl}^{\text{sp}}$ [21]. Thus, the total decay rate of the Rydberg atom, $\Gamma_{nl}^{\text{tot}}(T) = \Gamma_{nl}^{\text{BBR}}(T) + \Gamma_{nl}^{\text{sp}}$, which is the only one that can be measured in practice, contains only terms that depend on T , since Γ_{nl}^{sp} is compensated by the second term from expression (19). Note that this is the only term with an odd power of the transition frequency $|\omega_{n'n}|^3$. All subsequent terms of the series in (19) contain only even powers of frequencies $\omega_{n'n}^{2k+2}$. The result of their summation over the complete set of states $|n'l'\rangle$ can be formally called the oscillator-strength moment sum rules [22].

However, for states with a small orbital momentum l , the use of sums of moments with the exponent $k > l$ turns out to be impossible due to the divergence of the corresponding sums over the states $|n'l'\rangle$. The explicit form of expansion coefficients (18) indicates a rapid decrease in the terms of the

infinite series with Bernoulli coefficients in (19) at BBR temperatures $T > 100$ K. In practice, the term with $k = 1$ is quite sufficient to estimate the correction to the broadening $\Gamma_0(T)$ of states with moments $l \geq 1$. In fact, the terms of the infinite series in (17) with large values of the argument of the function $f(y)$ are exponentially small, so that the use of expansion (18) for $y > 1$ becomes impractical. Thus, the presence of the factor $f(y)$ ensures a rapid convergence of the series in (17) for any values of the orbital angular momentum l .

4. Contribution of the continuous spectrum to the thermally induced shift and broadening of Rydberg states with small orbital momenta

Infinite sums of expansions over complete sets of atomic eigenstates for BBR-induced shifts (10) and broadenings (11) of energy levels include integrals over continuum states with positive energy. In contrast to states with large orbital momenta, in which the contribution of the continuum is exponentially small [26], the contribution of integrals over the continuous spectrum for states with small orbital momentum ($l \ll n$) can be significant. The integrands of such integrals can be expressed in terms of the photoionisation cross section $\sigma_{nl}(\omega)$, for which the frequency dependence of BBR photons can be written in the semi-classical approximation [27] as $\sigma_{nl}(\omega) \approx \sigma_{nl}^{\text{th}} |E_{nl}(\omega)|^{7/3}$, where $\sigma_{nl}^{\text{th}} = \sigma_{nl}(\omega)|_{\omega=|E_{nl}|}$ is the threshold cross section, and $|E_{nl}|$ is the binding energy of the Rydberg state, which makes it possible to obtain closed analytical expressions for the integrals. In particular, for the shift

$$\varepsilon_{\text{cont}}^{\text{BBR}}(T) \approx \frac{(k_B T)^2}{18\pi c^2} \int_{|E_{nl}|}^{\infty} \sigma_{nl}(\omega) d\omega \approx \frac{(k_B T)^2}{24\pi c^2} \sigma_{nl}^{\text{th}} |E_{nl}| \quad (20)$$

quantities σ_{nl}^{th} and $|E_{nl}|$ determine the contribution of the continuum to the broadening, which represents the ionisation probability [17]

$$\Gamma_{nl}^{\text{ion}}(T) \approx \frac{3k_B T}{(\pi c)^2} \sigma_{nl}^{\text{th}} |E_{nl}|^2. \quad (21)$$

Thus, in the general expressions for shift (10) and broadening (11) of states with large principal quantum numbers and low binding energy, satisfying the relation $|E_{nl}| \ll k_B T$, it suffices to take into account only the sums over the states of the discrete spectrum, and replace the integrals over the continuum with expressions (20) and (21), respectively. An important characteristic in these expressions is the threshold ionisation cross section. The numerical values, as well as the parameters of the approximation formulae for simplified estimates of σ_{nl}^{th} for the nS , nP , and nD series of Rydberg states of alkali atoms, were obtained by the FMP method in [28]. In the present work, the numerical values of σ_{nl}^{th} for the nS , nP , nD , and nF series of Rydberg states of Rb and Cs atoms with principal quantum numbers $n = 20-300$ are calculated within the framework of the QDM. The values of the radial matrix elements of electric dipole transitions from the Rydberg state to the threshold state with zero energy are determined using the limiting relation

$$|\langle \varepsilon = 0, l' | r | nl \rangle|^2 = \lim_{n' \rightarrow \infty} (n')^3 |\langle n', l' | r | nl \rangle|^2.$$

Coefficients of a polynomial of asymptotic approximation quadratic in powers of $1/n$

$$\sigma_{nl}^{\text{th}} = s_l^{(0)} \left(1 + \frac{s_l^{(1)}}{n} + \frac{s_l^{(2)}}{n^2} \right) \quad (22)$$

are obtained by the standard method of polynomial interpolation with respect to the given values of function (22). Numerical values of the coefficients $s_l^{(i)}$ ($i = 0, 1, 2$) for the nS , nP , nD , and nF series of Rydberg states of Rb and Cs atoms, interpolated from the values of the threshold cross sections for the states with $n = 20, 60, 120$ are given in Table 4. It should be noted that similar interpolation formulae from [28] are in good agreement with the results of calculations of σ_{nl}^{th} using formula (22) with the parameters taken from Table 4. As for the spontaneous lifetime (2), the use of higher-order polynomials instead of quadratic polynomials does not reduce the approximation errors. Numerical calculations confirm the optimality of approximations (2) and (22) by second-order polynomials with coefficients taken from Tables 3 and 4, respectively.

Table 4. Coefficients of the polynomial of the asymptotic approximation (22) of the threshold ionisation cross section σ_{nl}^{th} (in megabarns, $1 \text{ Mb} = 10^{-18} \text{ cm}^2 = 0.03571 \text{ a. u.}$) for the nS , $nP_{3/2}$, $nD_{5/2}$, and $nF_{7/2}$ series of Rydberg states with the maximum total momentum $J = l + 1/2$ in rubidium and caesium atoms.

Ion	Series	$s_l^{(0)}/\text{Mb}$	$s_l^{(1)}$	$s_l^{(2)}$
Rb	nS	0.676395	-10.516	5.249
	$nP_{3/2}$	0.231548	63.406	-489.93
	$nD_{5/2}$	1.04094	-6.9262	52.682
	$nF_{7/2}$	2.60133	-1.7406	49.745
Cs	nS	0.689011	4.6493	-100.04
	$nP_{3/2}$	0.864189	80.377	-451.74
	$nD_{5/2}$	1.10844	14.023	-294.93
	$nF_{7/2}$	2.14534	-10.497	229.47

5. Conclusions

In this work, new information is obtained about the effect of thermal radiation on the shift and broadening of the energy levels of the nS , nP , nD , and nF series of Rydberg states in rubidium and caesium atoms. Analytical expressions for the cubic term of the Farley–Wing function (7) make it possible to calculate corrections to the asymptotic formulae for thermally induced broadening and shift, which depend on the BBR temperature, on the principal and orbital quantum numbers of the Rydberg state, and also to determine the accuracy and limits of applicability of the linear approximation for function (7). The threshold photoionisation cross sections σ_{nl}^{th} are calculated and an approximate asymptotic formula is obtained for simplified estimates of the numerical values of σ_{nl}^{th} needed to determine the contribution of the integral over the continuous spectrum (20) to the shift, as well as the contribution of the photoionisation rate (21) to the broadening of the energy levels (11) of the small-angular-momentum Rydberg states.

Thus, we can list the following main results of this work:

- approximation formula (2) and numerical values of the coefficients of the quadratic polynomial are obtained (see Table 3) for quick estimates of the natural lifetime of the Rydberg states of rubidium and caesium atoms;
- formulae (10)–(16) are derived for BBR-induced shifts, as well as broadenings (19), taking into account corrections to

the asymptotic expression (5), depending temperature T , principal n and orbital l quantum numbers; and

- the contributions of the states of the continuous spectrum to the BBR-induced shifts (20) and broadening (21) of the Rydberg states are expressed in terms of the threshold ionisation cross sections, represented by the asymptotic relation (22) with the coefficients of the approximation polynomial given in Table 4.

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