# **Spectroscopy of Radiative Decay Processes in Heavy Rydberg Alkali Atomic Systems**

### **Valentin B. Ternovsky, Alexander V. Glushkov, Olga Yu. Khetselius, Marina Yu. Gurskaya and Anna A. Kuznetsova**

**Abstract** We present the results of studying the radiation decay processes and computing the probabilities and oscillator strengths of radiative transitions in spectra of heavy Rydberg alkali-metal atoms. All calculations of the radiative decay (transitions) probabilities have been carried out within the generalized relativistic energy approach (which is based on the Gell-Mann and Low S-matrix formalism) and the relativistic many-body perturbation theory with using the optimized one-quasiparticle representation and an accurate accounting for the critically important exchange-correlation effects as the perturbation theory second and higher orders ones. The precise data on spectroscopic parameters (energies, reduced dipole transition matrix elements, amplitude transitions) of the radiative transitions  $nS_{1/2}$  $\rightarrow$  n'P<sub>1/2,3/2</sub> (n = 5, 6; n' = 10–70), nP<sub>1/2,3,2</sub>  $\rightarrow$  n'D<sub>3/2,5/2</sub> (n = 5, 6; n' = 10–80) in the Rydberg Rb, Cs spectra and the transitions  $7S_{1/2}$ -nP<sub>1/2,3/2</sub>,  $7P_{1/2,3,2}$ -nD<sub>3/2,5/2</sub>  $(n = 20-80)$  in the Rydberg francium spectrum are presented. The obtained results are analyzed and discussed from viewpoint of the correct accounting for the relativistic and exchange-correlation effects. It has been shown that theoretical approach used provides an effective accounting of the multielectron exchange-correlation effects, including effect of essentially non-Coulomb grouping of Rydberg levels and others.

**Keywords** Radiation decay processes ⋅ Multielectron atoms and multicharged ions ⋅ Relativistic energy approach

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### **1 Introduction**

Accurate radiative decay widths and probabilities, oscillator strengths of radiative transitions in spectra of the Rydberg atomic systems (atoms in the highly excited states with large values of the principal quantum number  $n \gg 1$ ) are of a great interest for astrophysical analysis, laboratory, thermonuclear plasma diagnostics, fusion research etc. (see, for example [[1](#page-9-0)–[60\]](#page-11-0)). In recent years intensive theoretical and experimental investigations of spectroscopic properties of the Rydberg atoms are also stimulated by a great number of their possible important applications in atomic and molecular optics and spectroscopy, quantum electronics, laser physics (for example, speech is about new lasing schemes in the short-wave range with using the Rydberg systems), quantum informatics and computing, astrophysics etc. It is well known that the Rydberg atoms make the contribution into interstellar clouds absorption spectrum (Rydberg states with  $n \sim 300-700$ ). The unique properties of the Rydberg atoms are associated with too small ionization potentials, sufficiently large size, enough long lifetime compared to conventional atomic states, finally, unprecedented sensitivity to external fields. Really, it is well known that the Rydberg atomic systems are very sensitive to electromagnetic fields and can be used for the detection and sensing static and AC electric and magnetic fields. Strongly interacting Rydberg systems have unique photon emission properties. These facts stimulate more intensive research of the Rydberg atoms, in particular, on the basis of new experimental methods of laser spectroscopy, beam-foil spectroscopy, using magneto-optical traps, synchrotron radiation sources, cryogenic devices and so on. It is worth to remind about such unique and interesting physical objects and phenomena such as the Rydberg matter, Bose-condensate in vapors of the Rydberg alkali-metal atoms, fountains of cold Rydberg atoms etc.

The well-known quasiclassical and quantum-mechanical approaches such as the Hartree-Fock (HF) and Dirac-Fock (DF) methods, quantum defect and the Coulomb approximations, the model potential and pseudopotential methods etc. have been used to calculate the spectroscopic properties of different light and middle Rydberg atoms. In a case of the heavy Rydberg atoms in a free state or in an external electromagnetic field a modern level of description of the Rydberg atoms is not sufficiently satisfactory. A precise accounting for the relativistic and exchange-correlation (XC) effects, including an effect of the non-Coulomb grouping levels in the Rydberg spectra (the effect, which, as a rule, is not considered within simplified Coulomb and quantum defect models) is of a great interest and importance.

The purpose of this work is to present the results of studying the radiation decay processes and computing probabilities and oscillator strengths of the radiative transitions in the spectra of heavy Rydberg atoms of alkali-metal elements. The precise data on spectroscopic parameters (energies, reduced dipole transition matrix elements, amplitude transitions) of the radiative transitions  $nS_{1/2} \rightarrow n'P_{1/2,3/2}$  (n = 5, 6; n' = 10–70), nP<sub>1/2,3,2</sub> → n'D<sub>3/2,5/2</sub> (n = 5, 6; n' = 10–80) in the Rydberg Rb, Cs spectra and the transitions  $7S_{1/2}$ -nP<sub>1/2,3/2</sub>,  $7P_{1/2,3,2}$ -nD<sub>3/2,5/2</sub> (n = 20–80) in the

Rydberg francium spectrum are listed. The data are discussed from the viewpoint of the correct accounting for the relativistic and exchange-correlation effects. It has been shown that the theoretical approach used provides a precise accounting for the important exchange-correlation effects, including the effect of essentially non-Coulomb grouping of Rydberg levels, continuum pressure etc.

All calculations of the radiative decay (transitions) probabilities (matrix elements) in the studied atomic systems have been performed with using the generalized relativistic energy approach and the relativistic many-body perturbation theory (PT) with using the optimized one-quasiparticle representation and an accurate accounting of the exchange-correlation effects, including the effect of essentially non-Coulomb grouping of Rydberg levels [\[61](#page-11-0)–[63](#page-11-0)].

Let us remind that the theoretical fundamentals of an energy approach in a case of the one-electron ions have been considered by Labzovsky et al. [[57,](#page-11-0) [58\]](#page-11-0). Originally the energy approach to radiative and autoionization processes in multielectron atoms and ions has been developed by Ivanova-Ivanov et al. [[59](#page-11-0)–[62,](#page-11-0) [64](#page-11-0)–[67](#page-11-0)]. More accurate, advanced version of the relativistic energy approach has been further developed in Refs. [[63,](#page-11-0) [68](#page-11-0)–[72](#page-11-0)]. The energy approach is based on the Gell-Mann and Low S-matrix formalism combined with the relativistic perturbation theory. In relativistic case the Gell-Mann and Low formula expressed an energy shift ΔE through the electrodynamical scattering matrix including interaction with as the photon vacuum field as a laser field. The first case is corresponding to determination of radiative decay characteristics for atomic systems. Earlier we have applied the corresponding generalized versions of the energy approach to many problems of atomic, nuclear and even molecular spectroscopy, including, cooperative electron-gamma-nuclear "shake-up" processes, electron-muon-beta-gamma-nuclear spectroscopy, spectroscopy of atoms in a laser field etc. [[73](#page-11-0)–[98\]](#page-12-0).

# **2 Relativistic Energy Approach and Many-Body Perturbation Theory with the Dirac-Kohn-Sham Zeroth Approximation**

Let us describe in brief the key moments of our theoretical approach (look for more details in Refs. [[63,](#page-11-0) [65](#page-11-0)–[69](#page-11-0), [73](#page-11-0)–[76](#page-11-0)]). As usually, the wave functions zeroth basis is found from the Dirac equation solution with self-consistent total potential.

The bare Hamiltonian is as follows:

$$
H = \sum_{i} \{ \alpha cp - \beta mc^2 + U(r_i|Z) \} + \sum_{i>j} \exp(i\omega_{ij}r_{ij}) \cdot \frac{(1 - \alpha_i\alpha_j)}{r_{ij}}, \qquad (1)
$$

where  $\alpha_i$ ,  $\alpha_j$ —the Dirac matrices,  $\omega_{ij}$ —the transition frequency, *c*—the light velocity, *Z* is a charge of the atomic nucleus. Within relativistic perturbation theory [\[3](#page-9-0), [4\]](#page-9-0) we introduce the zeroth–order Hamiltonian as:

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

<span id="page-3-0"></span>where  $V_{XC}(r_i)$ —one-particle exchange-correlation potential,  $U_{MF}(r_i|b)$ —a self-consistent Coulomb-like mean-field potential (b is the potential parameter, which is further determined within ab initio procedure), that potential interaction "quasiparticles-core" in the case of atomic system consisting of closed electron shells and external quasiparticles.

The relativistic wave functions are calculated by solution of the Dirac equation with the potential, which includes the Coulomb potential of the closed electron shells core of an alkali atomsplus the exchange Kohn-Sham potential and correlation Lundqvist-Gunnarsson potential (see details in Refs. [[63,](#page-11-0) [70,](#page-11-0) [73](#page-11-0)–[76\]](#page-11-0)).

In order to provide the construction of the optimized one-quasiparticle representation and improve an effectiveness of the numerical code we have used special ab initio procedure within relativistic energy approach [[68\]](#page-11-0) (see also [[69,](#page-11-0) [70](#page-11-0)]). It reduces to accurate treating the lowest order multielectron effects, in particular, the gauge dependent radiative contribution into imaginary part of the electron system energy Im  $\delta E_{\text{niniv}}$  for the certain class of the photon propagator calibrations and minimization of the corresponding density functional Im  $\delta E_{\text{niniv}}$ . Some known alternative approaches to construction of an optimized one-quasiparticle representation for multielectron atom can be found in Refs. [[11](#page-9-0)–[22\]](#page-10-0).

Within the relativistic energy approach  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  $[61, 62, 64, 65]$  an imaginary part of the electron energy shift of an atom is directly connected with the radiation decay possibility (transition amplitude). An approach, using the Gell-Mann and Low formula with the QED scattering matrix, is used in treating the relativistic atom. The total energy shift of the state is usually presented in the form:

$$
\Delta E = Re \Delta E + i \Gamma / 2 \tag{3}
$$

where  $\Gamma$  is interpreted as the level width, and the decay possibility  $P = \Gamma$ .

The imaginary part of an electron energy of the atomic system can be determined in the lowest second order of perturbation theory as:

$$
\text{Im}\Delta E(B) = -\frac{e^2}{4\pi} \sum_{\begin{bmatrix} \alpha > n > f \\ \alpha > n \le f \end{bmatrix}} V_{anan}^{|\omega_{an}|},\tag{4}
$$

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

$$
V_{ijkl}^{|\omega|} = \iint dr_1 dr_2 \Psi_i^*(r_1) \Psi_j^*(r_2) \frac{\sin|\omega| r_{12}}{r_{12}} (1 - \alpha_1 \alpha_2) \Psi_k^*(r_2) \Psi_l^*(r_1)
$$
 (5)

When calculating the matrix elements ([5\)](#page-3-0), one should use the angle symmetry of the task and write the corresponding expansion for  $\sin(\omega/r_{12}/r_{12})$  on spherical harmonics as follows [\[62](#page-11-0)]:

$$
\frac{\sin |\omega| r_{12}}{r_{12}} = \frac{\pi}{2\sqrt{r_1 r_2}} \sum_{\lambda=0}^{\infty} (\lambda) J_{\lambda+1/2}(|\omega| r_1) J_{\lambda+1/2}(|\omega| r_2) P_{\lambda}(\cos \widehat{r_1 r_2}) \tag{6}
$$

where *J* is the Bessel function of first kind and  $(\lambda) = 2\lambda + 1$ .

This expansion is corresponding to usual multipole one for probability of radiative decay (an amplitude approach of quantum mechanics). Substitution of the expansion ([5\)](#page-3-0) to matrix element allow to get the following expression:

$$
V_{1234}^{\omega} = [(j_1)(j_2)(j_3)(j_4)]^{1/2} \sum_{\lambda \mu} (-1)^{\mu} \binom{j_1 j_3}{m_1 - m_3} \frac{\lambda}{\mu} \times \text{Im}Q_{\lambda}(1234) \tag{7}
$$
  

$$
Q_{\lambda} = Q_{\lambda}^{\text{Qul}} + Q_{\lambda}^{\text{Br}}
$$

 $\mathcal{L}^{\text{max}}_{\text{max}}$ 

where  $j_i$  is the total single electron momentums,  $m_i$ —the projections;  $Q^{Qul}$  is the Coulomb part of interaction,  $Q^{Br}$ —the Breit part.

The total radiation width of the one-quasiparticle state can be presented in the following form:

$$
\Gamma(\gamma) = -2 \text{ Im} M^{1}(\gamma) = -2 \sum_{\lambda n l j} (2j + 1) \text{ Im} Q_{\lambda} (n_{\gamma} l_{\gamma} j_{\gamma} n l j)
$$
  
\n
$$
Q_{\lambda} = Q_{\lambda}^{Cul} + Q_{\lambda}^{Br}.
$$
  
\n
$$
Q_{\lambda}^{Br} = Q_{\lambda, \lambda - 1}^{Br} + Q_{\lambda, \lambda}^{Br} + Q_{\lambda, \lambda + 1}^{Br}
$$
\n(8)

The individual terms of the  $\Sigma_{nl}$  sum correspond to the partial contribution of the  $n_{\lambda}l_{\lambda}j_{\lambda} \rightarrow nlj$  transitions;  $\Sigma_{\lambda}$  is a sum of the contributions of the different multiplicity transitions. The detailed expressions for the Coulomb and Breit parts can be found in Refs. [[62](#page-11-0)–[66\]](#page-11-0).

The imaginary parts of the Coulomb part  $Q_{\lambda}^{Cul}$  and the Breit part contain the radial  $R_\lambda$  and angular  $S_\lambda$  integrals as follows (in the Coulomb units) [[65\]](#page-11-0):

$$
\operatorname{Im} Q_{\lambda}^{Cul}(12;43) = Z^{-1} \operatorname{Im} \left\{ R_{\lambda}(12;43) S_{\lambda}(12;43) + R_{\lambda} \left( \tilde{1}2; 4\tilde{3} \right) S_{\lambda} \left( \tilde{1}2; 4\tilde{3} \right) + R_{\lambda} \left( 1\tilde{2}; \tilde{4}3 \right) S_{\lambda} \left( 1\tilde{2}; \tilde{4}3 \right) + R_{\lambda} \left( 1\tilde{2}; \tilde{4}3 \right) S_{\lambda} \left( \tilde{1}\tilde{2}; \tilde{4}\tilde{3} \right) S_{\lambda} \left( \tilde{1}\tilde{2}; \tilde{4}\tilde{3} \right) \right\}.
$$
\n
$$
\operatorname{Im} Q_{\lambda,l}^{Br} = \frac{1}{Z} \operatorname{Im} \left\{ R_{\lambda} \left( 12; \tilde{4}\tilde{3} \right) S_{\lambda}^{l} \left( 12; \tilde{4}\tilde{3} \right) + R_{\lambda} \left( \tilde{1}\tilde{2}; 43 \right) S_{\lambda}^{l} \left( \tilde{1}\tilde{2}; 43 \right) + R_{\lambda} \left( \tilde{1}2; \tilde{4}3 \right) S_{\lambda}^{l} \left( \tilde{1}2; \tilde{4}\tilde{3} \right) S_{\lambda}^{l} \left( \tilde{1}\tilde{2}; \tilde{4}\tilde{3} \right) \right\}.
$$
\n(10)

Here  $\{\lambda l_1 l_3\}$  means that  $\lambda$ ,  $l_1$  and  $l_3$  must satisfy the triangle rule and the sum  $\lambda + l_1 + l_3$  must be an even number. The rest terms in (9), (10) include the small components of the Dirac functions. The tilde designates that the large radial component *f* must be replaced by the small one *g*, and instead of  $l_i$ ,  $\tilde{l}_i = l_i - 1$  should be taken for  $j_i < l_i$  and  $\tilde{l}_i = l_i + 1$  for  $j_i > l_i$ . The detailed definitions for the radial  $R_\lambda$ and angular  $S_\lambda$  integrals can be found in Refs. [\[59](#page-11-0), [64](#page-11-0)–[67\]](#page-11-0).

The total probability of a *λ*—pole transition is usually represented as a sum of the electric  $P_{\lambda}^{E}$  and magnetic  $P_{\lambda}^{M}$  parts. The electric (or magnetic)  $\lambda$ —pole transition *γ*  $\rightarrow$  *δ* connects two states with parities which by  $\lambda$  (or  $\lambda$  +1) units. In our designations (the radiative  $\gamma \rightarrow \delta$  transition) one could write:

$$
P_{\lambda}^{E}(\gamma \to \delta) = 2(2j+1)Q_{\lambda}^{E}(\gamma \delta; \gamma \delta) \qquad Q_{\lambda}^{E} = Q_{\lambda}^{Cul} + Q_{\lambda, \lambda-1}^{Br} + Q_{\lambda, \lambda+1}^{Br}.
$$
  
\n
$$
P_{\lambda}^{M}(\gamma \to \delta) = 2(2j+1)Q_{\lambda}^{M}(\gamma \delta; \gamma \delta) \qquad Q_{\lambda}^{M} = Q_{\lambda, \lambda}^{Br}.
$$
\n(11)

The adequate, precise computation of radiative parameters of the heavy Rydberg alkali-metal atoms within relativistic perturbation theory requires an accurate accounting for the multi-electron exchange-correlation effects (including polarization and screening effects, a continuum pressure etc.). These effects within our approach are treated as the effects of the perturbation theory second and higher orders. Using the standard Feynman diagram technique one should consider two kinds of diagrams (the polarization and ladder ones), which describe the polarization and screening exchange-correlation effects. The detailed description of the polarization diagrams and the corresponding analytical expressions for matrix elements of the polarization interelectron interaction (through the polarizable core of an alkali atom) potential is presented in Refs. [[63,](#page-11-0) [73](#page-11-0)–[76](#page-11-0)].

An effective approach to accounting of the polarization diagrams contributions is in adding the effective two-quasiparticle polarizable operator into the perturbation theory first order matrix elements. In Ref. [[65\]](#page-11-0) the corresponding non-relativistic polarization functional has been derived. More correct relativistic expression has been presented in the Refs. [\[34](#page-10-0), [35\]](#page-10-0) and used in our theory. The corresponding two-quasiparticle polarization potential looks as follows:

$$
V_{pol}^{d}(r_{1}r_{2}) = X \left\{ \int \frac{dr^{'} \left( \rho_{c}^{(0)}(r^{'} ) \right)^{1/3} \theta(r^{'} )}{|r_{1} - r^{'} | \cdot |r^{'} - r_{2}|} - \int \frac{dr^{'} \left( \rho_{c}^{(0)}(r^{'} ) \right)^{1/3} \theta(r^{'} )}{|r_{1} - r^{'} |} \int \frac{dr^{''} \left( \rho_{c}^{(0)}(r^{''}) \right)^{1/3} \theta(r^{''})}{|r^{''} - r_{2}|} \right/ \left( \rho_{c}^{(0)} \right)^{1/3} \right\}
$$
\n(12a)

$$
\langle \left(\rho_c^{(0)}\right)^{1/3} \rangle = \int dr \left(\rho_c^{(0)}(r)\right)^{1/3} \theta(r),\tag{12b}
$$

$$
\theta(r) = \left\{ 1 + \left[ 3\pi^2 \cdot \rho_c^{(0)}(r) \right]^{2/3} / c^2 \right\}^{1/2}
$$
 (12c)

where  $\rho_c^0$  is the core electron density (without account for the quasiparticle), X is numerical coefficient, c is the light velocity. The contribution of the ladder diagrams (these diagrams describe the immediate interparticle interaction) is summarized by a modification of the perturbation theory zeroth approximation mean-field central potential (look [[35,](#page-10-0) [65\]](#page-11-0)), which include the screening (anti-screening) of the core potential of each particle by the two others. The details of calculating this contribution can be found in Refs. [\[63](#page-11-0), [65](#page-11-0)–[76](#page-11-0)]. All computing was performed with using the modified PC code "Superatom-ISAN" (version 93).

### **3 Results and Conclusions**

In Tables 1 and [2](#page-7-0) we present the experimental and theoretical values (in atomic units: a.u.) of the reduced dipole transition matrix elements for the Fr and Cs atoms: experimental data—Exp; theoretical data: perturbation theory (PT)-DF<sup>SD</sup>—PT with the Dirac-Hartree-Fock zeroth approximation (single-double SD approximation in which single and double excitations of Dirac-Hartree-Fock wave functions are

Transition/ method	$\text{PT-DF}^{\text{SD}}$	$PT-DF^{SD}$ (corr)	<b>EMP</b>	PT-RHF (corr)	PT-RHF	DF	RPT-EA	Exp.
$7p_{1/2} - 7s$	4.256	-	-	4.279	4.304	4.179	4.272, 4.274	4.277
$8p_{1/2} - 7s$	0.327	0.306	0.304	0.291	0.301		0.339	
$9p_{1/2} - 7s$	0.110	0.098	0.096	$\overline{\phantom{0}}$	$\overline{\phantom{0}}$	-	0.092	
$10p_{1/2}$ -7s		-					0.063	
$7p_{3/2} - 7s$	5.851	-	-	5.894	5.927	5.791	5.891	5.898
$8p_{3/2} - 7s$	0.934	0.909	0.908	0.924	$\overline{\phantom{0}}$	$\overline{\phantom{0}}$	0.918	-
$9p_{3/2} - 7s$	0.436	0.422	0.420	$\overline{\phantom{0}}$	$\overline{\phantom{0}}$		0.426	-
$10p_{3/2} - 7s$	-						0.284	-
$7p_{1/2} - 8s$	4.184	4.237	4.230	4.165	4.219	4.196	4.228	$\overline{\phantom{0}}$
$8p_{1/2} - 8s$	10.02	10.10	10.06	10.16	10.00		10.12	-
$9p_{1/2} - 8s$	0.985	$\overline{\phantom{0}}$	0.977	$\overline{\phantom{0}}$	$\qquad \qquad -$	$\overline{\phantom{0}}$	0.972	$\overline{\phantom{0}}$
$10p_{1/2} - 8s$							0.395	
$7p_{3/2} - 8s$	7.418	7.461	7.449	7.384	7.470	7.472	7.453	-
$8p_{3/2} - 8s$	13.23	13.37	13.32	13.45	13.26		13.35	-
$9p_{3/2} - 8s$	2.245	-	2.236		-		2.232	-
$10p_{3/2} - 8s$	$\overline{\phantom{0}}$	-	-	$\overline{\phantom{0}}$	$\overline{\phantom{0}}$	-	1.058	-
$7p_{1/2} - 9s$	1.016	-	1.010	L,	-		1.062	
$8p_{1/2} - 9s$	9.280	-	9.342	-	$\overline{\phantom{0}}$	$\overline{\phantom{0}}$	9.318	-
$9p_{1/2} - 9s$	17.39	-	17.40	−	-		17.42	

**Table 1** The reduced dipole transition matrix elements for Fr (see text)

(continued)

Transition/ method	$\rm ^{\shortmid }PT\text{-}DF^{\text{SD}}$	$PT-DF^{SD}$ (corr)	EMP	PT-RHF (corr)	PT-RHF	DF	RPT-EA	Exp.
$10p_{1/2} - 9s$		-					1.836	
$7p_{3/2} - 9s$	1.393		1.380				1.41	
$8p_{3/2} - 9s$	15.88		15.92		-		15.96	
$9p_{3/2} - 9s$	22.59	$\overline{\phantom{a}}$	22.73		-		22.68	
$10p_{3/2} - 9s$	-	-	-				3.884	

<span id="page-7-0"></span>**Table 1** (continued)

Transition	$PT-DF^{SD}$	$PT-DF^{SD}$	DF	PT-RHF	<b>ODA</b>	<b>EF-RMP</b>	Exp.
		(corr)					
$6p_{1/2}$ -6s	4.482	4.535	4.510		4.282	4.489	4.4890(7)
$6p_{3/2}$ -6s	6.304	6.382	6.347		5.936	6.323	6.3238(7)
$7p_{1/2}$ -6s	0.297	0.279	0.280	0.2825	0.272	0.283	0.284(2)
$7p_{3/2}$ -6s	0.601	0.576	0.576	0.582	0.557	0.583	0.583(9)
$8p_{1/2}$ -6s	0.091	0.081	0.078		0.077	0.088	
$8p_{1/2}$ -6s	0.232	0.218	0.214	-	0.212	0.228	
$6p_{1/2} - 7s$	4.196	4.243	4.236	4.237	4.062	4.234	4.233(22)
$6p_{3/2} - 7s$	6.425	6.479	6.470	6.472	6.219	6.480	6.479(31)
$7p_{1/2} - 7s$	10.254	10.310	10.289	10.285	9.906	10.309	10.309
							(15)
$7p_{3/2} - 7s$	14.238	14.323	14.293	14.286	13.675	14.323	14.325
							(20)

**Table 2** The reduced dipole transition matrix elements for Cs (see text)

included to all PT orders); PT-DF<sup>SD</sup>-PT with the Dirac-Hartree-Fock zeroth approximation (plus compilation), EMP—empirical model potential method; PT-RHF—PT with the relativistic Hartree-Fock (RHF) zeroth approximation and PT-RHF(corr)—corrected version with using empirical data; DF-PT with the Dirac-Fock zeroth approximation; RPT-EA—our method (relativistic PT with the Dirac-Kohn-Sham zeroth approximation combined with an energy approach (EA)). All data have been taken from Refs. [[1](#page-9-0)–[6,](#page-9-0) [10,](#page-9-0) [30](#page-10-0), [31](#page-10-0)]).

In Fig. [1](#page-8-0) we present a dependence of the calculated reduced dipole matrix elements upon a principal quantum number for different states of the Rydberg atom Rb:  $5P_{3/2}$ -n $D_{5/2}$  (n ~ 70): available experimental data—the circles; Theory: continuous line—our data; the dotted line- data by Piotrowicz et al., obtained within the quasiclassical Dyachkov-Pankratov model [[4,](#page-9-0) [10](#page-9-0), [12,](#page-9-0) [30](#page-10-0), [31](#page-10-0)]. In Figs. [2](#page-8-0) and [3](#page-9-0) we present the same dependences for the Rydberg states of the Cs atom:  $6P_{3/2} \rightarrow nD_{5/2}$ and the Fr atom:  $7P_{3/2} \rightarrow nD_{5/2}$ , n = 10–70.

The detailed analysis of the computation data shows very important role of the relativistic and interelectron exchange-correlation effects (for example the contribution due to the perturbation theory second and higher orders, including the

<span id="page-8-0"></span>

**Fig. 1** A dependence of the calculated reduced dipole matrix elements upon principal quantum number for Rydberg atom Rb:  $5P_{3/2} \rightarrow nD_{5/2}$  (n ~ 70). The available experimental data are listed as a circle; Theory: continuous line—our data, dotted line- data by Piotrowicz et al. within the quasi-classical Dyachkov-Pankratov model (see text)



**Fig. 2** A dependence of the calculated reduced dipole matrix elements upon principal quantum number for Rydberg atom Cs:  $6P_{3/2}$  → nD<sub>5/2</sub> (n ~ 70). The available experimental data are listed as a circle; Theory: continuous line—our data, dotted line- data by Piotrowicz et al. within the quasi-classical Dyachkov-Pankratov model (see text)

interelectron polarization interaction and mutual screening ones reaches ∼40%), as well as the effect of the non-Coulomb grouping levels in the Rydberg spectra.

To conclude, we have presented the results of studying the radiation decay processes and computing reduced dipole matrix elements (radiative amplitudes) of transitions in spectra of heavy Rydberg atoms of alkali elements (Rb, Cs, Fr; for the states with the principal quantum number  $n = 10-80$  on the basis of the generalized relativistic energy approach and the relativistic many-body perturbation theory with the optimized one-quasiparticle representation. A critically important

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**Fig. 3** A dependence of the calculated reduced dipole matrix elements upon principal quantum number for Rydberg atom Fr:  $7P_{3/2} \rightarrow nD_{5/2}$ , n = 10–80 (our data)

role of the many-body exchange-polarization effects and effect of the non-Coulomb grouping levels in the Rydberg spectra has been found. The detailed numerical data on the dipole matrix elements and transition probabilities are listed in Refs. [\[99](#page-12-0), [100\]](#page-12-0).

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