Ion transition parameters and their present uncertainties: Electronic structure of Li-like Pb ions

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Transition energies in Pb^{79+} ions

The knowledge about $2s\text{-}2p_{1/2}$ (and $2s\text{-}2p_{3/2}$) transition energies is crucial for the Gamma Factory PoP experiment.

A number of calculations have been performed since 1990 (table from Jacek Bieron's contribution to YR).

Table 1: Transition energies of the $2s–2p_{1/2}$ and $2s–2p_{3/2}$ lines in the Pb Li-like ion

$2s–2p_{1/2}$	$2s–2p_{3/2}$	year	method	reference
231.374	2642.297	1990	MCDF VP SE	Indelicato and Desclaux (1990)
230.817	2641.980	1991	MCDF VP SE	Kim *et al.* (1991)
230.698	2641.989	1995	RCI QED NucPol	Chen *et al.* (1995)
231.16	2642.39	1996	3-rd order MBPT	Johnson *et al.* (1996)
230.68	—	2010	RCI QED NucPol	Kozhedub *et al.* (2010)
230.76(4)	2642.17(4)	2011	S-m. 2-l. NucPol	Sapirstein and Cheng (2011)
230.823(47)(4)	2642.220(46)(4)	2018	RCI QED NucPol	Yerokhin and Surzhykov (2018)
230.80(5)	2642.20(5)	2019	S-m. 2-l. NucPol	Sapirstein and Cheng (2019)
—	2642.26(10)	2008	EBIT	Zhang *et al.* (2008)

How shall we understand this table? Can we trust to some calculations more than to others?

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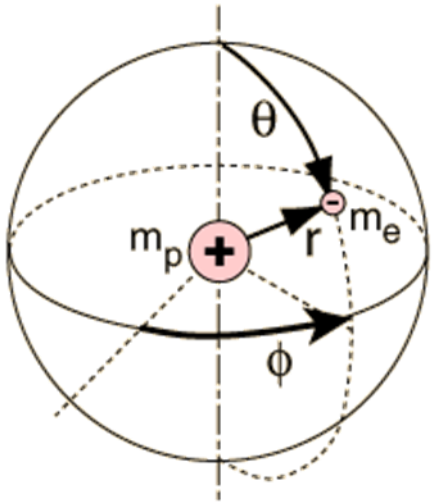
Hydrogen atom: Well-known solutions



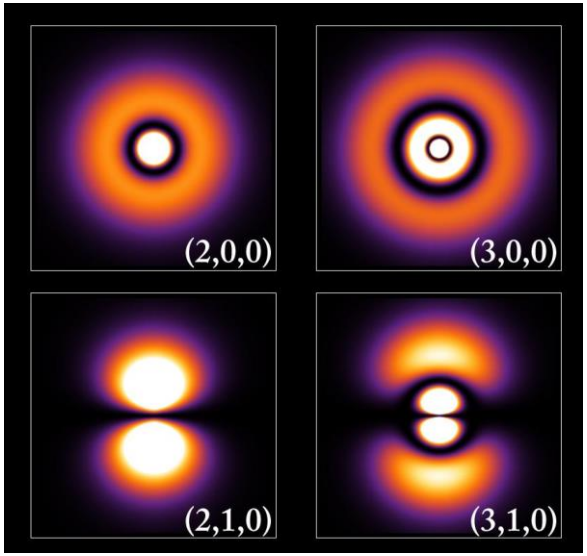
We all have studied in the quantum mechanics course the Schrödinger equation for a one-electron atom:

$$\left(-\frac{\hbar^2}{2m}\boldsymbol{\nabla}^2 - \frac{Ze^2}{r}\right)\psi(\boldsymbol{r}) = E\,\psi(\boldsymbol{r})$$

We know analytical solutions of this equation!

Energy: $E_n = -\mathrm{Ry}\,\dfrac{Z^2}{n^2}$

Wave function: $\psi_{nlm}(\boldsymbol{r}) = R_{nl}(r)Y_{lm}(\theta,\varphi)$



Three quantum numbers describe the system:

n = 1, 2, 3… (principal)
l = 0, … *n*-1 (orbital)
m = -*l*, …. +*l* (magnetic)

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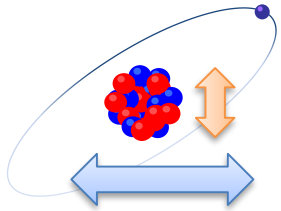
Heavy multiply-charged ions

What is so special about multiply-charged, heavy ions?



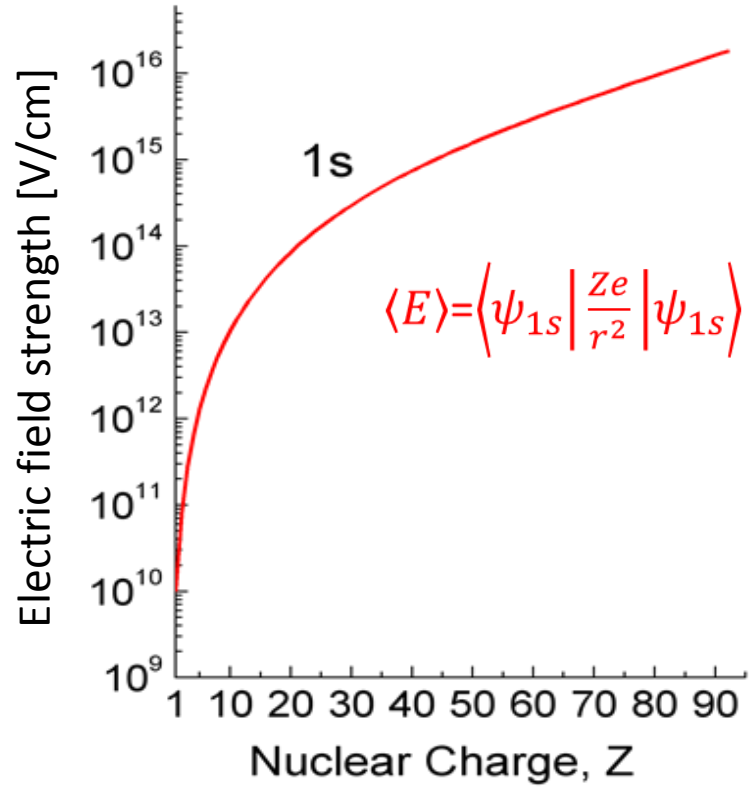
(nuclear size)
R ~ 10^{-14} m

(electron orbit)
r ~ 10^{-13} – 10^{-12} m

One can estimate the electron "velocity" in the ground state:

$$v_{el} \approx \alpha Z c \quad \Rightarrow \quad v_{el} \approx 0.7c$$

(for U^{91+})



Electron is exposed to huge fields (of microscopic) dimensions.

These ions are natural "laboratories" for studying simple atomic systems under critical conditions.

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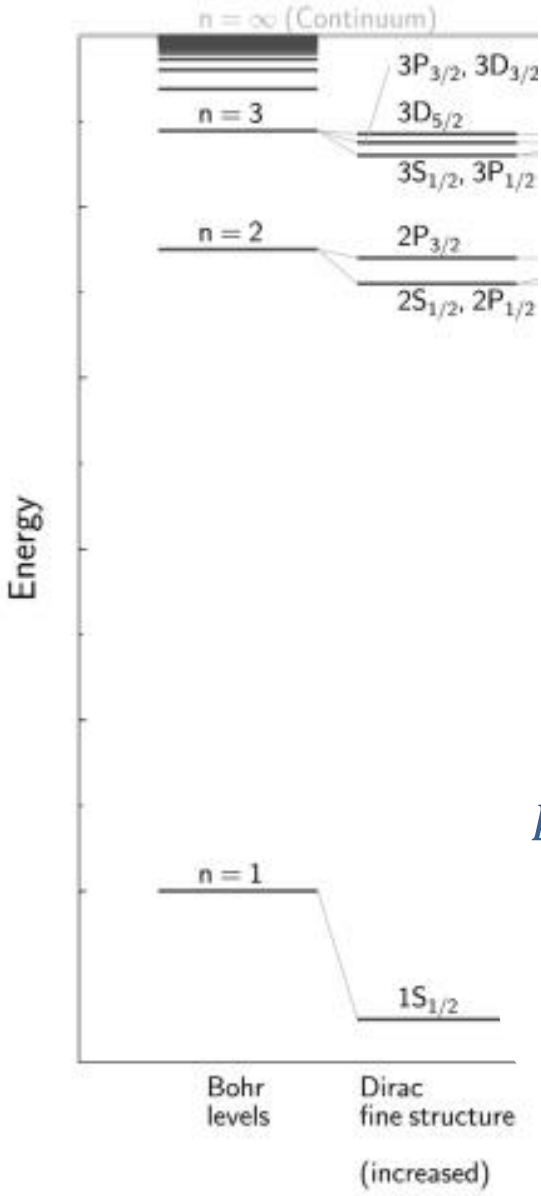
Dirac equation: Hydrogen-like ions



Dirac equation is the relativistic wave equation for a ½-spin electron in the nuclear field:

$$\left(-i\hbar c\boldsymbol{\alpha}\cdot\widehat{\boldsymbol{\nabla}}+V(\boldsymbol{r})+m_e c^2\beta\right)\psi(\boldsymbol{r})=E\,\psi(\boldsymbol{r})$$

Energy solutions of the Dirac equation for point-like, infinitely heavy nucleus:

$$E_{nj}\approx m_e c^2\left(1-\frac{1}{2}\frac{(\alpha Z)^2}{n^2}-\frac{1}{2}\frac{(\alpha Z)^4}{n^3}\left(\frac{1}{j+\frac{1}{2}}-\frac{3}{4n}\right)+\cdots\right)$$

Schrödinger energy

Leading relativistic term

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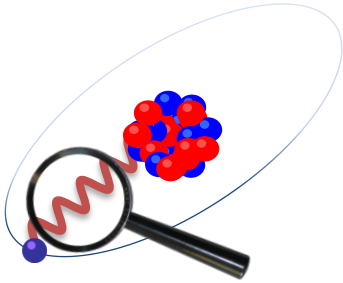
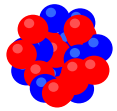
Nuclear effects



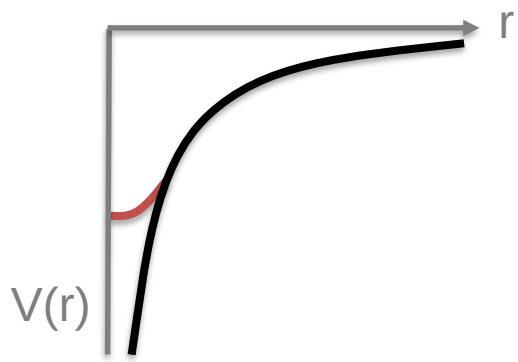
Is the electron-nucleus interaction just an interaction of two point-like charges, one of which (nucleus) is infinitely heavy?

$$V(r) = -\frac{Ze^2}{r}$$

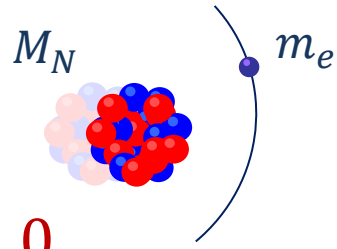
Field shift
due to non-zero charge radii



$\langle r^2 \rangle \neq 0$



Mass shift
due to finite nuclear mass



$M_N \neq 0$

Both corrections lead only to shift (not splitting!) of energy levels.

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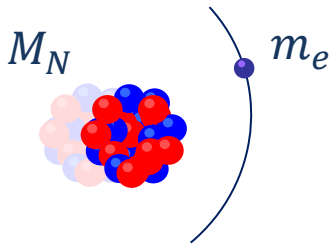
Nuclear recoil correction

Mass shift
due to finite nuclear mass



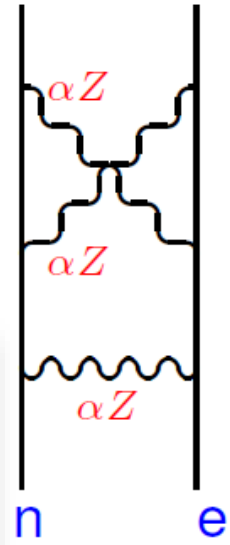
Mass shift has been discussed already in non-relativistic quantum mechanics.

In the non-relativistic approach, for single-electron ions, we have just to introduce the reduced mass:

$$m_e \to \frac{m_e M_N}{m_e + M_N}$$

Accurate relativistic treatment is possible only within the framework of quantum electrodynamics:

$$\begin{aligned}
\Delta E &= \Delta E_{\rm L} + \Delta E_{\rm H} \\
\Delta E_{\rm L} &= \frac{1}{2M}\langle a|[\vec{p}^{\,2} - (\vec{D}(0)\cdot\vec{p} + \vec{p}\cdot\vec{D}(0))]|a\rangle\,, \\
\Delta E_{\rm H} &= \frac{i}{2\pi M}\int_{-\infty}^{\infty} d\omega\, \langle a|\Big(\vec{D}(\omega) - \frac{[\vec{p}, V_{\rm C}]}{\omega + i0}\Big) G(\omega + E_a)\Big(\vec{D}(\omega) + \frac{[\vec{p}, V_{\rm C}]}{\omega + i0}\Big)|a\rangle
\end{aligned}$$



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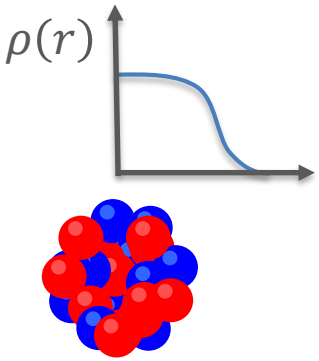
Finite nuclear size effect



The potential induced by the nuclear charge distribution $\rho(r)$ is defined as:

$$V_N(\boldsymbol{r}) = 4\pi\alpha Z \int_0^\infty dr' r'^2 \rho(r') \frac{1}{r_>} \qquad r_> = \mathrm{Max}\{r, r'\}$$

And can be plugged in the Dirac equation

$$\left(-i\hbar c\boldsymbol{\alpha}\cdot\hat{\boldsymbol{\nabla}} + V(\boldsymbol{r}) + m_e c^2\beta\right)\psi(\boldsymbol{r}) = E\,\psi(\boldsymbol{r})$$

Solution of the Dirac equation is most conveniently written in the bi-spinor form:

$$\psi_{nj\mu_j}(r) = \frac{1}{r}\begin{pmatrix} g_{nj}(r)\,\Omega_{lj\mu_j}(\hat{\boldsymbol{r}}) \\ i\,f_{nj}(r)\,\Omega_{l'j\mu_j}(\hat{\boldsymbol{r}}) \end{pmatrix}$$

We can re-write Dirac equation for the radial components!

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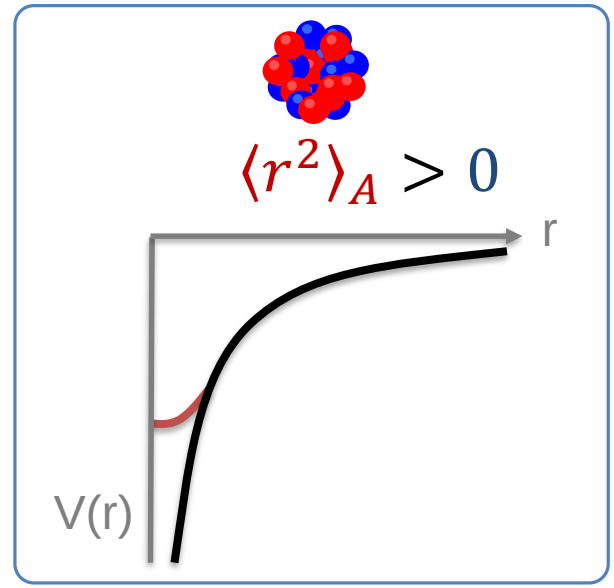
Finite nuclear size: Relativistic treatment

We need to choose proper charge distribution and plug it in Dirac equation:

$$\left(\frac{df_{nj}(r)}{dr} - \frac{\kappa}{r} f_{nj}(r)\right) = -(E - V(r) - m_e c^2) g_{nj}(r)$$

$$\left(\frac{dg_{nj}(r)}{dr} + \frac{\kappa}{r} g_{nj}(r)\right) = (E - V(r) + m_e c^2) f_{nj}(r)$$



One can find the relativistic correction (Shabaev 1993)

$$\Delta E \approx \frac{(\alpha Z)^2}{10\, n} \left[1 + (\alpha Z)^2\, \phi_{nj}(\alpha Z)\right] \left(2\, \frac{\alpha Z}{n}\, \frac{R}{\hbar/mc}\right)^{2\gamma} m_e c^2$$

Nuclear radii are „input data“ fo us! Uncertainty in nuclear data leads to uncertainty of our calculations.

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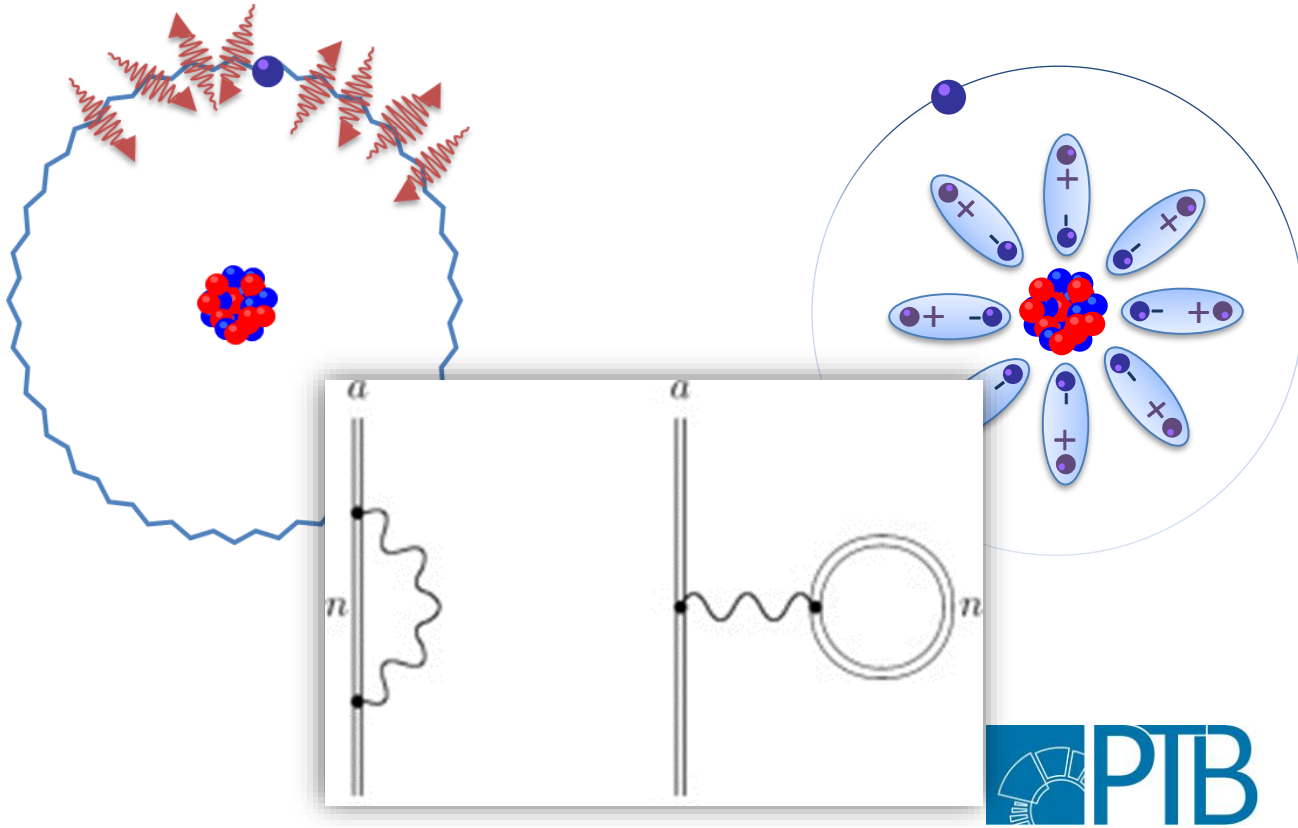
QED effects

Self-energy

Electron can also emit and absorb a virtual photon. We can see this process as interaction of electron with ist own electromagnetic field.

Vacuum polarization

We can for a short time to “borrow” energy from vacuum and to create electron-positron pairs.



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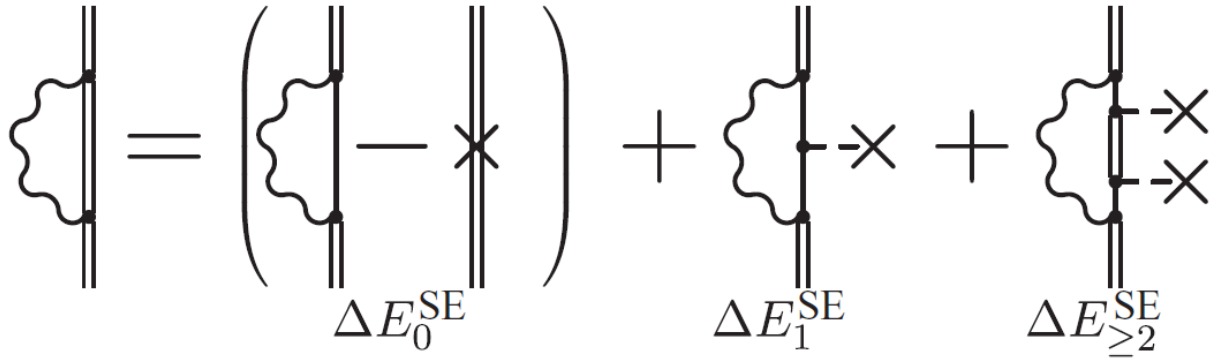
One-loop self-energy correction



The zero- and first-order terms are both divergent. These two terms should be renormalized together with the mass-counter term and are calculated within the momentum representation.

The remaining higher-order term is convergent and usually is calculated in the coordinate space by using a partial-wave decomposition for the electron propagators.

K.T. Cheng, W.R. Johnson, and J. Sapirstein, PRA, 1993
V.A. Yerokhin and V.M. Shabaev, PRA, 1999
A. Artemyev *et al.*, PRA, 2013

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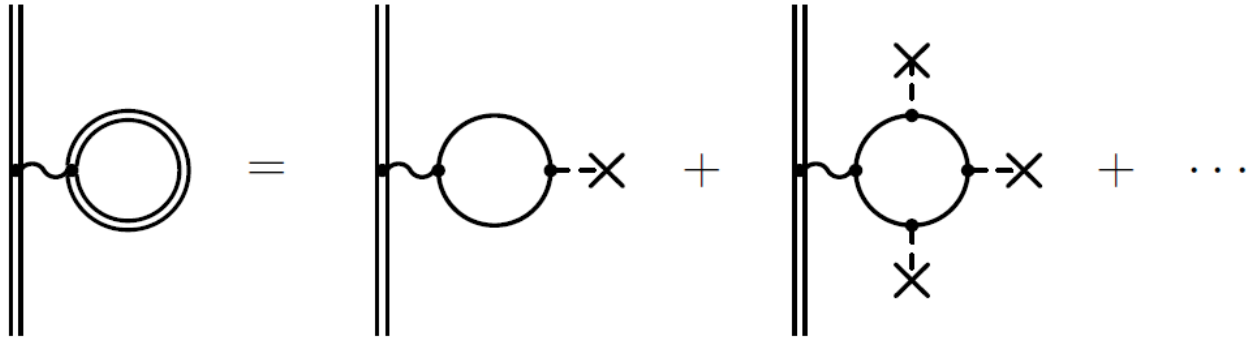
One-loop vacuum-polarization correction

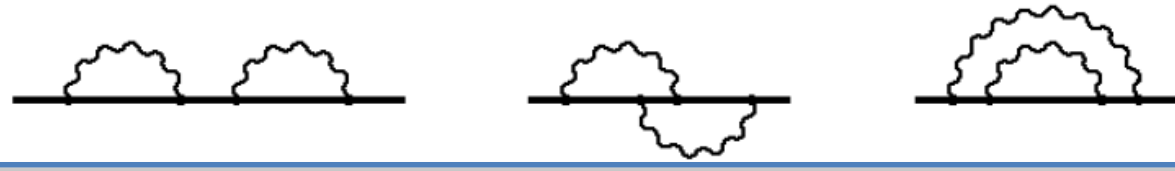


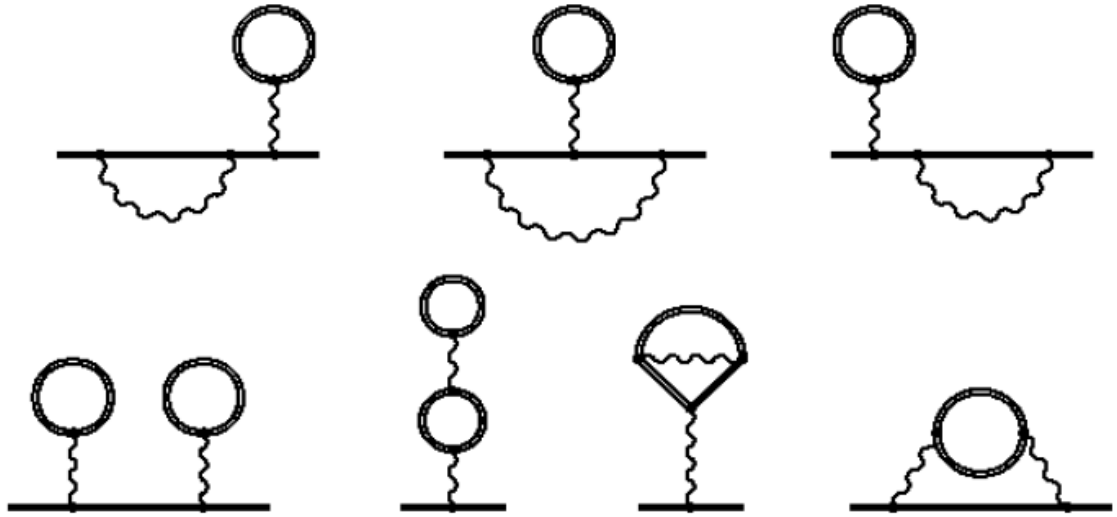
The one-loop vacuum polarization diagram can be computed in the traditional approach by a decomposition of the vacuum loop electron propagator in powers of the external potential. In this expansion, the first (also called the Uehling) term contains either *one* interaction with the full effective potential:

$$U_{\rm Uehl}(r) = -\alpha Z \frac{2\alpha}{3\pi} \int_0^\infty dr'\, 4\pi r' \rho(r') \int_1^\infty dt\, \left(1+\frac{1}{2t^2}\right) \frac{\sqrt{t^2-1}}{t^2} \times \frac{\left[\exp\left(-2m|r-r'|t\right) - \exp\left(-2m(r+r')t\right)\right]}{4mrt}$$

Two-loop QED corrections

Two-loop self-energy yields the dominant theoretical uncertainty for the Lamb shift in hydrogen and light hydrogen-like ions.





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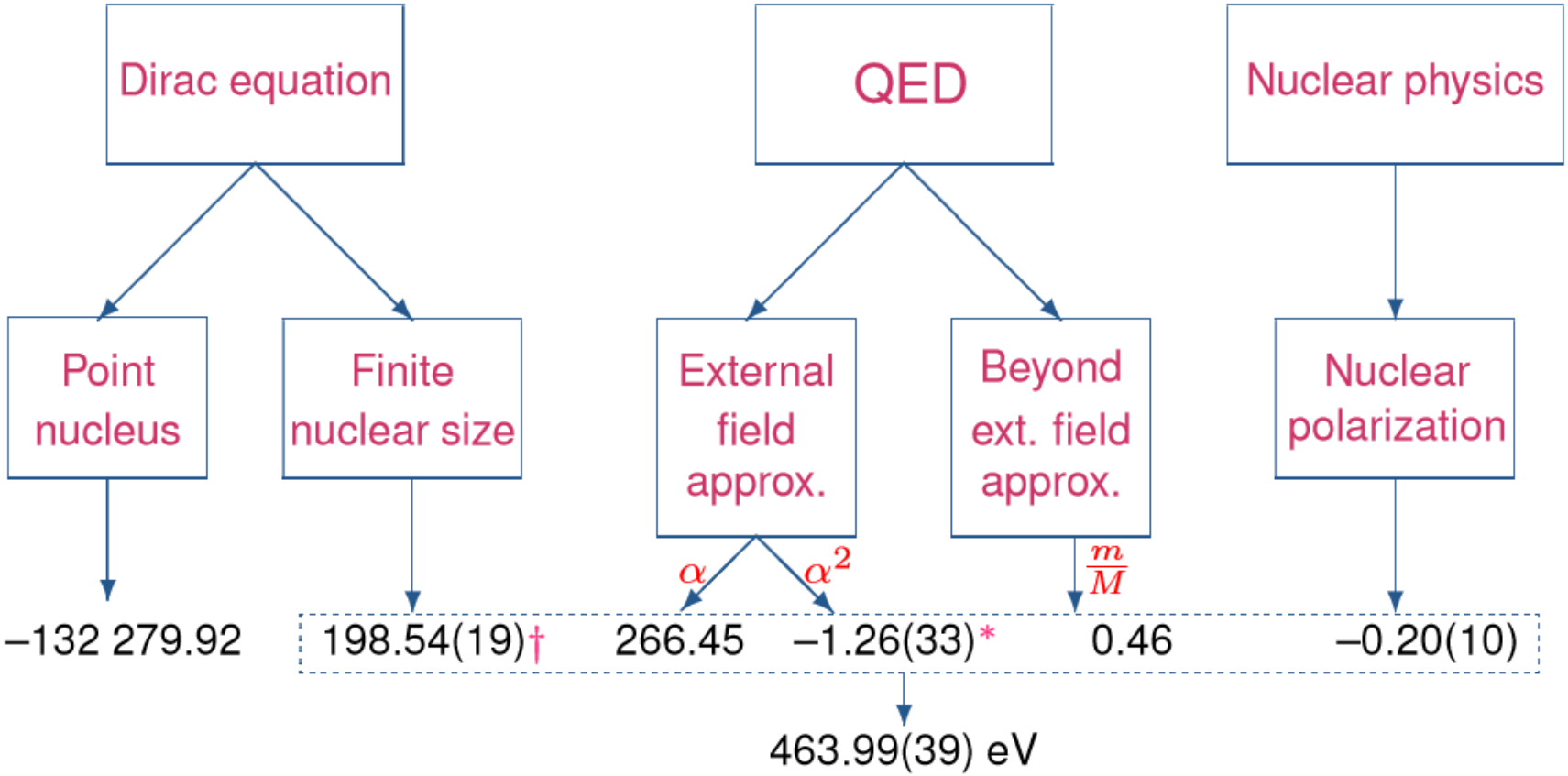
Uncertainty budget: Lamb shift

Example: 1s Lambshift in hydrogen-like uranium ,in eV



Picture from Vladimir Shabaev

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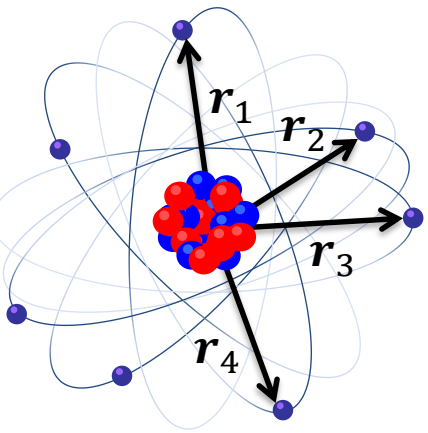
Many-electron ions: Theory



In quantum theory, states of an atom are described by their energy values and by wave-functions:

$$E_n\ ,\ \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N)$$

The wave function is a function of 3N coordinates, where N is the number of electrons! How to deal with this huge dimension?

For heavy ions like Pb the starting point is the Dirac equation:

$$\hat{H} = \sum_i \left(-i\hbar c\boldsymbol{\alpha}_i \cdot \hat{\nabla}_i + V(\boldsymbol{r}_i) + m_e c^2 \gamma^0\right) + \sum_{i<j} V(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$$

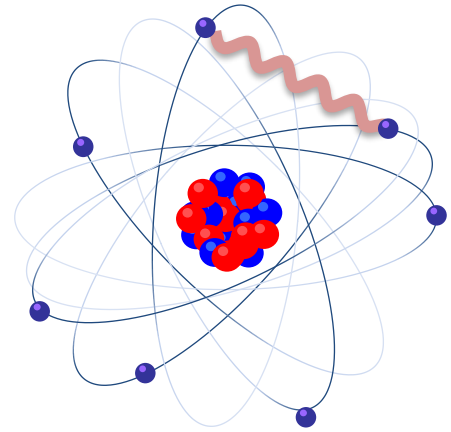
Hamiltonians of individual electrons

e-e interactions

How to describe e-e interactions?

Relativistic corrections to e-e interaction

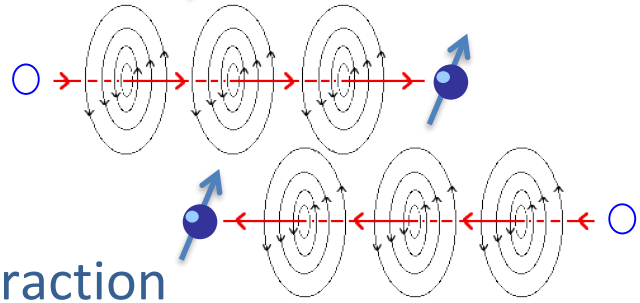
How do electrons interact with each other? Is it just Coulomb interaction of two static charges?



The relativistic electron-electron interaction is not anymore just Coulomb-type interaction $1/r_{12}$. It contains magnetic and retardation terms.

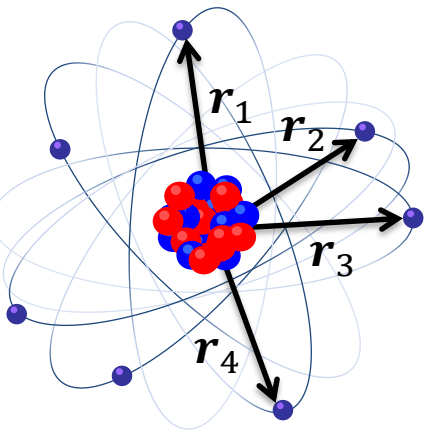
$$V_{ee} = V^C + V^B$$

$$= \frac{1}{r_{12}} + \left(-\boldsymbol{\alpha}_1 \cdot \boldsymbol{\alpha}_2 \frac{\cos \omega r_{12}}{r_{12}} + (\boldsymbol{\alpha}_1 \cdot \boldsymbol{\nabla}_1)(\boldsymbol{\alpha}_2 \cdot \boldsymbol{\nabla}_2) \frac{\cos \omega r_{12} - 1}{\omega^2 r_{12}} \right)$$



Breit interaction:
Retardation + magnetic interaction

Theory of many-electron systems



The Hamiltonian of many-electron atoms reads as:

$$\hat{H} = \sum_i \left(-i\hbar c\boldsymbol{\alpha}_i \cdot \hat{\nabla}_i + V(\boldsymbol{r}_i) + m_e c^2 \gamma^0\right) + \sum_{i<j} V(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$$

Hamiltonians of individual electrons

e-e interactions

We can easily construct the many-electron wavefunctions and energies if we neglect the electron-electron interaction term:

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N) = \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \cdots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \cdots & \varphi_N(r_N) \end{vmatrix}$$

Where $\varphi_i(r_i)$ are solutions of individual Hamiltonians:

$$\left(-i\hbar c\boldsymbol{\alpha}_i \cdot \hat{\nabla}_i + V(\boldsymbol{r}_i) + m_e c^2 \gamma^0\right) \varphi_i(r_i) = \varepsilon_i \varphi_i(r_i)$$

Which potential to use here?

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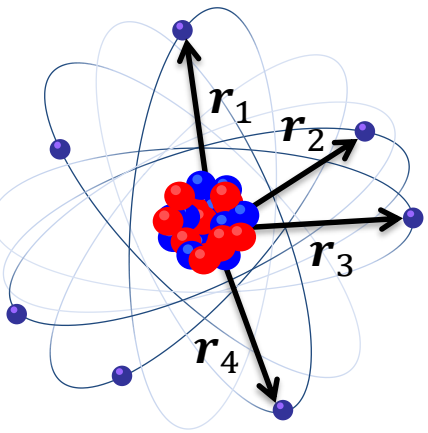
Theory of many-electron systems



We can construct the many-electron wavefunctions as:

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \dots, \boldsymbol{r}_N) = \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \dots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \dots & \varphi_N(r_N) \end{vmatrix}$$

Where $\varphi_i(r_i)$ are solutions of individual Hamiltonians:

$$\left(-i\hbar c\boldsymbol{\alpha}_i \cdot \hat{\nabla}_i + V_{eff}(\boldsymbol{r}_i) + m_e c^2 \gamma^0\right) \varphi_i(r_i) = \varepsilon_i \varphi_i(r_i)$$

Just Coulomb electron-nucleus interaction?

$$V_{eff}(\boldsymbol{r}_i) = -\frac{Ze^2}{r}$$

Too rough! We need to account for e-e interactions

It is usually convenient to include a part of the e-e interaction already in the one-electron orbitals by introducing a screening potential

$$V_{\mathrm{DF}}(\boldsymbol{r}_1)\,\psi(\boldsymbol{r}_1) = \sum_c \int d\boldsymbol{r}_2\, \psi_c^+(\boldsymbol{r}_2)\, \frac{\alpha}{r_{12}} \times [\psi_c(\boldsymbol{r}_2)\,\psi(\boldsymbol{r}_1) - \psi_c(\boldsymbol{r}_1)\,\psi(\boldsymbol{r}_2)]$$

But… We need to account for the rest of e-e interactions!

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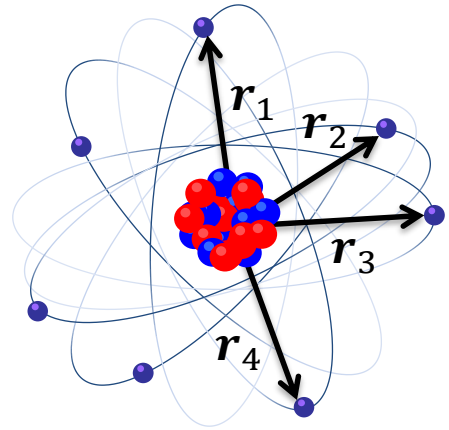
Configuration interaction method

We can use CSF as basis functions to construct more accurate wavefunction

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N) = \sum_r c_r \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \ldots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \ldots & \varphi_N(r_N) \end{vmatrix}$$

Summation over configurations

State of particular symmetry $|\gamma_r J^P M\rangle$



In the CI method, the energy levels of the system and the mixing coefficients c_r are obtained by solving the secular equation

$$det\left\{\left\langle \gamma_r J^P M \middle| \sum_i \hat{h}_i + \sum_{i<j} V(|\boldsymbol{r}_i - \boldsymbol{r}_j|) \middle| \gamma_s J^P M \right\rangle - E_r \delta_{rs}\right\} = 0$$

In present calculations the $V(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$ usually accounts for (frequency-dependent) Breit interaction corrections.

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Configuration interaction method

TABLE I. Contributions to the Coulomb energy (in a.u.) for the $1s2s2p\,^4P^o_{1/2}$ state of lithium-like iron ($Z = 26$), for the infinitely heavy nucleus. The values listed after the first row are the increments obtained on successively adding configurations while increasing the maximal value of the orbital quantum number $L_{\max}$ and enlarging the size of the one-electron basis. Label SD denotes single and double excitations, T denotes the triple excitations, n_a is the number of B splines in the one-electron basis set, and ϵ is the energy cutoff parameter; see text for the details.

$L_{\max}$	δ E
SD, $n_a = 30$, $\epsilon = 4$	
1	−497.762 342 48
2	−0.008 719 04
3	−0.000 662 72
4	−0.000 124 13
5	−0.000 034 81
6	−0.000 012 45
7	−0.000 005 18
$\geqslant 8$	−0.000 005 55
SD, $n_a = 40$, $\epsilon = 8$	
1	−0.000 173 23
2	−0.000 001 34
3	−0.000 001 13
$\geqslant 4$	−0.000 001 07
SD, $n_a = 50$, $\epsilon = 16$	
1	−0.000 006 46
2	−0.000 000 43
T, $n_a = 25$, $\epsilon = 4$	
1	−0.000 000 22
2	−0.000 000 77
3	−0.000 000 09
$\geqslant 4$	−0.000 000 04
Final result	**−497.772 091(7)**

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N)$$

$$= \sum_r c_r \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \cdots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \cdots & \varphi_N(r_N) \end{vmatrix}$$

The summation over configurations can not be infinite. The finite size of the basis is one of the sources of theoretical uncertainty.

One can control the convergence by enhancing the basis in a clear way.

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Transition energies in Pb^{79+} ions

Z	Present	Sapirstein and Cheng[5]	Kozhedub *et al.* [4]	Yerokhin *et al.* [3]
$2p_{3/2} - 2s$:				
18	35.03759 (73)	35.037 (1)	35.0378 (6)	
20	41.02497 (78)	41.024 (1)	41.0251 (7)	
21	44.30923 (84)	44.308 (1)	44.3092 (7)	
26	64.5646 (12)	64.562 (1)	64.5650 (9)	64.5647 (26)
28	74.9586 (14)	74.955 (1)	74.9586 (11)	74.9585 (28)
30	87.0274 (15)	87.023 (1)	87.0282 (12)	
32	101.0489 (17)	101.04		
36	136.1808 (22)	136.17	136.1818 (17)	
40	183.1095 (27)	183.10	183.1106 (23)	
42	211.9871 (29)(1)	211.98		
47	303.6714 (38)(2)	303.67	303.6709 (36)	303.6704 (52)
50	374.6774 (46)(2)	374.67	374.6757 (46)	
52	429.9055 (50)(3)	429.88	429.904 (5)	
54	492.2258 (58)(8)	492.21	492.225 (6)	
60	729.2038 (84)(8)	729.19 (1)	729.204 (6)	
64	937.376 (11)(2)	937.39 (1)		
66	1059.503 (13)(11)	1059.56 (1)		
70	1345.526 (17)(6)	1345.57 (2)		
74	1696.129 (24)(3)	1696.10 (3)		
79	2244.036 (36)(10)	2244.00 (3)		
80	2370.446 (39)(9)	2370.38 (4)		
82	2642.220 (46)(4)	2642.17 (4)		
83	2788.127 (52)(10)	2788.04 (5)		
90	4025.410 (96)(96)	4025.25 (7)		
92	4459.580 (94)(31)	4459.46 (8)		

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Transition energies in Pb^{79+} ions

The knowledge about 2s-$2p_{1/2}$ (and 2s-$2p_{3/2}$) transition energies is crucial for the Gamma Factory PoP experiment.

A number of calculations have been performed since 1990 (table from Jacek Bieron's contribution to YR).

Table 1: Transition energies of the 2s–$2p_{1/2}$ and 2s–$2p_{3/2}$ lines in the Pb Li-like ion

2s–$2p_{1/2}$	2s–$2p_{3/2}$	year	method	reference
231.374	2642.297	1990	MCDF VP SE	Indelicato and Desclaux (1990)
230.817	2641.980	1991	MCDF VP SE	Kim *et al.* (1991)
230.698	2641.989	1995	RCI QED NucPol	Chen *et al.* (1995)
231.16	2642.39	1996	3-rd order MBPT	Johnson *et al.* (1996)
230.68	—	2010	RCI QED NucPol	Kozhedub *et al.* (2010)
230.76(4)	2642.17(4)	2011	S-m. 2-l. NucPol	Sapirstein and Cheng (2011)
230.823(47)(4)	2642.220(46)(4)	2018	RCI QED NucPol	Yerokhin and Surzhykov (2018)
230.80(5)	2642.20(5)	2019	S-m. 2-l. NucPol	Sapirstein and Cheng (2019)
—	2642.26(10)	2008	EBIT	Zhang *et al.* (2008)

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Transition probabilities in Pb^{79+} ions

	$2p_{1/2} \rightarrow 2s$		$2p_{3/2} \rightarrow 2s$	
Z	E(a.u.)	A(s^{-1})	E(a.u.)	A(s^{-1})
52	4.1825	4.928[9]	15.799	2.852[11]
53	4.2934	5.104[9]	16.911	3.354[11]
54	4.4055	5.275[9]	18.090	3.941[11]
55	4.5200	5.460[9]	19.341	4.629[11]
56	4.6361	5.640[9]	20.667	5.432[11]
57	4.7543	5.834[9]	22.072	6.361[11]
58	4.8745	6.026[9]	23.560	7.444[11]
59	4.9968	6.233[9]	25.134	8.708[11]
60	5.1212	6.438[9]	26.799	1.018[12]
61	5.2476	6.660[9]	28.559	1.186[12]
62	5.3758	6.879[9]	30.417	1.383[12]
63	5.5072	7.123[9]	32.380	1.610[12]
64	5.6404	7.340[9]	34.451	1.873[12]
65	5.7766	7.606[9]	36.636	2.176[12]
66	5.9159	7.850[9]	38.941	2.522[12]
67	6.0544	8.107[9]	41.366	2.922[12]
68	6.2008	8.384[9]	43.926	3.388[12]
69	6.3458	8.644[9]	46.618	3.920[12]
70	6.4941	8.937[9]	49.452	4.526[12]
71	6.6461	9.236[9]	52.435	5.216[12]
72	6.7996	9.529[9]	55.571	6.015[12]
73	6.9573	9.865[9]	58.870	6.940[12]
74	7.1155	1.015[10]	62.335	7.972[12]
75	7.2797	1.050[10]	65.979	9.167[12]
76	7.4451	1.083[10]	69.806	1.052[13]
77	7.6132	1.121[10]	73.824	1.209[13]
78	7.7845	1.154[10]	78.043	1.383[13]
79	7.9586	1.193[10]	82.471	1.583[13]
80	8.1335	1.225[10]	87.116	1.815[13]
81	8.3137	1.263[10]	91.993	2.072[13]
82	8.4950	1.306[10]	97.106	2.368[13]

$$A(2p_{1/2} \rightarrow 2s) = 1.306 \times 10^{10}\ s^{-1}$$

$$A(2p_{3/2} \rightarrow 2s) = 2.368 \times 10^{13}\ s^{-1}$$

W. R. Johnson, K. T. Cheng, and J. Sapirstein, At. Data Nucl. Data Tables **64** (1996) 279.

$$A_{\mathrm{v}}(2p_{1/2} \rightarrow 2s) = 1.50 \times 10^{10}\ s^{-1}$$

$$A_{\mathrm{l}}(2p_{1/2} \rightarrow 2s) = 1.37 \times 10^{10}\ s^{-1}$$

$$A_{\mathrm{v}}(2p_{3/2} \rightarrow 2s) = 2.46 \times 10^{13}\ s^{-1}$$

$$A_{\mathrm{l}}(2p_{3/2} \rightarrow 2s) = 2.45 \times 10^{13}\ s^{-1}$$

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