Special Topics on Precision Measurement in Atomic Physics: Lecture 12

Two-photon decay and the Tune-out Wavelength

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Sponsored by USTC, Organized by WIPM

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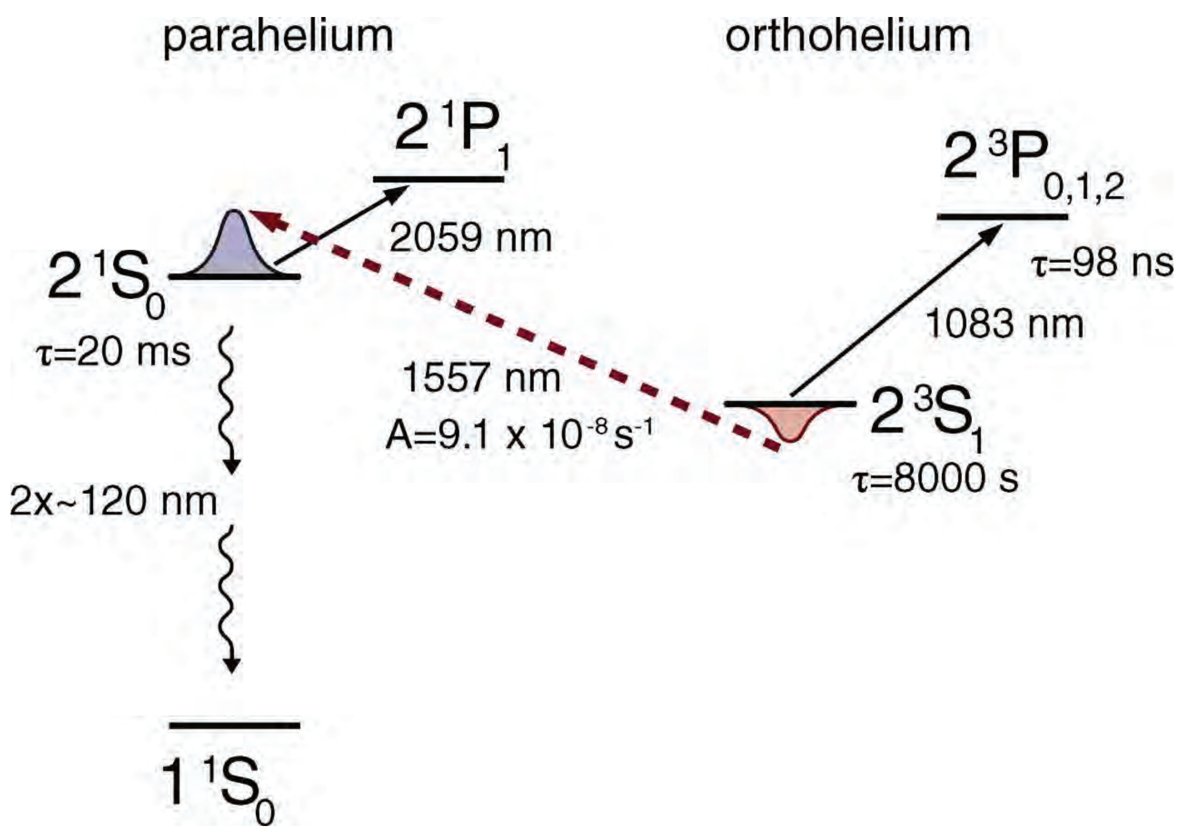
1 Two Photon Processes



R. van Rooij, J. S. Borbely, J. Simonet, M. D. Hoogerland, K. S. E. Eikema, R. A. Rozendaal, W. Vassen Science 333, 6039 (2011)

SELECTION RULES:

- Electric dipole (E1), In LS coupling:
  - $\Delta L = 0$ or ± 1, but $L = 0 \not\to L = 0$
  - $\Delta S = 0$
  - $\Delta J = 0$ or ± 1, but $J = 0 \not\to J = 0$
  - Parity $\mathcal{P}$ is odd
- Magnetic dipole (M1):
  - $\Delta L = 0$ or ± 1,
  - $\Delta S = 0$ or ± 1
  - $\Delta J = 0$ or ± 1, but $J = 0 \not\to J = 0$
  - Parity $\mathcal{P}$ is even

ORDERS OF MAGNITUDE

For the Einstein A-coefficient (per unit time)

- E1: $A \sim (\omega/c)^3 \langle r \rangle^2 \sim \alpha^3 Z^6 Z^{-2} = \alpha^3 Z^4$ if $\Delta n \neq 0$ $\quad(\sim 10^9\ \mathrm{s}^{-1})$
  $= \alpha^3 Z$ if $\Delta n = 0$
- M1: $A \sim (\mathrm{E1}) \times \alpha^2 Z = \alpha^5 Z^2$ if $\Delta n = 0$
- Relativistic M1: $A \sim (\mathrm{E1}) \times (\alpha^2 Z^2)(\alpha^2 Z^2)^2 = \alpha^9 Z^{10}$ $\quad(\sim 10^{-4}\ \mathrm{s}^{-1}$ for He$)$
- M2: $A \sim (\mathrm{E1}) \times (\alpha^2 Z^2)^2 = \alpha^7 Z^8$ $\quad$ (Exceeds E1 at $Z \sim 18$ if $\Delta n = 0$)
- spin-forbidden E1: $A \sim (\mathrm{E1}) \times \left(\frac{\alpha^2 Z^4}{Z}\right)^2 = \alpha^7 Z^{10}$
- 2E1: $A_{2\gamma} \sim \alpha^6 Z^6$

TWO-PHOTON TRANSITIONS

- First proposed by Maria Goeppert-Mayer M (1931) in her Ph.D. thesis and published in "Uber Elementarakte mit zwei Quantensprüngen". Ann. Phys. Lpz. **9** 27395 (1931).

- First approximate calculations for hydrogen $2s$ and helium $2\ 1S$ by Gregory Breit and Edward Teller in Astrophys. J. **91**, 215 (1940). (However, their result for helium $2\ {}^3S_1$ was incorrect and greatly over-estimated.)

- First accurate calculations for hydrogen $2s$ by Spitzer and Greenstein (1951), Shapiro and Breit (1959) ,Zon and Rapoport (1968), Klarsfeld (1968), and Drake (1969).

- First accurate calculations for He $2\ {}^1S$ by Drake et al. (1969) and including relativistic effects (1986).

- First correct calculation for He $2\ {}^3S_1 - 1\ {}^1S_0$ by Bely (1968) and independently by Drake (1969) (strongly suppressed since it is proportional to $|\hat{\boldsymbol{e}}_1 \times \hat{\boldsymbol{e}}_2|^2$ instead of $|\hat{\boldsymbol{e}}_1 \cdot \hat{\boldsymbol{e}}_2|^2$ and vanishes when $\omega_1 = \omega_2$ since it is a $J = 1 \to 0$ transition).

- Recent work on "Two-photon energy distribution from the decay of the $2\ {}^1S_0$ state in He-like uranium" by D. Banas et al., Phys. Rev. A **87**, 062510 (2013), and

  "Angular and polarization analysis for two-photon decay of 2s hyperfine states of hydrogenlike uranium" by L. Safari et al., Phys. Rev. A **90**, 014502 (2014).

- For a review, see P. H. Mokler and R. W. Dunford, Phys. Scr. **69** C1 (2004), and
  "QED theory of multiphoton transitions in atoms and ions," T.A. Zaliali-utdinov, A. Timur, D.A. Solovyev, L.N. Labzowsky, and G. Plunien, Phys. Rep. **737**, 1 (2018).

- "Two-photon decay rates of hydrogenlike ions revisited by using Dirac-Coulomb Sturmian expansions of the first order," Z. Bona,H.M.T. Nganso, T.B. Ekogo,M.G.K. Njock,and M.G. Kwato, Phys. Rev. A **89**, 022514 (2014).

PHYSICAL REVIEW VOLUME 180, NUMBER 1 5 APRIL 1969

Two-Photon Decay of the Singlet and Triplet Metastable States of Helium-like Ions

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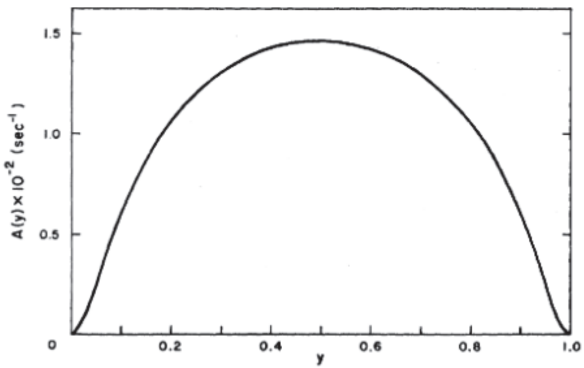

FIG. 1. Photon energy distribution for the $2\,^1S$-$1\,^1S$ two-photon decay of He I; y is the fraction of the energy transported by one of the two photons and $A=51.3$ sec^{-1}.

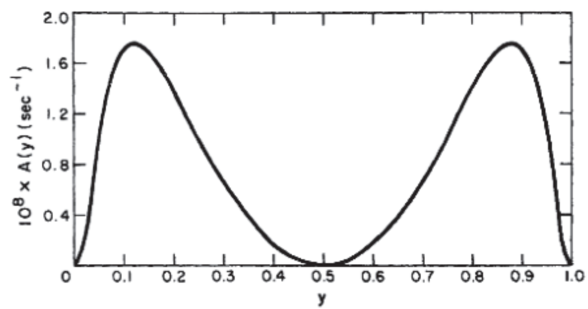

FIG. 5. Photon energy distribution for the $2\,^3S$-$1\,^1S$ two-photon decay of He I; y is the fraction of the energy transported by one of the two photons and $A=4.02\times10^{-9}$ sec^{-1}.

Comparison with Experiment

H-like $2\,^2S_{1/2} - 1\,^2S_{1/2}$**:**

$Z = 1$: H. Krüger and A. Oed, Phys. Lett. **54A**, 251 (1975).
$Z = 2$: E.A. Hinds, J.E. Clendenin, and R. Novick, Phys. Rev. A **17**, 670 (1978).
$Z = 8$: C.L. Cocke, B. Curnutte, J.R. MacDonald, J.A. Bednar, and R.Marrus, Phys. Rev. A **9**, 2242 (1974).
$Z = 9$: *ibid.*
$Z = 16$: R. Marrus and R.W. Schmieder, Phys. Rev. A **5**, 1160 (1972).
$Z = 18$: H. Gould and R. Marrus, Phys. Rev. A **28**, 2001 (1983).
$Z = 28$: R.W. Dunford, M. Hass, E. Bakke, H.G. Berry, C.J. Liu, M.L.A. Raphaelian, and L.J. Curtis, Phys. Rev. Lett. **62**, 2809 (1989).
$Z = 36$: S. Cheng, H.G. Berry, R.W. Dunford, D.S. Gemmell, E.P. Kanter, B.J. Zabransky, A.E. Livingston, L.J. Curtis, J. Bailey, and J.A. Nolen, Jr., Phys. Rev. A **47**, 903 (1993).

He-like $2\,^1S_0 - 1\,^1S_0$**:**

$Z = 2$: R.S. Van Dyck, Jr.,C.E. Johnson, and H.A. Shugart, Phys. Rev. A **4**, 1327 (1971).

$Z = 3$: M.H. Prior and H.A. Shugart, Phys. Rev. Lett. **27**, 902 (1971).

$Z = 18$: H. Gould and R. Marrus, Phys. Rev. A **28**, 2001 (1983).

$Z = 28$: R.W. Dunford, H.G. Berry, K.O. Groeneveld, M.Hass, E.Bakke, M.L.A. Raphaelian, A.E. Livingston, and L.J. Curtis, Phys. Rev. A **38**, 5423 (1988).

$Z = 35$: R.W. Dunford, H.G. Berry, S. Cheng, E.P. Kanter, C. Kurtz, B.J. Zabransky, A.E. Livingston, and L.J. Curtis, Phys. Rev. A **48**, 1929 (1993).

$Z = 36$: R. Marrus, V.S. Vicente, P. Charles, J.P. Briand, F.Bosch, D. Liesen, and I. Varga, Phys. Rev. Lett. **56**, 1683 (1986).

PHYSICAL REVIEW A VOLUME 48, NUMBER 4 OCTOBER 1993

Forbidden transitions in one- and two-electron nickel

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Includes and extensive discussion and survey of earlier work for Z = 1 to 36.

TABLE V. Experimental and theoretical lifetimes and branching ratios for the $2\,^2S_{1/2}$ level in H-like ions.

Z	Experiment (s)	Theory[a] (s)	nr Theory[a] (s)	$M1$ branching ratio (Theory)[a]
1	0.67 ± 0.29[b]	0.121 58	0.121 59	3.033×10^{-7}
2	$(1.922\pm0.082)\times10^{-3}$ [c]	$1.898\,2\times10^{-3}$	$1.897\,9\times10^{-3}$	4.853×10^{-6}
	$(2.04+0.81-0.34)\times10^{-3}$ [d]			
	$(1.905\pm0.018)\times10^{-3}$ [e]			
8	$(453\pm43)\times10^{-9}$ [f]	463.55×10^{-9}	462.55×10^{-9}	1.247×10^{-3}
9	$(237\pm17)\times10^{-9}$ [f]	228.62×10^{-9}	228.00×10^{-9}	1.996×10^{-3}
16	$(7.3\pm0.7)\times10^{-9}$ [g]	$7.153\,0\times10^{-9}$	$7.094\,7\times10^{-9}$	0.019 93
18	$(3.54\pm0.25)\times10^{-9}$ [g]	$3.494\,1\times10^{-9}$	$3.459\,4\times10^{-9}$	0.031 73
	$(3.487\pm0.036)\times10^{-9}$ [h]			
28	$(217.1\pm1.8)\times10^{-12}$ [i]	215.45×10^{-12}	212.34×10^{-12}	0.166 7
36	$(36.8\pm1.4)\times10^{-12}$ [j]	37.008×10^{-12}	26.982×10^{-12}	0.364 3

$Z=1$: H. Krüger and A. Oed, Phys. Lett. **54A**, 251 (1975).
$Z=2$: E.A. Hinds, J.E. Clendenin, and R. Novick, Phys. Rev. A **17**, 670 (1978).
$Z=8$: C.L. Cocke, B. Curnutte, J.R. MacDonald, J.A. Bednar, and R.Marrus, Phys. Rev. A **9**, 2242 (1974).
$Z=9$: *ibid.*
$Z=16$: R. Marrus and R.W. Schmieder, Phys. Rev. A **5**, 1160 (1972).
$Z=18$: H. Gould and R. Marrus, Phys. Rev. A **28**, 2001 (1983).
$Z=28$: R.W. Dunford, M. Hass, E. Bakke, H.G. Berry, C.J. Liu, M.L.A. Raphaelian, and L.J. Curtis, Phys. Rev. Lett. **62**, 2809 (1989).
$Z=36$: S. Cheng, H.G. Berry, R.W. Dunford, D.S. Gemmell, E.P. Kanter, B.J. Zabransky, A.E. Livingston, L.J. Curtis, J. Bailey, and J.A. Nolen, Jr., Phys. Rev. A **47**, 903 (1993).

TABLE VI. Experimental and theoretical results for the lifetime of the $2\,^1S_0$ level He-like atoms.

Z	Experiment (s)	Theory[a] (s)	nr Theory[a] (s)
2	$(38\pm8)\times10^{-3}$ [b]	$(19.630\pm0.028)\times10^{-3}$	19.600×10^{-3}
	$(19.7\pm1.0)\times10^{-3}$ [c]		
3	$(503\pm26)\times10^{-6}$ [d]	$(515.85\pm0.47)\times10^{-3}$	515.27×10^{-3}
18	$(2.3\pm0.3)\times10^{-9}$ [e]	$(2.3725\pm0.0056)\times10^{-9}$	2.3432×10^{-9}
	$(2.32\pm0.20)\times10^{-9}$ [f]		
28	$(150\pm16)\times10^{-12}$ [g]	$(154.28\pm0.50)\times10^{-12}$	150.00×10^{-12}
	$(156.1\pm1.6)\times10^{-12}$ [h]		
35	$(39.32\pm0.32)\times10^{-12}$ [i]	$(39.63\pm0.16)\times10^{-12}$	37.972×10^{-12}
36	$(34.08\pm0.34)\times10^{-12}$ [j]	$(33.41\pm0.13)\times10^{-12}$	31.943×10^{-12}

$Z=2$: R.S. Van Dyck, Jr.,C.E. Johnson, and H.A. Shugart, Phys. Rev. A **4**, 1327 (1971).

$Z=3$: M.H. Prior and H.A. Shugart, Phys. Rev. Lett. **27**, 902 (1971).

$Z=18$: H. Gould and R. Marrus, Phys. Rev. A **28**, 2001 (1983).

$Z=28$: R.W. Dunford, H.G. Berry, K.O. Groeneveld, M.Hass, E.Bakke, M.L.A. Raphaelian, A.E. Livingston, and L.J. Curtis, Phys. Rev. A **38**, 5423 (1988).

$Z=35$: R.W. Dunford, H.G. Berry, S. Cheng, E.P. Kanter, C. Kurtz, B.J. Zabransky, A.E. Livingston, and L.J. Curtis, Phys. Rev. A **48**, 1929 (1993).

$Z=36$: R. Marrus, V.S. Vicente, P. Charles, J.P. Briand, F.Bosch, D. Liesen, and I. Varga, Phys. Rev. Lett. **56**, 1683 (1986).

THEORETICAL FORMULATION

Regard the interaction with the electromagnetic field as a second-order perturbation resulting in the simultaneous emission of two photons $\hbar\omega_1$ and $\hbar\omega_2$ such that

$$E_i - E_f = \hbar\omega_1 + \hbar\omega_2 \tag{1}$$

Leads to broad distribution of photon energies such that the sum is equal to the atomic energy difference.

Just as in the single-photon case, the triply-differential transition rate from Fermi's Golden Rule is

$$w_{2\gamma} d\Omega_1 \, d\Omega_2 \, dE_1 = \frac{2\pi}{\hbar} |U_{if}^{(2)}|^2 \rho(\omega_1)\rho(\omega_2) \, dE_1 \tag{2}$$

where, as before, the density of states is

$$\rho_f = \frac{\mathcal{V}\omega^2}{(2\pi c)^3\hbar} d\Omega \tag{3}$$

for the number of photon states with polarization $\hat{\boldsymbol{\epsilon}}$ per unit energy and solid angle Ω in the normalization volume $\mathcal{V}$, and from QED, after integrating over time and factoring out an energy-conserving δ-function, the second-order interaction energy is

$$\begin{aligned} U_{i\to f} &= -e^2 \sum_{n\pm} \left[\frac{\langle f \mid \boldsymbol{\alpha}\cdot\mathbf{A}^*(\omega_1) \mid n\rangle\langle n \mid \boldsymbol{\alpha}\cdot\mathbf{A}^*(\omega_2) \mid i\rangle}{E_n - E_i + \hbar\omega_2} \right. \\ &\quad \left. + \frac{\langle f \mid \boldsymbol{\alpha}\cdot\mathbf{A}^*(\omega_2) \mid n\rangle\langle n \mid \boldsymbol{\alpha}\cdot\mathbf{A}^*(\omega_1) \mid i\rangle}{E_n - E_i + \hbar\omega_1} \right] \end{aligned} \tag{4}$$

summed over both positive and negative energy states. The vector potentials for the photons are

$$\mathbf{A}(\omega) = A_0 \hat{\boldsymbol{e}} e^{i\mathbf{k}\cdot\mathbf{r} - i\omega t} \tag{5}$$

with normalization factor

$$eA_0 = ce\sqrt{\hbar/(2\omega\mathcal{V})} \tag{6}$$

normalized to unit photon energy $\hbar\omega$ in the normalization volume $\mathcal{V}$. With these definitions, the two-photon decay rate becomes

$$w_{2\gamma} d\Omega_1 \, d\Omega_2 \, dE_1 = -\frac{\alpha^2\hbar\omega_1\omega_2}{(2\pi)^3} |Q(\omega_1, \omega_2|^2 d\Omega_1 \, d\Omega_2 \, dE_1 \tag{7}$$

with

$$Q(\omega_1,\omega_2) \;=\; \sum_{n\pm}\left[\frac{\langle f \mid \boldsymbol{\alpha}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_1 e^{-\mathbf{k}_1\boldsymbol{\cdot}\mathbf{r}} \mid n\rangle\langle n \mid \boldsymbol{\alpha}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_2 e^{-\mathbf{k}_2\boldsymbol{\cdot}\mathbf{r}} \mid i\rangle}{E_n - E_i + \hbar\omega_2} + \frac{\langle f \mid \boldsymbol{\alpha}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_2 e^{-\mathbf{k}_2\boldsymbol{\cdot}\mathbf{r}} \mid n\rangle\langle n \mid \boldsymbol{\alpha}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_1 e^{-\mathbf{k}_1\boldsymbol{\cdot}\mathbf{r}} \mid i\rangle}{E_n - E_i + \hbar\omega_1}\right] \tag{8}$$

summed over both positive and negative energy states.

Nonrelativistic 2E1 Approximation

Make the replacement

$$\boldsymbol{\alpha}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_i e^{-\mathbf{k}_i\boldsymbol{\cdot}\mathbf{r}} \to \frac{\mathbf{p}_i\boldsymbol{\cdot}\hat{\boldsymbol{e}}_i}{mc} \tag{9}$$

and restrict the sum in Eq. (12) to positive energy states.

The contribution from negative energy states can be evaluated in lowest order by making the approximation $E_n = -2mc^2$, and completing the sum over n by closure (see Akhiezer and Berestetskii *Quantum Electrodynamics*, p. 489) with the result

$$Q^-(\omega_1,\omega_2) \simeq \frac{1}{mc^2}\hat{\boldsymbol{e}}_1\boldsymbol{\cdot}\hat{\boldsymbol{e}}_2\langle f \mid e^{-i(\mathbf{k}_1+\mathbf{k}_2)\boldsymbol{\cdot}\mathbf{r}} \mid i\rangle. \tag{10}$$

Since $\mathbf{k} = \omega/c$, the matrix element vanishes in the long wavelength approximation if the initial and final states i and f are orthogonal. However, note that Q^- contributes to the relativistic corrections of relative order $\alpha^2 Z^2$ and must be included in an exact calculation.

With these substitutions, the expression for the positive frequency part reduces to

$$Q(\omega_1,\omega_2) \;=\; \frac{1}{m^2c^2}\sum_{n^+}\left[\frac{\langle f \mid \mathbf{p}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_1 \mid n\rangle\langle n \mid \mathbf{p}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_2 \mid i\rangle}{E_n - E_i + \hbar\omega_2} + \frac{\langle f \mid \mathbf{p}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_2 \mid n\rangle\langle n \mid \mathbf{p}\boldsymbol{\cdot}\hat{\boldsymbol{e}}_1 \mid i\rangle}{E_n - E_i + \hbar\omega_1}\right] \tag{11}$$

Averaged Decay Rate

For the case of $S_J - S_J$ transitions via intermediate P-states, the sum over magnetic substates results in a transition rate proportional to $|\hat{\boldsymbol{e}}_1 \cdot \hat{\boldsymbol{e}}_2|^2$.

Problem: Sum over two independent polarization vectors $\hat{\boldsymbol{e}}_1$ and $\hat{\boldsymbol{e}}_2$ perpendicular to $\mathbf{k}_1$ and $\mathbf{k}_2$.

Solution: Let $\mathbf{k}_1$ and $\mathbf{k}_2$ define the xy-plane (the collision plane). Then two possible independent choices for $|\hat{\boldsymbol{e}}_1 \cdot \hat{\boldsymbol{e}}_2|^2$ are

1. Choose $\hat{\boldsymbol{e}}_1 = \hat{\boldsymbol{e}}_z, \quad \hat{\boldsymbol{e}}_2 = \hat{\boldsymbol{e}}_z$. Then $\hat{\boldsymbol{e}}_1 \cdot \hat{\boldsymbol{e}}_2|^2 = 1$.
2. Choose $\hat{\boldsymbol{e}}_1 = \hat{\boldsymbol{k}}_1 \times \hat{\boldsymbol{e}}_z, \quad \hat{\boldsymbol{e}}_2 = \hat{\boldsymbol{k}}_2 \times \hat{\boldsymbol{e}}_z$. Then, since $\hat{\boldsymbol{k}}_i \cdot \hat{\boldsymbol{e}}_z = 0$,

$$\begin{aligned} |\hat{\boldsymbol{e}}_1 \cdot \hat{\boldsymbol{e}}_2|^2 &= |(\hat{\boldsymbol{k}}_1 \times \hat{\boldsymbol{e}}_z) \cdot (\hat{\boldsymbol{k}}_2 \times \hat{\boldsymbol{e}}_z)|^2 \\ &= |\hat{\boldsymbol{k}}_1 \cdot \hat{\boldsymbol{k}}_2|^2 \\ &= \cos^2\theta_{12} \end{aligned}$$

The sum of both polarization contributions is thus a factor of $1 + \cos^2\theta_{12}$.

Finally, integrating over $d\Omega_1$ and $d\Omega_2$ gives a multiplying factor of

$$\int_{4\pi} d\Omega_1 \int_{4\pi} d\Omega_2 (1 + \cos^2\theta_{12}) = 4\pi \times 4\pi \times (1 + 1/3)$$

and transform to the "length" gauge by use of the commutator

$$\begin{aligned} \langle i \mid \mathbf{p} \mid n \rangle &= \frac{im}{\hbar} \langle i \mid [H, \mathbf{r}] \mid n \rangle \\ &= \frac{im}{\hbar} (E_i - E_n) \langle i \mid \mathbf{r} \mid n \rangle \end{aligned}$$

The final result for the two-photon decay rate is then

$$w_{\gamma_1,\gamma_2}\, dE_1 = \frac{16\omega_1^3\omega_2^3 e^4}{3hc^6} \left| \sum_{n^+} \left[\frac{\langle f \mid z \mid n \rangle \langle n \mid z \mid i \rangle}{E_n - E_i + \hbar\omega_2} + \frac{\langle f \mid z \mid n \rangle \langle n \mid z \mid i \rangle}{E_n - E_i + \hbar\omega_1} \right] \right|^2 dE_1 \tag{12}$$

ADDITIONAL REFERENCES

- For relativistic corrections:
  G. W. F. Drake, "Spontaneous two photon decay rates in hydrogen-like and helium-like ions," Phys. Rev. A **34**, 2871 (1986), and

  S. P. Goldman and G. W. F. Drake, "Relativistic two-photon decay rates of $2s_{1/2}$ hydrogenic ions," Phys. Rev. A **24**, 183 (1981), and

  W.R. Johnson, Phys. Rev. Lett. **29**, 1123 (1972), and

  F.A. Parpia and W.R. Johnson, Phys. Rev. A **26**, 1142 (1982), and

  A. Derevianko and W.R. Johnson, Phys. Rev. A **56**, 1288 (1997). .

- For E1M1 two-photon tramsitions:
  G. W. F. Drake, "Energy level calculations and E1-M1 two photon transition rates in two-electron U^{90+}," Nucl. Instrum. and Methods B **9**, 465 (1985), and

  I.M. Savukov and W.R.Johnson, Phys. Rev. A **66**, 062507 (2002).

APPLICATION TO THE TUNE-OUT WAVELENGTH

Basic idea: treat as a two-photon process for coherent photon scattering (Rayleigh scattering).

Same as two-photon decay with the replacements

- $\mathbf{k}_2 \to -\mathbf{k}_2$
- $\omega_2 \to -\omega_2$
- $\mathbf{k}_1 = \mathbf{k}_2$ and $\omega_1 = \omega_2$.
- $\mathbf{A_2}^* \to \mathbf{A}_2$ (i.e. absorption in place of emission)

Collaborations

- **Experiment:** Ken Baldwin, Australian National University
- **Theory:** Li-Yan Tang, Wuhan Institute of Physics and Mathematics (WIPM)

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Helium tune-out wavelength: Gauge invariance and retardation corrections

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Abstract

The problem of calculating the tune-out wavelength for an atom interacting with a plane electromagnetic wave is formulated as a zero in the Rayleigh scattering cross section, rather than a zero in the dynamic polarizability. Retardation (finite wavelength) corrections are discussed in the velocity gauge, and possible gauge transformations to a length form are investigated. For the special case of S-states, it is shown that a pure length form exists for the leading $p_x z$ retardation correction, even though one does not exist in general. The results of high-precision calculations in Hylleraas coordinates are presented for the tune-out wavelength of helium near the $2\,^3S - 3\,^3P$ transition at 413 nm.

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Rapid Communications

QED and relativistic nuclear recoil corrections to the 413-nm tune-out wavelength for the $2\,^3S_1$ state of helium

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Comparison of high-accuracy calculations with precision measurement of the 413-nm tune-out wavelength of the He($2\,^3S_1$) state provides a unique test of quantum electrodynamics (QED). We perform large-scale relativistic-configuration-interaction (RCI) calculations of the tune-out wavelength that include the mass-shift operator and fully account for leading relativistic nuclear recoil terms in the Dirac-Coulomb-Breit (DCB) Hamiltonian. We obtain the QED correction to the tune-out wavelength using perturbation theory, and the effect of finite nuclear size is also evaluated. The resulting tune-out wavelengths for the $2\,^3S_1(M_J = 0)$ and $2\,^3S_1(M_J = \pm1)$ states are 413.084 26(4) nm and 413.090 15(4) nm, respectively. When we incorporate the retardation correction of 0.000 560 0236 nm obtained by Drake *et al.* [Hyperfine Interact **240**, 31 (2019)] to compare results with the only current experimental value of 413.0938($9_{\rm stat}$)($20_{\rm syst}$) nm for the $2\,^3S_1(M_J = \pm1)$ state, there is 1.4σ discrepancy between theory and experiment, which stimulates further theoretical and higher precision experimental investigations on the 413-nm tune-out wavelength. In addition, we also determine the QED correction for the static dipole polarizability of the He($2\,^3S_1$) state to be 22.5 ppm, which may enable a new test of QED in the future.

OUTLINE

- What is a tune-out wavelength
- Reformulation as a zero in the Rayleigh scattering cross section (photons) instead of the AC-Stark shift (optical lattices)
- Retardation (finite wavelength) corrections
- Gauge transformations and the non-existence of a “length” form
- Hylleraas wave functions for helium
- Relativistic and QED corrections
- Results and comparison with experiment for helium as a novel test of QED

Motivation

- Find new ways to detect and test quantum electrodynamic (QED) effects in atoms, other than energy differences (Lamb shift).
- So-called "tune-out" wavelengths can be measured to very high precision, and compared with theory.
- the tune-out wavelength is determined primarily by the frequency-dependent polarizability. It is the wavelength (or equivalent frequency) where the frequency-dependent polarizability vanishes.
- The polarizability in turn is determined by dipole matrix elements, as well as transition energies.

History

- First noted by LeBlanc and Thywissen (PRA **75**, 053612 (2007) in connection with species-specific optical lattices for alkali metals.

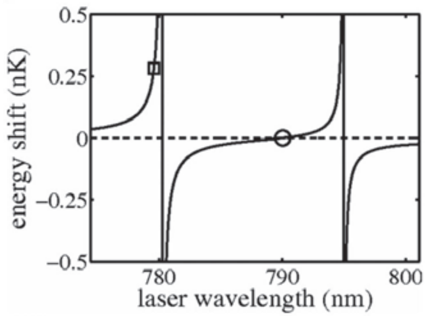

Figure 1
Energy shift as a function of wavelength for ^{87}Rb in the $|F, m_F\rangle = |2, 2\rangle$ state, under linear polarization, for

- Helium $2\ ^3S - 3\ ^3P$ experiment suggested by Jim Mitroy and Li-Yan Tang, Phys. Rev. A **88**, 052515 (2013).

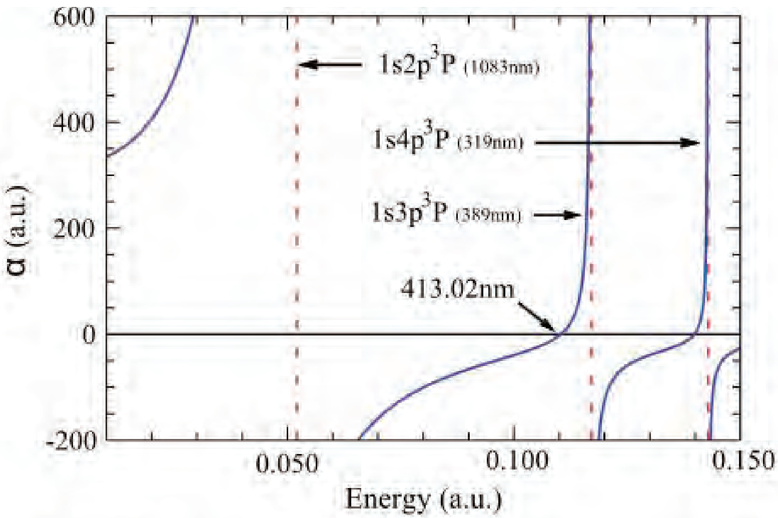

FIG. 1 (color online). Helium polarizability spectrum (solid curves) as a function of energy (a.u.). Triplet transition manifold positions are shown by the dotted vertical lines.

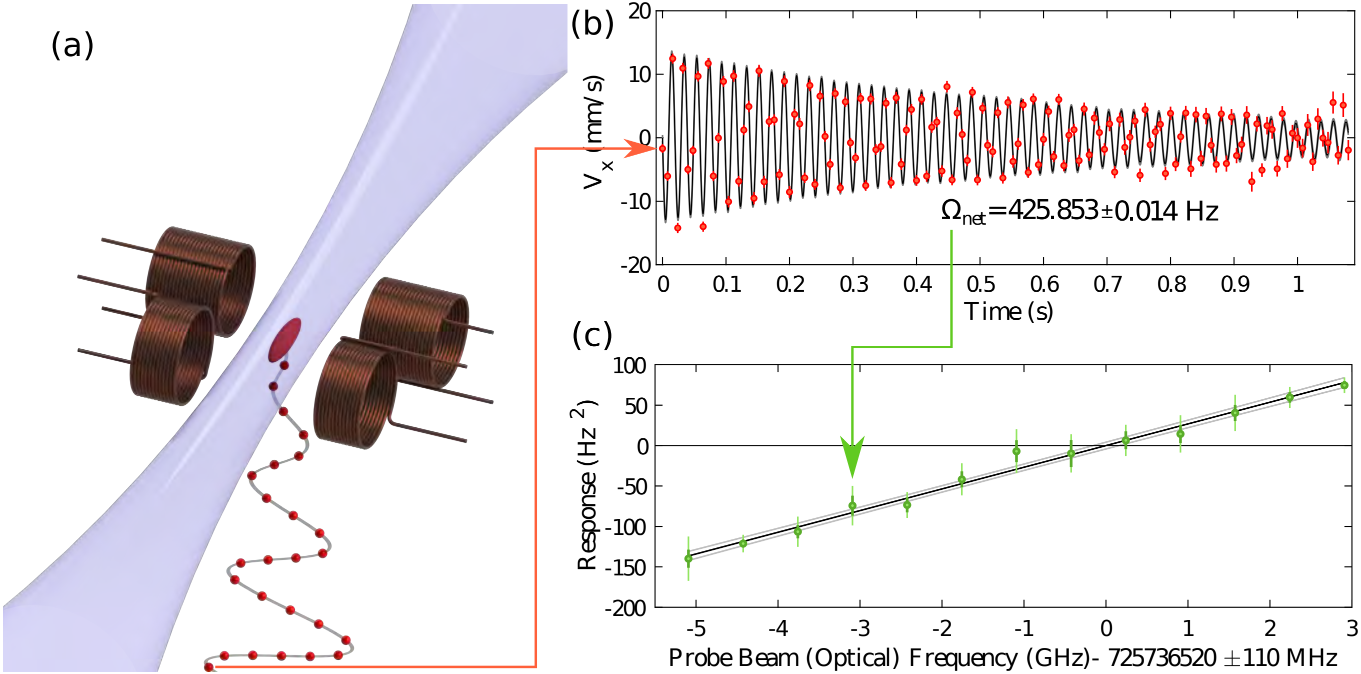

FIG. 2. Method to determine the ω_{TO} for a given polarization state. (a) As shown on this schematic, a magnetically trapped BEC of metastable helium atoms (red) is illuminated with a probe laser beam (blue) with an adjustable frequency. The BEC is made to oscillate and a sequence of pulses are outcoupled, each consisting of a small fraction of the BEC atoms. The mean position of each pulse is determined (b) and converted to a velocity v_x (red points - single experiential realization shown). By fitting the velocity as a function of time with a damped sine wave model (black line), the trap frequency can be extracted. (c) The difference in the trap frequency between runs with the probe beam on and off is then used to determine the change in the trap frequency (termed the response) due to the laser beam (green points). As the frequency of the probe laser beam is varied, the response observed to varies approximately linearly (black line). The x intercept is determined as the measurement of the tune-out frequency for this particular optical polarization state. [BMH:Clip fig a to show more detail, fix arrow]

Precision Measurement for Metastable Helium Atoms of the 413 nm Tune-Out Wavelength at Which the Atomic Polarizability Vanishes

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We present the first measurement for helium atoms of the tune-out wavelength at which the atomic polarizability vanishes. We utilize a novel, highly sensitive technique for precisely measuring the effect of variations in the trapping potential of confined metastable (2^3S_1) helium atoms illuminated by a perturbing laser light field. The measured tune-out wavelength of $413.0938(9_{\rm stat})(20_{\rm syst})$ nm compares well with the value predicted by a theoretical calculation [413.02(9) nm] which is sensitive to finite nuclear mass, relativistic, and quantum electrodynamic effects. This provides motivation for more detailed theoretical investigations to test quantum electrodynamics.

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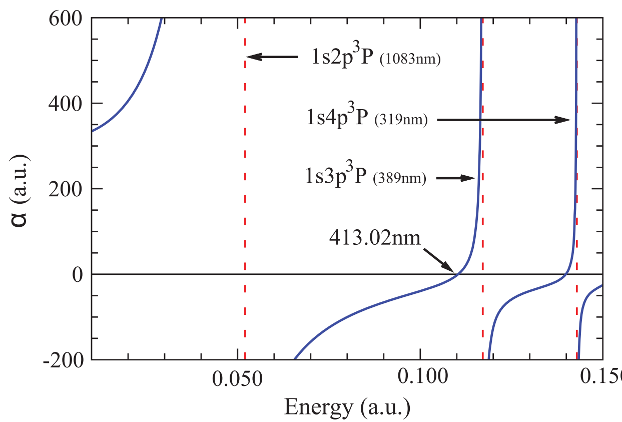

FIG. 1 (color online). Helium polarizability spectrum (solid curves) as a function of energy (a.u.). Triplet transition manifold positions are shown by the dotted vertical lines.

TABLE 1. Contributions to the tune-out wavelength and their orders of magnitude (in units of a_0, where a_0 is the Bohr radius)

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Magnitude	Physical origin
unity	nonrelativistic Schrödinger equation
$\mu/M \simeq 10^{-4}$	mass pol. operator $-(\mu/M)\nabla_1 \cdot \nabla_2$
$\alpha^2 \simeq 10^{-4}$	Breit (relativistic) interaction
$\alpha^2 \simeq 10^{-4}$	finite wavelength corrections
$\alpha^2\mu/M \simeq 10^{-7}$	Relativistic recoil + Stone term
$\alpha^3 \simeq 10^{-6}$	QED terms

Basic idea: treat as a two-photon process for coherent photon scattering (Rayleigh scattering).

Same as two-photon decay with the replacements

- $\mathbf{k}_2 \rightarrow -\mathbf{k}_2$
- $\omega_2 \rightarrow -\omega_2$
- $\mathbf{k}_1 = \mathbf{k}_2$ and $\omega_1 = \omega_2$.
- $\mathbf{A_2}^* \rightarrow \mathbf{A}_2$ (i.e. absorption in place of emission)

Scattering Theory

Treat as a Rayleigh scattering process instead of the AC Stark shift.

See e.g. Sindelka, Moiseyev and Cederbaum, Phys. Rev. A **74**, 053420 (2006).

Relativistic Case

The effective interaction energy corresponding to Rayleigh scattering of a photon with frequency ω, wave vector $\mathbf{k} = \hat{z}$ and polarization $\mathbf{e} = \hat{x}$ is

$$U(\omega) \ = \ \frac{2\pi\alpha}{\omega} \sum_{n\pm} \left[\frac{\langle i \mid \hat{\boldsymbol{e}} e^{-i\mathbf{k}\cdot\mathbf{r}} \mid n\rangle\langle n \mid \hat{\boldsymbol{e}} e^{i\mathbf{k}\cdot\mathbf{r}} \mid i\rangle}{E_n - E_i - \omega} + \frac{\langle i \mid \hat{\boldsymbol{e}} e^{i\mathbf{k}\cdot\mathbf{r}} \mid n\rangle\langle n \mid \hat{\boldsymbol{e}} e^{-i\mathbf{k}\cdot\mathbf{r}} \mid i\rangle}{E_n - E_f + \omega} \right]$$

summed over positive and negative energy states, where $\hat{\boldsymbol{e}} = \gamma_\mu e_\mu$. Approximate $E_n = -mc^2$ for the negative energy states, and complete the sum by closure to obtain

$$U(\omega) \ = \ \frac{2\pi\alpha}{\omega} \left\{ \sum_{n^+} \left| \langle i \mid \alpha_x e^{ikz} \mid n\rangle \right|^2 \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega} \right) - N \right\}$$

where $N =$ number of electrons. The last term is the 'seagull" A^*A term.

Nonrelativistic Approximation

With the replacement $\alpha_x \to p_x/(\mu c)$,

$$U(\omega) \;\propto\; \frac{1}{\omega}\left\{\left(1+\frac{\mu}{M}\right)\sum_n \left|\langle i \mid p_x e^{ikz} \mid n\rangle\right|^2 \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega}\right) - N\right\}$$

in reduced mass atomic units, where $\mu = mM/(m+M)$ is the electron reduced mass.

The Finite Wavelength Terms

Expand $e^{ikz} = 1 + ikz - \frac{1}{2}(kz)^2 + \cdots$ and correspondingly

$$U(\omega) = U_0(\omega) + (\omega/c)^2 U_2(\omega) + \cdots$$

where $k = \omega/c$. The dipole term (in the velocity gauge) is

$$U_0^{(V)}(\omega) \;=\; \frac{1}{\omega}\left\{\left(1+\frac{\mu}{M}\right)\sum_{n\,{}^3P} |\langle i \mid p_x \mid n\rangle|^2 \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega}\right) - N\right\}$$

and the finite wavelength (quadrupole-like) Q-term is

$$U_{2,\mathrm{Q}}^{(V)}(\omega) \;=\; \frac{1}{\omega}\left(1+\frac{\mu}{M}\right)\sum_{n\,{}^3D} |\langle i \mid p_x z \mid n\rangle|^2 \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega}\right)$$

and the finite wavelength (cross) X-term is

$$U_{2,\mathrm{X}}^{(V)}(\omega) \;=\; -\frac{1}{\omega}\left(1+\frac{\mu}{M}\right)\sum_{n\,{}^3P} |\langle i \mid p_x \mid n\rangle\langle n \mid p_x z^2 \mid i\rangle \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega}\right)$$

Gauge Invariance: The Length Gauge

Use the commutation relations

$$\begin{aligned} i\mu[H,x]/\hbar &= p_x \\ i\mu[H,xz]/\hbar &= xp_z + zp_x \\ &= 2zp_x \quad \text{for S-states} \end{aligned}$$

the expressions for the interaction energy become

$$U_0^{(L)}(\omega) = \omega \sum_n |\langle i \mid x \mid n\rangle|^2 \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega}\right)$$

and, for the finite wavelength Q-term (for S-states)

$$U_{2,\mathrm{Q}}^{(L)}(\omega) = \frac{\omega}{4} \sum_n |\langle i \mid xz \mid n\rangle|^2 \left(\frac{1}{E_n - E_i + \omega} + \frac{1}{E_n - E_i - \omega}\right)\left(\frac{E_n - E_i}{\omega}\right)^2$$

If the wave functions are exact, then

$U_0^{(V)}(\omega) = U_0^{(L)}(\omega)$ and
$U_{2,\mathrm{Q}}^{(V)}(\omega) = U_{2,\mathrm{Q}}^{(L)}(\omega)$

to all orders in μ/M (verified numerically).

Nonrelativistic Tune-out Wavelength (nm)

Infinite mass case:

$$\begin{aligned}
\text{Length form} &= 413.038\,304\,3869(20)\\
\text{Velocity form} &= 413.038\,304\,3858(1)\\
\text{Difference} &= \quad 0.000\,000\,0011(20)
\end{aligned}$$

^{4}He Finite mass case

$$\begin{aligned}
\text{Length form} &= 413.082\,590\,5829(20)\\
\text{Velocity form} &= 413.082\,590\,5819(1)\\
\text{Difference} &= \quad 0.000\,000\,0010(20)
\end{aligned}$$

Refresher on Gauge Transformations

Maxwell's equations are

$$\begin{aligned}\mathbf{E} &= -\nabla V - \frac{1}{c}\frac{\partial \mathbf{A}}{\partial t} \\ \mathbf{B} &= \nabla \times \mathbf{A}\end{aligned}$$

The physical fields $\mathbf{E}$ and $\mathbf{B}$ are invariant under an arbitrary transformation of the scalar and vector potentials according to

$$\begin{aligned}\mathbf{A} &\longrightarrow \mathbf{A} + \nabla f(\mathbf{r}, t) \\ V &\longrightarrow V - \frac{1}{c}\frac{\partial f(\mathbf{r}, t)}{\partial t}\end{aligned}$$

where $f(\mathbf{r}, t)$ is an arbitrary differentiable function of $\mathbf{r}$ and t.

For a wave propagating in the z-direction and polarized in the x-direction, choose

$$\begin{aligned}\mathbf{A} &= \hat{\mathbf{e}}_{\mathbf{x}}\, e^{ikz - i\omega t} \\ f(\mathbf{r}, t) &= \mathcal{C}\, e^{ikz - i\omega t}\end{aligned}$$

where $\mathcal{C}$ is an arbitrary constant.

For a wave propagating in the z-direction and polarized in the x-direction, choose

$$\begin{aligned} \mathbf{A} &= \hat{\mathbf{e}}_{\mathbf{x}}\, e^{ikz-i\omega t} \\ f(\mathbf{r},t) &= \mathcal{C}\, e^{ikz-i\omega t} \end{aligned}$$

where $\mathcal{C}$ is an arbitrary constant. Then

$$\begin{aligned} \mathbf{A} &\longrightarrow \hat{\mathbf{e}}_{\mathbf{x}}\, e^{ikz-i\omega t} + \mathcal{C} ik \hat{\mathbf{e}}_{\mathbf{z}}\, e^{ikz-i\omega t} \\ V &\longrightarrow V + \mathcal{C}\frac{i\omega}{c}\, e^{ikz-i\omega t} \end{aligned}$$

where $k = \omega/c$. Define a new arbitrary constant $\mathcal{C}' = ik\mathcal{C}$. Then

$$\begin{aligned} \mathbf{A} &= \hat{\mathbf{e}}_{\mathbf{x}}\, e^{ikz-i\omega t} + \mathcal{C}\hat{\mathbf{e}}_{\mathbf{z}}\, e^{ikz-i\omega t} \\ V &= V_0 + \mathcal{C}\, e^{ikz-i\omega t} \end{aligned}$$

Gauge Invariance and the Length Form

A "length form" for retardation corrections in general does not exist. Why?

Answer:

In the velocity form, the interaction operator is

$$U = \frac{1}{mc}\mathbf{p} \cdot \mathbf{A} + V$$

where

$$\begin{aligned} \mathbf{A} &= \frac{1}{\sqrt{2\Omega\omega}}(\mathbf{e_x} + \mathcal{C}\mathbf{e_z})\, e^{ikz-i\omega t} \quad \text{(vector part)} \\ V &= \frac{1}{\sqrt{2\Omega\omega}}\mathcal{C}\, e^{ikz-i\omega t} \quad \text{(scalar part)} \end{aligned}$$

$k = \omega/c$ and $\mathcal{C}$ is an arbitrary constant. Gauge invariance then requires for matrix elements

$$\frac{1}{mc}\langle a \mid p_z e^{ikz} \mid b\rangle = -\langle a \mid e^{ikz} \mid b\rangle$$

Expanding $e^{ikz} = 1 + ikz + \cdots$, the leading terms give

$$\frac{1}{mc}\langle a \mid p_z \mid b\rangle = -\frac{i\omega_{ab}}{c}\langle a \mid z \mid b\rangle$$

This is the usual equivalence of the length and velocity forms. However, the correct velocity operator is $\boldsymbol{p_x e^{ikz}}$, not $\boldsymbol{p_z e^{ikz}}$. In general, there is no equivalent length form valid beyond leading order.

Gauge Invariance of Multipole Expansions

Expand waves of definite direction of propagation $\mathbf{k}$ and direction of polarization $\hat{\mathbf{e}}$ in terms of definite angular momentum and parity. Since

$$\nabla^2 r^L Y_L^M(\theta, \phi) = 0$$

it is always possible to write

$$\omega_{ab}\langle a \mid r^L Y_L^M \mid b\rangle \;=\; \langle a \mid [H, r^L Y_L^M] \mid b\rangle$$

and (in atomic units)

$$\begin{aligned} [H, r^L Y_L^M] &= -\frac{1}{2}[\nabla^2, r^L Y_L^M] \\ &= -\nabla r^L Y_L^M \cdot \nabla \\ &= -i\nabla r^L Y_L^M \cdot \mathbf{p} \end{aligned}$$

which is the correct velocity form of the operator. However, this works only to lowest order. The function r^L is just the leading term in the power series expansion of the spherical Bessel function $j_L(kr) = j_L(\omega r/c)$.

Pseudospectral Theory

Diagonalize H_0 in a discrete variational basis set of functions ϕ_p, $p = 0, \ldots, N-1$ such that

$$\begin{aligned} \langle \phi_p | \phi_q \rangle &= \delta_{p,q} \\ \langle \phi_p | H_0 | \phi_q \rangle &= \varepsilon_p \, \delta_{p,q} \end{aligned}$$

.

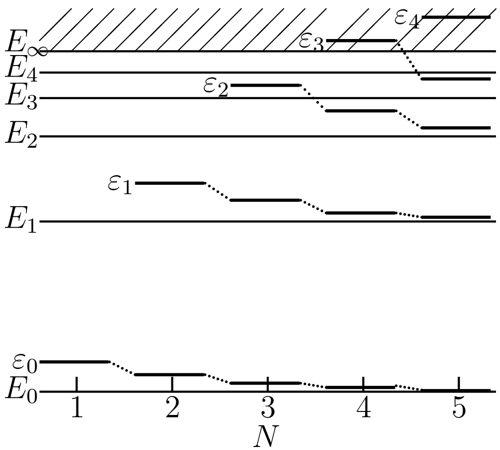


Figure 1: Diagram illustrating the HUM theorem. The ε_p, $p = 0, \ldots, N-1$ are the variational eigenvalues for an N-dimensional basis set, and the E_i are the exact eigenvalues of H_0. The highest ε_p lie in the continuous spectrum of H_0.

Replace the $\{\psi_n,\, E_n\}$, $n = 0, \ldots, \infty$ by $\{\phi_p,\, \varepsilon_p\}$, $p = 0, \ldots, N-1$ to obtain

$$\tilde{\alpha}_{\rm d}(\omega) \;=\; 2e^2 \sum_{p\neq 0}^{N-1} \frac{(\varepsilon_p - \varepsilon_0)\,\langle\phi_0|\hat{\boldsymbol{\epsilon}}^{\,*}\!\cdot\mathbf{r}|\phi_p\rangle\langle\phi_p|\hat{\boldsymbol{\epsilon}}\cdot\mathbf{r}|\phi_0\rangle}{(\varepsilon_p - \varepsilon_0)^2 - (\hbar\omega)^2}$$

Then $\tilde{\alpha}_{\rm d}(\omega) \to \alpha_{\rm d}(\omega)$ as $N \to \infty$, provided that the basis set is complete.

Variational Justification (Chan and Dalgarno, 1965)

Write $\psi^{(1)} = \sum_{p=1}^{N-1} a_p\phi_p$,

and construct $J_\pm(\omega) = \langle\psi^{(1)}|H - E_0 \pm \hbar\omega|\psi^{(1)}\rangle + 2\langle\psi^{(1)}|\hat{\boldsymbol{\epsilon}}\cdot\mathbf{r}|\psi_0\rangle$.

Then

$$\delta J_\pm(\omega) \;=\; \frac{\partial J_\pm(\omega)}{\partial a_p}\delta a_p = 0$$

$$\Rightarrow \quad a_p \;=\; \frac{\langle\phi_p|\hat{\boldsymbol{\epsilon}}\cdot\mathbf{r}|\phi_0\rangle}{E_0 - \varepsilon_p \mp \hbar\omega}$$

Convergence study for the nonrelativistic tune-out wavelength λ. N is the number of terms in the basis set.

N	λ (nm)	Difference (nm)
140	413.082 328 731 87	
190	413.082 581 514 32	0.000 252 782 45
246	413.082 578 777 26	–0.000 002 737 06
315	413.082 575 775 67	–0.000 003 001 59
393	413.082 574 808 89	–0.000 000 966 78
485	413.082 574 887 63	0.000 000 078 74
587	413.082 574 836 65	–0.000 000 050 98
705	413.082 574 825 76	–0.000 000 010 89
843	413.082 574 823 05	–0.000 000 002 71
981	413.082 574 822 39	–0.000 000 000 66
1140	413.082 574 822 16	–0.000 000 000 23
1319	413.082 574 821 98	–0.000 000 000 18
1906	413.082 574 821 91	–0.000 000 000 07

$$\begin{aligned}\alpha_{\mathrm{D}}(\omega) &= 2e^2 \sum_{n\neq 0} \frac{(E_n - E_0)|\langle 0 \mid z \mid n\rangle|^2}{(E_n - E_0)^2 - (\hbar\omega)^2} \\ &= 0\end{aligned}$$

Relativistic Corrections to the Dynamic Polarizability

Terms of second order in the external electric field and first-order in H_{rel} are

$$\alpha_{\text{D,rel}}(\omega) = \sum_{n,n' \neq 0} \left[\frac{-2(E_{n'} - E_0)\langle 0|H_{\text{rel}}|n\rangle\langle n|z|n'\rangle\langle n'|z|0\rangle}{(E_n - E_0)[(E_{n'} - E_0)^2 + \omega^2]} + \frac{\langle 0|z|n'\rangle\langle n'|(\langle H_{\text{rel}}\rangle - H_{\text{rel}})|n\rangle\langle n|z|0\rangle[(E_{n'} - E_0)(E_n - E_0) + \omega^2]}{[(E_n - E_0)^2 - \omega^2][(E_{n'} - E_0)^2 - \omega^2]} \right]$$

The Breit Interaction and Relativistic Recoil

The Breit interaction $H_{\mathsf{rel}} = B$ comes from lowest-order relativistic corrections (in atomic units)

$$B = \alpha^2 \sum_{i=1}^{2} \left[-\frac{1}{8}\nabla_i^4 + \frac{\pi Z}{2}\delta(\mathbf{r}_i) \right] + H_{\mathsf{orbit-orbit}} + H_{\mathsf{spin-spin}}$$

The "Stone" term (after A.P. Stone) of order $\alpha^2\mu/M$ comes from transforming the Breit interaction to c.m. plus relative coordinates.

$$\tilde{\Delta}_2 = \frac{Z\alpha^2}{2}\frac{\mu}{M}\left\{ \frac{1}{r_1}(\nabla_1 + \nabla_2)\cdot\nabla_1 + \frac{1}{r_1^3}\mathbf{r}_1\cdot[\mathbf{r}_1\cdot(\nabla_1 + \nabla_2)]\nabla_1 \right\}$$
$$+ 1 \leftrightarrow 2$$

QED Corrections

Include the additional "Lamb shift" type perturbations

$$
\begin{aligned}
C_1 &= \frac{8\alpha^3}{3}\left(\frac{19}{30} - 2\ln\alpha - \ln k_0\right)[\delta(r_1) + \delta(r_2)] \\
C_2 &= \alpha^3\left(\frac{164}{15} + \frac{14}{3}\ln\alpha\right)\delta(r_{12}) \\
C_3 &= -\frac{7\alpha^3}{6\pi}\left(\frac{1}{r_{12}^3}\right)_{\mathsf{PV}}
\end{aligned}
$$

in the same way as the relativistic corrections, where $\ln k_0$ is the Bethe logarithm (approximate by the field-free value).

TABLE 1. Nonrelativistic, relativistic, and QED contributions to the tune-out wavelength for the ^{4}He $1s2s\ ^3S$ state, including relativistic recoil of order $\alpha^2\mu/M$.

Terms included	λ_t (nm)	Zhang [1]
Nonrelativistic	413.038 304 3858	413.038 28(3)
NR + Rel. ($M = 0$)	413.079 958(2)	413.080 00(1)
NR + Rel. ($M = \pm 1$)	413.085 828(2)	413.085 89(1)
Finite wavelength Q	**0.000 5600**	
Finite wavelength X	**–0.000 0106**	
α^3 QED	0.004 1531	0.004 147 729(2)
α^4 QED	0.000 072 077 [a]	0.000 072 077
$\alpha^3\ \delta\ln(k_0)$	0.000 04(1) [a]	0.000 04(1)
Nuclear size	0.000 002 75 [a]	0.000 002 75
Total ($M = 0$)	413.084 78(1)	413.084 81(4) [b]
Total ($M = \pm 1$)	413.090 65(1)	413.090 70(4) [b]
Experiment [1]	413.093 8(9stat)(20syst)	
Difference	0.003 1(20)	

[a] From Zhang et al. [1] and private communication.

[b] adjusted by the missing 0.000 560 nm and –0.000 011 nm finite wavelength terms.

Conclusions

- Very high precision has been obtained in both the the length (L) and velocity (V) forms for the lowest-order nonrelativistic tune-out wavelength, including mass polarization and relativistic corrections, but the velocity form must be used for retardation corrections.

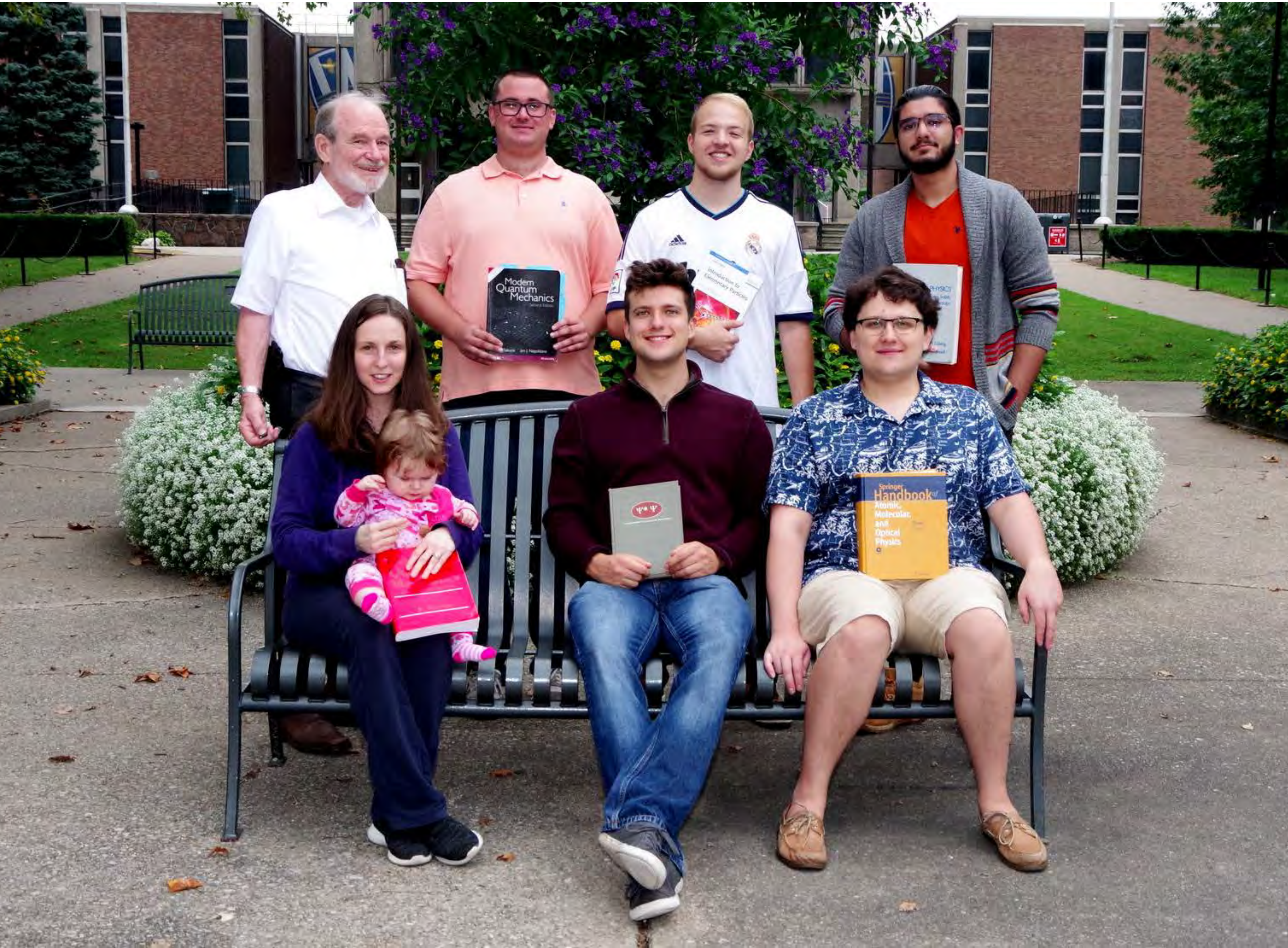
- Good agreement has been obtained with the less accurate calculations of Zhang et al. [2] obtained by the relativistic CI method. As a check, we also obtained good agreement with the corresponding QED correction to the polarizability [4,5] for the $1s^2\ ^1S$ state.

- The quadrupole shift of 0.000 5600 nm is significant compared with the QED shift, but is not sufficient to resolve the disagreement with experiment (see Table 1).

- The 1.4σ disagreement with experiment shown in Table 1 could be accounted for by further finite wavelength or binding-energy corrections from the sum over negative energy states not yet taken into account. Further experiments and calculations are in progress at ANU.

For further reading, see G.W.F. Drake et al. Hyperfine Int. **240**:31 (2019).

Web page for helium wave functions:
http://drake.sharcnet.ca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