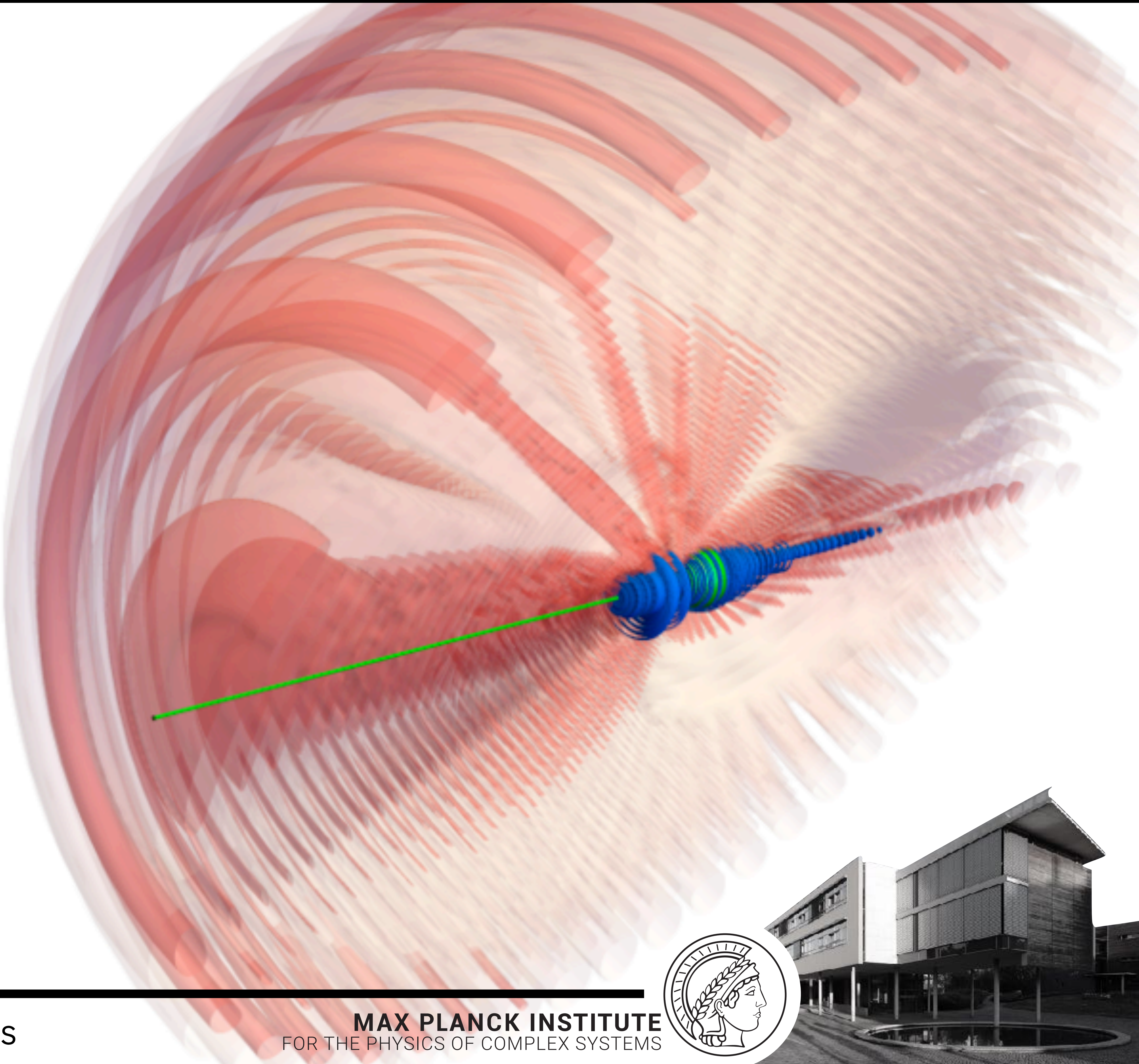
Rydberg Systems: Exciting Possibilities in Excited Atoms



Matt Eiles
MPI-PKS

Quantum Dynamics - Fundamentals and Realizations

MAX PLANCK INSTITUTE
FOR THE PHYSICS OF COMPLEX SYSTEMS

Scope of today's lecture

At the core of quantum simulation with Rydberg atoms: 150 years of spectroscopy

- From Rydberg to Pauli/Schrödinger to present day

As billed, it is a "lecture":

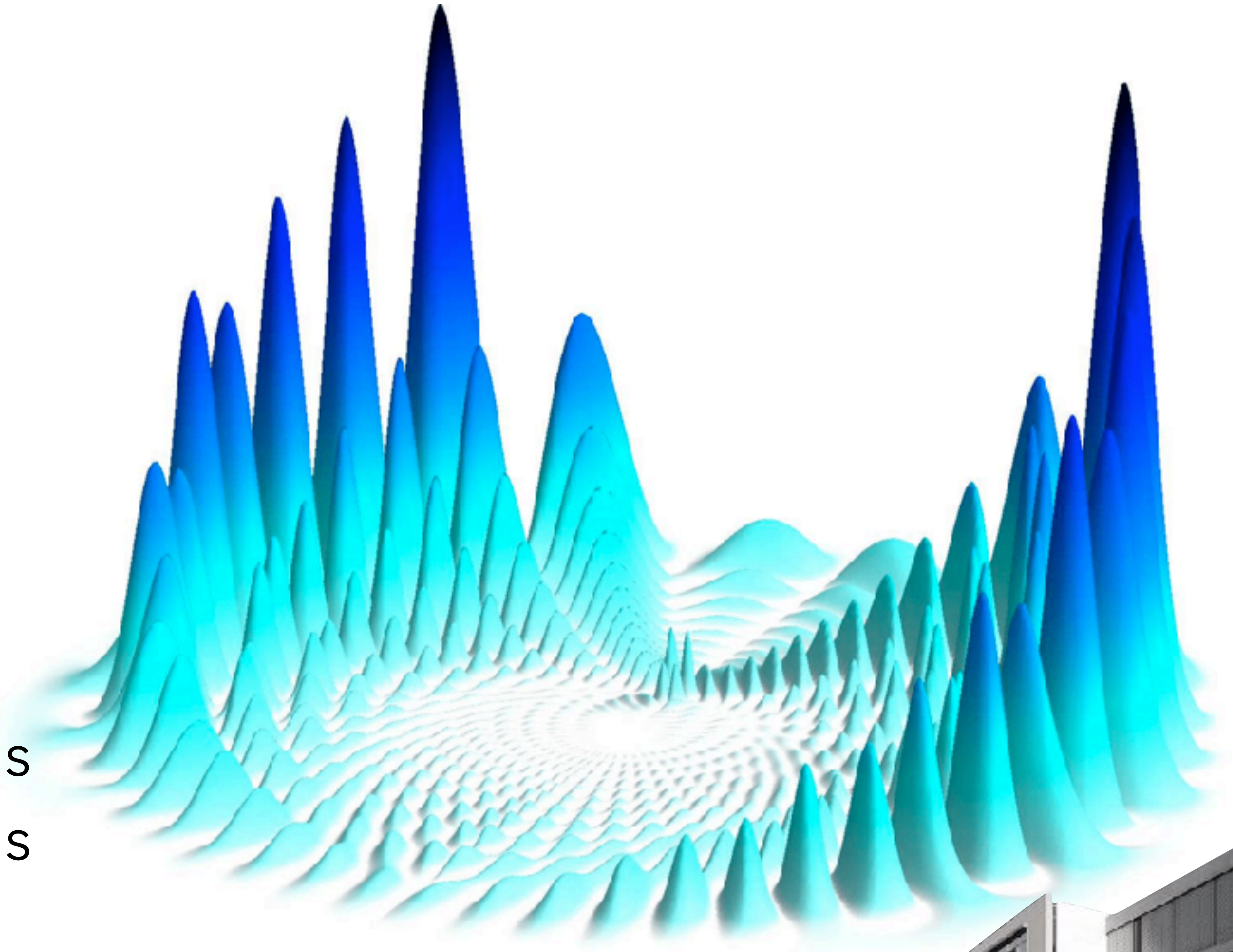
- …expect some equations…
- slides: https://www.pks.mpg.de/correlations-and-transport-in-rydberg-matter

What are Rydberg atoms?

- Quantum defect theory: alkali atoms
- Key properties of Rydberg atoms
- Multichannel quantum defect theory: many-electron atoms

What are they good for?

- Rydberg-Rydberg interactions
  - van der Waals / Rydberg blockade
  - dipole-dipole / "flip-flop" interactions
- Rydberg-ground-state-atom interactions



*we won't actually discuss this.
it's just to get your attention

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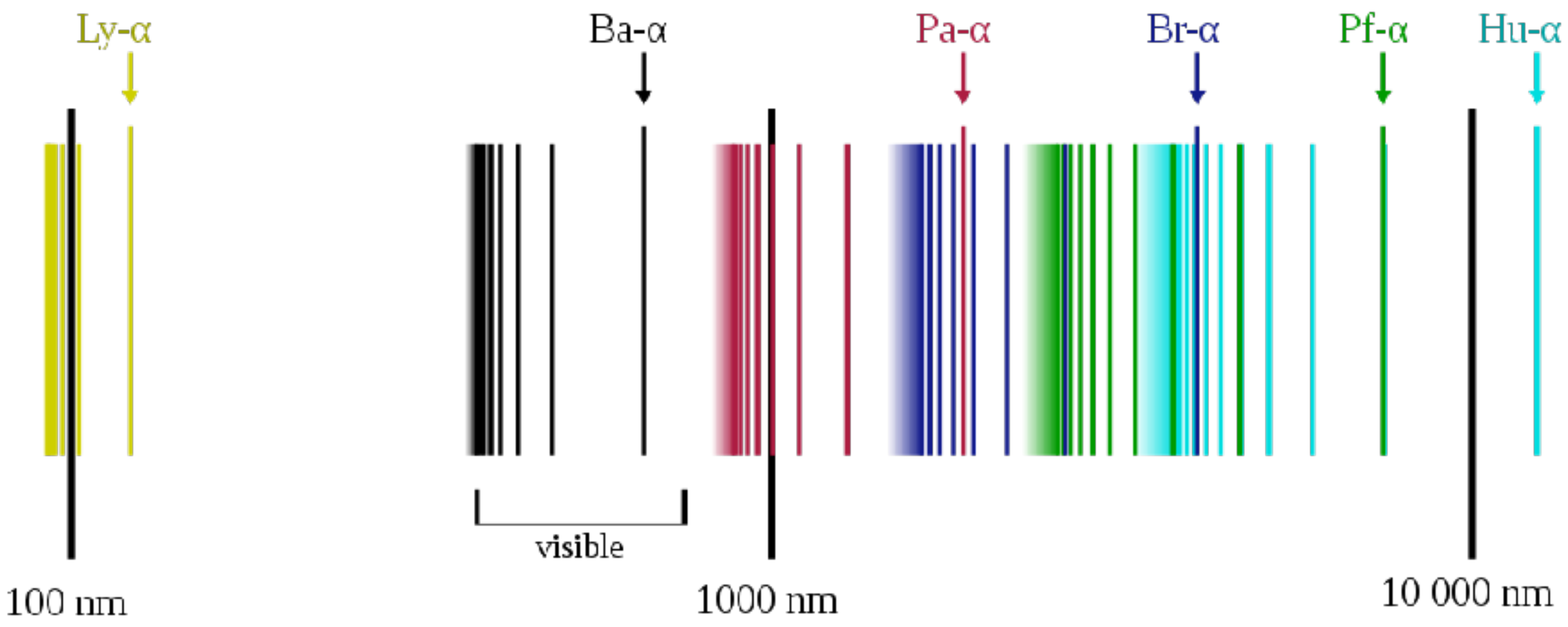
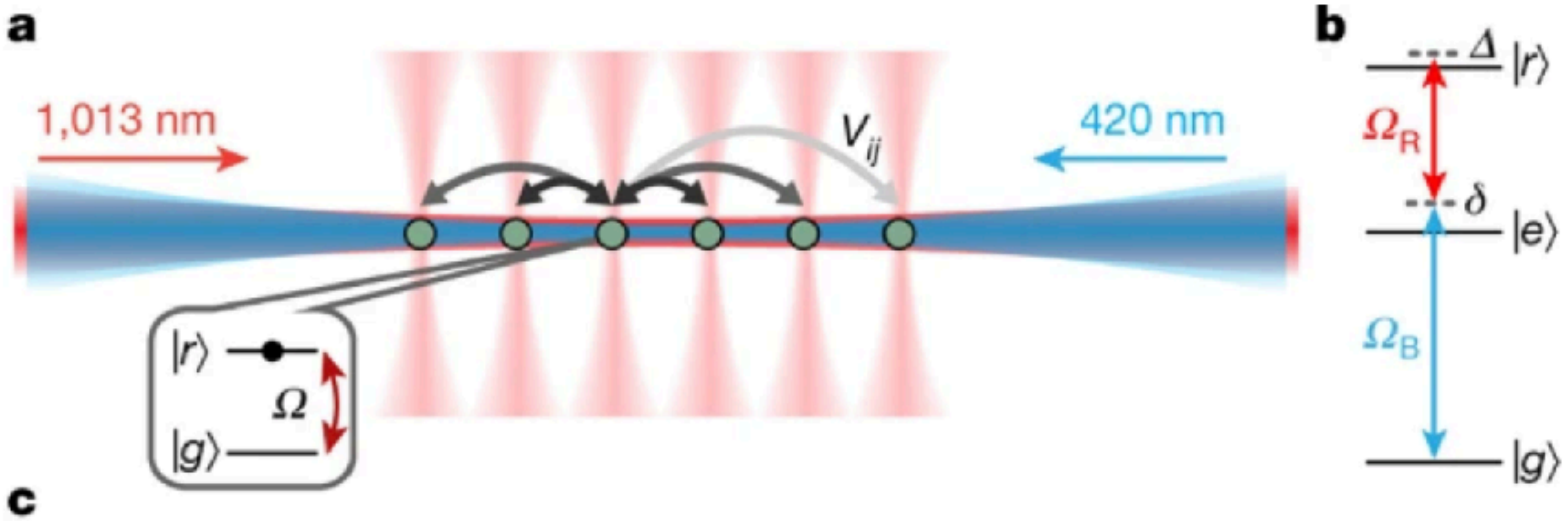




Bernien et al Nature **551** 579 (2017)

A little pre-history

What can the emission and absorption of light tell us about the structure of matter?

1868: **Ångström** publishes study of hydrogen spectrum

1885: Johann **Balmer** discovers a relationship between these observed lines.

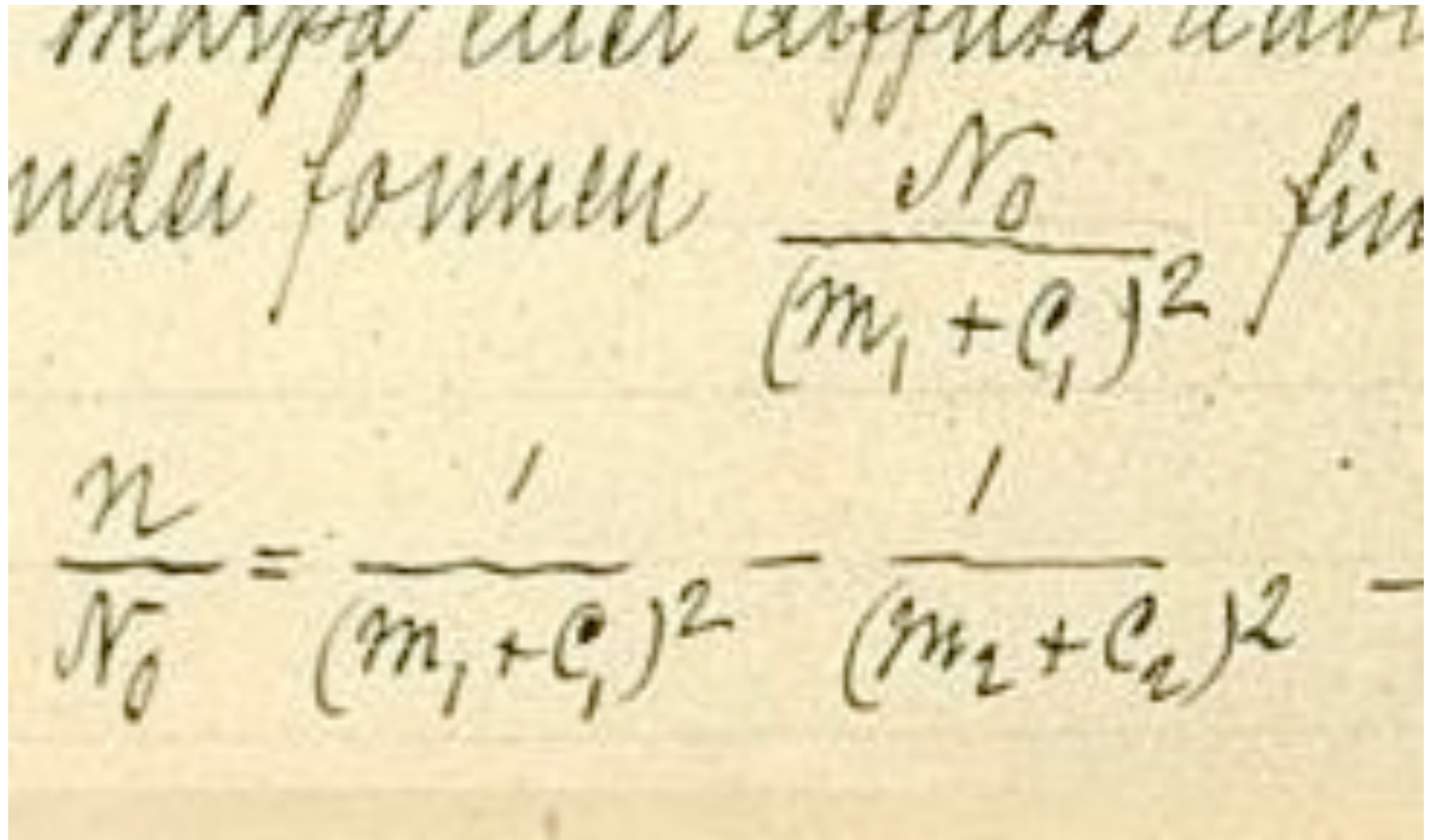
1888: Johannes **Rydberg** synthesizes empirical results, fully generalizing Balmer’s formula and kicking this all off.

1911: **Rutherford** presents his model of the atom: a compact, positive core with a cloud of electrons around it - no more plum pudding!

1913: **Bohr** and **Rutherford** present a semiclassical, “old quantum theory” argument. This fails for every atom with more than one electron.

17 January 1926: **Pauli** solves the quantum Kepler problem for the hydrogen atom.

27 January 1926: **Schrödinger** solves the quantum Kepler problem for the hydrogen atom.



our goal: to derive this formula that Rydberg figured out 30 years before quantum mechanics

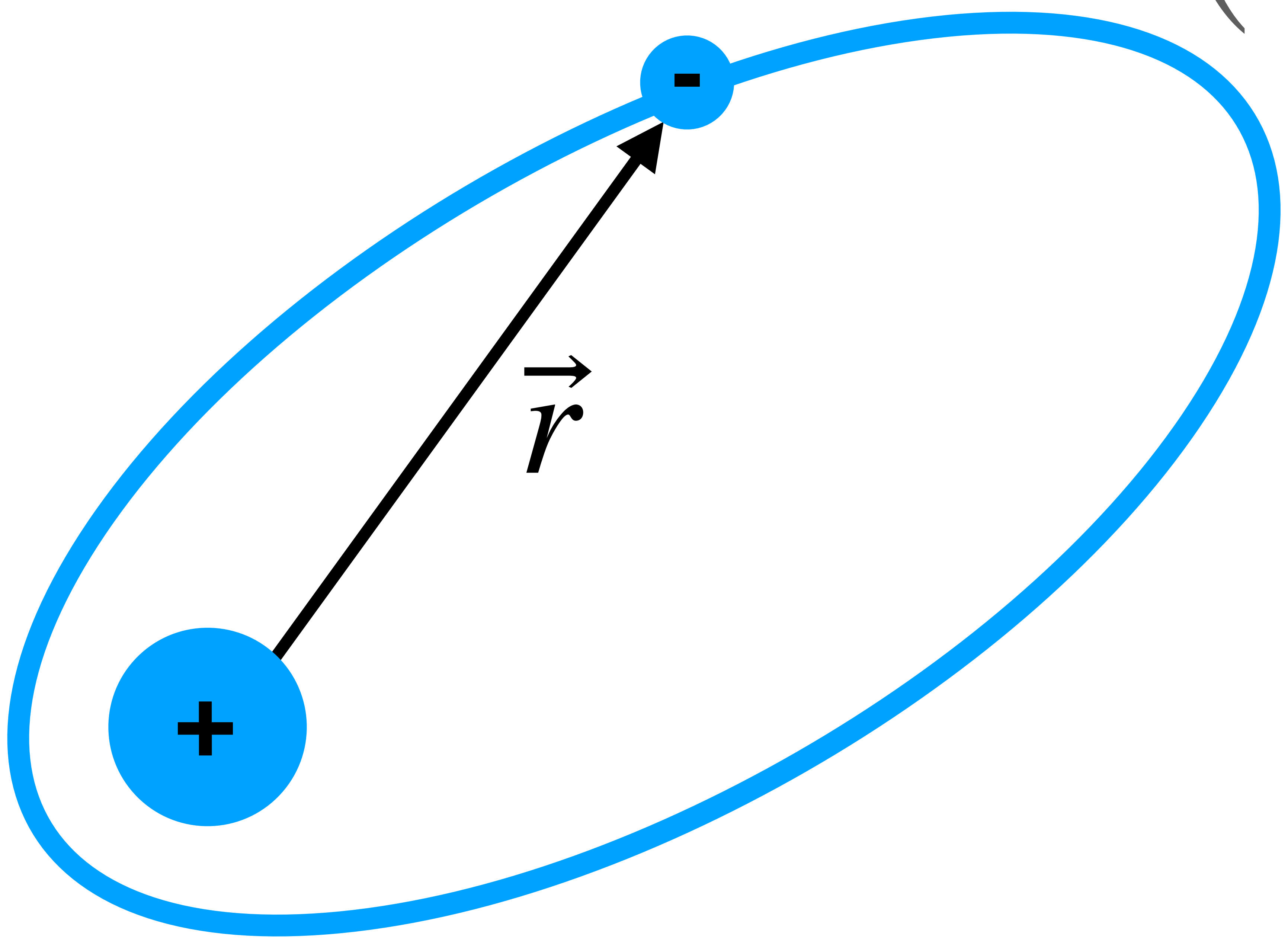
Lightning review

Before we can understand the rest of the periodic table, we need to understand H

Schrödinger equation:

$$0 = \left(-\frac{\nabla^2}{2} - \frac{1}{r} - E \right) \psi(\vec{r})$$

...in **atomic units** where $\hbar = e = m_e = 1$



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This separates in spherical coordinates (among many others - try it in parabolic coordinate in your vast spare time!)

$$\psi(\vec{r}) = \frac{u_{E\ell}(r)}{r} Y_{\ell m}(\hat{r})$$

…where ℓ and m are the eigenvalues of $\vec{L}^2$ and L_z

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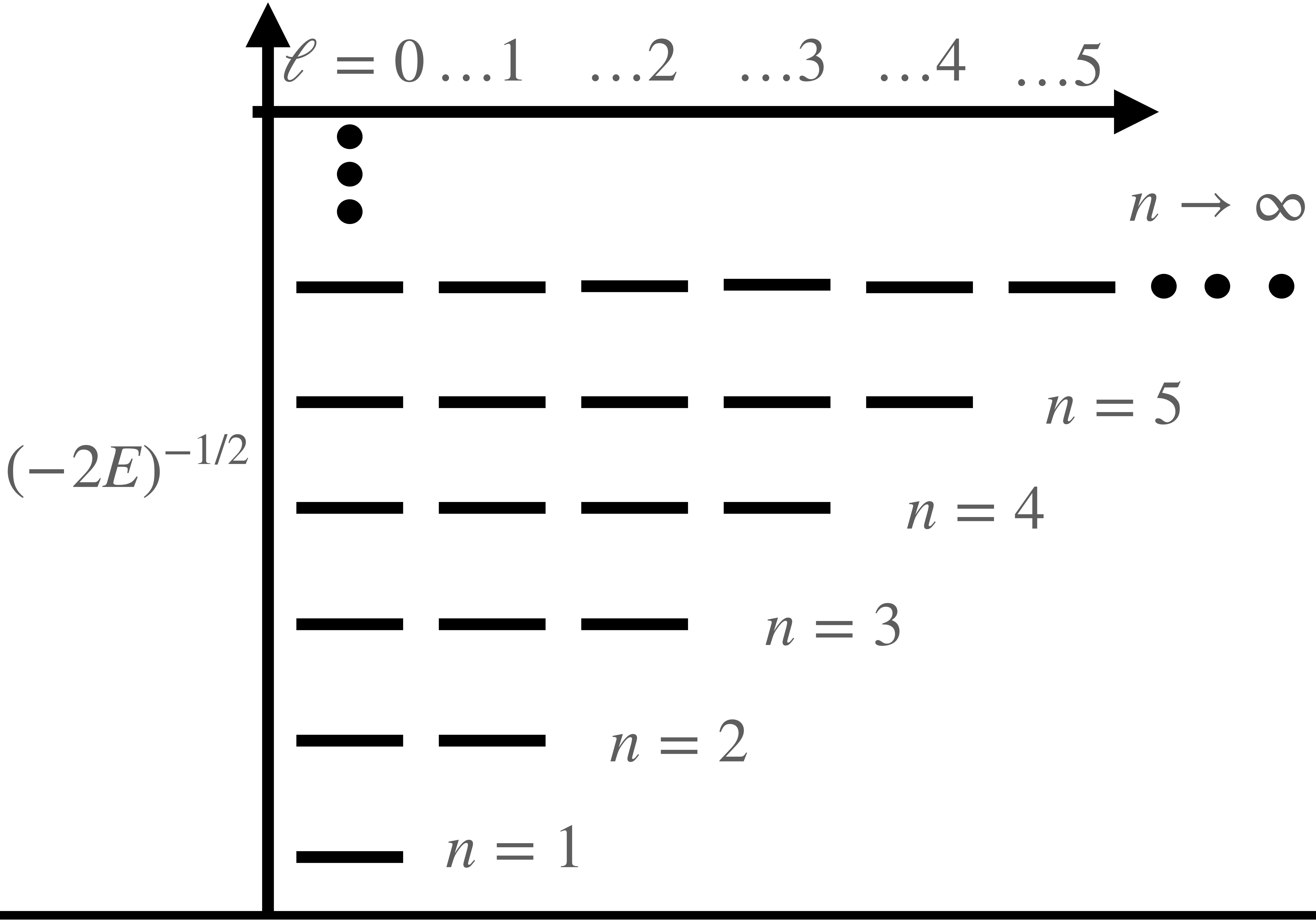
$$0 = -\frac{1}{2}u''_{E\ell}(r) + \left(\frac{\ell(\ell+1)}{2r^2} - \frac{1}{r} - E \right) u_{E\ell}(r)\,. \qquad E = -\frac{1}{2n^2},\ n > l\,.$$

...but we've all done this before!

From hydrogen atoms to Rydberg atoms

The hydrogen atom solution admits an infinite series of highly degenerate bound states

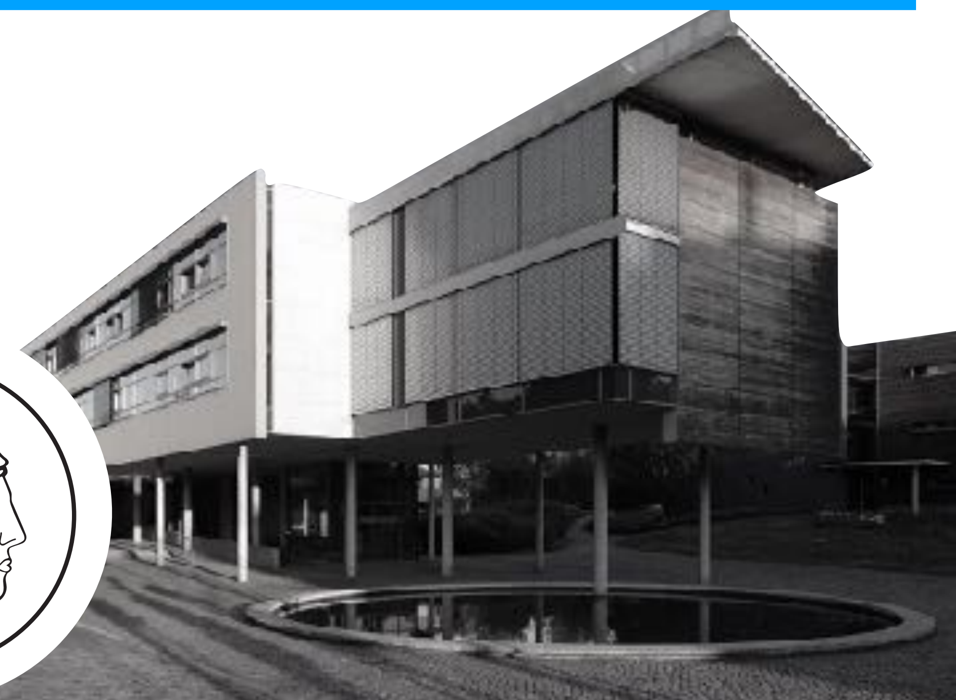


There is no restriction on n:
-infinite series of states converging to threshold at E = 0
-SO(4) symmetry: high degeneracy

Atoms with high *n* are called **Rydberg atoms**

Rydberg atoms get huge:

$$E = T + V \implies -\frac{1}{2n^2} = -\frac{1}{r_0}$$
$$\implies r_0 = 2n^2$$



A little bit of motivation

Why should you care about Rydberg physics?

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- **Fundamental**:
  - beyond a "measure zero" set of ground states, all spectroscopy is Rydberg physics.
  - Quantum-classical correspondence, chaos and quantum scarring

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- **Fundamental**:
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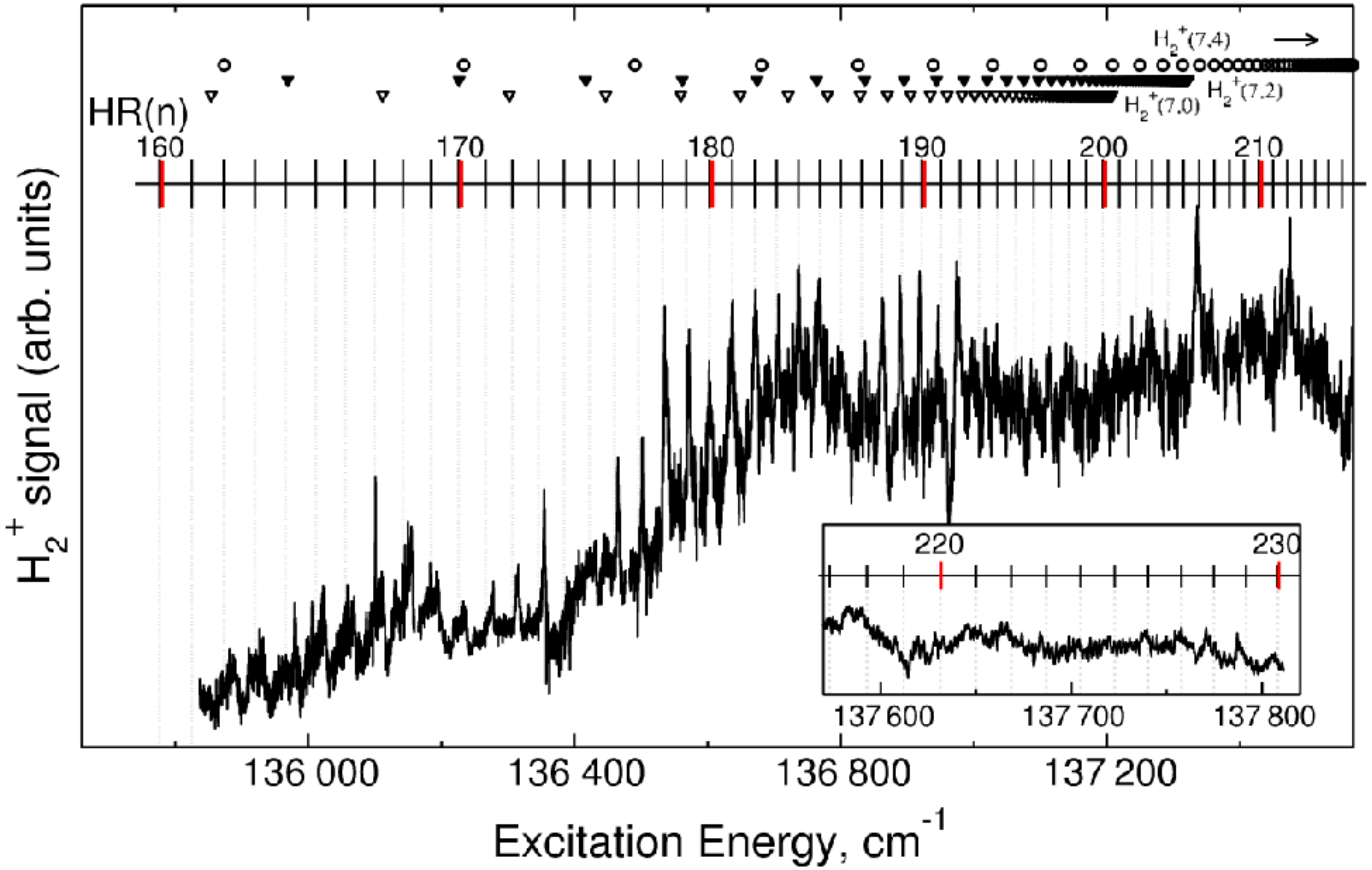
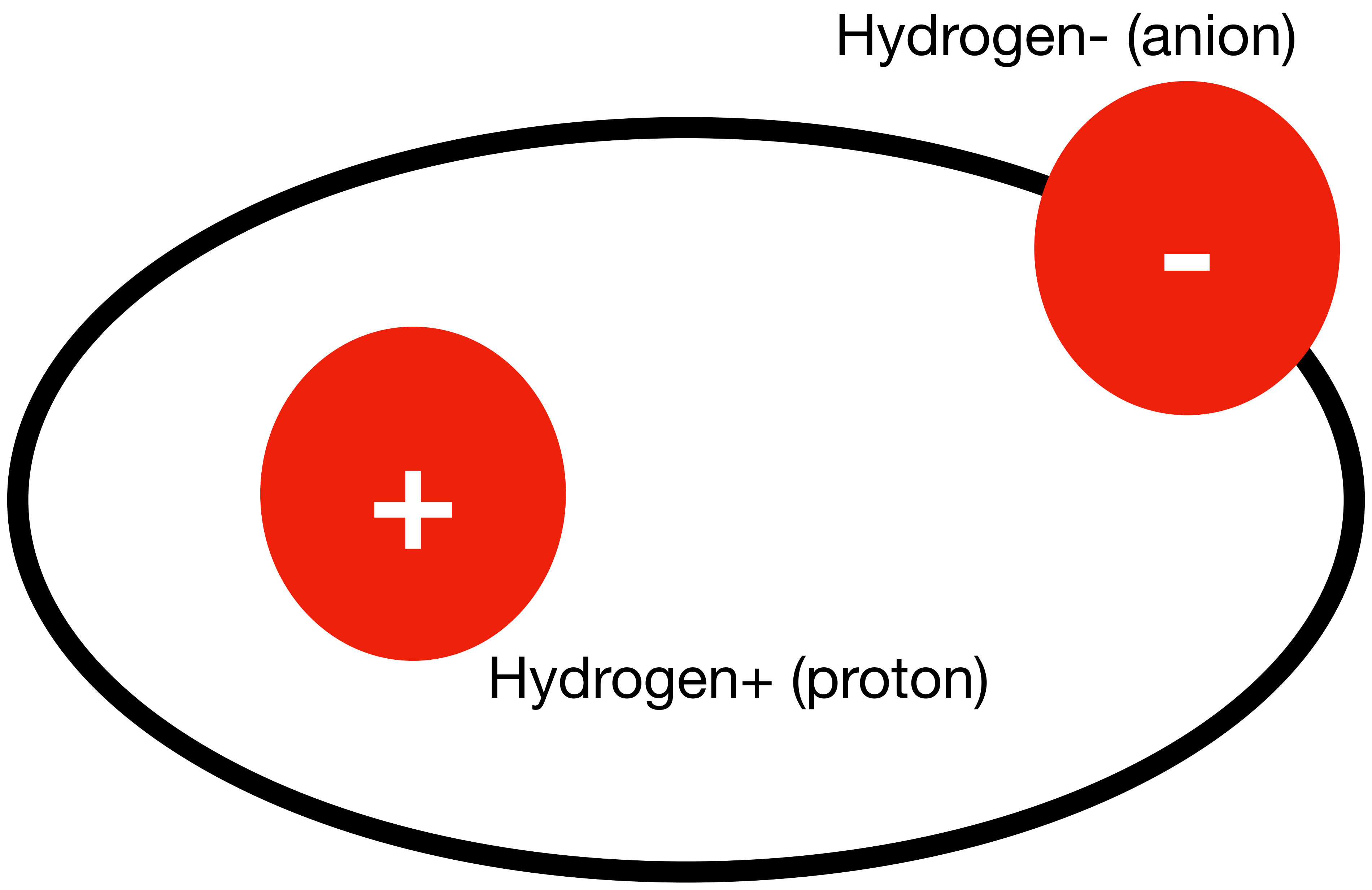
  - Quantum-classical correspondence, chaos and quantum scarring
- **Useful**:
  - sensing (highly responsive to external fields)
  - quantum computing
  - quantum simulation
  - quantum optics
  - many-body quantum scars
- **Versatile**
  - "Universal"

Exotic Rydberg systems

Heavy Rydberg states - atom-like molecules

- extremely different size / energy scales
- possible initial state for producing equal-mass plasmas



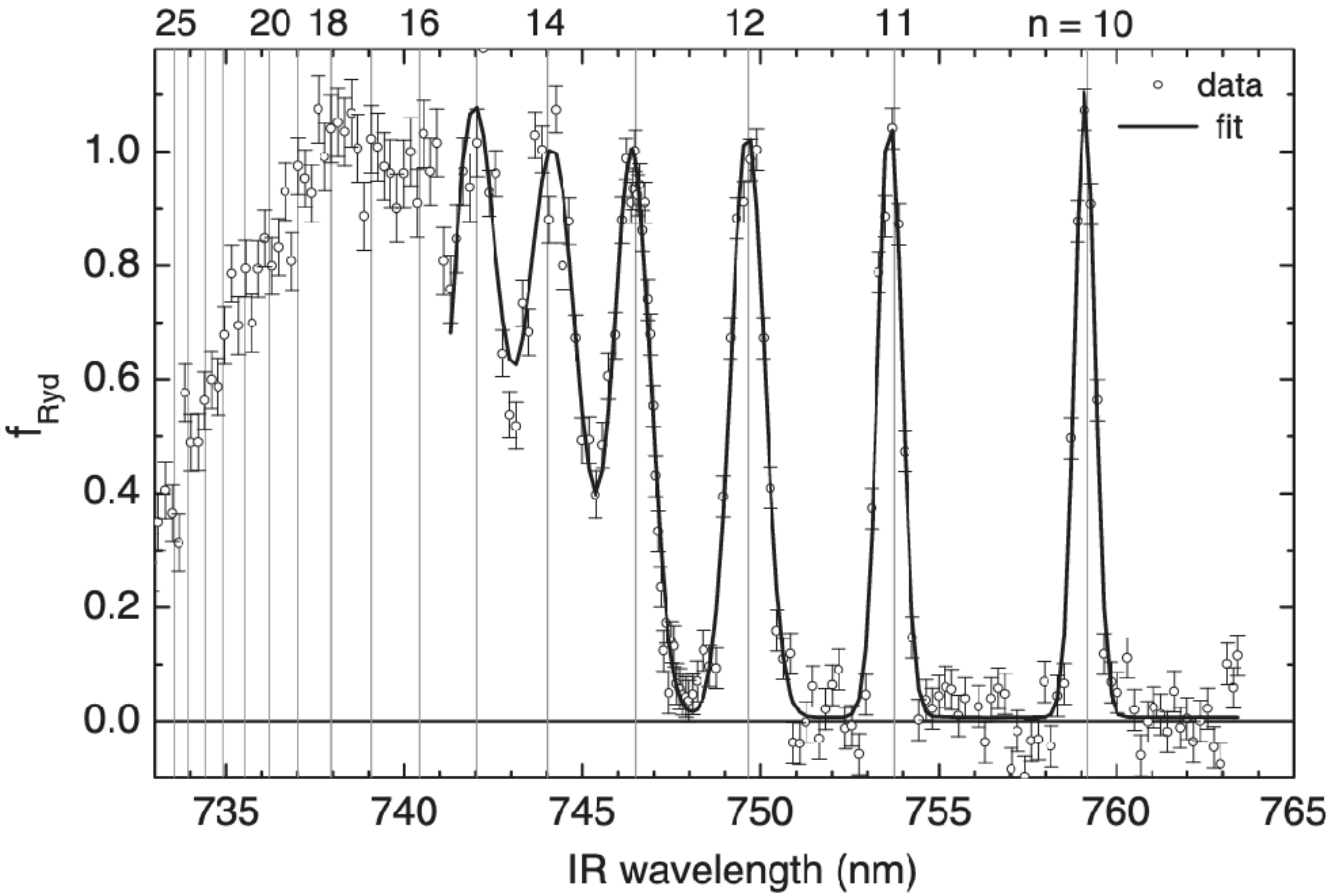


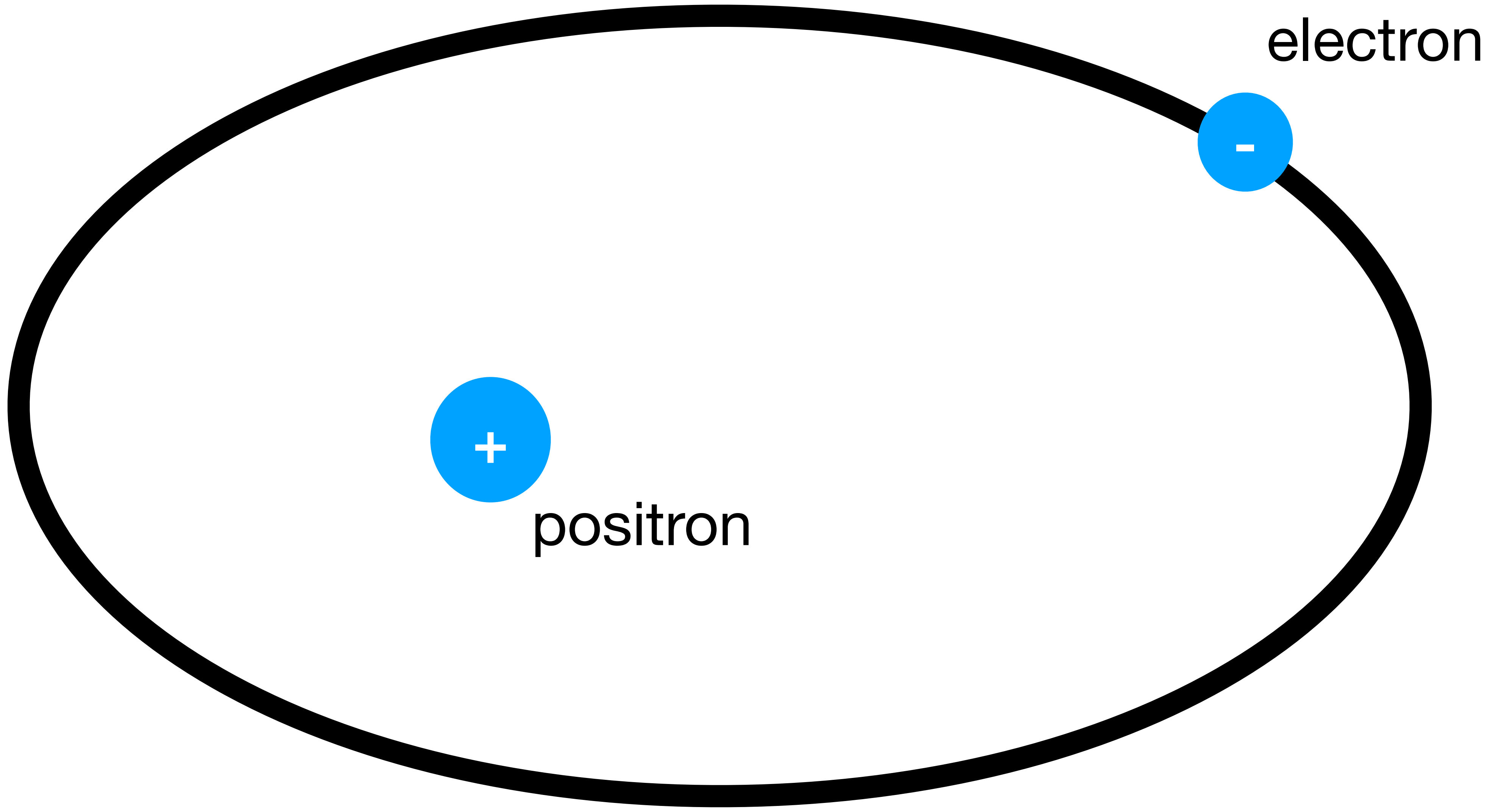
Vieitez et al PRL **101** 163001 (2008)

Exotic Rydberg systems

Rydberg positronium - long-lived matter/antimatter

- Annihilation of ground state Ps occurs $< 10^{-7}$ s. Rydberg states live $> 1^{-5}$ s.
- Precision QED or gravity tests



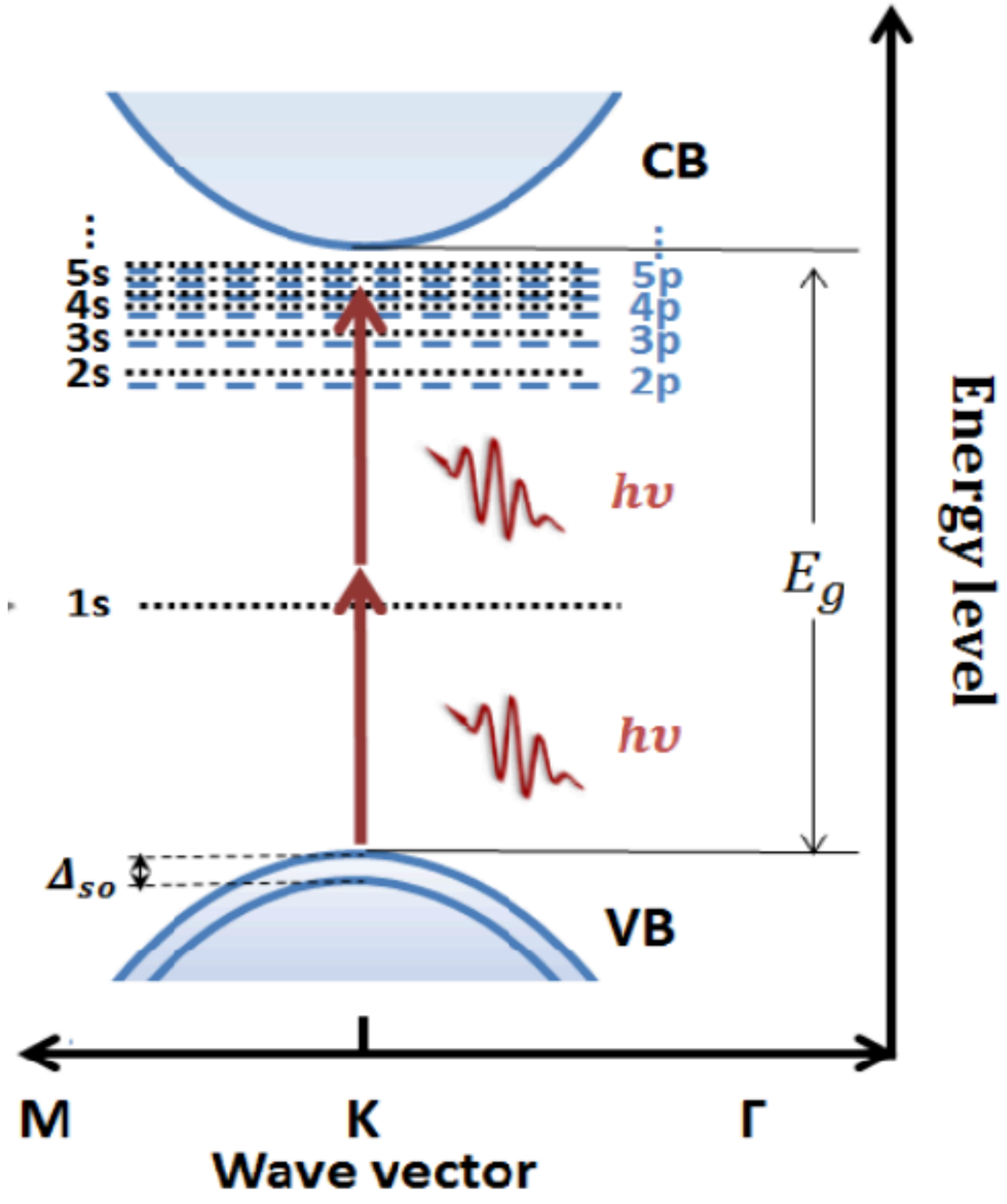


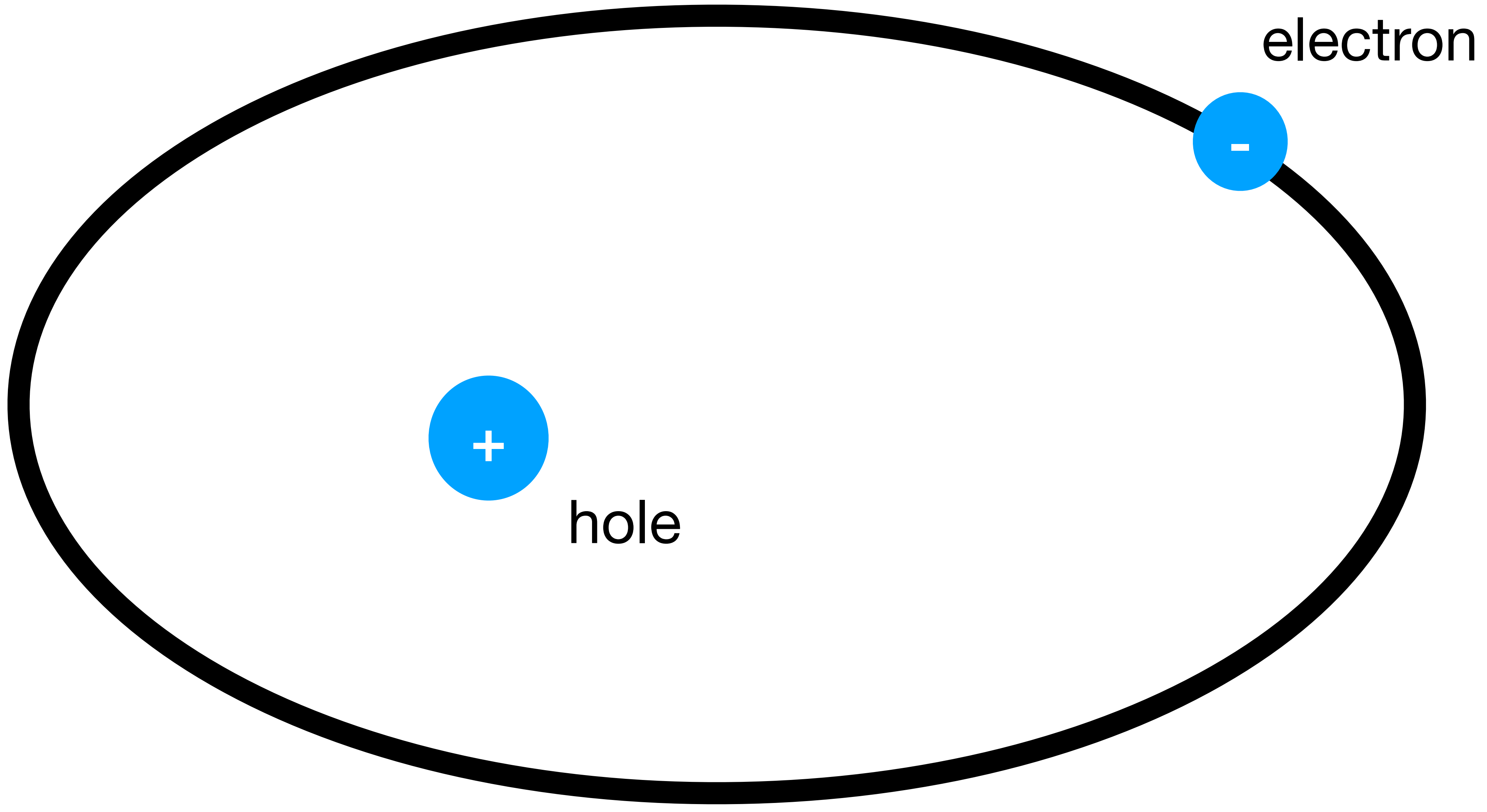
Cassidy et al PRL **108** 043401 (2012)

Exotic Rydberg systems

Rydberg excitons - bound electron-hole pairs in materials.

- particle + quasiparticle
- Not perfectly spherically symmetric - still living in a lattice!





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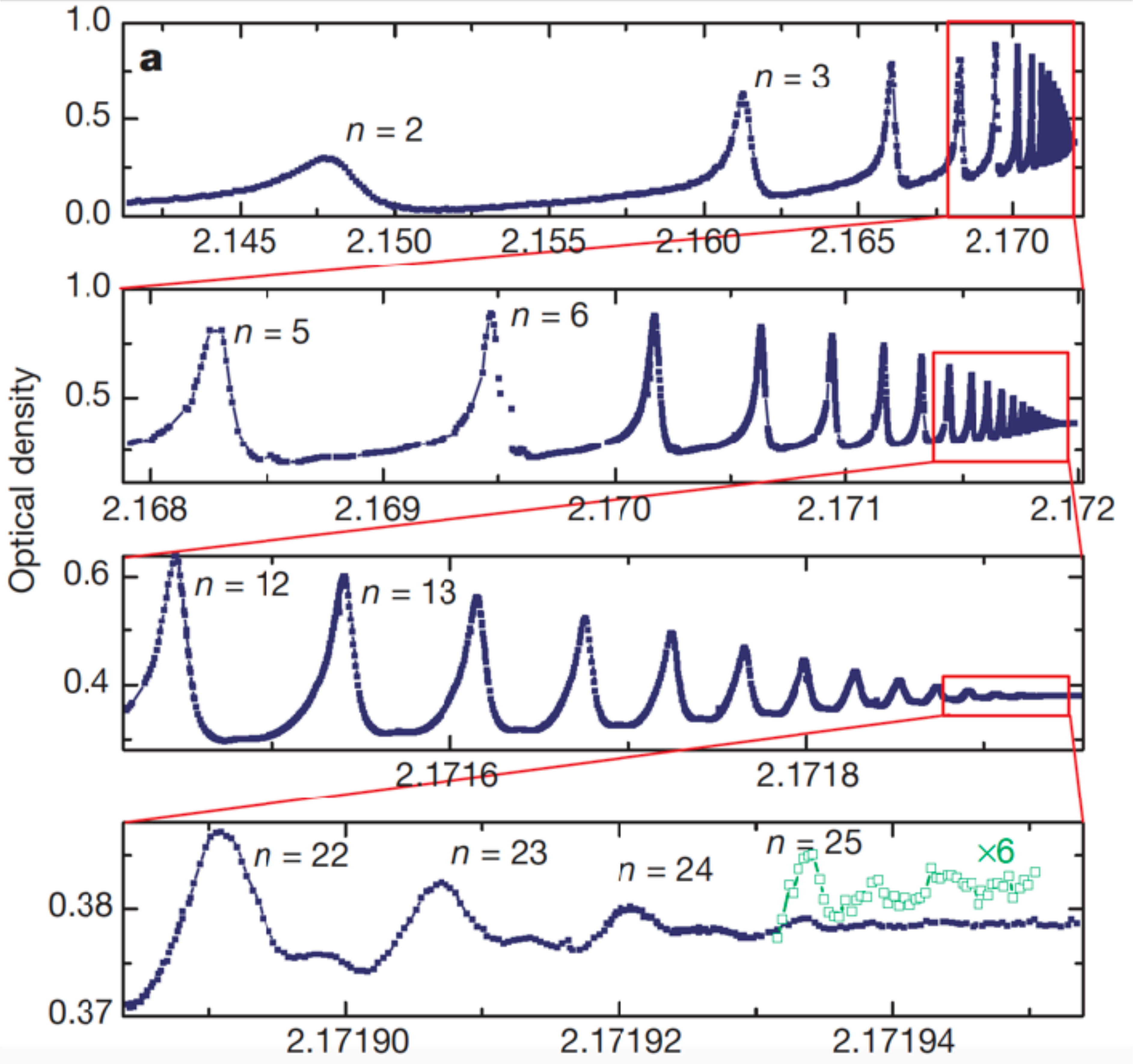
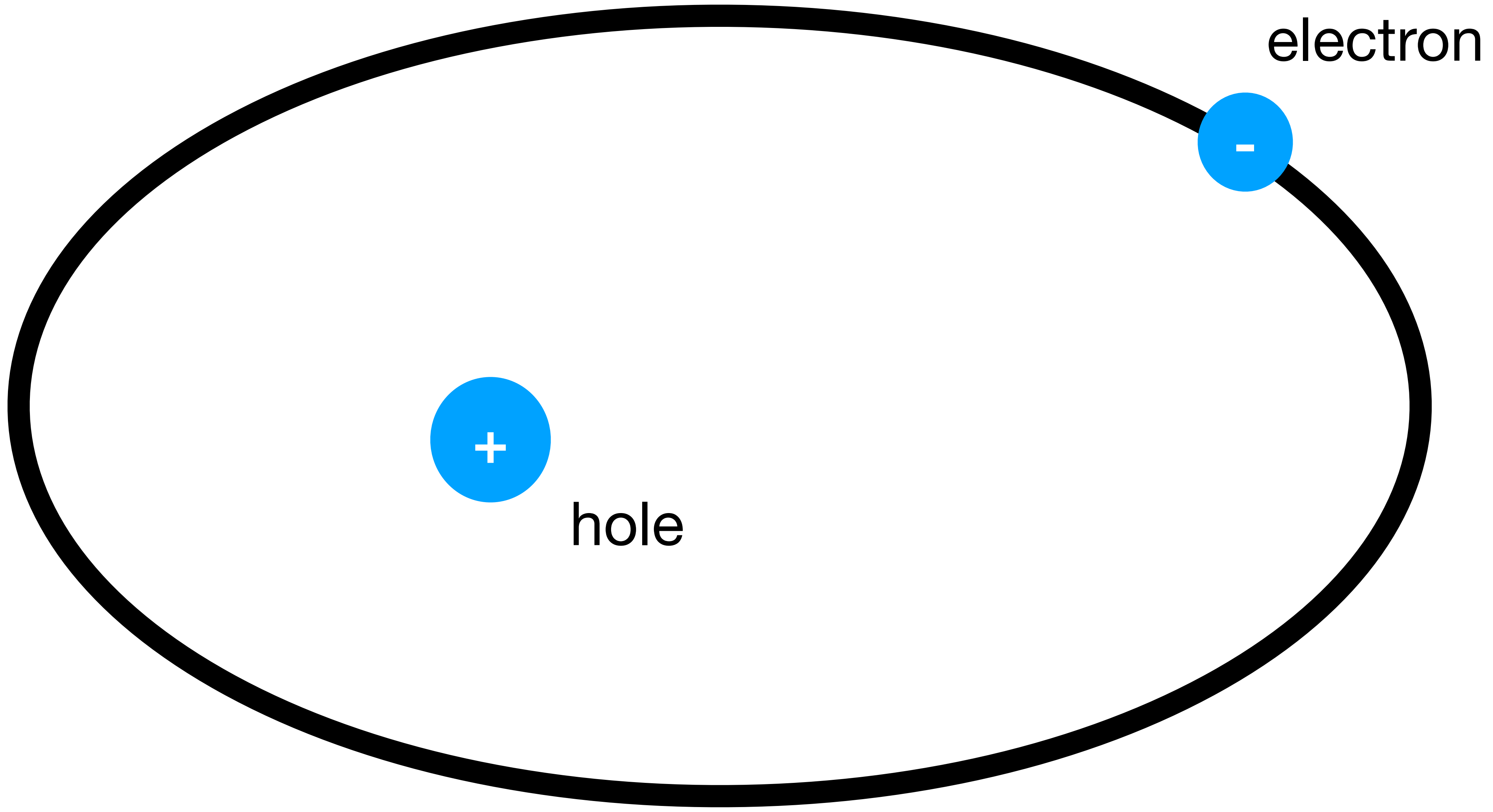


photon energy (eV)



Kazimierczuk et al Nature **514** 343 (2014)

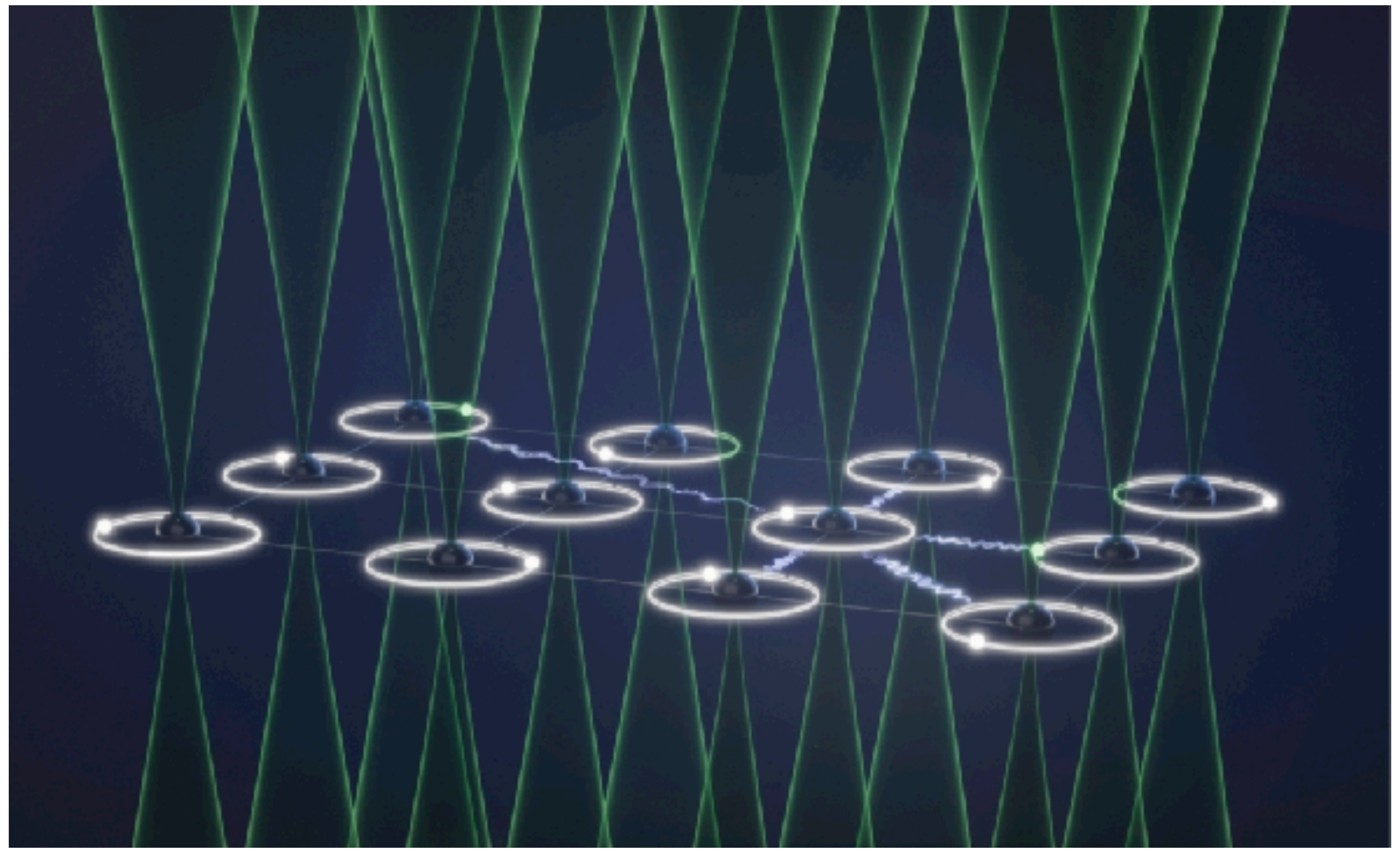
Exotic Rydberg systems

Rydberg molecules

- Frederic Merkt (ETH) / Tilman Pfau (Stuttgart) / Ed Grant (UBC) / Stephen Hogan (UCL) + more…

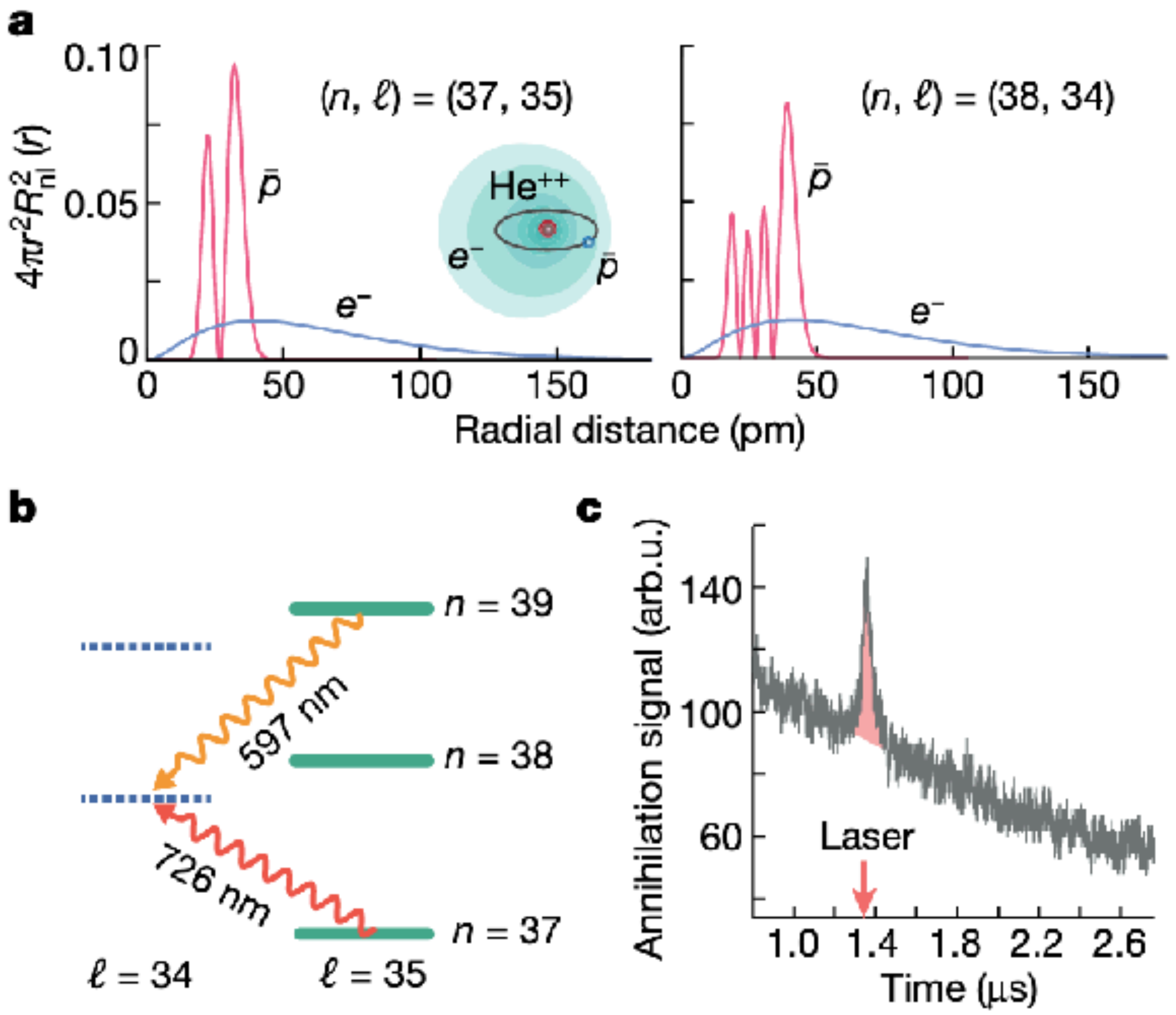
Circular Rydberg states

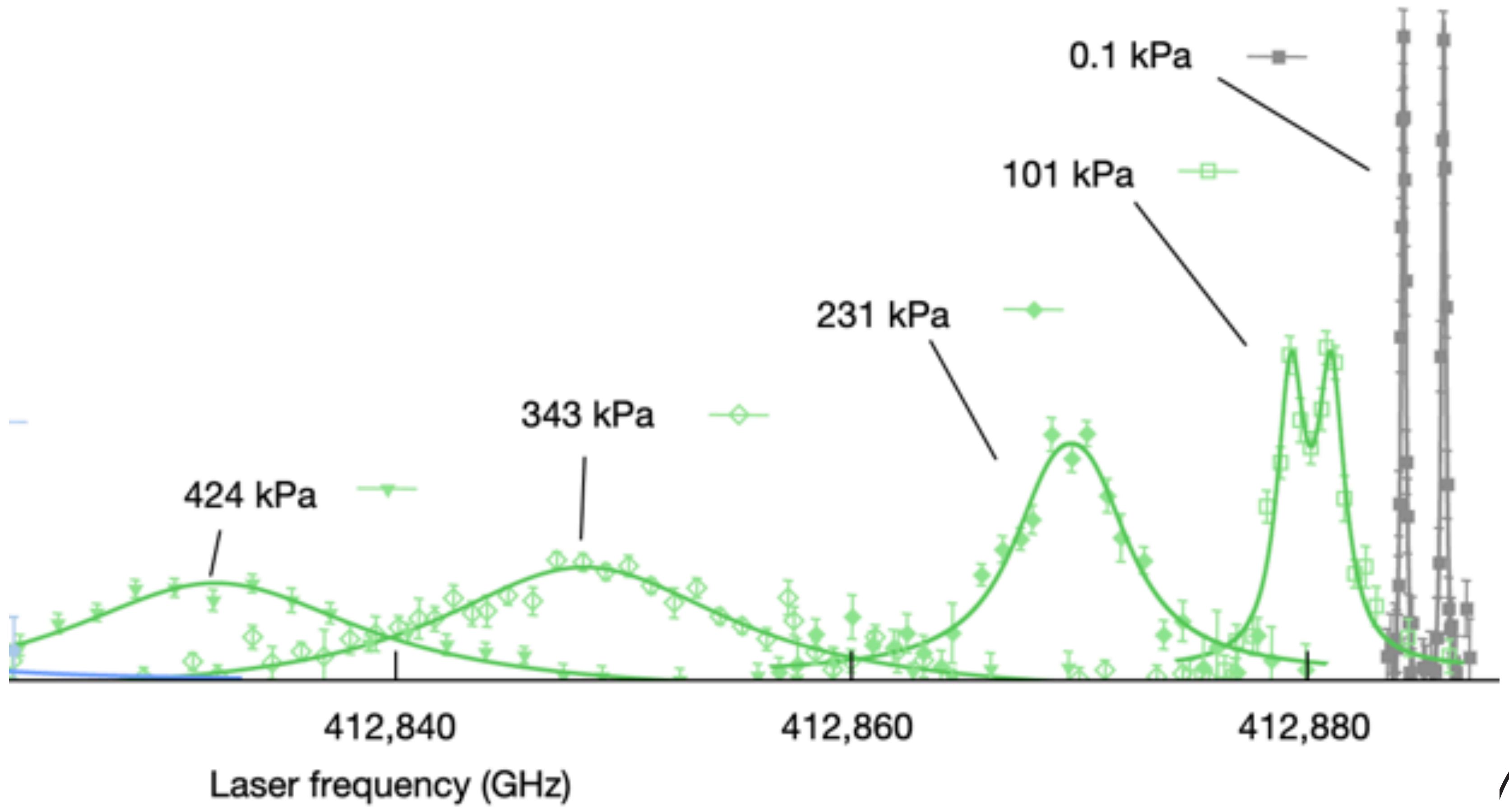
- Michel Brune (CNRS) / Florian Meinert (Stuttgart) + more…
- atoms with $l = m = n - 1$



Circular Rydberg states + antimatter + matter + ….

- Sótér et al Nature **603** 411 (2022)



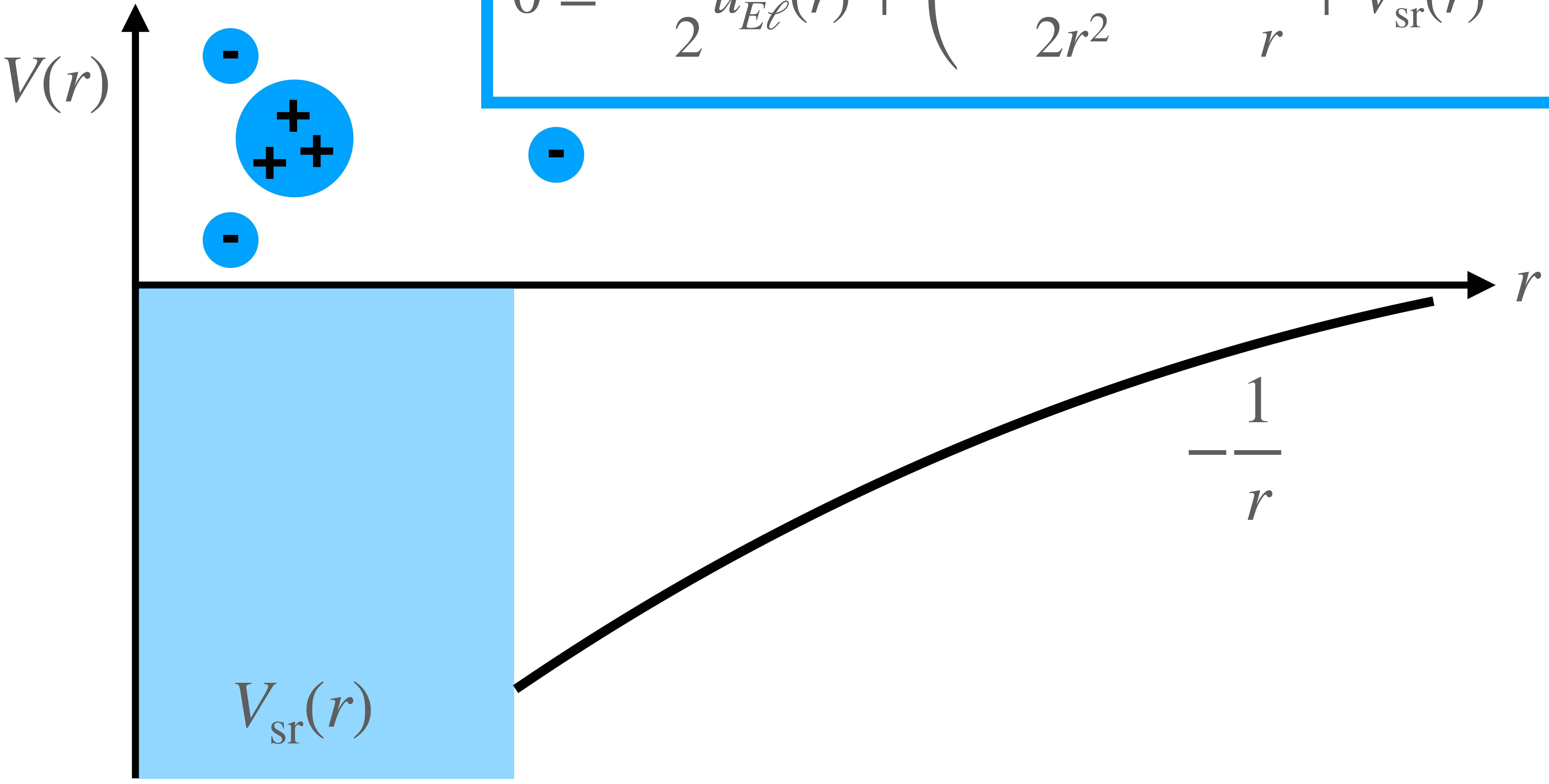


Rydberg transitions probe background density

How to go beyond hydrogen

"All" we have to do is to solve the radial equation in each angular momentum channel:

$$0 = -\frac{1}{2}u''_{E\ell}(r) + \left(\frac{\ell(\ell+1)}{2r^2} - \frac{1}{r} + V_{\rm sr}(r) - E\right)u_{E\ell}(r)\,.$$



Complicated… Coulomb…

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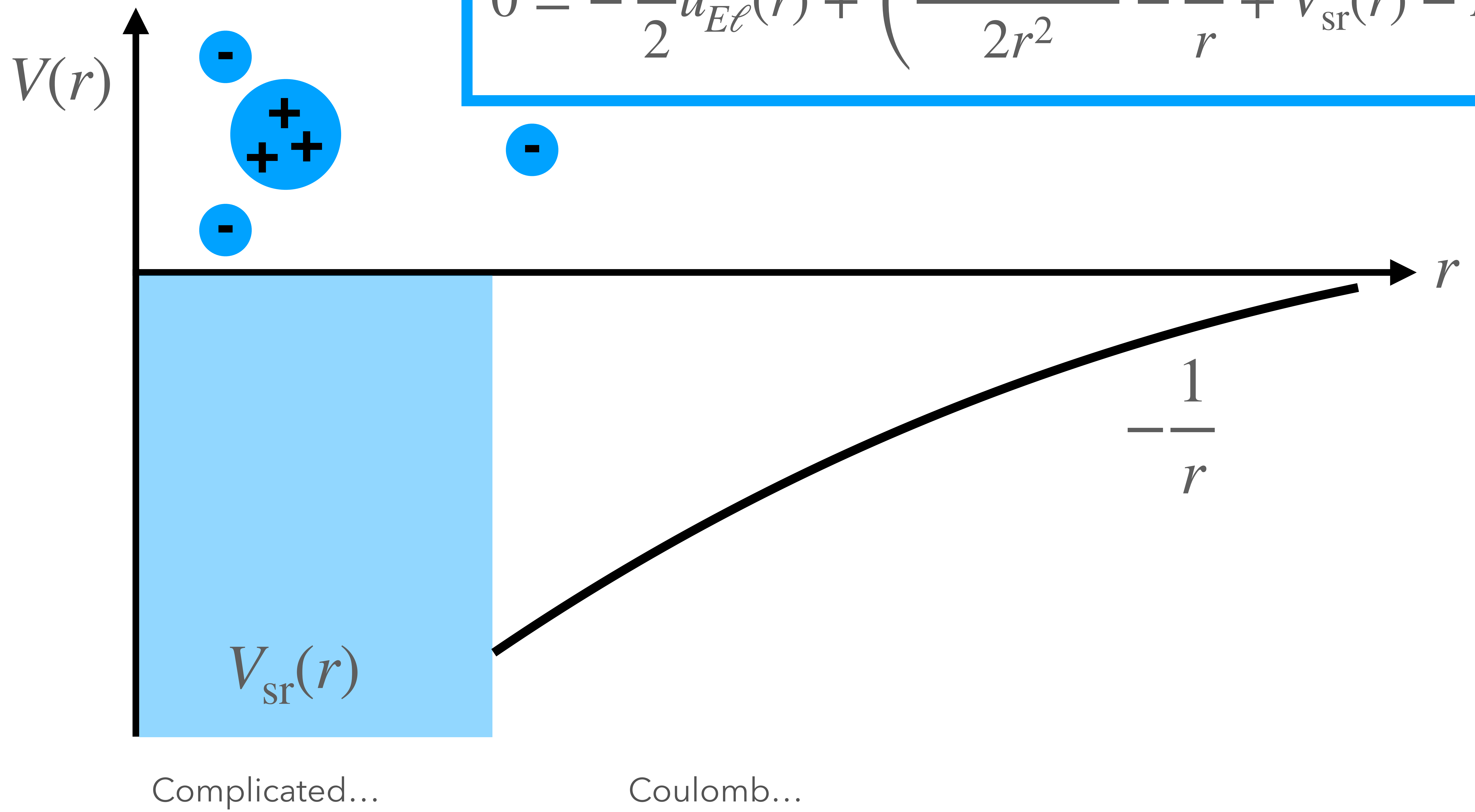

QUANTUM DEFECT THEORY:
A powerful framework for analyzing these (and much more complex) problems, built on two realizations:

- At large r we *know* the solution to this problem.
- At small r the solution is *nearly energy-independent*.

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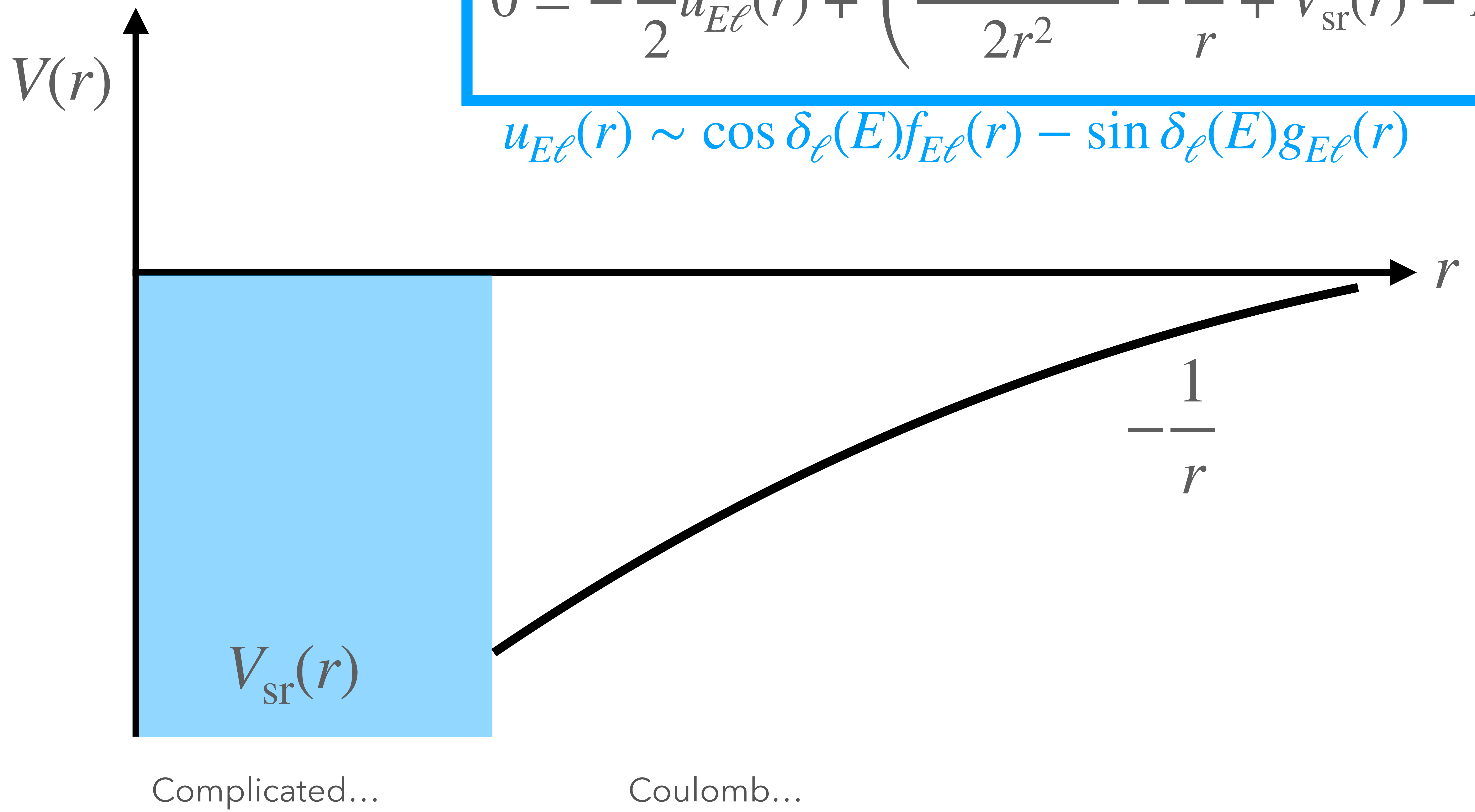

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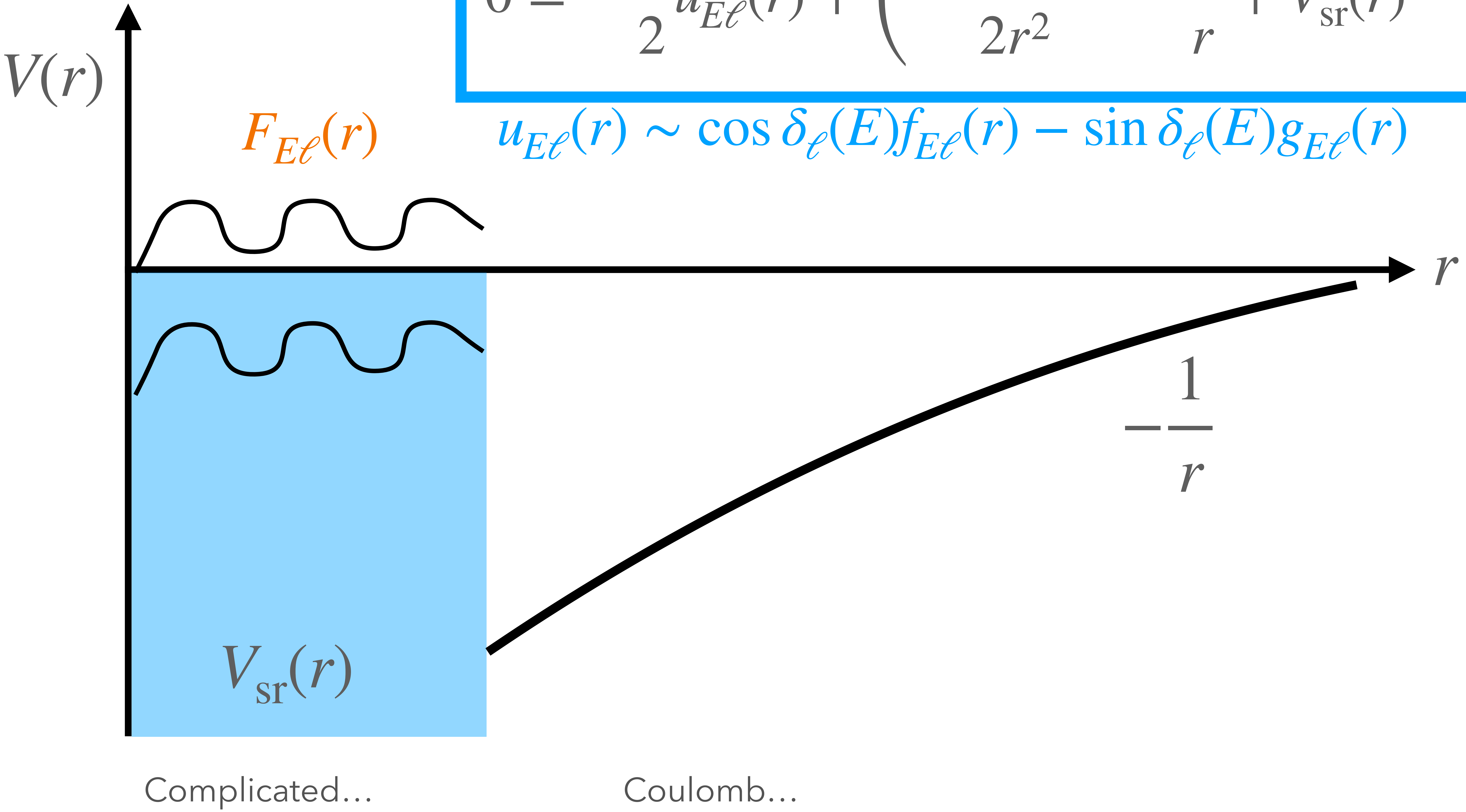

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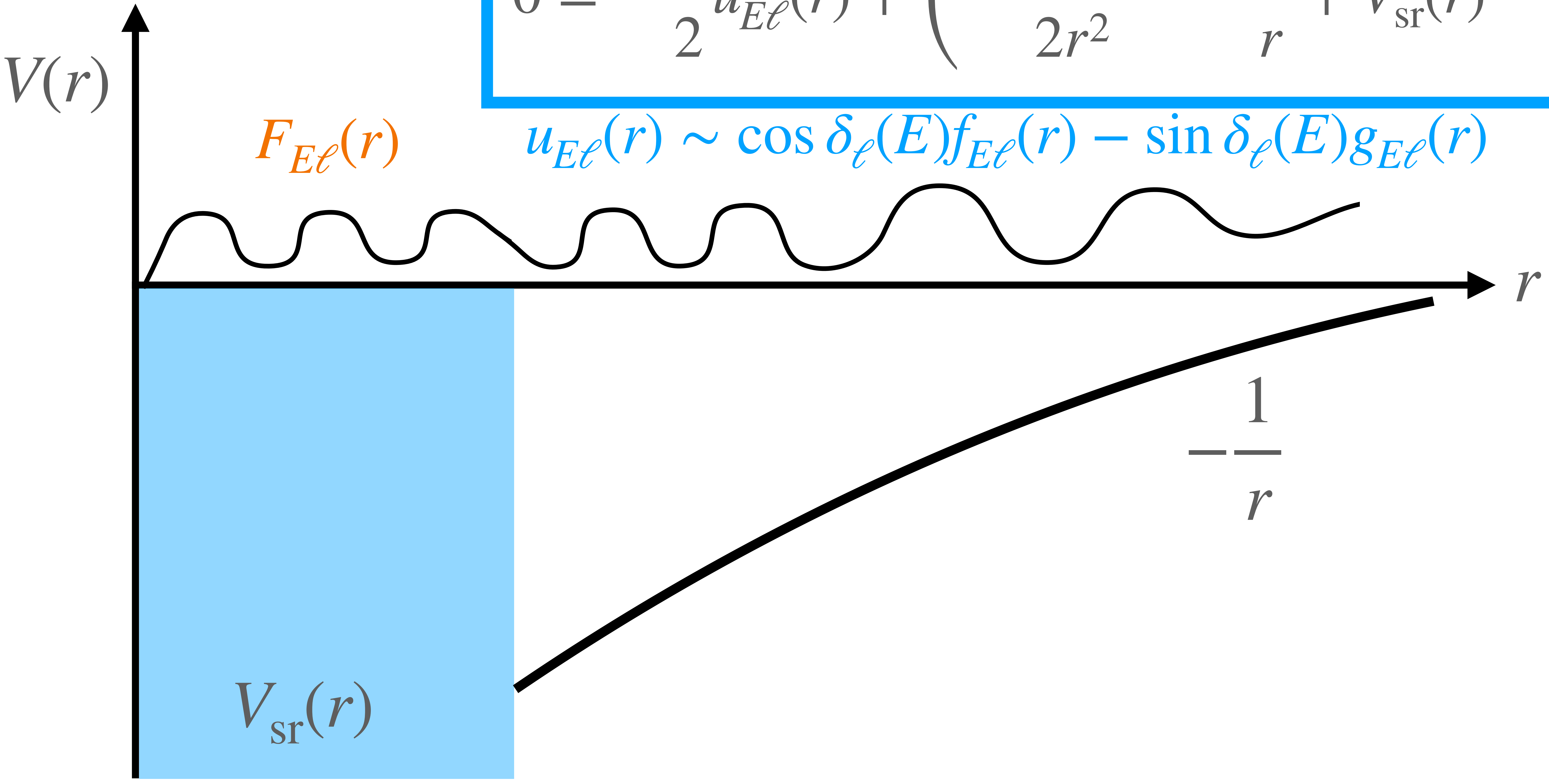

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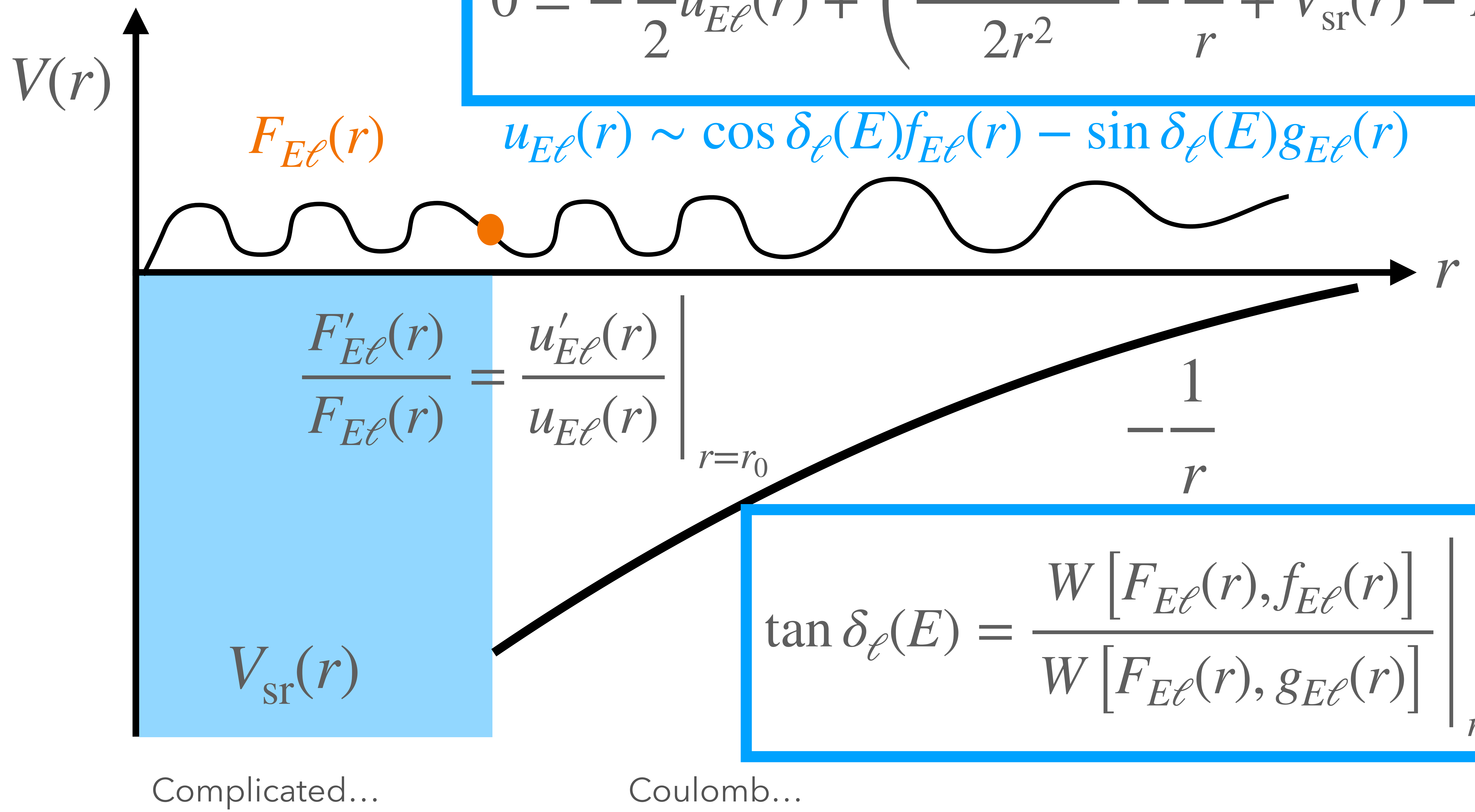

$$u_{E\ell}(r) \sim \cos\delta_\ell(E) f_{E\ell}(r) - \sin\delta_\ell(E) g_{E\ell}(r)$$

$$\frac{F'_{E\ell}(r)}{F_{E\ell}(r)} = \frac{u'_{E\ell}(r)}{u_{E\ell}(r)}\bigg|_{r=r_0}$$

$$\tan\delta_\ell(E) = \frac{W\left[F_{E\ell}(r), f_{E\ell}(r)\right]}{W\left[F_{E\ell}(r), g_{E\ell}(r)\right]}\bigg|_{r=r_0}$$

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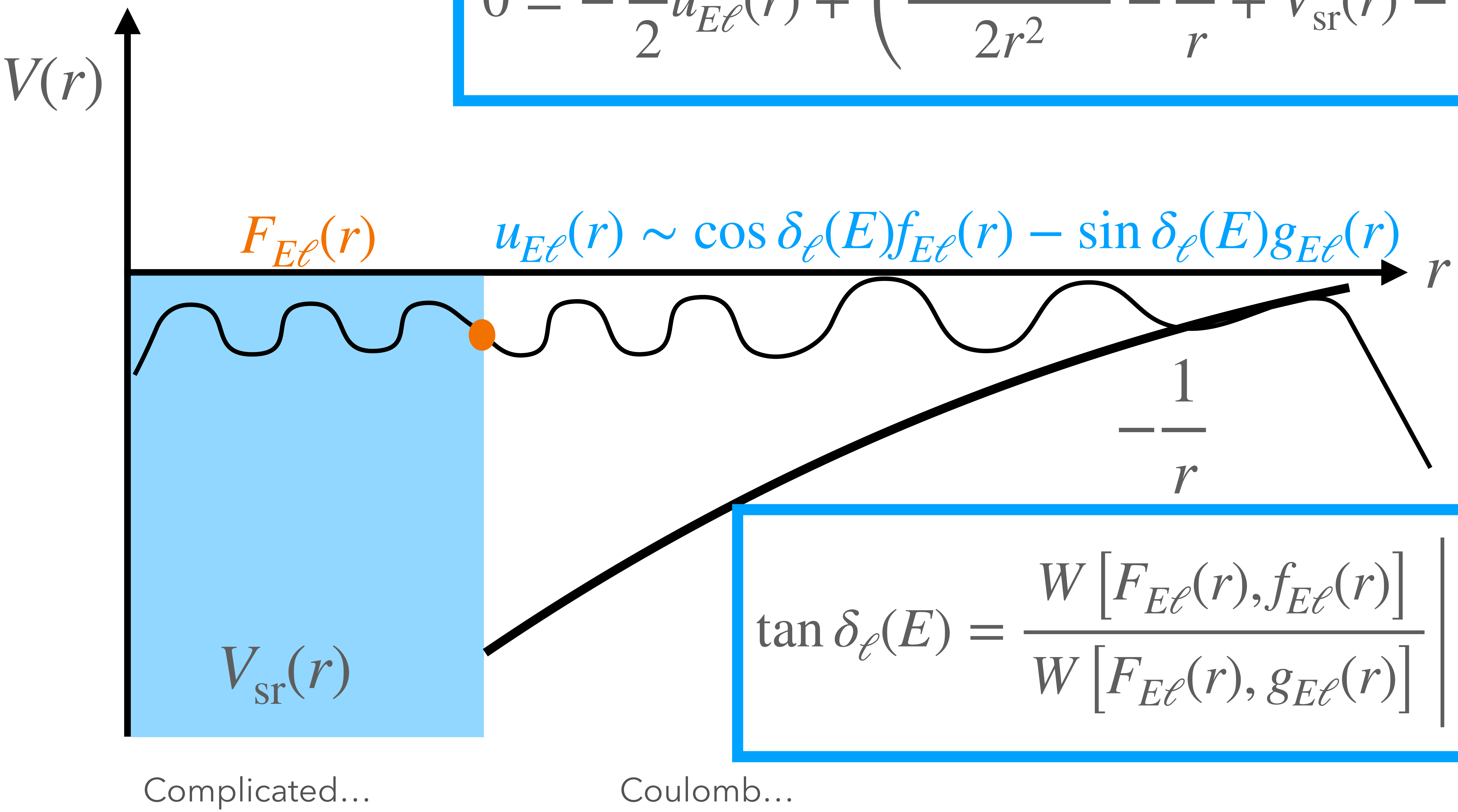

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A few formulas…

First - analytically continue to negative energies.
Second - obtain asymptotic expansions:

EVERY formula here assumes the limit $r \to \infty$

$$f_{E\ell}(r) \to Ar^{-\nu}e^{r/\nu}\sin\pi(\nu-\ell) - Br^{\nu}e^{-r/\nu}\cos\pi(\nu-\ell)$$

$$g_{E\ell}(r) \to -Ar^{-\nu}e^{r/\nu}\cos\pi(\nu-\ell) - Br^{\nu}e^{-r/\nu}\sin\pi(\nu-\ell)$$

$$E = -\frac{1}{2\nu^2}$$

(A and B are constants)

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This means that at an arbitrary energy our solution blows up **exponentially** at infinity (even single-particle quantum physics has problems with exponential growth!)

$$u_{E\ell}(r) \sim \cos\delta_\ell(E) f_{E\ell}(r) - \sin\delta_\ell(E) g_{E\ell}(r)$$

$$\sim Ar^{-\nu}e^{r/\nu}\left[\cos\delta_\ell(E)\sin\pi(\nu-\ell) + \sin\delta_\ell(E)\cos\pi(\nu-\ell)\right] + \mathcal{O}(e^{-r/\nu})$$

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$$u_{E\ell}(r) \sim A r^{-\nu} e^{r/\nu} \sin \pi \left[\nu - \ell + \frac{\delta_\ell(E)}{\pi}\right]$$

$$\implies \nu - \ell + \frac{\delta_\ell(E)}{\pi} = n_r$$

A few formulas…

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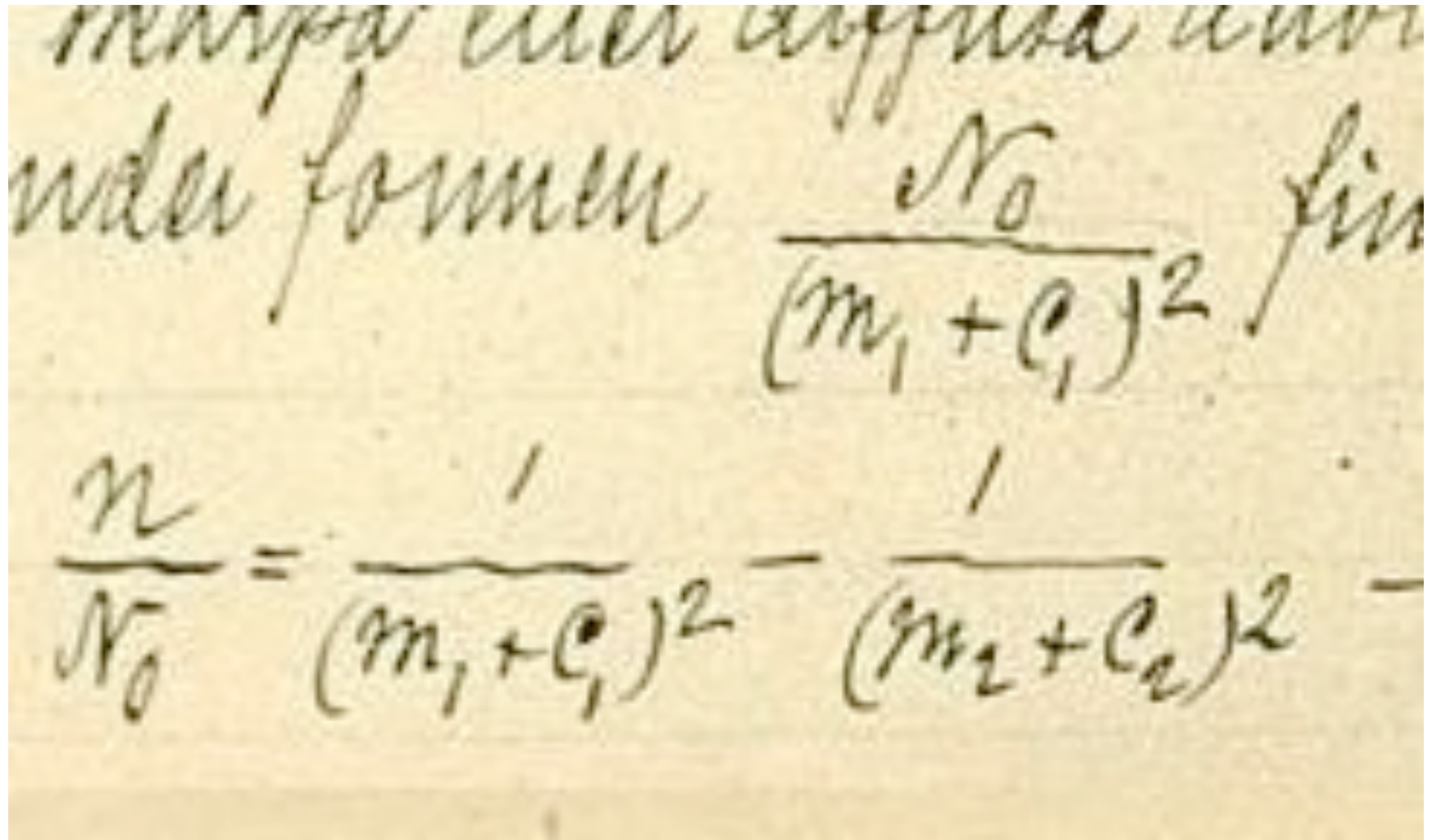
$$E_\ell = -\frac{1}{2(n-\mu_l)^2}$$

Where μ_l is the *quantum defect!*



(we did it!)

Some takeaways of quantum defect theory

KEY POINT #1: At sufficiently large r we have an *analytically solved problem*

KEY POINT #2: At small r the physics is nearly independent of energy

QDT **does not discriminate** between scattering physics (collisions) and bound state physics (spectroscopy) - this lets us describe a whole bunch of physics in a large energy range with just a **few parameters.**

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Table 2. Measured frequencies for the $nP_{3/2}$ states and respective quantum defects. E_n is measured from the centre of mass of the lower and upper states and contains a small correction to the wavemeter calibration. The third step data are reported exactly as measured.

n	Third step (MHz)	E_n (MHz)	δ	δ Error ($\times 10^{-5}$)
36	236 496 706	1007 068 254	2.641 87	2.3
37	236 666 310	1007 237 858	2.641 79	2.5
38	236 821 728	1007 393 277	2.641 70	2.7
39	236 964 479	1007 536 027	2.641 75	2.9
40	237 095 926	1007 667 475	2.641 77	3.2
41	237 217 235	1007 788 783	2.641 73	3.4
42	237 329 406	1007 900 954	2.641 76	3.7
43	237 433 360	1008 004 909	2.641 62	4.0
44	237 529 853	1008 101 402	2.641 60	4.3
45	237 619 595	1008 191 144	2.641 56	4.6
46	237 703 191	1008 274 740	2.641 63	5.0
47	237 781 211	1008 352 760	2.641 51	5.3
48	237 854 117	1008 425 666	2.641 54	5.7
49	237 922 362	1008 493 911	2.641 48	6.1
50	237 986 322	1008 557 870	2.641 55	6.5
51	238 046 352	1008 617 901	2.641 67	6.9
52	238 102 791	1008 674 339	2.641 44	7.3
53	238 155 879	1008 727 427	2.641 61	7.8
54	238 205 906	1008 777 455	2.641 59	8.2
55	238 253 103	1008 824 651	2.641 39	8.7
56	238 297 662	1008 869 210	2.641 39	9.2
57	238 339 780	1008 911 329	2.641 48	9.8
58	238 379 637	1008 951 185	2.641 58	10.3
59	238 417 400	1008 988 949	2.641 41	10.9
60	238 453 197	1009 024 746	2.641 51	11.5
61	238 487 172	1009 058 721	2.641 51	12.1
62	238 519 445	1009 090 994	2.641 51	12.7
63	238 550 123	1009 121 672	2.641 65	13.4

J. Phys. B: At. Mol. Opt. Phys. 42 (2009) 165004 (6pp) doi:10.1088/0953-4075/42/16/165004

Precision measurements of quantum defects in the $nP_{3/2}$ Rydberg states of ^{85}Rb

B Sanguinetti, H O Majeed, M L Jones and B T H Varcoe

School of Physics and Astronomy, University of Leeds, Leeds, LS2 9JT, UK

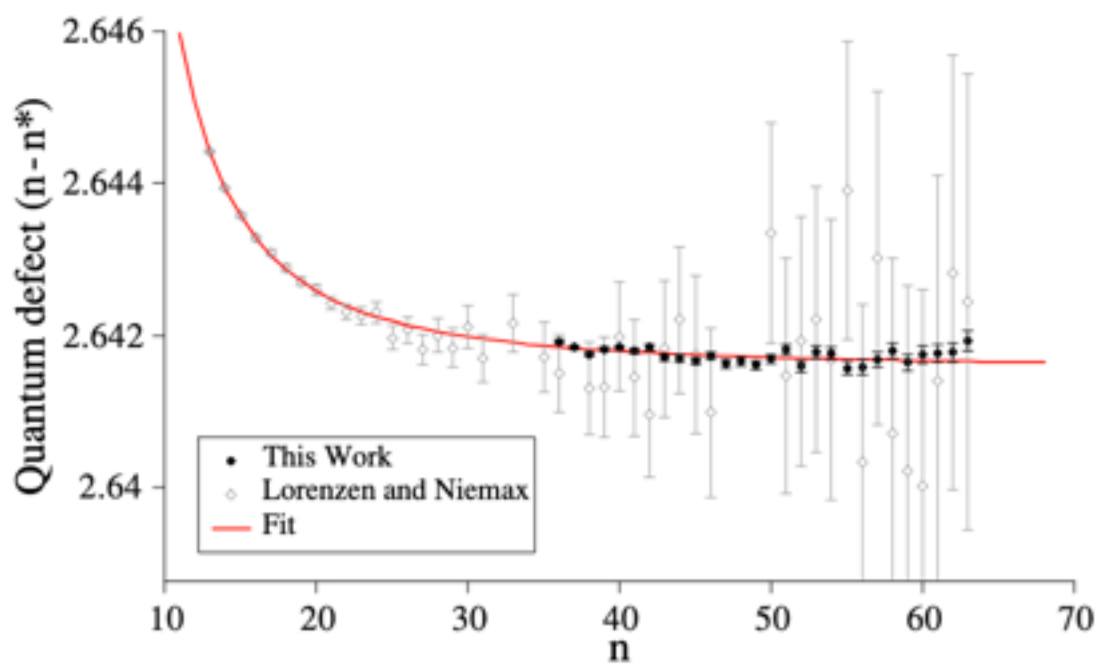
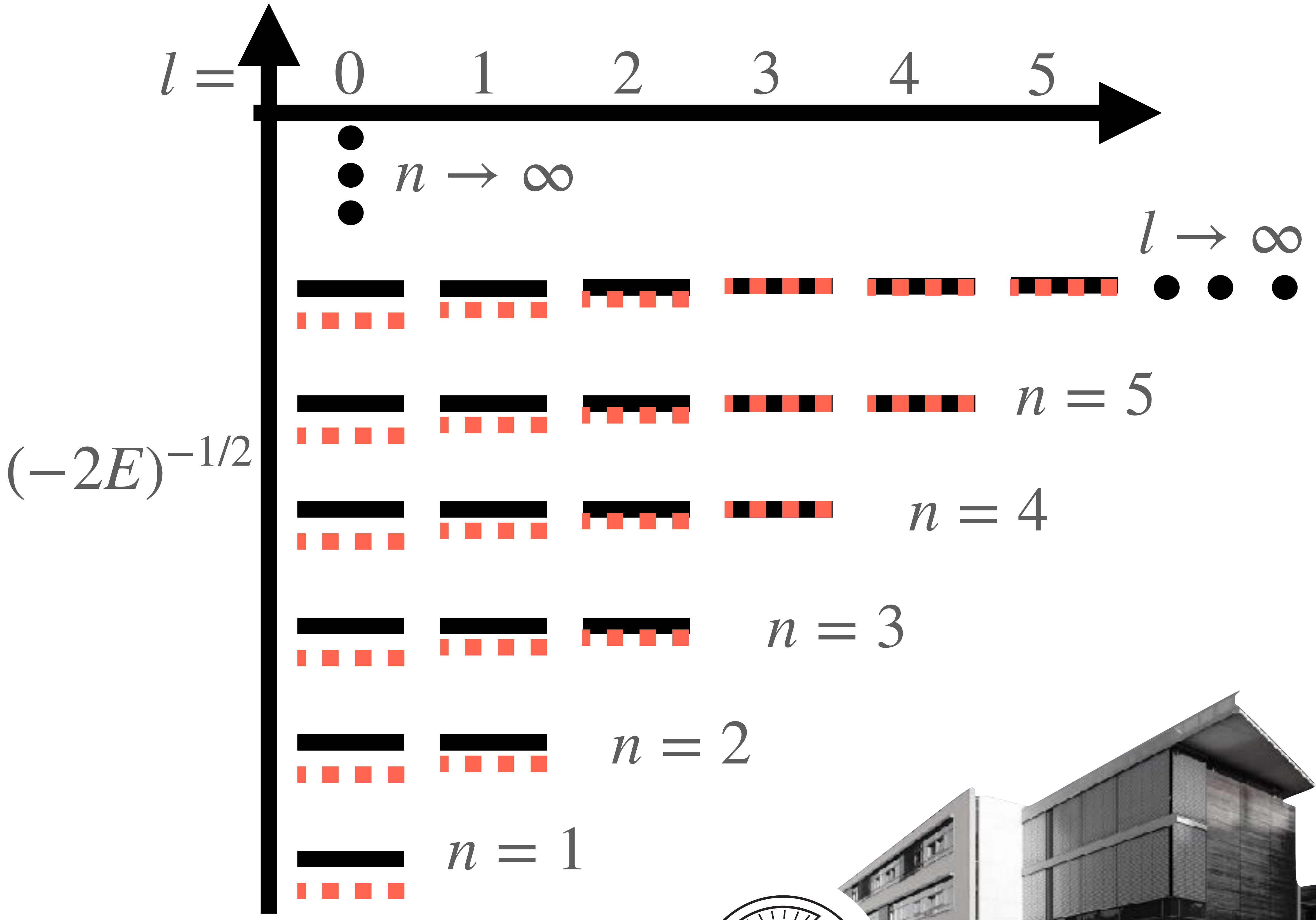


Figure 6. Quantum defects from the three different fitting methods. Data points for $n = 5$ and $n = 6$ were included in the calculations but are not shown, as their quantum defects are off the scale: 2.707 178 and 2.670 358, respectively.

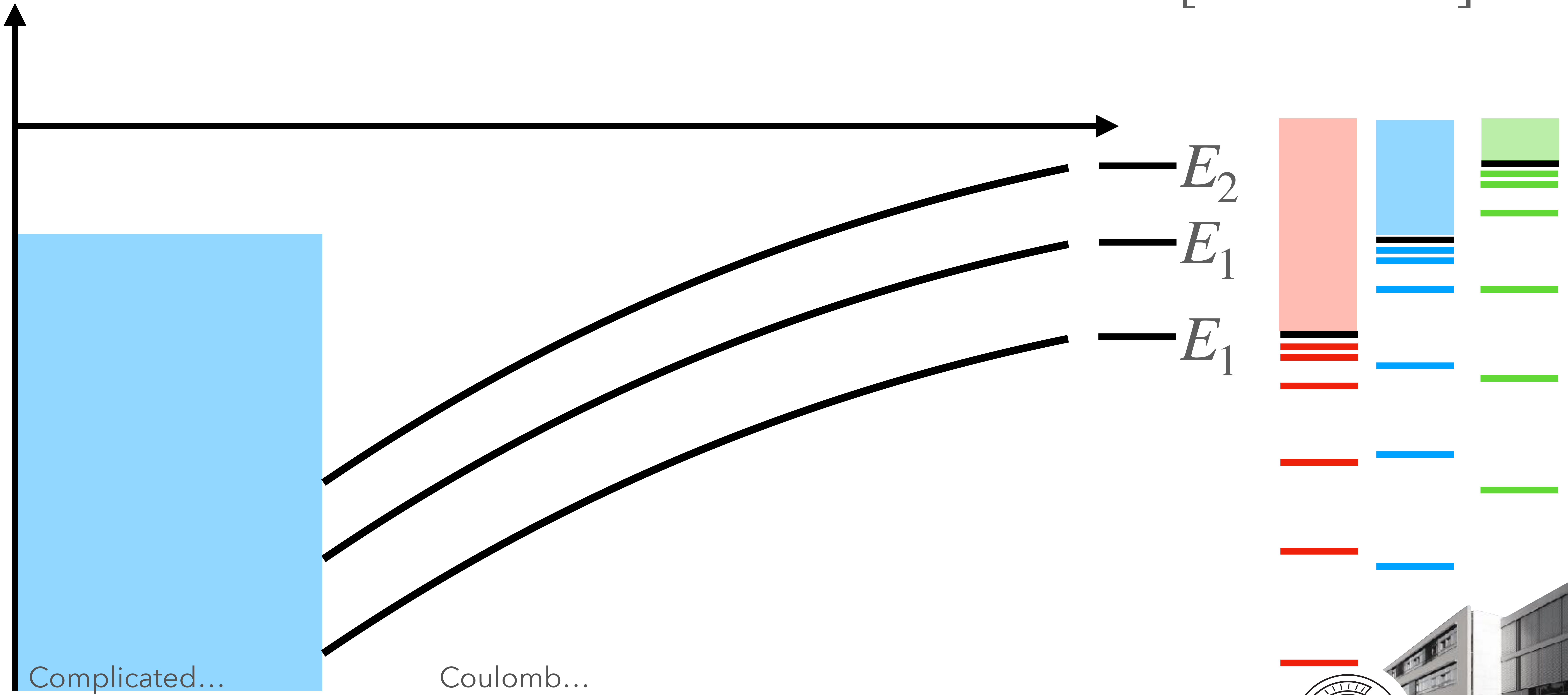


Multichannel quantum defect theory

KEY POINT #3: Most atoms are *multichannel* in nature - this is where QDT shines

$$u_{E\ell}(r) \sim \cos\delta_\ell(E) f_{E\ell}(r) - \sin\delta_\ell(E) g_{E\ell}(r)$$

$$0 = \sin\pi\left[\nu - \ell + \frac{\delta_\ell(E)}{\pi}\right]$$



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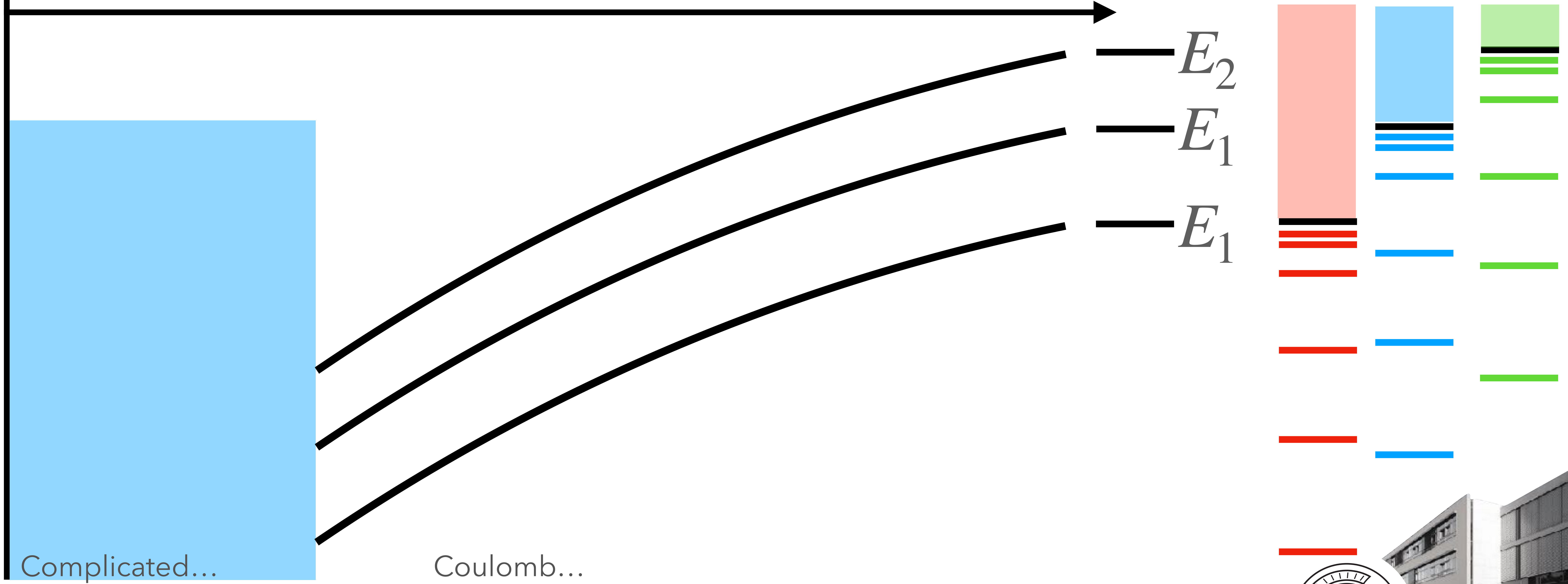
$$u_{E\ell}(r) \sim \cos\delta_\ell(E) f_{E\ell}(r) - \sin\delta_\ell(E) g_{E\ell}(r)$$

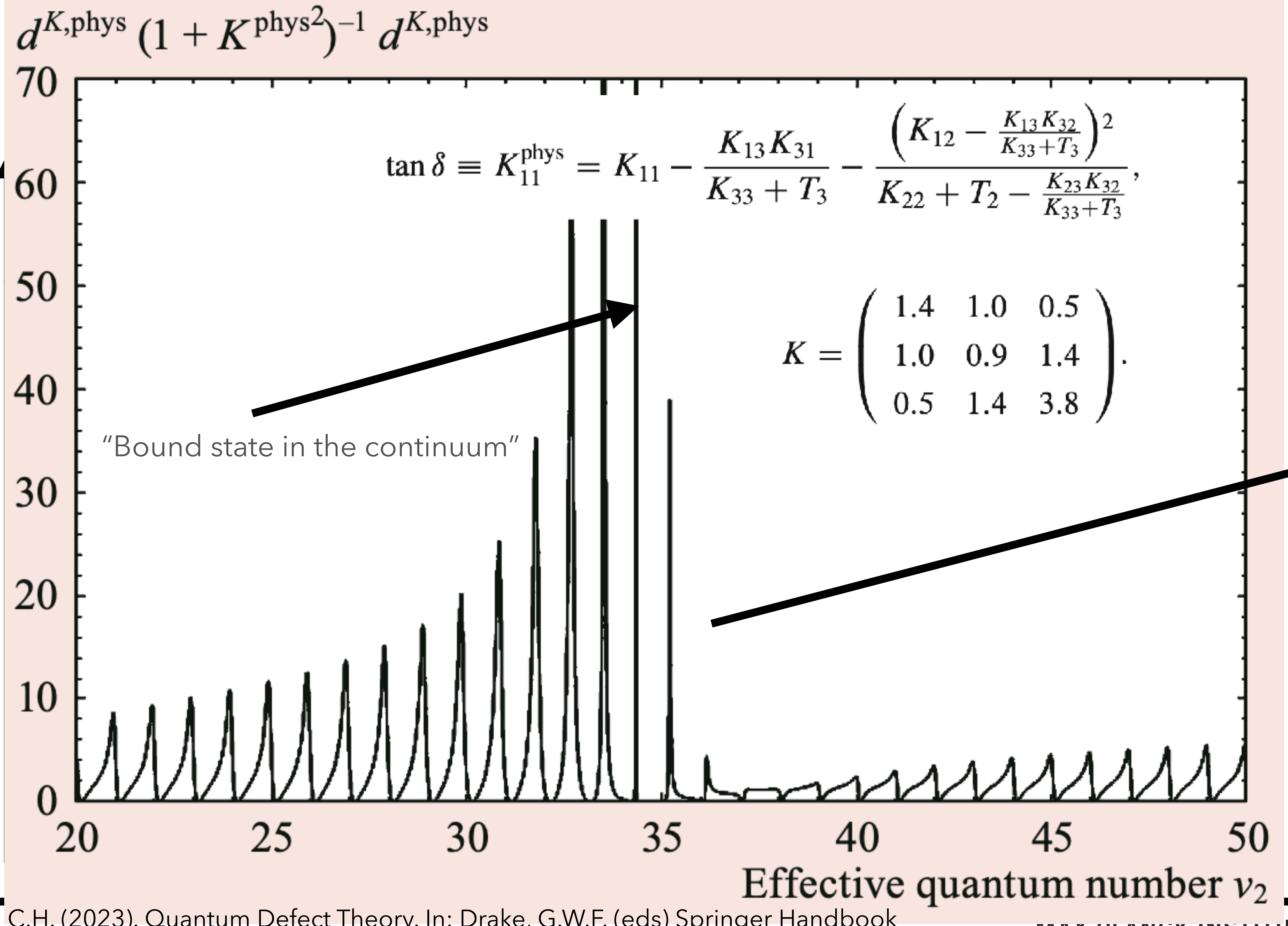
$$0 = \sin\pi\left[\nu - \ell + \frac{\delta_\ell(E)}{\pi}\right]$$

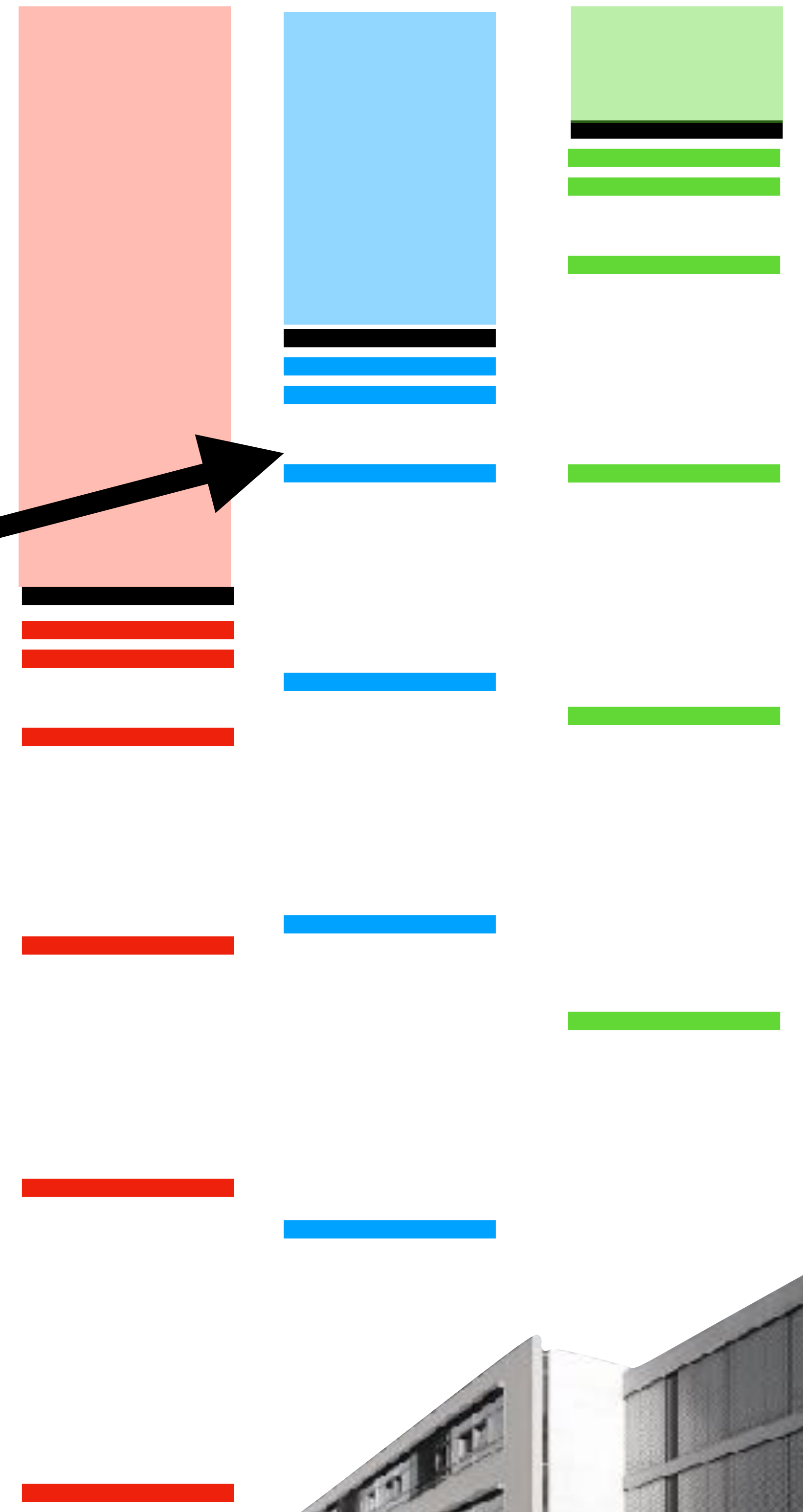
Instead of phase shifts…we get an energy-independent Scattering matrix S or Reactance matrix K

Applying boundary conditions at infinity gives either bound states or autoionizing resonances



Multichannel quantum defect theory





Greene, C.H. (2023). Quantum Defect Theory. In: Drake, G.W.F. (eds) Springer Handbook of Atomic, Molecular, and Optical Physics.

Scope of today's lecture

At the core of quantum simulation with Rydberg atoms: 150 years of spectroscopy

- From Rydberg to Pauli/Schrödinger to present day

As billed, it is a "lecture":

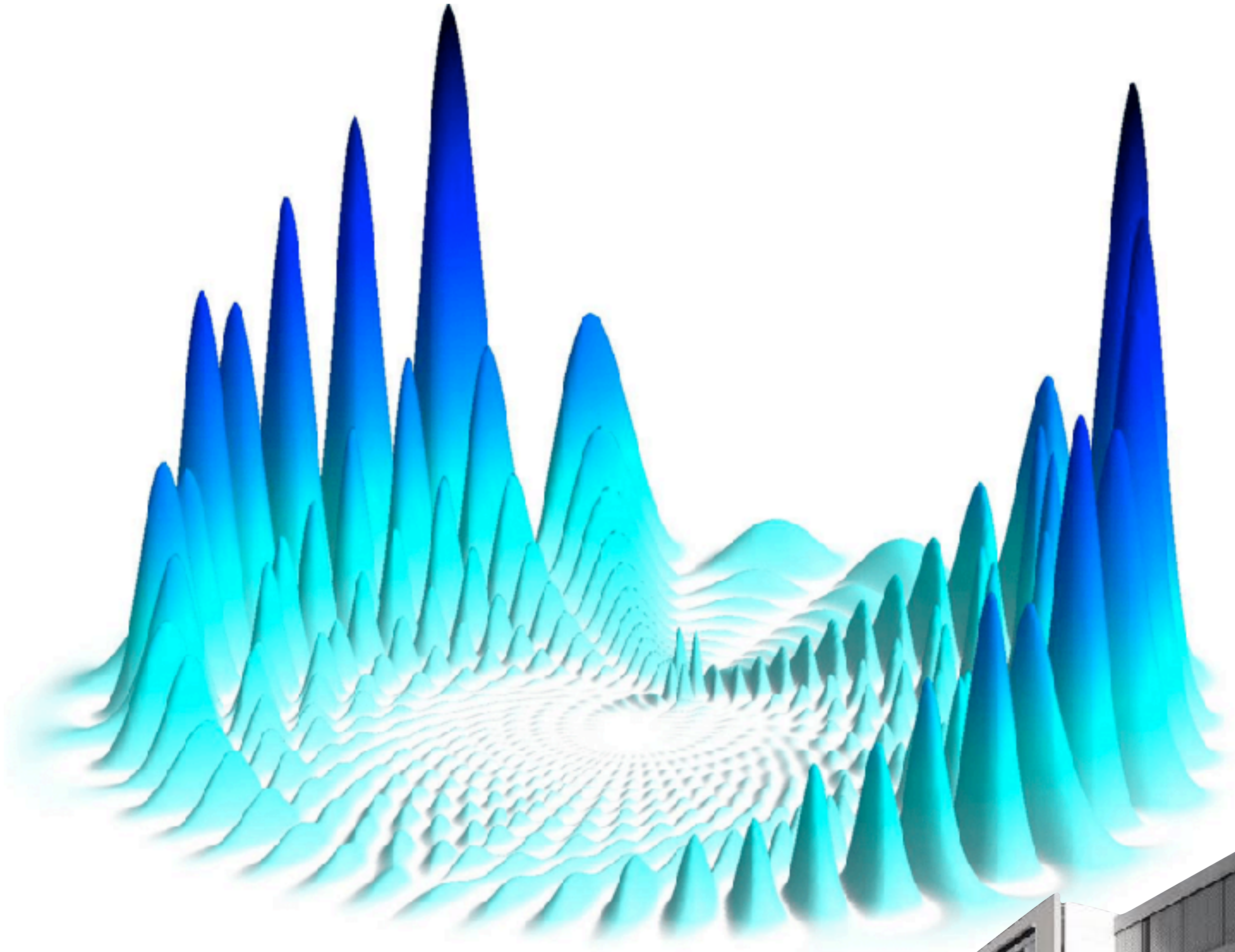
- …expect some equations…but hopefully not too many
- slides: https://www.pks.mpg.de/correlations-and-transport-in-rydberg-matter

What are Rydberg atoms?

- Quantum defect theory: alkali atoms
- Key properties of Rydberg atoms
- Multichannel quantum defect theory: many-electron atoms

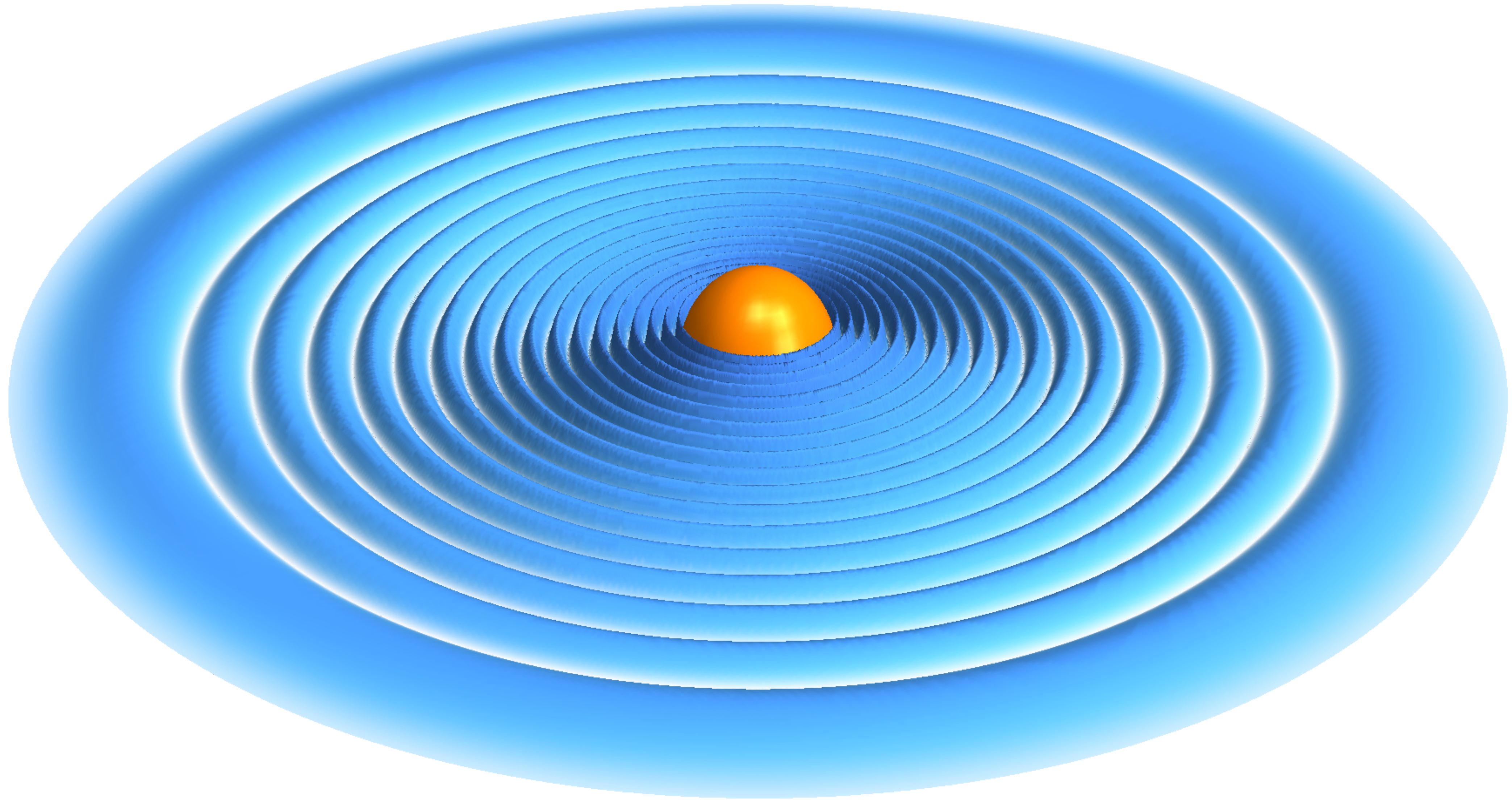
What are they good for?

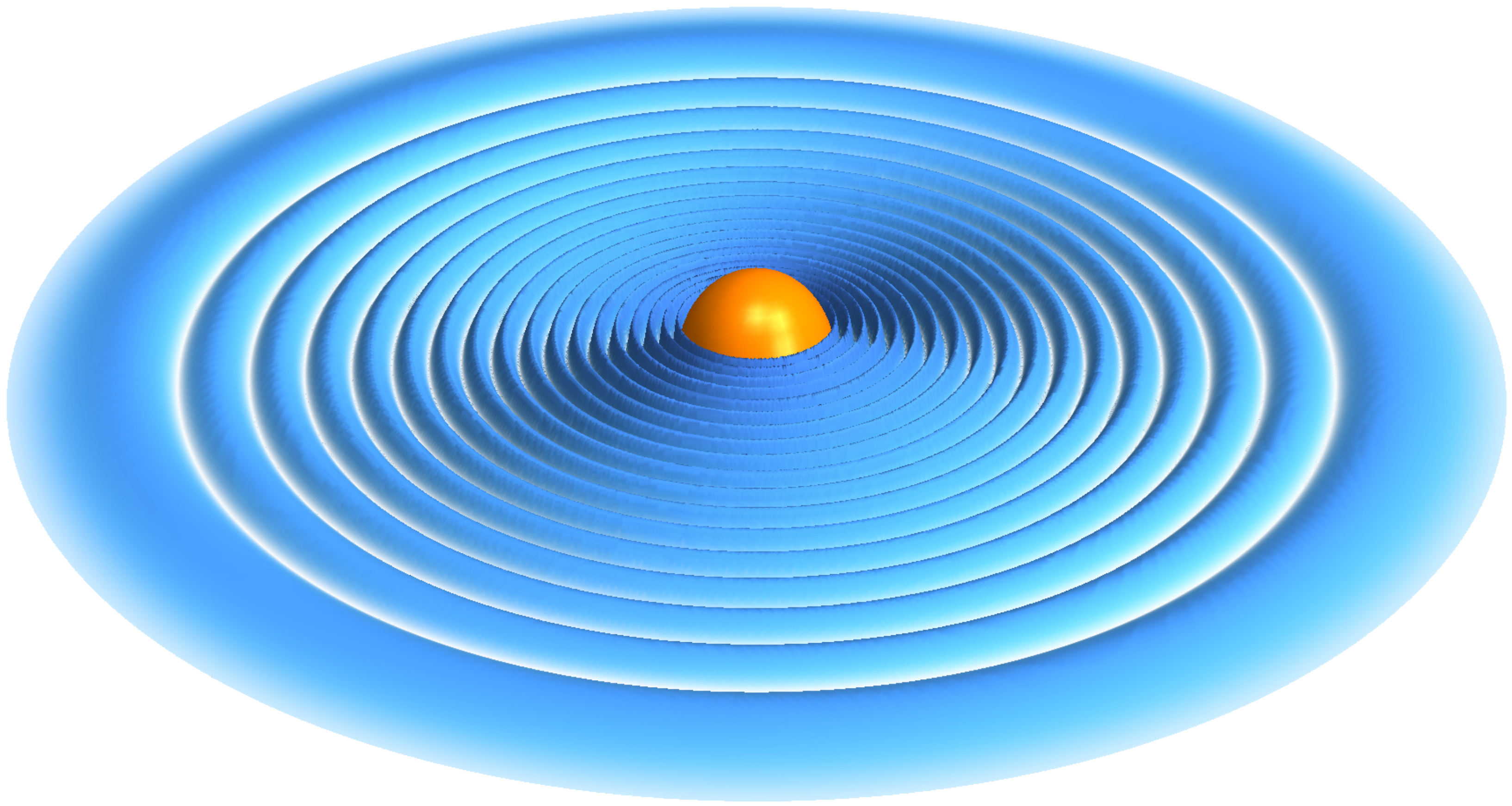
- Rydberg-Rydberg interactions
  - van der Waals / Rydberg blockade
  - dipole-dipole / "flip-flop" interactions
- Rydberg-ground-state-atom interactions



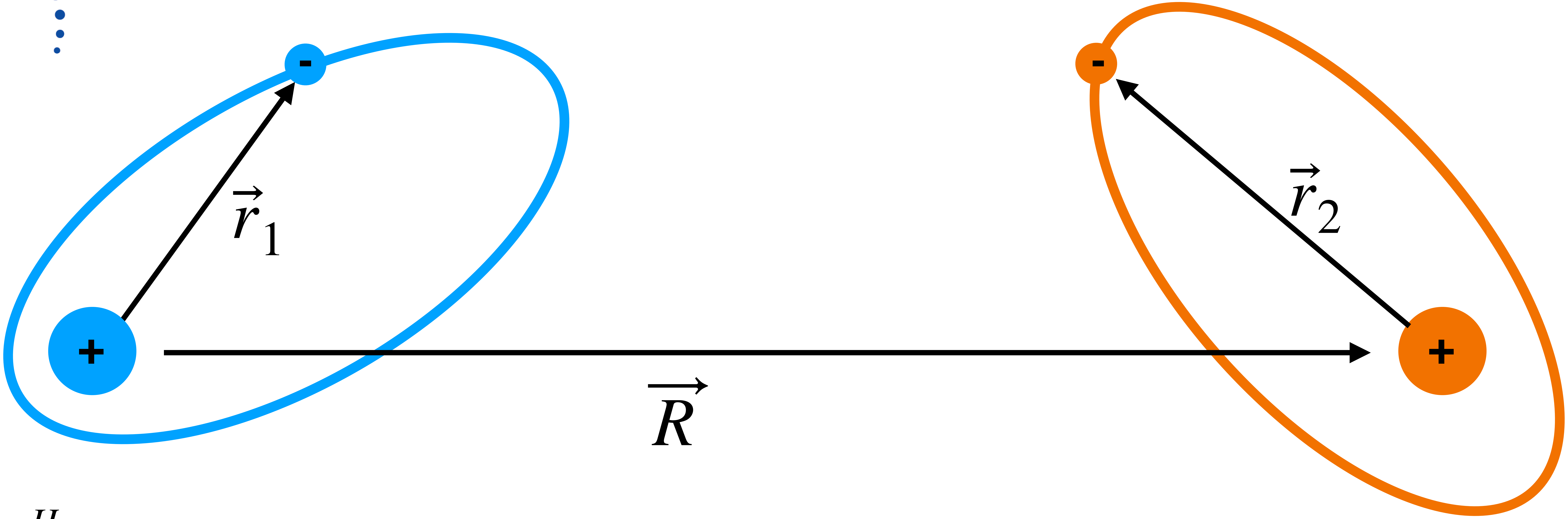
*we won't actually discuss this. it's just to get your attention

How do Rydberg atoms interact?



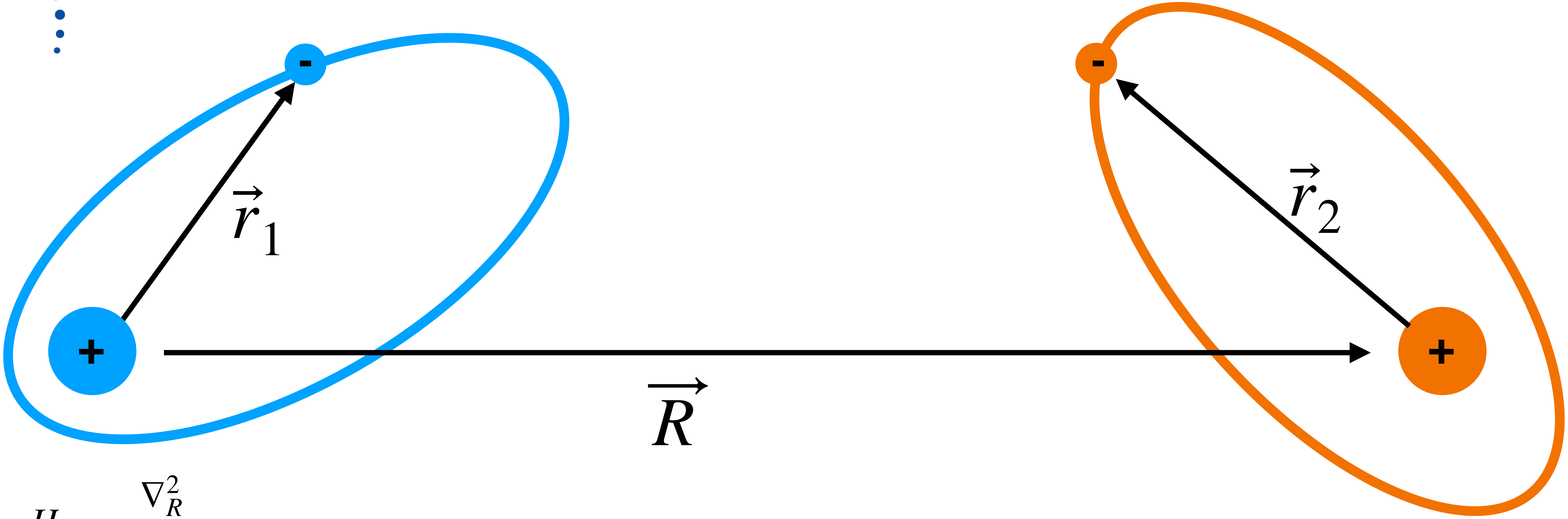


How do Rydberg atoms interact?



$H =$

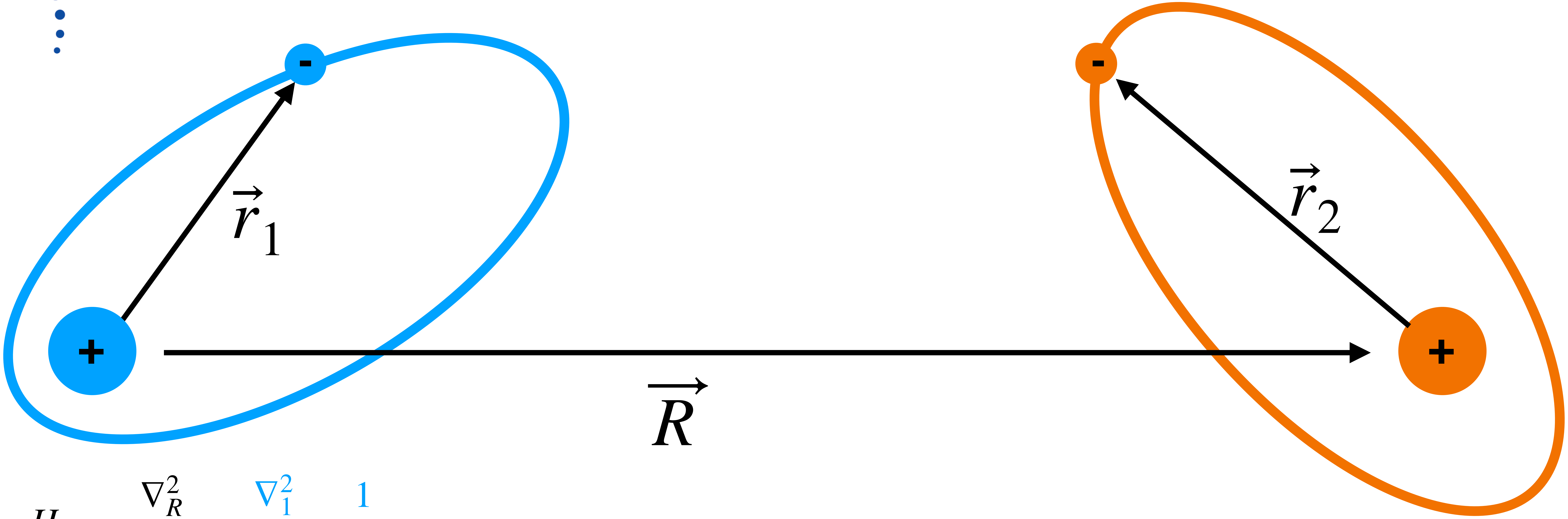
How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu}$$

kinetic energy of relative motion

How do Rydberg atoms interact?

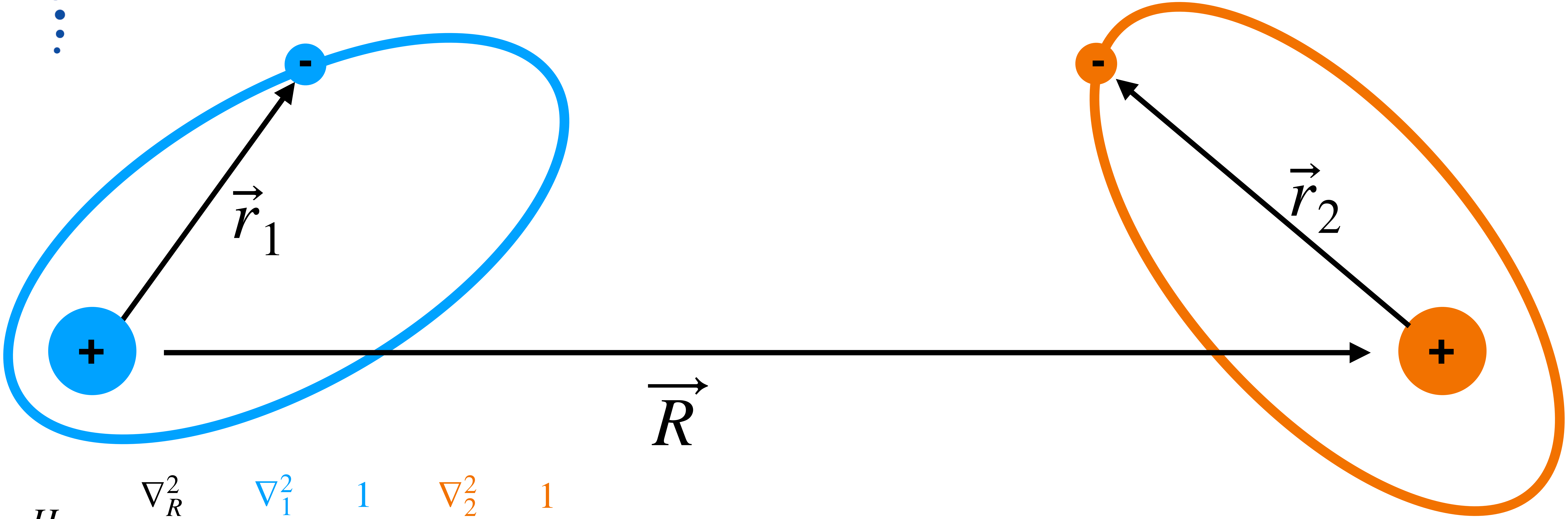


$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1}$$

kinetic energy of relative motion

Rydberg atom #1

How do Rydberg atoms interact?



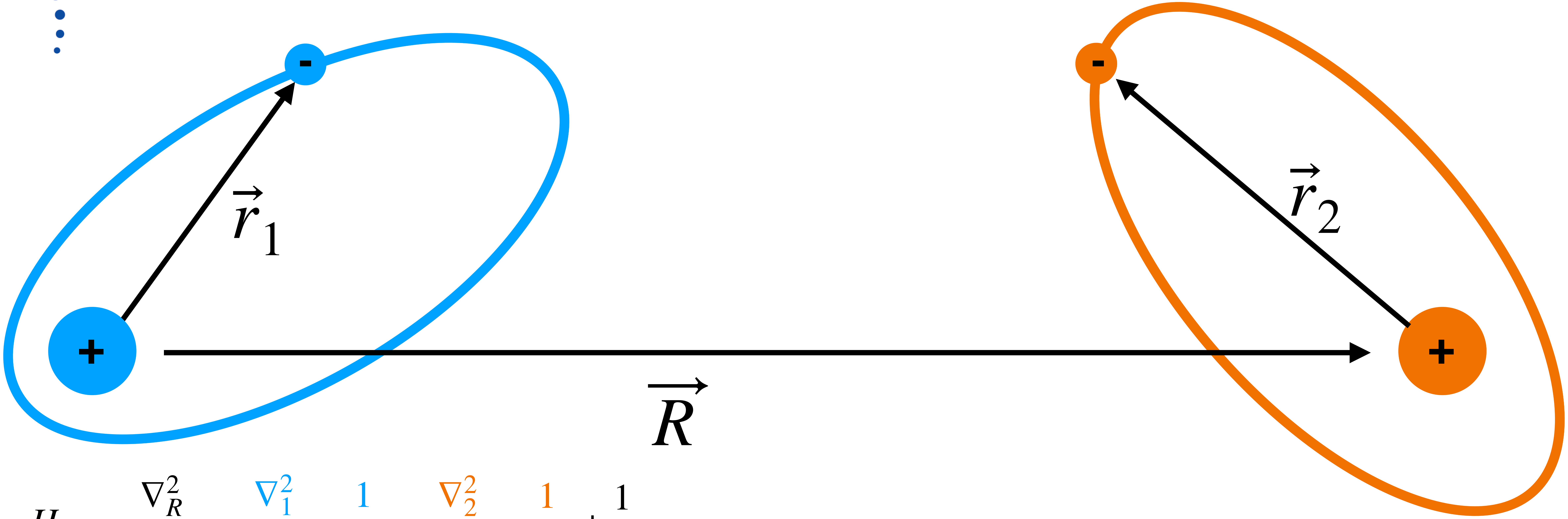
$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

kinetic energy of relative motion

Rydberg atom #1

Rydberg atom #2

How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{1}{R}$$

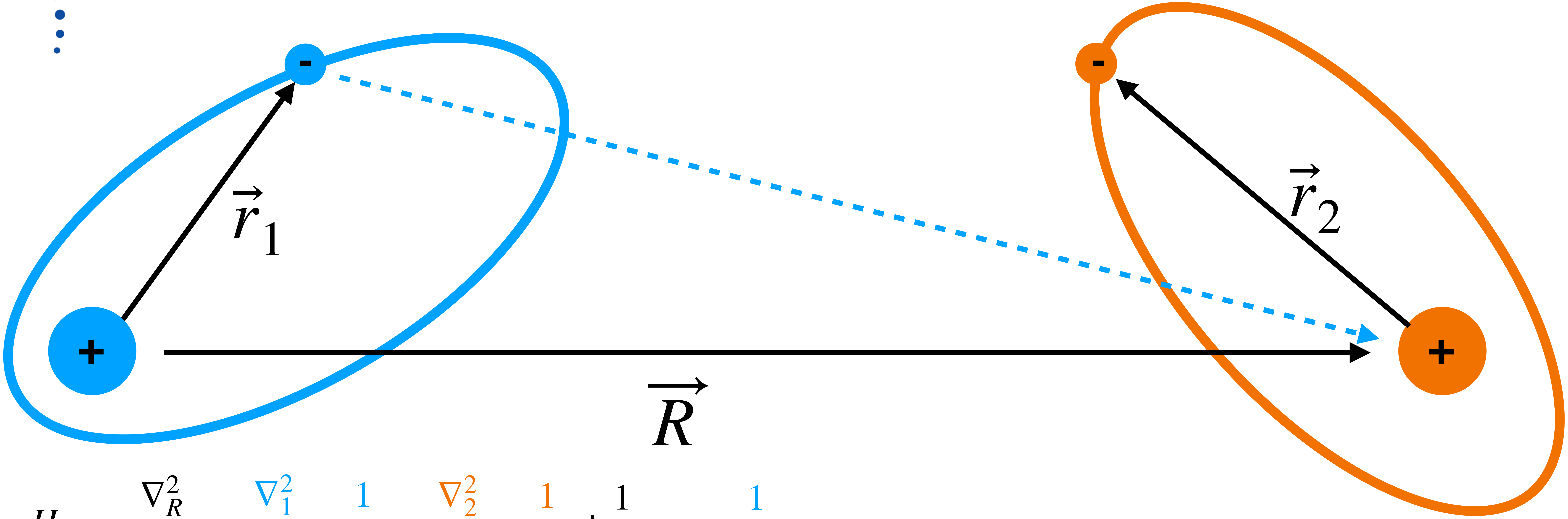
kinetic energy of relative motion

Rydberg atom #1

Rydberg atom #2

…repulsion…

How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{1}{R} - \frac{1}{\left|\vec{r}_1 - \vec{R}\right|}$$

kinetic energy of relative motion

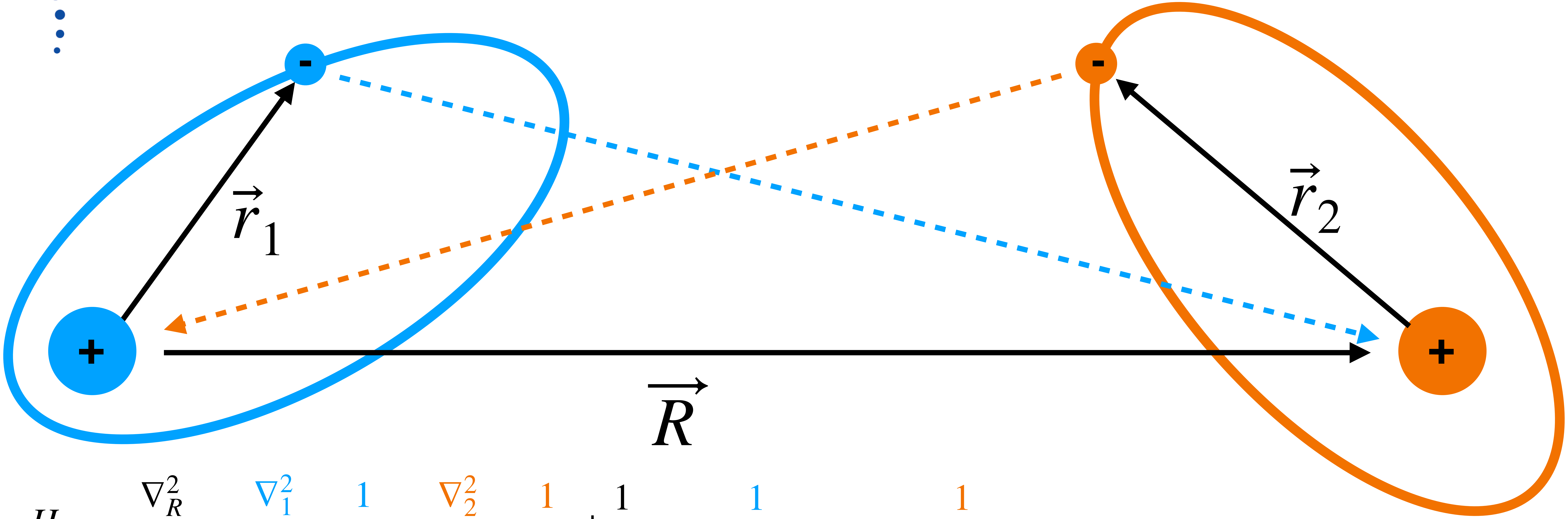
Rydberg atom #1

Rydberg atom #2

…repulsion…

…attraction…

How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{1}{R} - \frac{1}{\left|\vec{r}_1 - \vec{R}\right|} - \frac{1}{\left|\vec{r}_2 + \vec{R}\right|}$$

kinetic energy of relative motion

Rydberg atom #1

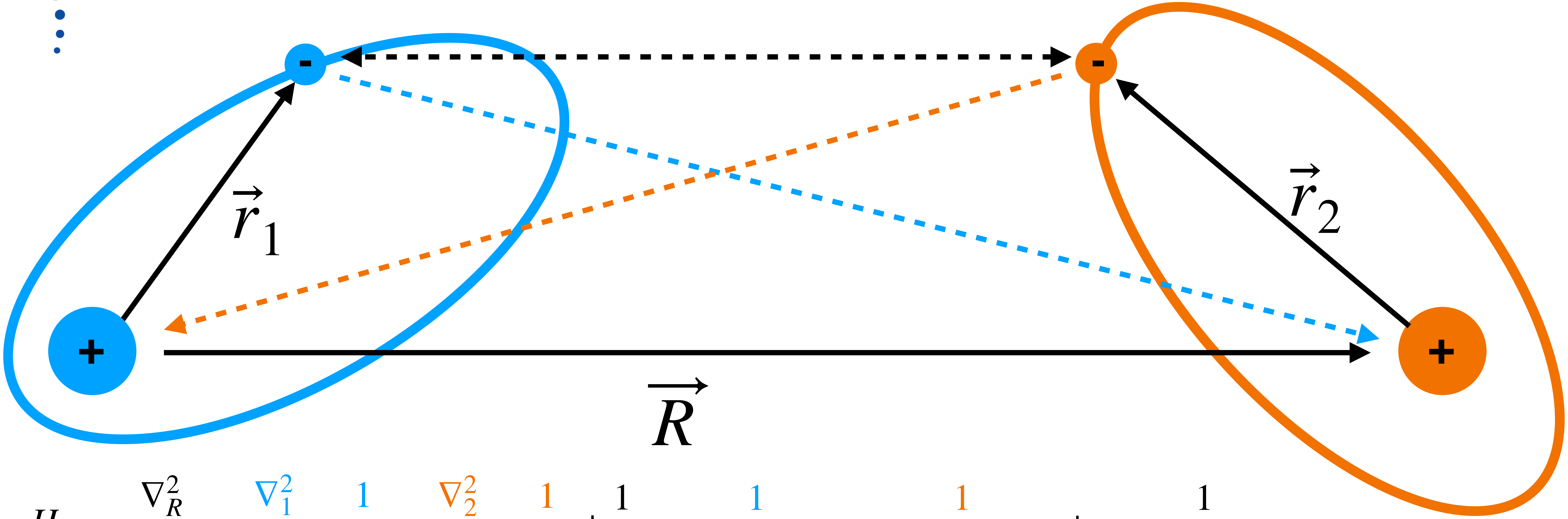
Rydberg atom #2

…repulsion…

…attraction…

…attraction…

How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{1}{R} - \frac{1}{\left|\vec{r}_1 - \vec{R}\right|} - \frac{1}{\left|\vec{r}_2 + \vec{R}\right|} + \frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

kinetic energy of relative motion

Rydberg atom #1

Rydberg atom #2

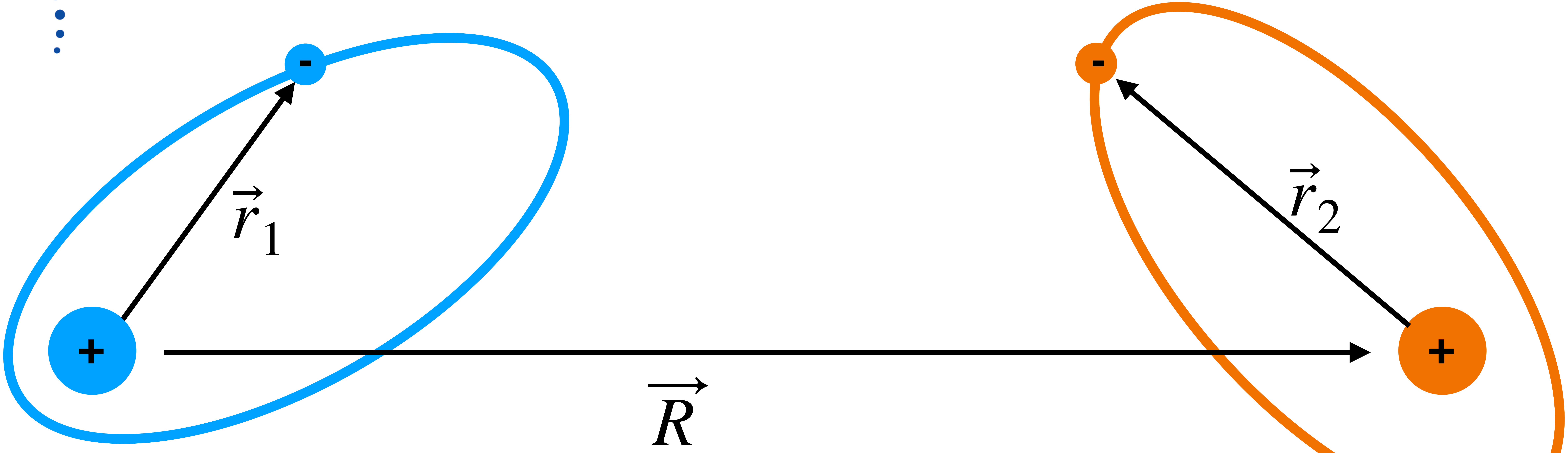
…repulsion…

…attraction…

…attraction…

…repulsion…

How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{1}{R} - \frac{1}{\left|\vec{r}_1 - \vec{R}\right|} - \frac{1}{\left|\vec{r}_2 + \vec{R}\right|} + \frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

kinetic energy of relative motion | Rydberg atom #1 | Rydberg atom #2 | …repulsion… | …attraction… | …attraction… | …repulsion…

Let R be much larger than the Rydberg orbits…looks like a great opportunity to do a Taylor expansion!

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} - \frac{1}{\left|\vec{r}_1 - \vec{R}\right|} - \frac{1}{\left|\vec{r}_2 + \vec{R}\right|} + \frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} \quad \longrightarrow \quad +\frac{1}{R}$$

$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|}$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|}$$

$$+\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

mpipks

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} \longrightarrow +\frac{1}{R}$$

$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|} \longrightarrow -\frac{1}{R\sqrt{1 - \frac{2\vec{r}_1 \cdot \hat{z}}{R} + \frac{r_1^2}{R^2}}}$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|}$$

$$+\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} \longrightarrow +\frac{1}{R}$$

$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(1 + \frac{\vec{r}_1 \cdot \hat{z}}{R} - \frac{1}{2}\frac{r_1^2}{R^2} + \frac{3(\vec{r}_1 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|}$$

$$+\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

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$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(1 - \frac{\vec{r}_2 \cdot \hat{z}}{R} - \frac{1}{2}\frac{r_2^2}{R^2} + \frac{3(\vec{r}_2 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$+\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|}$$

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} \longrightarrow +\frac{1}{R}$$

$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(1 + \frac{\vec{r}_1 \cdot \hat{z}}{R} - \frac{1}{2}\frac{r_1^2}{R^2} + \frac{3(\vec{r}_1 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(1 - \frac{\vec{r}_2 \cdot \hat{z}}{R} - \frac{1}{2}\frac{r_2^2}{R^2} + \frac{3(\vec{r}_2 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$+\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|} \longrightarrow -\frac{1}{R}\left(-1 - \frac{\vec{r}_1 \cdot \hat{z}}{R} + \frac{r_2 \cdot \hat{z}}{R} - \frac{-r_1^2 + 2\vec{r}_1 \cdot \vec{r}_2 - r_2^2}{2R^2} - \frac{3}{2R^2}\left((\vec{r}_1 \cdot \hat{z})^2 - 2(\vec{r}_1 \cdot \hat{z})(\vec{r}_2 \cdot \hat{z}) + (\vec{r}_2 \cdot \hat{z})^2\right)\right)$$

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} \longrightarrow \cancel{+\frac{1}{R}}$$

$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} + \frac{\vec{r}_1 \cdot \hat{z}}{R} - \frac{1}{2}\frac{r_1^2}{R^2} + \frac{3(\vec{r}_1 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} - \frac{\vec{r}_2 \cdot \hat{z}}{R} - \frac{1}{2}\frac{r_2^2}{R^2} + \frac{3(\vec{r}_2 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} - \frac{\vec{r}_1 \cdot \hat{z}}{R} + \frac{r_2 \cdot \hat{z}}{R} - \frac{-r_1^2 + 2\vec{r}_1 \cdot \vec{r}_2 - r_2^2}{2R^2} - \frac{3}{2R^2}\left((\vec{r}_1 \cdot \hat{z})^2 - 2(\vec{r}_1 \cdot \hat{z})(\vec{r}_2 \cdot \hat{z}) + (\vec{r}_2 \cdot \hat{z})^2\right)\right)$$

mpipks

How do Rydberg atoms interact?

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = +\frac{1}{R} \longrightarrow \cancel{+\frac{1}{R}}$$

$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} + \cancel{\frac{\vec{r}_1 \cdot \hat{z}}{R}} - \frac{1}{2}\frac{r_1^2}{R^2} + \frac{3(\vec{r}_1 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} - \cancel{\frac{\vec{r}_2 \cdot \hat{z}}{R}} - \frac{1}{2}\frac{r_2^2}{R^2} + \frac{3(\vec{r}_2 \cdot \hat{z})^2}{2R^2}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|} \longrightarrow -\frac{1}{R}\Bigg(\cancel{1} - \cancel{\frac{\vec{r}_1 \cdot \hat{z}}{R}} + \cancel{\frac{r_2 \cdot \hat{z}}{R}} - \frac{-r_1^2 + 2\vec{r}_1 \cdot \vec{r}_2 - r_2^2}{2R^2}$$

$$-\frac{3}{2R^2}\left((\vec{r}_1 \cdot \hat{z})^2 - 2(\vec{r}_1 \cdot \hat{z})(\vec{r}_2 \cdot \hat{z}) + (\vec{r}_2 \cdot \hat{z})^2\right)\Bigg)$$

How do Rydberg atoms interact?

$$V(\vec{R},\vec{r}_1,\vec{r}_2) = +\frac{1}{R} \longrightarrow \cancel{+\frac{1}{R}}$$

$$-\frac{1}{\left|\vec{r}_1-\vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1}+\cancel{\frac{\vec{r}_1\cdot\hat{z}}{R}}-\cancel{\frac{1}{2}\frac{r_1^2}{R^2}}+\frac{3(\vec{r}_1\cdot\hat{z})^2}{2R^2}\right)+\mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{r}_2+\vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1}-\cancel{\frac{\vec{r}_2\cdot\hat{z}}{R}}-\cancel{\frac{1}{2}\frac{r_2^2}{R^2}}+\frac{3(\vec{r}_2\cdot\hat{z})^2}{2R^2}\right)+\mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{R}-\vec{r}_1+\vec{r}_2\right|} \longrightarrow -\frac{1}{R}\Bigg(\cancel{-1}\,1-\cancel{\frac{\vec{r}_1\cdot\hat{z}}{R}}+\cancel{\frac{r_2\cdot\hat{z}}{R}}-\frac{\cancel{-r_1^2}+2\vec{r}_1\cdot\vec{r}_2\cancel{-r_2^2}}{2R^2}$$

$$-\frac{3}{2R^2}\left((\vec{r}_1\cdot\hat{z})^2-2(\vec{r}_1\cdot\hat{z})(\vec{r}_2\cdot\hat{z})+(\vec{r}_2\cdot\hat{z})^2\right)\Bigg)$$

How do Rydberg atoms interact?

$$V(\vec{R},\vec{r}_1,\vec{r}_2) = +\frac{1}{R} \longrightarrow \cancel{+\frac{1}{R}}$$

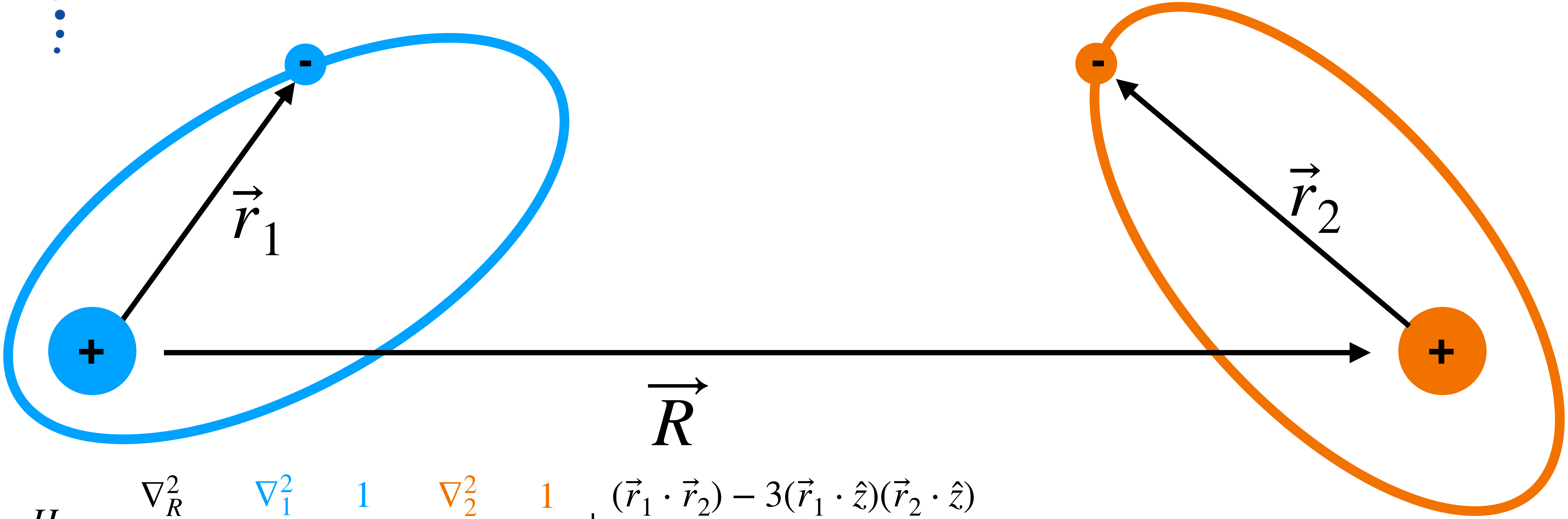
$$-\frac{1}{\left|\vec{r}_1 - \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} + \cancel{\frac{\vec{r}_1\cdot\hat{z}}{R}} - \cancel{\frac{1}{2}\frac{r_1^2}{R^2}} + \cancel{\frac{3(\vec{r}_1\cdot\hat{z})^2}{2R^2}}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{r}_2 + \vec{R}\right|} \longrightarrow -\frac{1}{R}\left(\cancel{1} - \cancel{\frac{\vec{r}_2\cdot\hat{z}}{R}} - \cancel{\frac{1}{2}\frac{r_2^2}{R^2}} + \cancel{\frac{3(\vec{r}_2\cdot\hat{z})^2}{2R^2}}\right) + \mathcal{O}(R^{-4})$$

$$-\frac{1}{\left|\vec{R} - \vec{r}_1 + \vec{r}_2\right|} \longrightarrow -\frac{1}{R}\left(\cancel{-}1 - \cancel{\frac{\vec{r}_1\cdot\hat{z}}{R}} + \cancel{\frac{r_2\cdot\hat{z}}{R}} - \frac{\cancel{-r_1^2} + 2\vec{r}_1\cdot\vec{r}_2 - \cancel{r_2^2}}{2R^2}\right.$$

$$\left. -\frac{3}{2R^2}\left(\cancel{(\vec{r}_1\cdot\hat{z})^2} - 2(\vec{r}_1\cdot\hat{z})(\vec{r}_2\cdot\hat{z}) + \cancel{(\vec{r}_2\cdot\hat{z})^2}\right)\right)$$

How do Rydberg atoms interact?



$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{(\vec{r}_1 \cdot \vec{r}_2) - 3(\vec{r}_1 \cdot \hat{z})(\vec{r}_2 \cdot \hat{z})}{R^3}$$

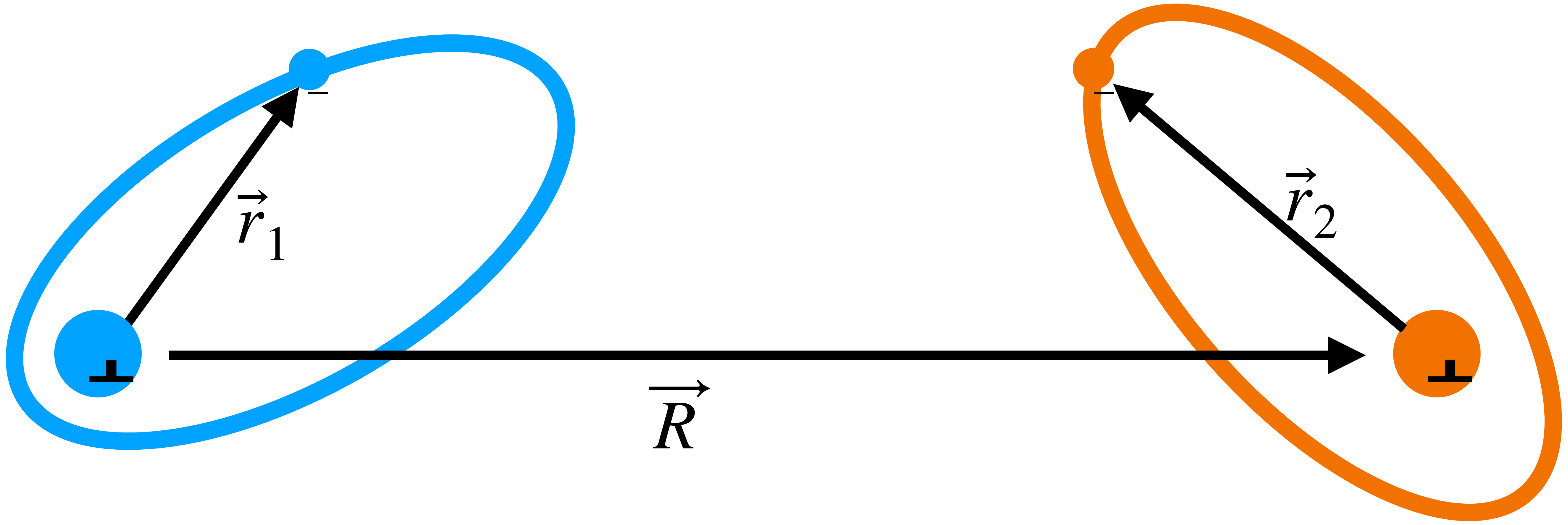
kinetic energy of relative motion | Rydberg atom #1 | Rydberg atom #2 | Dipole-dipole interaction

After the dust has settled, we are left with a dipole-dipole potential.
What next?

How do Rydberg atoms interact?

$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

$$+\frac{(\vec{r}_1 \cdot \vec{r}_2) - 3(\vec{r}_1 \cdot \hat{z})(\vec{r}_2 \cdot \hat{z})}{R^3}$$

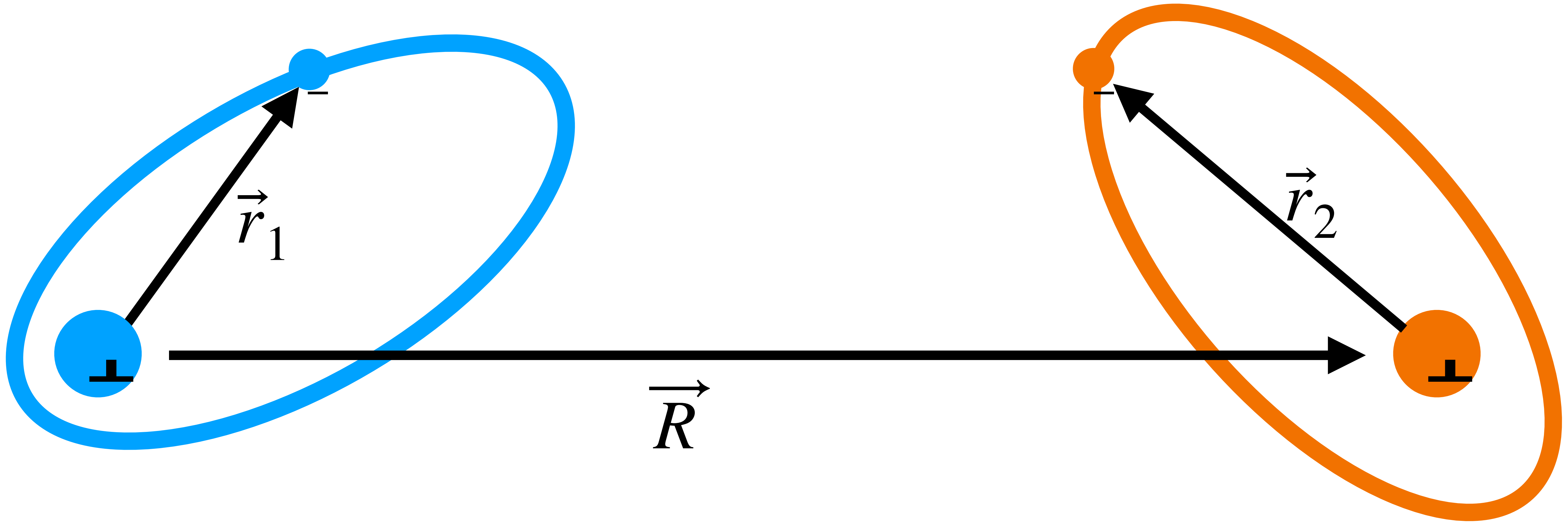


At the distances where this formula is valid, the bottom row is a perturbation to the upper row.

How do Rydberg atoms interact?

$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

$$+\frac{r_1 r_2}{R^3}$$

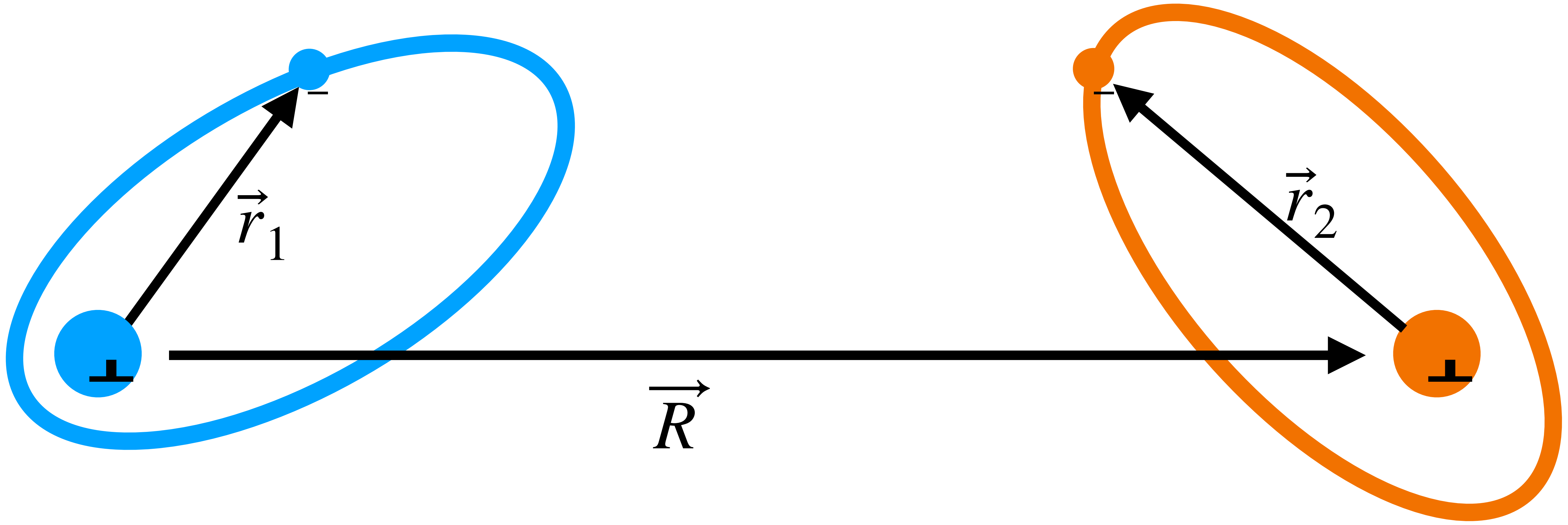


At the distances where this formula is valid, the bottom row is a perturbation to the upper row.

How do Rydberg atoms interact?

$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

$$+\frac{r_1 r_2}{R^3}$$



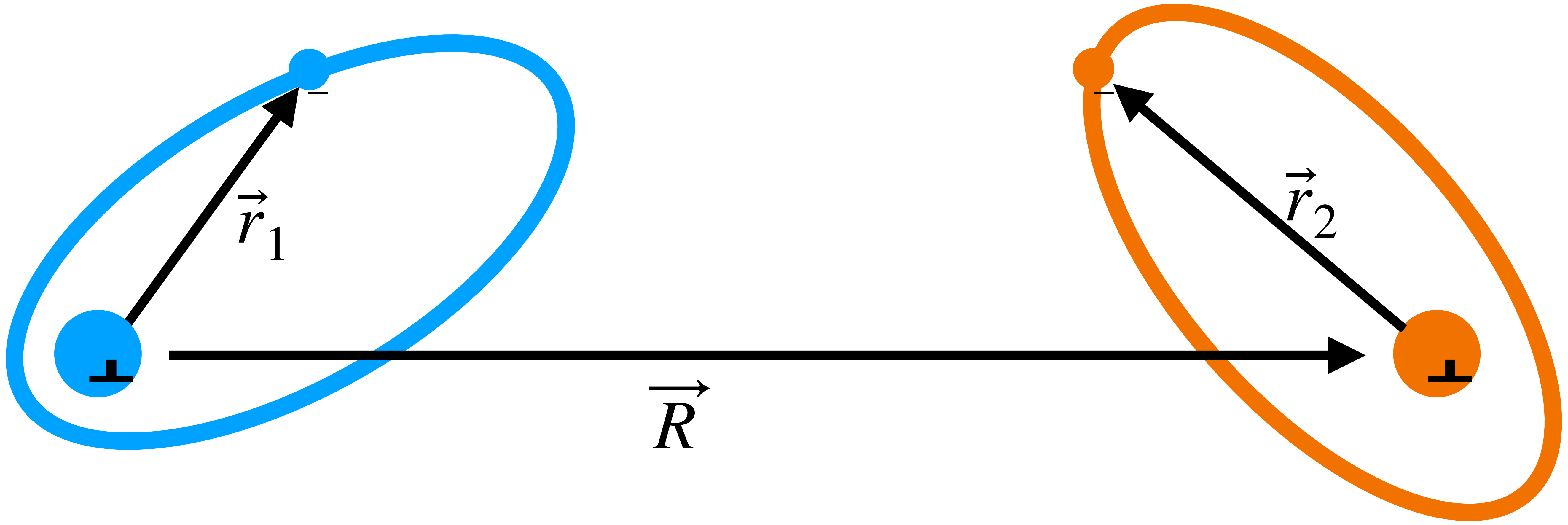
At the distances where this formula is valid, the bottom row is a perturbation to the upper row.

$g = |ns\rangle$ $g = |ns\rangle$

How do Rydberg atoms interact?

$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

$$+\frac{r_1 r_2}{R^3}$$



At the distances where this formula is valid, the bottom row is a perturbation to the upper row.

$$0 = \langle n\ell \,|\, r \,|\, n\ell \rangle$$

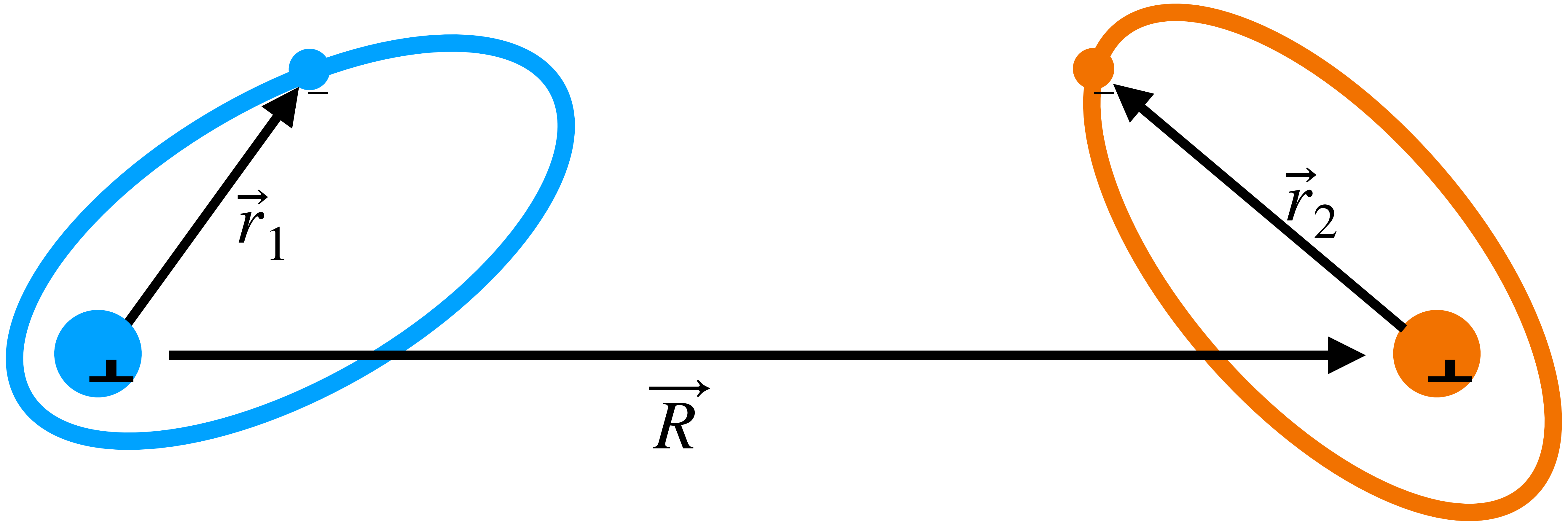
$g = |ns\rangle$ $g = |ns\rangle$

mpipks

How do Rydberg atoms interact?

$$H = -\frac{\nabla_R^2}{2\mu} \quad -\frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} \quad -\frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

$$+\frac{r_1 r_2}{R^3}$$



At the distances where this formula is valid, the bottom row is a perturbation to the upper row.

$$0 = \langle n\ell \,|\, r \,|\, n\ell \rangle$$

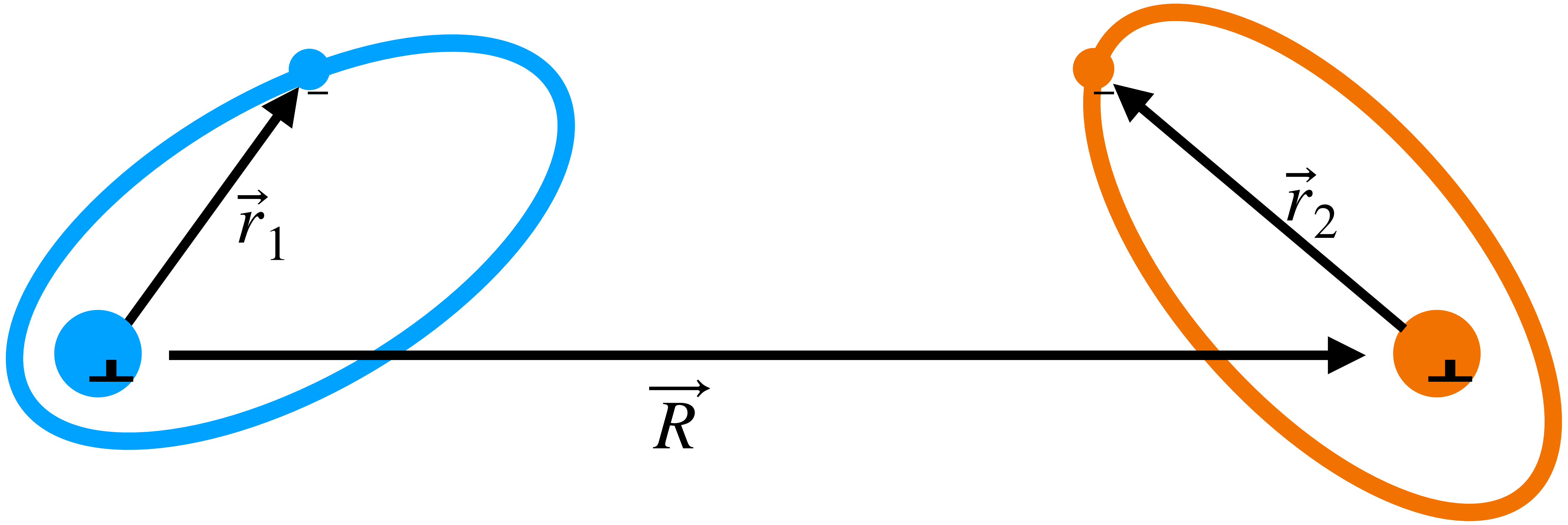
When life gets hard, make it a two level system:

Atom 1	Atom 2
$e = \lvert np \rangle$	$e = \lvert np \rangle$
Δ	Δ
$g = \lvert ns \rangle$	$g = \lvert ns \rangle$

How do Rydberg atoms interact?

$$H = -\frac{\nabla_R^2}{2\mu} - \frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2}$$

$$+\frac{r_1 r_2}{R^3}$$



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$e = |np\rangle$ $\quad$ $e = |np\rangle$

Δ $\quad$ Δ

$g = |ns\rangle$ $\quad$ $g = |ns\rangle$

Four state basis: $|ee\rangle, |gg\rangle, |eg\rangle, |ge\rangle$ $\qquad$ $d = \langle ns | r | np \rangle$

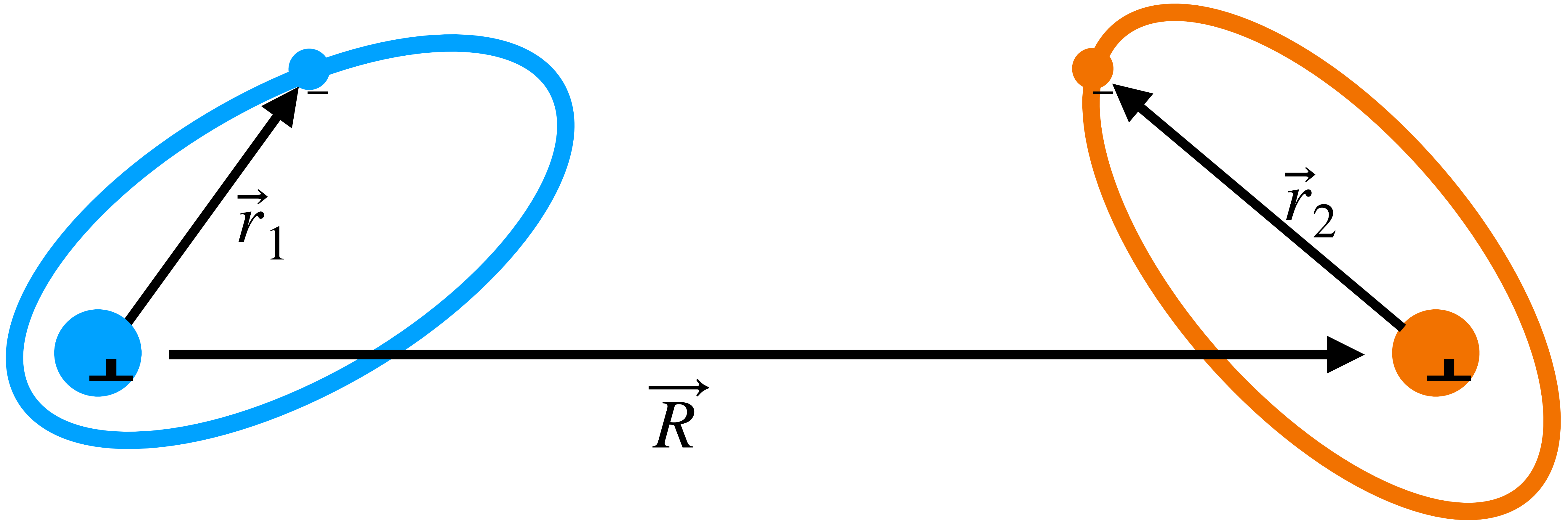
$$\implies \langle ge | r_1 r_2 | eg \rangle = \langle g | r_1 | e \rangle \langle e | r_2 | g \rangle = d^2$$

$$\implies \langle gg | r_1 r_2 | ee \rangle = \langle g | r_1 | e \rangle \langle g | r_2 | e \rangle = d^2$$

$$\implies \langle ge | r_1 r_2 | ge \rangle = \langle g | r_1 | g \rangle \langle e | r_2 | e \rangle = 0$$

How do Rydberg atoms interact?

$$V = -\frac{\nabla_1^2}{2m_e} - \frac{1}{r_1} - \frac{\nabla_2^2}{2m_e} - \frac{1}{r_2} + \frac{r_1 r_2}{R^3}$$



$$\underline{V}(R) = \begin{pmatrix} 0 & \frac{d_1 d_2}{R^3} & 0 & 0 \\ \frac{d_1 d_2}{R^3} & 2\Delta & 0 & 0 \\ 0 & 0 & \Delta & \frac{d_1 d_2}{R^3} \\ 0 & 0 & \frac{d_1 d_2}{R^3} & \Delta \end{pmatrix}$$

When life gets hard, make it a two level system:

$e = |np\rangle$ $\quad\Delta\quad$ $g = |ns\rangle$ (atom 1)

$e = |np\rangle$ $\quad\Delta\quad$ $g = |ns\rangle$ (atom 2)

Four state basis: $|ee\rangle, |gg\rangle, |eg\rangle, |ge\rangle$ $\qquad d = \langle ns | r | np \rangle$

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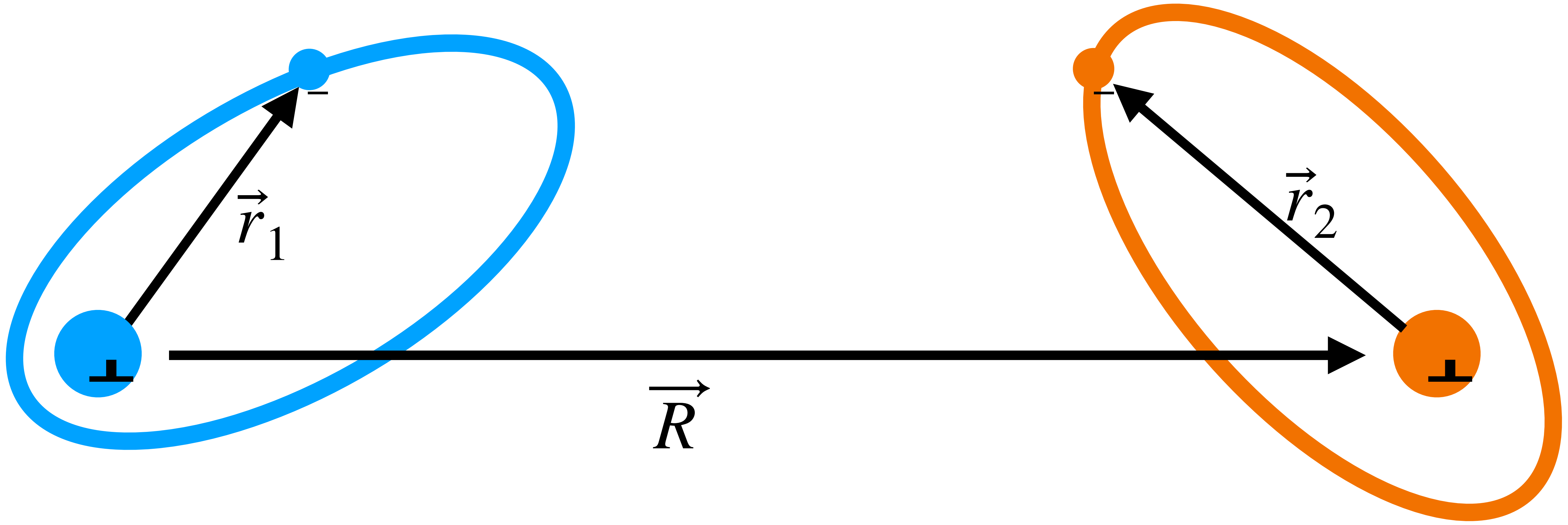

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When life gets hard, make it a two level system:

$e = |np\rangle$ Δ $g = |ns\rangle$ $e = |np\rangle$ Δ $g = |ns\rangle$

$$\begin{pmatrix} a & b \\ b & c \end{pmatrix}$$

$$2u_\pm = (a+c) \pm \sqrt{4b^2 + (a-c)^2}$$

(of course we're gonna Taylor expand again)

Two classes of interaction:

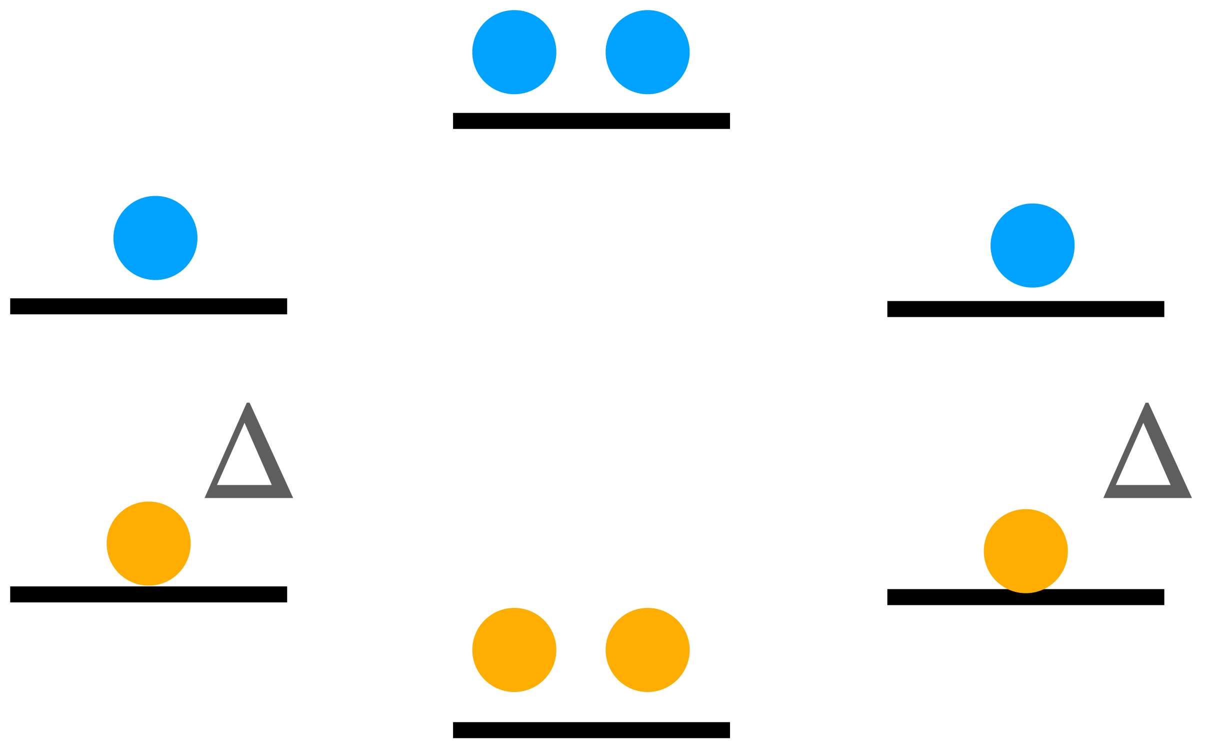
$$u_+ = \Delta + \frac{d_1 d_2}{R^3}. \qquad u_+ \approx 2\Delta + \frac{(d_1 d_2)^2/(2\Delta)}{R^6}.$$

$$u_- = \Delta - \frac{d_1 d_2}{R^3}. \qquad u_- \approx 0 - \frac{(d_1 d_2)^2/(2\Delta)}{R^6}.$$

How do Rydberg atoms interact?

This simple two-state model shows that atoms interact at long-range in two different regimes:

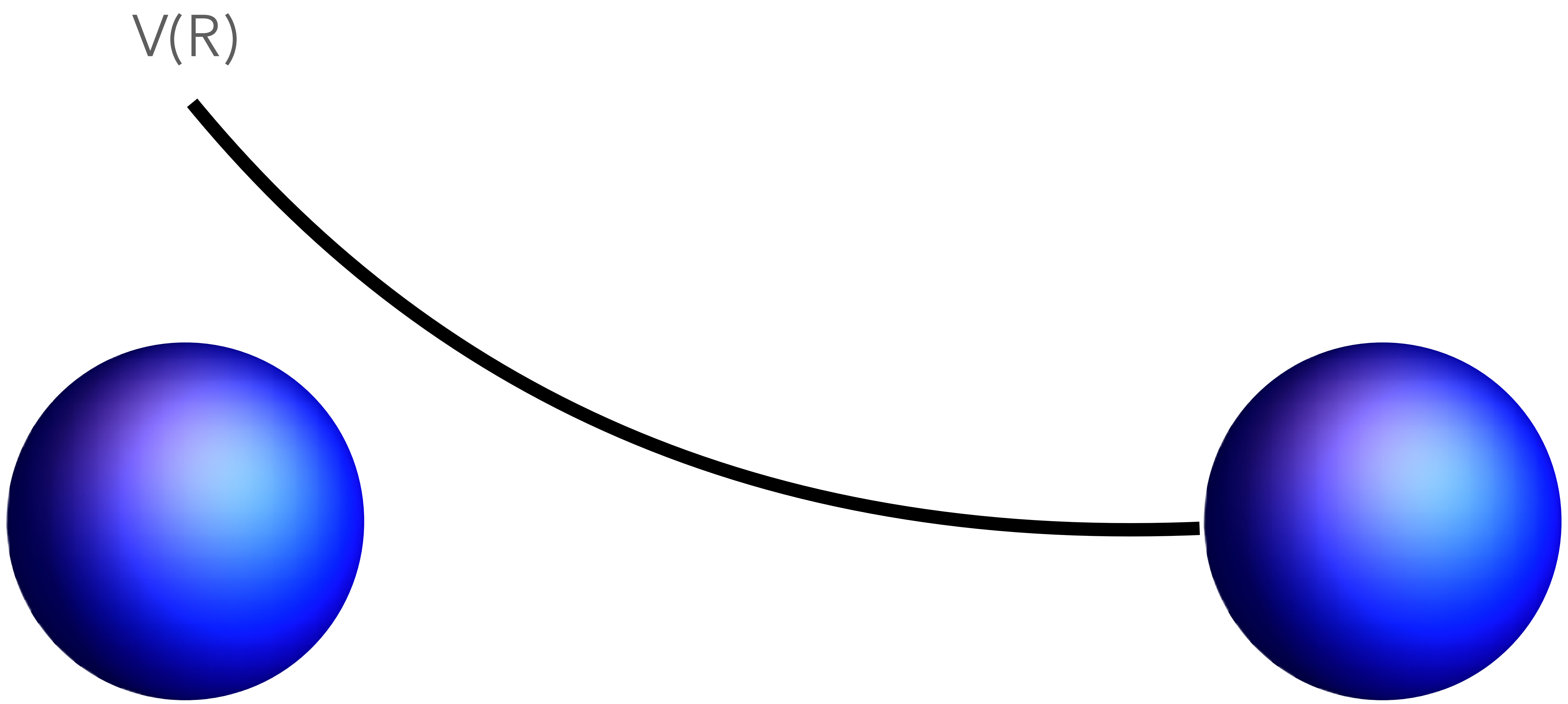
Both atoms in same state:



$$E_+ \approx 2\Delta + \frac{(d_1 d_2)^2/(2\Delta)}{R^6}.$$

$$E_- \approx -\frac{(d_1 d_2)^2/(2\Delta)}{R^6}.$$

This *non-resonant* van der Waals interaction is at the core of ground-state – ground-state atom scattering as well as the source of Rydberg blockade: the ultra-strong interaction between Rydberg atoms prevents their mutual excitation!



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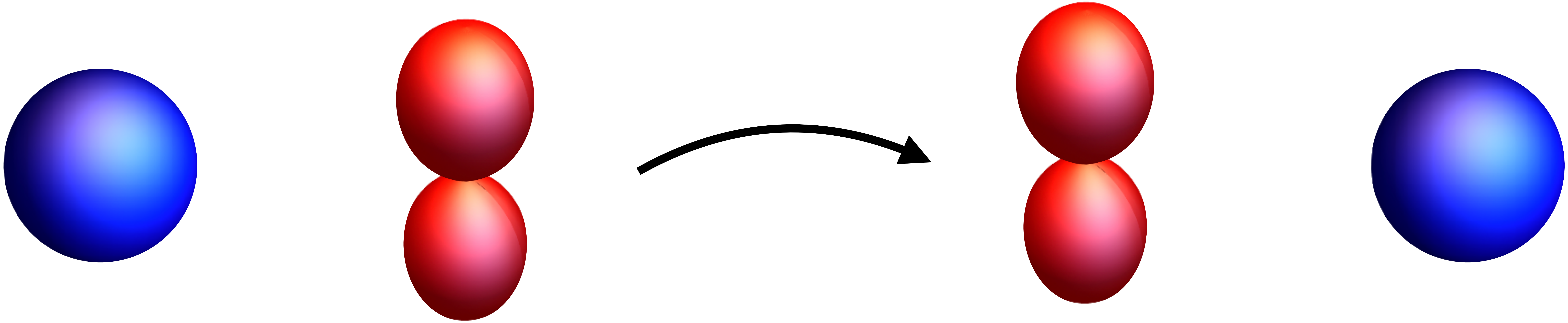
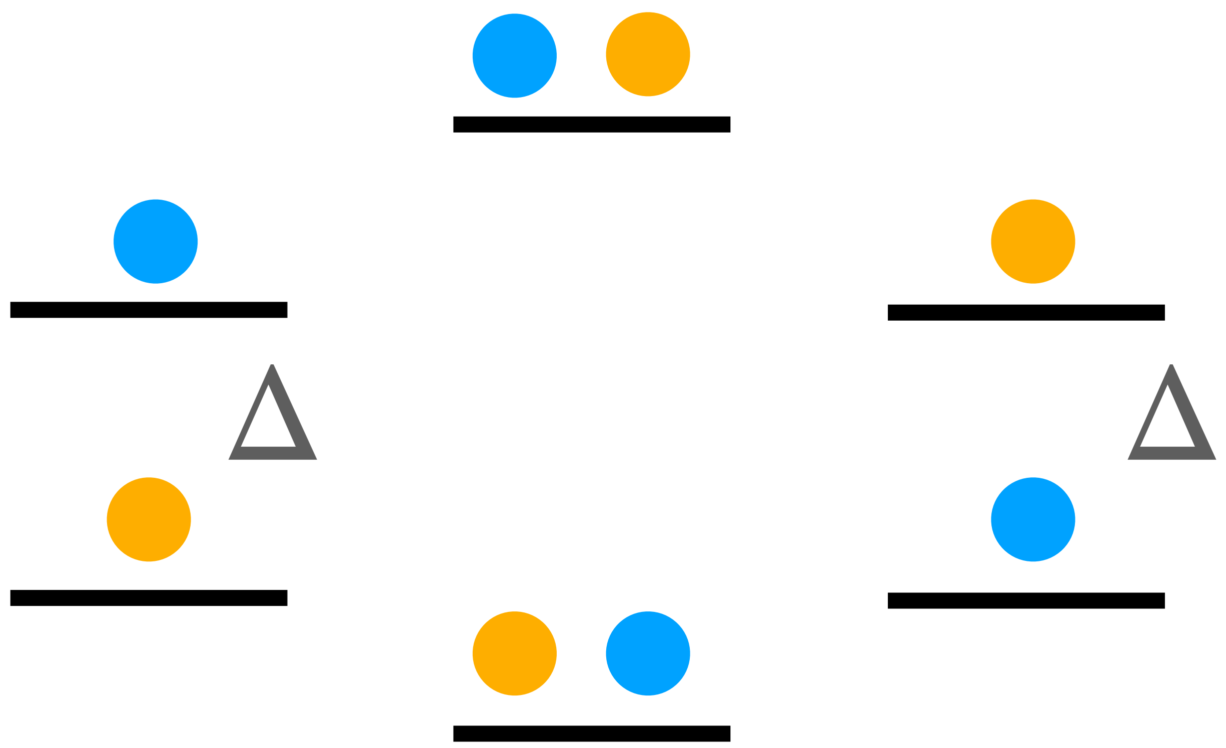


Each atom in a different state:



$$u_{\pm} = \Delta \pm \frac{d_1 d_2}{R^3} \,.$$

This *resonant* dipolar interaction leads to a "flip-flop" or exchange interaction between atoms; in the full picture this interaction is *anisotropic*!

How do Rydberg atoms interact?

The general problem is just a little bit harder:

$$H_{\text{int}}(\vec{R}) = \sum_{\kappa_1,\kappa_2=1}^{\infty} \frac{V_{\kappa_1\kappa_2}}{R^{\kappa_1+\kappa_2+1}}$$

The exact form of $V_{\kappa_1\kappa_2}$ depends on the choice of the coordinate systems used to label the positions of the electrons. If we choose the coordinate systems such that the z-axis points along $\boldsymbol{R}$, i.e. along the interatomic axis, we get the comparatively simple result

$$V_{\kappa_1\kappa_2} = (-1)^{\kappa_2} \sum_{q=-\kappa_<}^{\kappa_<} \sqrt{\binom{\kappa_1+\kappa_2}{\kappa_1+q}\binom{\kappa_1+\kappa_2}{\kappa_2+q}}\, p^{(1)}_{\kappa_1 q} p^{(2)}_{\kappa_2 -q}, \quad (7)$$

where we use $\kappa_< = \min(\kappa_1, \kappa_2)$ and binomial coefficients to shorten our notation.

And the equivalent pieces to our $\vec{r}_1, \vec{r}_2$ are…

$$\hat{p}^{(i)}_{\kappa q} = e\, \hat{r}_i^{\kappa} \cdot \sqrt{\frac{4\pi}{2\kappa+1}}\, Y_{\kappa q}(\hat{\vartheta}_i, \hat{\varphi}_i)$$

How do Rydberg atoms interact?

The general problem is just a little bit harder:

Instead of dipole moments d we have…

$$\langle lsjm_j|\hat{T}_{\kappa q}|l's'j'm_j'\rangle = (-1)^{j-m_j}(lsj||\hat{T}_{\kappa 0}||l's'j')\begin{pmatrix} j & \kappa & j' \\ -m_j & q & m_j' \end{pmatrix}$$

$$(lsj||\hat{T}_{\kappa 0}||l'sj') = (-1)^{l+s+j'+\kappa}(l||\hat{T}_{\kappa 0}||l')\sqrt{(2j+1)(2j'+1)} \times \begin{Bmatrix} l & j & s \\ j' & l' & \kappa \end{Bmatrix},$$

How do Rydberg atoms interact?

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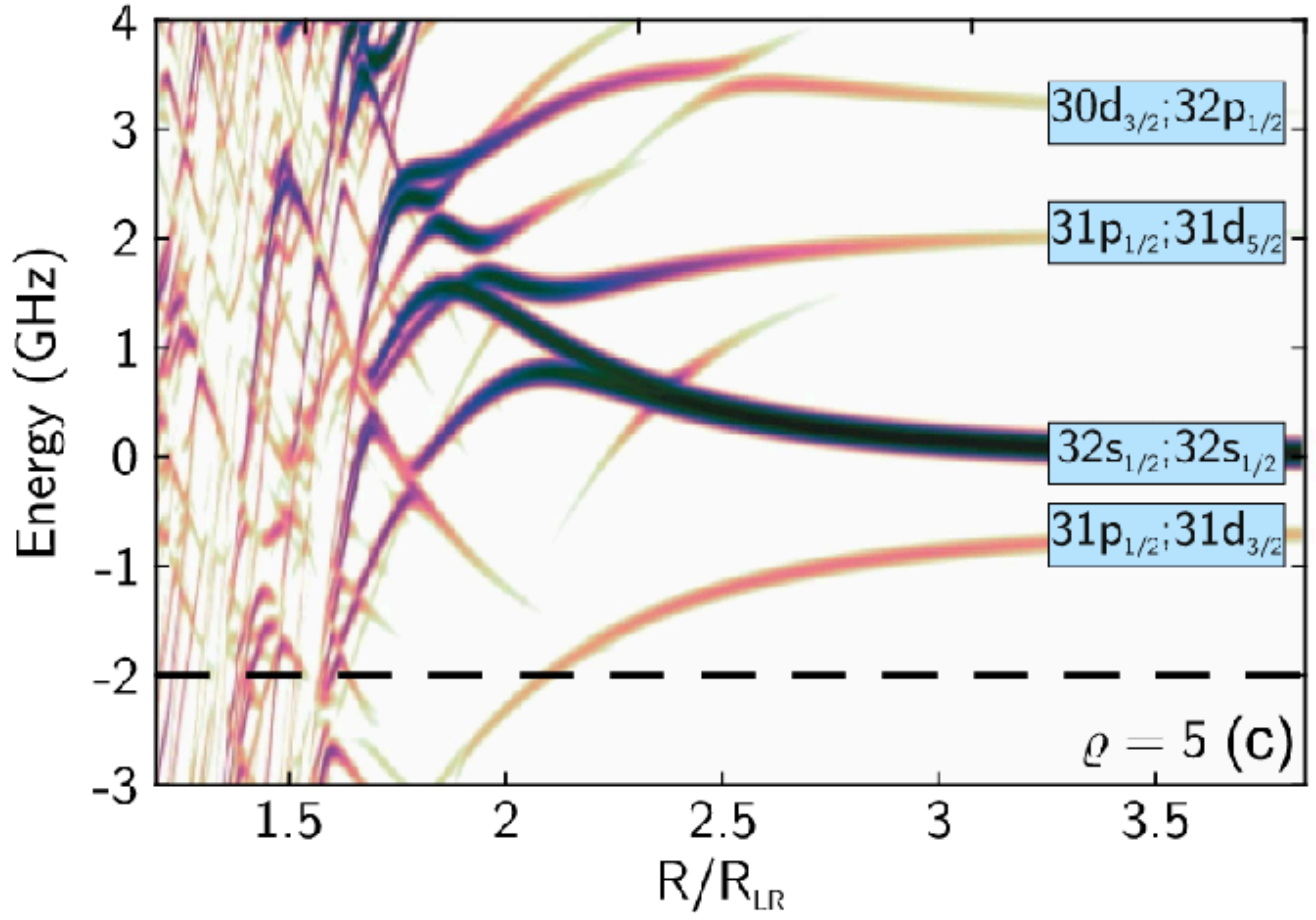
https://github.com/pairinteraction/pairinteraction



IOP Publishing — Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **50** (2017) 133001 (18pp) https://doi.org/10.1088/1361-6455/aa743a

Tutorial

Calculation of Rydberg interaction potentials

Sebastian Weber[1,7], Christoph Tresp[2,3], Henri Menke[4], Alban Urvoy[2,5], Ofer Firstenberg[6], Hans Peter Büchler[1] and Sebastian Hofferberth[2,3,7]

Scope of today's lecture

At the core of quantum simulation with Rydberg atoms: 150 years of spectroscopy

- From Rydberg to Pauli/Schrödinger to present day

As billed, it is a "lecture":

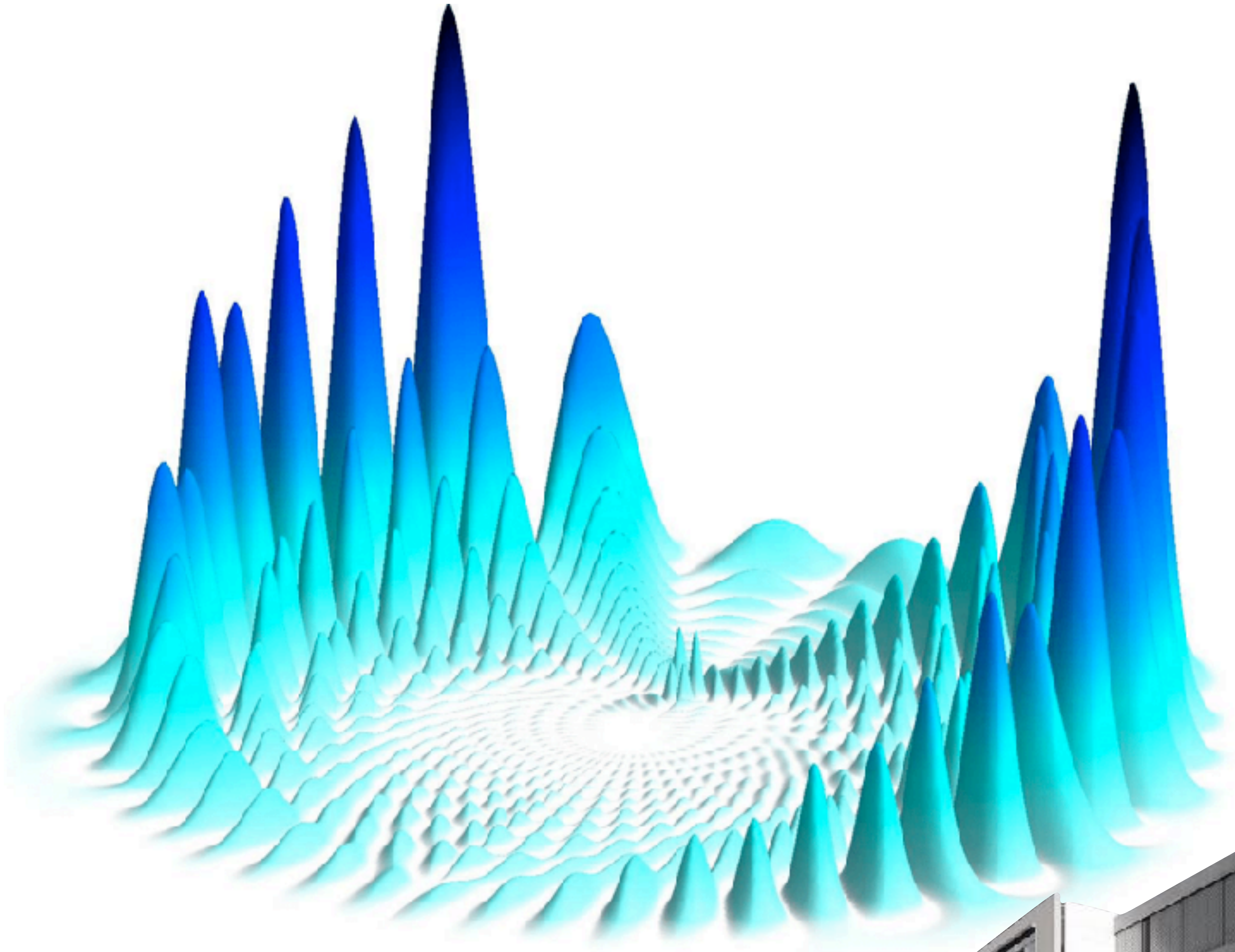
- …expect some equations…but hopefully not too many
- slides: https://www.pks.mpg.de/correlations-and-transport-in-rydberg-matter

What are Rydberg atoms?

- Quantum defect theory: alkali atoms
- Key properties of Rydberg atoms
- Multichannel quantum defect theory: many-electron atoms

What are they good for?

- Rydberg-Rydberg interactions
  - van der Waals / Rydberg blockade
  - dipole-dipole / "flip-flop" interactions
- Rydberg-ground-state-atom interactions



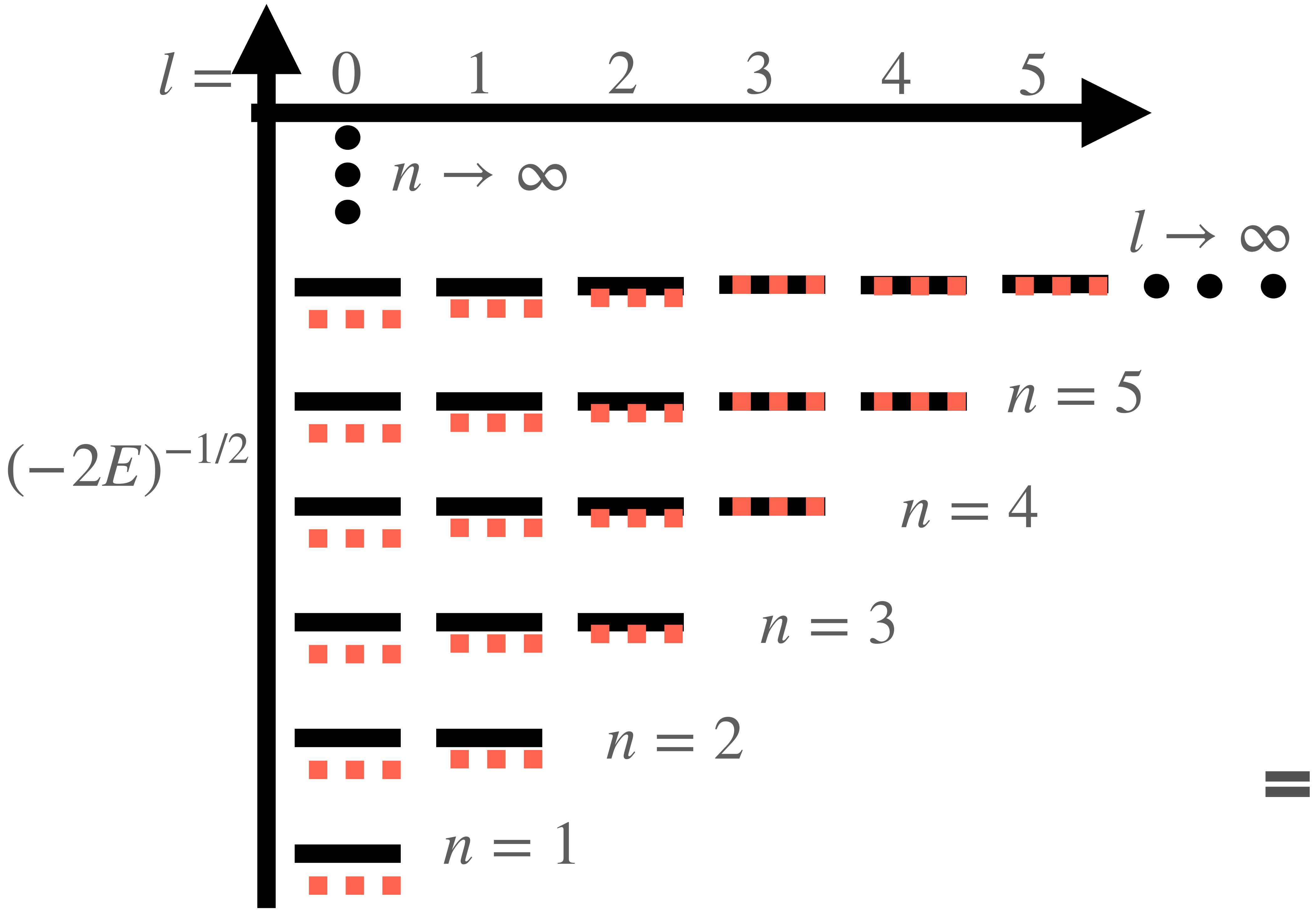
*we didn't actually discuss this. it's just to get your attention

For more details: feel free to shoot me an email at meiles@pks.mpg.de

Rydberg molecules, polarons, and composites

Two **ingredients** make a **long-range Rydberg molecule:**

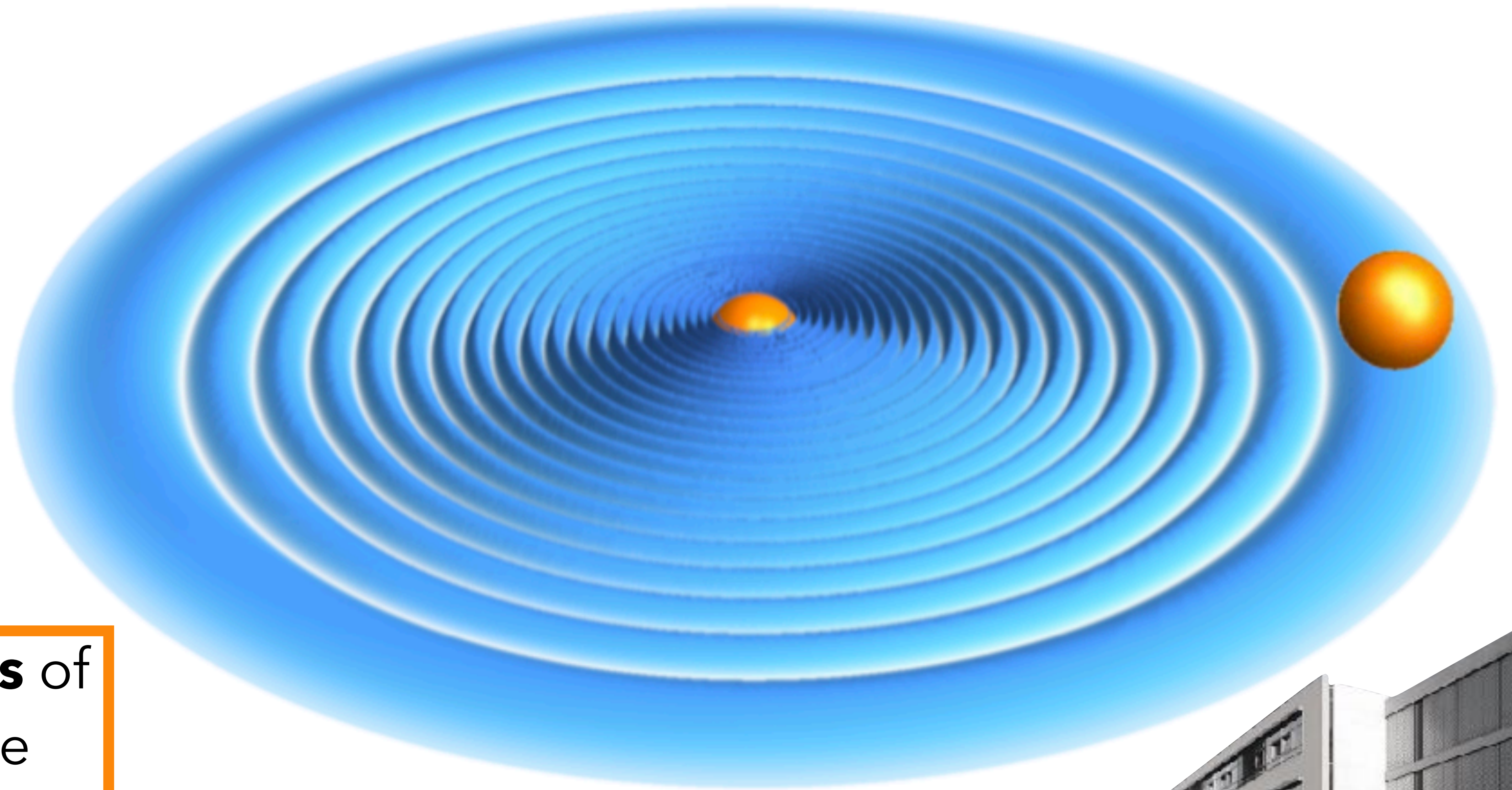
Rydberg atom + A ground state atom



$$V(\vec{r}, \overrightarrow{R}) = 2\pi a_e \delta^3(\vec{r} - \overrightarrow{R}) \,.$$

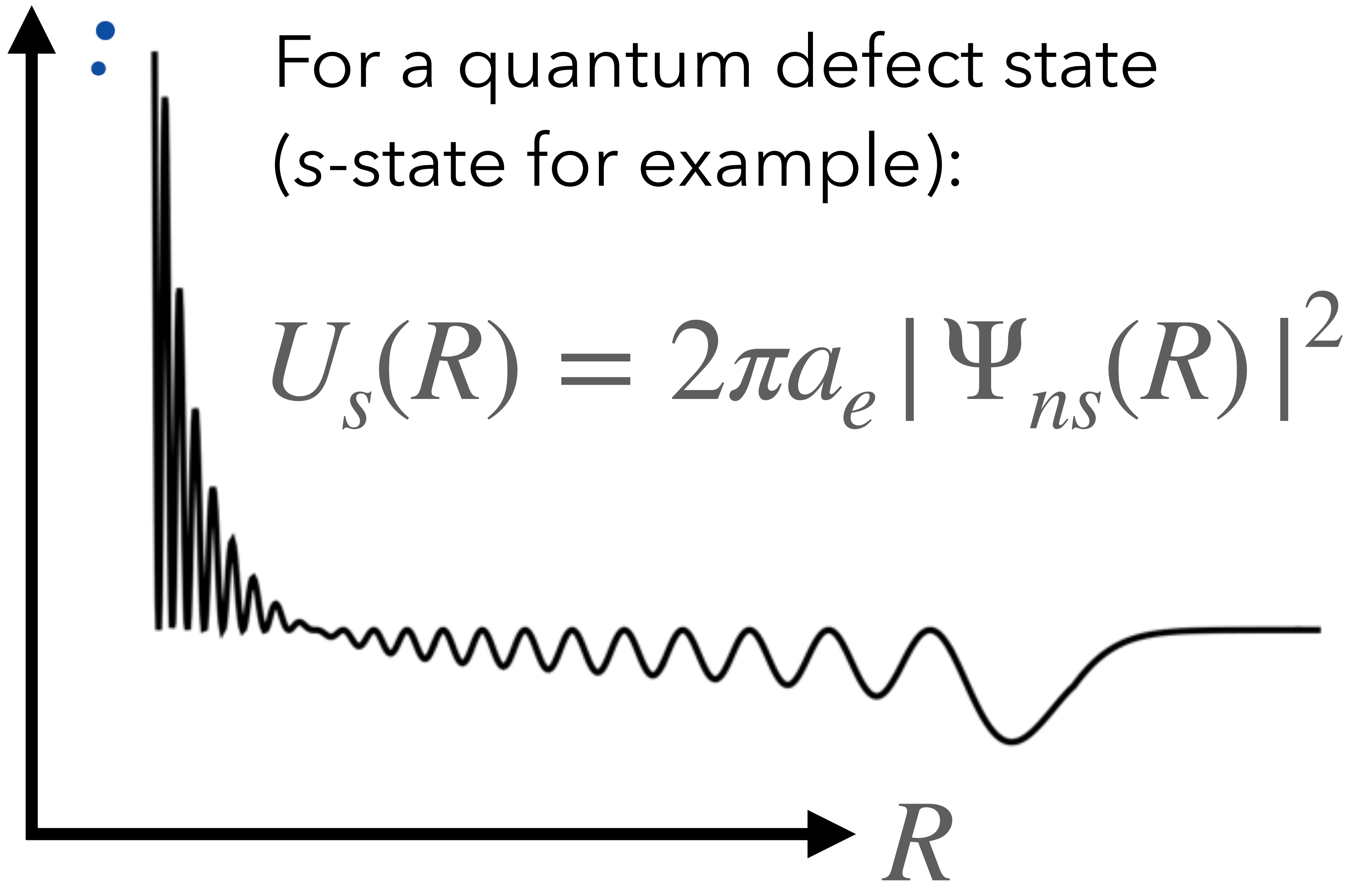
(electron-atom interaction given by Fermi's pseudopotential)

=



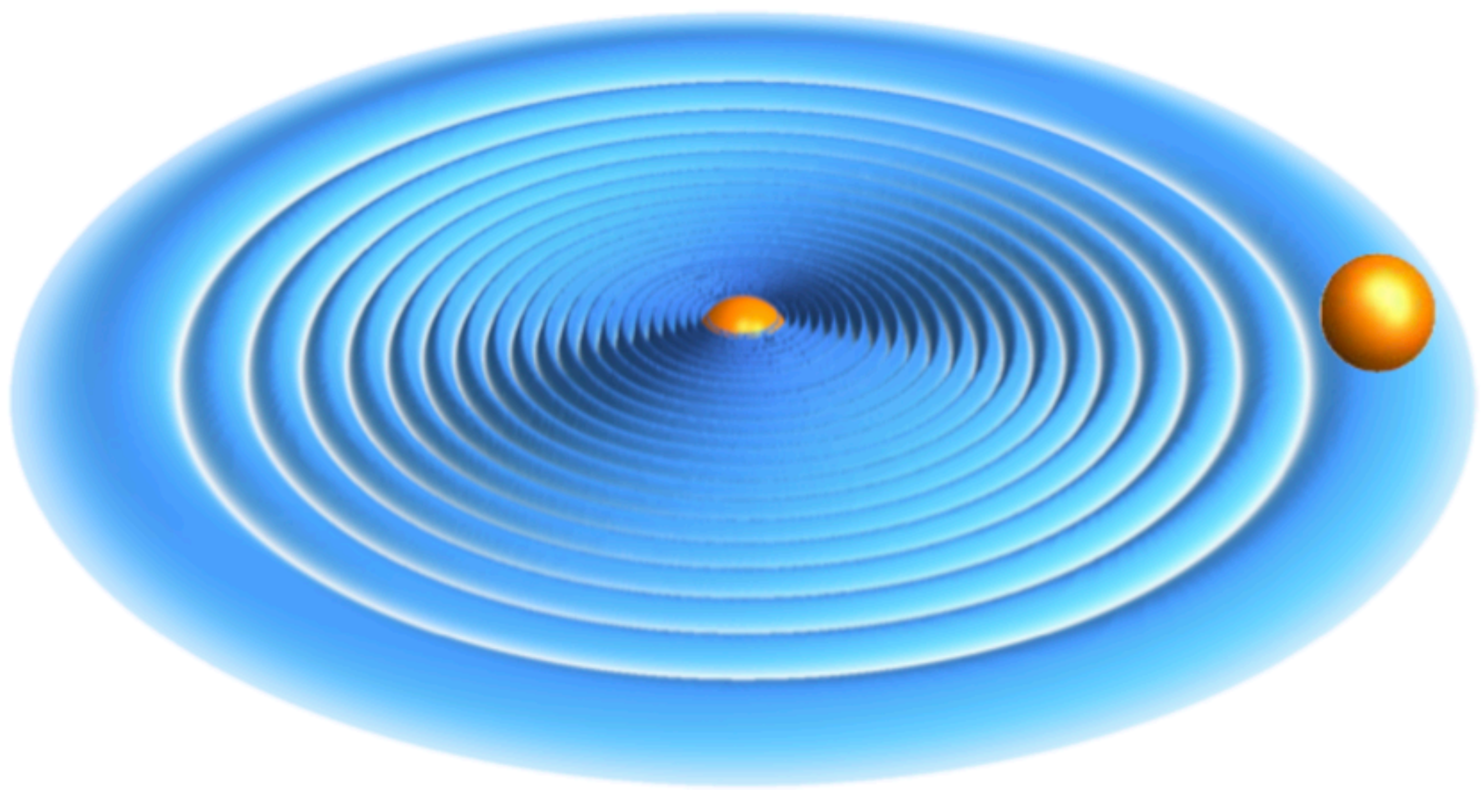
Two **types** of molecules exist because there are two **types** of Rydberg states: te high-l states remain hydrogenic and the SO(4) symmetry is partially preserved!

Rydberg molecules, polarons, and composites



For a quantum defect state
(*s*-state for example):

$$U_s(R) = 2\pi a_e |\Psi_{ns}(R)|^2$$



Rydberg molecules, polarons, and composites

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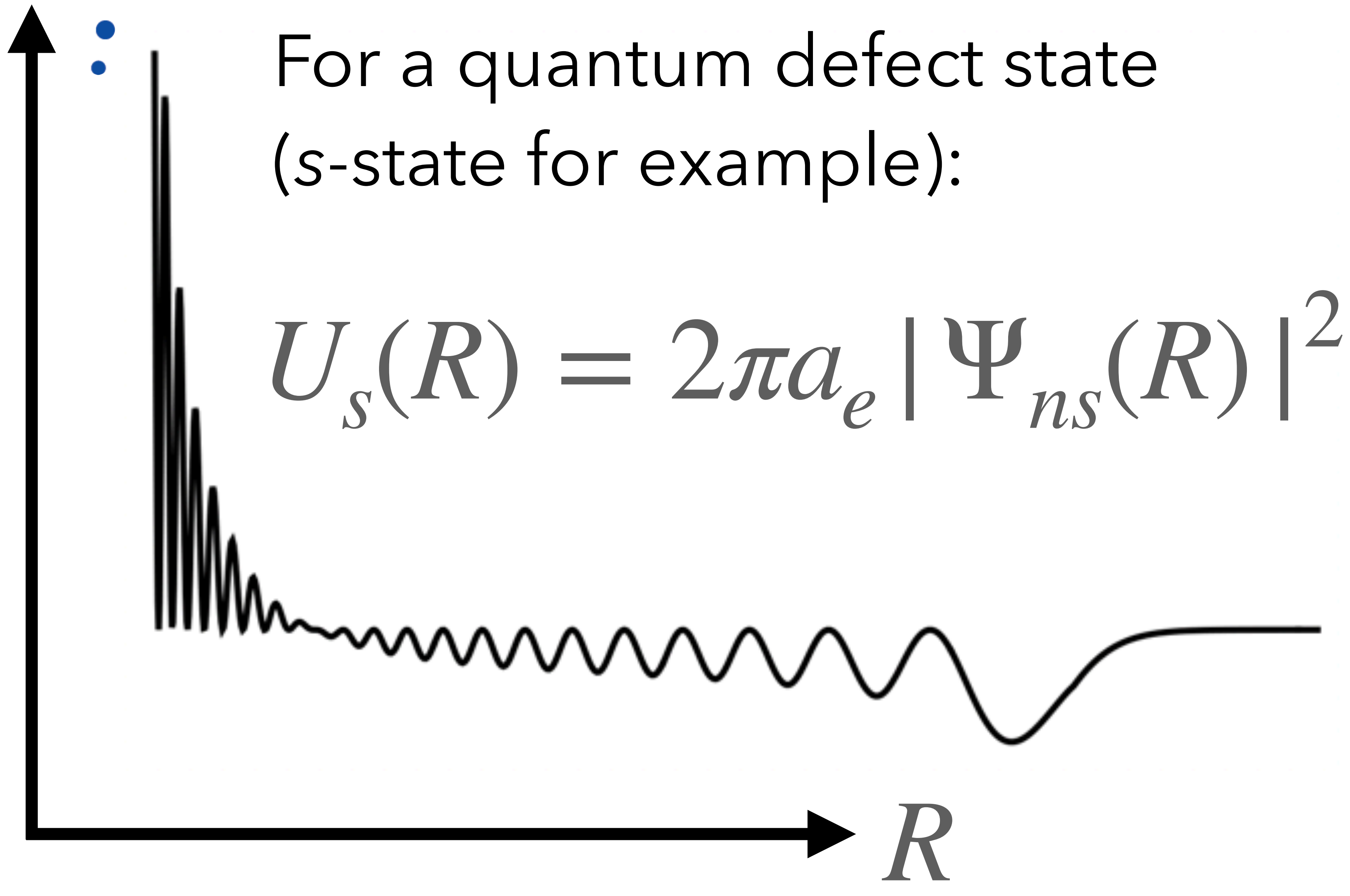
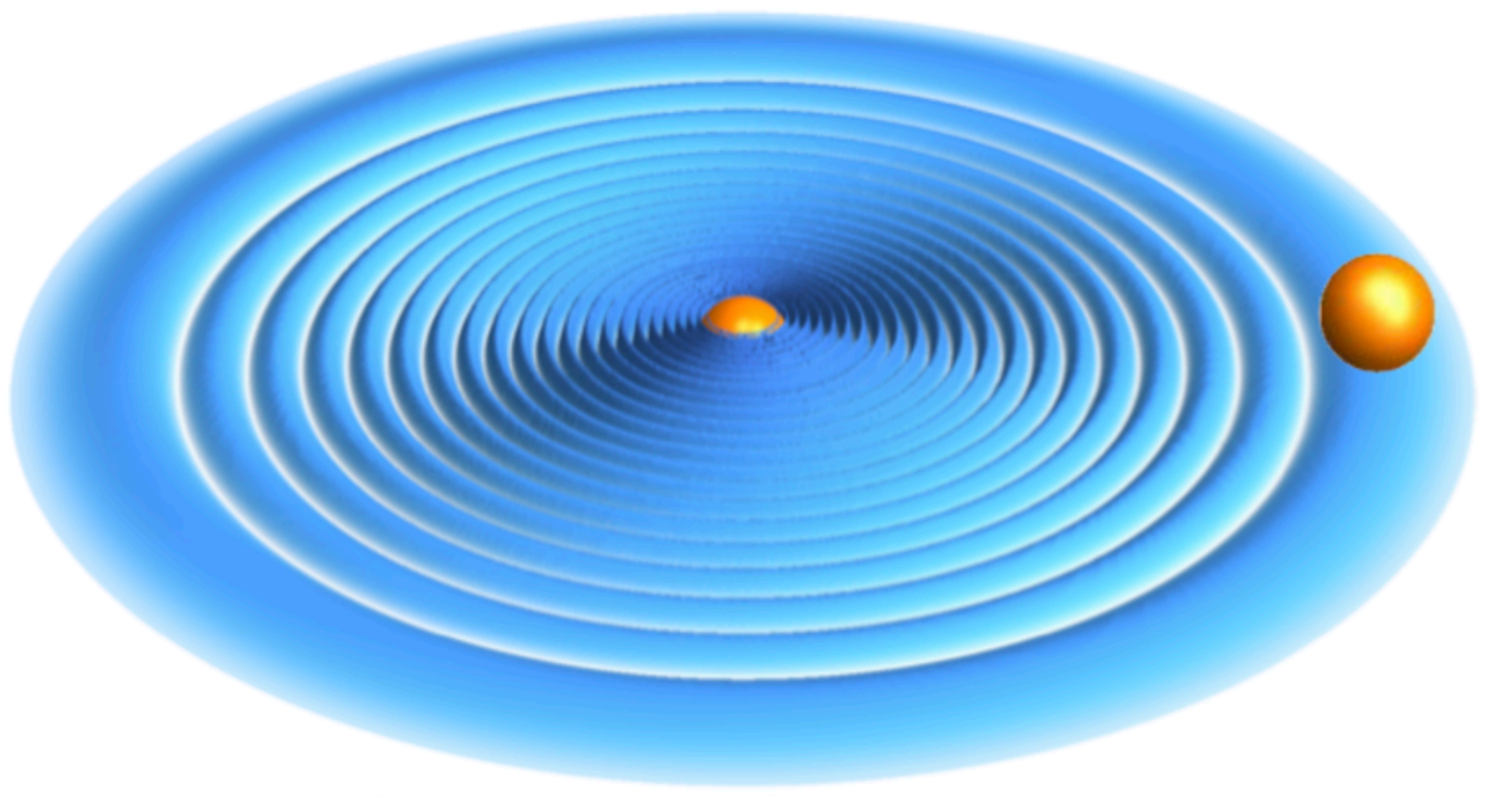
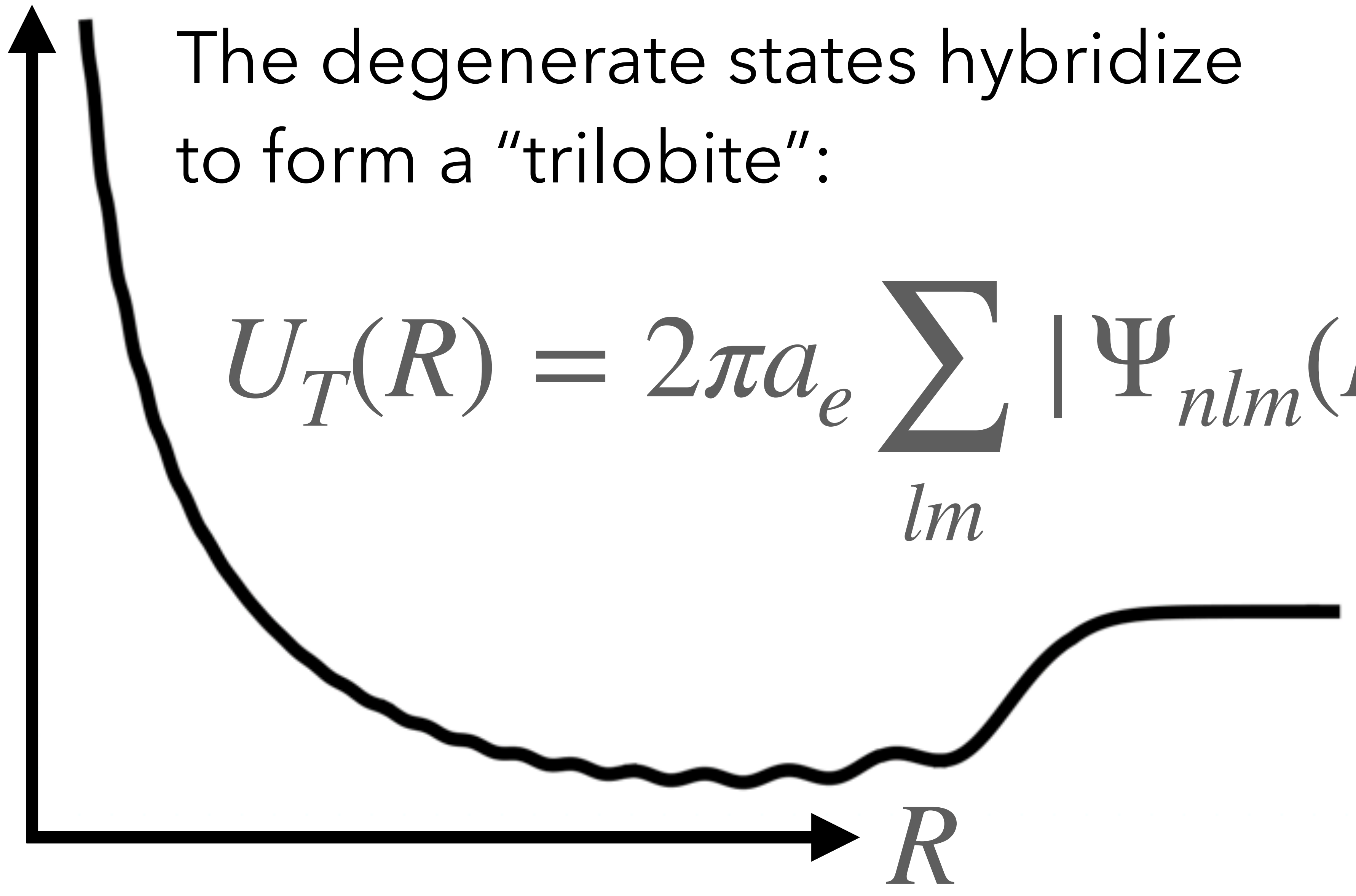
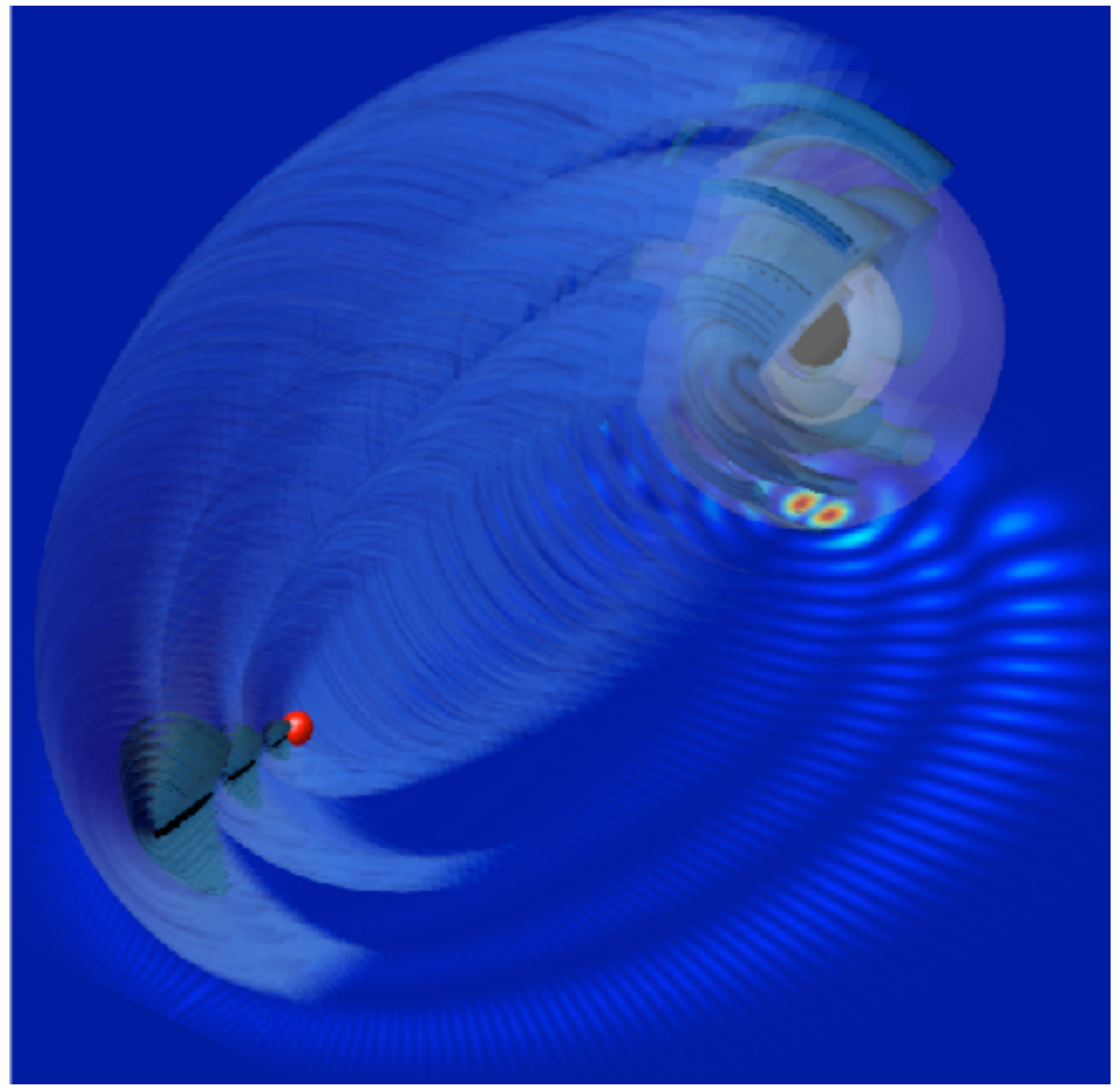




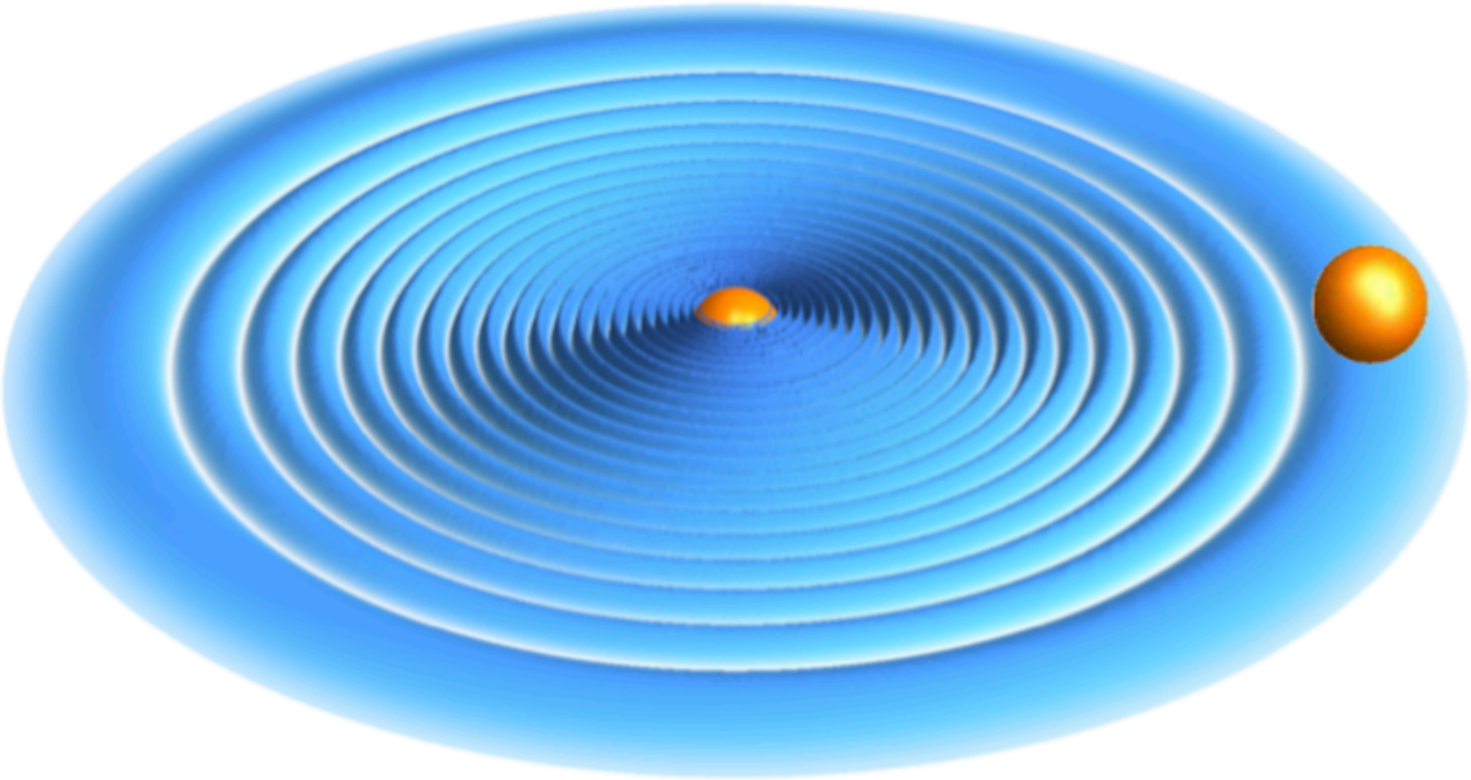
The degenerate states hybridize
to form a “trilobite”:

$$U_T(R) = 2\pi a_e \sum_{lm} |\Psi_{nlm}(R)|^2$$



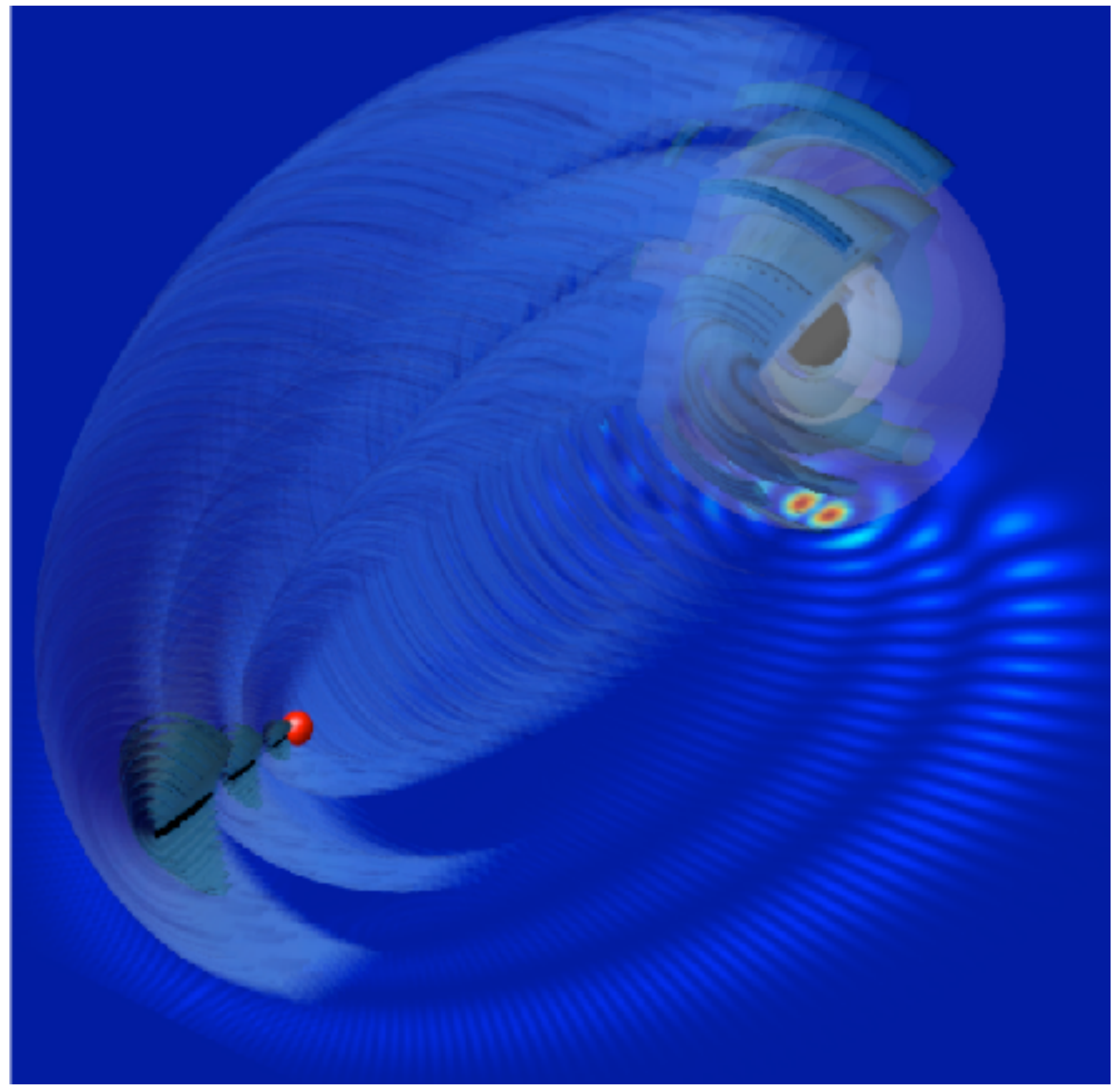


Rydberg molecules, polarons, and composites



Rydberg molecule **with** a quantum defect: simple electronic structure (no back-action on the electron); interaction with each ground state atom in a gas is independent of the others: **polaron physics.**

- **Simple** electronic dynamics, **complex** atomic dynamics



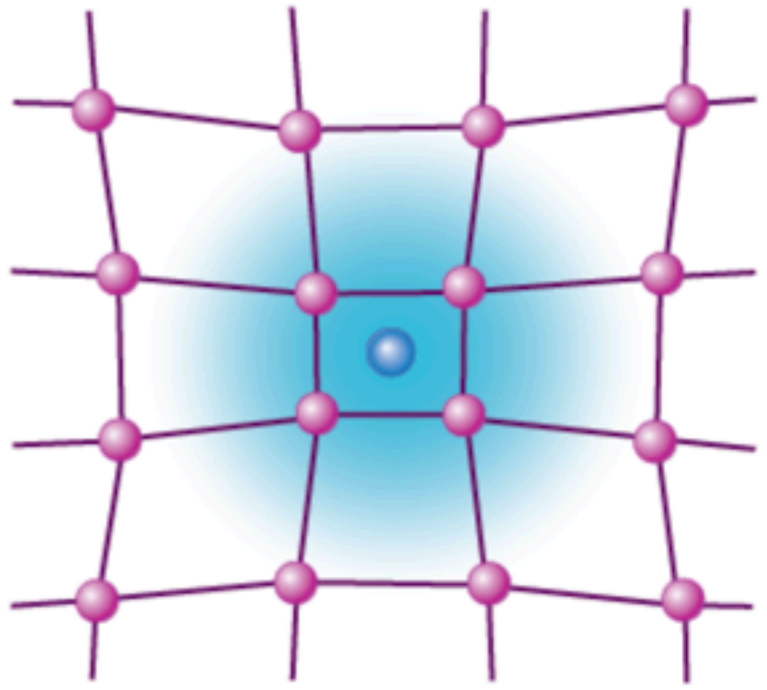
Rydberg molecule **without** a quantum defect: electronic character is sculpted by the ground state atom; each atom added modifies the potential all of the rest feel: **Rydberg composites**

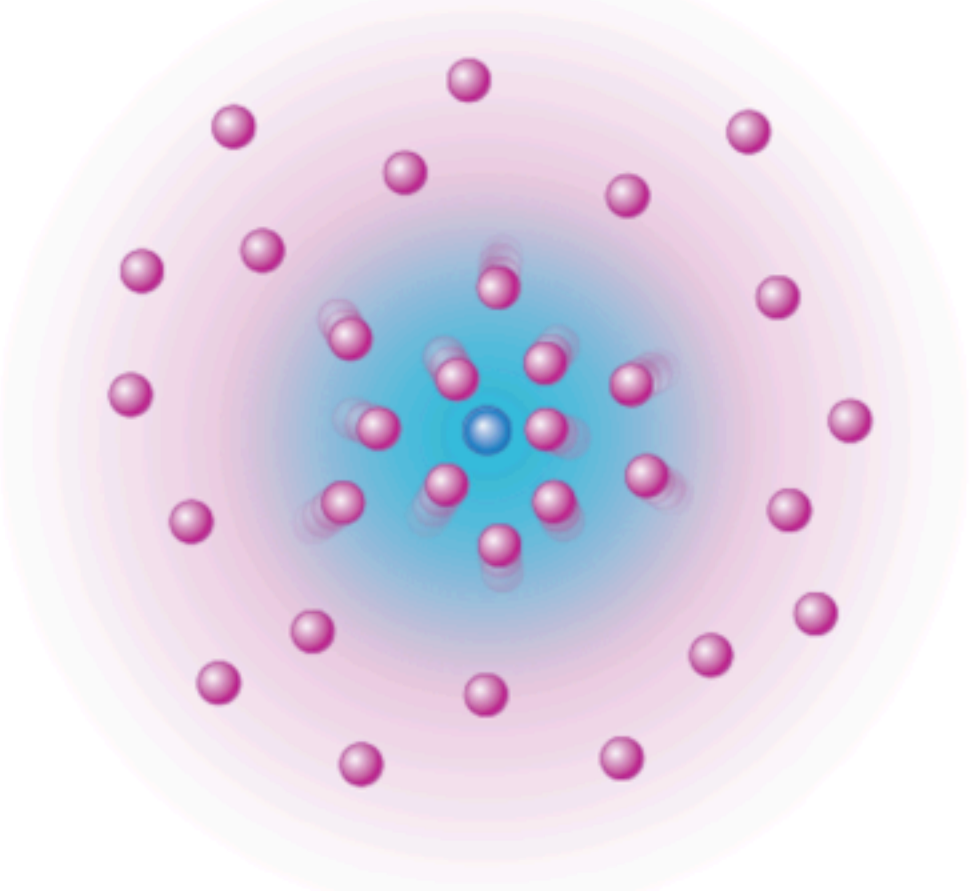
- **Complex** electronic dynamics, **simple** atomic dynamics

Rydberg molecules, polarons, and composites

- **An impurity particle interacts with a non-interacting BEC at T=0**

$$H = \sum_k \frac{k^2}{2M} d_k^\dagger d_k + \sum_k \frac{k^2}{2M} b_k^\dagger b_k + \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} V(\mathbf{q}) d_{\mathbf{k}-\mathbf{q}}^\dagger d_{\mathbf{k}} b_{\mathbf{k}'+\mathbf{q}}^\dagger b_{\mathbf{k}'}$$





- **atomic impurity:** short-ranged interactions

$$V(r) \equiv V_{sr}(r) = \frac{2\pi a_{IB}}{M}\delta(r)$$

- **Rydberg impurity:** long-ranged interactions

$$V(r) \equiv V_{\text{Ryd}}(r) = \frac{2\pi a_e}{m_e} |\psi_{n00}(r)|^2 .$$

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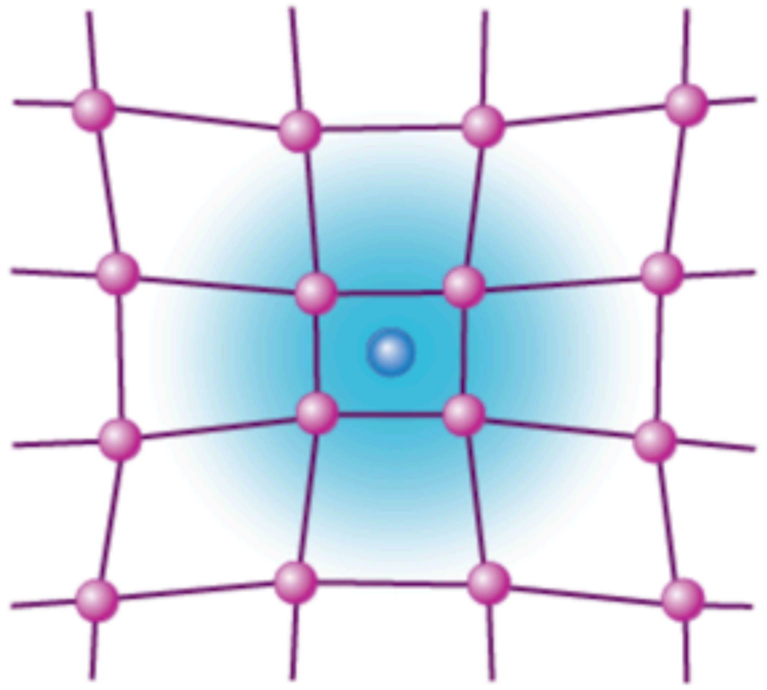

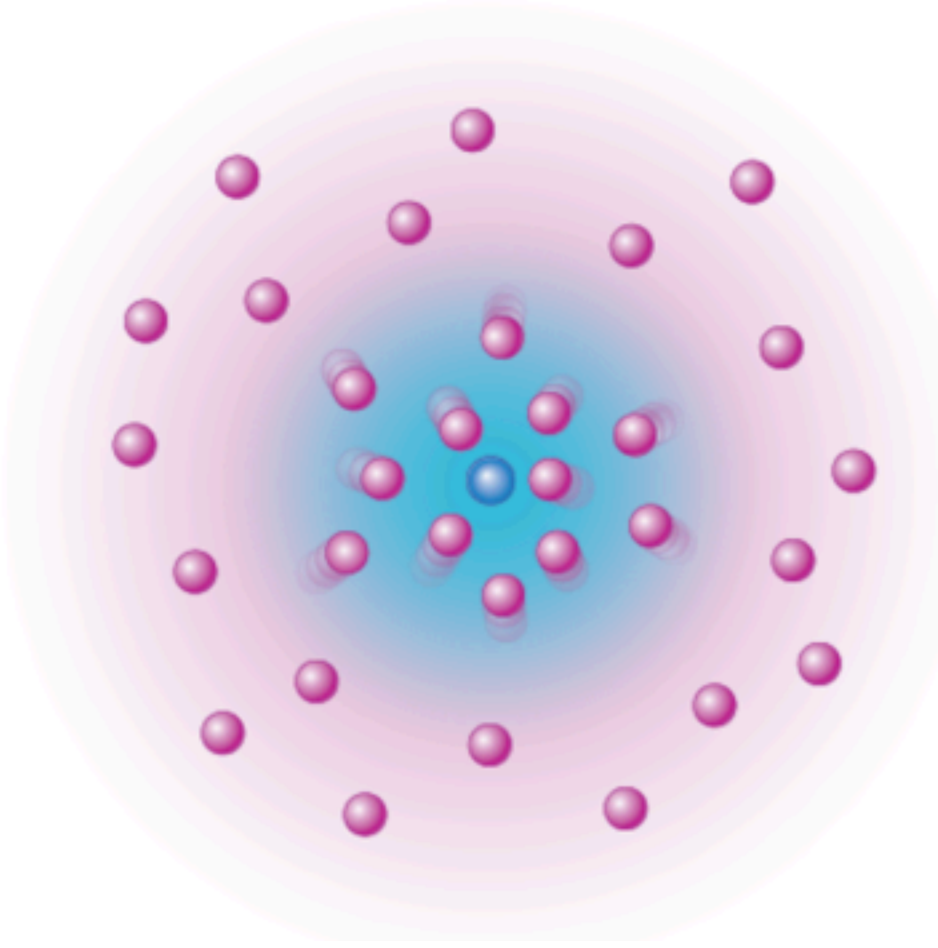

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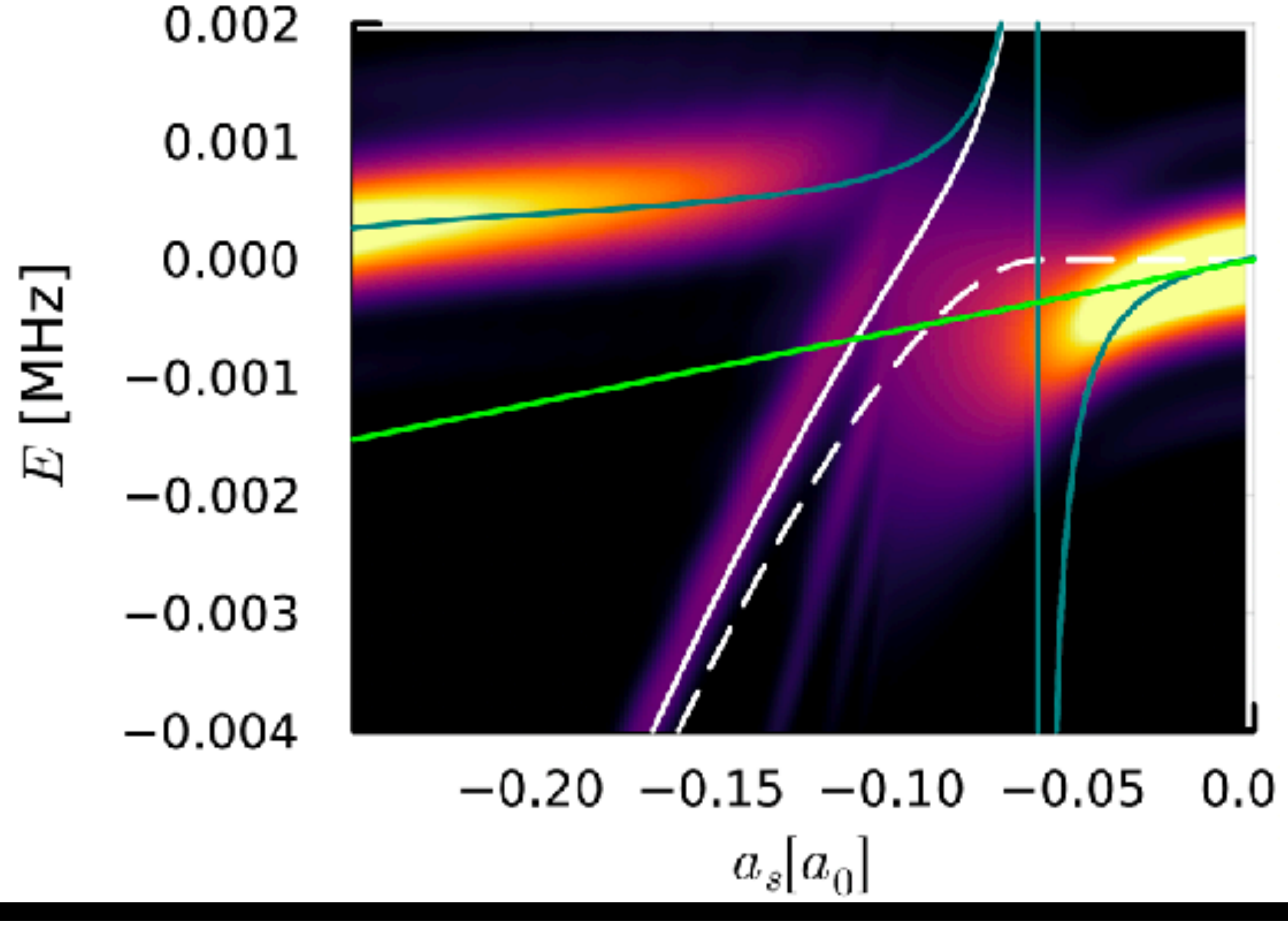

In the right limit, the Rydberg polaron behaves identically to the “normal” Bose polaron.

Rydberg molecules, polarons, and composites

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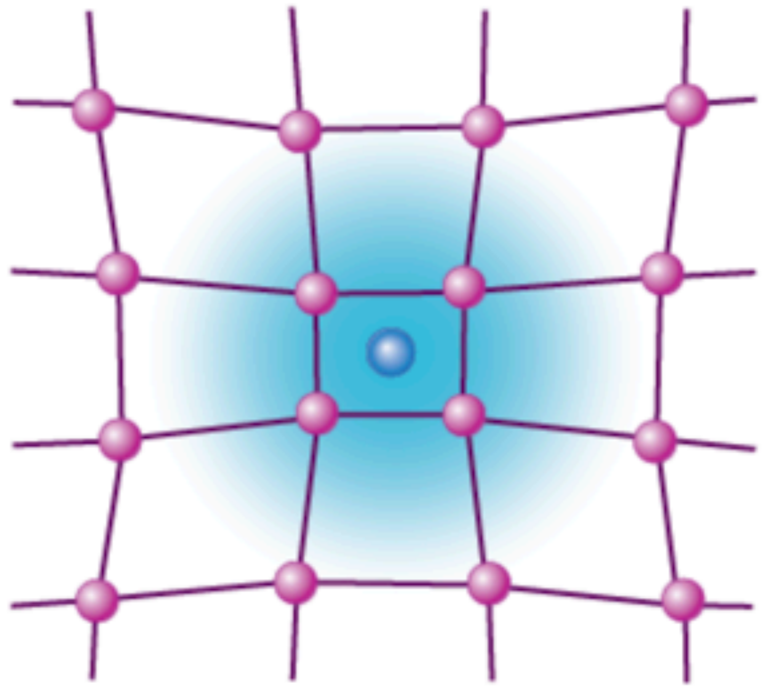

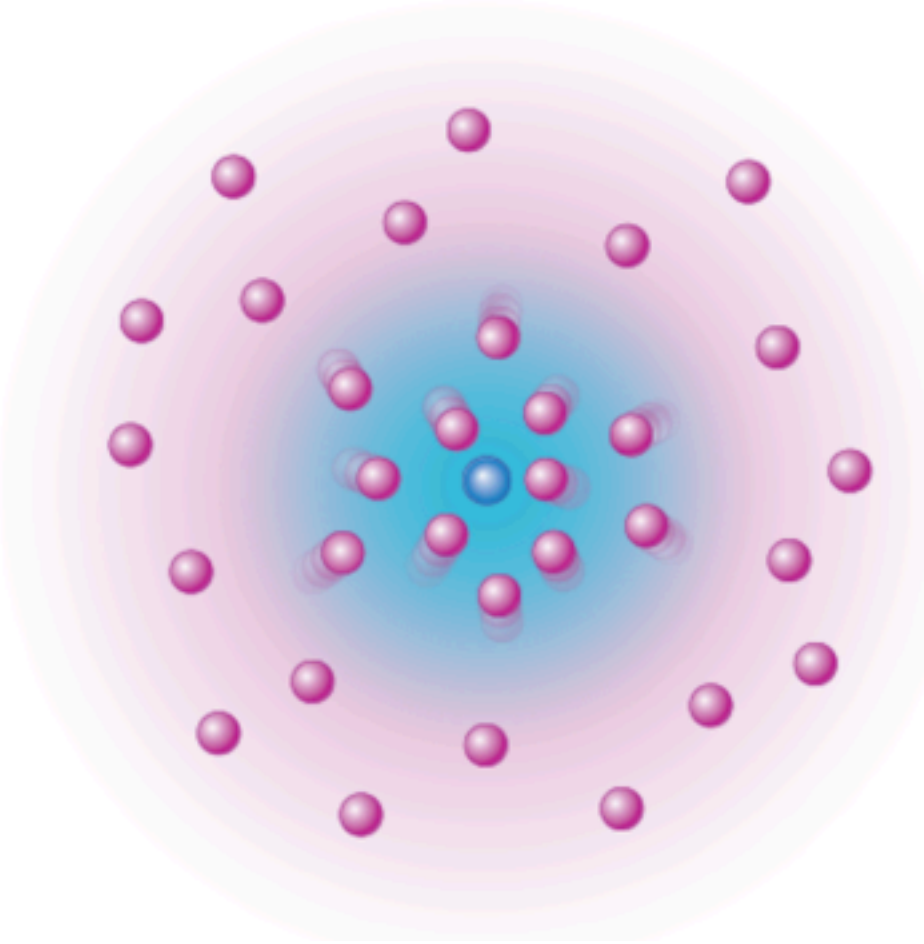

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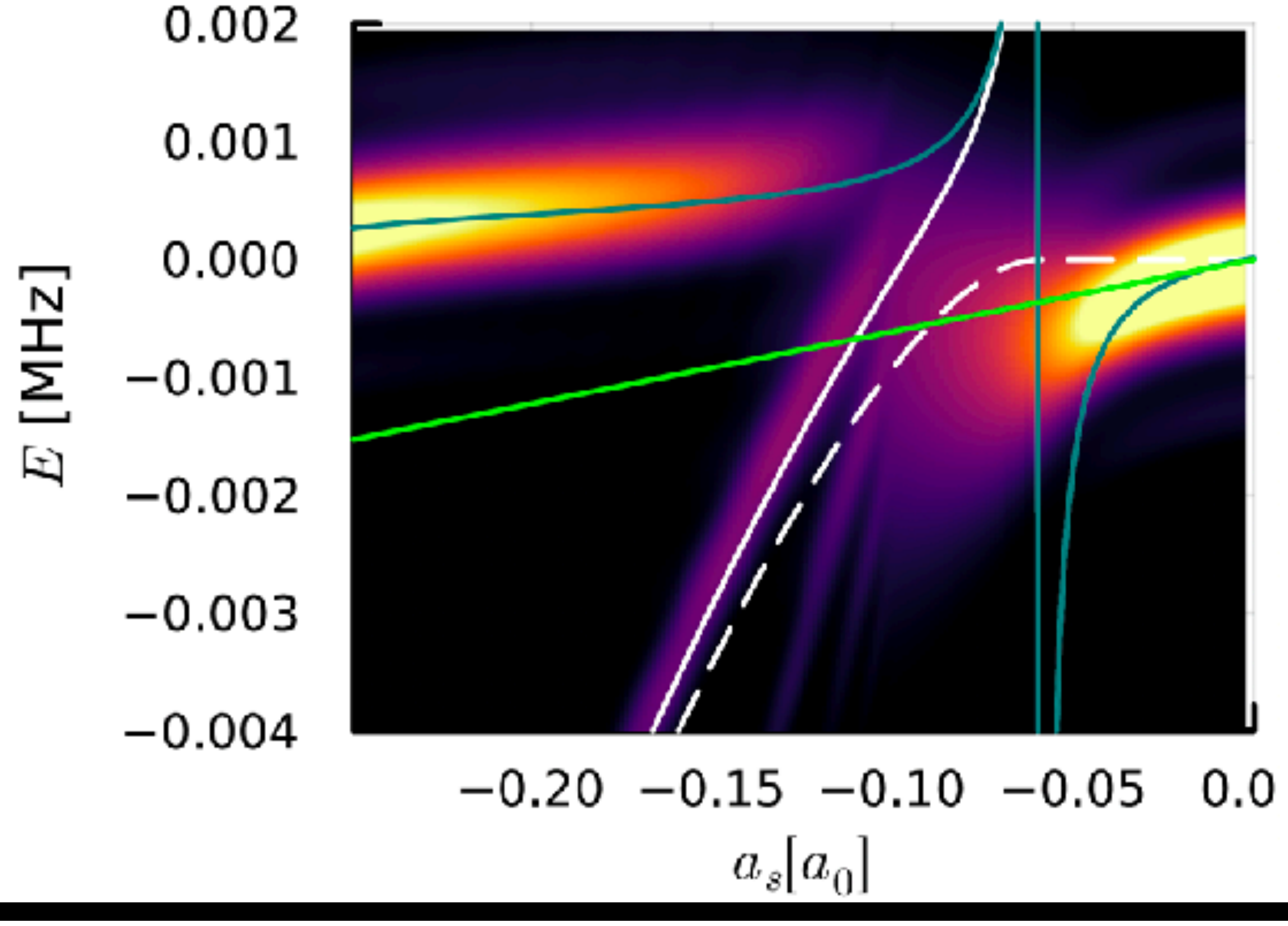

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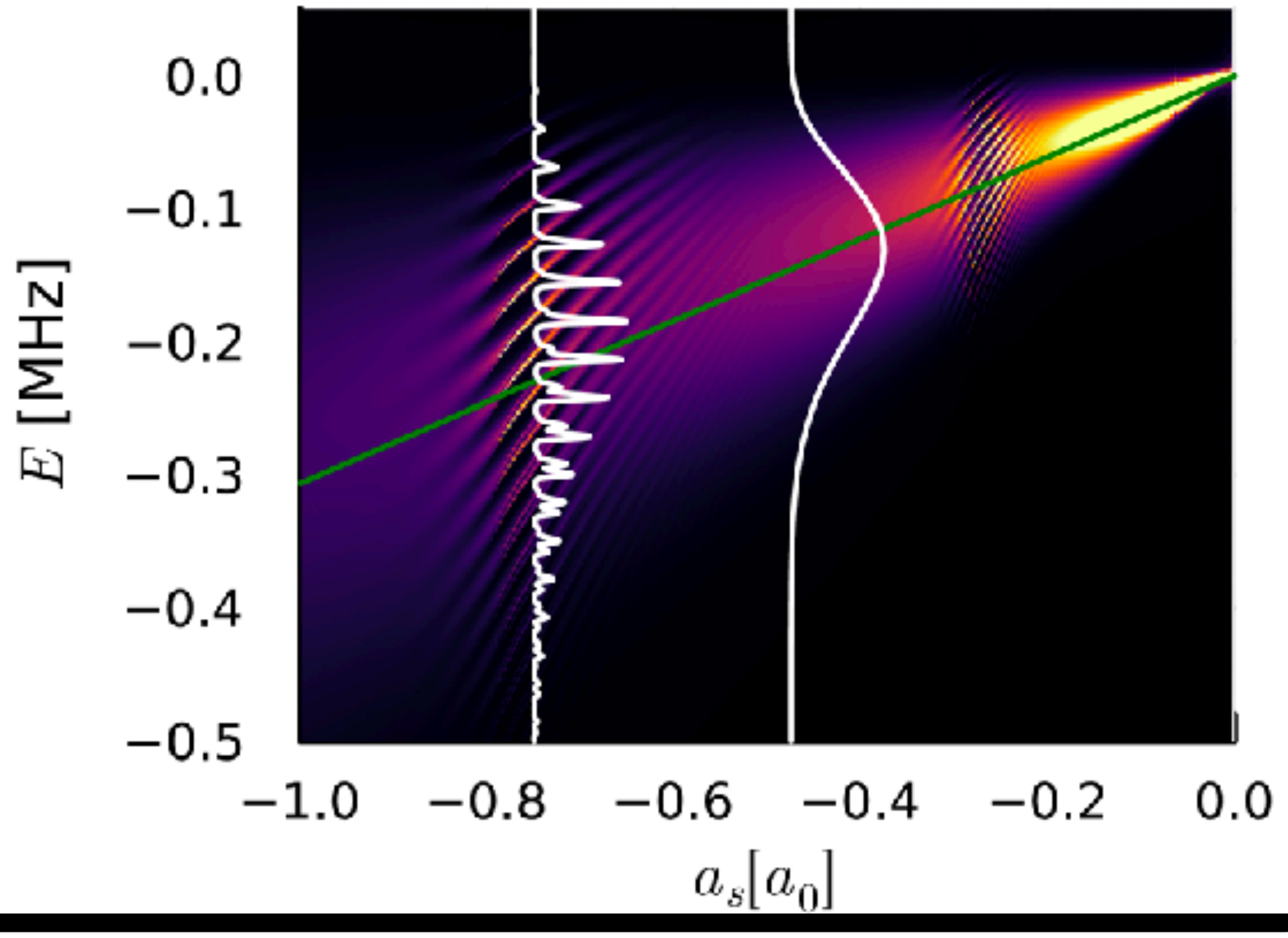


…but it can do lots more!

A. A. T. Durst and **MTE** *in prep*

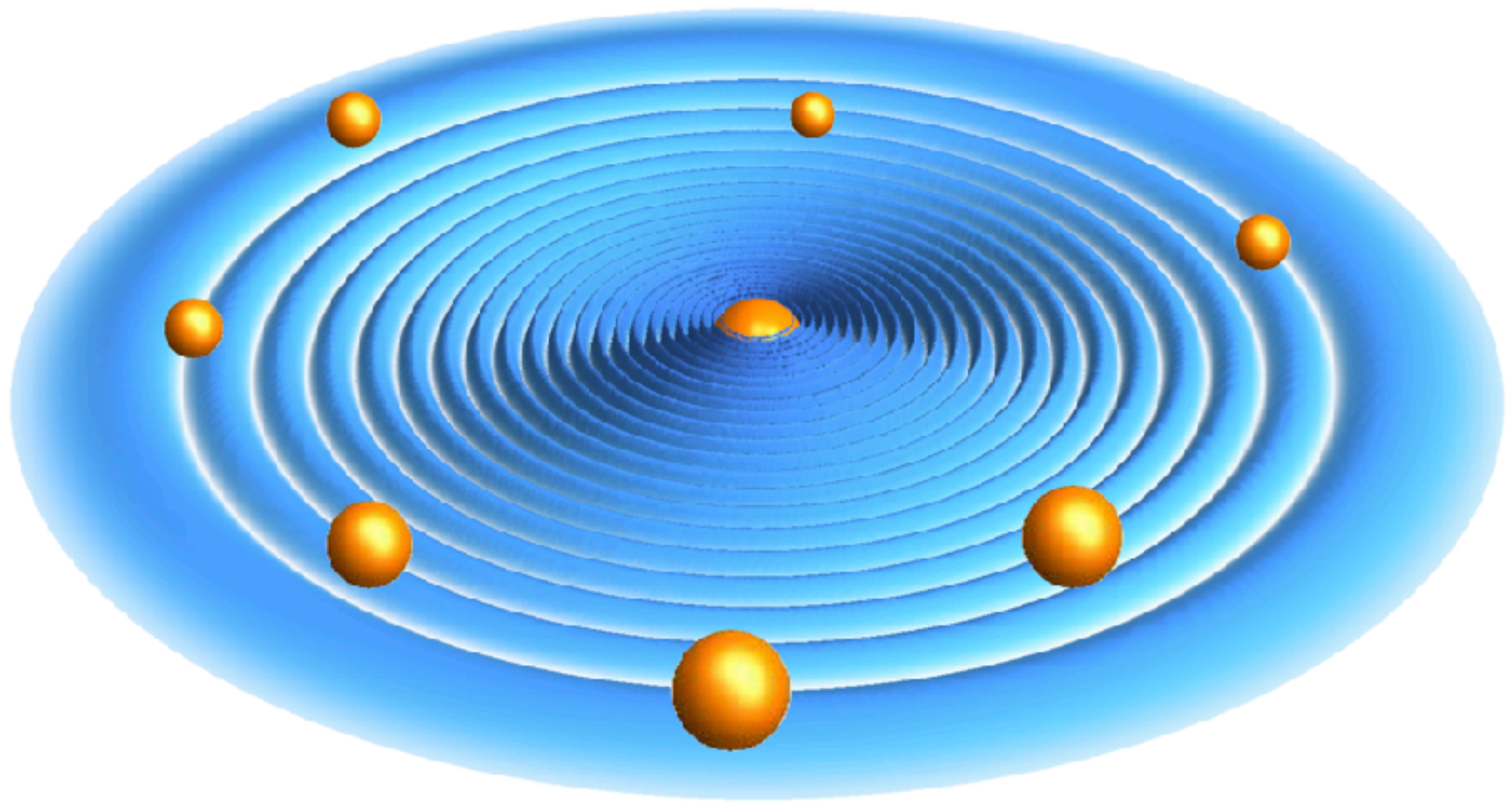
Rydberg molecules, polarons, and composites

"Rydberg composite" Hamiltonian

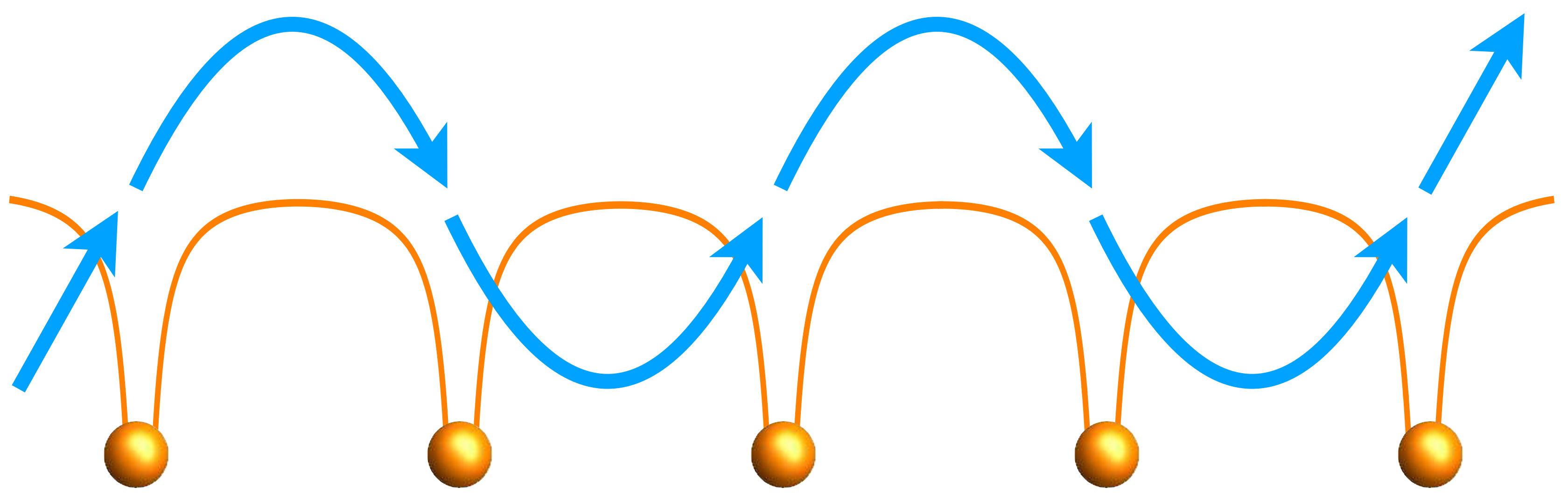
$$H_{RC} = -\frac{\nabla^2}{2} - \frac{1}{r} + 2\pi \sum_{q=1}^{M} a_e \delta^3(\vec{r} - \vec{R}_q)$$

Tight-binding Hamiltonian

$$H_{TB} = \sum_{q=1}^{M} E_q |q\rangle\langle q| + \sum_{q=1}^{M} \sum_{q' \neq q}^{M} V_{qq'} |q\rangle\langle q'|$$



M ground state atoms immersed in a Rydberg electron's wave function



A "particle" hopping through a lattice of M sites

this is made possible by the "accidental *degeneracy*" of the Coulomb potential.

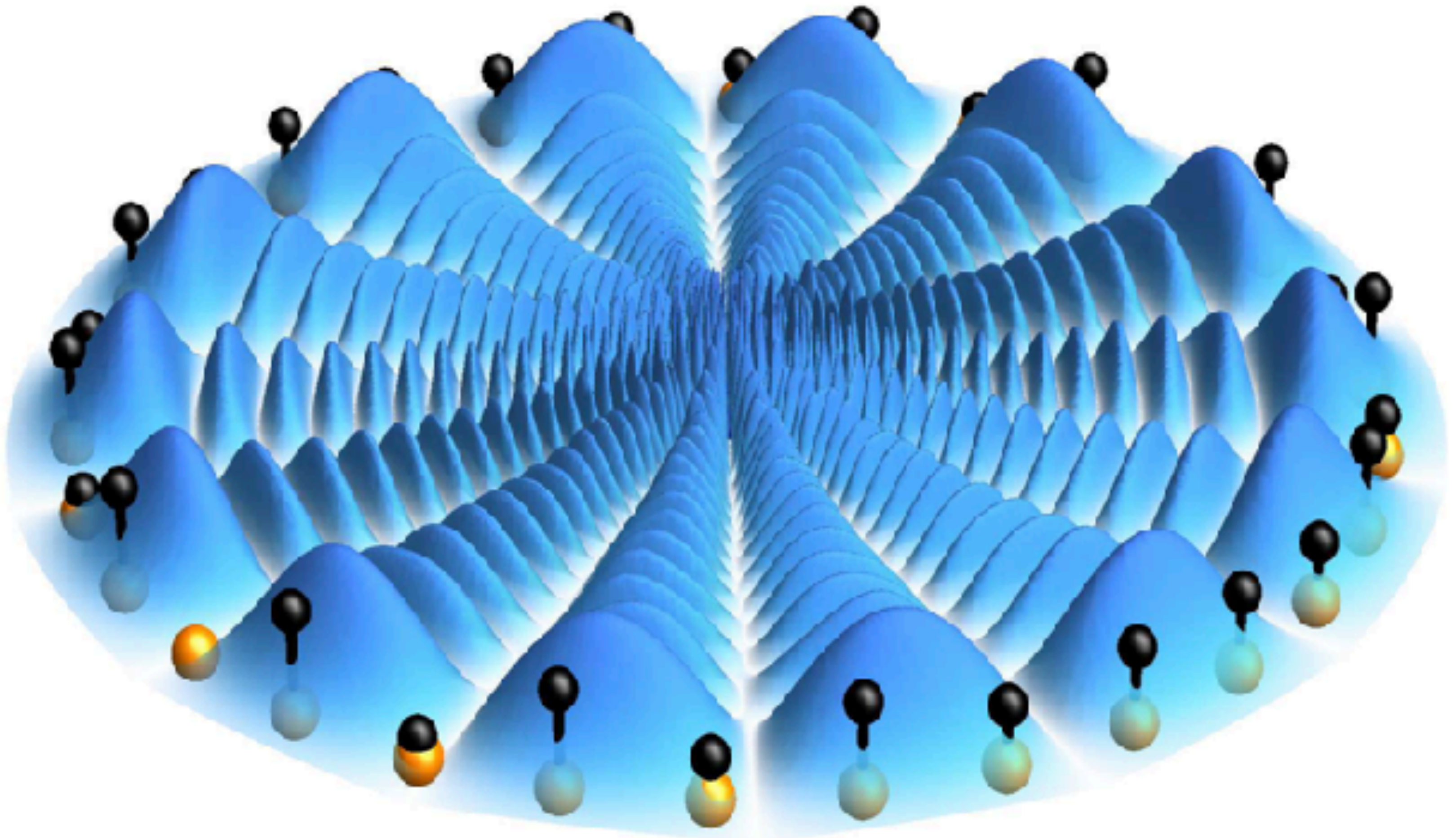
MTE, A. Eisfeld, J. M. Rost PRR **5**, 033032 (2023)

MTE, C. W. Wächtler, A. Eisfeld, J. M. Rost arXiv:2309.03039

Rydberg molecules, polarons, and composites

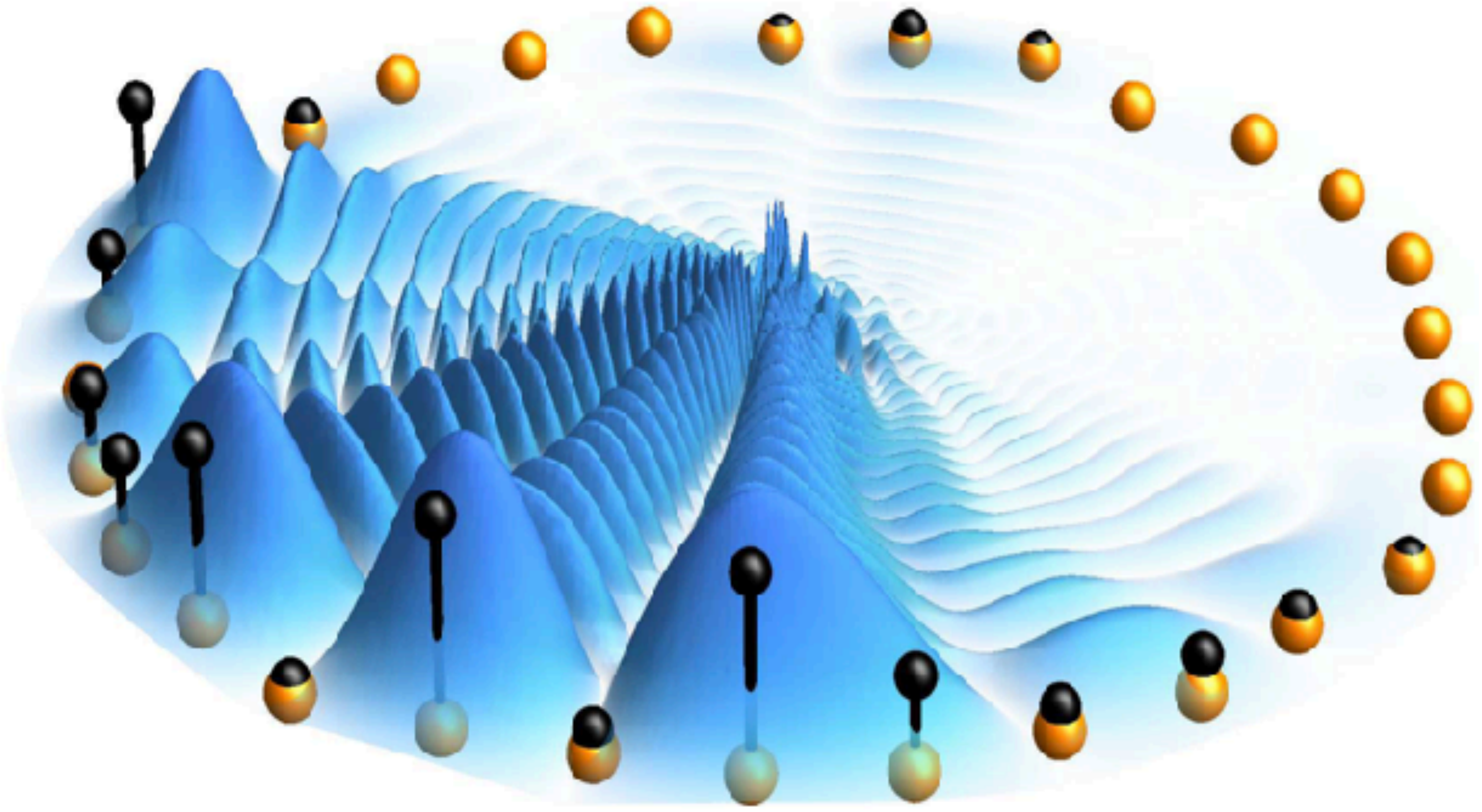
When you get a tight-binding Hamiltonian, why not study Anderson localization and disordered systems?

Extended



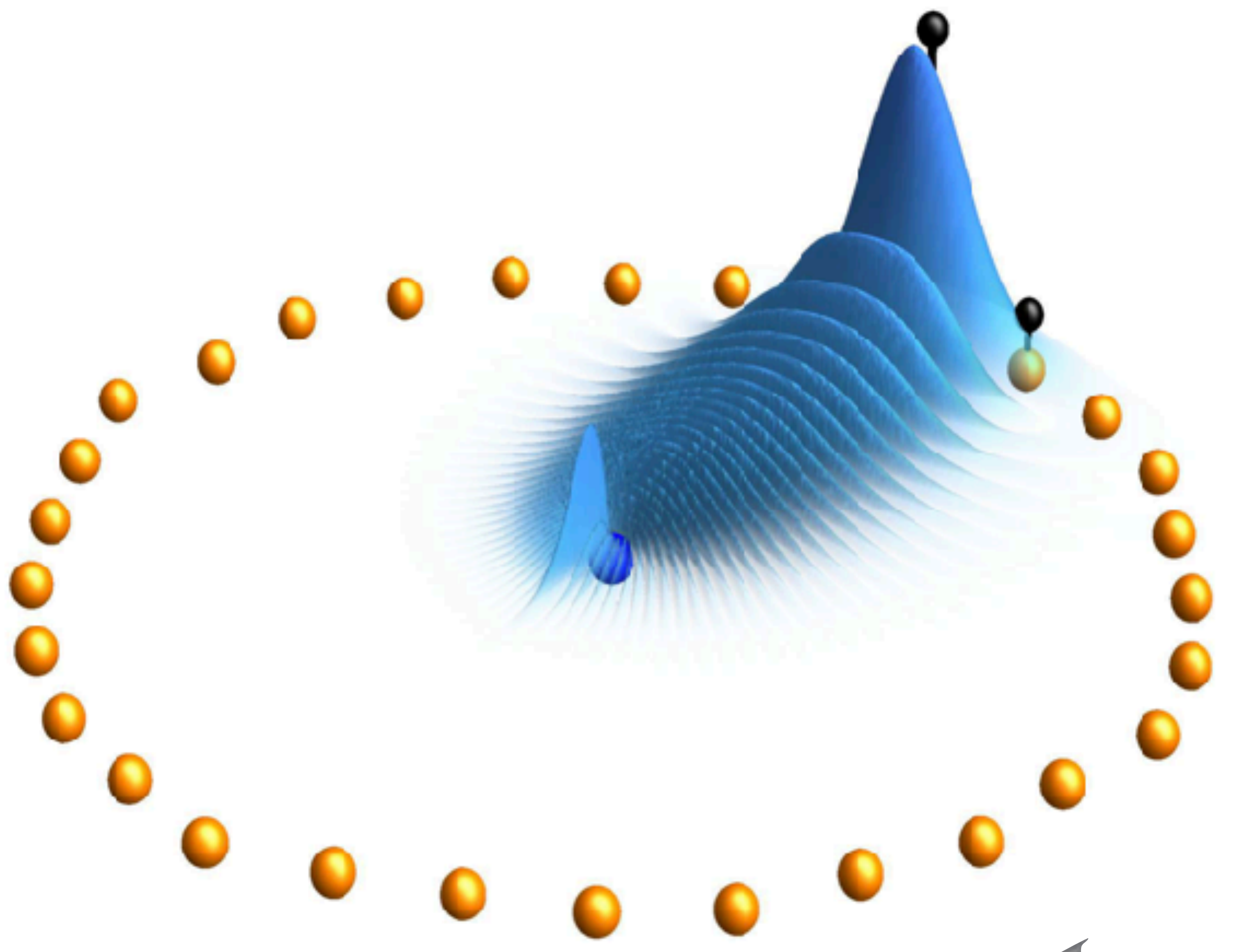
No disorder

Mixed



Disorder; band middle

Localized

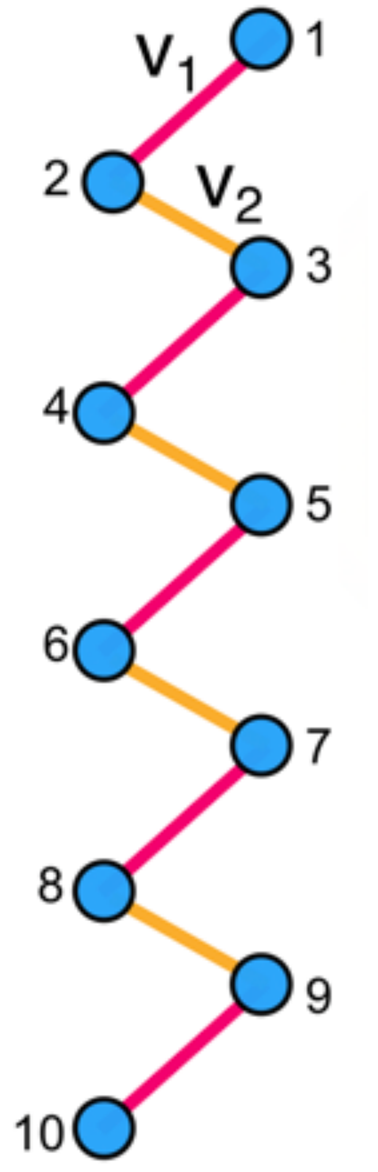


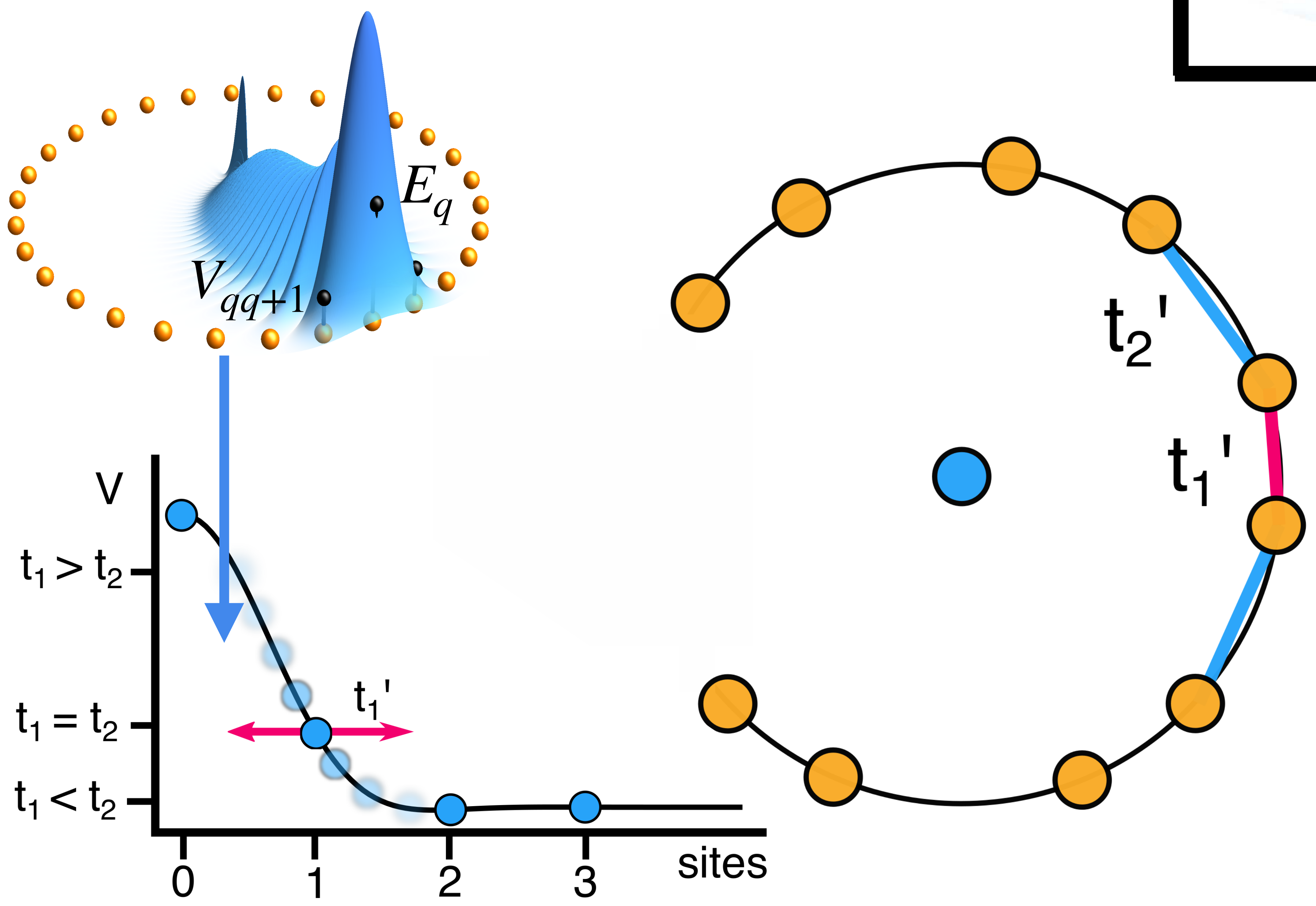
Disorder; band edge

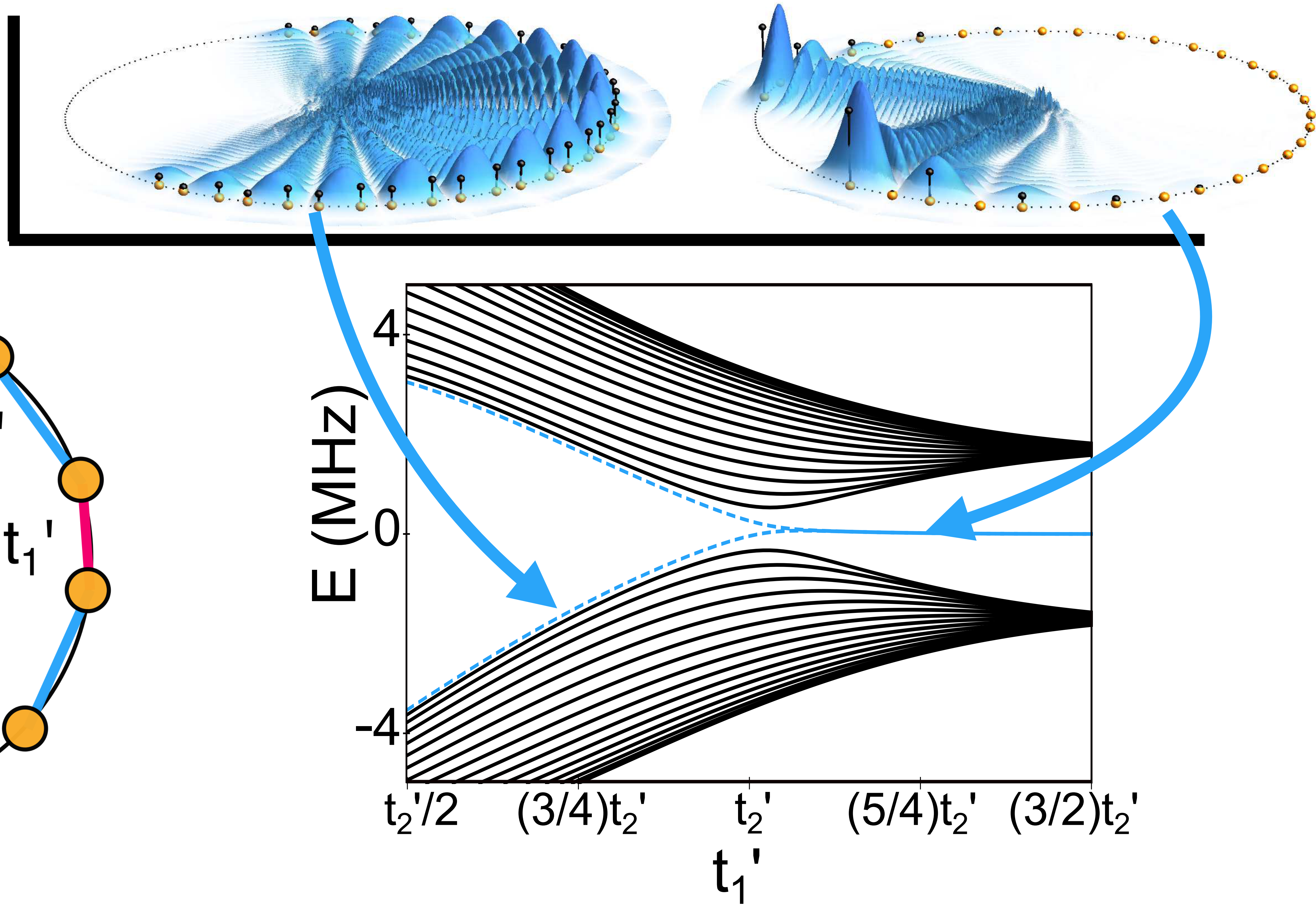
MTE, A. Eisfeld, J. M. Rost PRR **5**, 033032 (2023)

Rydberg molecules, polarons, and composites

When you have disorder, why not seek out topological protection?







Su-Schriefer-Heeger:
- Staggered hopping
- Chiral symmetry
- Polyacetylene model

Ring Rydberg composite:
- Same configuration for NN hopping
- Staggered angles ⟶ staggered hopping

Rydberg spectrum and wave functions:
- Bulk-boundary correspondence
- Topologically protected edge states

MTE, C. W. Wächtler, A. Eisfeld, J. M. Rost arXiv:2309.03039

MAX PLANCK INSTITUTE
FOR THE PHYSICS OF COMPLEX SYSTEMS

Scope of today's lecture

At the core of quantum simulation with Rydberg atoms: 150 years of spectroscopy

- From Rydberg to Pauli/Schrödinger to present day

As billed, it is a "lecture":

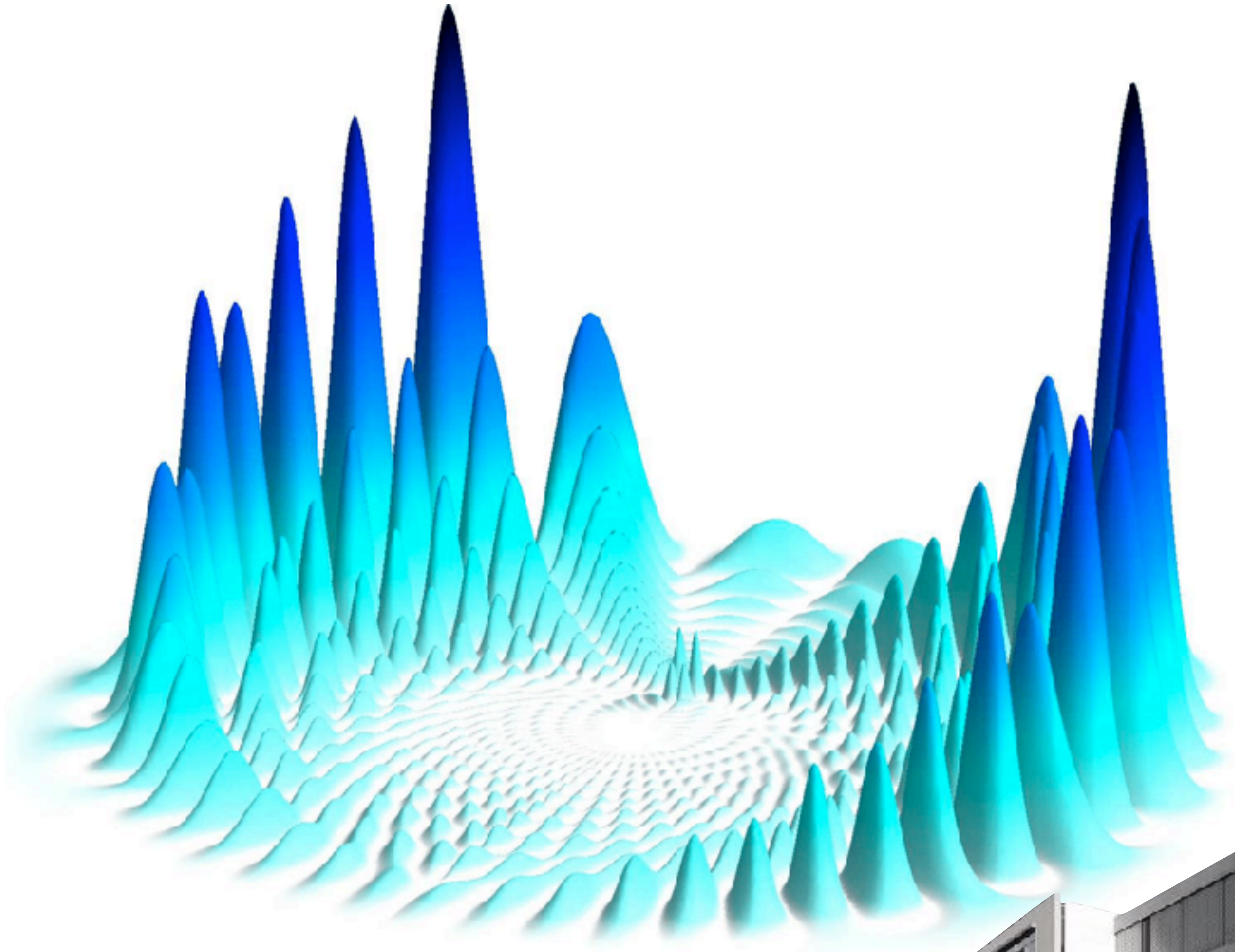
- …expect some equations…but hopefully not too many
- slides: https://www.pks.mpg.de/correlations-and-transport-in-rydberg-matter

What are Rydberg atoms?

- Quantum defect theory: alkali atoms
- Key properties of Rydberg atoms
- Multichannel quantum defect theory: many-electron atoms

What are they good for?

- Rydberg-Rydberg interactions
  - van der Waals / Rydberg blockade
  - dipole-dipole / "flip-flop" interactions
- Rydberg-ground-state-atom interactions



*we didn't actually discuss this. it's just to get your attention

For more details: feel free to shoot me an email at meiles@pks.mpg.de