Non-perturbative calculation of Rydberg interaction

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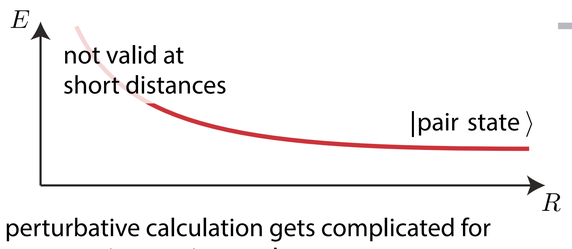
Motivation

Recent Rydberg experiments show the requirement of a deeper theoretical understanding of the full Rydberg interaction physics at short atomic distances. Prominent examples are antiblockade effects due to level crossings or high bandwidth Rydberg excitations as in thermal vapor cells. Here we present the non-perturbative calculation of interaction potentials by numerical diagonalization of the Hamiltonian up to arbitrary order in the multipole expansion.

Perturbatively calculated pair potential

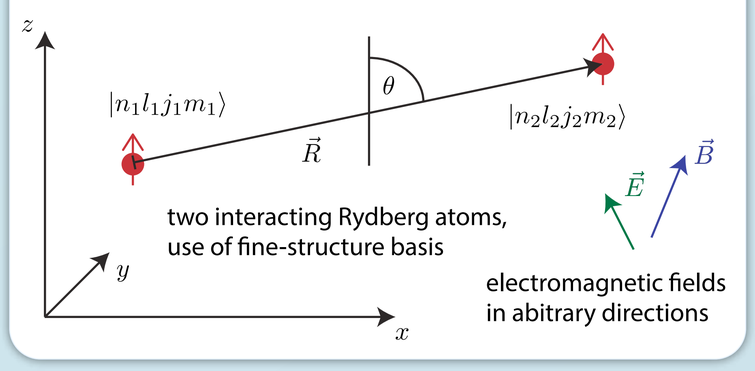


perturbative calculation gets complicated for non-zero interaction angles or near resonances

Non-perturbative calculation of pair potentials by numerical diagonalization of the interaction Hamiltonian

pairinteraction.github.io
Get the open source software.

Setup



two interacting Rydberg atoms, use of fine-structure basis

electromagnetic fields in abitrary directions

Electronic Hamiltonian in Born–Oppenheimer approximation

$H(\vec{R}) = H_0 + H_{\text{int}}(\vec{R}) + H_{\text{fields}}$

For calculation of matrix elements: write operators as product of radial and an angular operators and express angular operators in the spherical basis $\{\vec{e}_\pm = \mp(\vec{e}_x \mp i\vec{e}_y)/\sqrt{2}, \vec{e}_0 = \vec{e}_z\}$.

Energies of the unperturbed levels

$$H_0 = \sum_{n_1,l_1,j_1,m_1} E_{n_1 l_1 j_1} |n_1 l_1 j_1 m_1\rangle\langle n_1 l_1 j_1 m_1| \otimes \mathbb{1} + \mathbb{1} \otimes \sum_{n_2,l_2,j_2,m_2} E_{n_2 l_2 j_2} |n_2 l_2 j_2 m_2\rangle\langle n_2 l_2 j_2 m_2| \quad \text{with energy} \quad E_{nlj} = -\frac{R^*}{(n-\delta_{nlj})^2}$$

Multipole interaction between atoms ($|\vec{R}|$ > Le Roy radius)

$$H_{\text{int}}(\vec{R}) = \sum_{\kappa_1,\kappa_2=0}^{\infty} \frac{V_{\kappa_1,\kappa_2}}{4\pi\epsilon_0 |\vec{R}|^{\kappa_1+\kappa_2+1}} \quad \text{with} \quad V_{\kappa_1,\kappa_2} = (-1)^{\kappa_2} \sum_{q=-\kappa_<}^{\kappa_<} \sqrt{\binom{\kappa_1+\kappa_2}{\kappa_1+q}\binom{\kappa_1+\kappa_2}{\kappa_2+q}}\, \hat{p}^{\text{atom }1}_{\kappa_1 q} \hat{p}^{\text{atom }2}_{\kappa_2 -q}$$

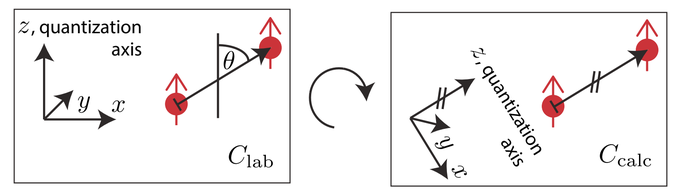
(in rotated frame C_{calc}), $\hat{p}_{\kappa q} = e\,\hat{r}^\kappa \cdot \sqrt{\frac{4\pi}{2\kappa+1}} \hat{Y}_{\kappa q}$ spherical harmonic

Interaction with (static and homogeneous) fields

$$H_{\text{fields}} = -\hat{\vec{d}}\cdot\vec{E} - \hat{\vec{\mu}}\cdot\vec{B} + \frac{1}{8m_e}|\hat{\vec{d}}\times\vec{B}|^2 \quad \text{with} \quad \hat{\vec{d}} = e\hat{\vec{r}} \quad \text{and} \quad \hat{\vec{\mu}} = -\frac{\mu_B}{\hbar}(g_l\hat{\vec{l}} + g_s\hat{\vec{s}})$$

, in the spherical basis $\hat{\vec{J}}_\pm = \mp\frac{1}{\sqrt{2}}(\hat{\vec{J}}_x \pm i\hat{\vec{J}}_y)$, $\hat{\vec{J}}_0 = \hat{\vec{J}}_z$, with $\hat{\vec{J}} \in \{\hat{\vec{d}}, \hat{\vec{l}}, \hat{\vec{s}}\}$

Rotation of coordinate system



$|n,l,j,m\rangle_{\text{lab}} = \sum_{m'} d^j_{mm'}(\theta)\,|n,l,j,m'\rangle_{\text{calc}}$, where $d^j_{mm'}$ is the Wigner d-matrix

$$\vec{E}_{\text{calc}} = \begin{pmatrix} \cos\theta & 0 & -\sin\theta \\ 0 & 1 & 0 \\ \sin\theta & 0 & \cos\theta \end{pmatrix} \vec{E}_{\text{lab}}$$

Non-perturbative calculation of pair potentials

We would like to determine non-perturbatively on which pair potential curves can we find the state $|\Phi^0\rangle$ and what do these curves look like.

Basis

Can be restricted to states with

- similar *energy* as $|\Phi^0\rangle$.

  Reason: interaction dicreases with 1/energy difference.

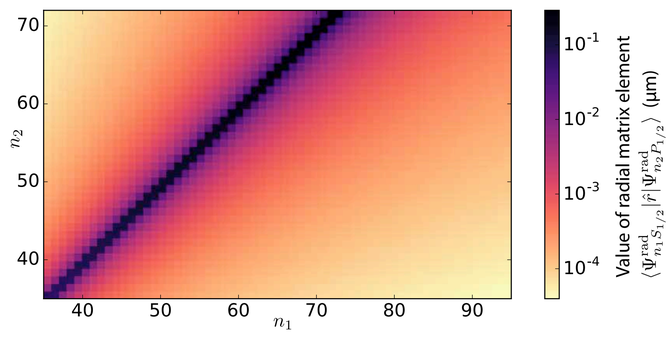
- similar *l, j, m quantum numbers* as $|\Phi^0\rangle$.

  Reason: interaction of lowest order can change these quantum numbers at most by one, for large changes interaction of high order is needed that is weak.

- similar *n quantum number* as $|\Phi^0\rangle$.

  Reason: radial matrix elements are small for large differences in n.

Strong restriction of basis important as matrix diagonalization scales like $O(n^3)$

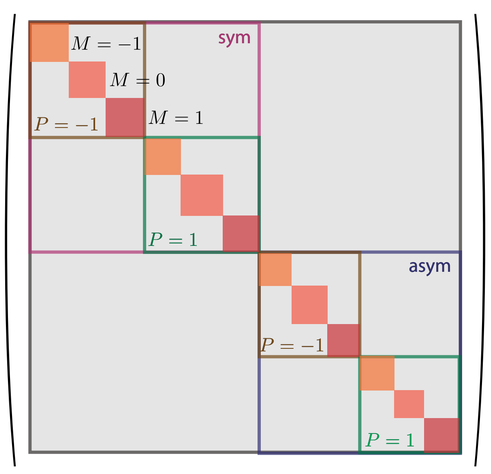


Note: Treat atom-field interaction in a large one-atom basis and atom-atom interaction in a strongly restricted pair basis.

To find the optimal basis size we have to increase the number of basis states until the pair potentials have converged.

Hamiltonian in matrix form

Construct it for distances at whitch we would like to know the pair potential.



Operators conserve some properties of the states.

Example: pure dipole-dipole interaction conserves

- $M = m_1 + m_2$
- parity P of $l_1 + l_2$
- symmetry of pair states

Hamiltonian matrix is block diagonal, only diagonalization of the block that contains $|\Phi^0\rangle$ is needed.

Eigenenergies make up pair potentials

Diagonalize the Hamiltonians and plot eigenenergies over distances.

Extraction of potential curves

Draw lines between the eigenenergy-points for which the overlap of the eigenvectors is maximal.

Formalisms for calculating matrix elements

Radial matrix elements $\langle\Psi^{\text{rad}}_{nlj}|\hat{r}^\kappa|\Psi^{\text{rad}}_{n'l'j'}\rangle$

Solve radial Schrödinger equation

- numerically for model potential considering core polarizability and SO coupling [1] *or*
- analytically via Whittaker functions [2].

Determine radial matrix elements by numerical integration.

[1] Marinescu et al., Phys. Rev. A **49** 982, 1994
[2] Seaton, Rep. Prog. Phys. **47** 167, 1983

Angular matrix elements of spherical operators $\langle Y_{lsjm}|\hat{T}_{\kappa q}|Y_{l's'j'm'}\rangle$

Angular wave functions are spin spherical harmonics. Calculate angular matrix elements with Wigner-Eckart theorem [3].

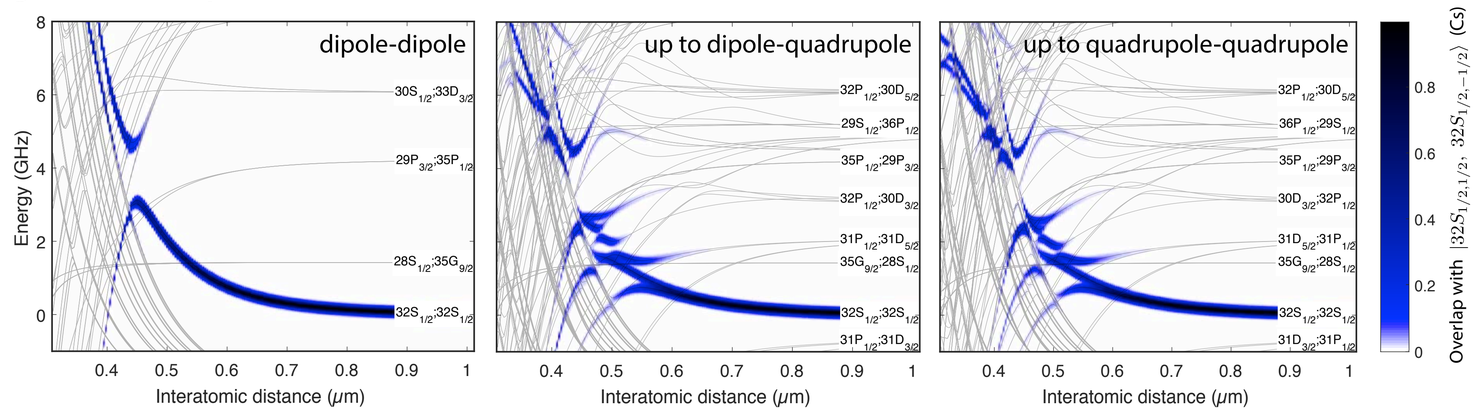
[3] Sobelman, *Atomic Spectra and Radiative Transitions*, Springer, 1979

Selection rules

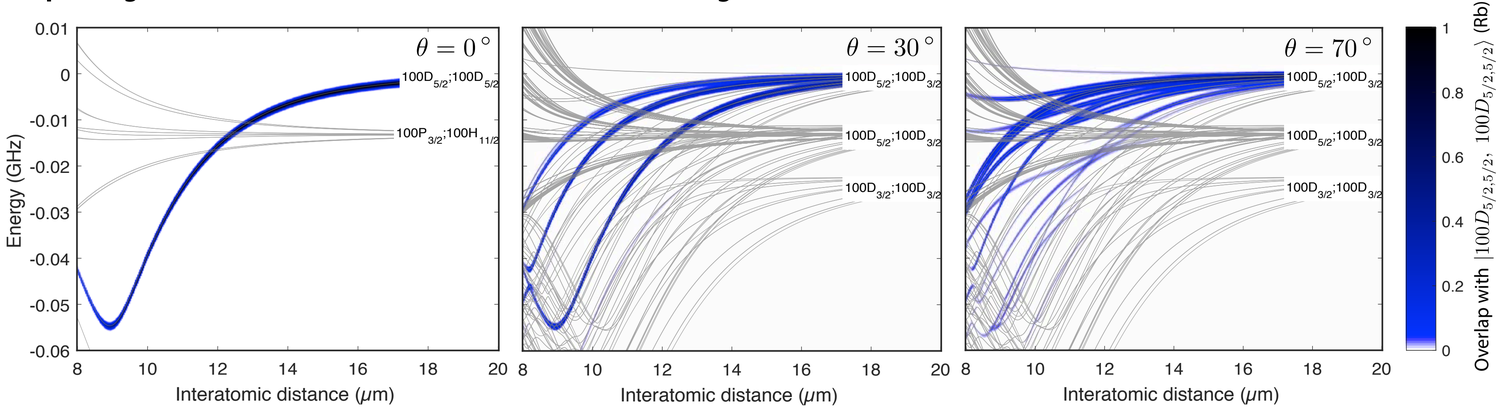
Spherical harmonics $\hat{Y}_{\kappa q}$ and multipole operator $\hat{p}_{\kappa q}$	Momentum operator $\hat{J}_{1q}$
n not restricted	n not restricted
$l = \begin{cases} l' \pm 0, 2, ..., \kappa & \text{even } \kappa \\ l' \pm 1, 3, ..., \kappa & \text{odd } \kappa \end{cases}$	$l = l'$
$s = s'$	$s = s'$
$j = j' \pm 0, 1, ..., \kappa,\ j + j' \geq \kappa$	$j = j' \pm 0, 1$
$m = m' + q$	$m = m' + q$

Applications

Higher-order multipole interaction



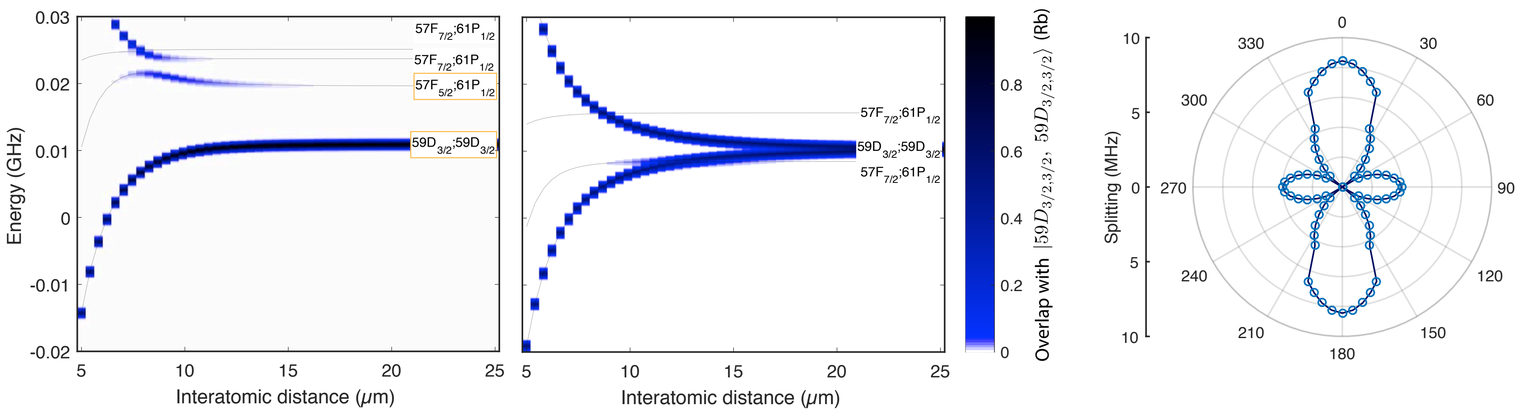
Dephasing into Zeeman sublevels for non-zero interaction angles



Förster resonance tuned by electric and magnetic fields

$B = 3.3\,\text{G}$, no electric field

$B = 3.3\,\text{G}$, electric field adjusted to $E = 34\,\text{mV}$



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