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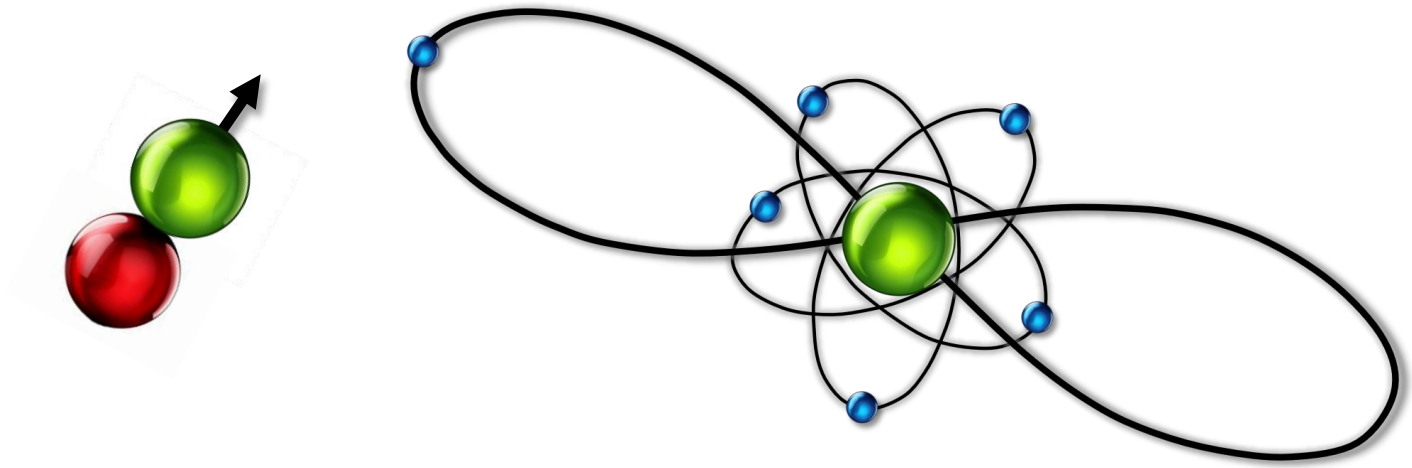
ULTRALONG-RANGE Cs-RbCs RYDBERG MOLECULE: NON-ADIABATICITY OF DIPOLE MOMENTS

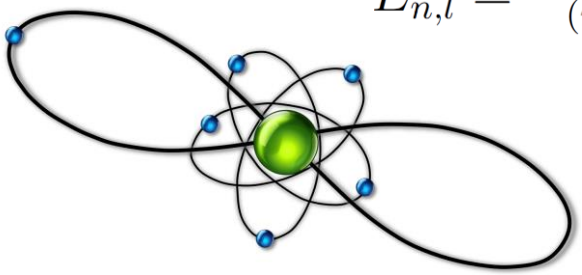


DAVID MELLADO-ALCEDO

September 8th, 2023

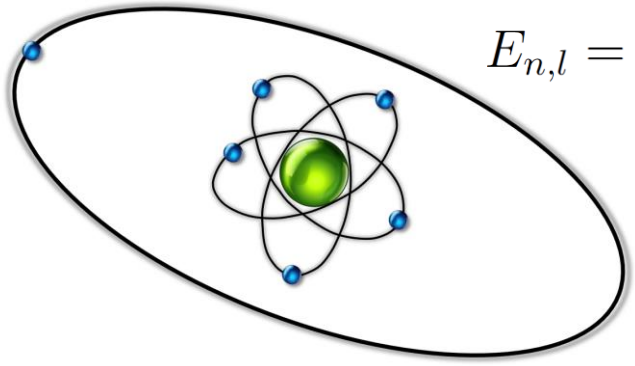
RYDBERG ATOM

$$E_{n,l} = -\frac{Ry}{(n-\delta_{nl})^2}$$



Low angular momentum Rydberg states $l \leq 3$

$$E_{n,l} = -\frac{Ry}{n^2}$$



High angular momentum Rydberg states $l \geq 4$

Rydberg-Ritz expression

$$\delta_{nl} = \delta_0 + \frac{\delta_2}{(n-\delta_0)^2} + \frac{\delta_4}{(n-\delta_0)^4} + \cdots$$

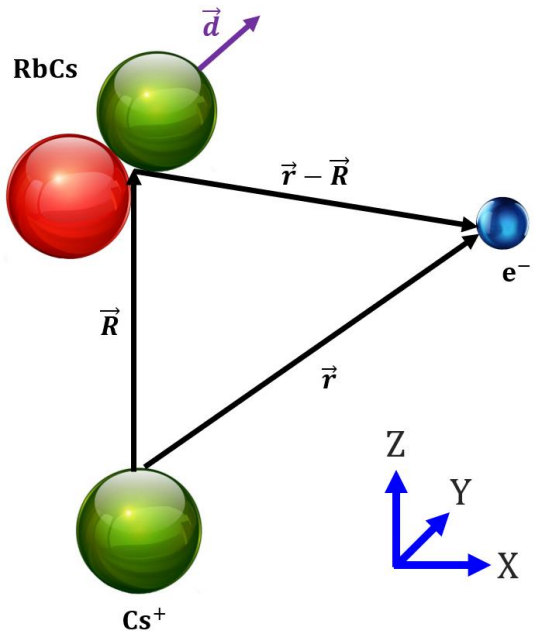
- **Strong permanent electric dipole moment**
- **Sensitivity to small external fields**
- **Applications: ultracold chemistry or quantum information processing**

Binding energy	n^{-2}
Orbital radius	n^2
Dipole moment	n^2
Polarizability	n^7
Radiative lifetime	n^3

THE ADIABATIC HAMILTONIAN

- Subcritical dipole moment: $d < 1.639D$ (Fermi-Teller)

$$H_{ad} = H_A + H_{rot} + H_{ryd} + H_{sw}$$



$$H_A = -\frac{\hbar^2}{2m_e}\nabla_r^2 + V_l(r)$$

$$H_{rot} = B\boldsymbol{N}^2$$

$$H_{ryd} = -\boldsymbol{d}\cdot\left(e\frac{\boldsymbol{R}}{R^3} + e\frac{\boldsymbol{r}-\boldsymbol{R}}{|\boldsymbol{r}-\boldsymbol{R}|^3}\right)$$ Charge-dipole interaction

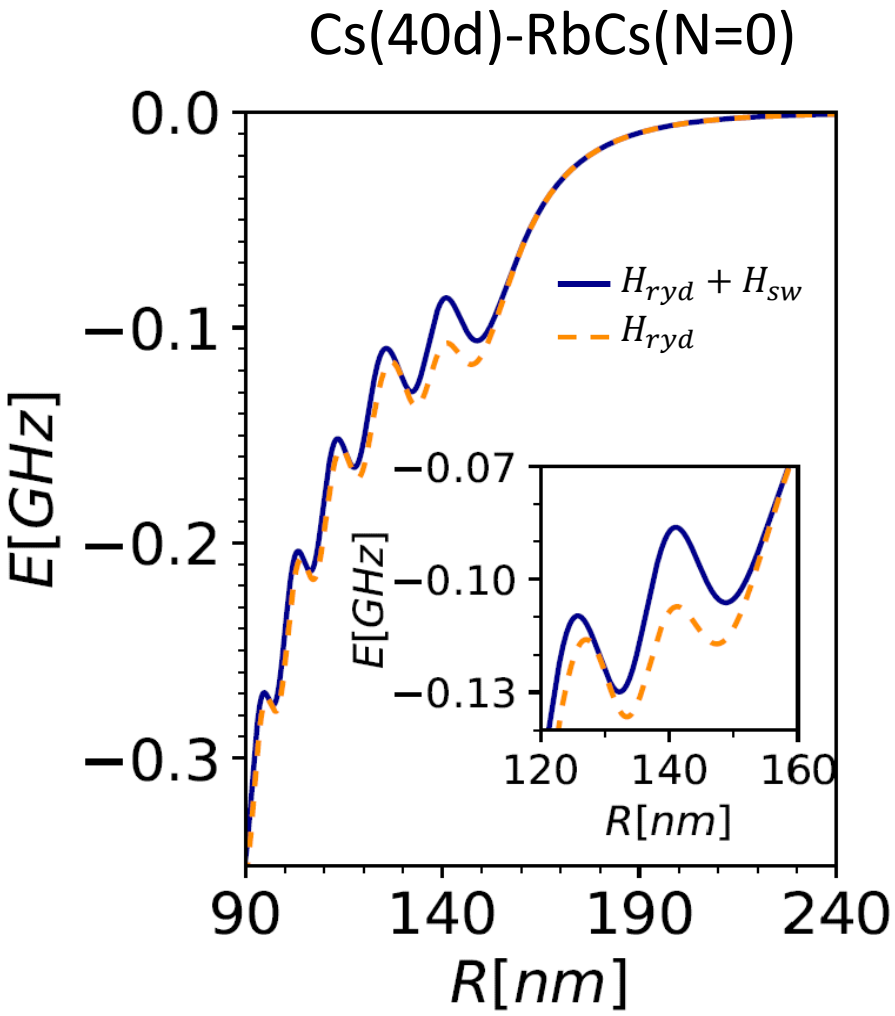
$$H_{sw} = 2\pi a_S(k)\delta(\boldsymbol{r}-\boldsymbol{R})$$ s-wave fermi pseudopotential

$$\boldsymbol{J} = \boldsymbol{N} + \boldsymbol{l} \qquad [J_Z, H] = 0 \rightarrow$$

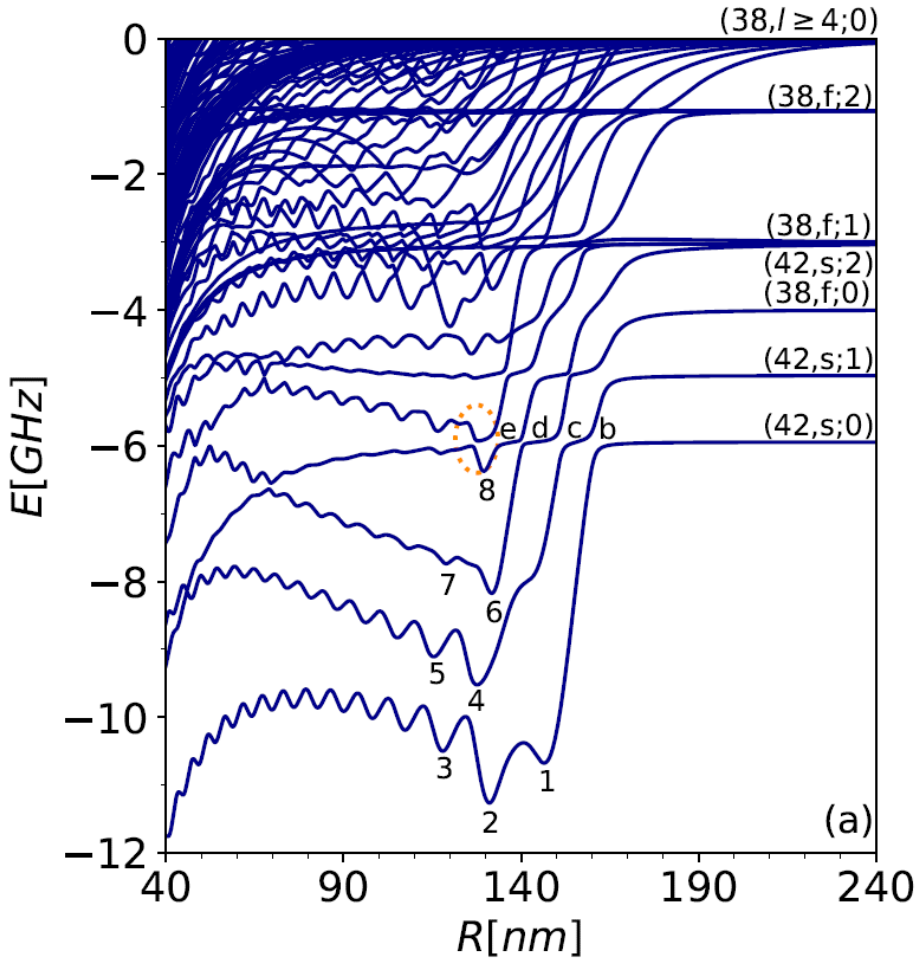
$$\Psi(\mathbf{r},\Omega;R) = \sum_{n,l,N,J} C^J_{n,l,N}(R)\Psi^{JM_J}_{nl,N}(\mathbf{r},\Omega)$$

$$\Psi^{JM_J}_{nl,N}(\mathbf{r},\Omega) = \sum_{m_l=-l}^{m_l=l}\sum_{M_N=-N}^{M_N=N}\langle lm_lNM_N|JM_J\rangle\,\Psi_{nlm_l}(\mathbf{r})Y_{NM_N}(\Omega)$$

ZERO-RANGE ELECTRON-MOLECULE SCATTERING

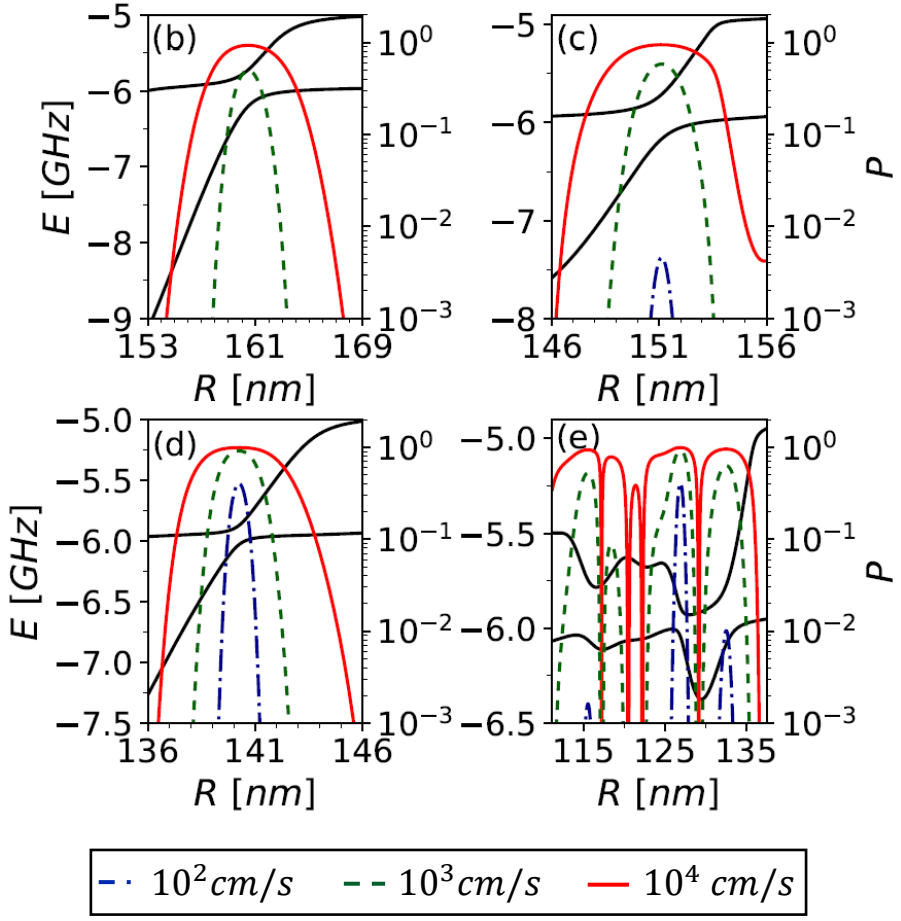


ELECTRONIC STRUCTURE OF Cs(38)-RbCs



$$P = \exp\left(-\frac{\pi \Delta E}{4\hbar g v}\right)$$

Landau-Zener



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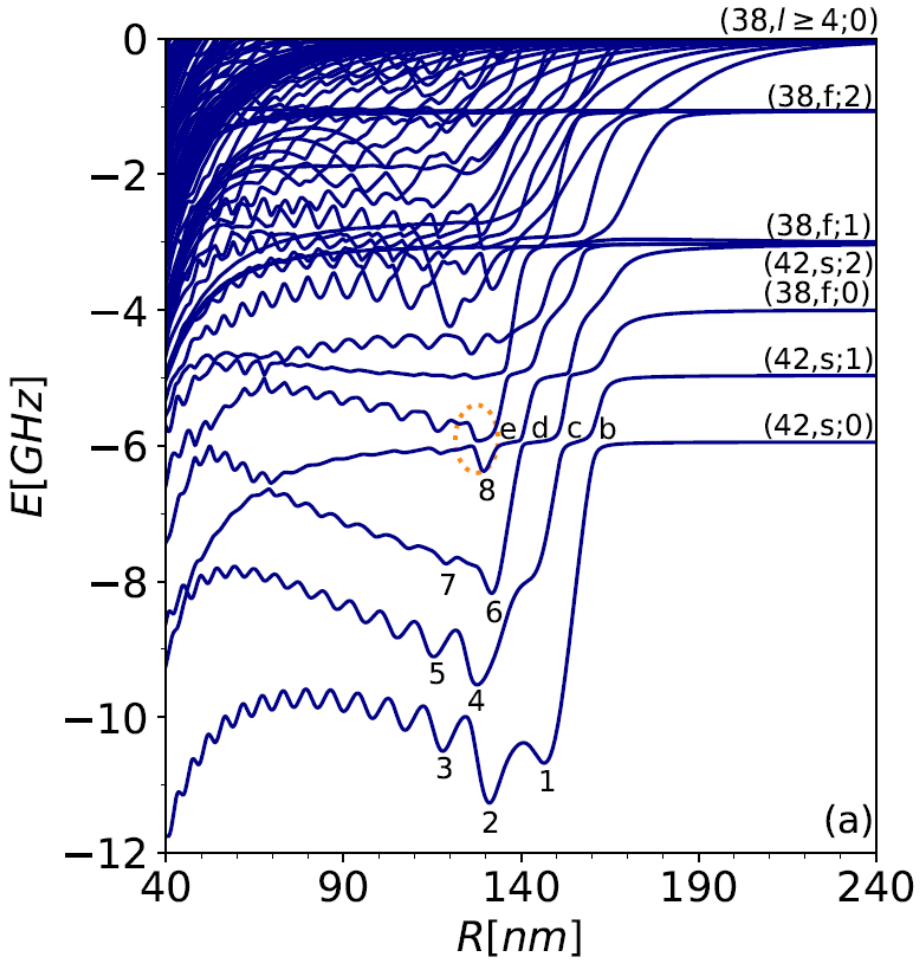
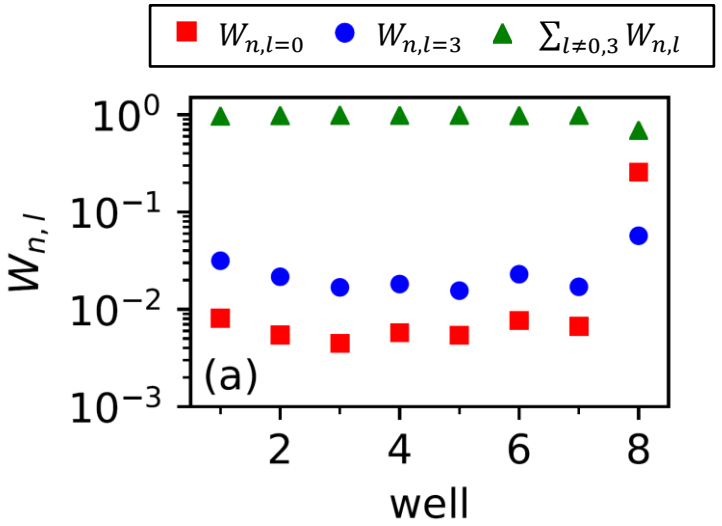
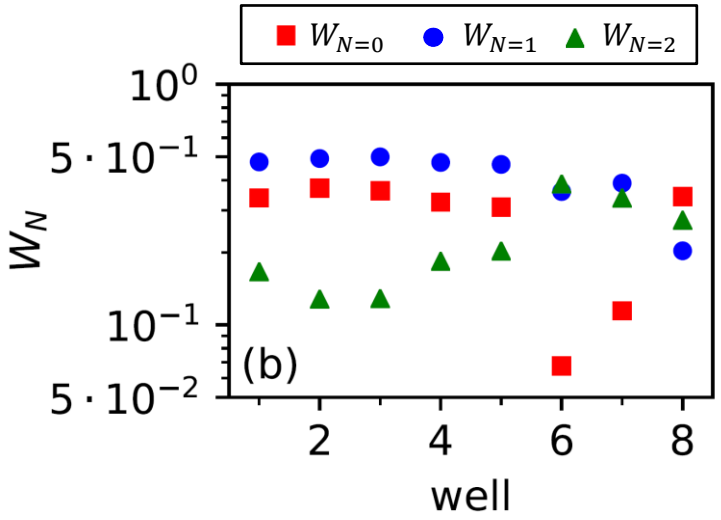
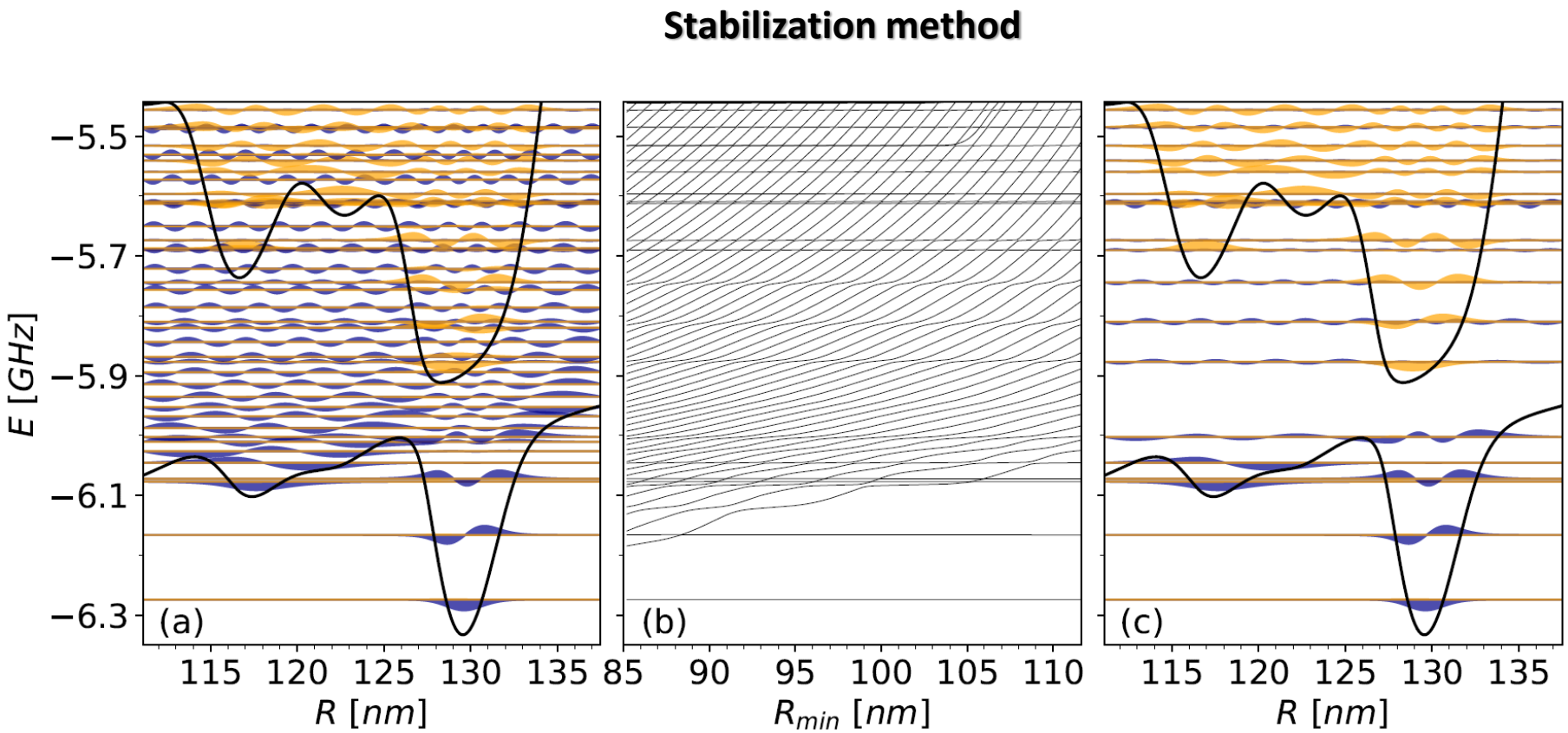


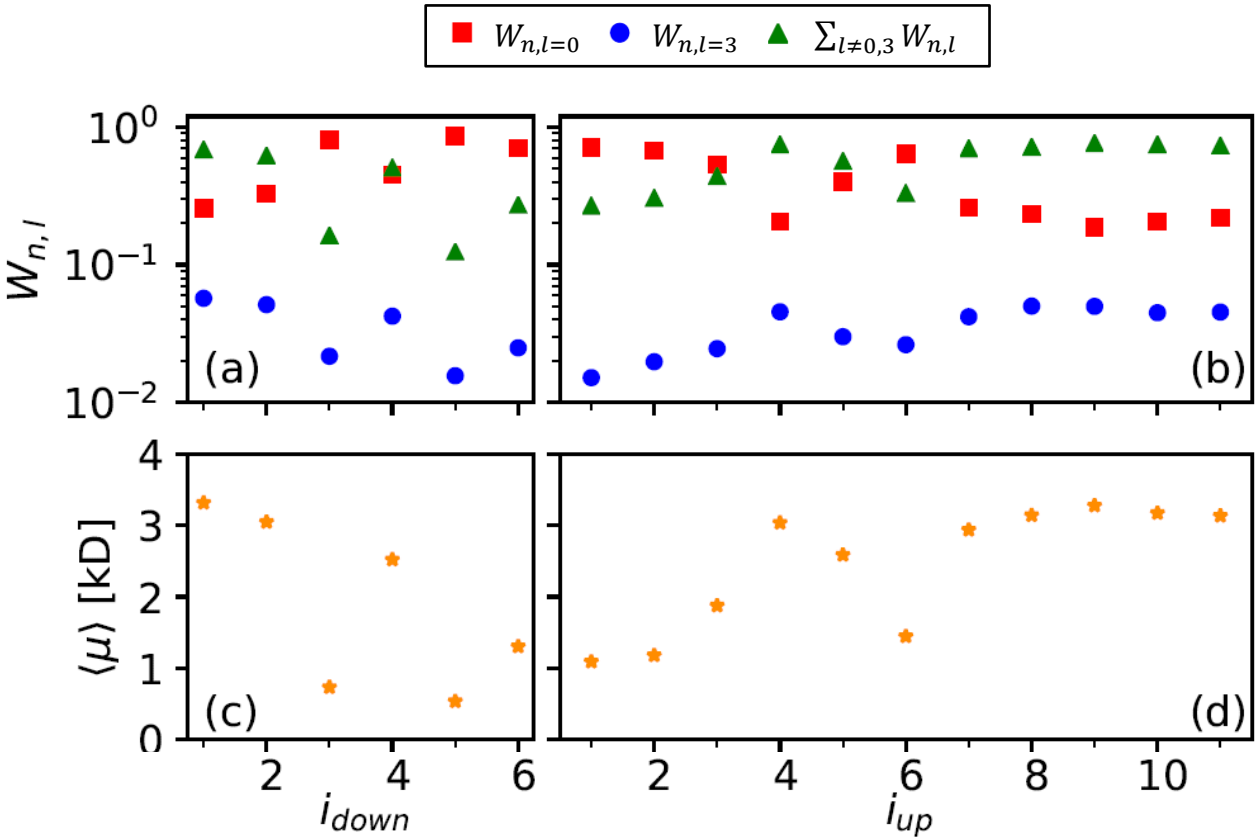


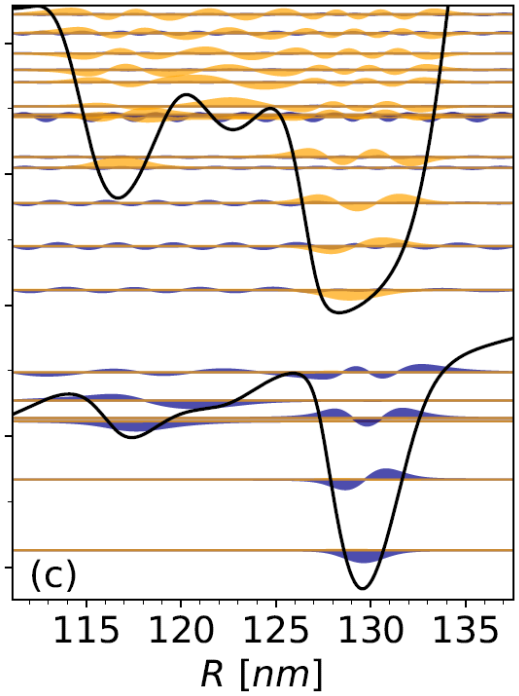


VIBRATIONAL STATES OF Cs(42s)-RbCs(N=0)

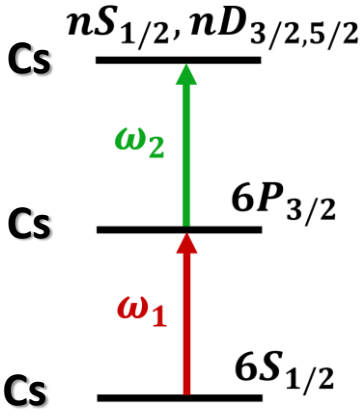


VIBRATIONAL STATES OF Cs(42s)-RbCs(N=0)





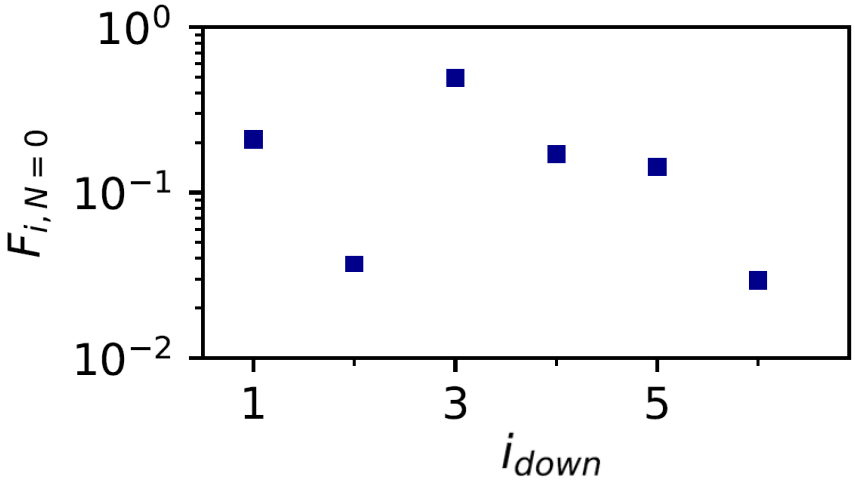
VIBRATIONAL STATES OF Cs(42s)-RbCs(N=0)

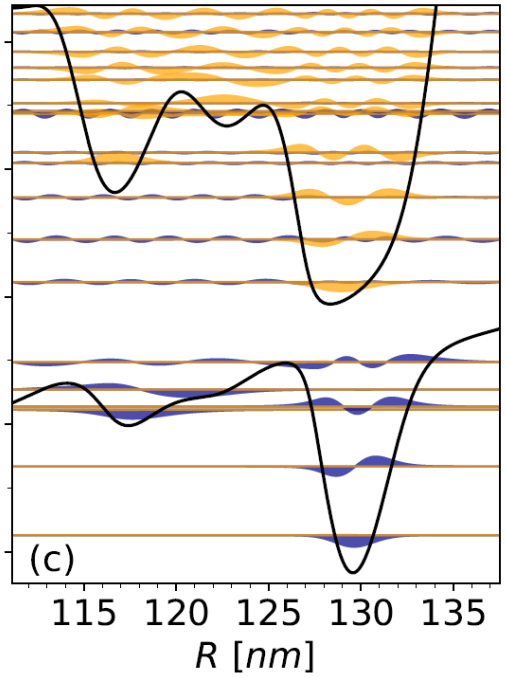


two-photon scheme

Franck-Condon factors

$$F_{i,N} = \sum_{k=d,u} \int (\chi_i^k(R))^* \Omega_{ns} C^{J,k}_{n,l=0,N}(R) \psi_{scat}(R) R dR$$





CONCLUSIONS AND OUTLOOK

- **Analysis of the electronic structure -> H_{sw} cannot be neglected**
- **Including non-adiabatic couplings in the neighboring adiabatic potential curves**
- **Identification of vibrational states that could be created by photoassociation**
- **Future works-> including hyperfine, and electric and magnetic field interactions**


Thank you!
In collaboration with:
Hossein R. Sadeghpour (ITAMP)
Rosario González-Férez (U. Granada)
Simon Cornish (Durham University)
Alexander Guttridge (Durham University)

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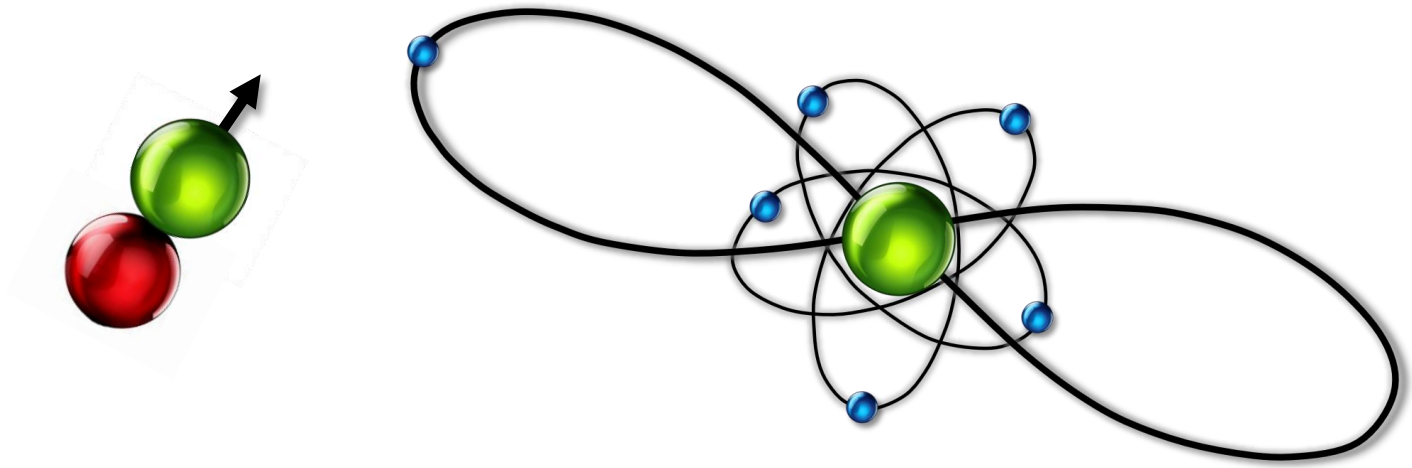

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SUPPLEMENTARY MATERIAL

- **Landau-Zener**:

$$P = \exp\left(-\frac{\pi \Delta E}{4\hbar g v}\right) \qquad g = \left|\langle \Psi_i | \frac{\partial}{\partial R} | \Psi_j \rangle\right|$$

- **Decoupling:**

$$\Big[-\frac{\hbar^2}{2m_R}\frac{d^2}{dR^2} + E(R)\Big]\chi(R) = \mathcal{E}\chi(R)$$

- **Coupling:**

$$\begin{pmatrix} T + E_d + \mathcal{A}_{dd} & \mathcal{A}_{ud} \\ \mathcal{A}_{du} & T + E_u + \mathcal{A}_{uu} \end{pmatrix} \begin{pmatrix} \chi^d \\ \chi^u \end{pmatrix} = \mathcal{E} \begin{pmatrix} \chi^d \\ \chi^u \end{pmatrix}$$

Reduced kinetic energy: $T = -\frac{\hbar^2}{2m_R}\frac{d^2}{dR^2}$

Electronic adiabatic potential energy: $E_i = \langle \phi_i | H_{ad} | \phi_i \rangle$

Coupling terms: $\mathcal{A}_{ij} = \langle \Psi_i | T | \Psi_j \rangle - \frac{\hbar^2}{m_R}\langle \Psi_i | \frac{\partial}{\partial R} | \Psi_j \rangle \frac{d}{dR} \quad \left(\langle \Psi_i | \frac{\partial}{\partial R} | \Psi_i \rangle = 0\right)$

SUPPLEMENTARY MATERIAL

- **Weights of the Rydberg atom partial waves**:

$$W_{n,l} = \sum_{k=d,u} \int \chi^{k*}(R)\mathcal{C}_{n,l}(R)\chi^{k}(R)dR \qquad \mathcal{C}_{n,l}(R) = \sum_{N,J} |C^{J}_{n,l,N}(R)|^2$$

- **Weights of the polar molecule partial waves:**

$$W_{N} = \sum_{k=d,u} \int \chi^{k*}(R)\mathcal{C}_{N}(R)\chi^{k}(R)dR \qquad \mathcal{C}_{N}(R) = \sum_{n,l,J} |C^{J}_{n,l,N}(R)|^2$$

- **Electric dipole moments:**

$$\langle \mu \rangle = \sum_{k_1,k_2=d,u} \int (\chi^{k_1}(R))^* D^{k_1,k_2}_{ryd}(R)\chi^{k_2}(R)dR$$

$$D^{k_1,k_2}_{ryd}(R) = \int \Psi^*_{k_1}(\mathbf{r},\Omega;R) r\cos\theta_e \Psi_{k_2}(\mathbf{r},\Omega;R) d^3r d\Omega$$