Dressed Ion-Pair States of an Ultralong-Range Rydberg Molecule

P. Giannakeas*,[1], M. T. Eiles[1], F. Robicheaux[2], and Jan-Michael Rost[1]

[1]Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Strasse 38, 01187 Dresden, Germany
[2]Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47907, USA

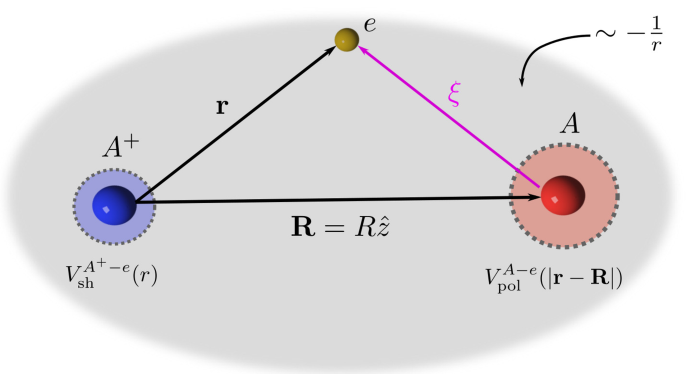
System & Motivation

The main premise of this project is to explore exotic states of Ultralong-range Rydberg Molecules (ULRMs) which share characteristics with Heavy Rydberg States (HRS) [1].

System: Two ^{85}Rb atoms: one in a highly excited Rydberg state and a ground state one located at distance R.

Motivation: The impact of high-angular momentum electron-atom interactions

Ultralong-range Rydberg molecules and Hamiltonian



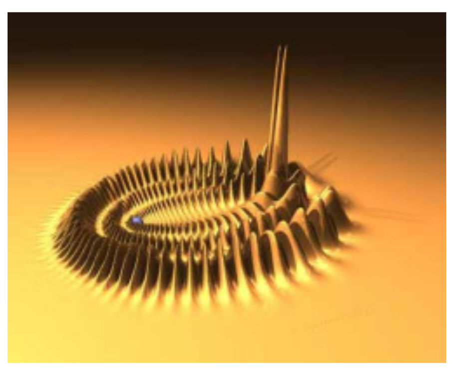
- The Hamiltonian of the electronic degrees-of-freedom reads:

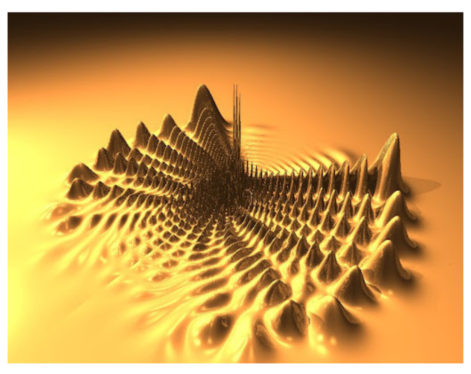
$$H(r) = -\frac{1}{2}\frac{\partial}{\partial r} - \frac{1}{r} + V_{sh}^{A^+-e}(r) + V_{pol}^{A-e}(|\boldsymbol{r}-\boldsymbol{R}|) \quad (1)$$

- The polarization potential V_{pol}^{A-e} between the electron and the ground state atom reads:

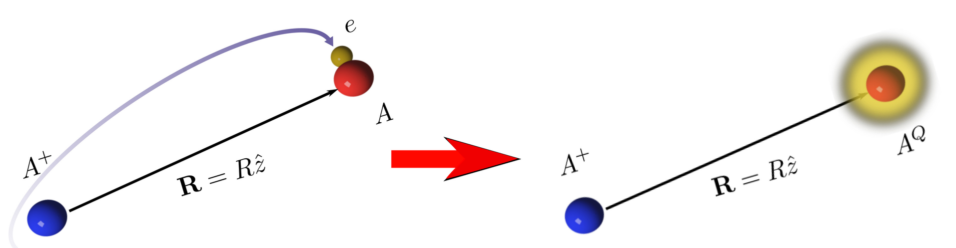
$$V_{pol}^{A-e}(|\boldsymbol{r}-\boldsymbol{R}|) = 2\pi a_S(k)\delta(\boldsymbol{r}-\boldsymbol{R}) + 6\pi a_P^3(k)\overleftarrow{\nabla}\delta(\boldsymbol{r}-\boldsymbol{R})\overrightarrow{\nabla} \quad (2)$$

- $S-$ and $P-$wave electron-atom interactions $\to$ **two types of molecular states:** the **trilobites** and **butterflies**, respectively [2-4].





Dressed ion-pair model



- The **ground state** atom is **dressed by a charge distribution:**

$$\langle Q_L(R)\rangle = -\frac{1}{\nu^3\pi k}\frac{d\delta_L(k)}{dk} \quad (3)$$

- The **positively charged core** interacts with the **dressed anion** via Coulomb forces $\to F_L(R) = \langle Q_L(R)\rangle / R^2$
- The molecular potential curves then read: $E_L(R) = -\frac{1}{2(n-\delta_L(k)/\pi)^2}$

What if the fractional charge Q is independent of R?

- The **fractional charge** Q is constant if $\delta_L \sim k^2$
- The phase shifts in Born approximation for $L > 1$ read:

$$\tan\delta^B_{L>1} = \pi\bar{a}_L k^2 \text{ with } \bar{a}_L = \frac{\alpha}{(2L+1)(2L-1)(2L+3)} \quad (4)$$

- Taylor expanding the high-L potential curves:

$$E_L(R) = \frac{\bar{a}_L}{n^5} - \frac{2\bar{a}_L}{n^3 R} - \frac{6\bar{a}_L}{n^4 R^2} - \ldots \quad (5)$$

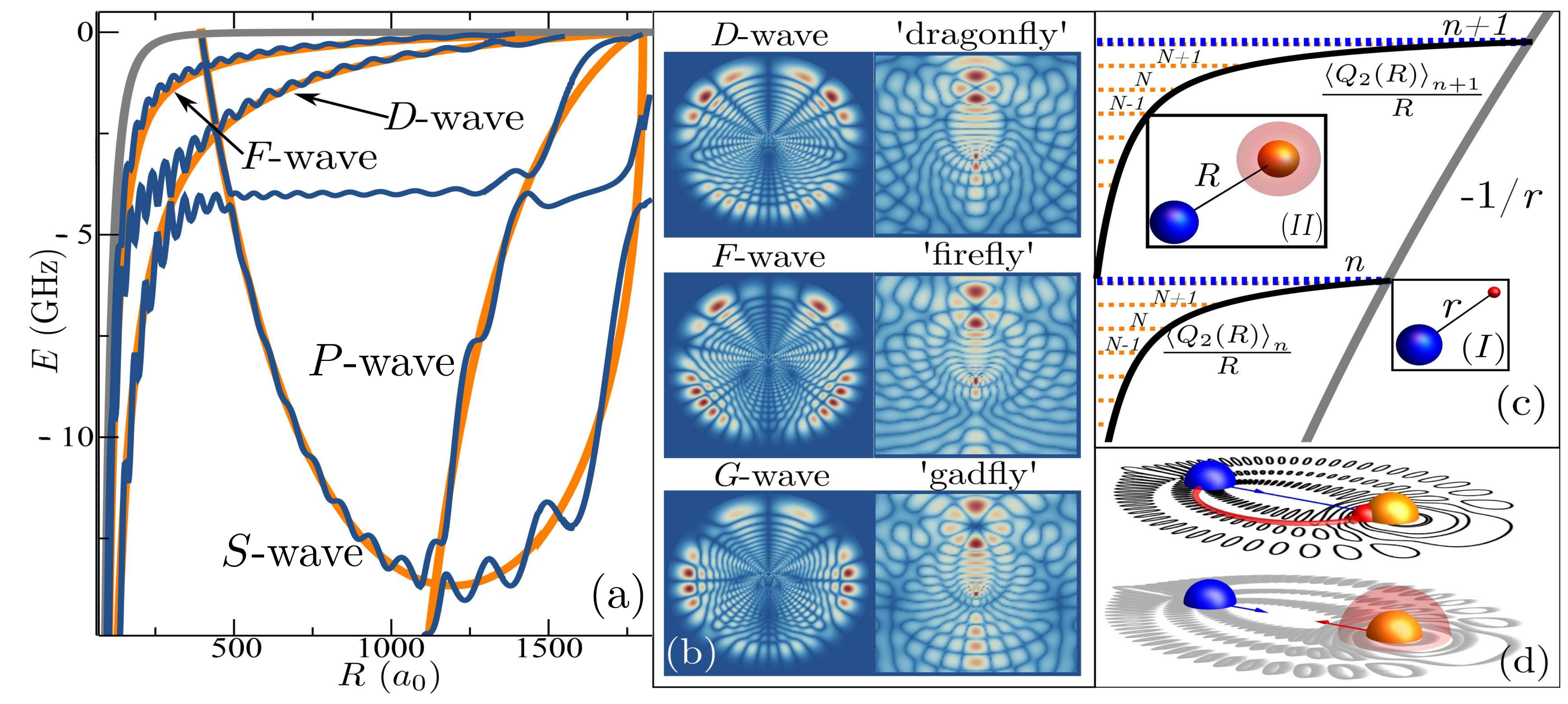
- The **leading term** is a **Coulomb tail** attached to each electronic Rydberg manifold n
- **Vibrational spectrum** $\to$ WKB analysis:

$$E^{nL}_{\nu J} = \frac{\bar{a}_L}{n^5} - \frac{R'_{nL}}{(\nu-\eta_J)^2};\ R'_{nL} = \frac{2\mu\bar{a}_L}{n^6} \quad (6)$$

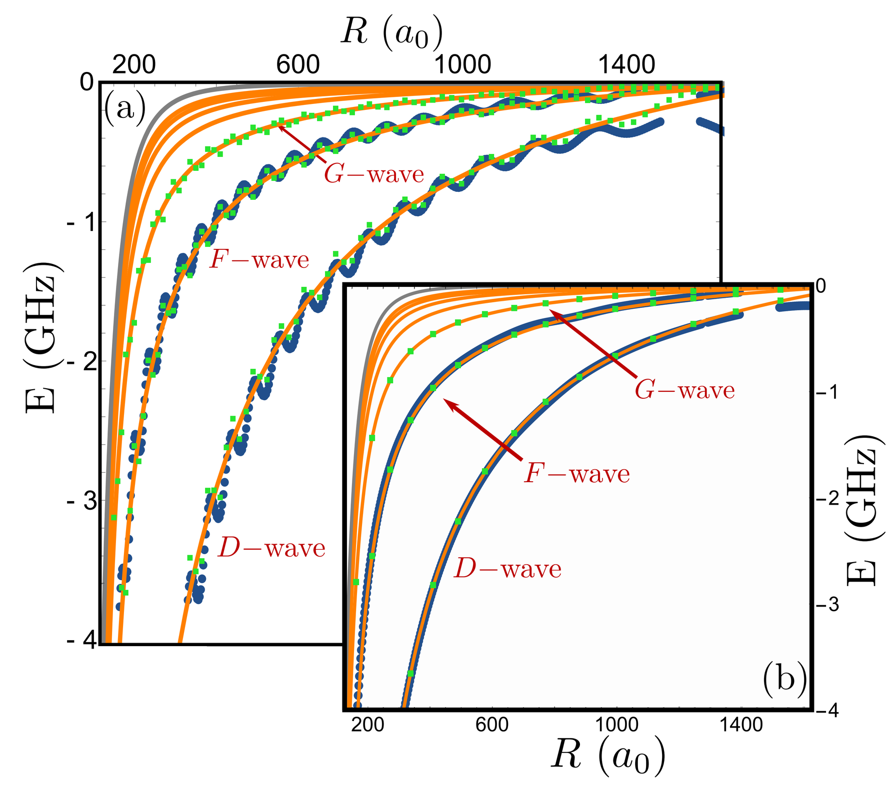
- **The core and the dressed anion** form a **vibrational heavy Rydberg series** with a **small Rydberg constant**

A new class of Rydberg molecules

- **A new theoretical framework**: **Generalized Local Frame Transformation** (GLFT) theory for Rydberg molecules [5]
- For Rb atoms we include $D-$, $F-$ and $G-$wave $e-Rb$ phase shifts
- **A family of high-L ULRMs** $\to \Sigma$ molecular symmetry



- **Neglecting atomic quantum defects**: **Comparison** of numerical (green dots) calculations, dressed-ion pair predictions (orange lines) and GLFT results (blue dots)



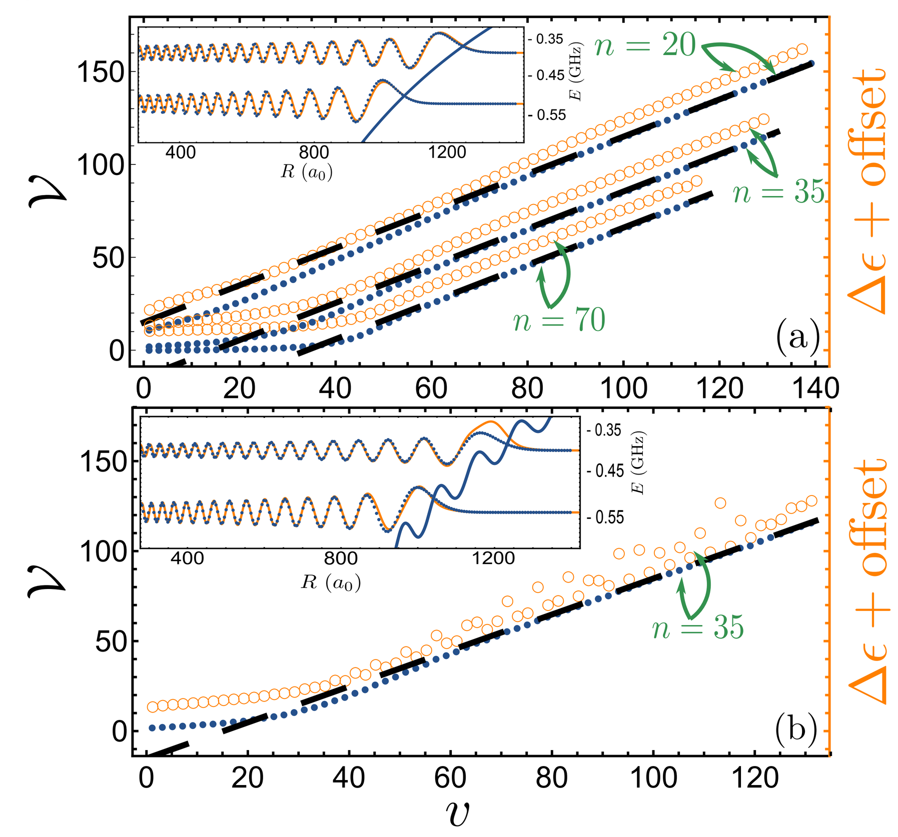
- Panels (a) and (b) show Σ and Δ molecular symmetry

Trimmed Rydberg series in Dragonfly potential curves

- The **effective nuclear quantum number** is $\mathscr{V}$ (blue dots)

$$\mathscr{V} = \sqrt{R_{nL}/(\bar{a}_L/n^5 - \varepsilon^{nL}_{\nu J})} \quad (7)$$

- The rescaled **difference** of **successive energy levels** $\Delta\varepsilon$ (orange circles)



- Panels (a) and (b) show the **vibrational spectra** for the Δ and Σ **dragonfly molecular curves** at different electronic n manifolds
- The **straight lines** of blue, orange and black-dashed lines denote a **Rydberg series** in the **vibrational spectrum**

References

[1] P. Giannakeas, M.T. Eiles, F.Robicheaux, and J.-M. Rost, Phys. Rev. Lett. 125, 123401 (2020).
[2] C. H. Greene, A. S. Dickinson, and H. R. Sadeghpour, Phys. Rev. Lett. 85, 2458 (2000).
[3] E. L. Hamilton, C. H. Greene, and H. R. Sadeghpour, J. Phys. B 35, L199 (2002).
[4] M. I. Chibisov, A. A. Khuskivadze, and I. I. Fabrikant, J. Phys. B 35, L193 (2002).
[5] P. Giannakeas, M. T. Eiles, F. Robicheaux, and J.-M. Rost, Phys. Rev. A 102, 033315 (2020).

* pgiannak@pks.mpg.de