Precision spectroscopy and vibrational stabilization of trilobite Rydberg molecules

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In this talk, I report on a joint experimental-theoretical work in which we perform three-photon photoassociation to obtain high resolution spectra of ^{87}Rb trilobite dimers for the principal quantum numbers $n = 22, 24, 25, 26,$ and 27. The large binding energy of the molecules in combination with a relative spectroscopic resolution of 10^{-4} provides a rigorous benchmark for existing theoretical models. A recently developed Green's function framework, which circumvents the convergence issues that afflicted previous studies, is employed to theoretically reproduce the vibrational spectrum of the molecule with high accuracy. With this, we extract the $^3S_1$ scattering phase shift with unprecedented accuracy, at low energy regimes inaccessible to free electrons [1]. Additionally, for $n = 25$, we observe a regularly spaced series of highly excited ($\nu \sim 100$) vibrational states close to the dissociation threshold. The existence and observed stability of these states requires the almost complete suppression of the adiabatic decay pathway induced by the P-wave shape resonance of Rb. This stabilization is predicted to occur only for certain Rydberg levels where the avoided crossing between trilobite and P-wave dominated butterfly potential energy curves nearly vanishes, allowing the vibrational states to diabatically traverse the crossing with almost unit probability. This is the first direct measurement of beyond-Born-Oppenheimer physics in long-range Rydberg molecules, and paves the way for future experiments to access and manipulate wavepackets formed from high-lying vibrational states [2].

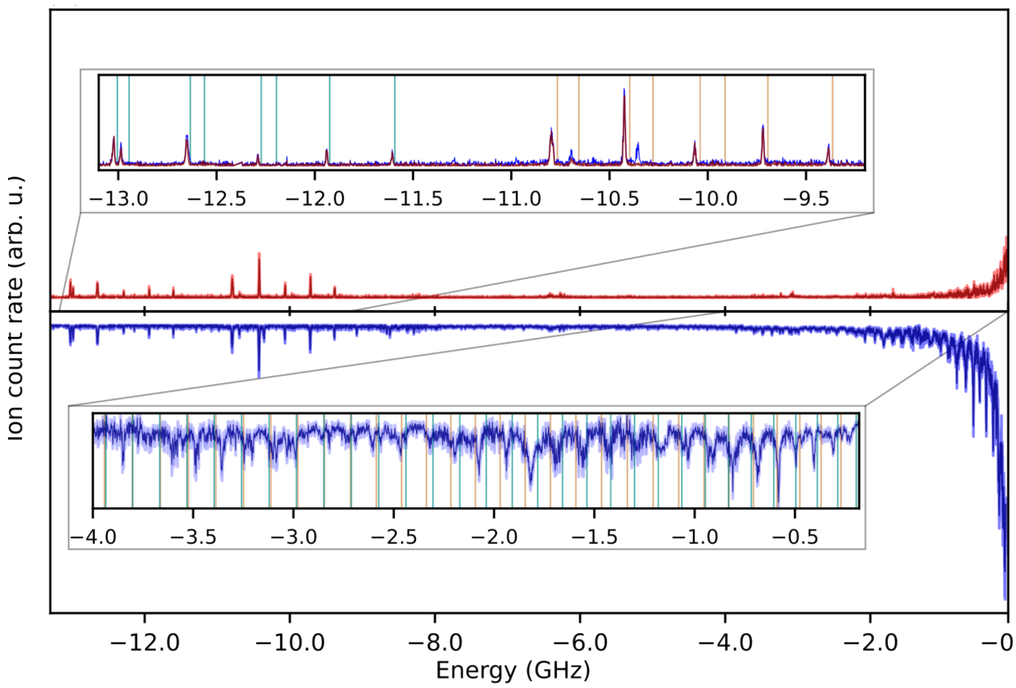

Figure 1: Experimental and theoretical spectrum for an $n = 25$ trilobite molecule, showing both the deeply bound molecular states utilized for precision extraction of scattering phase shifts (upper panel) and the vibrationally excited series stabilized by non-adiabatic coupling (lower panel).

References

[1] M. Exner, R. Srikumar, R. Blättner, M. T. Eiles, P. Schmelcher, and H. Ott, arXiv:2412.19710.

[2] R. Srikumar, M. Exner, R. Blättner, P. Schmelcher, M. T. Eiles, and H. Ott, arXiv:2502.15509.