Rydberg Macrodimers: From Polariton Decay to Molecular Interactions

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Rydberg macrodimers are molecules of two Rydberg atoms measuring up a micron in diameter [1]. Bound to by polarization forces, the bond energy energy can be as low as a few MHz, opening up new perspectives for light-mediated control of molecular bonds.

Here, I will revisit the familiar concept that coupling of an isolated quantum state to a continuum is associated with decoherence and decreased lifetime [3]. Due to their special properties [2], Rydberg macrodimers can overcome this dissipative mechanism and instead form bound states with the continuum of free motional states. This is enabled by the unique combination of extraordinarily slow vibrational motion in the molecular state and the optical coupling to a non-interacting continuum. Under conditions of strong coupling, we observe the emergence of distinct resonances where the macrodimer is hybridized with the continuum. For atoms arranged on a lattice, we predict molecules consisting of more than two atoms that appear in atom loss correlations in a quantum gas microscope. Our results present an intriguing light-mediated mechanism to control decoherence and bind multiatomic molecules using macrodimers.



Figure 1: A macrodimer molecule is exposed to a strong dissociating laser field, and yet does not fall apart.

References

- S. Hollerith, J. Zeiher, J. Rui, A. Rubio-Abadal, V. Walther, T. Pohl, D. M. Stamper-Kurn, I. Bloch, & C. Gross, *Science*, 364, 664 (2019)
- [2] S. Hollerith, K. Srakaew, D. Wei, A. Rubio-Abadal, D. Adler, P. Weckesser, A. Kruckenhauser, V. Walther, R. van Bijnen, J. Rui, C. Gross, I. Bloch, & J. Zeiher, *Phys. Rev. Lett.*, **128**, 113602 (2022)
- [3] S. Hollerith, V. Walther, K. Srakaew, D. Wei, D. Adler, D. Agrawal, P. Weckesser, I. Bloch, & J. Zeiher, PRX Quantum, 5, 030335 (2024)