

Quantum Many-Body Chemistry: Phase Matching and Entanglement Generation

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Chemical reactions in the quantum degenerate regime differ fundamentally from their normal gas counterparts. At quantum degeneracy, atoms and molecules form macroscopic matterwaves, and their reactions are governed by the nonlinear mixing of these matterwave fields. In this regime, quantum coherence and bosonic enhancement become central features, defining a new paradigm known as *quantum many-body chemistry*.

The realization of molecular quantum gases through coherent pairing of atomic Bose-Einstein condensates (BECs) opens new avenues for exploring guiding principles in quantum many-body chemistry. In our previous work, we confirmed Bose-enhanced reactive coupling in BECs of g -wave $^{133}\text{Cs}_2$ molecules [1,2]. Separately, we observed atom pairing mediated by a degenerate Fermi gas in a quantum degenerate Cs-Li Bose-Fermi mixture [3].

In this talk, I present a new study of matterwave phase matching and entanglement generation in the pairing reactions of atoms, inspired by analogies to nonlinear quantum optics. We perform matter-wave diffraction experiments on atom-molecule BECs to precisely measure the phase of the atoms and molecules. By imprinting a spatial phase modulation ϕ_a onto the atomic BEC, we observe doubling of the phase in the emerging molecular BEC $\phi_m = 2\phi_a$, consistent with coherent second-harmonic generation of matterwaves. Additionally, we detect signatures of weak entanglement in the molecular field, which we attribute to the intrinsic nonlinearity of the wave-mixing process.

These results reveal new mechanisms in quantum many-body chemistry and demonstrate the potential of molecular matterwave interferometry as a tool for precision studies of quantum correlations and entanglement.

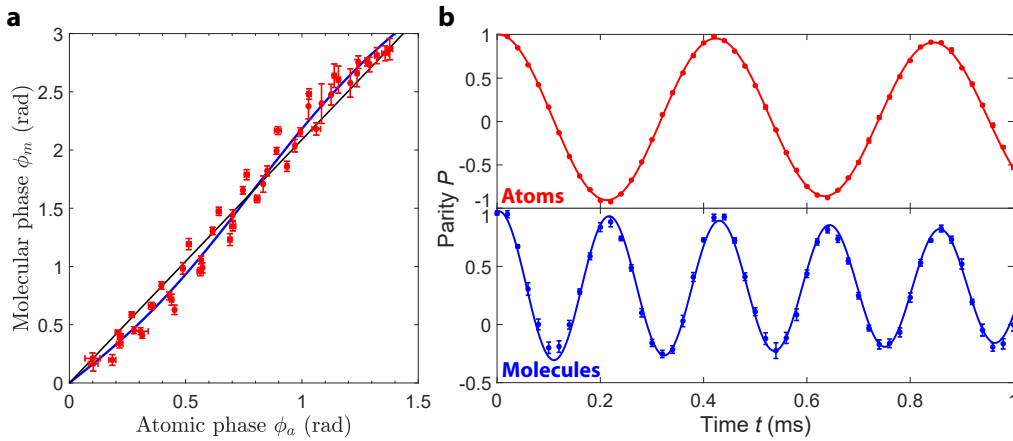


Figure 1: Phase matching and entanglement generation in quantum many-body chemistry. From atomic BECs with phase modulation ϕ_a , molecular BECs are produced with phase modulation ϕ_m near a Feshbach resonance. The phases ϕ_a and ϕ_m are measured from Bragg diffraction experiments. The data (red circles) are compared with the model (blue line) $\phi_m = \eta\phi_a - \epsilon \sin 2\phi_a$, from which we extract the linear ratio $\eta = 2.09(2)$ and the nonlinear parameter $\epsilon = 0.13(2)$. Black solid line is a linear fit with slope $\eta = 2.09$. **b**, Pseudo-spin parities P for atoms and molecules. The parities oscillate at the frequencies of 2.374(3) and 4.68(2) kHz for atoms and molecules, respectively, confirming phase doubling. Molecular states with negative parity indicates non-separability and entanglement of the constituent atomic pair.

References

- [1] Z. Zhang, L. Chen, K. Yao, and C. Chin, *Nature* **592**, 708 (2021).
- [2] Z. Zhang, S. Nagata, K. Yao, and C. Chin, *Nature Physics* **19**, 1466 (2023).
- [3] G. Cai, H. Ando, S. McCusker, and C. Chin, ArXiv:2502.06266