# Abstract


## Observation of 'Missing' Vibrational Levels and Extending Photoassociation Spectra of Excited Ultracold Cesium Dimers

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This thesis presents novel observations in photoassociation spectroscopy of cesium molecules, achieved through a series of benchmarking experiments conducted with the apparatus designed for realizing the creation of ultracold ground state KCs molecules and advancing the study of their properties. The benchmarking measurements on ultracold cesium atoms confirmed a theoretical prediction made over a decade ago, which postulated the existence of two 'missing' lowest-lying vibrational levels in the external well of the $0_g^-(6S_{1/2} + 6P_{3/2})$ state. We also provide unambiguous evidence that previously reported experimental works that claimed the priority in observing these new levels, in fact, observed levels belonging to other molecular states. As a complementary investigation, we utilize the same experimental apparatus to extend the observed range of vibrational energy levels of these other molecular states $0_u^+(6S_{1/2}+6P_{3/2})$ and $1_g(6S_{1/2}+6P_{3/2})$ to -300 $cm^{-1}$ from the $(6S_{1/2}+6P_{3/2})$ asymptote, surpassing the previously reported experimental limit of -120 $cm^{-1}$.

Following these novel observations, the potential energy curves of the molecular states under study were numerically reviewed. Our analysis of the states $0_u^+(6S_{1/2} + 6P_{3/2})$ and $1_g(6S_{1/2}+6P_{3/2})$ indicates that the extended measurements fall short for the molecular models used for long-range interactions, necessitating a refined model formulated for the intermediate range. The analysis of the potential energy curve of the $0_g^-(6S_{1/2} + 6P_{3/2})$ state on the other hand yielded results in agreement with the developed theoretical postulates. While our experimental measurements demonstrated accuracy below 1 MHz, it is essential to acknowledge that experimental data obtained in prior reported experiments from other groups exhibit accuracy of less than 60 MHz. We rely on prior experimental data for the previously probed vibrational levels, necessitating precise measurements of all vibrational energy levels to achieve a more accurate assessment of potential energy curves and important atomic and molecular constants.

Overall, our experimental apparatus for photoassociation shows promise for further studies within our research group, particularly showcasing its efficacy. Moving forward, our focus will shift towards conducting photoassociation spectroscopy of excited molecular states of KCs, with the aim of facilitating the formation of ground-state KCs molecules. Additionally, our future plans involve extending these investigations to KAg and CsAg molecules as part of a recently initiated research project.