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Photoassociation Spectroscopy of Purely Long-Range Krypton Molecules

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Synopsis Photoassociation spectra for purely long-range (PLR) krypton molecules are both computed and measured. The photoassociation laser is tuned just red of the $5s[3/2]_2-5p[5/2]_2$ transition at 810.6 nm, over a frequency range in which fourteen different PLR potential wells support hundreds of vibrational bound states. In the experiment, laser light is directed into a cloud of magneto-optically trapped ^{84}Kr atoms, and the emission of both ultraviolet photons and ions is monitored. Peaks in these signals are observed for laser frequencies within 200 MHz of the atomic resonance. Ongoing work is aimed at improving the experimental signal and understanding the degree of agreement between empirical data and computed molecular spectra.

We investigate the photoassociation of metastable krypton atoms in a magneto-optical trap. The initial goal of our experiments is the detection and spectroscopic characterization of Kr bound states in purely long-range (PLR) molecular potential wells. Similar states have been studied in the case of "giant helium dimers" by a group at ENS, Paris [1, 2]. The most notable achievement of that effort was the characterization of the PLR 0_u^+ potential, which has a local minimum at an internuclear distance of $R \approx 10$ nm and forms a shallow well only 2 GHz in depth. Measured and computed ro-vibrational spectra were in agreement to within 1 MHz. Our present work aims at a comparable level of understanding for several PLR states that have been computed for krypton [3].

We expose a cloud of magneto-optically trapped ^{84}Kr atoms with light from a frequency-tunable photoassociating (PA) laser, and we detect ultraviolet fluorescence from the spontaneous decay of the molecules that are produced during this exposure. Our trap holds roughly 10^6 atoms at a density of about $10^{11}/\text{cm}^3$ and a temperature on the order of 10 μK . The PA pulses last 10 microseconds and are introduced while the trapping lasers are momentarily turned off. The PA frequency is tuned slightly red of the $5s[3/2]_2-5p[5/2]_2$ transition ($\lambda = 810.6$ nm), in the range over which PLR states are predicted. Scans of this frequency are calibrated through a saturated-absorption signal and the transmission spectrum of a Fabry-Perot interferometer. We use a photon multiplier in photon-counting mode to measure the ultraviolet fluorescence from the cloud ($\lambda = 123.6$ nm). We also record an ion signal that indicates the rate of Penning ionization produced by atomic collisions.

There are fourteen PLR wells (s + p potentials)

in Kr that are expected to be accessible in our experiments. We have computed the array of potentials and reproduced the results of [3]. In addition, we have computed the bound state energies in each of the wells and generated theoretical spectra that guide the experimental search for resonances. Our experiments themselves are ongoing and are still aimed at optimizing the signal. Preliminary data seem to show molecular peaks within 200 MHz of the atomic resonance, but evidence for states lying deeper in the PLR potentials is not yet definitive.

A longer-term goal of this project involves spectroscopy of PLR molecules composed of various pairings of Kr isotopes. Pairing-dependent shifts in the spectra will provide an empirical test of theoretical molecular potentials in which the dominant interaction in PLR potential wells is expected to exhibit an unusual crossover, changing from resonant dipole-dipole (C_3/R^3) to non-resonant coupling that is higher-order in $(1/R)$ at longer range. Mixed-isotope photoassociation spectra can serve as a probe of the interactions that are expected to modify the PLR potentials described in reference [3].

The poster presentation will give additional theoretical details on the atomic and molecular structure involved in the experiment, the experimental setup and procedure, and the progress of our effort. Work supported by grant NSF-PHYS-0554807.

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