Two-Color Photoassociation Spectroscopy of the Triplet Ground State of Na Dimers

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Abstract

We use a type of Autler-Townes spectroscopy to measure the position of approximatley 100 individual states of the lowest triplet state of Na_2. The spectroscopy starts with the photoassociation of ultracold atoms in a magneto-optical trap. These states have binding energies of up to 811 GHz and were measured with a typical resolution of 20 MHz, an improvement of three orders of magnitude over previous results. These vibrational levels are found by inducing an Autler-Townes splitting between an intermediate excited state and a particular ground state. By choosing states of different angular momentum character within the excited vibrational level, we were able to resolve the vibrational, rotational and hyperfine features of these states.

Recent work in cold collisions and Bose-Einstein condensation of alkali gases has led to a renewed interest in the interaction potentials of alkali dimers. Further interest has been kindled by the prospects for the creation of a Bose-Einstein condensate of molecules. Such an accomplishment will likely entail detailed knowledge of the lowest triplet-state potential of an alkali dimer and detailed control of the molecule formation process. With much of this in mind, the lowest singlet- and triplet-state potentials ($X\ ^1\Sigma_g^+$ and $a\ ^3\Sigma_u^+$) have been recently refined for a number of alkali dimers. Na_2 spectroscopy has traditionally been particularly well-studied because of its accessibility with tunable dye lasers. Recently, the Na_2 singlet ground-state potential has seen substantial refinement by molecular-beam spectroscopy methods [1, 2, 3]. Here, we similarly improve the spectroscopy of the lowest triplet potential of Na_2 by performing multi-color photoassociation spectroscopy of laser-cooled sodium atoms.

The technique involves the perturbation of an ionization signal by the Autler-Townes splitting of a line that is an intermediate in a photoassociation-ionization process. Such a technique has been used

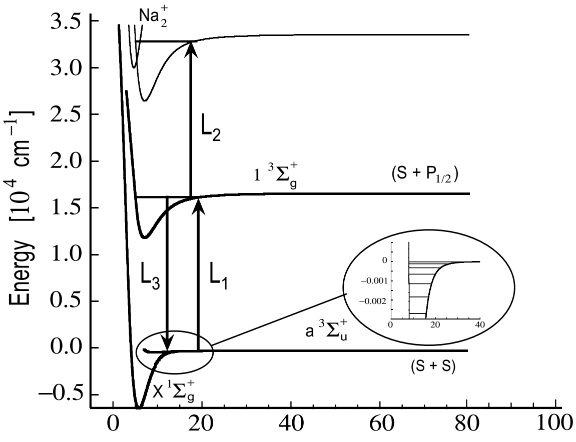

Figure 1: Adiabatic potential energy diagram for the potentials in our experiment. The zero of energy is defined as the $f = 1 + f = 1$ free-atom, hyperfine asymptotic limit. The inset is a blow-up of the $a\ ^3\Sigma_u^+$ potential, showing the location of the last few bound levels.

previously in combination with the photoassociation of alkali dimers. Our implementation is discussed in detail in Ref. [4], where we also reported the observation of the highest (v = 15) bound and quasi-bound levels of the $a\ {}^3\Sigma_u^+$ potential of $\mathrm{Na_2}$.

Briefly, a laser is tuned to photoassociate atoms held in a dark-spot magneto-optical trap (MOT) [5]. The atoms are held at a temperature of approximately 0.4 mK and a density on the order of 10^{10}–10^{11} cm^{-3}, primarily in the $f = 1$ hyperfine state. As shown in Figure 1, the photoassociation laser ($\mathrm{L_1}$) is tuned to a bound state of the $1\ {}^3\Sigma_g^+$ molecular potential. A second laser ($\mathrm{L_2}$) is then introduced to resonantly ionize this state through an autoionizing doubly-excited state of $\mathrm{Na_2}$. Fixing the frequency of these two lasers, a steady ionization signal is obtained. A third laser ($\mathrm{L_3}$) is then introduced and scanned to perform the spectroscopy. When this third laser hits a bound-bound resonance between the excited intermediate state and another state in the $a\ {}^3\Sigma_u^+$, it produces an Autler-Townes splitting of this pair of levels. This splitting frustrates the photoassociation, thus reducing the ion signal.

The $1\ {}^3\Sigma_g^+$ potential is chosen as the intermediate state for both a strong photoassociation transition and good access to the $a\ {}^3\Sigma_u^+$ potential. The initial cold collision state has both $X\ {}^1\Sigma_g^+$ and $a\ {}^3\Sigma_u^+$ character, allowing the initial photoassociation transition from the $a\ {}^3\Sigma_u^+$ component of the scattering state. The spectrum of the excited intermediate state shows a rotational progression with an underlying, resolved hyperfine structure. The second, bound—bound transition is allowed by conventional optical selection rules between these triplet states.

Scanning laser $\mathrm{L_3}$ produces the spectra shown in Figure 2 for v = 10 of $a\ {}^3\Sigma_u^+$. The different traces are obtained starting from intermediate states of different angular momentum character: By choosing different rotational-hyperfine intermediate states, we observe a variety of rotational and hyperfine states of the a $a\ {}^3\Sigma_u^+$ potential. The structure of the other vibrational levels is similar.

We have used this multi-color photoassociation technique to measure the binding energies of the 8 highest vibrational levels ($v = 8$–15) in the lowest triplet potential of $\mathrm{Na_2}$. (Lower vibrational levels (v ¡ 8) of the $a\ {}^3\Sigma_u^+$ potential could not be observed through either intermediate state, due to suppressed Franck-Condon factors. The limited tuning range of our photoassociation and ionization lasers prevented us from using more deeply bound intermediate states, which would have better overlap with $v < 8$.) The precise location of the $a\ {}^3\Sigma_u^+$ levels is found by fitting Bohn and Julienne's [6, 7] lineshape formula to the data.

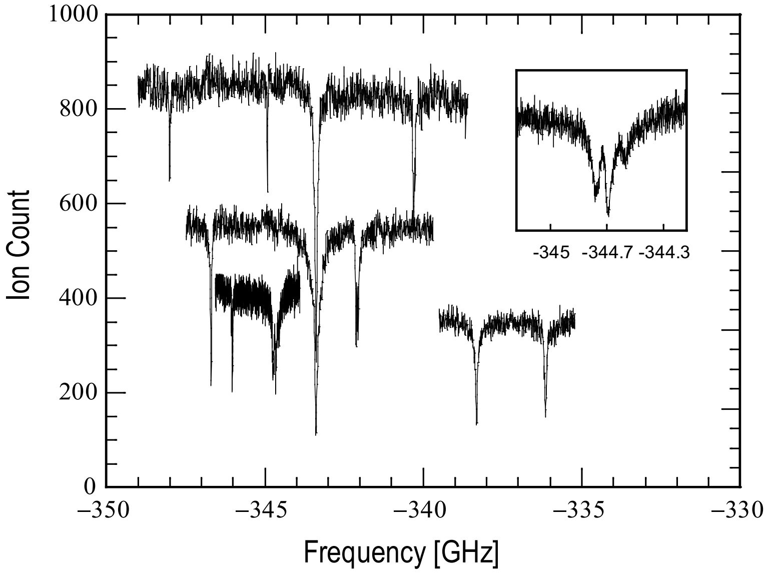

Figure 2: The spectra of the v = 10 vibrational level of the $a\ {}^3\Sigma_u^+$ potential, as seen through different intermediate states. The ion count is plotted versus the frequency difference of lasers $\mathrm{L_1}$ and $\mathrm{L_3}$. The inset shows the splitting of a line due to the magnetic dipole interactions of the electron spins of the two atoms.

Both the resolution and accuracy of the measurements have improved, by about 3 orders of magnitude (to 20 MHz and 15 MHz, respectively), over previous observations. In addition, we have observed a number of new vibrational lines. While we have measured lines only in the upper part of the potential ($v = 8$–15), we have observed spectral features corresponding to rotational, hyperfine, and spin-spin structure in the molecule. We find that the observed spectra are well described by a model in which hyperfine splitting dominates over spin-spin splitting. A simple perturbative model qualitatively describes the observed structure.

Acknowledgements

We thank Craig Sansonetti and Bill Degraffenreid for assistance in confirming the frequency accuracy of our iodine cell. We thank Jon T. Hougen for helpful conversations and reading the manuscript. Partial funding was provided by the U.S. Office of Naval Research.

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