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PHOTOASSOCIATION OF ULTRACOLD ATOMS

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ABSTRACT

Cold-atom photoassociation is important as a novel molecular bond formation mechanism, as a loss mechanism in optical atom traps, as a possible source of ultra-cold molecules, and as a new spectroscopic method.¹⁻³ In this process, a colliding pair of laser-cooled atoms A and B absorbs a photon of frequency ν to produce a bound, excited molecule AB^{*}. Photoassociation resonances occur that are associated with the excitation of specific ro-vibrational states of AB^{*}; these resonances can be very narrow because the kinetic energy spread of the atoms is small at low temperature (e.g. $k_BT/h = 21$ MHz at T = 1 mK). Unique, long-range molecular states are readily studied because the initial collisional state has a large amplitude at long-range.

We have carried out photoassociation studies of trapped, laser-cooled Rb atoms, and explored Rb₂[•] states with binding energies from less than 0.3 cm⁻¹ to more than 1000 cm⁻¹, with outer turning points from 17 a₀ to 250 a₀, and with a resolution of order 1 MHz. The Rb atoms are held in a far-off resonance optical dipole force trap, have a density of order $10^{12}/\text{cm}^3$, and a temperature of about 1 mK. We detect the photoassociation resonances by monitoring the loss of atoms from the trap, which occurs because the excited Rb₂[•] dimers decay predominantly to free states with an energy much greater than the trap depth. These studies have revealed many new features of long-range molecular states. Several vibrational series are observed, with intensity oscillations that reflect the structure of the initial collisional state wavefunction. We observe the bound levels of the unique 0_g⁻ molecular "pure long-range state", which has an inner turning point beyond 25 Bohr. We also observe well-resolved rotational structure and predissociation broadening of the photoassociation resonances.

Long range interaction potentials can be derived from the photoassociation spectra. We can extract the coefficient C_3 of the dominant C_3/R^3 resonant dipole interaction between the atoms to an accuracy of about 1%, and may eventually be able to realize 0.1% accuracy. This analysis indirectly yields the excited atomic lifetime to comparable accuracy. Information on the ground state interaction potential is contained in the free-bound Franck-Condon factors, and in the intensities and lineshapes of rotationally resolved lines. These lines exhibit asymmetric lineshapes that reflect the distribution of initial collisional state energies, as modified by transmission of the colliding atoms through the ground state centrifugal barrier.

Analysis of the photoassociation spectra is simplified if the atoms are prepared in the spin-polarized (F=3, $M_F=3$) state, because to a good approximation the collisions then occur on a single, triplet potential curve. As illustrated in Fig. 1, we have obtained photoassociation spectra with spin-polarized atoms. An analysis of this spectrum, in progress, should yield the ground state triplet scattering length and coefficient C₆ of the long range -C₆/R⁶ atomic interaction.

In summary, we have obtained highly resolved cold atom photoassociation spectra which provide a wealth of information on the long-range interactions between the atoms. With further analysis, an almost complete characterization of both excited and ground state interactions should be possible. We gratefully acknowledge the support of the National Science Foundation, the A. P. Sloan Foundation, and the R. A. Welch Foundation.



Fig. 1. High-resolution photoassociation spectrum of ⁸⁵Rb atoms, showing the rotationally resolved structure of a single 0_g^- vibrational line. a) Atoms are optically pumped into the F = 3, $M_F = 3$ ground state sublevel. b) Atoms are pumped into F = 3, but are in a random distribution of Zeeman sublevels.

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