Saturation of Cs₂ Photoassociation in an Optical Dipole Trap

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We present studies of strong coupling in single-photon photoassociation of cesium dimers using an optical dipole trap. A thermodynamic model of the trap depletion dynamics is employed to extract absolute rate coefficients. From the dependence of the rate coefficient on the photoassociation laser intensity, we observe saturation of the photoassociation rate coefficient at the unitarity limit in quantitative agreement with the theoretical model by Bohn and Julienne [1]. Also the corresponding power broadening of the resonance width is measured. We could not observe an intensity dependent light shift in contrast to findings for lithium and rubidium, which is attributed to the absence of a p or d-wave shape resonance in cesium.

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Ultracold thermal and quantum degenerate atomic ensembles have allowed to investigate powerful coupling schemes between continuum scattering states of two atoms and bound molecular states of the corresponding dimer. Both photoassociation light [2,3] or magnetic field sweeps across Feshbach resonances [4,5,6] have lead to the formation of ultracold molecules. Three-body recombination of fermionic atoms near a Feshbach resonance has finally lead to the creation of molecular Bose Einstein condensates [7,8,9]. Recently, molecular photoassociation has been performed using a two-photon Raman process in a two-atom Mott insulator phase [10], which may prove to be a route to a molecular Mott insulator and, subsequently, an alternative way to a molecular BEC [11]. In contrast to this, the successful formation of ultracold molecular ensembles in the absolute rovibrational ground state, which is important if one wants to study molecular collisions and reactions over long interaction times, is still at large. Future routes for the production of absolute ground state molecules from a pair of atoms will certainly involve Raman type coupling schemes [12,13,14]. A very interesting approach to transfer highly vibrationally excited molecules into the vibrational ground state proposes the application of optimally controlled femtosecond laser pulses [15].

In order to understand the regime of strong coupling in photoassociation, which is important for both continuous and femtosecond Raman processes, we report in this Rapid Communication investigations of saturation effects in the first photoassociation step from the two-atom continuum to an excited molecular level. This is also the determining step in the overall molecule formation rate [16]. Using an ultracold ensemble of cesium atoms trapped in a quasi-electrostatic optical trap [17] we are able to employ trap loss measurements to extract absolute photoassociation rate coefficients [18], from which the dependence of the rate coefficient on the photoassociation laser intensity is derived.

Previously, Schlöder et al. have investigated strong coupling in a ⁶Li²Li two-species magneto-optical trap and observed broadening and center shifts of several photoassociation resonances [19]. Comparing their results to the semiclassical model of Bohn and Julienne [1] a saturation intensity of 30-50 W/cm² was derived [19]. McKenzie et al. [20] and Prodan et al. [21] have studied photoassociation out of a Bose-Einstein condensate of Na and ⁷Li, respectively, to determine the limit for the photoassociation rate coefficient. While McKenzie et al. have observed a linear relationship between laser intensity and association rate up to about 1 kW/cm², Prodan et al. clearly find that the photoassociation rate saturates at the unitarity limit of the inelastic scattering cross section, due to probability conservation in the quantum mechanical scattering process [21]. These results are conveniently interpreted in the framework of the two-body scattering model of Ref. [1], as long as the low intensity regime is concerned. Effects of high photoassociation laser intensities have been investigated in several other papers, emphasizing the multichannel character of the photoassociation process which have to be described within a molecular dressed-state approach. Specific discussions on cold sodium and rubidium photoassociation are proposed in Ref. [22], on strontium photoassociation in Ref. [23], and on photoassociation in a Na Bose-Einstein condensate in Ref. [24].

The photoassociation experiments are performed with a thermal ensemble of cesium atoms trapped in an optical dipole trap formed by the focus of a CO₂ laser [13]. As described in Refs. [25,26], the cesium atoms are loaded from a magneto-optical trap which is superimposed in the focus of the CO₂ laser. The MOT is operated in five-beam configuration and loads from a Zeeman slowed beam up to 10⁸ particles at a density of 10⁹ cm⁻³ as inferred from absorption imaging.

After turning of the magnetic field of the MOT a brief mol-lases cooling phase transfers about 5 × 10⁵ atoms at a density of 5 × 10¹³ cm⁻³ and a temperature of 40 μK into the optical dipole trap [25,26]. For the cold cesium atoms the CO₂ laser focus with its potential depth of 0.8 mK represents a harmonic trap with axial and radial trap frequencies of 12.8 Hz and 625 Hz respectively. The lifetime of the atoms in the dipole trap is of the order of 100 s, due to collisions with residual gas atoms.
The photoassociation light is provided by a widely tunable Titanium:Sapphire laser (Coherent MBR 110) system with a typical output power of 200 mW, and a line width of about 100 kHz. Relative frequency changes are monitored by measuring the transmission signal of the Ti:Sapphire laser through a confocal cavity with a free spectral range of 500 MHz. In order to decrease the frequency spacing of the transmission peaks, two side-bands are modulated onto the Ti:Sapphire beam at ±166 MHz using a double-pass AOM setup. The length of the cavity is stabilized by locking one of the cavity mirrors with a piezo actuator to the fringe of the transmission signal of a superimposed spectroscopy stabilized diode laser. This provides a relative frequency accuracy of about 5 MHz. The absolute laser frequency is measured with a commercial wavemeter (Burleigh WA 1000) with an accuracy of 500 MHz. The photoassociation laser is passed through the trapped cesium cloud in the focus of the CO$_2$ laser at an angle of 22.5° with respect to the CO$_2$ laser beam. The width of the Ti:Sapphire beam at the trap center amounts to about 150 µm. The intensity of the photoassociation beam has been changed between 30 to 400 W/cm$^2$.

Once the cesium atoms are loaded into the dipole trap, the shutter of the photoassociation laser is opened and the atom cloud is illuminated for 1 s. Then the CO$_2$ laser light is extinguished and all remaining cesium atoms are recaptured into the magneto-optical trap. The number of recaptured cesium atoms is obtained from the fluorescence signal of the MOT with an accuracy of 30%. Since the storage time of atoms inside the magneto-optical trap can range up to 100 s the atom loss signal is an accuracy of 30%. Since the storage time of atoms inside the magneto-optical trap. The number of recaptured cesium guished and all remaining cesium atoms are recaptured into

By fitting a single Lorentzian to the $v = 6, J = 2$ resonances, as shown in Fig.2 we obtain the maximum photoassociation rate coefficient, the resonance width and the relative resonance position as a function of the photoassociation laser intensity (see Fig. 3). The peak width is corrected for the 3.5 MHz inhomogeneous AC stark shift that the atoms experience in the dipole trap. The thermal broadening of the resonances at the temperature of the cesium atoms at 40 µK corresponds to less than 1 MHz and is also corrected for.

From the fitted rate coefficient $G$ the quantum mechanical scattering probability $p_{\text{scatt}}$ is given by

$$p_{\text{scatt}} = \frac{G}{G_{\text{unitarity}}}$$

where the rate coefficient at the unitarity limit denotes

$$G_{\text{unitarity}} = \frac{\Gamma \phi (|\epsilon_v|)}{\hbar^2} = \frac{2\pi}{\sqrt{\frac{\mu^2}{\hbar^2} k_B T}}.$$  

When plotting the scattering probability as a function of the intensity for intensities up to 500 W/cm$^2$ (see Fig. 3) saturation is clearly observable. Within the experimental accuracy the measured scattering probability always stays below unity, in agreement with the unitarity limit. The resonance width increases linearly with intensity (see Fig. 3), whereas the position of the resonance remains unchanged (see Fig. 3).

These results are compared to the theoretical model of Bohn and Julienne for photoassociation in strong laser fields [1], giving the scattering probability:

$$p_{\text{scatt}} = \frac{\gamma I}{\pi (\Gamma + \delta_f)^2} = \frac{I I_S}{\pi (I + I_S)^2},$$

where $\gamma$ is the natural linewidth of the transition. $\Gamma$ denotes the coupling strength defined by the overlap of the initial continuum wavefunction $|\phi(E_{\text{kin}} \rightarrow 0)\rangle$ with the final bound state wavefunction $|\psi(v)\rangle$

$$\Gamma = 2\pi (V_{eg})^2 |\langle \psi(v)| \phi(E_{\text{kin}} \rightarrow 0)\rangle|^2.$$  

$V_{eg}$ is the Rabi frequency of the electronic transition, which is proportional to the square-root of the laser intensity. The saturation intensity $I_S$ is introduced according to $I/I_S = \Gamma/\gamma$. In this formalism the resonance width is given by

$$\delta_f \text{FWHM} = \gamma + \Gamma = \gamma (1 + \frac{I}{I_S}).$$

The saturation intensity is obtained from the measured data for the scattering probability and the resonance width by a fit with Eqs. 2 and 3 (see Fig. 3). The two fits agree very well with each other and yield a saturation intensity of 460±90 and 450±150 W/cm$^2$, respectively. The fit to the scattering probability used the maximum scattering probability as a second free parameter because the overall accuracy of the scattering probabilities of about 40%, due to the accuracy of the extracted rate coefficients, is larger than the accuracy of the fit. The fit value of 1.36±0.07 for the maximum scattering probability is in good agreement with unity within this experimental accuracy.
By extrapolation to zero intensity the natural line width $\gamma$ of the photoassociation transition can be extracted. From the fit, a value of $17 \pm 2$ MHz is derived. This value is larger than the predicted theoretical resonance width of 10.4 MHz that amounts to twice the atomic line width of the $6s - 6p$ transition. Thus, the measured width may be the consequence of additional broadening mechanisms. One mechanism that could be responsible for the discrepancy between the prediction and the experiment is the lifting of the degeneracy of the magnetic sub-levels of the $J = 2$ state in the electric field of the CO$_2$ laser [27]. We have estimated the energy difference between the different $m_J$ levels to be about 7 MHz, assuming that the molecular polarization along the internuclear axis is twice the atomic polarization. This interpretation of aligned molecules in the field of the CO$_2$ laser trap will be investigated further in future experiments. Another possible explanation could be found in Ref. [28], where photoassociation is described within the molecular dressed state approach at low intensities. Due to the very low kinetic energy in the entrance channel, the continuum wavefunction and the photoassociation wavefunction are predicted to be coupled at low photoassociation intensities (typically 10 W/cm$^2$) to the last bound state lying very close to the Cs(6s)+Cs(6s) limit, inferred by its large scattering length. This contribution is not included into the model of Ref. [1]. The consequent multi-channel character of the photoassociation process described now as a two-level plus continuum coupled system, would induce a non-linear variation of the linewidth, which could well converge to the expected doubled atomic value. This hypothesis, which is also discussed in Ref. [23], is not easy to check in experiments due to the weakness of the photoassociation signal at very low intensities.

Ref. [1] also predicts a linear shift of the absolute resonance position, previously observed in a $^7$Li quantum degenerate gas [21], and in a Na Bose-Einstein condensate [20]. In the latter case, the shift is attributed to the presence of a shape resonance in the $d$-wave collisional entrance channel according to the prediction of Ref. [22]. Simoni et al. [22] also predict a light shift enhanced by a $d$-wave shape resonance. In both the Na and Rb case, the calculated light shift ignoring the shape resonance is found negligible. The clear absence of a shift in our measurements for intensities up to the saturation intensity seems to support the absence of any $p$- or $d$-wave shape resonance in cesium. The calculations performed in Ref. [28] also predicts a negligible light shift in the presently explored intensity range. It should be noted that the absence of a light shift in the present experiment also leads to a correspondingly negligible inhomogeneous broadening of the photoassociation resonance, in contrast to previous saturation studies where it had to be accounted for [14, 20].

Photoassociation experiments inside a quasi-electrostatic optical dipole trap allow for precise measurements of absolute photoassociation rate coefficients, due to the high pair density, the long interaction times, and the well defined internal hyperfine state of the trapped atoms. This allows us in this work to investigate quantitatively the saturation of the photoassociation rate coefficient with increasing photoassociation laser intensity. It is found that the scattering probability saturates at unity, corresponding to the the unitarity limit of this inelastic scattering process. The same result was concluded in the experiment with quantum degenerate $^7$Li [21]. Saturation at the unitarity limit has to be distinguished from saturation in an optical transition, thus Rabi oscillations between the pair of atoms and the molecular dimer, for which we see no indication, are not expected under these conditions. We also observe that the photoassociation resonance width exhibits a linear increase with intensity. Both the saturation and the width increase are in good quantitative agreement with the theoretical model of Bohn and Julienne using the saturation intensity as the only parameter of the model, for which a value of $450 \pm 100$ W/cm$^2$ is obtained. A light shift is found to be strongly suppressed, which is attributed to the lack of a $p$ or $d$-wave shape resonance in cesium, in contrast to the findings for sodium or rubidium.

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FIG. 1: Overview of the important potential energy curves for the photoassociation of cesium molecules.
FIG. 2: Upper panels: trap loss spectra of the studied photoassociation resonance $v = 6, J = 2$ of the $0^g_2$ outer well for different laser intensities. Lower panels: Spectra of the photoassociation rate coefficients extracted from the trap loss signal. The solid lines represent Lorentzian fits to the resonances.

FIG. 3: Photoassociation scattering probability (upper panel), resonance width (middle panel) and resonance line shift (lower panel) as a function of the photoassociation laser intensity. The solid lines represent the fit to the saturation model of Bohn and Julienne.