Dipolar effects and collisions in an ultracold gas of LiCs molecules

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Abstract. We review our recent experiments on the formation and optical trapping of LiCs molecules, which are photoassociated from a mixed ultracold gas of Li and Cs atoms. The electric dipole moment of the atoms in the lowest vibrational states is measured, yielding the predicted value of 5.5 Debye, and vibrational redistribution due to spontaneous emission and blackbody radiation is observed. We also show preliminary results on inelastic collision between optically trapped LiCs molecules and Cs atoms.

1. Introduction
In the past two years, the quest for ultracold molecules in deeply bound internal states, in particular the rovibrational ground state, has progressed with breath-taking speed. Previous work demonstrated the conceptual possibility to populate the vibrational ground state of ultracold homonuclear molecules (K2) [1] and heteronuclear molecules (RbCs) [2] by applying photoassociation (PA). In 2008, several groups achieved major breakthroughs to increase the number of molecules in the vibrational ground state and to selectively populate the rotational ground state. The molecules were either optically pumped into the vibrational ground state by shaped broadband laser pulses following PA [3], or directly formed by PA of ultracold atomic gases [4]. In an alternative approach, the molecules were created by magnetoassociation at high phase-space density in the vicinity of a Feshbach resonance from a quantum-degenerate gas, and subsequently adiabatically transferred into the rovibrational ground state by one or two consecutive stimulated Raman adiabatic passage (STIRAP) steps [5, 6, 7] (see also Christoph Nägele’s contribution to these Proceedings).

In two of these experiments, one in the group of Deborah Jin and Jun Ye at Boulder and one in our group, formerly in Freiburg, now in Heidelberg, the molecules were formed from a mixture of ultracold atomic gases, thus creating molecules with a permanent body-fixed electric dipole moment (EDM) (KRb in Boulder, LiCs in Freiburg/Heidelberg). Ultracold gases of polar molecules (see the recent reviews [8, 9]) offer intriguing perspectives to the exploration of complex many-body quantum systems [10, 11], the development of quantum computation

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techniques [12], precision measurements of fundamental constants [13], and the investigation and control of ultracold chemical reactions [14]. Current experimental efforts are therefore aiming at the investigation of effects related to the polar nature of the ultracold molecules. Here, we will concentrate on progress being made with a gas of ultracold LiCs molecules in low rovibrational states. LiCs is of particular interest as its dipole moment is as large as 5.5 Debye [15, 16], thus featuring the largest electric dipole moment of all bialkali molecules. We will briefly review our experimental approach to the formation of an ultracold gas of LiCs molecules and present a measurement of the LiCs dipole moment. Then, we will show evidence for vibrational relaxation and blackbody redistribution as a direct consequence of the large dipole moment. In the last part, we show first humble steps towards Ultracold Chemistry by presenting inelastic collisions between atoms and molecules trapped in a quasi-electrostatic optical dipole trap.

2. Photoassociation and detection of an ultracold gas of LiCs

The experimental sequence for formation, detection, and high-resolution spectroscopy of ground state molecules is schematically shown in Fig. 1. Ultracold LiCs molecules are formed by photoassociation (PA) [17, 18] of laser cooled $^{133}$Cs and $^7$Li atoms. The atoms are cooled and trapped in two overlapped magneto-optical traps (MOTs) which are loaded via a single Zeeman slower from a double species oven. For the Cs atoms the MOT is operated in the configuration of a dark spontaneous force optical trap (dark SPOT) [19] to reduce inelastic inter-species collisions. In this configuration $4 \times 10^7$ $^{133}$Cs and $10^8$ $^7$Li atoms are trapped simultaneously at densities of $3 \times 10^9 \text{cm}^{-3}$ and $10^{10} \text{cm}^{-3}$, and temperatures of 250(50) $\mu$K and 600(150) $\mu$K, respectively. Details of the experimental setup for the formation and detection of ultracold LiCs molecules have been given elsewhere [4, 20, 21]. For PA of LiCs molecules the two overlapped atom clouds are continuously illuminated by light from a tunable cw TiSa laser. The excited LiCs molecules decay spontaneously within few tens of nanoseconds into different electronic ground state levels. Molecules in given vibrational ground state levels are then ionized by resonant-enhanced multi-photon ionization (REMPI) and the resulting ions are detected in a time-of-flight (TOF) mass spectrometer [21].

Rotationally resolved spectroscopy of ground state molecules is achieved by depletion spectroscopy [22]: a narrow band laser pumps population out of a selected rovibrational ground state level leading to a reduction in the detected ion signal from this level. Further details have been given previously [20]. Electric fields between 0 and 500 V/cm can be applied to the atomic and molecular samples by two round electrodes inside the vacuum with an outer diameter of 80 mm and a distance of 40 mm, centered around the trapped atom clouds. The electrodes have a central hole (ø 30 mm) to allow optical access and extraction of produced ions (these electrodes are also part of the mass spectrometer used for the detection of the ground state molecules [21]). The electric field created at the trap position is derived from the measured dimensions and voltages using the simulation package SIMION (v8.0) with an uncertainty of 3% due to estimated inaccuracies in the electrode geometry and voltages. For the detection of molecules the fields are switched 0.6 ms before ionization to a fixed value of 38 V/cm.

In order to obtain trapped samples of LiCs molecules, PA of atoms stored simultaneously in a quasi-electrostatic trap (QUEST) is performed. The QUEST is formed by the single focus of a CO$_2$ laser (power 64(4) W, waist $w_0=89(4) \mu$m) emitting at 10.6 $\mu$m. Details of the QUEST setup can be found in [23] and references therein. The resulting trap depths for the different species are calculated from measured values for beam power and waist, experimental values for atomic polarizabilities [24], and ab-initio calculated values for the molecular polarizabilities [25] as 194(21) $\mu$K for Li and 479(53) $\mu$K for Cs. Due to the large detuning from all electronic transitions the trap depth for LiCs molecules varies only slowly with the vibrational index $v''$, from a minimum value of 435(48) $\mu$K for $X^\Sigma^+,v''=0$ to a maximal trap depth of 700(77) $\mu$K for $v''=42$. Despite its significant detuning, the optical trapping field could drive Raman transitions...
between neighboring levels due to its high intensity. The rate for such transitions can be estimated from a simple classical model of the Raman process [26]. Using the \textit{ab-initio} calculated polarizability tensor for the $X^1\Sigma^+$ state of LiCs [25] and the experimental laser intensity we derive transition rates which are several orders of magnitude below the transition rates for spontaneous decay and black-body radiation (BBR) transitions. Thus vibrational or rotational heating by the trapping CO$_2$ laser can be neglected even on longer timescales.

After a variable hold time in the dipole trap, molecules in given vibrational ground state levels are ionized by a REMPI pulse (typical intensities 2 J/cm$^2$) and the resulting ions are detected in the TOF spectrometer. The slowest measured decay of a molecular signal from the trap corresponds to a time constant of 24(4) s. This value is comparable to the lifetime of pure atomic samples in the trap and is thus attributed to trap loss due to collisions with thermal background gas at a pressure of $2 \times 10^{-11}$ mbar. Further details of the experimental setup for the formation and detection of ultracold LiCs molecules can be found elsewhere [4, 20, 21].

3. Measurement of the electric dipole moment

In order to determine the permanent EDM of a given vibrational level of the $X^1\Sigma^+$ state we measure the electric-field induced shift in the transition frequency between the $J''=0$ component of this level and a rovibrational level in the $B^1\Pi$ state [16]. A shift in the transition frequency originates in the combined Stark shifts of the $X^1\Sigma^+$ level and the excited $B^1\Pi$ level. A rotationless level $v''$ ($J''=0$) of the $X^1\Sigma^+$ state experiences an electric-field induced shift $\Delta W = -\mu_v E^2/(6\hbar B_v)$, where $\mu_v$ is the permanent EDM, $E$ the external field, and $B_v$ the rotational constant [27]. Rotational constants are calculated from an experimental $X^1\Sigma^+$ potential energy curve [28] using Level 8.0 [29]. Due to selection rules the upper level in the depletion transition has to be rotationally excited ($J'=1$). The $B^1\Pi$ state is predicted to have a smaller EDM than the ground state: a pseudopotential configuration interaction calculation as described in ref. [15] yields for the $B^1\Pi$ state a permanent EDM of 0.47 Debye at the equilibrium
distance. One can thus expect a significantly smaller Stark effect for the upper level than for the ground state level, as verified experimentally.

For the formation of deeply bound molecules we use the PA resonance $B^{11}\Pi, v'=18, J'=1$. After spontaneous decay the rotational states $J''=0$ of $X^{1}\Sigma^{+}, v''=2$ and higher are significantly populated. Then depletion spectroscopy of $v'=1, J'=0$ and $v'=3, J''=0$ is performed in electric fields varying from 0 to 488 V/cm. We did not obtain depletion spectra with sufficient signal to noise ratio for Stark spectroscopy from the $J''=0$ component of the $v'=0$ and $v''=1$ levels. Molecules in the lowest vibrational levels $v''=0$ and $v''=1$ are also formed by PA, but mostly in the rotationally excited $J''=2$ components [4, 30] for which the Stark effect at feasible electric fields could not be experimentally observed, yet. The change in the frequency of the transition $X^{1}\Sigma^{+}, v''=2, J''=0 \rightarrow B^{11}\Pi, v'=18, J'=1$ is determined via depletion spectroscopy. We find a permanent EDM of $\mu=5.5(2)$ Debye for $v''=2$, where the reported error stems from the uncertainty of the fit and the estimated uncertainty in the electric field determination. The same measurement and analysis was performed for molecules in $X^{1}\Sigma^{+}, v''=3, J''=0$. Using $B_{v''=3}=0.1836$ cm$^{-1}$ the value for the EDM is determined as $\mu=5.3(2)$ Debye [16].

4. Dipolar relaxation of LiCs molecules

The presence of a permanent electric dipole moment in a molecule leads to a coupling of the internal molecular state to the thermal environment via black-body radiation (BBR). Thus if the system is prepared in a certain internal state, this state will be altered after a finite time. In particular this yields a limited lifetime even for molecules prepared in the absolute rovibrational ground state. In the case of LiCs one can expect significant dipole transition rates between rovibrational ground state levels, enhanced by the large slope of the dipole moment function associated with its large average value. In the following the redistribution of population due to spontaneous decay and BBR will be modeled.

A transition between levels $i$ and $j$ with wave functions $\Psi_i$ and $\Psi_j$ is proportional to the dipole matrix element $\mu_{i,j} = \langle \Psi_i | \mu(R) | \Psi_j \rangle$, where $\mu(R)$ is the EDM function. For vibrational transitions only the variation of $\mu$ with the internuclear distance $R$ leads to a finite transition dipole moment. The EDM function was calculated ab-initio and published previously [15]. Rovibrational wave functions and overlap integrals are calculated with Level 8.0 (http://leroy.uwaterloo.ca/programs/) using a precise experimental potential energy curve [28]. The resulting permanent EDMs of low lying vibrational levels are in very good agreement with measured values [16], as presented in Sect. 3. For modeling the spontaneous decay we calculate the Einstein A coefficients, and the H"onl-London factor $S_{J', J''}$ as defined in Ref. [31]. The coupling to BBR from the surrounding walls of the vacuum chamber is given by the Einstein B coefficients for stimulated absorption and emission multiplied by the spectral density of the BBR from an environment at a given temperature $T$.

For the rovibrational ground state $X^{1}\Sigma^{+}, v''=0, J''=0$ we calculate a depopulation time constant of 65.4 s, in good agreement with previously calculated values [32, 33]. It is noteworthy that at cryogenic temperatures (77 K) this depopulation time constant is lengthened to $1.3 \times 10^4$ s. Already at room temperature rotational heating due to BBR is greatly suppressed by the low spectral density at the relevant transition frequencies. Only for $J'' > 12$ rotational heating rates become comparable to molecular trap loss rates, which are far below 0.1 s$^{-1}$.

The evolution of an initial distribution of populated ground state levels is finally given by the following rate equation for the population $n_i$ in a level $i$ (the single index $i$ enumerates rovibrational levels $v'', J''$ in energetic ordering)

$$\frac{dn_i(t)}{dt} = \sum_{j<i} A_{ij} n_j(t) - \sum_{j<i} A_{ji} n_i(t) + \sum_{j \neq i} \tilde{B}_{ij} n_j(t) - \sum_{j \neq i} \tilde{B}_{ji} n_i(t) - \Gamma n_i(t) \tag{1}$$

where $\tilde{B}_{ij} = A/(\exp(h\nu/c/kT) - 1)$ is the black-body induced redistribution rate and $\Gamma$ is a decay
Figure 2. Time evolution of the population in the lowest nine vibrational levels of the X$^1\Sigma^+$ after spontaneous decay from B$^1\Pi,v'=26,J'=1$. The population is given as a percentage of the initial total ground state population. Also shown is the population (rescaled to unity) after $t_{eq} \equiv 5 \times A_{0,1}^{-1} \sim 200$ s.

Figure 3. Simulated time evolution of the population in X$^1\Sigma^+,v''=3$. Shown are the total signal (solid line) and the population of the rotational sublevels (dashed lines, labeled by $J''$).

constant accounting for losses from the trap. The value of $\Gamma$ is taken from measurements (as discussed in section 2) and is assumed to be independent of $v''$.

In the formation of ultracold molecules by PA (PA) [17, 18], the initial population of ground state levels is determined by the Einstein A coefficients for the spontaneous decay from the excited PA state. For the B$^1\Pi$ state of LiCs we have calculated this initial population for different PA levels and found agreement with experimental observations [20]. Starting from this initial distribution the rate equation Eq. (1) is integrated numerically in order to derive the time evolution of the population, where convergence of the results is reached for time steps smaller than 50 ms. As can be seen in Fig. 2 the initial population distribution in the lowest vibrational
Figure 4. Measured rate coefficient for inelastic Cs-Cs\textsubscript{2} collisions in relation to the p- and d-wave barriers and the expected rate coefficient from capture theory and in the unitarity limit. The measured rate coefficient is found to be independent of the vibrational and rotational states.

levels of the X\textsuperscript{1}\Sigma\textsuperscript{+} state is strongly modified. Most levels show either a steady increase or decrease essentially due to vibrational redistribution with adjacent states. As an exception the population in the v\textsuperscript{″}=3 level exhibits a clear maximum after roughly 3 s, presenting a concise feature for testing the validity of the model. The time evolution of the population in this level is shown in more detail in Fig. 3 together with the individual contributions from the rotational sublevels. The significant increase of the population in v\textsuperscript{″}=3, J\textsuperscript{″}=1, being caused by the decay of molecules from v\textsuperscript{′}=4 and the cycling of population between neighboring levels due to BBR with comparable rates, indicates stronger dynamics of the process than is visible from the vibrational populations only.

Our model shows that for LiCs population redistribution caused by BBR and spontaneous decay leads to features on timescales accessible to the experiment. We have indeed found experimental evidence for population redistribution in ground state LiCs molecules, which is in good qualitative agreement with the simulation presented here.

5. Cold atom-molecule collisions
As a first step towards chemistry at ultralow temperatures, we have studied inelastic collisions between atoms and molecules simultaneously trapped in the CO\textsubscript{2} dipole trap [23]. Ultracold collisions between weakly bound, magneto-associated molecules and atoms have been studied by several groups [34, 35]. In our case, the PA process prepares the molecules in deeply bound vibrational states. Since the energy released by inelastic collisions transforming either rotation or vibrational energy into kinetic energy is much larger than the trap depth, the reaction partners are expelled from the trap with near-unity probability. There are several predictions on the rate coefficients for specific inelastic atom-molecule collisions involving alkali atoms [36, 37], not including Cs. However, as Julienne and coworkers recently pointed out [38], the rate coefficient can be derived from a universal relation depending only on the long-range properties of the atom-biatom potential, if the inelastic process takes place with unity probability at short range. This assumption is safely fulfilled in the cases studied here, and typical rate coefficients predicted by this approach are in the range 10\textsuperscript{-10} cm\textsuperscript{3}/s. Similar results have been found by the group of Pierre Pillet under equivalent conditions [39].

A measurement of the rate coefficient for these inelastic collisions proceeds in the following
way. The atoms are loaded into the optical trap and associated to molecules by illuminating the atomic cloud with the PA laser. Typical PA times are of the order of 100 ms to a second. Typically 5000 molecules are formed in a distribution of internal states which is determined by the Franck-Condon overlap of the excited rovibrational state addressed by the PA laser and the ground state. The molecules then interact with the atomic cloud for an adjustable time interval, until the number of remaining molecules is probed by REMPI with a short laser pulse. From the temporal decay of the number of trapped molecules one deduces a loss rate. By measuring the dependence of the molecular loss rate on the atomic density, one can directly determine the rate coefficient which is \( \beta = 1.5 \times 10^{-10} \text{ cm}^3/\text{s} \) as the slope of the curve. As expected from the prediction by Julienne et al., the measured rate coefficients show no dependence on the Cs\(_2\) vibrational level, even though they cover many vibrational quantum numbers. It should, however, be noted that, due to the temperature of the gas around 100 \( \mu \)K, there is a small contribution of p-wave scattering to the rate coefficient. Figure 4 shows the measured rate coefficient in relation to the p- and d-wave barrier and the expectation from capture theory as well as from the unitarity limit.

Figure 5 shows preliminary results for collision experiments between LiCs molecules and Cs atoms (this process is currently under closer investigation). From measured ion yields and a detection efficiency of 1% [20] one can estimate that around 5000 molecules are stored in the trap. This corresponds to roughly 10% of the number of trapped cesium atoms. As the trapping potentials for cesium and LiCs are nearly identical and the LiCs and Cs temperatures are very close, the molecular density is at least one order of magnitude smaller than the cesium density. This yields an upper limit of \( 5 \times 10^9 \text{ cm}^{-3} \) for the LiCs density. It is therefore not likely that molecule-molecule collisions are observable in the current experiment. From a first evaluation, we infer that the rate coefficient is of the same order of magnitude as the rate coefficient measured in the Cs-Cs\(_2\) case.
6. Conclusions
Currently, we are setting up a new experimental apparatus which will allow us to enter the regime of quantum degeneracy for the atomic gases. Based on the knowledge gained by the experiments presented here and by other groups in the field, we will develop a path towards the formation and trapping of LiCs molecules at high phase-space densities. The approach will rely upon magnetoassociation and STIRAP, eventually combined with photoassociation techniques. Due to the large dipole moment, we expect that the dynamics of the gas will be determined by the long-range dipolar interactions, allowing us to study new quantum phases and other many-body phenomena. As a second direction, we will move further towards “Chemistry without Entropy”, using electric fields and/or shaped laser fields for quantum control. One of the big issues in this direction will be the development of appropriate detectors for state-resolved product state detection, e.g. using velocity-map imaging techniques adapted to ultracold atomic and molecular gases. It is truly amazing how much progress has been made in the quest for molecular quantum gases within the last few years, with many unforeseen discoveries on this way. In view of the many intriguing perspectives of molecular quantum gases in quantum many-body physics, quantum information and quantum chemistry, even more will be discovered in the forthcoming years.

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