The permanent dipole moment of LiCs in the ground state

J. Deiglmayr† A. Grochola§ M. Repp, O. Dulieu† R. Wester‡ and M. Weidemüller§
Ruprecht-Karls-Universität Heidelberg, Physikalisches Institut, Philosophenweg 12, 69120 Heidelberg, Germany
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Recently we demonstrated the formation of ultracold polar LiCs molecules in deeply bound levels of the $X^1\Sigma^+$ ground state, including the rovibrational ground state $[5]$, after a single step of PA $[7,8]$. Here we report on the first experimental determination of the permanent electric dipole moment of deeply bound LiCs molecules. For $X^1\Sigma^+, v''=2$ and $v''=3$ we measure values of $\mu=5.5(2)$ Debye and $5.3(2)$ Debye respectively.

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Ultracold gases of dipolar molecules have been proposed as candidates for the exploration of quantum phases in dipolar gases $[1]$, the development of quantum computation techniques $[2]$, or precision measurements of fundamental constants $[3]$. LiCs is predicted to have the largest permanent electric dipole moment (EDM) of all alkali dimers $[4]$ and is thus very advantageous for such schemes. Recently we have reported the EDM of all alkali dimers $[4]$ and is thus very advantageous for such schemes. Recently we have reported the formation of LiCs molecules in deeply bound levels of the $X^1\Sigma^+$ ground state $[5]$, including the rovibrational ground state $[J. Deiglmayr et al., Phys. Rev. Lett. 101, 133004 (2008)]$. Here we report on the first experimental determination of the permanent EDM of deeply bound LiCs molecules.

The experimental sequence for formation, detection, and high-resolution spectroscopy of ground state molecules is shown in fig. 1. Ultracold LiCs molecules are formed by photoassociation (PA) $[7,8]$ 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) $[9]$ to reduce inelastic interspecies collisions. In this configuration $4 \times 10^7$ $^{133}$Cs and $10^8$ $^7$Li atoms are trapped simultaneously at densities of $3 \times 10^9$ cm$^{-3}$ and $10^{10}$ 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 $[3,6,10]$. For PA of LiCs molecules the two overlapped atom clouds are continuously illuminated by light from a tunable cw TiSa laser (typically 500 mW, collimated to 1.0 mm). The frequency of the laser is stabilized to a transfer cavity which is in turn stabilized via a frequency-offset locked diode laser $[11]$ to an atomic cesium resonance. This locking scheme results in residual shifts of less than 1 MHz for the central PA frequency. 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 mass spectrometer $[10]$.

Rotationally resolved spectroscopy of ground state molecules is achieved by depletion spectroscopy $[12]$: 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 $[5]$. The light for depumping is provided by a cw dye laser and transferred to the experimental setup by a single-mode optical fiber. Here
the collimated beam with a waist of 0.7 mm is overlapped and aligned colinear with the PA light using a selectively coated mirror. The intensity of the depumping light after the fiber is monitored using a beam sampler and a photodiode (PD). The PD signal is fed back to an electro-optical modulator in front of the fiber to compensate intensity fluctuations originating from the laser and the fiber coupling. The frequency of the cw dye laser is stabilized via a transfer cavity to an atomic cesium resonance.

Using an AOM in the locking branch of the dye laser, relative frequency shifts of few hundred MHz can be controlled with an accuracy of few MHz. In the depletion spectroscopy we reach a resolution of typically 5 MHz for shifts in transition frequencies, mostly limited by the hyperfine-induced broadening of the excited level used for depletion, and an absolute accuracy of 500 MHz, given by the resolution of the employed wavemeter (Burleigh WA-1000).

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 (ø30 mm) to allow for optical access and extraction of trapped atom clouds. The electrodes are coated mirror. The intensity of the depumping light after fiber coupling. The frequency of the cw dye laser is stabilized via a transfer cavity to an atomic cesium resonance.

For the formation of deeply bound molecules we use the photodissociation resonance B^3Π,ν′=18,J′=1. After spontaneous decay the rotational states J′′=0 of X^1Σ^+,ν′′=2 and higher are significantly populated. Then depletion spectroscopy of ν′′=2,J′′=0 and ν′′=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 ν′′=0 and ν′′=1 levels[19]. The change in the frequency of the transition X^1Σ^+,ν′′=2→B^3Π,ν′=18,J′=1 is determined via depletion spectroscopy, as shown in fig. 2a), where X^1Σ^+,ν′′=2 molecules are selectively detected via the intermediate state B^3Π,ν′=19 at a REMPI wavelength of 16 860.0 cm^{-1}. The position of the depletion resonance is obtained from a fit of two Lorentzian line profiles to the spectrum, one for the shifted resonance and one for an unshifted feature, which is introduced by switching the fields to a small value during detection of molecules.

Fitting eq. (1) to the data and using $B_{ν''=2}=0.1848$ cm^{-1} we find a permanent EDM of $μ=5.5(2)$ Debye for ν′′=2 (see fig. 2b)), where the reported error stems from the uncertainty of the fit and the estimated uncertainty on the electric field determination. The same measurement was performed for molecules in X^1Σ^+,ν′′=3,J′′=0. These molecules are detected at a REMPI frequency of 16 886.0 cm^{-1} (the intermediate level is a vibrational level in a Ω=0 state, probably associated to A^1Σ^+). Using $B_{ν''=3}=0.1836$ cm^{-1} the value for the EDM is determined as μ=5.3(2) Debye.

While this is the first experimental determination of the permanent EDM of LiCs in the ground state, few results of ab-initio calculations have been published. Igel-Mann et al. perform pseudopotential configuration interaction calculations and report a vibrational average for X^1Σ^+,ν′′=0 of 5.48 Debye[20]. Aymar and Dulieu use a similar approach with refined basis sets and obtain a value of 5.478 Debye[4]. Sørensen et al. employ a coupled cluster model with full iterative triple excitations correlating 10 electrons and report a value of 5.440 Debye[21]. Only Ref. 4 also shows vibrational averages for higher vibrational levels. Due to the coincidence of the equilibrium distance of the X^1Σ^+ state potential with a linear part of the dipole moment curve[21] the dependence of the EDM on the vibrational level is only weak. For the here considered levels ν′′=2 and ν′′=3 the values are found to be at most 1% larger than the value for ν′′=0. Therefore we would not expect a significantly different measurement for the dipole moment of the ν′′=0 ground state.
The large permanent EDM of deeply bound LiCs makes this molecule a promising candidate for the realization of an ultracold gas with strong dipolar interactions. However, a permanent EDM also couples the internal molecular state to the thermal environment via blackbody radiation (BBR) and thus leads to a finite lifetime even for molecules in the absolute ground state. As a second consequence of the existence of a permanent EDM, spontaneous decay of electronic ground state molecules into more deeply bound rovibrational levels is possible. In the case of LiCs, the timescale for such processes is expected to be in the range of few tens of seconds and should thus be observable during the lifetime of the molecules in an optical dipole trap. We will discuss the importance of these processes in a future publication.

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