Spectroscopy of the 1001 nm transition in atomic dysprosium

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We report on spectroscopy of cold dysprosium atoms on the 1001 nm transition and present measurements of the excited state lifetime which is at least 87.2(6.7) ms long. Due to the long excited state lifetime we are able to measure the excited state polarizability at 1064 nm by parametric heating to be $\alpha_e = 172(92)$ a.u., which is in fair agreement to theoretical predictions. In addition we measure the isotope shifts of the three most abundant bosonic isotopes of dysprosium on the 1001 nm transition with an accuracy better than 30 kHz.

Quantum gases of magnetic atoms enable the study of many-body physics with long-range, inhomogeneous interactions [1]. Due to their large magnetic moments especially erbium (Er) and dysprosium (Dy) experiments became more prominent in recent years and led to the realization of the extended Bose-Hubbard Hamiltonian [2], the observation of the roton mode in the excitation spectrum of a dipolar Bose Einstein condensate [3], and the discovery of self-bound quantum droplets [4,5]. In addition to its large ground state magnetic moment Dy features seven stable isotopes of which four have natural abundancies around 20%. Due to its submerged and not completely filled 4f-electron shell its energy spectrum, which is partially depicted in Fig. 1(a), is quite complex. Among the many possible transitions there are at least two candidates for ultra-narrow-linewidth ground state transitions. One at 1322 nm with a predicted lifetime of 6.9 ms and one at 1001 nm with a predicted lifetime of 3 ms [6]. In this work we investigate the latter transition.

Having an ultra-narrow-linewidth transition at hand would enrich Dy quantum gas experiments with a versatile tool. Such transitions serve as sensitive probes for interactions between the atoms for example on lattice sites of an optical lattice [7] or as probes for the external trapping potential e.g. to selectively probe different lattice sites in an optical superlattice. Furthermore, because of its long lifetime, the excited state can be used as a second species in the experiment, whose population can be precisely controlled, e.g. to study Kondo-lattice physics [8,9]. Ultra-narrow-linewidth transitions can also be used to probe the inner atomic potentials with a high sensitivity to investigate more fundamental physical questions. It has been proposed to use precise isotope shift measurements of two ultra-narrow-linewidth transitions of the same element to search for high-energy physics contributions to the inner potentials and for physics beyond the standard model [11,12]. For this purpose the element needs to have at least four (zero nuclear spin) isotopes, which is fulfilled by Dy.

Recently, the 1001 nm transition has for the first time been studied by laser spectroscopy and isotope shifts of all seven stable isotopes had been measured on the 40 MHz level [13].

The experimental results presented here demonstrate that the lifetime of the excited state of the 1001 nm transition with 87.2(6.7) ms exceeds the previous theoretical prediction [6]. In addition we refine the precision of the isotope shift measurements to the 30 kHz level for the three most abundant bosonic isotopes and present measurements of the excited state polarizability at the commonly used wavelength of 1064 nm.

I. EXPERIMENTAL SCHEME

The experimental setup is depicted in Fig. 1(b). A magneto-optical trap (MOT) operated on the $\Gamma_{626} = 2\pi \times 136$ kHz transition from the ground-state at 626 nm is loaded from a Zeeman slower, which is using the $\Gamma_{421} = 2\pi \times 32$ MHz broad 421 nm transition. Details of the experimental setup can be found in [11]. Typically, $3 \cdot 10^6$ $^{162}$Dy or $^{164}$Dy atoms or $3 \cdot 10^5$ $^{166}$Dy atoms are trapped and cooled to temperatures on the order of 20 $\mu$K. The atoms can be transferred to a single beam optical dipole trap (ODT) at 1064 nm. Absorption imaging...
is done in the horizontal plane in 45° to the ODT beam using the 421 nm transition. The spectroscopy light at 1001 nm is generated by an extended cavity diode laser (ECDL), which is stabilized by the Pound-Drever-Hall method to a cylindrical cavity made of ultra-low expansion glass (ULE). We use a fiber EOM and an offset sideband locking technique to shift the laser frequency relative to the cavity resonances, which are spaced by about 1 kHz and observe drifts of the stabilized laser frequency below 15 kHz/h. The 1001 nm light is scanned in frequency by a double pass acousto-optical modulator (dAOM). The radio frequencies driving the single pass and double pass AOMs and the fiber EOM are generated by direct digital synthesis. The circularly polarized elliptical 1001 nm spectroscopy beam is coming from top of the main chamber under a small angle to the z-axis and has beam diameters of 9.2 mm and 2.9 mm at the position of the atoms, where the broader half axis of the ellipse points along the ODT beam. From the same direction a resonant 421 nm beam can be applied to the atoms in the ODT to remove all ground state atoms from the trap. The magnetic field at the position of the MOT and the ODT is compensated during the application of the spectroscopy pulses.

II. MEASUREMENT OF THE ISOTOPE SHIFTS AND ABSOLUTE TRANSITION WAVELENGTH

The spectroscopic measurements to obtain the isotope shifts are carried out in a pulsed manner. Atoms are released from the MOT and after 6 ms time of flight (TOF) a pulse of 6 mW/cm² light at 1001 nm is applied for 12 ms with a fixed detuning to the atomic resonance. After the pulse the remaining ground state population is measured by absorption imaging and then a new detuning is set and the measurement is repeated. This way the atomic resonance appears as a dip in the ground state population like in the three exemplary spectra presented in Fig. 2. The data points in each spectrum are obtained in sequence. Since there were slow drifts of the overall atom number the background of the spectra exhibits a slope. To determine the atomic resonance frequency from each spectrum an inverted Gaussian function with a linear offset is fitted to the data points. We reference the atomic resonances of 160Dy and 162Dy to the resonance of 164Dy and conduct a 164Dy measurement immediately before and after each measurement of one of the other isotopes. This way we can detect and account for drifts of the ULE cavity, which is the optical frequency reference in our setup. As result we obtain the following isotope shifts for 160Dy in Eq. (1) and 162Dy in Eq. (2) relative to 164Dy:

$$\delta \nu_{160-164} = -2514.277(29) \text{ MHz}, \quad (1)$$

$$\delta \nu_{162-164} = -1195.773(20) \text{ MHz}. \quad (2)$$

| Error budget contribution | $\delta \nu_{160-164}$ (kHz) | $\delta \nu_{162-164}$ (kHz) |
|---------------------------|-----------------------------|-----------------------------|
| ULE cavity FSR            | 20                          | 10                          |
| ULE frequency drift       | 14                          | 6.7                         |
| RF measurement uncertainty| 10                          | 10                          |
| Fit errors                | 7.4                         | 7.8                         |
| **Total:**                | **29**                      | **20**                      |

TABLE I. Summary of error contributions taken into account for the calculation of the total uncertainty of the measured isotope shifts. RF: radio frequency.

The error budget is summarized in Table I and takes contributions from ULE cavity FSR uncertainties, ULE frequency drifts, RF measurement uncertainties and fit errors into account. By using a wavelength meter we determine the absolute frequency of the transition for 162Dy to be:

$$\nu_{162} = 299.521643(30) \text{THz} \quad (3)$$

corresponding to a wavenumber of:

$$\nu_{162} = 9990.9666(10) \text{ cm}^{-1}. \quad (4)$$

The accuracy of the Rubidium calibrated wavelength meter of 30 MHz is the dominant contribution to the measurement uncertainty by three orders of magnitude.

III. MEASUREMENT OF THE EXCITED STATE LIFETIME

The lifetime of the excited state is measured by the following sequence, which is outlined in Fig. 3(a). About 2.1 · 10⁶ 162Dy atoms are trapped in the ODT after a holding time of 800 ms. By applying a linear ramp of the laser detuning from ~300 kHz below the atomic resonance to 300 kHz above the atomic resonance in 984 µs with 4.88 Hz wide frequency steps and with a peak intensity of 450 mW/cm² about 71 % of the ground state population is transferred to the excited state of the 1001 nm transition by means of a rapid adiabatic passage (RAP). The mixture of ground and excited state atoms is trapped for a variable amount of time between 0.01 ms and 300 ms during which excited state atoms can decay back to the ground state. After this variable holding time, resonant 421 nm light is applied to remove the ground state population. Then another RAP transfers 71 % of the excited state atoms back to the ground state and subsequently the ground state atom number $N_e$ is measured by absorption imaging which is proportional to the number of excited state atoms before the second RAP pulse was applied. This sequence is repeated without applying the RAP pulses and the ground state removing 421 nm beam to obtain the total atom number $N_t$. For each holding time $N_e$ and $N_t$ are measured 15 times in turns and the resulting excitation ratios $N_e/N_t$ are averaged. The decay of the excitation ratio is plotted in Fig. 3(a). The
order in which the excitation ratio was measured for different holding times was randomized. By fitting an exponential decay function to the data we find the lifetime of the excited state to be

$$\tau_{1001} \geq 87.2(6.7)\text{ ms.}$$ (5)

This is a lower limit for the lifetime since other effects that could decrease the excited state population over time like larger trap losses for the excited state atoms compared to the ground state atoms can not be fully excluded. Compared to the theoretical prediction of $\tau_{1001,\text{th.}} = 3\text{ ms}$ this is almost a factor 30 longer than expected [6].

IV. MEASUREMENT OF THE EXCITED STATE POLARIZABILITY

Since the excited state lifetime is more than one order of magnitude larger than expected we are able to use parametric heating [16–18] of excited state atoms in the ODT to estimate the ratio of the excited state dynamic polarizability $\alpha_e$ to the ground state dynamic polarizability $\alpha_g$ at 1064 nm. For this purpose the intensity of the ODT beam is modulated for 60 ms with an amplitude of 21% (Fig. 3(b)) and modulation frequencies $f_{\text{mod}}$ ranging from 10 Hz to 4010 Hz. In the case of the excited state the sequence depicted in Fig. 3(c) is applied, where the ODT intensity modulation is switched on after a RAP transfers about 62% of the atoms to the excited state. Then the ground state atoms are removed by resonant 421 nm light before a second RAP transfers part of the excited state population to the ground state and an absorption image is taken. The parametric heating spectrum for excited state atoms is depicted in Fig. 3(c) on the top and features a resonance at $f_e = 1.05(22)\text{ kHz}$. Due to a low amount of atoms left in our atomic beam source the total ground state population is reduced to about 20·10^3 atoms in the ODT. The parametric heating resonance for ground state atoms under the same trapping conditions is measured by using the same sequence as for the excited state but without applying the RAPs and the resonant 421 nm light. The resulting spectrum is depicted in the lower half of Fig. 3(c) and it shows a resonance at $f_g = 1.09(20)\text{ kHz}$. From the resonances we obtain the ratio of the polarizabilities in analogy to [19]:

$$\frac{\alpha_e}{\alpha_g} = \left(\frac{f_e}{f_g}\right)^2 = 0.93(50).$$ (6)

Theoretical calculations lead to $\alpha_e/\alpha_g = 157\text{ a.u.}/181\text{ a.u.} = 0.870$ [20], while for the ground state

$$\alpha_g = 184.4(2.4)\text{ a.u.}$$ (7)
is experimentally determined by Ravensbergen et al. [19]. From Eq. (6) and Eq. (7) we then obtain

$$\alpha_e = 172(92) \text{ a.u.}$$

for the excited state polarizability.

V. CONCLUSIONS

We have measured the relative isotope shifts of the three most abundant bosonic isotopes of dysprosium on the 1001 nm transition with an accuracy better than 30 kHz while the absolute frequencies were determined with an uncertainty of 30 MHz. In addition, we have determined a lower boundary for the excited state lifetime which is more than one order of magnitude larger than expected from theoretical predictions [6]. The dynamical polarizability of the excited state was determined relatively to the ground state dynamical polarizability and the ratio is in fair agreement with theory [20].

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[1] T. Lahaye, C. Menotti, L. Santos, M. Lewenstein, and T. Pfau. The physics of dipolar bosonic quantum gases. Reports on Progress in Physics, 72(12):126401, 2009.
[2] S. Baier, M. J. Mark, D. Petter, K. Aikawa, L. Chomaz, Z. Cai, M. Baranov, P. Zoller, and F. Ferlaino. Extended bose-hubbard models with ultracold magnetic atoms. Science, 352(6282):201–205, 2016.
[3] L. Chomaz, R.M.W. Bijnen, D. Petter, G. Faraoni, S. Baier, J.H. Becher, M.J. Mark, F. Waeclhter, L. Santos, and F. Ferlaino. Observation of roton mode population in a dipolar quantum gas. Nature physics, 14(5):442, 2018.
[4] I. Ferrier-Barbut, H. Kadau, M. Schmitt, M. Wenzel, and T. Pfau. Observation of quantum droplets in a strongly dipolar bose gas. Physical review letters, 116(21):215301, 2016.
[5] M. Schmitt, M. Wenzel, F. Böttcher, I. Ferrier-Barbut, and T. Pfau. Self-bound droplets of a dilute magnetic quantum liquid. Nature, 539(7628):259, 2016.
[6] V.A. Dzuba and V.V. Flambaum. Theoretical study of some experimentally relevant states of dysprosium. Physical review A, 81(5):052515, 2010.
[7] F. Scazza, C. Hofrichter, M. Höfer, P.C. De Groot, I. Bloch, and S. Fölling. Observation of two-orbital spin-exchange interactions with ultracold su (n)-symmetric fermions. Nature Physics, 10(10):779, 2014.
[8] M. Foss-Feig, M. Hermele, and A.M. Rey. Probing the kondo lattice model with alkaline-earth-metal atoms. Physical Review A, 81(5):051603, 2010.
[9] L. Rieger, N. D. Oppong, M. Höfer, D.R. Fernandes, I. Bloch, and S. Fölling. Localized magnetic moments with tunable spin exchange in a gas of ultracold fermions. Physical review letters, 120(14):143601, 2018.