Optical Ramsey interferences on laser cooled and trapped atoms, detected by electron shelving

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Based on a new detection scheme optical Ramsey fringes on the magnesium intercombination transition (λ =457 nm) have been demonstrated with a resolution of 4 kHz and an accuracy of 1 Hz ($\Delta \nu / \nu \approx 2 \times 10^{-15}$) using laser cooled and trapped atoms. Applying a pulsed excitation scheme to the trapped ensemble, the Ramsey signals are nearly undisturbed by the relativistic Doppler effect and phase errors of the Ramsey zones. The detection is based on the quantum amplification due to the electron shelving effect in cooperation with the trap dynamics, monitored as decrease of the trap fluorescence induced by the fast trapping transition. Simultaneously recorded Ramsey interferences on a thermal atomic beam allowed a direct measurement of the second order Doppler shift. The relevance of the experiment to future optical frequency standards achieving a stability and an accuracy of better than 10⁻¹⁵ as well as applications of this system for atom interferometry are discussed.

1. Introduction

Besides many others, some of the basic motivations for laser cooling, laser manipulation and trapping of neutral atoms [1] are possible applications of cold atoms for ultra high resolution spectroscopy, new frequency standards [2,3] and collective quantum phenomena [4]. During the last years also the new emerging area of atom interferometry [5-11]received benefits from laser cooled atoms [8,11].

Using thermal atomic samples, optical high resolution spectroscopy is limited by the second-order Doppler effect that broadens and shifts spectral lines by an amount in the order of $\Delta \nu / \nu \approx 10^{-12}$ [12]. In addition transit time broadening drastically reduces the resolved linewidth of interesting transitions. Recently several groups demonstrated microwave spectroscopy on cold atoms in atomic fountains [13-17]. On their ballistic flight due to earth gravitation the slow atoms travel twice through a microwave region [15-17], which reduces systematic field phase errors, usually present in Ramsey spectroscopy. In the optical domain the compensation of the first order Doppler effect demands more elaborate techniques, e.g., optical Ramsey spectroscopy with three standing [18] or four running waves [19]. But for beam

experiments, laser cooled or thermal, even with time reversal of the laser beams residual phase errors are one of the main uncertainties [20].

Recently optical Ramsey interferences on the calcium 657 nm intercombination line on laser trapped atoms, detected on the direct 657 nm fluorescence, were reported [21].

In the experiment described here magneto-optically trapped atoms are directly utilized for a new spectroscopic scheme, based on a pulsed Ramsey excitation cycle, which is inherently free of phase errors. The method uses the advantage of a three-level V-system with a fast transition for trapping and cooling and a slow transition for spectroscopy, like the ${}^{1}S_{0}-{}^{1}P_{1}$, ${}^{3}P_{1}$ system in ${}^{24}Mg$.

The atoms are captured, stored and cooled on the fast ${}^{1}S_{0}{}^{-1}P_{1}$ transition ($\lambda = 285 \text{ nm}, \tau({}^{1}P_{1}) = 2.02 \text{ ns}$). To avoid light shifts due to the trap laser light the trapping beams are switched off for a short time to allow the atoms to interact with the clock transition, the intercombination line ${}^{1}S_{0}{}^{-3}P_{1}$ ($\lambda = 457 \text{ nm}, \tau({}^{3}P_{1}) = 5.1 \text{ ms}$). The sequential interaction of an atomic beam crossing four optical Ramsey zones is replaced by two pairs of counterpropagating laser pulses.

The scheme of periodically trapping and probing

is different from schemes which separate the spectroscopic process from the trap in space, e.g., in microwave spectroscopy on atomic fountains [13-17]. Atoms excited to the metastable ${}^{3}P_{1}$ state leave the trap volume, not influenced by the trap forces, when the trap laser beams are switched on again. By the subsequent decrease in the strong trap fluorescence the Ramsey excitation is easily detectable. This decrease is further amplified by the dynamics of the trapping process itself, as will be explained below. Besides providing high resolution this detection scheme opens new possibilities for investigations of cold collisions, atom trap analysis, and pulsed atom interferometry.

2. The experimental setup

The experimental setup is shown in fig. 1. It consists of two Ramsey spectrometers, one on a thermal atomic beam and a second one on a laser cooled and trapped ensemble of cold magnesium atoms. Both sections are supported by the same laser beam tuned to the $457 \text{ nm} \, {}^{1}\text{S}_{0} - {}^{3}\text{P}_{1}$ clock transition. The laser light

is generated by a frequency-stabilized stilbene 3 dye laser with a linewidth of 5 Hz relative to a stable reference cavity. The short term stability due to residual seismic influence on the reference cavity is in the order of one kHz, as deduced from the spectra on the thermal beam.

This setup allows the simultaneous recording of Ramsey fringes on a thermal atomic beam using a four-zone excitation geometry [19,20,22,23] and a laser cooled ensemble with pulsed excitation. The signals provide the correction for the reference cavity's frequency drift of about 10 Hz/s and the measurement of the second-order Doppler shift of the thermal atomic beam. An acousto optical modulator (AOM 1) switches the available power between the thermal beam apparatus and the trap. Two further AOMs (AOM 2/3) form the pulses for the trap excitation scheme, discussed in detail in a later section.

We capture and cool an ensemble of magnesium atoms in a magneto-optical trap similar in concept to that first demonstrated by Raab et al. [24]. The trap consists of a spherical magnetic quadrupole field and three perpendicular pairs of $\sigma^+-\sigma^-$ polarized laser beams. The trap can be filled either from an



Fig. 1. Overview of the experimental setup with thermal beam apparatus (left) and magneto-optical trap (MOT) apparatus (right). The acousto-optic modulators (AOM 1-3) switch between the thermal beam apparatus and the trap. The photomultiplier (PM1) detects the ${}^{3}P_{1}-{}^{1}S_{0}$ fluorescence, PM2 detects the strong ${}^{1}P_{1}-{}^{1}S_{0}$ trap fluorescence. The small inset shows the relevant part of the magnesium level scheme.

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atomic beam, decelerated to an adjustable velocity of about 50 m/s by the Zeeman tuning technique [25], or, more simply but about a factor of 100 less efficiently, from the low velocity tail of the Maxwell-Boltzmann distribution of a thermal atomic beam. The trapping beams with a diameter of 2.6 mm and intensities of 2 mW/mm² (saturation parameter S=0.45) as well as the stopping laser beam are obtained by frequency doubling the 570 nm light of two rhodamine 6G dye lasers in external cavities using ADA crystals. To make efficient use of the available uv power one single laser beam is threaded through the trap assembly, using six mirrors inside the vacuum chamber. The frequency of the trap laser is tuned 40 MHz (one half linewidth) below the ${}^{1}S_{0}$ -¹P₁ transition ($\lambda = 285$ nm). The magnetic field is provided by two coils in anti-Helmholtz configuration (radius 3 cm) which generate an adjustable gradient of up to 160 G/cm along their axis.

For magnetic field gradients of $80 \text{ G/cm} 2 \times 10^5$ atoms (with decelerated atomic beam) or 2×10^3 atoms (without beam deceleration) are stored within a diameter of 300 µm. In most of the experiments presented here the trap was filled directly from a thermal beam. The temperature of the trapped atoms was inferred from the Doppler width of the 457 nm intercombination line measured on the trapped ensemble. The temperature is a factor of 6.7 above the Doppler limit whereas with our parameters we would expect for the closed two-level system $({}^{1}S_{0}-{}^{1}P_{1})$ of ${}^{24}Mg$ a temperature of 2.4 times the Doppler limit [26] (1.9 mK). This increased temperature is probably due to a intensity imbalance between the six trapping laser beams, because of the unexpected poor reflectivity of the mirrors used to steer the uv laser beam through the trap.

3. The pulsed excitation method

The pulsed spectroscopy is performed at a repetition rate of typically 70 Hz. Each cycle (see fig. 2) is started by shutting off the trapping beams within 50 ns by an uv acousto-optic modulator (uv-AOM in fig. 2). Because of the AOM's limited extinction ratio, an additional mechanical shutter closes several μ s later to prevent any uv light from perturbing the Ramsey interferences.



Fig. 2. Time scheme for the pulsed Ramsey spectroscopy on laser trapped atoms (not to scale, see text).

Simultaneously with the trapping lasers the magnetic field configuration is switched from a quadrupole to a homogeneous field of 15 G perpendicular to the direction of the Ramsey pulses in order to shift the $\Delta m = \pm 1$ transitions out of resonance. About 50 µs later the cold atoms are irradiated by two pairs of counterpropagating laser pulses. Each of the pulses has a width of 2 µs and they are separated by a time *T* between 5 and 50 µs. Due to this Ramsey excitation a part of the atoms is shelved in the metastable ³P₁ state (τ =5.1 ms) thus leaving the trap region undisturbed by the trap laser beams, which are switched on 50 µs after the last excitation pulse. The decrease in the trap fluorescence is monitored by a photomultiplier tube and completes the cycle.

4. The Ramsey excitation

The frequency dependence of the excitation probability for a four-beam experiment was calculated by Bordé et al. [22], but the calculation performed in the atomic rest frame is actually that of a four-pulse experiment; it is therefore directly applicable to the trap arrangement. The incoherent excitation leads to the Doppler-broadened line and to the Lamb dip. The coherent part

$$P_{\mathbf{R}} = A_{+}(\Delta) \cos[2T(\Delta + \delta) + \phi]$$

+
$$A_{-}(\Delta) \cos[2T(\Delta - \delta) + \phi]$$
(1)

is the sum of two fringe patterns corresponding to

the two recoil components. Here $\Delta = \gamma \omega_{\rm L} - \omega_0$ denotes the detuning between the laser frequency $\omega_{\rm L}$ and the atomic frequency ω_0 corrected for the relativistic second order Doppler effect $\gamma = (1 - v^2/c^2)^{1/2}$, $\delta = \hbar k^2/2M$ denotes the recoil shift, with the wavevector k of the laser beams and the atomic mass M. The phase ϕ is the phase difference between the phases $\phi_1 - \phi_4$ of the four laser pulses

$$\phi = \phi_2 - \phi_1 + \phi_4 - \phi_3, \tag{2}$$

and the factors A_+ and A_- are weakly frequency-dependent amplitude factors taking into account the single pulse excitation probability.

One of the most important features of the pulsed scheme is its inherent phase compensation. As in our experiment the pulses are simply cut from a cw beam, the phase difference of eq. (2) exactly cancels to zero. Mechanical vibration of the optical elements on the beam paths during the relevant spectroscopic times T lead to phase fluctuations of the exciting pulses, that reduce the fringe contrast without frequency shift. Pulsing the AOM for 2 µs increases its temperature by 2 µK, which amounts to a negligible systematic phase shift of 2×10^{-6} fringe periods. This thermal effect was checked experimentally with an interferometric setup, using longer pulses.

In contrast to experiments on a thermal beam, where the width of the fringe pattern is given by the contributing velocity distribution or the longitudinal coherence length of the atomic wave respectively, the width in the trap experiment is only limited by the frequency-dependence of the single-pulse excitation probability leading to the factors A_+ and A_- , which are only slowly dependent on the detuning (typically 500 kHz for the experimental parameters given above). Therefore a wide range of Ramsey fringes filling the complete Lamb dip is visible.

5. The signal amplification due to loss accumulation

Excited atoms, shelved in the metastable ${}^{3}P_{1}$ state, moving with the mean trap velocity, fly a distance of about 5 mm before they return to the ground state by spontaneous emission. Those atoms are not recaptured and leave the trap region. This is different from the quantum amplification due to electron shelving used in ion traps [27], where the excited atoms remain trapped. Therefore the Ramsey excitation constitutes an additional loss mechanism for the trap. As in our trap with its low density the number of stored atoms is not limited by the trap density but mainly by collisions with the background gas, this additional loss will, after reaching a new steady state, lead to a lower equilibrium in the number of trapped atoms and hence to a decrease in the trap fluorescence.

The resulting number of trapped atoms N is given by

$$N = N_0 \frac{1}{1 + P\tau_0/t_{\rm C}}.$$
 (3)

Here N_0 denotes the number of atoms without Ramsey excitation, t_C the duration of one complete cycle (fig. 2) and τ_0 the undisturbed lifetime of the trap. *P* denotes the excitation probability for one excitation cycle. Typical values in the experiment described here are: $N_0=2\times10^3$, $P\approx0.01$, $t_C=1/70$ s, $\tau_0=1$ s. With these parameters we obtain an amplification factor $\tau_0/t_C=70$ that allowed us to detect a 1% excitation probability as a 40% decrease in trap fluorescence. Because of this strong amplification and the efficient detection of the electron shelving via the fast trap transition, we were able to detect Ramsey fringes with integration times of two seconds per frequency point even with only two thousand trapped atoms.

Compared to schemes which separate trapping and spectroscopy in space this method offers the advantage of accumulating the loss of many cycles and of retrapping atoms, which have not taken part in the spectroscopic process, for the next cycle.

6. Results and discussion

Figure 3 shows the Doppler broadened line profile with the central Lamb dip measured on the trapped atoms. In this measurement only one pulse of each direction has been applied to the atomic sample. The Doppler width of 8.8 MHz corresponds to a mean velocity of 2.1 m/s of the trapped atoms, which is a factor 1.7 above the expected velocity for a two-level atom. This example demonstrates the powerful tool, offered by the spectroscopy on that narrow transiVolume 103, number 1,2



Fig. 3. Fluorescence of trapped atoms when irradiated by two counterpropagating 457 nm laser pulses, showing the central part of the Doppler profile with the Lamb dip.

tion, for temperature measurements and trap analysis.

The Lamb dip is broadened due to the spectral width of each pulse. The experimental pulse width of $2 \mu s$ is an optimum, determined by the available laser power. Increasing the pulse length leads to an increasing excitation probability but reduces the number of contributing velocity classes because of the smaller homogeneous width of longer pulses.

Typical high resolution scans of Ramsey fringes are shown in fig. 4. The pulse separations T of $6 \,\mu s$ (top), $24 \,\mu s$ (second curve), and $44 \,\mu s$ (third curve) lead to fringe periodicities of 80 kHz, 20 kHz, and 12 kHz, respectively. The simultaneously recorded Ramsey fringes on the thermal atomic beam (T=700 K, lowest curve, fwhm: 4 kHz) clearly show the relativistic red shift due to the second order Doppler effect, measured in a series of scans to (-1.5 ± 0.4) kHz. The error of 400 Hz is mainly determined by phase uncertainties of the thermal signal. From a numerical simulation of the signals taken with our experimental conditions we obtain a theoretical value of (-1.5 ± 0.3) kHz, where the error is due to the strong dependence of the signal on the contributing velocity distribution [28].

On the trap, the second order Doppler effect of the



Fig. 4. Ramsey fringes on trapped atoms (upper three curves) for different pulse separations in comparison with a signal from the thermal beam apparatus (lowest curve). The incoherent Doppler- and Lamb-dip background have been subtracted. The dots are measured data, the solid curves show numerical simulations.

residual velocity spread of 2 m/s will negligibly contribute to a broadening of 2×10^{-17} . Also the phase error which amounts to 400 Hz in the thermal beam experiment contributes to less than 1 Hz in the trap experiment. Residual magnetic and electric fields shift the transition frequency only in second order and contribute in both cases to less than 1 Hz. Thus the uncertainty of 500 Hz on the atomic beam experiment is reduced in the trap experiment to the Hzlevel ($\Delta \nu / \nu \approx 2 \times 10^{-15}$).

The experimental resolution of 4 kHz is at the moment mainly limited by residual vibrations and fluctuations of the trap apparatus. With a more elaborate setup, a resolution of the order of the natural linewidth of 31 Hz seems to be feasible. The magnesium atom furthermore offers the ${}^{1}S_{0}{}^{-3}P_{2}$ magnetic quadrupole transition with a lifetime of the upper state of 5500 s [29], thus offering resolutions in the sub-Hz regime with $\Delta \nu / \nu \approx 10^{-17}$.

Besides spectroscopic applications the trap setup offers many opportunities for atom interferometry [23,30]. The pulsed interferometer described here is best suited for the study of phase shifts due to time dependent interactions, like the scalar AharonovBohm effect [31,32] or the Berry phase [33]. The physical mechanisms inducing these small phase shifts (additional electric, magnetic, and/or laser fields) can easily be introduced in the time between the Ramsey pulses [23].

7. Conclusion

Ultrahigh spectroscopy of an optical transition has been performed on laser cooled and trapped neutral atoms. The detection scheme utilizing the electron shelving and a related amplification (atom recycling) leads to a strong increase in the signal to noise ratio and a strongly improved detection sensitivity, which can be used for many other purposes. Because of the low atomic velocities, the influence of the second order Doppler effect can be neglected and the excitation scheme is inherently free of phase errors. Other systematic errors are reduced to the Hz level. This experimental setup with its advantages compared to conventional spectroscopy is a further step towards improved future optical frequency standards based on laser manipulated particles.

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