AN OPTICAL FREQUENCY STANDARD WITH ULTRACOLD Ca-ATOMS

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Optical frequency standards utilizing narrow atomic transitions with $Q \approx 10^{12}$ and low sensitivities to external fields offer the possibility to reach relative uncertainties below 10^{-15}. Using ensembles of more than10^7 neutral alkaline earth atoms laser cooled in a magneto-optical trap, excellent short time stabilities below 10^{-16} in 1 s seem possible. In our optical Ca-frequency standard with an ensemble Doppler cooled to a temperature of 3 mK we have realized a relative uncertainty of $2{\cdot}10^{-14}$ limited mainly by a residual Doppler effect of the atomic motion and by the effect of cold collisions. With a new quench cooling method we were able to reduce the temperature to below 10 µK. First measurements showed the potential for a reduced uncertainty and an improved stability in utilizing these ultracold atoms in the frequency standard.

optical frequency standard, Doppler effect, ultracold atoms, state-selective detection method

1. INTRODUCTION

The realization of frequency standards with Cs-fountains has reached a relative uncertainty level of $1{\cdot}10^{-15}$. Here, the line center has to be resolved to within 10^{-5} of the linewidth, which is achieved by averaging over measurement times of more than 10^4 s (Ref. 1).
In optical frequency standards the clock transitions provide frequencies higher by 4 orders of magnitude. Resolutions close to the natural linewidths, which are in the range of 320 Hz for Ca atoms at 456 THz (Ref. 2) down to below 10 Hz for $^{199}Hg^+$ ions at 1 PHz (Ref. 3) have been achieved. At the same time these transitions provide very low sensitivities to external electric and magnetic fields. Two different concepts are used.
Either single ions are trapped in an electrodynamic potential and additionally laser cooled. In this case the ion is confined in a volume of around 1 µm diameter and can be kept up to years and probed for extended periods of time. Frequency measurements with a relative uncertainty of $1{\cdot}10^{-14}$ for $^{171}Yb^+$ (Ref. 4) and $^{199}Hg^+$ (Ref. 5) have been demonstrated.
Neutral-atom standards based on alkaline earth atoms like Mg, Sr and Ca use Doppler cooling and trapping in a magneto-optical trap (MOT) to prepare ensembles of about 10^7 atoms in a volume with 1 mm radius reaching temperatures in the one millikelvin range (Refs 6,7). With such high numbers of atoms very high signal-to-noise ratios (*SNR*) can be reached in a single measurement cycle. A stability of $4{\cdot}10^{-15}$ has been demonstrated for Ca by Oates et al. (Ref. 2).
In neutral-atom standards the uncertainty contributions are dominated by a residual Doppler effect due to the cloud of atoms freely expanding in non-perfect wave fronts of the spectroscopy laser beams and by frequency shifts due to cold collisions.
In this paper we present the status and future prospect of our Ca-frequency standard. By careful investigation and control of the Doppler effect and cold collisions we have reached a relative uncertainty of $2{\cdot}10^{-14}$.
With a new quench cooling method (Ref. 8) we were able to reduce the velocity distribution of the laser cooled atoms. We will present first measurements that show the potential to reduce the uncertainty of the Ca-frequency standard by more than an order of magnitude and to improve the stability using a novel detection scheme to the theoretical limit given by the quantum projection noise (QPN) (Ref. 9) of $4{\cdot}10^{-17}$.

2. CA-FREQUENCY STANDARD WITH COLD ATOMS

2.1. Experimental set-up

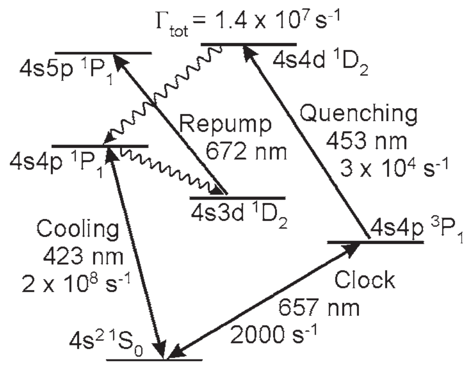

Fig. 1: Excerpt from the energy level scheme of ^{40}Ca.

An effusive beam of Ca-atoms from a Ca-oven at 900 K is directed into a MOT. On the cooling transition at 423 nm (Fig. 1) atoms from the low velocity part of the atomic velocity distribution are slowed down, trapped and cooled without use of a Zeeman-slower. In 15 ms about 10^7 atoms are collected in a diameter of 1.6 mm at a temperature of 3 mK which is close to the Doppler limit for the cooling transition. This corresponds to a width of the velocity distribution of v_{rms} = 75 cm/s and a density of a few times 10^9 atoms/cm^3.
Within 200 µs the trap is switched off and a well-defined magnetic Helmholtz-field of 0.2 mT is applied to split the Zeeman-components ($\Delta m_J = \pm 1$) of the $^1S_0$-$^3P_1$ transition from the ($\Delta m_J = 0$)-clock transition. Then the clock transition is probed at the freely expanding ensemble with linearly polarized light using atom interferometers of the Ramsey-Bordé type (Ref. 10) in the time domain. A 4-pulse time sequence of two anti-parallel pairs of laser pulses is applied. Regarding the atoms as matter waves the pulses serve as 50% beam splitters (Rabi angle of $\pi/2$) and split the matter waves coherently into partial waves being in the ground or excited states. The partial waves follow different paths defined by the photon recoil.

After redirection of the partial waves with the intermediate pulses the final pulse leads to a recombination, so that the geometry can be considered as a Mach-Zehnder-interferometer for matter waves. The interferometric signal is given by the excitation probability for each atom and is detected using the photons emitted during the decay of the excited state. It oscillates with the frequency detuning of the laser beams from the clock frequency and is centered around the clock transition (*Ramsey-Bordé-fringes*, Fig. 2). The resolution $\Delta \approx 1/(4T)$ can be set by the pulse distance T. Values between 1.2 kHz and 600 Hz close to the natural linewidth are chosen for the best stability while the Doppler-broadened linewidth of the ensemble is more than 2 MHz. For the frequency standard the spectroscopy laser is stabilized to the central fringe.

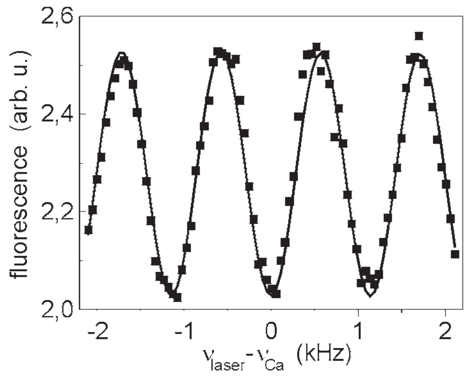

Fig. 2 Interferogram of a Ramsey-Bordé-interferometer. Fluorescence of the excited Ca-atoms as function of laser detuning.

2.2. Residual Doppler effect

One special feature of the atom interferometer is that in each of the four interactions zones i the instantaneous phases Φ_i of the laser pulses are added to the wave functions of the atoms. This results in a phase shift $\Delta\Phi = \Phi_2 - \Phi_1 + \Phi_4 - \Phi_3$ of the Ramsey-Bordé-fringes and may lead to a systematic frequency shift.
The main contribution to this shift is given by a residual Doppler effect. Due to the atomic movement during interferometry through the spatial wave fronts of the spectroscopic laser beams (Fig. 3), the atoms see a different spatial phase in each interaction zone (Ref. 11). If the wave fronts are perfectly plain ($R_l = \infty$) and the anti-parallel k-vectors $\mathbf{k}_l$ (laser beam direction l = 1,2) of the beams are perpendicular to gravitational acceleration $\mathbf{g}$ ($\alpha_l = \pi/2$) then $\Delta\Phi = 0$. Otherwise, there is a frequency shift given by $\Delta\nu = \Delta\Phi/4\pi T$. To first order $\Delta\nu$ depends linearly on the time between switching of the trap and the pulse i and on $A_l = (kg{\cdot}\cos\alpha_l + kv^2/R_l)$. v is given by the velocity distribution in the plane perpendicular to $\mathbf{k}_l$ and $k = |\mathbf{k}_l|$. Additionally, a time independent offset as well as higher order contributions have to be taken into account (Tab. 1). Even for carefully pre-aligned laser beams ($|R_l|$ > 40 m,
$|\alpha_l - \pi/2|$ < 5 mrad) total relative frequency shifts up to $3{\cdot}10^{-14}$ can occur.
To evaluate and correct for these frequency shifts we apply a method based on a proposal by Trebst et al. (Ref. 11).

For this purpose we measure the phase shifts in two purely phase dependent atom interferometers as a function of pulse distance. Each interferometer uses three parallel pulses from one of the two Ramsey-Bordé interferometer directions l with equal pulse distances T.

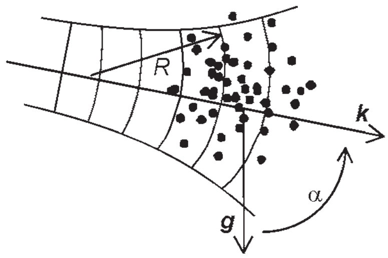

Fig. 3 Atoms moving in curved and tilted wave fronts.

The coefficient of the phase shift $\propto T^2$ is again given by A_l. Hence, they can be explicitly deduced from measurements. Together with v known from Doppler-spectra in the three spatial dimensions upper limits for the offsets and residuals of the frequency shifts can be calculated and the linear frequency shift with pulse distance can be corrected for with a relative uncertainty of $1{\cdot}10^{-14}$.

2.3. Uncertainty budget

Besides the Doppler effect we have investigated several systematic influences contributing to the overall uncertainty budget of the Ca-frequency standard. This includes the effect of changes in free-running laser drift, the effect of black body radiation from the hot Ca-oven and the 2nd order Zeeman effect. We eliminated a possible AC-Stark shift due to residual cooling laser light by using a synchronized mechanical chopper. By varying the density of the trapped atoms we measured the relative frequency shift due to cold collisions between the atoms to $(3 \pm 4.4){\cdot}10^{-24}$ cm^3. This allowed us to reduce the relative uncertainty contribution of this effect by a factor of 5 to $4{\cdot}10^{-15}$.

contribution (in Hz)	**correction**	**uncertainty**
2nd order Zeeman	-2.8	0.1
technical	0	3.2
blackbody oven (900 K)	2.1	4.3
blackb. environ. (300 K)	1	0.05
DC-Stark	0.04	0.02
1st order Doppler		
residuals/offsets	0	2.4
1.2 kHz resolution	1.5	1.4
spurious phase chirps	0	4
collisions ($1.3{\cdot}10^{15}$/m^3)	-1.2	1.8
total	**0.6**	**7.5**

Tab. 1 Uncertainty budget achieved for the PTB's Ca optical frequency standard with cold atoms.

Tab. 1 summarizes the individual uncertainty contributions. We have measured the clock frequency using a frequency comb generator for the comparison with the frequency of the Cs-fountain of the PTB to (455 986 240 494 151 ± 9) Hz with a relative uncertainty of $2{\cdot}10^{-14}$.

The measurements indicated another spurious phase shifting effect leading to a frequency shift dependent on the pulse distance in the range of 10^{-14}. We attribute this effect to phase chirps in the switching AOMs for the spectroscopy beams due to the amplitude modulation of the RF-pulses. Such an effect has been observed also by Udem et al. (Ref. 5).

3. UTILIZING ULTRACOLD ATOMS

With a new cooling scheme for alkaline earth atoms realized in our laboratory we were able to reduce the width of the velocity distribution by a factor of 15. We performed first atom interferometric investigations with ultracold atoms that show their high potential to substantially improve the uncertainty as well as the stability for optical frequency standards.

3.1. Quench cooling method

The cooling method is a purely optical quench cooling scheme using the clock transition in a MOT configuration allowing us to cool the Ca-atoms far below the Doppler limit for the cooling transition down to temperatures below 10 µK (Ref. 8). The cooling force reached with the low scattering rate can hardly hold the atoms against gravity. Therefore, a quench laser at 453 nm is used to quench the $^3P_1$-state by transferring the excited atoms into the 4*s*4*d* $^1D_2$-state (Fig. 1) from where they decay quickly to the ground state and can take part in another cooling cycle. Thus, the cooling force was increased by a factor of 15. To prevent the atoms from escaping from resonance on the narrow transition due to the recoil shift the 657 nm cooling laser is broadened to 1.4 MHz (broadband cooling). The scheme is applied as a 2nd stage cooling method directly after the regular cooling cycle to 3 mK. For a typical 2nd stage cooling duration of 20 ms we already managed to transfer 12 % of the atoms into an ultracold ensemble with a velocity distribution with v_{rms} < 5 cm/s.

3.2. Improvement of the contrast

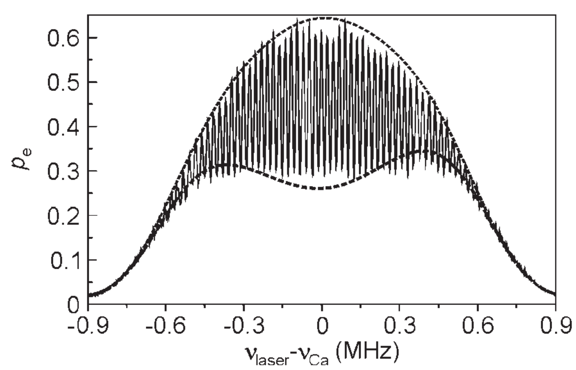

Fig. 4 Ramsey-Bordé interferogram with ultracold atoms. The dashed line shows the envelope of a spectrum for a v_{rms} = 7.5 cm/s calculated with a spinor approach according to Ref. 10. The solid line shows the scaled measured fluorescence of the decay of excited atoms. Resolution set to 11.6 kHz, τ = 1.1 µs.

To reach maximum contrast in atom interferometry it is necessary to have equal excitation probabilities of 50 % for all atoms in the cooled ensemble. This can only be achieved when the Fourier-width of each pulse ($\propto 1/\tau$) is substantially broader than the width of the Doppler-broadened spectrum of the clock transition. For a $\pi/2$-pulse the product of intensity I in the laser beam and τ^2 has to be constant. The laser power available, therefore, limits the Fourier width to about 1 MHz ($\tau \approx$ 1.1 µs). For the 3 mK ensemble this was more than two times below the Doppler-width leading to a contrast of not more than 0.2. The ultracold ensemble has a Doppler-width that is below 200 kHz. Fig. 4 shows a Ramsey-Bordé interferogram for an ensemble of ultracold atoms. The envelope for the ultracold atoms is given by the Fourier transform of the rectangular pulses applied. In contrast to the spectrum of the 3 mK ensemble, the velocity distribution gives only minor contributions to the line shape. The excitation probability and contrast are increased close to the optimum. The measured contrast of 0.36 is already 70 % of the maximum value achievable for atoms at rest (v_{rms} = 0).

3.3. State-selective detection method

The Ramsey-fringes shown in Fig. 2 and Fig. 4 were taken by detecting photons from the decay of the excited state. The aperture of the detection set up and the quantum efficiency limits the overall detection efficiency to a few times 10^{-3} giving a high contribution of the photon shot noise to the overall noise. Additionally, atom number fluctuations between the measurement cycles further reduce the SNR and, therefore, the stability achievable.
To overcome these problems and get closer to the quantum projection noise (QPN) limit of the atoms, methods using *electron shelving* (Refs 6,12) can be utilized.
The strong fluorescence of the broad cooling transition allows to reduce the photon shot noise level below the limit given by the QPN.
Using the slow expansion of the ultracold atoms we modified the method to detect the ground and the excited state atoms within one measurement cycle after spectroscopy.
Directly after spectroscopy the ground state atoms are probed with a 50 µs pulse of resonant cooling laser light and their fluorescence is measured. At the same time the atoms are accelerated. After waiting for one to two lifetimes of the excited clock state, the ground state atoms have left the detection region and the excited atoms have decayed to the ground state. To speed up the decay the quenching laser is switched on after the first detection pulse. A second pulse then probes the formerly excited atoms. With the two signals a normalized excitation probability for the atoms can be measured. The method eliminates the influence of atom number fluctuations, avoids heating of the ensemble before spectroscopy and should allow to reach the QPN-limit for the stability of $4 \cdot 10^{-17}$ in 1 s for 40 measurement cycles in 1 s. In preliminary measurements with no optimization an improvement of the SNR by a factor of 6 for a resolution of 1.2 kHz has been achieved already. Here the stability was limited by the lack of intensity stabilization of the detection laser and a high background together with contributions from the spectroscopy laser noise.

3.4. Reduced Doppler effect

The reduced width of the velocity distribution of the ultracold atoms leads to a substantial reduction in sensitivity to curved wave fronts. Fig. 5 shows a comparison of the residual Doppler effect for cold and ultracold atoms for an intentionally misaligned laser beam. For the cold atoms (a)) the phase shift in the three pulse interferometer is dominated by the contribution of kv^2/R, so that the contribution by $kg \cdot \cos \alpha$ is not visible any more. For the ultracold atoms (b)) only the gravitational contribution shows up while the velocity dependent shifts are reduced by more than two orders of magnitude.

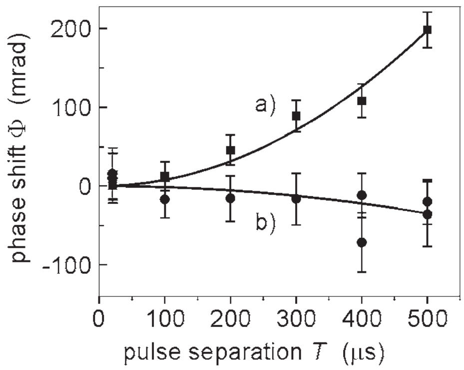

Fig. 5 Phase shift $\Delta\Phi$ in an three pulse interferometer with parallel pulses as function of the pulse distance T for a wave front curvature of $R = (12 \pm 5)$ m and $\alpha = (1.6 \pm 1)$ mrad. a) $v_{rms} \approx 1.1$ m/s,
b) ultracold atoms with $v_{rms} \approx 7.5$ cm/s.

3.5. Uncertainty budget for ultracold atoms

From these results we have estimated an uncertainty budget for the application of ultracold atoms in our Ca optical frequency standard summarized in Tab. 2.

contribution (in mHz)	correction	uncertainty
technical	0	100
2^{nd} order Zeeman	-2560	80
blackbody envir. (300 K)	970	50
DC-Stark	35	20
1^{st} order Doppler		
offsets	0	35
linear in T (resol. 400 Hz)	0	135
higher order in T	0	50
asymmetry of line shape	0	13
collisions ($1.3 \cdot 10^{15}/m^3$)	-1800	260
total	**-3355**	**330**

Tab. 2 Uncertainty budget estimated for a best case assumption of the utilization of ultracold Ca atoms for the optical frequency standard.

For this purpose we have assumed a number of $3 \cdot 10^7$ atoms. An optimized alignment regarding velocity distribution (v_{rms} = 3.5 cm/s) and laser wave fronts (adjusted to $\boldsymbol{k}_l$ within 6 µrad vertical to gravity and R_l > 300 m) is assumed that could be achieved, for instance, with the help of a mercury mirror and pentaprism and the application of three pulse interferometers with cold and ultracold atoms (compare Fig. 5). In this way a reduction of the Doppler-effect by a factor of 17 seems possible.

A substantially increased stability of the standard should allow systematic measurements of density dependent frequency shifts only limited by the accuracy of the density measurements leading to an uncertainty reduced by a factor of 7.
Hence, a total relative uncertainty of an optical frequency standard with ultracold Ca atoms of $8 \cdot 10^{-16}$ seems possible. With the stability we aim at, this could be realized in less than one second.

4. CONCLUSIONS AND OUTLOOK

In conclusion we have shown the present status of our Ca optical frequency standard with cold atoms that has reached a total relative uncertainty of $2 \cdot 10^{-14}$ making the clock transition of Ca one of the most accurately known optical transition frequencies. We have presented first measurements with ultracold atoms showing the potential for an optical frequency standard with ultracold neutral Ca atoms that can reach an accuracy better than 10^{-15} and an unprecedented stability of $4 \cdot 10^{-17}$ in 1 s. This will make the standard competitive in accuracy to the best existing microwave standards and superior in stability to single ion optical clocks.

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