An Optical Clock with Ultracold Neutral Atoms

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We demonstrate how to realize an optical clock with neutral atoms that is competitive to the currently best single ion optical clocks in accuracy and superior in stability. Using ultracold atoms in a Cs optical frequency standard we show how to reduce the relative uncertainty to below \(10^{-15}\). We observed atom interferences for stabilization of the laser to the clock transition with a visibility of 0.36, which is 70% of the ultimate limit achievable with atoms at rest. A novel scheme was applied to detect these atom interferences with the prospect to reach the quantum projection noise limit at an exceptional low instability of \(4 \times 10^{-17}\) in 1 s.

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Progress of science and technology has been closely connected to the development of sophisticated methods to measure time and frequency. From the highly developed pendulum clocks of around 1900 to today’s most advanced Cs atomic clocks, realizing the SI time unit, the increase in relative accuracy from \(10^{-7}\) to \(10^{-15}\) was mainly made possible by an increase in the operational frequency from the Hz to the GHz range. With optical comb generators based on femtosecond lasers \([1]\) now optical frequencies can be connected to radio frequencies with an uncertainty below \(10^{-18}\) \([3]\), enabling optical clocks at frequencies that are five orders of magnitude higher than the frequency of the Cs transition.

With this ‘clockwork’ available the performance of an optical clock primarily depends on the ‘pendulum’, i.e. on the atomic or molecular transition, and equally important on the method that is used to interrogate the ‘pendulum’ with the least noise while keeping all disturbances at a minimum. In an optical clock this corresponds to probing the transition at the best possible signal-to-noise ratio (SNR) and stabilizing the laser to the true undisturbed line center.

The achieved uncertainties of optical clocks based on single ions of \(\lesssim 10^{-14}\) (Hg \([2]\), Yb \([2]\)) and on large numbers of neutral absorbers of \(\gtrsim 10^{-15}\) (H \([2]\), Cs \([6]\)) are still about an order of magnitude worse than the best Cs atomic clocks \([3]\). In contrast to the single ion standards, where the absorber is confined to a small volume, the neutral atom standards are ultimately limited by the residual velocity of the absorbers, but, on the other hand, benefit from the higher signal to noise ratio due to the large number of atoms which may lead to exceptionally low instabilities.

In this letter we show on the example of the calcium standard how the use of ultracold atoms makes neutral atom optical frequency standards competitive in accuracy to the best microwave standards and improves the stability to unprecedented levels.

For the interrogation of optical transitions of laser-cooled atoms generally a separated field excitation \([4]\) in the time domain \([4]\) with two consecutive pairs of laser pulses from opposite directions is used. Such a pulsed excitation scheme represents an atom interferometer with laser pulses acting as beam splitters in analogy to an optical Mach-Zehnder interferometer \([2]\).

The excitation sequence leads to a cosine-shaped signal where the argument of the cosine is given by the phase \(\Phi = 4\pi T_{\text{rep}}(\nu_{\text{laser}} - \nu_{\text{Ca}}) + (\phi_2 - \phi_1) + (\phi_4 - \phi_3)\) which depends on the time \(T_{\text{rep}}\) between the pulses in each pulse pair, the laser detuning \(\nu_{\text{laser}} - \nu_{\text{Ca}}\) and the laser phases \(\phi_i\) in the i-th interaction. These phases appear in this atom-light interferometer because the phase of each beamsplitting laser pulse is transferred to the atomic partial wave. For perfect alignment these phase differences cancel, however, net phase differences remain when atoms of the cloud move between the pulses to a position where the local phase of the laser is different, e.g. due to curved wavefronts or tilted laser beams \([4]\).

To achieve optimum visibility of the resulting interference fringes, 50% splitters are necessary. For resonant excitation this requires \(\pi/2\)-pulses, defined by the Rabi angle \(\Omega_R \propto \tau = \pi/2\) with the Rabi frequency \(\Omega_R \propto \sqrt{\gamma}\). In particular for narrow linewidth \(\gamma\), as needed in a frequency standard, the Rabi frequency at the available laser intensity \(I\) is rather low, setting a lower limit on the pulse duration \(\tau\) which is \(\tau \approx 1\ \mu s\) in our experiment. For non-resonant excitation, the excitation probability is diminished with a half width equal to the Fourier width of the single pulse. Due to the Doppler effect this translates to an acceptance range of atomic velocities, that effectively contribute to the interference signal. For our setup this acceptance range is in the order of 15 cm/s. For broader velocity distributions only a part of the atomic ensemble contributes to the atom interferences, thereby reducing the contrast and the SNR.

In addition, the velocity dependent excitation probability influences the amplitude and the incoherent background of the observed signal. This background may shift the central interference fringe from the undisturbed line center. To correct for this shift and to recover the undisturbed center the signal has to be calculated by an integration over the actual velocity distribution, which be-
comes less reliable as the velocity spread becomes wider than the acceptance range.

We realize a neutral atom optical frequency standard by stabilizing a laser ($\lambda = 657$ nm) to the intercombination transition $^1S_0-^3P_1$ of $^{40}\text{Ca}$ (fig. 1) which has a natural linewidth of $\gamma/2\pi = 320$ Hz. Like other alkaline earth elements, $^{40}\text{Ca}$ has a $^1S_0$ ground state that is little sensitive to external fields, making it an ideal candidate for an optical frequency standard, and a nearly closed dipole transition $^1S_0-^1P_1$ for effective laser cooling. About $10^7$ atoms at a temperature of a few millikelvin are prepared by means of a magneto-optical trap (MOT). The corresponding velocity width of this cold ensemble previously limited the uncertainty of the Ca standard to $2 \times 10^{-14}$ [13] as a result of the influences mentioned above.

To overcome this limitation we applied a new scheme for Doppler-cooling on the narrow intercombination transition [16] leading to ultracold atoms with a theoretical limit for calcium of 30 mK [17]. The small scattering force achievable on that transition of only 1.5 times the gravitational force was increased by quenching the $^3P_1$ state via the $^3P_1-^1D_2$ transition (fig. 1). With this cooling scheme the atoms can be trapped and the temperature is reduced by more than two orders of magnitude to below $T = 10 \mu$K.

The interrogation is performed with a ballistically expanding ensemble of ultracold Ca atoms released from the MOT, where the interference pattern is measured by detecting the fluorescence emitted in the decay of the $^3P_1$ state after the last pulse of the atom interferometric sequence. Now, the frequency width of the whole signal is given by the Fourier width of the exciting pulses (fig. 2) which is dramatically different from the signal of cold atoms ($T = 2.7$ mK, inset of fig. 2) where the width is given by the Doppler width. Therefore nearly all atoms contribute to the interference signal, leading to an observed contrast of 0.36 that is 70% of the optimum contrast achievable with atoms at rest. The measured interference pattern is in good agreement with calculations based on a spinor approach [18]. The small deviations from theory can be attributed to slow variations of atom number during the long scanning time of 40 min.

Because the Fourier width of the exciting pulses is much better controllable than the velocity distribution of the atoms, theoretical calculations for the ultracold atoms will now allow us to estimate systematic shifts of the interference pattern with reduced uncertainty. From the experience with the Cs atomic clock [19] where an accuracy of $10^{-5}$ of the fringe width is realized in part due to the good theoretical knowledge of the line profile, it may now be possible to stabilize a laser to an optical transition with similar accuracy.

The SNR of the detection scheme used so far is limited by fluctuations in the atom number and by the shot noise of the detected fluorescence photons, because only one photon per excited atom is emitted and from those only a small fraction can be detected. The ultimate stability $\sigma_\nu$ of a frequency standard is given by the quantum projection noise limit to $\sigma_\nu \propto \sqrt{N}$ [20] where $N$ is the number of atoms contributing to the signal. To reach this limit a detection probability close to unity is necessary. It can be obtained by detecting ground state atoms via their resonance fluorescence when cycled on the cooling transition in the singlet system, which gives a high number of photons per atom. However, such electron shelving detection schemes [19] for optical frequency standards with cold atoms have suffered from atom number fluctuations [20] or from heating of the atoms when a normalisation

\[ \Gamma_{\text{tot}} = 1.4 \times 10^7 \text{s}^{-1} \]

\[ \text{Repubm transition 672 nm} \]

\[ \text{Quench transition 452.7 nm} \]

\[ 4.09 \times 10^8 \text{s}^{-1} \]

\[ \text{Clock transition, 2nd stage cooling 657.5 nm} \]

\[ 2000 \text{s}^{-1} \]
was applied [1]. The novel scheme presented here measures both the ground and the excited state atoms after the interferometry. Immediately after the last pulse of the atom interferometric sequence the number of atoms in the ground state is measured by cycling on the \(^1S_0\)\(^1P_1\) transition by a resonant single laser beam and detection of the fluorescence. By the radiation pressure the atoms are accelerated and should leave the interaction region. After one to two natural lifetimes of the \(^3P_1\) state this measurement is repeated, giving a measure for the number of atoms that have been in the excited state after the interferometry. In addition to waiting for the spontaneous decay also a pulse on the quenching transition was applied. From both values a normalized excitation probability independent of atom number fluctuations can be derived. Fig. 3 shows interference fringes with a period of 2.3 kHz that are detected with the new state selective scheme. The SNR is increased by a factor of 6 compared to the SNR reached with the previous detection method, mainly limited by amplitude noise of the detection laser.

This scheme is particularly suited for ultracold atoms due to the low average velocity and the slow expansion of the atomic cloud by less than 50 \(\mu\)m, while for a cold ensemble, part of the excited atoms would have left the detection region before decaying to the ground state. Our new simple scheme avoids any heating of the ensemble before the spectroscopy and allows the elimination of atom number fluctuations. Hence, no basic limitation exists any longer that prevents one from reaching the quantum projection noise limit. For typical values of our apparatus a stability of \(4 \times 10^{-17}\) in only 1 s seems possible, which would be better than that of any existing optical and microwave frequency standard.

At the same time the utilization of ultracold atoms leads to a considerably increased accuracy. The main contribution to the uncertainty of the Ca frequency standard arises from residual first order Doppler effects due to wavefront imperfections of the exciting laser beams. E.g., in a symmetric three-pulse atom interferometer with equal pulse separation \(T_{sep}\) the movement of the atoms through the wavefronts (radius of curvature \(R\)) of the exciting laser beam results in a phase shift

\[
\Phi = A T_{sep}^2 \text{ with } A = \vec{k} \cdot \vec{g} + \vec{k} \cdot \vec{v}_L^2 / R,
\]

where \(\vec{g}\) denotes the gravitational acceleration and \(v_L\) the velocity component perpendicular to the wavevector of the exciting laser pulse \(\vec{k}\). For an intentionally misaligned beam \((R \approx 12 \mu m)\) this phase shift in dependence of the time between the pulses is shown in fig. 4 for a Doppler-cooled ensemble \((T = 2.8 \mu K)\) and for a quench-cooled ensemble \((T = 14 \mu K)\). Because of the reduction of \(v_L^2\) by a factor of more than 200, the second term in the above equation becomes negligible and the remaining phase shift can be fully explained by the slight deviation from horizontal alignment of the laser beam \((\vec{k} \cdot \vec{g} \neq 0)\) while for the Doppler-cooled ensemble this effect is hidden by the much stronger influence of the wavefront curvature. In a four-pulse interferometer these phase shifts lead to an frequency error of \(\Delta \nu = (A_1 (2T_1 + T) + A_2 (2T_1 + 3T))/8\pi\) where \(A_1\) and \(A_2\) denote the factor \(A\) in eq. 1 for the direction of the first and the second pair of pulses and \(T_1\) denotes the time between releasing the atoms from the MOT and the first interrogation pulse. Antiparallel directions \((\vec{k}_1 = -\vec{k}_2)\) are assumed as this can be achieved.
to a very high degree by optical means. A measurement of the phase shifts in two 3-pulse interferometers therefore can be used to correct this frequency offset to a large amount [4, 5]. Extensive investigations show, that the low velocity of the ultracold atoms combined with an optimized alignment of the exciting laser beams will reduce the contribution of velocity-dependent effects to the uncertainty of the Cs optical frequency standard from 4 Hz to only 150 mHz [5].

At this level other contributions like collisions between the atoms will play the dominant role. From our present measurements at $T \approx 3$ mK we estimate the coefficient of the density-dependent frequency shift $\alpha = (3.0 \pm 4.4) \times 10^{-30}$ m$^{-3}$ [5] which is more than a factor of 200 smaller than in the Cs microwave clock and still smaller than in the Rb microwave clock. With the accuracy and stability now at hand using ultracold atoms this dependence can be measured more accurately and then be used to correct the clock frequency accordingly. Altogether, the expected total relative uncertainty will be $8 \times 10^{-16}$.

In conclusion, we showed how the neutral atom optical frequency standard can reach an expected uncertainty of below $10^{-15}$ and a quantum projection noise limited stability of $4 \times 10^{-17}$ in only 1 s, which will make it competitive in accuracy and superior in stability to existing microwave standards and single ion optical clocks. Besides the use as an optical clock the sensitivity, the high signal-to-noise ratio and the reliable theoretical modeling of the atom interferences can be used for various sensor applications.

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