Towards a Mg Lattice Clock: Observation of the $^1S_0$-$^3P_0$ Transition and Determination of the Magic Wavelength


Phys. Rev. Lett. 115, 240801 — Published 9 December 2015

DOI: 10.1103/PhysRevLett.115.240801
Towards a Mg lattice clock: Observation of the $^1S_0^−^3P_0$ transition and determination of the magic wavelength


1 Institut für Quantenoptik, Leibniz Universität Hannover, Welfengarten 1, 30167 Hannover, Germany
2 Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802, USA

M. S. Safronova, U. I. Safronova, S. G. Porsev

1 Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA
2 Joint Quantum Institute, NIST and the University of Maryland, College Park, Maryland 20899, USA
3 Department of Physics, University of Nevada, Reno, Nevada, 89557, USA
4 Petersburg Nuclear Physics Institute, Gatchina 188300, Russia

(Dated: November 5, 2015)

We optically excite the electronic state $3s3p\ ^3P_0$ in $^{24}$Mg atoms, laser-cooled and trapped in a magic-wavelength lattice. An applied magnetic field enhances the coupling of the light to the otherwise strictly forbidden transition. We determine the magic wavelength, the quadratic magnetic Zeeman shift and the transition frequency to be $468.46(21)$ nm, $−206.6(2.0)$ MHz/T$^2$ and $655.058$ × $10^9$ Hz/T$^2$, respectively. These are compared with theoretical predictions and results from complementary experiments. We also developed a high-precision relativistic structure model for magnesium, give an improved theoretical value for the blackbody radiation shift and discuss a clock based on bosonic magnesium.

The frequencies of optical clocks are currently measured with a fractional accuracy and precision of nearly $10^{-18}$ [1–4]. A potentially limiting systematic frequency shift of both ion and optical lattice clocks is the AC Stark shift from room-temperature black body radiation (BBR) [5, 6]. Clock transitions with small BBR sensitivities are an attractive approach to even higher accuracies. Among these are neutral Hg and Mg, In+*, and especially the A1+ ion clock transition, which all have significantly smaller BBR sensitivities than Sr and Yb lattice clocks and Cs microwave clocks.

In this Letter we report the spectroscopy of the Mg clock transition in a magic wavelength optical lattice, which gives equal AC Stark shifts of the clock states. We measure the transition frequency [7, 8] and its magic wavelength and quadratic Zeeman shift, which were recently predicted [9–12].

Along with our measurements, we developed a more refined atomic structure model to calculate both the magic wavelength as well as the static BBR shift. For less massive atoms, such as Mg, these models are more accurate than for heavier elements like Sr and Yb, and spectroscopy of low-mass elements generally represents an interesting testbed for validating improved theoretical models [13]. Both our theoretical and experimental results for the magic wavelength agree at a level of better than 1% and restrict the value, which was estimated to fall in the range between 466 and 480 nm [9–11]. Our model for Mg predicts a static BBR shift to be eight and five times lower than those of Sr and Yb, which were recently measured [14, 15]. Apart from the static contribution, the total BBR shift also includes a dynamic contribution, which is derived from the combination of theoretical calculations and measurements of $^3D_1$ state lifetime [5, 16]. Ref. [17] estimated the dynamic contribution in Mg to be 0.1 a.u. for the $^3P_0$ state, being considerably smaller than for Yb (1.86 a.u.) and Sr (12.37 a.u.).

For bosonic atoms, optical dipole excitation of the electronic ground state $^1S_0^−^3P_0$ is strongly suppressed. A magnetic field enhances the dipole coupling, enabling nHz linewidth by mixing the $^3P_1$ electronic state [12, 18]. Refs. [12, 19] calculated the associated second order Zeeman effect for Mg to be $−217(11)$ MHz/T$^2$ (equivalent to a fractional frequency shift of $−3.31(17)\times 10^{-7}$/T$^2$), a systematic effect that must be evaluated. We precisely measured the magnetic field dependence, which is consistent within the uncertainty of [12], estimated to be 5% [19]. This second-order Zeeman shift is larger than those of Yb ($−6.6(4)$ MHz/T$^2$ or $−1.27(8)\times 10^{-8}$/T$^2$ [18]) and Sr ($−23.5(2)$ MHz/T$^2$ or $−5.47(47)\times 10^{-8}$/T$^2$ [20]).

In this way we directly measure the transition frequency, which agrees with the difference of the measured frequencies of the $^1S_0^−^3P_1$ and $^3P_0^−^3P_1$ transitions [7, 8]. Due to its low mass and the short magic wavelength, Mg has a large photon recoil frequency shift, $\Delta\nu_R = h/(\lambda_{\text{magic}}m_{\text{Mg}})$, where $\lambda_{\text{magic}}$ is the magic wavelength, as well as greater tunneling. A deeper lattice is therefore required to suppress tunneling [21], as compared to heavier species.

We briefly summarize the steps required for the optical lattice spectroscopy in Fig. 1. A thermal beam of Mg atoms is slowed and loaded into a “singlet”-magneto-optical trap (MOT) using laser light tuned near the $^1S_0^−^1P_1$ transition. Atoms are optically transferred, after excitation to the $^3P_1$ state, into a second, simulta-
Afterwards, a spatially-overlapped 1D optical lattice with procedure reproducibly generates about 1000 atoms at ear intensity variations is performed within 100 ms. This a lower depth and subsequently increased to a final value fore the dipole trap is rapidly switched off. To select the coldest atoms, the optical lattice intensity is ramped to nearly 10^3 cold atoms are permitted to accumulate in 3P_0 in the ODT. These atoms are optically depumped to the ground state via 3P_1 and the magic wavelength optical lattice is adiabatically turned on. The dipole trap and the optical lattice laser beams are depicted by the bold arrows. The 458 nm light interro-gates the magnetic field enhanced clock transition.

FIG. 1. Optical transitions in ²⁴Mg relevant for performing optical lattice spectroscopy. Atoms are continuously loaded in the long lived electronic state ¹P_0 in an optical dipole trap (ODT) at 1064 nm using a dual magneto-optical trap (MOT) [22]. Atoms trapped in a MOT using the ¹S_0−¹P_1 transition are optically transferred with 457 nm to the ³P_1 state and then to ³P_0. The atoms are further cooled in a MOT with 383 nm light that excites the ³D-manifold, and cold atoms are permitted to accumulate in ³P_0 in the ODT. These atoms are optically pumped to the ground state via ³P_1 and the magic wavelength optical lattice is adiabatically turned on. The dipole trap and the optical lattice laser beams are depicted by the bold arrows. The 458 nm light interro-

gates the magnetic field enhanced clock transition.

controller mirror. An additional feedback loop driving an acousto-optical modulator (AOM) can set and stabilize the depth of the lattice. The light transmitted through the cavity is used to measure the circulating light intensity in the cavity.

The clock transition spectroscopy is performed with a home-built external cavity diode laser stabilized to an ultra-stable resonator with finesse F = 600,000 at 916 nm, similar to [25]. The infra-red light is fiber-guided to the spectroscopy setup, a tapered amplifier chip, and a commercial second-harmonic generation (SHG) stage. The system generates 10 mW of 458 nm light with a short-term frequency instability as low as 5 × 10^-16 in 1 s, inferred with the help of a second ultra-stable res-
onator. The spectroscopy is performed by irradiating the atoms for 100 ms with a pulsed, Gaussian shaped laser beam with a waist of 300 µm and a peak intensity of 7 W/cm². The MOT coils, operated in Helmhotz config-
ura tion, generate a magnetic field of 2.49(1) G/A, deter-

mined via optical Zeeman spectroscopy of the ¹S_0(m_J = 0)−³P_1(m_J = ±1) transitions, increasing the dipole coupling of ¹S_0 and ³P_0. We normally use a magnetic field of 249 G, which yields a predicted linewidth of 8.07 µHz and a Rabi frequency of 205 Hz [12]. In this way, we resonantly excite up to a thousand atoms to ³P_0, which are then optically pumped to ³P_3 and detected with 80 ms of fluorescence from the “triplet”-MOT. This detection scheme yields a sensitivity of a few tens of atoms. To obtain the line centre and profile of the transition, we record the number of excited atoms as we step the frequency of the 458 nm laser. Using the atoms, the linear drift of the laser is determined at the beginning and end of the spectroscopic measurements. The initial drift drift of the laser is determined at the beginning and end of the spectroscopic measurements. The initial drift is compensated with a feed-forward of an AOM that shifts the laser frequency to a resonance of our ultrastable cav-
ity. A scan over the resonance typically comprises 30 measurements, each lasting 1.9 s.

The magic wavelength for ²⁴Mg is inferred from measure-
ments of the line centre for different lattice depths and several wavelengths. Figure 2 a) shows two sets of measurements of the transition probability (red and blue dots) versus clock laser frequency and correspond-
ing Gaussian fits (red and blue solid curves) for three depths of a 466.9 nm lattice. The line profiles for different trap depths were measured successively. To evaluate and correct the residual laser drift, the measurement sequence was repeated three times and the shift of the line centers for a specific trap depth is determined from the Gaussian fits. From the frequencies for a specific lattice depth, we infer the residual clock laser drift, which can be as large as 2-3 kHz within several minutes. The line profiles in Figure 2 a) are three superposed scans. The linewidth of each profile, on the order of a few kHz, is mostly due to tunneling in our shallow optical lattice. According to [21], the total line broadening is on the or-
der of twice the ground state band width. This is in

We generate 160 mW of lattice light near the magic wavelength at λ_magic = 469 nm with a frequency-doubled Ti:Sa laser. A horizontal build-up cavity, with a finesse of 80, twines around the vacuum chamber and, with a circulating power of ∼2.3 W, produces trap depths of 10 recoil energies hνR. The cavity length is stabilized to the frequency of the laser with a Pound-Drever-Hall [24] lock driving an electro-optical modulator (EOM) and a piezo-
circulating power of ∼80, twines around the vacuum chamber and, with a circulating power of 2.3 W, produces trap depths of 10 recoil energies hνR. The cavity length is stabilized to the frequency of the laser with a Pound-Drever-Hall [24] lock driving an electro-optical modulator (EOM) and a piezo-
agreement with our findings. In our current apparatus, the trap depth was as low as 6 recoil energies, which gives rise to a carrier width of approximately 16 kHz. Figure 2 b) shows the line centers (dots) and the corresponding linear regression (solid lines) of the AC Stark shifts versus lattice depth for different lattice wavelengths. An offset frequency was subtracted from the linear regressions for each lattice wavelength to account for the laser drift between measurements. The uncertainty of the experimental determination of the lattice depth is about 5% dominated by the uncertainty of the lattice waist. The uncertainty of the AC Stark shift is a combination of the statistical uncertainty of the linear regression and the systematic uncertainty of residual (non-linear) frequency drifts of the clock laser, on the order of a few kHz. During the measurements for 466.97 nm, the clock laser was less stable, leading to more noise in the number of detected atoms, larger error bars, and a larger variation of the observed AC Stark shifts. The differences in the linear regression from the two measurement campaigns agree within these uncertainties. Separately, the two data sets yield magic wavelengths of 468.47(22) nm (blue data) and 468.45(19) nm (red data). Applying a linear regression to the combination of both measurement sets, we determine the magic wavelength of the 468.46(21) nm and the linear AC Stark shift dependence on lattice depth and wavelength to be 1.67(12) kHz/ER/\text{nm} (equivalent to a fractional frequency shift of 2.55(18) × 10^{-12}/ER/\text{nm}).

The experimentally determined magic wavelength agrees well with our theoretical prediction. We use a state-of-the-art relativistic approach that combines configuration interaction and all-order linearized coupled-cluster methods (CI+all-order). To evaluate the uncertainty of our calculations, we use a combination of the CI and second-order many body perturbation theory (CI+MBPT), which does not include all-order corrections to the effective Hamiltonian. The difference of the CI+MBPT and CI+all-order values serves as an estimate of the theoretical accuracy [26–28]. The results are summarized in Table I. Our final recommended value for the theoretical Mg magic wavelength, listed in the “Final” row, uses measured instead of calculated transition energies for the dominant contributions. While our calculated Mg transition energies agree with the observed values to a few cm^{-1}, even these small differences affect the magic wavelength in the fourth significant figure. We note that this correction is quite small and the final value differs from the \textit{ab initio} CI+all-order value by only 0.05%.

In Table I, we give CI+MBPT and CI+all-order values for the magic wavelength \lambda_{\text{magic}}. We also give the static ground state \alpha(n{s}^{2}\ ^{1}S_{0}) and excited clock state \alpha(n{s}np\ ^{3}P_{0}) polarizabilities, and their difference \Delta\alpha, which is proportional to the static BBR shift [17].

To demonstrate the extremely high accuracy of the theoretical calculations in Mg, we compare the magic wavelength and polarizabilities of Mg, Sr, and Yb in Table I. In our theoretical method, the main difference between Mg, Sr, and Yb are the much larger and more complicated cores of Sr and Yb than of Mg. As a result, core-valence correlations are much smaller in Mg than in Sr and Yb, leading to substantially higher accuracy for the theoretical predictions for Mg.
CI+MBPT and CI+all-order Sr and Yb values illustrate the significance of higher-order effects in these heavier systems. We note that the difference of the CI+MBPT and CI+all-order values for Sr and Yb is much larger than the difference of the CI+all-order results with experiment for both magic wavelength and $\Delta\alpha$, confirming the validity of using such difference as an uncertainty estimate of the Mg theoretical values. The excellent agreement of the CI+MBPT and CI+all-order polarizabilities indicates an uncertainty of the Mg BBR shift of less than 1%.

The second order Zeeman shift drops out of the determination of the magic wavelength but is a significant correction to our measured transition frequency. Fig. 3 shows the measured transition frequency versus applied magnetic field (squares), a parabolic fit (black curve) of the measurements, and a theoretical prediction by [12] (red curve). The predicted dependence on the magnetic field is -217(11) MHz/T², which agrees with the experimental result of -206.6(2.0) MHz/T².

![FIG. 3. Quadratic Zeeman shift of the clock transition versus magnetic field strength (black squares), a parabolic fit (black curve), and the theoretical prediction of [12] (red curve). The predicted dependence on the magnetic field is -217(11) MHz/T², which agrees with the experimental result of -206.6(2.0) MHz/T².](image)

Our measurements at the magic wavelength, with the correction of the second-order Zeeman shift, yield a direct measurement of the optical transition frequency of 655 058 646 691(101) kHz. The absolute frequency is measured by beating the spectroscopy laser with an optical frequency comb that is stabilized to a 10 MHz GPS frequency reference. The measured transition frequency agrees with the difference of previous spectroscopic measurements of the $^1S_0-^3P_1$ and $^3P_0-^3P_1$ transitions of 655 659 923 839 730(48) Hz and 601 277 157 870.0(0.1) Hz to better than 100 kHz, which corresponds to the estimated uncertainty of this measurement [7, 8].

In summary, we report the direct optical spectroscopy of the $^1S_0-^3P_0$ clock transition of laser cooled bosonic $^{24}$Mg in a magic-wavelength optical lattice. Our measurements determine precisely the magic wavelength and confirm the high precision obtained with a new theoretical atomic model of Mg. Our experimental determination of the quadratic Zeeman effect and clock transition frequency agree with a prediction [12] and previous indirect frequency measurements. Lattices with a larger depth of more than 40 recoil energies, accessible with higher laser power and a higher finesse of the enhancement cavity, will display a reduced width of the lowest vibrational band to 20 Hz and thus allow more precise spectroscopic measurements.

The demonstrated agreement of our combination of theory and experimental measurements is an important ingredient for exploring a future bosonic and fermionic Mg optical lattice clock. For bosonic magnesium, atoms can be optically prepared at μK temperatures, which has not yet been demonstrated for the fermionic isotope $^{25}$Mg. In our experiment, a dilute atomic cloud of 1,000 atoms is distributed over 130,000 lattice sites (~0.008 atoms per lattice site), which is a factor of 100 lower density than that reported for other clocks with approximately the same number of atoms [31, 32], significantly reducing the limitations from collisional shifts. The quadratic Zeeman shift can be sufficiently controlled [33] using the suitably narrow Mg transitions for Zeeman spectroscopy, a higher clock laser intensity, and a smaller...
A clock laser intensity of 7 W/cm² yields a Rabi frequency of 20 Hz, with a magnetic field that is 10 times smaller and a corresponding reduction in the uncertainty of the quadratic Zeeman shift. Here, in the context of Sr, Yb and Hg, Mg can use higher clock laser intensities because the clock and magic wavelengths are nearly equal, leading to small differential polarizabilities of the Mg clock states and a naturally smaller clock-laser AC Stark shift. Combining all of these techniques can exploit the small Mg sensitivity to blackbody radiation to make a highly accurate and stable lattice clock and further precisely test atomic models for precision spectroscopy.

We thank A. Bauch and H. Schnatz from Physikalisch Technische Bundesanstalt (PTB) for providing a passive H-maser and assistance for the frequency measurement during the early stage of the experiment as well as C. Lisdat for helpful discussions.

This work was supported in part by Deutsche Forschungsgemeinschaft within QUEST, Center for Quantum Engineering and Space-Time Research (APK, SR, SS, WE and EMR), NASA (KG) and U. S. NSF Grant Nos. PHY-1404156 (MSS and SGP) and PHY-1311570 (KG). DF and KHZ acknowledge financial support from Research Training Group RTG1729. NJ is supported by the Marie Curie Initial Training Networks (ITN) Program, call: FP7-PEOPLE-2013-ITN.

[33] Other clock spectroscopy schemes can suppress it, as has been explored in [34–36].