Laser Sources for Precision Spectroscopy on Atomic Strontium

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ABSTRACT

We present a new laser setup suited for high precision spectroscopy on atomic strontium. The source is used for an absolute frequency measurement of the visible $5s^2\,{}^1S_0$-$5s\,5p^3P_1$ intercombination line of strontium which is considered a possible candidate for a future optical frequency standard. The optical frequency is measured with an optical comb generator referenced to the SI through a GPS signal. We developed also an all solid state blue laser source that will be used for laser cooling of strontium, which will result in a better control on the systematic effects and a great improvement in the precision of the measurement.

Keywords: Laser frequency stabilization, atomic strontium, laser spectroscopy, spectroscopic techniques, visible and ultraviolet spectra, time and frequency.

The accurate measurement of frequency is far less fraught with systematic error effects than is a corresponding measurement in wavelength space; e.g. wavefront curvature and diffraction effects don't bias frequency measurements. The accuracy limit to absolute wavelength measurements is of the order of a part in 10^{10}. While simple, commercial frequency counters can perform at a part in 10^{12} in a one second measurement and laboratory measurements in excess of a part in 10^{18} are rather routinely made. For this reason, the development of the optical-frequency comb generators[1,2] was a real quantum leap in the potential of precision optical spectroscopy as it made possible, for the first time, relatively easy direct optical-frequency measurements. Applied to frequency metrology, optical transitions have potential for greatly improved accuracy and stability relative to conventional atomic clocks based on microwave frequency transitions. While it is clear that optical clocks represent the future of time and frequency metrology,[3] many groups are searching for the best candidate of a future frequency reference in terms of performances and experimental requirements.[4] Sr has long been considered one of the most interesting candidates[5] among the neutral atoms.

The Strontium intercombination lines (5^1S-5^3P) from the ground state are in the visible and easily accessible with semiconductor lasers (fig.1). Depending on the specific fine-structure component and on the isotope - Sr has four natural isotopes, three bosonic, ^{88}Sr (82%), ^{86}Sr (10%), ^{84}Sr (0.5%) with nuclear spin I=0 and one fermionic, ^{87}Sr (7%) with I=9/2 - a wide choice of transitions with different natural linewidths is possible. These span from the 7.5 kHz linewidth of the 5^1S_0-5^3P_1 line, which is the subject of this work, down to the highly forbidden 5^1S_0-$5^3P_{0,2}$ transitions. In ^{87}Sr, the presence of hyperfine mixing has allowed the observation of the 0-0 transition which is expected to have a natural width of only 1 mHz.[6]

In this paper we present the laser sources suited for the realization of a frequency standard referenced on the visible $5s^2\,{}^1S_0$-$5s\,5p^3P_1$ intercombination line of atomic strontium and we demonstrate the potentiality of this transition by measuring its frequency with a simple and compact apparatus improving by 4 orders of magnitude the accuracy with respect to the best wavelength measurement.[7]

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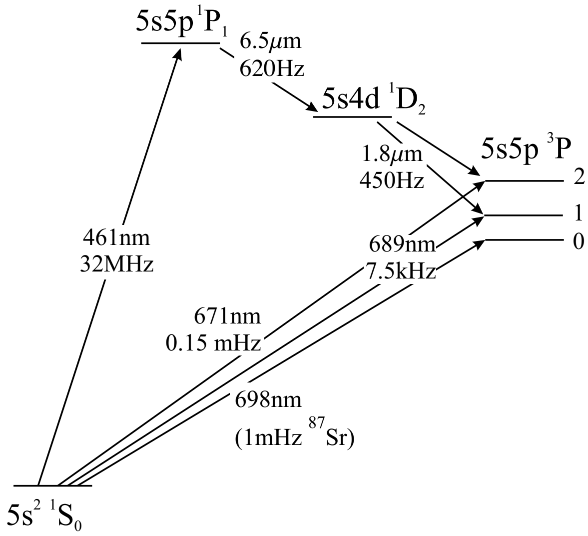

Figure 1. Relevant energy levels and transition linewidth for high resolution spectroscopy and atomic manipulation of bosonic strontium.

Section 1 describes the red diode laser including the frequency narrowing and stabilization against the atomic reference. The measurement of the optical transition under locking conditions is presented in section 2. In section 3 we illustrate a generalization of the experimental setup used to measure the ^{88}Sr-^{86}Sr isotopic shift on the intercombination line with an accuracy of the order of one part in 10^6, and we conclude presenting an all solid state blue laser source that will be used for laser cooling and trapping of atomic strontium (section 4).

1. EXPERIMENTAL SETUP

Our experimental setup is composed of a laser-diode frequency-locked to an optical cavity whose modes are locked to keep the laser on resonance with the atomic line. The optical frequency is measured with a self-referenced optical-comb stabilized against a GPS controlled quartz reference oscillator. A scheme of the experimental setup is given in fig. 2. The extended cavity laser-diode (ECDL) is a Hitachi HL6738MG mounted in the Littrow configuration which delivers typically 15 mW. Optical feedback to the ECDL is prevented by a 40 dB optical isolator and a single pass acusto-optic modulator in cascade. The laser linewidth is reduced by locking the laser to an optical reference cavity (RC) with the classic Pound-Drever-Hall scheme[8]; the phase modulation is produced by a resonant electro-optic modulator (EOM) driven at 21 MHz and leaves 85% of the power in the carrier . To avoid residual standing wave in the EOM, which induces spurious AM on the locking signal, a 25 dB optical isolator is placed between the EOM and the cavity. The reference cavity has a free spectral range (FSR) of 1.5 GHz and a finesse of 10000. On one side of the quartz spacer we glued a concave mirror (R=50 cm) while on the other side a piezoelectric transducer (PZT) is glued between the spacer and a flat mirror in order to steer the modes of the cavity by more than one FSR.

The lock of the laser onto the cavity includes a low frequency loop acting on the PZT of the ECDL (1 kHz bandwidth), and a high frequency loop acting on the laser-diode current supply (1 MHz bandwidth). Under lock condition more than 55 % of the incident light is transmitted through the cavity. From the noise spectra of the locking signal and by comparison with another cavity we can infer a laser linewidth less than 2 kHz, and more than 90 % of the optical power in the carrier.[9] We do not passively stabilize the RC in a vacuum chamber[10] since the acoustic and sub acoustic noise is sufficiently reduced by the servo to the atomic signal which acts on the PZT of the RC with a 200 Hz bandwidth.

The strontium atomic beam is obtained from the metal heated to 830 K in an oven using a bundle of stainless steel capillaries for collimation.[11] The capillary geometry insures a residual atomic beam divergency of 25 mrad with typical atomic density in the detection region of 10^8 cm^{-3}. The vacuum system is pumped by a 20 l/s ion

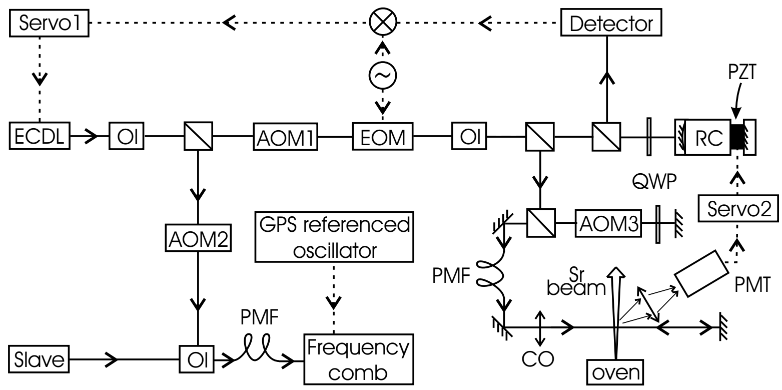

Figure 2. Experimental setup used for the frequency measurement on the Sr intercombination line. Optical isolators (OI) and acusto-optic modulators (AOM) eliminate feedback among the master laser (ECDL), the slave laser, the electro-optic modulator (EOM) and the reference cavity (RC). Solid lines represent the optical path, dashed lines represent electrical connections. QWP: quarter wave-plate. CO: collimation optics. PMF: polarization maintaining fiber. PMT: photo multiplier tube.

pump; the windows towards the detection region are anti-reflection coated on both sides and they are sealed using a modified copper gasket.[12]

The Doppler-free atomic line is resolved by saturation spectroscopy using two counterpropagating laser beams perpendicular to the atomic beam. The fluorescence light from the laser excited atoms (fig. 3) is detected on a photomultiplier tube with an overall efficiency of 0.4 %. Orthogonality between atomic and laser beams is optimized by centering the Lamb dip with respect to the residual Doppler profile.

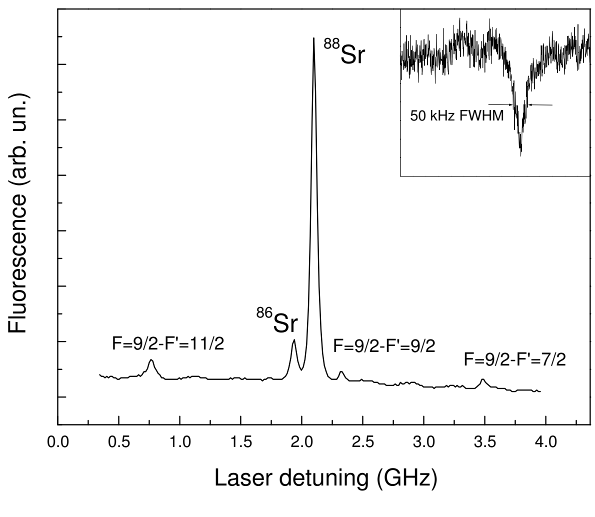

Figure 3. Fluorescence spectrum of the strontium $^1S_0$-$^3P_1$ line at 689 nm. The lines of the two bosonic isotopes ^{86}Sr and ^{88}Sr, together with the hyperfine structure of the fermionic ^{87}Sr, can be resolved. The linewidth corresponds to the residual 1st order Doppler broadening in the thermal beam. Inset: sub-Doppler resonance of ^{88}Sr recorded by saturation spectroscopy using two counter-propagating laser beams. The amplitude of the dip is 10 % of the Doppler signal.

The laser beam is spatially filtered using a single mode fiber and it is collimated at a $1/e^2$ diameter of 14 mm (wavefront distortion less than $\lambda/6$); the beam is retro-reflected using a mirror at a distance of 65 mm from

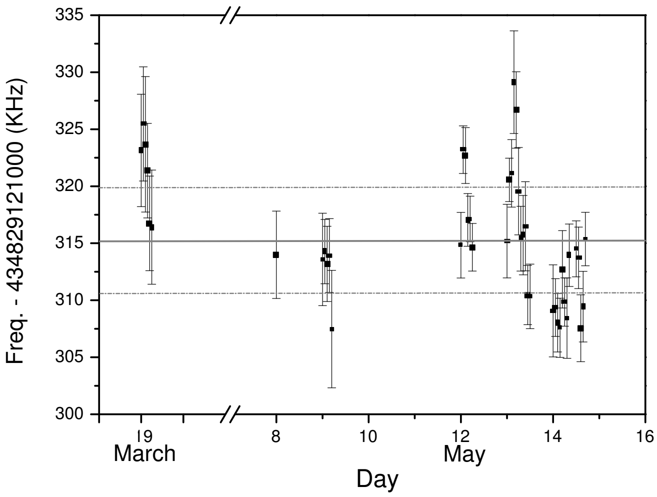

Figure 4. Measurements used to determine the transition frequency. The error bars correspond to the standard deviation for each data set.

the interaction region and coupled back into the fiber. We estimate the indetermination on the angle of the retroreflected beam to be less than 10 μrad maximizing the transmitted power through the fiber. The peak beam intensity of 60 μWcm^{-2} (to be compared to the saturation intensity of 3 μWcm^{-2}) was chosen to obtain sufficient signal to noise for the RC lock onto the atomic resonance. A uniform magnetic field of 10 Gauss defines the quantization axis in the interrogation region such that the light is π polarized.

The acusto-optic modulators between the ECDL and the EOM (AOM1) and between the ECDL and the slave laser (AOM2) are driven from the same oscillator and both deliver the -1 order such that the frequency instability and indetermination of their driving RF does not affect the optical frequency measurement. The double pass AOM next to the atomic detection (AOM3) is frequency modulated at 10 kHz to derive the locking signal of the cavity onto the atomic line, and its RF is counted against the same GPS clock used to reference the frequency comb. Figure 3 shows the Doppler broadened resonances of ^{88}Sr, ^{86}Sr and part of the hyperfine structure of ^{87}Sr. The residual atomic beam divergency produces a Doppler broadening of 60 MHz FWHM. In the inset, the sub-Doppler signal for ^{88}Sr is shown. Two independent measurements[13] of the sub-Doppler resonance show a FWHM of about 50 kHz, which is in agreement with the expected value considering the saturation, transit time broadening, and the recoil splitting.

2. OPTICAL FREQUENCY MEASUREMENTS

We measure the optical frequency through a commercial optical frequency comb generator[14] based on a Kerr-lens mode-locked Ti:Sa laser with a repetition rate of 1GHz, which is spectrally broadened in a microstructured fiber. Stability and accuracy of the comb generator are established by referencing the repetition and carrier offset envelope frequencies to a GPS stabilized local oscillator (LO). The light is delivered form the Sr setup to the frequency comb generator through a 20 m long polarization maintaining optical fiber. Figure 4 shows the result of the measurement of the ^{88}Sr transition frequency taken over a period of several days. Each data point corresponds to the averaging of the values resulting from consecutive measurements taken with a 1 s integration time over 100-200 s. The error bars correspond to the standard deviation for each data set. The Allan deviation of each set shows a flicker floor varying between 1 and 2 KHz in the region from 1 to 100 seconds (see fig. 5).

Analysis of potential systematic errors (see table 1) indicate that we should have no uncontrolled bias of more than 5 kHz. We have evaluated 1st and 2nd order Doppler and Zeeman effects, AC Stark shift, collisional shifts and recoil effects. The 1st order Doppler shift, resulting from imperfect alignment in our standing wave, is evaluated at 7 kHz by comparing consecutive measurements with independent alignments. The shift was

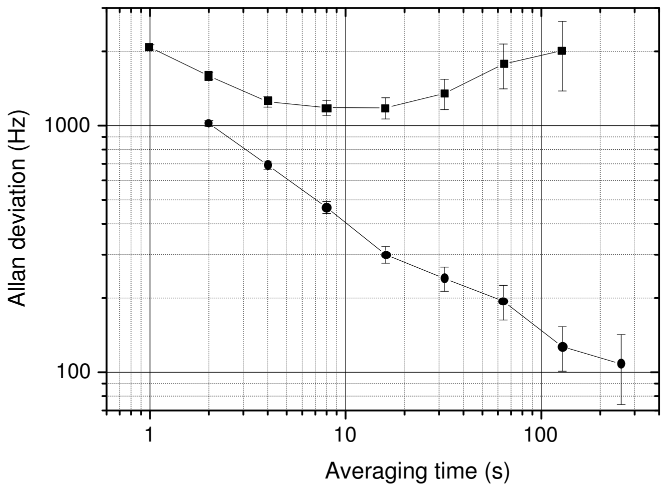

Figure 5. Typical Allan variance of the ^{88}Sr optical frequency measurement (squares) and of the ^{88}Sr-^{86}Sr isotopic shift measurement (circles).

quasi-randomized by realigning the retroreflected beam after each measurement and the resulting contribution in the final uncertainty is included as 2 kHz. Frequency noise induced by atmospheric turbulence modulating the beam pointing is measured to be less than $1\,\mathrm{kHz}/\sqrt{\mathrm{Hz}}$. The offsets and lineshape asymmetries introduced by the recoil and 2nd order Doppler are calculated by numerically integrating the 1D optical Bloch equations along the atomic trajectories considering the experimental conditions.[15] The resonance linewidth obtained from this simulation is in good agreement with the experimental value proving that we do not have unexplained line broadening mechanisms. Since we observe a closed transition we estimate the offset introduced by unbalanced counter-propagating beams and curved wavefront,[16] and wavefront distortion less than 2 kHz.

Table 1. Budget of corrections and uncertainties for the ^{88}Sr optical frequency measurement; all values are in kHz.

Statistical value	434 829 121 316.5	(5.0)
1st order Doppler	0	(2)
Recoil and 2nd order Doppler	-5.6	(0.1)
2nd order Zeeman	-0.006	(0.003)
Collisional shift	0	(0.05)
Spectral purity	0	(0.5)
Integrator offset	0	(0.2)
Curvature and unbalanced intensity	0	(2)
Final value	434 829 121 311	(10)

There is no first order Zeeman shift because we observe the J = 0 to J = 1, Δm = 0 transition. Because the excited state fine structure splitting is of the order of few THz, the second-order Zeeman shift in our magnetic field is of the order of few Hz and totally insignificant to this measurement. The collisional shift coefficient for this transition has not been measured. The self broadening coefficient is known to be about 50 MHz/torr[17] and the shift is generally considerably smaller. Hence, assuming as an upper limit for the collisional shift the self broadening coefficient and considering the background pressure of the order of 10^{-6} torr, we expect a pressure induced shift of less than 50 Hz. The spectral purity of the interrogating laser is an important subject. Unbalanced

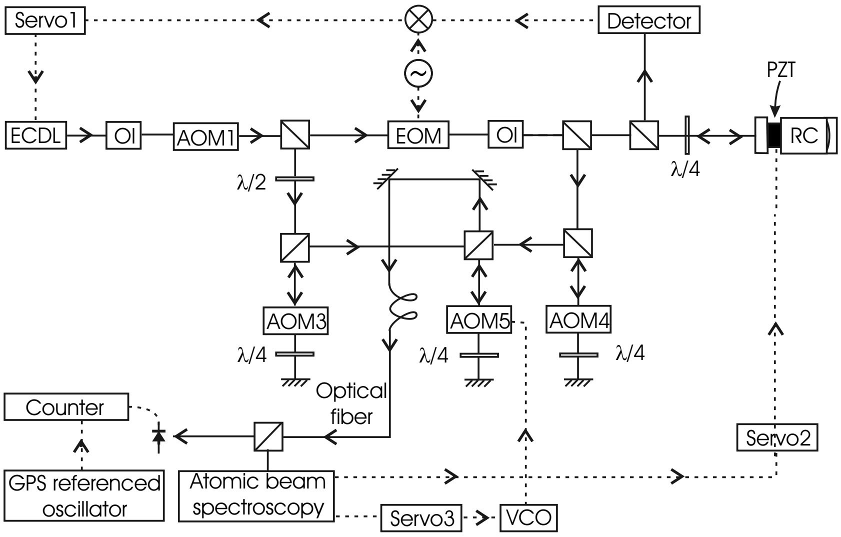

Figure 6. Experimental setup used for the isotopic shift measurement. The notation is the same as in fig. 2. VCO: voltage-controlled oscillator.

sidebands within the range of atomic linewidth will lead to a frequency pulling effect. Such unbalanced sidebands must be accompanied by a synchronous AM component; pure frequency and pure phase modulation sidebands will not lead to a pulling. Hence, a simple power detector and FFT suffice to place an upper bound on their presence. In our case, all such sidebands are more than 40 dB below the carrier and lead to a pulling that is less than 1% of the atomic linewidth. We did not experimentally observe any dependence of the measured optical frequency on the modulation depth and laser intensity, which is in agreement with numerical simulations. The resulting value for the ^{88}Sr transition frequency, including the corrections discussed previously, is 434 829 121 311 (10) kHz, corresponding to a $1\,\sigma$ relative uncertainty of 2.3×10^{-11}.

3. ISOTOPIC FREQUENCY MEASUREMENTS

With a minor change in the apparatus, we locked simultaneously and independently the frequency of two laser beams to the sub-Doppler signals of ^{86}Sr and ^{88}Sr. This system allowed us to measure the isotope shift by counting the beatnote between the two interrogating beams. For this purpose, the reference cavity is locked to ^{88}Sr resonance as described previously. The light for ^{86}Sr is derived from the same diode laser and it is brought to resonance through AOM4 (fig. 6). The two beams are overlapped in a polarization-maintaining single-mode optical fiber and sent to the interrogation region. By frequency modulating the two beams at different rates and using phase sensitive detection we get the lock signal for both the isotopes from the fluorescence signal. The lock on ^{86}Sr acts on the voltage-controlled oscillator (VCO) that drives AOM4. The ^{86}Sr lock bandwidth of 1 Hz, limited by lower signal to noise, is enough since the short term stability is insured by the lock to the reference cavity and ^{88}Sr.

In this isotope shift measurement most of the noise sources are basically common mode and rejected, the Allan variance shows a white noise spectrum of 1 kHz at 2 s and does not show any flicker noise for times longer than 500 s (fig. 5), resulting in precision better than 100 Hz. At this level of precision we observe the servo loop offset compensation limiting the reproducibility to 200 Hz. The measured ^{88}Sr-^{86}Sr isotope shift for the $^1S_0$-$^3P_1$ transition is 163 817.4 (0.2) kHz. This value represents an improvement in accuracy of more than 3 orders of magnitude with respect to previously available data.[18] The ^{86}Sr optical frequency then amounts to 434 828 957 494 (10) kHz. We did not measure the hyperfine structure and isotope shift of the ^{87}Sr since an accurate measurement requires a low magnetic field environment not compatible with our spectroscopic scheme.

The isotope-shift experiment provides also an indication of the laser frequency stability when locked to the atomic signal for periods longer than 2 s.[10] We conclude that the observed flicker noise at 5×10^{-12} in the absolute frequency measurement may be attributed to the optical frequency comb including its frequency reference. Moreover the relative uncertainty of 1.2×10^{-11} due to uncontrolled systematic effects does not explain completely the data scatter of 5×10^{-11} in the absolute frequency measurement. We did not evaluate the noise performance in the GPS disciplined quartz oscillator that is our local frequency reference. Possible sources of noise are oscillation frequency sensitivity of quartz to vibration and the behavior of the complex, adaptive filter used to discipline the quartz LO to the GPS signal[19] in the 10^3-10^4 s region, which is the time period in which we are making our measurements.

4. BLUE LASER SOURCE

Future improvements and developments of the Sr-based optical reference involve cooling and trapping of Sr atoms. Using cold atoms a precision in the range of one part in 10^{14} in one second can be expected with the transition investigated in this work, while probing the ultranarrow $0-0$ or $0-2$ transitions in cold trapped atoms should lead to a dramatic improvement in stability and accuracy opening the way to the $10^{-17}-10^{-18}$ range.

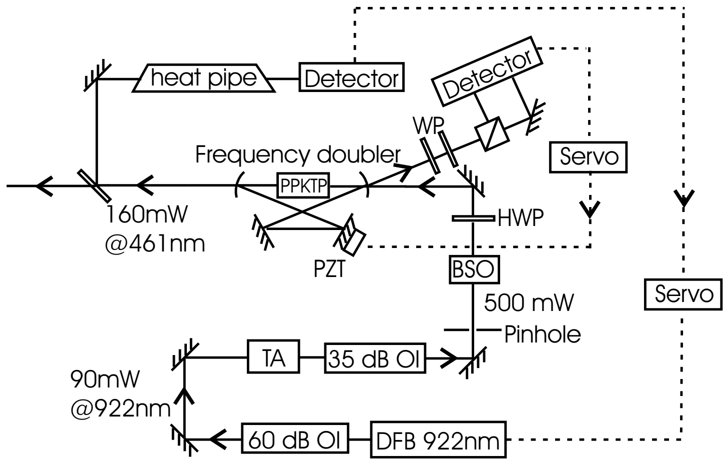

Figure 7. Setup of the blue laser source: the light from a distributed feedback diode laser (DFB) is amplified on a semiconductor tapered amplifier (TA) and then it is frequency doubled on a periodically poled KTP crystal (PPKTP) placed in an enhancement cavity. The optical cavity is kept on resonance with the incoming light with a Hänsch-Couillaud lock, while the frequency of the DFB is kept on resonance with the atomic vapor produced in a heatpipe. BSO: beam shaping optics. WP: wave plate. HWP: half-wave plate. HP: heatpipe.

Keeping the idea of realizing a compact and eventually transportable system, which would enable future tests of fundamental physics on Earth and in space, we developed an all solid state laser source at 461 nm which is required for the first stage cooling and trapping. The laser is based on a distributed feedback (DFB) diode laser at 922 nm which is amplified in a semiconductor tapered amplifier (TA) and then frequency doubled with a periodically-poled KTP crystal placed in an optical resonator. A scheme of the laser setup is reported in fig. 7. The DFB (Eagleyard Photonics, model EYP-DFB-0923-00100-1500-SOT02-0000) is operated at -16 degrees C under dry air to match the resonance condition with the atomic line and to avoid water condensation. The light from the DFB (typically 90 mW) is sent to the TA through a 60 dB optical isolator. Under operation condition the DFB linewidth is less than 3 MHz FWHM. The TA is based on a chip (Toptica Photonics, model TA-940) mounted on a temperature stabilized holder which includes the optics for injection and the output collimation. Injecting the TA with 75 mW and driving 1.6 A we obtain typically 500 mW after a 35 dB optical isolator, spatial mode filtering through a pinhole, and the beam shaping optics for the coupling into the doubler resonator. The frequency doubler is composed by a non-linear crystal, a 25 mm long periodically-poled KTP crystal, placed in an optical build up cavity. The crystal facets are anti-reflection coated both at 922 and 461 nm ($R < 0.2\,\%$) and the poling period is chosen to fulfill the phase matching of our wavelength at room temperature. The resonator has an input coupler with 11 % transmission and it is held in resonance with the input light feeding the error signal from a Hänsch-Couillaud detection[20] to a PZT controlled folding mirror. Under optimal conditions we obtain 160 mW in the blue and routinely we work with 120 mW. The overall efficiency of 32 % is explained by the yet not optimized choice of the mirrors used for the resonator.

5. CONCLUSION

We presented a new semiconductor laser source well suited for high precision spectroscopy on atomic strontium. By locking the laser to the visible $5s^2\,{}^1S_0$-$5s\,5p^3P_1$ intercombination line of Sr we measured the frequency of the transition using an optical-frequency comb-generator referenced to the SI second through a GPS stabilized quartz oscillator. The optical frequency measurement is obtained with a relative uncertainty of 2.3×10^{-11}, which represents an improvement of more than 4 orders of magnitude with respect to previous data.[7] We also obtain an accurate value for the ^{88}Sr-^{86}Sr isotope shift improving the accuracy by more than 3 orders of magnitude. We present also a compact and reliable all-solid state blue laser source that will be used of laser cooling and trapping of atomic strontium, in order to improve the accuracy and stability of the Sr-based optical reference.

The simple and compact experimental setup developed represent one of the first step towards the realization of transportable optical standards to be employed in future tests of fundamental physics on Earth and in space.

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