

# Precision frequency measurement of visible intercombination lines of strontium

G. Ferrari, P. Cancio<sup>1</sup>, R. Drullinger, G. Giusfredi<sup>1</sup>, N. Poli,  
M. Prevedelli<sup>2</sup>, C. Toninelli, and G.M. Tino

*Dipartimento di Fisica and LENS, Università di Firenze, INFN-UdR Firenze, Polo Scientifico, 50019 Sesto Fiorentino, Italy*

*1) Istituto Nazionale di Ottica Applicata, Largo E. Fermi 2, 50125 Firenze, Italy*

*2) also Dipartimento di Chimica Fisica, Università di Bologna, Via del Risorgimento 4, 40136 Bologna, Italy*

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We report the direct frequency measurement of the visible  $5s^2\ ^1S_0$ - $5s5p^3\ ^3P_1$  intercombination line of strontium that is considered a possible candidate for a future optical frequency standard. The frequency of a cavity-stabilized laser is locked to the saturated fluorescence in a thermal Sr atomic beam and is measured with an optical-frequency comb-generator referenced to the SI second through a GPS signal. The  $^{88}\text{Sr}$  transition is measured to be at 434 829 121 311 (10) kHz. We measure also the  $^{88}\text{Sr}$ - $^{86}\text{Sr}$  isotope shift to be 163 817.4 (0.2) kHz.

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The recent development of optical-frequency comb generation has made possible, for the first time, relatively easy optical frequency measurements [1,2]. This, in turn, opened the way to atomic clocks based on optical frequency transitions. Because of their higher frequency, these transitions have potential for greatly improved accuracy and stability relative to conventional atomic clocks based on microwave frequency transitions [3]. Different transitions are now considered as optical frequency standards, involving single ions and neutral atoms [4]. While single ions offer an excellent control on systematic effects, clouds of laser cooled atoms have the potential for extremely high precision. Amongst the neutral atoms, Sr has long been considered one of the most interesting candidates [5]. Several features, some of which are specific to this atom, allow different possibilities for the realization of a high precision optical clock. The intercombination  $5^1S_0$ - $5^3P$  lines 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,  $^{88}\text{Sr}$  (82%),  $^{86}\text{Sr}$  (10%),  $^{84}\text{Sr}$  (0.5%) with nuclear spin  $I=0$  and one fermionic,  $^{87}\text{Sr}$  (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 the present paper, down to the highly forbidden  $5^1S_0$ - $5^3P_{0,2}$  transitions. In  $^{87}\text{Sr}$ , the presence of hyperfine mixing makes the 0-0 transition weakly allowed with an expected natural width of about 1 mHz.

From the point of view of laser cooling and manipulation, Sr has several interesting features which are also important for the final operation of a precise frequency stan-

dard: two-stage cooling using the intercombination transition allows extremely low temperatures and magneto-optical trapping; atoms can be trapped in optical lattices with negligible shift of the optical clock transition [6]. After initial laser spectroscopy experiments based on wavelength metrology and discharges as atomic sources [7], recently Sr has been the subject of several experiments aiming to all-optical cooling down to quantum degeneracy for bosonic and fermionic isotopes [8–10], continuous atom laser [6], and detection of ultra-narrow transitions [11,12]. This atom is also considered interesting for the understanding of its spectrum [13] and for the investigation of cold collisions [14,15].

In this paper, we report the first precision frequency measurements on the intercombination  $5^1S_0$ - $5^3P_1$  transition. Using a femtosecond laser comb, we determine the absolute frequency of the transition for  $^{88}\text{Sr}$  and  $^{86}\text{Sr}$  and a very accurate value for the isotope shift. The improvement by several orders of magnitude with respect to previous data and the use of a relatively simple and compact apparatus demonstrate the potentialities of this system.

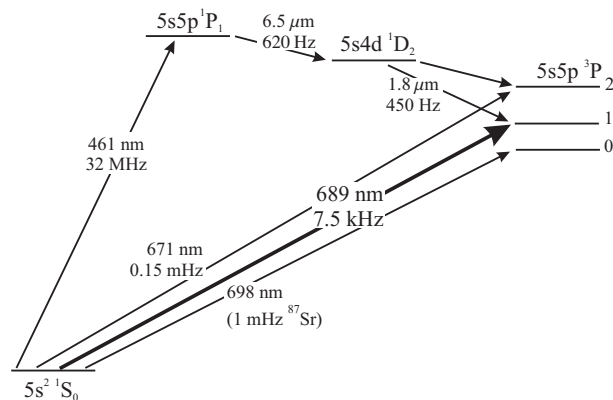


FIG. 1. Relevant energy levels (not to scale) and transition linewidth for high resolution spectroscopy and atomic manipulation of bosonic strontium.

The experimental setup we use 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 Global Positioning System (GPS) controlled quartz. A scheme of the experimental setup is given in Fig. 2. The extended cav-

ity 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 [16]; the phase modulation is produced by a resonant electro-optic modulator (EOM) driven at 21 MHz. 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.

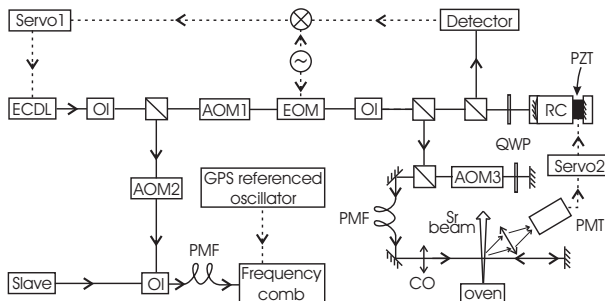


FIG. 2. Experimental setup used for the frequency measurement on the Sr intercombination line. Optical isolators (OI) and acousto-optic modulators (AOM) eliminate feedback among the master laser (ECDL), the slave laser, the electro-optic modulator (EOM) and the reference cavity (RC). Solid lines represent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

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 [17]. We do not passively stabilize the RC in a vacuum chamber [18] since the acoustic and sub acoustic noise is removed 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 and using a bundle of stainless steel capillaries to collimate it [19]. The residual atomic beam divergency is 25 mrad and the typical atomic density in the detection region is  $10^8$  cm $^{-3}$ .

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 is collected on a photomultiplier tube with an efficiency of 0.4% including quantum efficiency and solid angle. Orthogonality between atomic and laser beams is optimized by centering the Lamb dip with respect to the Doppler profile.

The laser beam is filtered using a single mode fiber and 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 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  $\mu$ rad maximizing the transmitted power through the fiber. The peak beam intensity of  $60 \mu$ Wcm $^{-2}$  (to be compared to the saturation intensity of  $3 \mu$ Wcm $^{-2}$ ) was chosen to obtain sufficient signal to noise for the RC lock onto the atomic resonance. A uniform magnetic field of 10 G defines the quantization axis in the interrogation region such that the light is  $\pi$  polarized.

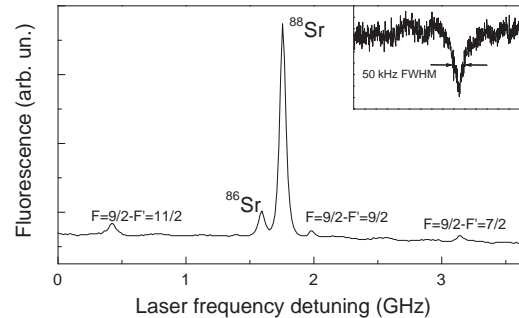


FIG. 3. Fluorescence spectrum of the strontium  $1S_0-3P_1$  line at 689 nm. The lines of the two bosonic isotopes  $^{86}\text{Sr}$  and  $^{88}\text{Sr}$ , together with the hyperfine structure of the fermionic  $^{87}\text{Sr}$ , can be resolved. The linewidth corresponds to the residual 1<sup>st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

The acousto-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.

Fig. 3 shows the Doppler broadened resonances of  $^{88}\text{Sr}$ ,  $^{86}\text{Sr}$  and the hyperfine structure of  $^{87}\text{Sr}$ . The resid-

ual atomic beam divergency produces a residual Doppler broadening of 60 MHz FWHM. In the inset, the sub-Doppler signal for  $^{88}\text{Sr}$  is shown. Two independent measurements [20] of the sub-Doppler resonance show a FWHM of about 50 kHz, which is in agreement with the expected value considering the saturation and transit time broadening, and the recoil splitting.

We measure the optical frequency through a commercial optical frequency comb generator based on a Kerr-lens mode-locked Ti:Sa laser with a repetition rate of 1 GHz (MenloSystems GmbH, Model FC8003). The repetition rate and carrier offset envelope frequency are locked to a GPS stabilized quartz oscillator, as well as counters and RF generators for AOM's. Figure 4 shows the result of the measurement of the  $^{88}\text{Sr}$  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 100-200s. 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.

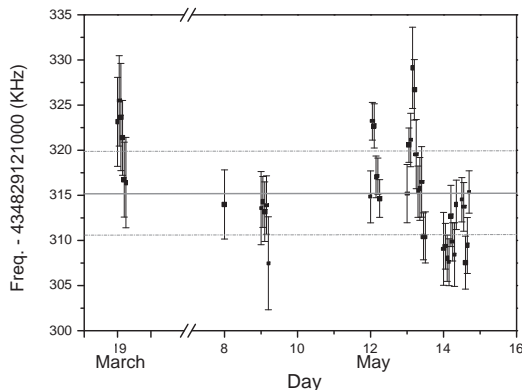


FIG. 4. Measurements used to determine the transition frequency. The error bars correspond to the standard deviation for each data set.

We evaluated 1<sup>st</sup> and 2<sup>nd</sup> order Doppler and Zeeman effects, AC Stark shift, collisional shifts, and mechanical effects of light (table I). The 1<sup>st</sup> order Doppler shift resulting from imperfect alignment in the standing wave was randomized by realigning the retroreflected beam after each measurement; the resulting contribution in the final uncertainty is included as 2 kHz. The offsets and lineshape asymmetries introduced by the recoil, atom deflection induced by the light field, and 2<sup>nd</sup> order Doppler were calculated by numerically integrating the 1D optical Bloch equations along the atomic trajectories considering the experimental conditions [21]. 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 [22], and wavefront distortion less than 2 kHz. There is no first order Zeeman shift because we observe the  $J=0$  to  $J=1$ ,  $\Delta m=0$  transition. The second-order Zeeman shift in our magnetic field is of the order of few Hz. The collisional shift coefficient for this transition has not been measured but the self broadening coefficient is known to be about 50 MHz/torr [23]. 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. Unbalanced sidebands on the interrogating laser were measured to be more than 40 dB below the carrier leading to a shift smaller than 1% of the atomic linewidth. In our experimental conditions, 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  $^{88}\text{Sr}$  transition frequency, including the corrections discussed previously, is 434 829 121 311 (10) kHz, corresponding to a  $1\sigma$  relative uncertainty of  $2.3 \times 10^{-11}$ .

With a minor change in the apparatus, we locked simultaneously the frequency of two laser beams to the sub-Doppler signals of  $^{86}\text{Sr}$  and  $^{88}\text{Sr}$ . 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  $^{88}\text{Sr}$  resonance as described previously and the light for  $^{86}\text{Sr}$  is derived from the same laser beam and brought to resonance through AOM's. The two beams are overlapped in a single mode optical fiber and sent to the interrogation region. By frequency modulating the beams at different rates and using phase sensitive detection we get the lock signal for both the isotopes from the same photomultiplier. The lock on  $^{86}\text{Sr}$  acts on the voltage-controlled oscillator that drives one of its AOMs. The  $^{86}\text{Sr}$  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  $^{88}\text{Sr}$ . 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 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  $^{88}\text{Sr}$ - $^{86}\text{Sr}$  isotope shift for the  $^1\text{S}_0$ - $^3\text{P}_1$  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 [24]. The  $^{86}\text{Sr}$  optical frequency then amounts to 434 828 957 494 (10) kHz.

The isotope-shift experiment provides also an indication of the stability of the lock to the atomic line for

periods longer than 2 s [18]. We conclude that the observed flicker noise at  $5 \times 10^{-12}$  in the absolute frequency measurement may be attributed to the optical frequency comb including its frequency reference. Moreover the relative uncertainty of  $1.2 \times 10^{-11}$  due to uncontrolled systematic effects does not explain completely the data scatter of  $5 \times 10^{-11}$  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 behaviour of the complex, adaptive filter used to discipline the quartz local oscillator to the GPS signal in the  $10^3$ - $10^4$  s region, which is the time period in which we are making our measurements.

In conclusion, we demonstrated locking of a laser-diode to the visible  $5s^2\ ^1S_0$ - $5s5p\ ^3P_1$  intercombination line of Sr and measured its frequency using an optical-frequency comb-generator referenced to the SI second through a GPS signal. The optical frequency measurement is obtained with a relative uncertainty of  $2.3 \times 10^{-11}$ , 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  $^{88}\text{Sr}$ - $^{86}\text{Sr}$  isotope shift improving the accuracy by more than 3 orders of magnitude.

Future improvements and developments 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. Probing the ultra-narrow  $0 - 0$  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. A Sr-based optical reference could employ all-solid-state laser sources (including light at 461 nm required for cooling and trapping). The realization of ultra-precise optical frequency standards based on compact and eventually transportable systems will enable future tests of fundamental physics on Earth and in space.

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TABLE I. Budget of corrections and uncertainties for the  $^{88}\text{Sr}$  optical frequency measurement; all values are in kHz.

STATISTICAL VALUE	434 829 121 316.5 (5.0)
1 <sup>st</sup> order Doppler	0 (2)
Recoil and 2 <sup>nd</sup> order Doppler	-5.6 (0.1)
2 <sup>nd</sup> 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)

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