We report the direct frequency measurement of the visible $5s^2 1S_0-5s 5p^3 P_1$ 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 $5^1S-5^3P$ 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 has allowed the observation of the 0-0 transition which is expected to have a natural width of only 1 mHz [6].

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 standard: 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; all-optical cooling down to quantum degeneracy should be possible for bosonic and fermionic isotopes [7–9]. After initial laser spectroscopy experiments based on wavelength metrology and discharges as atomic sources [10], recently Sr has been the subject of several experiments aiming to all-optical Bose-Einstein condensation [7], continuous atom laser [11] and detection of ultra-narrow transitions [6,12]. From the theoretical point of view, this atom is also considered interesting for the understanding of its spectrum [13,14] and for the investigation of cold collisions [15,16].

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 with respect to previous data, obtained with a relatively simple and compact apparatus, demonstrate the potentialities of this system.

![Energy Levels and Transition linewidth](image)

**FIG. 1.** Relevant energy levels 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 Posi-
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 locked of the cavity 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. We do not passively stabilize the RC in a vacuum chamber [19]. The strontium atomic beam is obtained from the metal vaporization System (GPS) controlled quartz. 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 [17]; 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.

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 [20]. The residual atomic beam divergency is 25 mrad and the typical atomic density in the detection region is $10^{15}$ cm$^{-3}$.

The Doppler-free atomic line is resolved by saturation spectroscopy using two counter-propagating 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 (Fig. 3).

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 Gauss defines the quantization axis in the interrogation region such that the light is $\pi$ 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

FIG. 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: wave-plate. PMT: photomultiplier tube. CO: collimation optics. PMF: polarization maintaining fiber.

FIG. 3. Fluorescence spectrum of the strontium $^1$S$_0$-$^3$P$_1$ 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 1$^\text{st}$ 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.
against the same GPS clock used to reference the frequency comb. Fig. 3 shows the Doppler broadened resonances of $^{88}$Sr, $^{86}$Sr and the hyperfine structure of $^{87}$Sr. The residual atomic beam divergency produces a residual Doppler broadening of 60 MHz FWHM. In the inset, the sub-Doppler signal for $^{88}$Sr is shown. Two independent measurements [21] 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 [22] 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 oscillator. 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.

Analysis of potential systematic errors (see table I) 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 mechanical effects of light. 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 1st order Doppler shift was 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 kHz/√Hz. The offsets and lineshape asymmetries introduced by the recoil, atom deflection induced by the light field, and 2nd order Doppler are calculated by numerically integrating the 1D optical Bloch equations along the atomic trajectories considering the experimental conditions [23]. 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 [24], 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. 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 but the self broadening coefficient is known to be about 50 MHz/torr [25] 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 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 FM 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 σ 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 $^{86}$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 and the light for $^{88}$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 photomulti-
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 $^{86}\text{Sr}^{86}\text{Sr}$ isotope shift for the $^1S_0-^3P_1$ transition is $163.817.4(0.2)\text{ kHz}$. This value represents an improvement in accuracy of more than 3 orders of magnitude with respect to previous data \cite{26}. The $^{86}\text{Sr}$ optical frequency then amounts to $434.828.957.494(10)\text{ kHz}$. We did not measure the hyperfine structure and isotope shift of the $^{87}\text{Sr}$ 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 \cite{19}. 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 \cite{27} in the $10^3-10^4\text{ s}$ 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 $5s^21S_0-5s5p^3P_1$ 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 \cite{10}. 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 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. 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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\end{thebibliography}
<table>
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