PHYSICAL REVIEW A **68**, 030501(R) (2003)

Clock transition for a future optical frequency standard with trapped atoms

Irène Courtillot, Audrey Quessada, Richard P. Kovacich, Anders Brusch, Dmitri Kolker, Jean-Jacques Zondy, Giovanni D. Rovera, and Pierre Lemonde*

BNM-SYRTE, Observatoire de Paris 61, Avenue de l'Observatoire, 75014 Paris, France

(Received 5 March 2003; published 9 September 2003)

We report direct excitation of the strongly forbidden $5s^2\,{}^1S_0$-$5s5p\,{}^3P_0$ transition in ^{87}Sr. Its frequency is 429 228 004 235(20) kHz. A resonant laser creates a small leak in a magneto-optical trap (MOT): atoms build up to the metastable $^3P_0$ state and escape the trapping process, leading to a detectable decrease in the MOT fluorescence. This line has a natural width of 10^{-3} Hz and can be used for a new generation of optical frequency standards using atoms trapped in a light-shift-free dipole trap.

DOI: 10.1103/PhysRevA.68.030501 PACS number(s): 32.30.Jc, 06.30.Ft, 32.80.−t, 39.30.+w

In addition to being at the heart of the international system of units, extreme frequency metrology finds a wide range of applications throughout physics. As an example, tests of the stability of fundamental constants are performed by comparing the frequencies of different atomic transitions [1,2]. These laboratory tests are already competitive with those performed at the cosmological scale [3,4] and improve with the accuracy of the atomic transitions frequency measurements. Microwave frequency standards such as atomic fountains now have relative accuracies better than 10^{-15} with potential improvements down to 10^{-16} [5]. Going further, however, seems extremely difficult due to the limited quality factor of the atomic resonance in these devices ($\sim 10^{10}$). Optical frequency standards exhibit much higher line-Q, up to 10^{14} as recently demonstrated in Ref. [6]. These devices reach fractional frequency instabilities below 10^{-14} over 1 s [7], almost one order of magnitude better than fountains [5]. Their current accuracy lies near 1 part in 10^{14} with anticipated large room for improvement [2,8–11].

Two different approaches are commonly used to develop optical frequency standards [12]. The first one is based on the spectroscopy of a single trapped ion, the second on the spectroscopy of a large ensemble of free falling neutral atoms. It is a commonly shared opinion that the ion approach may lead to a better ultimate frequency accuracy due to the "perfect" control of the ion motion, while the atomic approach should lead to a better frequency stability thanks to the numerous quantum references contributing to the signal. Recently Katori proposed a scheme that combines the advantages of both approaches [13]. The idea is to trap neutral atoms in the Lamb-Dicke regime in an optical lattice operating at a wavelength where the light shift of the clock transition vanishes.

Katori proposed to use ^{87}Sr probed on the $5s^2\,{}^1S_0$-$5s5p\,{}^3P_0$ line at 698 nm, which indeed seems an ideal system for the realization of this scheme. This $J=0-J=0$ resonance is only slightly allowed by hyperfine coupling and its metrological properties are exquisite. A natural linewidth of ~ 1 mHz can be derived from the hyperfine data found in Ref. [14]. It has a high insensitivity to external electromagnetic fields. The light-shift cancellation is expected to occur near 800 nm, a wavelength far from any atomic resonance and for which powerful and practical laser sources are readily available. Apart from a residual effect due to hyperfine structure, the light-shift cancellation is independent on polarization. Finally, higher-order effects due to the trapping field are expected to be extremely small on this particular line [15]. We report here the direct excitation and frequency measurement of this transition, the most forbidden optical transition of a neutral atom ever directly detected.

The experiment is performed on a sample of cold atoms collected in a magneto-optical trap (MOT). An atomic beam is decelerated in a Zeeman slower and captured at the crossing point of three retroreflected beams tuned 40 MHz to the red of the ${}^1S_0$-${}^1P_1(F=9/2)$ transition (Fig. 1). The setup is described in details in Ref. [16]. For this particular experiment 3×10^6 ^{87}Sr atoms are trapped at steady state at a temperature of 2 mK. The atomic cloud has a $1/e^2$ diameter of 2 mm. To probe the 698-nm line, a 14-mW laser beam passes four times through the atomic cloud in a standing wave configuration. The probe beam waist radius is 1.3 mm. Generally speaking, the detection of extremely forbidden transitions is an experimental challenge, and at first sight it seems a desperate task with our system: the resonance is expected to be Doppler broadened to 1.5 MHz [full width at half-maximum (FWHM)] more than three orders of magnitude higher than the Rabi frequency (0.8 kHz). Thus only 10^3 atoms are ex-

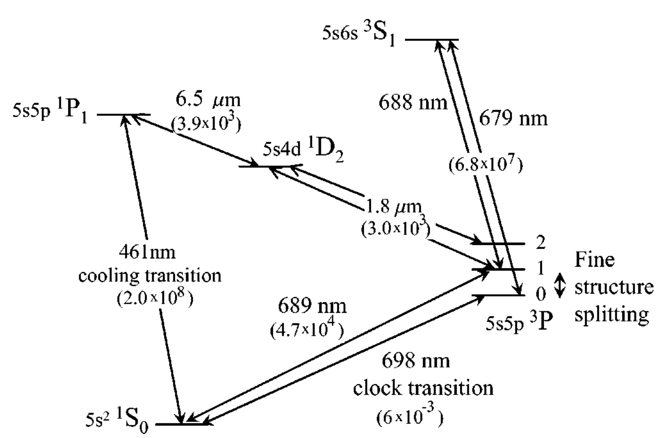

FIG. 1. Energy diagram of ^{87}Sr with wavelength and decay rate (s^{-1}) of the transitions involved in the experiment. For clarity, the hyperfine structure is not represented ($I=9/2$).

*Electronic address: pierre.lemonde@obspm.fr

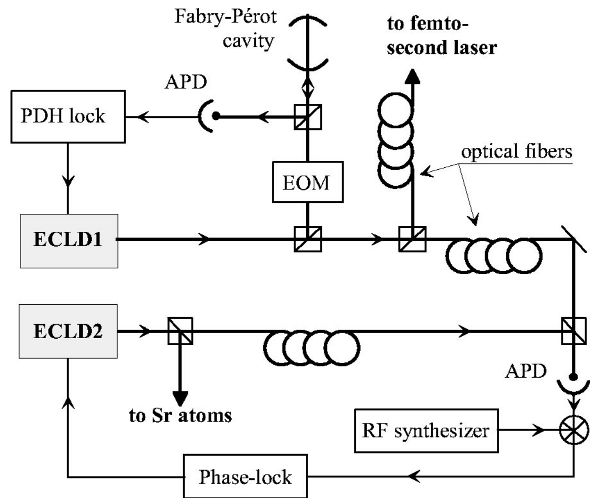

FIG. 2. Experimental setup used for frequency measurements. An extended cavity laser diode (ECLD1) is locked to a high finesse Fabry-Pérot cavity ($F=27\,000$). 100 μW are sent to a frequency chain based on a femtosecond laser for absolute frequency measurement. A second laser (ECLD2) which is used to probe the strontium atoms is offset-phase locked to ECDL1. The beat note collected by an avalanche photodiode (APD) is mixed with a radio-frequency reference. Noise and frequency offsets introduced by optical fibers are negligible in this experiment. EOM: electro-optical modulator.

pected to be resonant with the laser at a time. In addition, an efficient fluorescence detection of atoms in the $^3P_0$ state is made difficult by the absence of cycling transition from this state. Finally the energy of the $^3P_0$ state is known from databases to within a few hundred megahertz only.

To overcome these problems we have first determined the resonance frequency with an accuracy of 110 kHz by measuring the frequency of the $^1S_0$-$^3P_1$ line at 689 nm and of the frequency difference between the $^3P_1$-$^3S_1$ and $^3P_0$-$^3S_1$ transitions at 688 nm and 679 nm, respectively (see Fig. 1) [17]. Second, to amplify the fraction of cold atoms transferred to the $^3P_0$ state by direct excitation, we have taken advantage of the fact that the lifetime of the MOT is 40 times larger than the Rabi oscillation period. A laser tuned to resonance induces a leak in the MOT leading to a detectable decrease in the number of trapped atoms of 1%.

The experimental setup used for all frequency measurements is described in Fig. 2. An extended cavity laser diode (ECLD1) is locked to a high finesse cavity using the Pound-Drever-Hall method [18] with performances reported in Ref. [19]. Its frequency is continuously measured vs a hydrogen maser with a scheme based on a self-referenced femtosecond Ti:sapphire laser [20,21]. The relative resolution of this measurement is typically 3×10^{-13} for a 1-s averaging time. A second laser (ECLD2) is offset-phase locked to ECLD1. The beat note between both lasers is mixed with the output of a radio-frequency synthesizer to generate the offset-phase lock error signal. The bandwidth of the servo control is 2 MHz. With this scheme the light of ECLD2, which is sent to the atoms, can be tuned to any frequency between two modes of the cavity of free spectral range 1.5 GHz by actuating the rf synthesizer. Two sets of lasers are used. One can be tuned from 675 to 685 nm, the other from 685 to 698 nm.

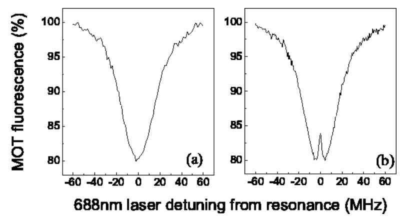

FIG. 3. Relative MOT fluorescence signals obtained when a 688-nm laser is swept around the $^3P_1,F=9/2$-$^3S_1,F=11/2$ transition. (a) 688-nm laser only; (b) with an additional 679-nm laser tuned to the $^3P_0,F=9/2$-$^3S_1,F=11/2$ resonance. In this case the CPT dip is observed. Intensities of the 688-nm and 679-nm lasers are 5 μW/mm^2 and 2 mW/mm^2, respectively.

The frequency of the $^1S_0$-$^3P_1(F=9/2)$ transition is measured with an atomic beam, which appeared to be simpler than with the cold atom setup. The obtained frequency is 434 829 342 950(100) kHz, with an uncertainty mainly dominated by statistical noise and imperfect knowledge of the magnetic field.

The determination of the fine-structure splitting between $^3P_0$ and $^3P_1$ is performed with the cold atoms. While trapped and cycling on the $^1S_0$-$^1P_1$ transition, atoms eventually emit a spontaneous photon which brings them to the $^1D_2$ state (Fig. 1). This state has two main decay channels: to the $^3P_1$ and $^3P_2$ states. Atoms in the $^3P_1$ state decay back to the ground state and are kept in the trap while atoms in the metastable $^3P_2$ state are lost. This process limits the lifetime of the MOT to some 50 ms. We make the fine-structure measurement by modifying this escape process. If a laser resonant to one of the hyperfine components of the $^3P_1$-$^3S_1$ transition is added to the trap, atoms in the corresponding $^3P_1$ state are pumped to the $^3P_2$ and $^3P_0$ metastable states. They escape the trap instead of decaying back to the ground state. This decreases the trapped atom number. Figure 3(a) shows the fluorescence of the MOT as a function of the 688-nm laser detuning from the $^3P_1,F=9/2-{}^3S_1,F=11/2$ resonance.

A direct detection of the $^3P_0$-$^3S_1$ transition by the same technique is not possible because $^3P_0$ is not populated in the MOT. Instead, a first signal is obtained by detecting the light shift of the $^3P_1$-$^3S_1$ transition induced by a 679-nm laser close to the $^3P_0$-$^3S_1$ resonance. With an intensity of 1.8 mW/mm^2, a shift of 100 kHz is observed for a detuning of 100 MHz. For a 679-nm detuning smaller than the width of the 688-nm resonance, a dip appears in the resonance profile due to coherent population trapping (CPT): when the frequency difference between both lasers matches the atomic fine structure, there exists a coherent superposition of $^3P_1$ and $^3P_0$ states which is not coupled to $^3S_1$ [22]. Atoms in this dark state are not pumped to $^3P_2$ but decay back to the ground state in a few 10 μs due to the $^3P_1$ instability. They are kept in the MOT. When the CPT dip is centered on the 688-nm resonance, the 679-nm laser is tuned to resonance. The observed signal in this configuration is shown in Fig. 3(b). The fine-structure measurement is performed light-shift-free with both lasers locked on resonance. We measured a frequency of 5 601 338 650(50) kHz with an uncertainty mainly due to the magnetic field gradient of the MOT. Ac-

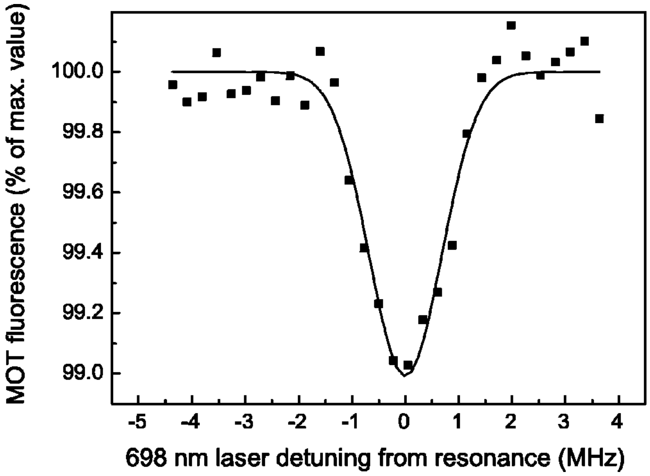

FIG. 4. Direct observation of $^1S_0$-$^3P_0$ transition. The line is broadened by the Doppler effect due to the finite temperature of the atoms to 1.4 MHz (FWHM).

cording to these measurements, the $^1S_0$-$^3P_0$ transition is expected to have a frequency of 429 228 004 300(110) kHz.

For the direct observation, we induce in the MOT a leak to the $^3P_0$ state with a laser tuned to resonance. The lifetime of the trap is two orders of magnitude longer than the duration of a π pulse on the forbidden transition with the laser parameters given above. This leads to a buildup by the same factor of the fraction of atoms escaping the MOT if the transfer rate to $^3P_0$ is constant and if the atoms, once in the $^3P_0$ state, actually escape the trapping process. The trapped atom number should then decrease by several perecent. One way to fulfill the transfer rate condition could consist in using the MOT to rethermalize atoms to fill the dip in the velocity distribution created by excitation to $^3P_0$. For the escape condition, one could think of optical pumping to $^3P_2$ with a laser tuned to $^3P_0$-$^3S_1$. In our experiment however, the Doppler effect induced by gravity is sufficient to fulfill both conditions. The experiment is operated sequentially: by means of acousto-optic modulators we alternate a capture and cooling phase with the blue lasers and a probe phase with the 698-nm laser. During the probe phases atoms are free falling both in the ground and exciting states. The frequency sweep caused by acceleration amounts to several times the Rabi frequency per millisecond with the 45° angle formed by the 698-nm probe beam and vertical. Atoms transferred to the $^3P_0$ state and the corresponding dip in the velocity distribution of the ground state are then rapidly detuned from the excitation laser. The pulsed operation also avoids any light shift of the forbidden transition by the trap beams. The optimization of the time sequence results from a trade-off between the capture efficiency of the MOT, the ballistic expansion of the atomic cloud during probe phases, and the efficiency of excitation to $^3P_0$. With phases of capture of 3 ms duration and probe phases of 1 ms duration, we have 1×10^6 atoms at steady state in the MOT and the contrast of the resonance is 1%. In Fig. 4 is shown the fluorescence of the trapped atoms vs the 698-nm laser detuning from resonance. A narrow sub-Doppler structure is expected at the center of the resonance due to the standing-wave configuration. With the present signal-to-noise ratio, we are not able to detect it.

TABLE I. Measured frequencies of ^{87}Sr transitions ($F=9/2$ for all atomic states).

Transition	Frequency (kHz)
$5s^2\ ^1S_0$-$5s5p\ ^3P_1$	434829342950 ± 100
$5s5p\ ^3P_0$-$5s5p\ ^3P_1$	5601338650 ± 50
$5s^2\ ^1S_0$-$5s5p\ ^3P_0$	
Indirect measurement	429228004300 ± 110
Direct measurement	429228004235 ± 20

We have locked the laser to the $^1S_0$-$^3P_0$ resonance for the determination of its frequency. Measurements on both sides of the resonance are alternated with a 140 ms duration. The resulting error signal is integrated and fed back to the synthesizer driving the offset-phase lock of ECLD2. The average frequency is 429 228 004 235 kHz with a residual statistical uncertainty of 20 kHz after 2 h of integration. At that level, the systematic effects are negligible. The first-order Doppler effect vanishes due to the standing-wave configuration. The Zeeman effect induced by the MOT field gradient is less than 1 kHz, since the Landé factors of the $^1S_0$ and $^3P_0$ levels are of the order of 10^{-4}. The light shift which would result from imperfect extinction of the blue light is also estimated below 1 kHz. Table I summarizes the indirect and direct measurements and confirms the good agreement between both methods.

The technique used for the direct detection of this line can be extended to other atomic species. As an example, Yb has the same level scheme as Sr with several isotopes having nonzero nuclear spin. With Yb atoms trapped on the $^1S_0$-$^3P_1$ transition of natural width 200 kHz, the lifetime of the MOT can be much longer than the blue Sr MOT, up to several seconds in Ref. [24]. The contrast of the forbidden line would then be close to 100%.

The observation of the $^1S_0$-$^3P_0$ transition is a first step towards the realization of an optical frequency standard using trapped neutral atoms. Clearly the next step is the measurement of the wavelength of the dipole trap beam where light-shift cancellation occurs. This measurement requires better frequency resolution than achieved here, i.e., higher laser intensity at 698 nm and/or colder atoms.

In this type of optical frequency standards a line-Q as high as 10^{15} is achievable if the spontaneous emission rate in the optical trap is less than one per second. With a laser intensity of 8×10^7 W/m^2, the trap oscillation frequency can be 50 kHz and the spontaneous emission rate 0.4 s^{-1}. With a line-Q of 10^{15} and a reasonable trapped atom number of the order of 10^6, ultimate performances are orders of magnitude better than existing devices. The frequency noise of the laser used to probe the atoms will then be of decisive importance [19]. State-of-the-art ultrastable lasers use macroscopic resonators as a reference and exhibit $1/f$ frequency noise at Fourier frequencies below a few Hertz [23]. We propose to circumvent this problem with a first stage servo control to the atomic transition with a separate setup fully optimized for frequency stability: large number of atoms, favorable duty cycle, etc. At the price of an increase of the experimental width of the atomic resonance the response time of the servocontrol can be as fast as desired. With this additional degree of freedom, the optimization of ultrastable lasers should lead to a large improvement of their performances. One could then approach the demanding requirements of the optical standard using trapped neutral atoms.

We thank Ouali Acef, André Clairon, Michel Lours, and Giorgio Santarelli for helpful discussions, and the optoelectronic group of the University of Bath (UK) for providing the photonic crystal fiber of the frequency chain. A.B. acknowledges a grant from the European Research Training Network CAUAC. BNM-SYRTE is Unité Associée au CNRS (UMR 8630).

[1] H. Marion *et al.*, Phys. Rev. Lett. **90**, 150801 (2003).
[2] S. Bize *et al.*, Phys. Rev. Lett. **90**, 150802 (2003).
[3] T. Damour and F. Dyson, Nucl. Phys. B **480**, 37 (1996).
[4] J. Webb *et al.*, Phys. Rev. Lett. **87**, 091301 (2001).
[5] S. Bize *et al.*, in *Sixth Symposium on Frequency Standards and Metrology*, edited by P. Gill (World Scientific, Singapore, 2002).
[6] R.J. Rafac *et al.*, Phys. Rev. Lett. **85**, 2462 (2000).
[7] S.A. Diddams *et al.*, Science **293**, 825 (2001).
[8] M. Niering *et al.*, Phys. Rev. Lett. **84**, 5496 (2000).
[9] F. Riehle, G. Wilpers, T. Binnewies, and J. Helmcke, in *Sixth Symposium on Frequency Standards and Metrology* (Ref. [5]).
[10] E. A. Curtis *et al.*, in *Sixth Symposium on Frequency Standards and Metrology* (Ref. [5]).
[11] J. Stenger *et al.*, Opt. Lett. **26**, 1589 (2001).
[12] See *Sixth Symposium on Frequency Standards and Metrology* (Ref. [5]).
[13] H. Katori, in *Sixth Symposium on Frequency Standards and Metrology* (Ref. [5]).
[14] H.-J. Kluge and H. Sauter, Z. Phys. **270**, 295 (1974).
[15] V.G. Pal'chikov, J. Opt. B: Quantum Semiclassical Opt. **5**, S131 (2003).
[16] I. Courtillot *et al.*, Opt. Lett. **28**, 468 (2003).
[17] We also performed absolute frequency measurements of $^1S_0$-$^3P_1$, $^3P_0$-$^3S_1$, and $^3P_1$-$^3S_1$ transitions for all hyperfine sublevels of ^{87}Sr and ^{88}Sr. These measurements will be reported subsequently.
[18] R.W.P. Drever *et al.*, Appl. Phys. B: Photophys. Laser Chem. **B31**, 97 (1983).
[19] A. Quessada *et al.*, J. Opt. B: Quantum Semiclassical Opt. **5**, S150 (2003).
[20] R. Holzwarth *et al.*, Phys. Rev. Lett. **85**, 2264 (2000).
[21] D.J. Jones *et al.*, Science **288**, 635 (2000).
[22] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento B **36**, 5 (1976).
[23] B. Young, F. Cruz, W. Itano, and J.C. Bergquist, Phys. Rev. Lett. **82**, 3799 (1999).
[24] T. Kuwamoto, K. Honda, Y. Takahashi, and T. Yubuzaki, Phys. Rev. A **60**, R745 (1999).