High-Precision Measurement of the Laser-Trapping Frequencies for $^{209,210,211}$Fr Atoms

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We present the accurate measurement of the frequency of the $7S - 7P$ laser-trapping transition for three francium isotopes. Our approach is based on an interferometric comparison to deduce the unknown laser frequency from a secondary laser frequency-standard. After careful investigation of systematics, with samples of about 100 atoms the final accuracy reaches 8 MHz, an order of magnitude better than the best previous measurement for $^{210}$Fr, and opens the way to improved tests of the theoretical computation of Fr atomic structure.

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Francium, the heaviest alkali, is a promising atom for probing fundamental symmetries in physics: there are projects to measure parity violation in electron-nucleon interactions (parameterized by weak charges) and in nucleon-nucleon forces (via anapole moments), and also to search for an electric dipole moment of the electron (EDM). All the isotopes of Fr being radioactive make this research field extremely challenging, due to the small quantities of atoms available. Laser trapping is an effective tool that allows to concentrate the samples in a small volume. This technique was successfully employed to trap Na, K, Rb and Fr radioactive isotopes, in view of measurements of fundamental interactions: parity violation in nuclear beta decay of Rb and $\beta - \nu$ correlation in Na, K. Wieman’s group performed spectroscopic studies on trapped $^{221}$Fr coming from radioactive decay of $^{229}$Th. The group at Stony Brook produced and trapped Fr online with their accelerator: an impressive amount of spectroscopic information about wavelengths and lifetimes was achieved, mainly for $^{210}$Fr.

The availability of high quality experimental data stimulated the improvement of theoretical studies on the atomic structure of Fr and its isoelectronic sequence: an example with extensive reference to the relevant literature is Ref. \cite{1}. The success in trapping small samples of Fr also triggered the proposal of a new approach to measure parity violation in atoms \cite{2}. Here we report on the measurement of transition frequencies with a final accuracy an order of magnitude better than the best one made so far. This will lead to a more accurate comparison between experiment and the theory of Fr energy levels, which constitutes a necessary step to properly investigate fundamental symmetries.

Our experiment to produce and collect Fr in a magneto-optical trap (MOT) was set up in Legnaro: the apparatus is described in Ref. \cite{3}. In short, Fr ions are produced by nuclear fusion of a 100 MeV $^{18}$O beam colliding on a heated gold target; the ions are then injected in a secondary electrostatic beam line, pass through a low-resolution mass selector and are conveyed to a heated yttrium neutralizer inside the pyrex MOT cell, where the atoms are captured by the lasers. The cold cloud is imaged on a high sensitivity CCD camera, able to detect as few as ten trapped atoms. We mainly trap the most abundant isotope ($^{210}$Fr), but we also succeeded in trapping the $^{209}$Fr and $^{211}$Fr isotopes.

We use a standard configuration for the MOT, with a tunable Ti:Sa laser for trapping, a diode laser for repumping and a magnetic field gradient of about 7 gauss/cm. For each isotope, the repumping laser is tuned to the $7S(F = I - 1/2) - 7P_{1/2}(F = I + 1/2)$ $D_1$ transition at 817 nm and the trap laser to the $718$ nm $7S(F = I + 1/2) - 7P_{3/2}(F = I + 3/2)$ cycling transition; for atom cooling the second laser is slightly red-detuned. Tuning is very critical: in order to reproducibly trap each Fr isotope, it is important to know the laser absolute frequency with an uncertainty comparable to the natural width ($\Gamma = 7.6$ MHz) of the line. However the best available measurement was performed only for $^{210}$Fr with a wavemeter and was affected by a 90 MHz uncertainty \cite{4}. So we decided to implement a system that allowed us to improve the information about the trapping frequencies and the energy of the atomic levels at stake, and here we present the principle of the experimental method.

The measurement accuracy that can be expected for the frequencies of trap transitions in Fr is limited to a few MHz by the natural width $\Gamma$ and in our experiment the precision is also constrained by the modest signal-
to-noise ratio allowed by the small number of atoms. In these conditions it was reasonable to choose the classical methods of optical interferometry, without resorting to the complex technique of femtosecond frequency combs.

We then developed a methodology based on the use of a Fabry-Perot resonator to measure the frequency \( \nu_1 \) of the Ti:Sa laser with respect to a well-known frequency \( \nu_0 \) of a different laser (\( \nu_0 \) is attained by comparison with a third laser, the frequency standard). The principle of the experimental apparatus is described in Fig. 1. The idea is to overlap the two laser beams and observe the interferometer transmission: for very accurate measurements the interferometer should be operated in vacuum and at different mirror spacings [11]. Unfortunately, in the first implementation of our setup we could not satisfy these requirements, therefore much of the following discussion is devoted to explain how we treated and evaluated the systematic effects of the air refractive index and of the reflective phase shift. Actually when both lasers are transmitted by the Fabry-Perot, their frequencies are linked by the resonance condition [11, 12]

\[
\nu_i \cdot n(\nu_i) = \frac{c}{2d} \left( N_i - \frac{\psi(\nu_i)}{2\pi} + \frac{\phi_i}{\pi} \right), \quad i = 0, 1, \quad (1)
\]

where \( n \) is the frequency-dependent refractive index of air, \( c \) is the speed of light, \( d \) the mirror separation and \( N_i \) the interference order (integer). \( \psi(\nu_i) \) is the phase shift due to the reflection on both mirrors in a round-trip while \( \phi_i \) is the Fresnel phase shift, which affects the laser beam in a single pass between the mirrors: for a TEM\(_{00}\) mode in a cavity with mirrors of equal curvature \( R \), \( \phi_1 = (p + q + 1) \arccos(1 - d/R) \).

We first discuss the effects of the dispersion in the refractive index of the air. We can determine the relation between the two frequencies \( \nu_0 \) and \( \nu_1 \) imposed by the resonance condition (1) by taking the difference

\[
\nu_1 \cdot n_1 / n_0 - \nu_0 = \frac{c}{2d \cdot n_0} \left( \Delta N - \frac{\psi_1 - \psi_0}{2\pi} + \frac{\phi_1 - \phi_0}{\pi} \right), \quad (2)
\]

where \( n_i, \psi_i \) stand for \( n(\nu_i), \psi(\nu_i) \) and \( \Delta N = N_1 - N_0 \).

For measurements in air we must know the refractive index \( n(\nu) \) in order to derive the frequency \( \nu_1 \) from Eq. (2). \( n(\nu) \) depends on several environmental parameters: temperature, atmospheric pressure, humidity, CO\(_2\) concentration. During each run, we estimated these quantities and used the results to find \( n(\nu) \), computed according to Ref. [12]. The final precision for \( n \), better than \( 2 \cdot 10^{-6} \), is limited essentially by the accuracy of the temperature and pressure data (< 2 °C and < 3 Torr respectively). The contribution due to the uncertainties of the humidity and of the CO\(_2\) concentration were completely negligible, and also the error (0.2 ppm) on the frequency \( \nu_1 \) initially measured with a wavemeter, did not affect the accuracy of \( n(\nu) \).

It is worth noting that the error for the ratio \( n_1 / n_0 \) is even lower than the absolute accuracy for \( n \): we found that for our frequencies \( \nu_0 \simeq 385 \) THz and \( \nu_1 \simeq 417 \) THz, it is better than \( 5 \cdot 10^{-9} \). This is the reason why we expressed the result in Eq. (2) as a function of \( n_1 / n_0 \). We will show below that the factor depending on \( n_0 \) in the second member of that equation can be calibrated with a dedicated procedure. In summary, the contribution of the refractive index of air to the final error for the frequency measurements was conservatively estimated to be less than 2 MHz.

Now we discuss how we managed to evaluate the uncertainty produced by the reflective phase shift of the mirrors. We used a confocal Fabry-Perot cavity \( (d = R) \). In this case, \( \phi_i \) is a multiple of \( \pi/2 \), therefore it is not strictly necessary to match the laser beam to the TEM\(_{00}\) mode of the cavity: simply, with both \( (p + q) \) even- and odd-order modes, the free spectral range (FSR) appears to be \( c/4d \) instead of \( c/2d \). However, if we manage to align the laser beam very well on the cavity axis, we observe that the odd modes are strongly suppressed for symmetry reasons and we recover the FSR = \( c/2d \). We took advantage of this property to optimize the alignment of the laser beams in the cavity, by minimizing the intensity of transmission peaks corresponding to odd modes: this ensures that the travelled distance \( d \) is the same for both lasers. For even modes, we obtain that \((\phi_1 - \phi_0) / \pi\)
is integer, therefore

\[ \nu_1 \frac{1}{n_0} - \nu_0 = \frac{c}{2d n_0} \left( N - \frac{\psi_1 - \psi_0}{2\pi} \right), \tag{3} \]

where the integer \( N = \Delta N + (\phi_1 - \phi_0)/\pi \) in our apparatus is typically near 8000.

In general, the reflective phase shift \( \psi \) is not zero and depends on the frequency of the light. If we assume that the dependence is linear in a frequency interval corresponding to the high-reflectivity range of the mirrors \([14, 15]\), then \( \psi(\nu') = \psi(\nu) + 2\pi\alpha(\nu' - \nu) \). The validity of the linear approximation is discussed in the following. From Eq. (3), we find that

\[ \nu_1 \frac{1}{n_0} - \nu_0 = \frac{c}{2d n_0} \left( N - \alpha(\nu_1 - \nu_0) \right), \]

and with a bland approximation,

\[ \nu_1 \frac{1}{n_0} - \nu_0 = \frac{c}{2d n_0} \frac{N + \epsilon}{1 + \frac{\epsilon}{2d n_0} \alpha} \simeq \mathcal{F} \cdot N, \tag{4} \]

where the factor \( \mathcal{F} \), implicitly defined by the last equation, has the meaning of an effective FSR. The error in the approximation is \( \epsilon = \alpha (\nu_1/n_0 - 1) \nu_1 \): it is expected to be negligible at our level of accuracy. To get an idea of its order of magnitude, let’s recall the operation of a multi-layer dielectric quarter-wave stack: for a typical structure of several alternating layers of high and low refraction, a reasonable value for \( \alpha \) is \(-3 \cdot 10^{-15} \text{ Hz}^{-1}\).

We describe the calibration procedure by which we can determine the factor \( \mathcal{F} \), necessary to compute the unknown frequency from Eq. (4). The idea is to tune the Ti:Sa laser to several different frequencies \( \nu_1 \) corresponding to the transmission peaks of the Fabry-Perot while its length is locked to the resonance value found for the trap frequency; the scanned range is 350 – 430 THz and we measure them with our commercial wavemeter (10 MHz resolution, 0.2 ppm quoted accuracy).

For each measurement, we know that the condition (4) has to be satisfied, with an integer \( N \) corresponding to the number of FSRS separating \( \nu_1 \) from \( \nu_0 \). In practice, with frequencies \( \nu_1 \) near \( \nu_0 \), it is possible to directly count the number of FSRs, find \( N \) and obtain a first estimate of \( \mathcal{F} \). This value, together with the integer condition for \( N \) and the wavemeter measurement, is used in Eq. (4) to determine a new \( N \) for a frequency \( \nu_1 \) farther from \( \nu_0 \), and to find a more accurate value of \( \mathcal{F} \). But more accurate values of \( \mathcal{F} \) allow us to extract \( N \) for frequencies ever more distant: by iterating the procedure, we find \( N \) for all the measured frequencies. We are then able to fit the function

\[ \nu_1 \frac{1}{n_0} = a + b \cdot N \]

to the whole set of \( \nu_1 n_1/n_0 \) acquired values, to find the parameters \( a \) and \( b \). \( a \) is compared to the known value of the transfer laser frequency \( \nu_0 \) to deduce the systematic error of the wavemeter, which was found to be 50 MHz at \( \nu_0 \simeq 385 \text{ THz} \). This estimation allows us to introduce a small correction to compensate for the bias of the wavemeter and determine \( \mathcal{F} \):

\[ \mathcal{F} = \frac{b \cdot \nu_0}{a}. \]

Note that the fitting procedure also allows us to check that Eq. (4) is satisfied in a large range of frequencies, and therefore gives an experimental support to the assumption of negligible non-linearity for the reflective phase shift in our spectral region.

A crucial element for the evaluation of the final uncertainty is the accuracy of the reference laser. As explained in Fig. 11, the calibrated frequency is provided by the coordinated operation of the transfer laser and the reference laser. They are diode lasers in an extended cavity configuration, with a Littrow-mounted grating for wavelength selection and piezoelectric stacks for fine tuning. The frequency of the reference laser is defined by a secondary frequency standard, namely the 5\text{S}_2/\text{D}_5/2\) two-photon transition in Rb atoms, at 778 nm [16]. In order to implement this standard, we lock the red emission of the laser by observing the blue fluorescence at 420 nm from the 6\text{P}_3/2 level [17]. Usually, we excite the 5\text{S}(F = 2) – 5\text{D}_{5/2}(F = 4) two-photon hyperfine component of \( ^8 \text{Rb} \) (nominally at 385284566.366 ± 0.008 MHz), because it is relatively intense and well separated from the contiguous hyperfine lines. A fast photo-diode followed by a radio-frequency amplifier is used to detect the beat signal between the transfer laser and the reference laser and its frequency is measured by a counter: so the absolute frequency of the transfer laser can be obtained.

In fact, to check the reliability of our frequency standard, we set up two laser systems, independently locked to the two-photon transition of Rb in separate cells, and compared their frequencies. We measured the beat frequency of the two lasers in different conditions: cell temperature, beam alignment, selected hyperfine component. In particular we changed the focusing of the laser beams inside the Rb cell, to observe the effects caused by the light shift and we also measured the frequency shift as a function of an external applied magnetic field, for testing the efficiency of our mu-metal shield. In normal conditions, we can safely estimate an absolute accuracy of the reference frequency better than 300 kHz.

Our Fabry-Perot resonator has a nominal free spectral range \( c/4d = 2 \text{ GHz} \) and a finesse 200 in the range 700 – 860 nm. The mechanical structure supporting the mirrors is a thick super-invar cylinder and the spacing can be finely adjusted by means of a piezo. At the beginning of each measurement run, both lasers are carefully aligned according to the procedure described above.

As explained in Fig. 11 we first obtain the resonance condition for the transfer laser, then we search for the
transmission of the Ti:Sa laser. It was not necessary to separate the two transmitted beams: we directly observe the sum of the two signals with the same detector. The feedback loop is robust, so for reasonable Ti:Sa intensities the cavity lock is not disturbed.

Once we have maximized the transmission for both lasers, we can use Eq. (4) and the performed calibration to find the accurate value of the Ti:Sa laser frequency. By repeating the measurement several times, we found that the reproducibility is better than 2 MHz, mostly limited by the localization of the transmission peak for the Ti:Sa laser. We measured the number of trapped atoms for $^{209}\text{Fr}$, $^{210}\text{Fr}$, and $^{211}\text{Fr}$, as a function of the trapping-laser frequency (Fig. 2). Due to a limited Fr production at that run-time, we did not operate in optimal conditions but the signal was high enough to determine the frequency corresponding to the maximum number of atoms. Then we measured the absolute frequencies corresponding to the peak signal for the three isotopes. The data for the calibration procedure ($33 \nu_1$ frequencies in total) were acquired at the cavity lengths corresponding to the three trapping frequencies and allowed to obtain the most accurate calibration of the $F$ factors ($\Delta F < 400 \text{ Hz}$), which means a contribution of about 3 MHz for the error on the final frequencies.

We report in Table I the results of our measurements, along with the frequencies of the repumping transitions, measured with the wavemeter after calibration with the secondary frequency standard. If we assume that the trapping laser detuning corresponds to $5\Gamma \pm 2\Gamma$ (typical values for a MOT), we can deduce the frequency of the atomic lines involved in the trapping process (Table II). In order to confirm the correctness of this approach, we used it to measure the frequency of the $5S(F = 3) - 5P_{3/2}(F = 4)$ transition in a $^{85}\text{Rb}$ MOT. After applying the detuning correction, we measured 384229.250(18) GHz, compatible with the most accurate value presently available, namely 384229.2428(4) GHz.$^{15}$

In conclusion we presented accurate measurements of the laser-trapping frequencies of $^{209}\text{Fr}$, $^{210}\text{Fr}$, and $^{211}\text{Fr}$. Most of the results displayed in Table I are new; in cases where previous measurements are available, our results agree with existing data, but feature an improved accuracy. The interferometric method, much simpler than other spectroscopic techniques, allowed us to reach a precision of 7-9 MHz with samples of about 100 atoms. The experimental setup is presently being upgraded by operating in vacuum and using the virtual mirrors method.$^{11}$ and we expect an improvement of about a factor 2 in the final accuracy.

The importance of precision measurements on Fr, particularly if available on several different isotopes, lies in the interest of this heavy atom for testing fundamental symmetries: both parity violation and EDM require a theoretical analysis of the atomic structure that must be checked against experimental data. Accurate absolute measurements are also very useful in the design of future experiments. Therefore the results presented here are an important step in this direction.

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FIG. 2: Number of trapped atoms as a function of the Ti:Sa laser scan frequency $\nu_1$, for the three isotopes $^{209}\text{Fr}$, $^{210}\text{Fr}$, and $^{211}\text{Fr}$. Each scan begins with no trapped atoms: two of the curves are down-shifted for clarity.

| Isotope | Trapping laser | Repumper | Trapping transition |
|---------|----------------|----------|---------------------|
| $^{209}\text{Fr}$ | 417415.087(8) | 366897.428(50) | 417415.125(17) |
| $^{210}\text{Fr}$ | 417412.448(7) | 366898.698(50) | 417412.486(17) |
| $^{211}\text{Fr}$ | 417412.627(9) | 366895.568(50) | 417412.665(18) |