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# Microwave-Optical Double Resonance on a Single Laser-Cooled $^{171}Yb^{+}$ Ion.

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**Abstract.** – A single ion whose energy levels are hyperfine split, $^{171}Yb^{+}$, was electrodynamically trapped and laser cooled. The residual energy of both its driven motion and its secular motion was 11 μeV. Radiofrequency-optical double-resonance excitation yields spectra whose 40 Hz wide lines are determined so far by the duration of the ion's interaction with the 12.6 GHz radiation. These results prove that a microwave clock is feasible with its fractional systematic uncertainty not exceeding $10^{-16}$.

The dynamic confinement of ions in Paul and Penning traps has been shown to provide the ideal means for precision spectroscopy on these ions, aimed in particular at the representation of a time and frequency standard. The times of ion storage are virtually unlimited, and with the trap placed in ultra-high vacuum there are no collisions with background gas. The ensuing long times of interaction with applied electromagnetic fields may give rise to the recording of extremely narrow lines. Trapped clouds of several ion species have been used to demonstrate atomic clocks [1-5], and it was shown that their short-term performance can exceed that of a primary Cs standard. In addition, and according to its inherent lack of gross perturbations, a clock based on an ion trap shows long-term stability that may be superior to the stability of a hydrogen master [6]. The major source of systematics in these demonstrations was the residual ion motion which shifts the signal frequency and limits the ultimately achievable accuracy of the standard frequency. This drawback is eliminated by laser-cooling the ions [7, 8]. Although the efficiency of laser-cooling an ion cloud is poor, this technique is routinely and successfully applied on a single trapped ion [9]. However, for the precise control of a standard frequency, a narrow and therefore weak signal-driven transition has to be employed whose resonance is hardly detected, with sufficient signal-to-noise ratio, in the concomitant single-ion fluorescence on

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that signal line. This problem is readily overcome by observing the individual acts of absorption of the signal radiation by the detection of additional laser-driven resonance scattering from the ion that is switched off and on when the ion is excited and de-excited, respectively, on the signal line [10, 11]. This scheme of quantized fluorescence amounts to the detection of signal absorbtion with unit quantum efficiency. So far the potential of this technique has been demonstrated with a narrow *optical* transition [12]. Although standard radiofrequencies cannot compete with optical ones as far as their potential for fractional stability is concerned, their preparation remains nonetheless important for applications, since radiofrequency cycles are readily countable. Recently, a precise measurement of the ground-state hyperfine splitting on a string of few laser-cooled $Hg^+$ ions has been performed whose result was a 250 mHz wide resonance [13].

In this letter we report the detection of microwave-optical double resonance on a *single* laser-cooled $^{171}Yb^+$ ion. Trapping a single ion in a field node thoroughly eliminates potential perturbative interaction with neighbouring ions, unlike an experiment on several ions in a linear trap, where the light of one of the ions was singled out with the detection [14]. For some time, the ytterbium ion has been considered a candidate for the control of a standard frequency [15]. Optical transitions to long-living metastable states of the ytterbium ion have been studied recently [16, 17]. In particular, the mass-171 species of this ion stands out, since it shows narrow resonances in the microwave as well as in the optical regime. The ground-state hyperfine splitting of $^{171}Yb^+$ has been used previously as the basis of an atomic clock [3].

Figure 1 shows the lowest relevant levels and the corresponding transitions of $^{171}Yb^+$. Its ground state $S_{1/2}$ splits into states $F = 1$ and $F = 0$ with 12.6 GHz separation by the hyperfine interaction, whereas the $P_{1/2}$ resonance level splits by 2.1 GHz. Uninterrupted, continuous scattering of fluorescence light requires the simultaneous illumination of the ions with laser light at 369 nm *and* 2.44 μm in order to avoid pumping the ion into the $D_{3/2}$ level. With a cloud of trapped ions, the first-order Doppler effect spectrally distributes the resonances over a range of some 2 GHz, and the hyperfine splitting in the excited $P$ and $D$ states is not resolved. Thus, microwave-optical double resonance is obtained when the laser at 369 nm is tuned to one of the hyperfine components of the resonance line, and the scattered light is detected, the additional laser at 2.44 μm is tuned to undo optical pumping into the $D_{3/2}$ state, and microwave radiation at 12.6 GHz couples the ground-state hyperfine components. With a single trapped $Yb^+$ ion and laser cooling, this simple scheme is no longer

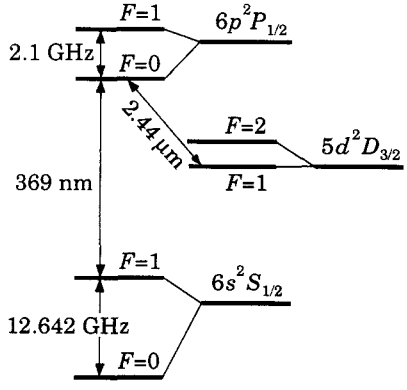


Fig. 1. – Relevant levels and transitions of $^{171}Yb^+$. Zeeman splitting not shown.

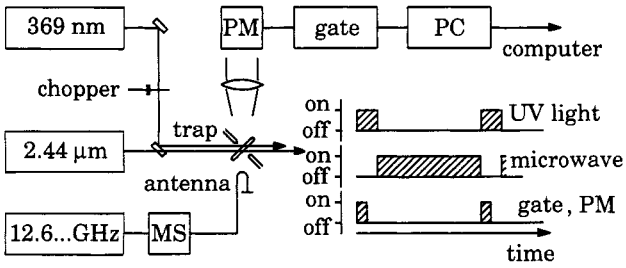


Fig. 2. – Experimental set-up for single-ion laser cooling and microwave-optical double resonance. Time sequence of ion irradiation and fluorescence detection. PM: photomultiplier, MS: microwave switch, PC: photon counter.

feasible, since the resolved hyperfine levels admit various modes of optical pumping and selective excitation.

Figure 2 shows the experimental set-up used for the single-ion double-resonance experiment. The $S_{1/2}$, $F = 1 \rightarrow P_{1/2}$, $F = 0$ resonance line is cyclically excited by scattering light of the 369 nm laser that is slightly down-tuned from this resonance for optically cooling the ion. Strong microwave radiation at 12.6 GHz emitted from a hairpin antenna irradiates the ion and excites the $\Delta m_F = 0$ transition between ground-state hyperfine levels such that optically pumping the ion occasionally into the $F = 0$ ground state in the far wing of the $F = 1 \rightarrow F = 1$ resonance line is immediately undone. Optical pumping the ion into particular Zeeman sublevels is eliminated by setting the light polarization at an oblique angle with the weak magnetic field at the ion location. The microwave interaction should take place when no UV light is present in order to avoid power broadening and light shifts: the microwave signal is applied for 20 ms and switched off for the subsequent 4 ms, whereas the UV light alternates with it. The 2 ms gating of the photomultiplier is synchronized with the appearance of the UV light, whose irradiating the ion during the remaining 2 ms ensures the eventual preparation of the ion in the $S_{1/2}$, $F = 0$ ground state before interaction with the microwave signal starts anew. Thus, the scattered light probes the microwave-induced absorptive ion transitions into the $S_{1/2}$, $F = 1$, $m_F = 0$ level. The counting rate of the scattered light is accumulated for typically 4*s*, and the microwave frequency is stepwise scanned, mapping the population of the $S_{1/2}$, $F = 1$, $m_F = 0$ level that appears as a result of the microwave excitation, *vs.* the microwave frequency scan.

An individual $^{171}Yb^+$ ion is loaded into the 1 mm wide trap when a faint atomic beam from an oven loaded with isotope-enriched Yb crosses the trap and is ionized by electron impact. The presence of no more than one ion is verified by the observation of dark intervals in the resonance fluorescence at 369 nm which occur occasionally due to quantum jumps of the ion from the $D_{3/2}$ level to the $D_{5/2}$ level that is almost uncoupled with the laser light. These jumps seem to be induced by collisions with hot background gas during the loading process, while the ytterbium oven is heated. As soon as an individual ion is prepared, the oven is switched off, and the trap is operated at the base background pressure, $2 \cdot 10^{-10}$ mbar.

The localization and cooling of the ion in the trap was first explored on an ion of the even isotope $^{174}Yb^+$. Figure 3*a*) shows an excitation spectrum of the UV resonance scattering of a single $^{174}Yb^+$ ion obtained after the initial cooling process. The spectrum shows the characteristics of high-index phase modulation by driven harmonic vibration [18-20] that results in a U-shaped spectral response. The transition from laser cooling to heating is indicated by the sharp drop of the fluorescence rate at zero detuning of the UV laser. Upon heating the ion, the first-order Doppler width of the resonance line becomes so large that

resonance scattering is drastically reduced. In order to determine the cold ion's residual kinetic energy of its driven vibration, the line shape of phase-modulated emission [16]

$$I(\omega) = \sum_{n=-50}^{n=50} \frac{J_n^2(p)}{(\Delta + n\omega_a)^2 + \Gamma_{\text{sat}}^2/4} \tag{1}$$

was used for fitting the data, and the modulation index $p$ was adjusted. Here, $\omega_a$ is the frequency of the driving field, 9.5 MHz $\times 2\pi$, $\Gamma_{\text{sat}}$ denotes the power-broadened natural linewidth of the $S$-$P$ resonance line, $\Delta = \omega - \omega_0$ the detuning of the light off-resonance, and $J_n(p)$ is the Bessel function of the first kind of order $n$. The resulting modulation index is $p = 18$ which corresponds to the kinetic energy $1.8 \times 10^{-3}$ eV. Note that this residual energy corresponds to the ion's driven micromotion in the trap which persists due to a d.c. offset potential that shifts the ion off the trap's r.f. centre. This energy is minimized by compensating the offset potential by a d.c. field superimposed by means of additional electrodes [21, 22]. The result of this compensating procedure is shown in the excitation spectrum of fig. 3*b*). This spectrum is of Lorentzian shape, and the residual energy is 4.5 μeV. In order to determine this value, eq. (1) has been fitted to the data of fig. 3*b*) with the linewidth *and* the modulation index $p$ taken as free parameters. This fitting is sensitive to $p$ (and to the ion energy) since half the linewidth nearly equals the trapping frequency. The result is $p_{\text{opt}} = 0.9$. Smaller values seem compatible with the data, whereas larger values ($p \gtrsim 1.0$) are *not*. Although this procedure also works on an ion of the odd isotope $^{171}Yb^+$, the light flux required for the recording of a satisfactory excitation spectrum is higher. Therefore, these spectra are affected, to some extent, by power broadening, see fig. 3*c*). When comparing this line shape with eq. (1) one concludes that the linewidth is entirely attributed to power broadening, and the kinetic energy of the residual ion motions is 11 μeV.

After having loaded a single $^{171}Yb^+$ ion into the trap, eliminated the micromotion, and cooled the ion, the double-resonance interaction is activated by sequential application of UV light and microwave radiation. Figure 4*a*) shows the counting rate of the photomultiplier signal *vs.* microwave tuning for 20 ms long interaction of microwave and ion per data point.

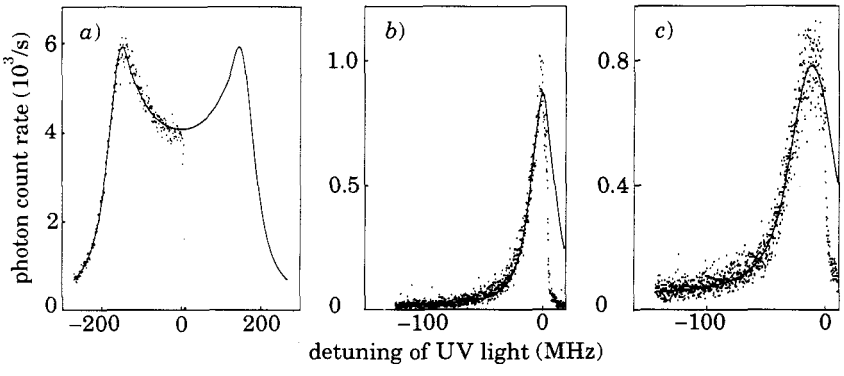


Fig. 3. – Excitation spectra of single $Yb^+$ ion. *a*) $^{174}Yb^+$ subject to strong residual micromotion, *b*) $^{174}Yb^+$ micromotion compensated by d.c. potential on auxiliary electrodes, *c*) $^{174}Yb^+$ ion, micromotion compensated.

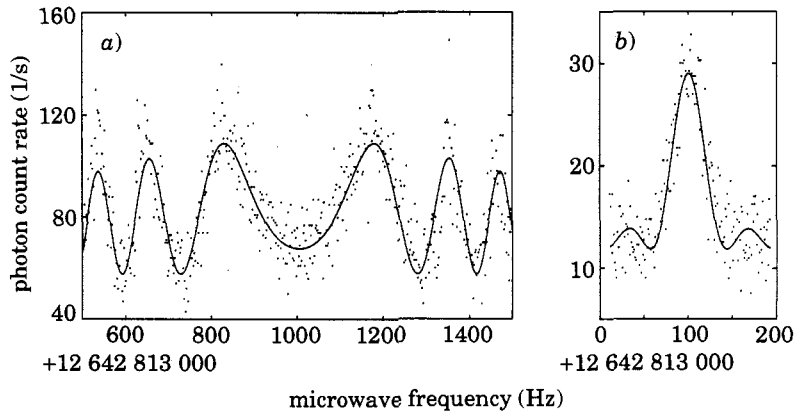


Fig. 4. – Microwave-optical double-resonance spectra of the $\Delta m_F = 0$ ground-state hyperfine transition: *a*) high Rabi frequency, *b*) low Rabi frequency, matched to the ion interaction with the signal wave for the generation of a narrow single-peaked line.

As expected, the line shape is determined by the finite-interaction time and obeys the Rabi formula

$$P(\Delta_m) = \frac{\Omega^2}{\Delta_m^2 + \Omega^2} \sin^2\left[\frac{1}{2}\tau(\Delta_m^2 + \Omega^2)^{1/2}\right], \tag{2}$$

with the microwave Rabi frequency $\Omega$ and detuning $\Delta_m$ from the hyperfine line, and where $\tau$ denotes the interaction time. The appearance of side lobes in the observed line shape (cf. fig. 4*a*)) is caused by the high Rabi frequency in this measurement. Matching Rabi frequency and interaction time such that $\Omega\tau < \pi$ allows one to produce a narrow, single-peaked line structure. Upon reducing the Rabi frequency to 157 Hz, the line shape shown in fig. 4*b*) was obtained. The FWHM of this line is 40 HZ, determined by the interaction time $\tau$. In principle, this interaction time is limited only by relaxation among the optically pumped hyperfine levels provided that the ion remains cooled during the entire time of the measurement. For the data shown in fig. 4*b*) their total acquisition took about 15 minutes. During this time the ion undergoes several collisions with molecules of the background gas which spuriously heat the ion. Since optical pumping into the $S_{1/2}$, $F = 0$ level is rather fast during the probe intervals—*i.e.* the periods of time when the laser at 369 nm is switched on for the excitation of resonance scattering and repumping the ion, cf. fig. 2—, the effective time of interaction with the light is short, and optical cooling does not suffice to overcome the residual collisional heating during more extended interaction and detection cycles. However, this limitation is not of a principal nature, since an extra cooling interval could be sandwiched between every two successive cycles of probing and detection.

The observed spectra demonstrate that well-resolved microwave optical double-resonance spectra are obtainable from an individual trapped and laser-cooled ion, although linewidth and signal-to-noise ratio do not yet match corresponding data from double-resonance detection on ion clouds. However, a 100 mHz linewidth of the signal seems thoroughly achievable with $S/N \simeq 5$, which would give rise to frequency stability (or «precision») on the order $10^{13}$ with 3 min intergation time. On the other hand, the long-term stability and the *accuracy* of a standard frequency controlled by the resonance of a single cold

ion are expected to far exceed the characteristics of a standard frequency based on an ion cloud. To this end, the measured frequency of the ground-state hyperfine splitting is to be corrected by extrapolating it to zero magnetic field, a procedure which is well known from primary atomic clocks and which requires the measurement of the $\Delta m_F \pm 1$ transitions with sufficient precision [3]. Systematic frequency shifts are small, since the cold ion, when localized in the node of the trapping field, is neither afflicted by field inhomogeneities nor by motional shifts. For our experimental set-up, we estimate that the largest uncertainty of the resonance frequency is due to the Stark shift of the hyperfine splitting. From the residual motion of the ion we calculate its maximum excursion in the trap and the corresponding electric field. Using the known Stark shift coefficient [21] we estimate an upper limit of the fractional Stark shift, $3 \cdot 10^{-15}$. However, improved optical cooling making use of Raman transitions would reduce the residual kinetic energy below the Doppler limit to less than 10 neV, as has been proved with a comparable system, and diminish the fractional Stark shift by at least three orders of magnitude. At the kinetic energy of the ion so far achievable, the fractional second-order Doppler shift is $10^{-16}$. From the measured pressure dependence of the hyperfine splitting [23], we obtain the fractional pressure shift, $5 \cdot 10^{-17}$. The black-body radiation at the ion's location and at ambient temperature shifts the signal resonance by $-1.6 \cdot 10^{-15}$, which could be reduced when using a cryogenic trap, or else included as a correction.

The reported observations demonstrate that microwave-optical double-resonance spectroscopy on an individual cold ion shows promising features for its use as the basis for an improved microwave atomic clock. The reported experimental data indicate that such a frequency standard can be obtained with absolute inaccuracy on the order of $10^{-16}$, two orders of magnitude better than currently used primary standards.

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