Experimental Demonstration of a Terahertz Frequency Reference based on Coherent Population Trapping

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A novel protocol of interrogation based on coherent population trapping in an N-level scheme atomic system leads to dark resonances involving three different photons. An ensemble of several hundreds of radiofrequency-trapped ions is probed by three lasers simultaneously locked onto the same optical frequency comb, resulting in high-contrast spectral lines referenced to an atomic transition in the THz domain. We discuss the cause of uncertainties and limitations for this method and show that reaching a sub-kHz resolution is experimentally accessible via this interrogation protocol.

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laser-atom interaction on this transition, it can play a major role in the internal state dynamics provided that a resonance condition is fulfilled \[16\]. This condition can be extrapolated from two-photon Λ-scheme dark resonances \[17\] and it writes

\[
\Delta_R = \Delta_B - \Delta_C - \delta_C
\]

where \(\Delta_R, \Delta_B, \Delta_C\) are the detuning of the three lasers and \(\delta_C\) is the light-shift induced by the quadrupole coupling on the 729 nm transition \[16\] (see Supplemental Material for further explanation). The trapping state is a coherent superposition of the three stable and metastable dressed states that is not coupled by laser excitation and once trapped in this state, the ions do not emit any photon, provided the trapping state is stationary.

When fulfilled, the three-photon resonance condition implies a strong relation between the three laser frequencies

\[
\omega_R + \omega_C - \omega_B + \delta_C = \omega_{THz}
\]

with \(\omega_{THz}\) the frequency of the magnetic dipole transition between \(3D_{3/2}\) and \(3D_{5/2}\), which appears as the atomic reference. The frequency \(\omega_{THz}\) is 1.82 THz in Ca\(^+\) and its absolute value is known with a ±8 Hz uncertainty through Raman spectroscopy on a single trapped ion \[18\] [19]. Considering the typical intensity and detuning for the laser at 729 nm, \(\delta_C\) ranges 1 to 100 Hz. In the following, we present the experimental conditions of observing a three-photon dark resonance in the fluorescence of a cloud of trapped ions. We review the major effects which contribute to the line-width, frequency shift and contrast of the dark line as there are the Doppler effect, the Zeeman effect and power-induced effects.

In the experiments presented here, the three lasers co-propagate along the symmetry axis of a linear quadrupole RF-trap (see Fig. 1b) and the effective wave-vector \(\Delta \vec{k}\) is the one of the magnetic dipole transition \(k_{THz} = 2\pi/\lambda_{THz}\) with \(\lambda_{THz}\) the \(3D_{3/2}/3D_{5/2}\) transition wavelength, equal to 165 μm. The quadrupole trap is described in \[21\] [21], its main characteristics are an inner radius of 3.93 mm for a rod radius of 4.5 mm \[22\] and an RF trapping frequency of 5.2 MHz. For the work presented here, it is operated with an RF-voltage difference between neighboring rods of 826 V \(pp\) (Mathieu parameter \(q_x = 0.24\)). The Doppler-laser cooling drives the ion cloud from a gas, through the liquid, to a crystal phase \[23\] with a temperature estimated to be of the order of 10 mK. Once in the liquid and crystal phase, the ion cloud forms an ellipsoid \[24\] [24] with a diameter ranging from 80 to 280 μm and a length ranging from 120 to 740 μm for a number of ions comprised between 40 and 2750 (see Fig. 1c). The 397 nm and 866 nm lasers have an elliptical section, with an aspect ratio of 2 and a mean-squared diameter at the position of the cloud equal to 4.0 mm and 4.7 mm, respectively. The laser intensity and wave-vectors can be considered as uniform all over the ion cloud. The 729 nm laser has the smallest size with a waist diameter measured to 300(±20) μm. It is
still larger than the largest of the cloud diameters but its intensity is not uniform over the largest clouds. This geometric configuration does not cancel the Doppler effect which broadens the three-photon dark lines by 20 kHz (FWHM) for a sample at 10 mK, but it eliminates any broadening induced by finite interaction time, which is an identified limitation on two-photon CPT line-width when observed on an atomic beam or a gas in a cell.\cite{12,20}

For the observation of the dark lines, the three involved lasers are admitted continuously on the cloud. They are phase-locked through a commercial offset-free optical frequency comb \cite{22} and their frequency is measured with an uncertainty in the kHz range (see Supplemental Material and Fig.\textsuperscript{1}(d)). A magnetic field of the order of 1 Gauss is applied and the three laser polarisations are linear, perpendicular to the trap axis and nearly perpendicular to the local magnetic field. Spectra as the one shown on figure\textsuperscript{1}(e) are observed when collecting the photons emitted at 397 nm on the $4P_{1/2} \rightarrow 4S_{1/2}$ transition while the frequency of the $R$-laser is scanned. Typical laser powers are 10 to 20 mW at 397 nm ($P_B$), 0.5 to 5 mW at 866 nm ($P_R$) and 5 to 25 mW at 729 nm ($P_C$). A maximum contrast of 22\% is observed for the dark line and the bright atoms are still laser-cooled and maintain the ion cloud dynamical stability by sympathetic cooling of the dark atoms (see Fig.\textsuperscript{1}(c)). This cooling is efficient enough to keep the ion cloud in a liquid phase all over the frequency scan, which offers the great advantage of keeping the number of trapped ions constant during the whole recording. The splitting of the dark line into several pairs of lines is due to the local magnetic field which lifts the degeneracy of the Zeeman sub-states. We identify each transition according to its Zeeman shift $\delta f$ on the THz transition $m_{THz} = m_{THz} \mu_B B$ (see Supplemental Material for the relation between the Zeeman shift and the electronic level properties). The stability of each line center frequency is limited by the long term fluctuations of the local magnetic field measured to 0.4 mG (pk-pk). It contributes an uncertainty proportional to $m_{THz}$ and of the order of 1 kHz. The short term fluctuations of the total local magnetic field are measured to 6 mG (pk-pk) and are responsible for a $m_{THz}$-dependent broadening of the order of 20 kHz (pk-pk).

In a first step, to prove that these dark lines result from the 3-photon process and are referenced to the THz transition, we focus on the dark line defined by $m_{THz} = -13/5$ because of its high contrast. The $R$-laser frequency $\omega_R$ is scanned for different values of $\omega_C$ in an interval of 16 MHz, while $\omega_B$ is kept constant. The frequency step is 1 kHz and signal is accumulated for 150 ms at each step. Each scan is reproduced 4 times and averaged. Each observed line profile is fitted to a Lorentzien profile and the center of the line $\omega_R^c$ is pointed with an uncertainty of the order of 1 kHz (1 \sigma) conditioned by the frequency step and the signal to noise ratio. The frequency combination $\Delta_{RCB} = \omega_R^c + \omega_C - \omega_B$ is expected to give access to the magnetic transition frequency, once the experimental shifts removed, the Zeeman shift being the largest one identified. Exploiting several multiline spectra as the one of Fig.\textsuperscript{1}(e), the Zeeman shift $\delta f(-13/5)$ is evaluated with an uncertainty of $\pm 6$ kHz dominating the total uncertainty of the THz frequency. As shown on Fig\textsuperscript{2} the Zeeman corrected transition frequencies are shifted from the 3D$_{3/2}$ to 3D$_{5/2}$ transition frequency $f_{DD}$ of reference \cite{10}, by values which evolve between $+5$ and $-15$ ($\pm 6$) kHz. These shifts are plotted against the detuning $\Delta f_R^Z = \omega_C - \omega_{D_3/D_5/2} + \delta f(-13/5)$ with $\omega_{D_3/D_5/2}$ the frequency of the atomic transition measured in $\omega_R^c$ and $\delta f(-13/5)$ the Zeeman shift on the $R$-transition. This plot shows a correlation between the shifts and the detunings that cannot be explained by any drifts in the experimental set-up, neither by the coupling on the quadrupole transition $\delta C$.

When the 729 nm laser power is increased, the frequency shift of the $m_{THz} = -13/5$ line reaches a stationary value for a laser power $P_C$ larger than 10 mW. This is consistent with the model developed to explain the three-photon CPT by a laser mediated two-photon CPT \cite{16} and can be interpreted as a minimum coupling strength requirement to reach an effective CPT. We further use the analogy with two-photon CPT to track laser-induced shifts. In this scheme, the strongest laser coupling is the one driving the $R$-transition. The observed dependence of the THz-frequency shifts with the power $P_R$ for different Zeeman transitions shows that several power-induced effects add up to build a global frequency shift. Furthermore, the extrapolations of the shifts to zero laser power do not cross the same value, ranging 5 to 15 kHz and speak also for non $R$-laser power induced effects, like the
one identified above. There are several causes for laser-induced shifts in two-photon CPT and some are known to depend on the one-photon detuning and on the ratio of the effective intensity responsible for the laser coupling on the two legs of the Λ-scheme. In the case of a three-photon CPT, the corresponding one-photon detuning is $Δ_R$ and the corresponding intensity ratio $R_I$ is $\left(\Omega_R Δ_C/(\Omega_B Δ_C)\right)^2$ with $\Omega_R$, $\Omega_B$, $\Omega_C$ the Rabi frequencies on each transition. On Figure 3 we plot these measured frequency shifts against the modified ratio $R_I \times |\Delta_R^2|$ for each transition $m_{THz}$. To that purpose, we take into account the differences in $\Omega_R$, $\Omega_C$, $Δ_R^2$ and $Δ_Z^2$ for the transition between different Zeeman sub-levels ($Δ_Z^2$ is the same for all the shown transitions—see Supplemental Material). These plots group the shifts measured for 8 different transitions on 4 different lines, meaning that the foreseen dependence matches only partially the experimental observations. The interplay of the three laser coupling parameters to explain the measured shift deserves a more detailed study to identify the condition for control and/or cancellation of these shifts. One solution to reduce the impact of the power-induced shift may be the implementation of pulsed Ramsey-type protocols which prove to be very efficient to reduce power-induced effect on two-photon CPT-clocks.

To quantify the metrological performance of the 3-photon CPT dark line as a THz reference, let’s assume these shifts are under control and focus on the line-width, the absolute signal level as well as the contrast of the dark line. We recall that within the present experimental set-up, each line is broadened by the Doppler effect (estimated to a minimum of 20 kHz FWHM) and by a fluctuating Zeeman effect (estimated to $8.4 \times m_{THz}$ kHz pk-pk). Furthermore, in the range of our experimental parameters, the observed power-induced broadening is only due to the coupling on the R-transition. The narrowest observed dark lines (line-width of 45 kHz for $P_R = 0.7$ mW) are the one with the smallest coupling on the R-transition, identified by $|m_{THz}| = 11/5$ and 13/5 (see Table 1 in Supplemental Material). The maximum dark line contrast, reaching 25%, is observed for the lines $|m_{THz}| = 13/5$ and 21/5 and they are the ones with the largest coupling on the C-transition. These two independent conditions point $|m_{THz}| = 13/5$ as the transition of the best contrast/broadening compromise in the context of our present experimental set-up.

Coming back to the results of Fig. 2 where the $m_{THz} = -13/5$ dark line is observed for different frequencies of the C-laser, we can plot the line-width and contrast for these five different sets of $\{ω_C, ω_B\}$ (see Fig. 4). The data show a dependence of the line-width and contrast with the effective value of the detuning $Δ_R$ for which the CPT occurs while all the laser powers are kept constant. We think this effect depends on the relative position of the dark line in the broader fluorescence spectra profiles of the trapped ions, due to a competition with the strong transitions involved in laser cooling. Further inquiries are required to identify the best condition to reach the contrast/line-width optimum but these curves open very positive perspectives as we find in the same detuning range the smallest shifts, the largest contrast and the smallest line-width.

Considering that the Doppler broadening can be cancelled by obeying the phase matching condition $k_B - k_R - k_C = 0$, or eventually by a Lamb-Dicke effect on the THz-wavelength scale, the signal over noise ratio could be increased with a larger number of ions building the dark line. When varying the number of trapped ions we recall that within the present experimental set-up, each line is broadened by the Doppler effect (estimated to a minimum of 20 kHz FWHM) and by a fluctuating Zeeman effect (estimated to $8.4 \times m_{THz}$ kHz pk-pk). Furthermore, in the range of our experimental parameters, the observed power-induced broadening is only due to the coupling on the R-transition. The narrowest observed dark lines (line-width of 45 kHz for $P_R = 0.7$ mW) are the one with the smallest coupling on the R-transition, identified by $|m_{THz}| = 11/5$ and 13/5 (see Table 1 in Supplemental Material). The maximum dark line contrast, reaching 25%, is observed for the lines $|m_{THz}| = 13/5$ and 21/5 and they are the ones with the largest coupling on the C-transition. These two independent conditions point $|m_{THz}| = 13/5$ as the transition of the best contrast/broadening compromise in the context of our present experimental set-up.

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from 60 to 400 ions, with all other parameters fixed, we observe no variation of the THz-frequency shift, neither of the dark line width. As the magnetic field fluctuations can be actively reduced by a factor 50 and the sensitivity to these fluctuations can also be reduced by using the transitions $m_{\text{THz}} = \pm 1/5$ if other laser polarisation and propagation directions are permitted by the set-up, a line-width in the kHz range, or lower, together with a large contrast seems very accessible. It would enable to access a resolution in the $10^{-9}$ range, which is the state of the art in the THz precision spectroscopy [36, 37]. In a system without any experimentally induced decoherence, on can show that sub-kHz line-widths can be observed [11]. Nevertheless, with a 25% contrast and an average fluorescence signal of 4000 counts/ms which is the typical value for the fluorescence of one thousand trapped ions in the optimum condition for a narrow dark line, the signal to noise ratio at 1 ms reaches 16. Even with a kHz line-width, such a large signal to noise ratio allows the resolution to be increased to the $10^{-11}$ range by averaging data over seconds.

This new 3-photon CPT has a large potential for high-resolution spectroscopy. Very similar to 2-photon CPT, the interrogation protocol depends on numerous parameters that have to be further explored. The originality of our approach allows not only to access an insufficiently explored and very promising spectral domain but also to implement a Doppler-free technique which opens the route to its implementation on a large variety of atomic systems.

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