Direct spectroscopy of the $^{2}S_{1/2} - ^{2}P_{1/2}$ and $^{2}D_{3/2} - ^{2}P_{1/2}$ transitions and observation of micromotion modulated spectra in trapped $^{40}\text{Ca}^+$

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Abstract
We present an experimental scheme to perform spectroscopy on ions and atoms with a lambda-level structure. By rapidly switching lasers between both transitions, we circumvent the complications of both dark resonances and the ac-Stark effect. We demonstrate the scheme on the $^{2}S_{1/2} - ^{2}P_{1/2}$ and $^{2}D_{3/2} - ^{2}P_{1/2}$ transitions in $^{40}\text{Ca}^+$ and, within a measurement time of 10 min, extract the centre frequencies of both dipole transitions with a statistical uncertainty on the order of 200 kHz. We also apply this method to directly observe the micromotion modulated fluorescence spectra of both transitions and for the first time measure the dependence of the micromotion modulation index on the trap frequency for both transitions.

Keywords: spectroscopy, trapped ions and atoms, micromotion

Introduction
Trapped ions provide an excellent experimental platform for the measurement of atomic properties such as transition frequencies, branching fractions and lifetimes of atomic energy levels. Trapped ions can be cooled easily into the millikelvin regime and are well isolated from their environment. In addition, all relevant experimental parameters such as trap frequency, magnetic and electric fields are well controlled. Because of these features, trapped ions are particularly well-suited to implement atomic clocks [1] and a large body of work has been carried out to study dipole forbidden narrow atomic transitions [2, 3]. However, outside the context of atomic clocks, absolute frequency measurements of dipole allowed transitions are also of interest. For example, in the astrophysics community, spectra from galactic objects stem mainly from dipole allowed transitions in ions and it is desirable to compare them to precisely measured laboratory values [4, 5].

A serious complication in performing spectroscopy of commonly used trapped ions and atoms is that the excited state does not only decay into the ground state but also into metastable levels (see figure 1 as an example). For instance, experiments with Ca$^+$, Ba$^+$, Yb$^+$ and Sr$^+$ ions require additional repumping lasers to depopulate those so-called dark states. However, the presence of the repumping laser strongly modifies the spectrum of the primary spectroscopy laser applied between the ground and the excited state. To take those uncertainties into account, one might resort to modelling the spectra with Bloch equations. For this the laser linewidth, intensity and polarization of both beams need to be either precisely controlled or extracted in addition to the magnitude and orientation of the magnetic field [6].

Moreover, changing the frequency of the spectroscopy and repumping lasers during the spectroscopy measurements modifies the cooling dynamics and influences the effective profile of the spectrum. This complication is present even in the case of two level systems. To circumvent this complication, experiments on Mg$^+$ ions, with an essentially two-level
that couple between states are shown for \( ^{40}\text{Ca} \) probability \( p \) to either the ground state, \( |g\rangle \), or the metastable state, \( |d\rangle \), with probability \( p \) and \( 1 - p \), respectively. The wavelengths of the lasers that couple between states are shown for \(^{40}\text{Ca}^+\).

structure, use sympathetic cooling of additional ions to maintain low temperature throughout the experiment \([7, 8]\). Another possibility is to first cool the ions and apply the spectroscopy laser only for a short duration, as carried out in \( \text{Ca}^+ \) [9].

In this work, we present an experimental scheme that circumvents the complications of dark resonances, ac-Stark shifts and cooling dynamics. Thus, our technique eliminates the most important systematic effects present in typical measurements in lambda-systems. We apply the scheme to a trapped linear chain of \(^{40}\text{Ca}^+\) and perform spectroscopy of the \(^2S_{1/2} \rightarrow ^2P_{1/2}\) and \(^2D_{3/2} \rightarrow ^2P_{1/2}\) transitions. Instead of controlling all the parameters of the eight-level Bloch equations to extract the line centre, we only need to control the polarization of the spectroscopy laser while scanning its frequency. The scheme relies on fast photon counting and rapid switching of laser pulses similar to the one presented for the purposes of measuring the branching fractions in [10]. The method is applicable to ion and atom species with a lambda-level scheme, provided that the metastable state has a significantly longer lifetime compared to the excited state. This shows that for \( R_0/\gamma \ll 1 \) (low saturation) and \( (1 - p)R_0T \ll 1 \) (short probe duration), the number of photons detected in a duration \( T \) is directly proportional to the coupling rate of the probe laser, \( R_0 \). Therefore, for a small duration \( T \), we are able to perform spectroscopy of the transition with no repumping laser and without perturbations from the cooling dynamics.

Similarly, we analyse the spectroscopy of the \( |d\rangle \) to \( |e\rangle \) transition. A probe laser couples these two states with the rate \( R_0 \). The experimental scheme is similar to the case of the spectroscopy of the \( |g\rangle \) to \( |e\rangle \) transition except that we do not directly detect photons scattered from the \( |d\rangle \) to \( |e\rangle \) transition. However, we can detect the photons emitted from \( |e\rangle \) to \( |g\rangle \), where the photon emission rate is directly proportional to the population in \( |e\rangle \).

We again describe the dynamics of the atomic population using rate equations:

\[
\frac{d\rho_e(t)}{dt} = p\gamma \rho_e(t) \\
\frac{d\rho_e(t)}{dt} = R_0\rho_d(t) - (\gamma + R_0)\rho_e(t)
\]
Since experimentally, we only detect photons emitted from spectroscopy on the fluorescence spectrum for the transition for a duration \(T\) is directly proportional to the coupling rate of the probe laser, \(R_\gamma\).

We apply this method to measure fluorescence spectra of trapped \(^{40}\text{Ca}^+\). In this case, the laser wavelengths that couple \(|g\rangle \rightarrow |e\rangle\) and \(|d\rangle \rightarrow |e\rangle\) are at 397 nm and 866 nm, respectively (see [10] for details on the experimental apparatus), and \(p \sim 0.936\) for \(^{40}\text{Ca}^+\) [10]. To observe the fluorescence spectrum for \(|g\rangle \rightarrow |e\rangle\) transition, in each measurement iteration, the 397 nm laser (the probe) is first turned on for a short duration. We adjust the intensity and the duration of the probe beam such that the fluorescence from the ions is constant during the probing time, as shown in figure 2(a). This ensures that the probability that ions are excited is small. After switching off the probe light, the ions are reset to \(|g\rangle\) with a strong pulse at 866 nm, preparing all the ions at the ground state for the next iteration.

To observe the fluorescence spectrum for \(|d\rangle \rightarrow |e\rangle\) transition, the roles of the 397 and 866 nm are reversed, as shown in figure 2(b). In both cases, only the 397 nm photons scattered from ions are collected during the probe duration. For our measurement, the probe beam duration is typically 2 \(\mu\)s and the reset beam duration is typically 10 \(\mu\)s. For switching the laser intensities, we use acousto-optical modulators in a double-pass configuration.

To minimize heating and cooling effects of the ions due to the probe and reset lasers, we perform Doppler cooling with a period of 1 ms after every 50 repetitions of the measurement cycle. Since the fluorescence is constant during the detection window (as shown in figure 2), using the 866 nm laser as a probe, each ion scatters less than one photon from the 866 nm laser during each measurement cycle. Thus, the ions do not heat up significantly and they remain close to the initial temperature after 50 measurement cycles.

Using the 397 nm laser as a probe, equation (4) shows that the decay constant of the fluorescence is suppressed by a factor of \((1 - p) \sim 0.064\) as compared to \(p \sim 0.936\) in equation (11) for the 866 nm probe laser. Hence, constant fluorescence does not guarantee that each ion scatters less than one photon during each measurement cycle. Instead, we estimate from the number of photons collected and our detection efficiency (\(\sim 1.2 \times 10^{-3}\) [10]) that our 397 nm probe beam has a saturation parameter of \(s_0 \sim 0.003\). With this, after 50 measurement cycles, the 397 nm probe beam increases the temperature of the ions by less than \(\sim 8\%\). For the ions close to the Doppler temperature (\(\sim 1\) mK), this change in the temperature has a negligible effect.

\[ \alpha = \frac{\hbar^2 \Delta}{(1 + s_0 + 4(\Delta^2)^2)^2} \]

\(\Delta\) is the detuning and \(\Delta\) is the wavenumber of the laser [12]. By integrating the equation of motion of the ion, the kinetic energy of the ions is given by \(E_k(t) = E_0 e^{\alpha t/m}\) where \(m\) is the mass of the ion and \(E_0\) is the initial energy. For \(s_0 \sim 0.003\), \(\alpha\) is maximum at \(\frac{\Delta}{\Delta^2} \sim \frac{1}{16}\). With this, after 50 measurement cycles \((t_f = 50 \times 2\mu\text{s})\), we have \(\frac{E_k(t_f)}{E_0} \sim 1.08\).
is negligible compared to the statistical uncertainty extracted from the fit of trapped ions of both $^{40}\text{Ca}$. The magnetic field extracted from the fit has a statistical uncertainty of $\sim 1\text{ mK}$, an 8% increase in the temperature from the current applied to the magnetic coil. The micromotion of the ions is $\sim 1\text{ mK}$, an 8% increase in the temperature from the current applied to the magnetic coil. The micromotion of the ions is dominated by the excess micromotion in the radial directions caused by a stray static electric field. We also include a possibility of having an additional micromotion caused by a relative phase difference of the potentials between trap RF-electrodes.

Due to the Doppler effect, the electric field of the probe laser as seen by ions undergoing micromotion is given by

$$E(t) = \text{Re}[E_0 e^{i(\omega_0 t + \vec{k} \cdot \vec{u})}]$$

where $E_0$ is the amplitude of the laser field, $\vec{k}$ is the wavevector of the laser, $\omega_0$ is the frequency of the laser, $\vec{u}_0$ and $\vec{u}$ are the displacements of the regular motion and excess micromotion, respectively. At small excess micromotion amplitude, we can write

$$\vec{k} \cdot \vec{u} = \beta \cos (\Omega t + \delta),$$

where $\Omega$ is the trap drive frequency, $\beta$ is the modulation index of the micromotion and $\delta$ is the associated phase. The dependence of the micromotion modulation index on the two radial trap frequencies, $f_x$ and $f_y$, is given by equation (22) in [15]:

$$\beta = \frac{1}{\lambda_2 \Omega^2} \left( \frac{A}{f_x^2} + \frac{B}{f_y^2} \right)^2 + C,$$

where $\lambda_2$ is the wavelength of the probe laser, $A$ and $B$ are constants proportional to the projection of the stray electric field on the two radial directions and $C$ describes additional micromotion from a relative phase difference of the potentials between trap electrodes. From equations (13) and (14), the fluorescence of the ion at low probe laser intensity can be expanded using Bessel functions:

$$P \propto |E_0|^2 \sum_{n=-\infty}^{\infty} \frac{J_n(\beta)}{(\Delta + n\Omega)^2 + (\gamma/2)^2}.$$

Micromotion modulated spectrum of trapped ions

In this section we study micromotion modulated spectra of trapped ions of both $^{2}\text{S}_{1/2} \rightarrow ^{2}\text{P}_{1/2}$ and $^{2}\text{D}_{3/2} \rightarrow ^{2}\text{P}_{1/2}$ transitions in $^{40}\text{Ca}$. We consider a case where the micromotion of the ions is dominated by the excess micromotion in the radial directions caused by a stray static electric field. We also include a possibility of having an additional micromotion caused by a relative phase difference of the potentials between trap RF-electrodes.

Due to the Doppler effect, the electric field of the probe laser as seen by ions undergoing micromotion is given by

$$E(t) = \text{Re}[E_0 e^{i(\omega_0 t + \vec{k} \cdot \vec{u})}]$$

where $E_0$ is the amplitude of the laser field, $\omega_0$ is the frequency of the laser, $\vec{u}_0$ and $\vec{u}$ are the displacements of the regular motion and excess micromotion, respectively. At small excess micromotion amplitude, we can write

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$$P \propto |E_0|^2 \sum_{n=-\infty}^{\infty} \frac{J_n(\beta)}{(\Delta + n\Omega)^2 + (\gamma/2)^2}.$$

We use the experimental scheme described in the previous section to observe the modulations of fluorescence spectra of the $^{2}\text{S}_{1/2} \rightarrow ^{2}\text{P}_{1/2}$ transition from ion micromotion. We measure the modulation index, $\beta$, as a function of the radial trap frequencies. By changing the amplitude of the RF-drive applied to the trap electrodes, the relationship between the two radial trap frequencies is measured to be

$$f_y = 1.18(4) \cdot f_x - 0.63(8) \text{ (MHz)}.$$

The fluorescence spectra obtained for different trap frequencies, $f_x$, fitted to the model given in equation (16), are shown in figure 4. The micromotion sidebands at the drive frequency ($\omega \sim 2\pi \times 30.7 \text{ MHz}$) are clearly visible. The dependence of the modulation index, $\beta$, on $f_x$ is plotted in figure 5. The data is fitted to a model according to equations (15) and (17) with the errorbars in $\beta$ (comparable to the size of the markers) derived from the fit in figure 4. The fit yields $(A, B) = (1.1 \pm 0.6, 1.0 \pm 0.2) \times 10^7 \text{ nm} \cdot \text{MHz}^2$ and

Figure 3. The fluorescence spectrum of the $^{2}\text{S}_{1/2} \rightarrow ^{2}\text{P}_{1/2}$ transition for $^{40}\text{Ca}$ as the frequency of the laser, $f_0$, is scanned across the transition. After measuring for 10 min (10$^6$ cycles per each laser frequency), we extract the centre frequency with a statistical uncertainty of 200 kHz. The solid line is a Lorentzian profile including the Zeeman effect and the micromotion of the ions (see text).

on the lineshape of the fluorescence spectrum$^2$. Moreover, the value of $x_0$ suggests that power broadening is also negligible.

Figure 3 shows the spectrum obtained with this method on the $^{2}\text{S}_{1/2} \rightarrow ^{2}\text{P}_{1/2}$ transition. The method is not restricted to a single ion, which allows us to increase signal-to-noise by performing measurement with many ions. For the data shown in figure 3, we use a linear crystal of 7 ions and $\sim 10^6$ cycles per each probe laser frequency. This corresponds to a total of measurement time of 10 min. We describe the data by summing over Lorentzian profiles between different Zeeman sublevels. We also take into account the effects of micromotions, as described in the following section.

The fit in figure 3 yields $\chi^2_{\text{red}} \approx 1.02$ and a linewidth of 24.1 $\pm$ 1.3 MHz, consistent with the natural linewidth of 22.4 MHz [13] with additional Doppler broadening for an ion temperature of 5.3 $\pm$ 4.6 mK. This is consistent with the ion temperature of 0.8 mK extracted from the previous measurement in [14]. The magnetic field extracted from the fit is 1.4 $\pm$ 0.5 gauss. This is consistent with $\sim 0.8$ gauss which we estimate from the current applied to the magnetic coil. The centre of the spectrum extracted from the fit has a statistical uncertainty of $\sim 200$ kHz.

In this section we study micromotion modulated spectra of trapped ions of both $^{2}\text{S}_{1/2} \rightarrow ^{2}\text{P}_{1/2}$ and $^{2}\text{D}_{3/2} \rightarrow ^{2}\text{P}_{1/2}$ transitions in $^{40}\text{Ca}$ using the method described in the previous section.

To describe the effect of the micromotion caused by a radio-frequency (RF) drive in a linear Paul trap, we follow the analysis by Berkeland et al [15]. We consider a case where the micromotion of the ions is dominated by the excess micromotion in the radial directions caused by a stray static electric field. We also include a possibility of having an additional micromotion caused by a relative phase difference of the potentials between trap RF-electrodes.

Due to the Doppler effect, the electric field of the probe laser as seen by ions undergoing micromotion is given by

$$E(t) = \text{Re}[E_0 e^{i(\omega_0 t + \vec{k} \cdot \vec{u})}]$$

where $E_0$ is the amplitude of the laser field, $\omega_0$ is the frequency of the laser, $\vec{u}_0$ and $\vec{u}$ are the displacements of the regular motion and excess micromotion, respectively. At small excess micromotion amplitude, we can write

$$\vec{k} \cdot \vec{u} = \beta \cos (\Omega t + \delta),$$

where $\Omega$ is the trap drive frequency, $\beta$ is the modulation index of the micromotion and $\delta$ is the associated phase. The dependence of the micromotion modulation index on the two radial trap frequencies, $f_x$ and $f_y$, is given by equation (22) in [15]:

$$\beta = \frac{1}{\lambda_2 \Omega^2} \left( \frac{A}{f_x^2} + \frac{B}{f_y^2} \right)^2 + C,$$

where $\lambda_2$ is the wavelength of the probe laser, $A$ and $B$ are constants proportional to the projection of the stray electric field on the two radial directions and $C$ describes additional micromotion from a relative phase difference of the potentials between trap electrodes. From equations (13) and (14), the fluorescence of the ion at low probe laser intensity can be expanded using Bessel functions:

$$P \propto |E_0|^2 \sum_{n=-\infty}^{\infty} \frac{J_n(\beta)}{(\Delta + n\Omega)^2 + (\gamma/2)^2}.$$
Figure 4. Micromotion modulated fluorescence spectra of the $S_{1/2} \rightarrow P_{1/2}$ transition for $^{40}$Ca$^+$ at different modulation indices, $\beta$, due to different trap frequencies.

Figure 5. Micromotion modulation index, $\beta$, measured as a function of trap frequency, $f_x$. The line is the theoretical fit according to equation (15) with the value of the radial trap frequencies given by equation (17). The errorbars in $\beta$ are comparable to the size of the markers.

$C = 0.00 \pm 0.03$ with $\chi^2_{\text{red}} \approx 0.94$. The value of $C$ suggests that the effect from the phase difference of the potentials on the trap electrodes is small.

We also observe the micromotion modulated fluorescence spectra of the $^2D_{3/2} \rightarrow ^2P_{1/2}$ transition, as shown in figure 6, which are measured at two different trap frequencies. At the trap frequency of $f_x = 1.07$ MHz, the spectra of the $^2D_{3/2} \rightarrow ^2P_{1/2}$ and $^2S_{1/2} \rightarrow ^2P_{1/2}$ transitions yield the modulation indices to be $\beta_{866} = 1.04(5)$ and $\beta_{397} = 2.31(5)$. The ratio $\beta_{397}/\beta_{866} = 2.22(12)$ is consistent with the ratio of the laser wavelengths $\lambda_{866}/\lambda_{397} \approx 2.18$ according to equation (15).

Since we are able to directly observe the effect of micromotion on the fluorescence spectra, this method can be used to compensate excess micromotion in the direction along the laser beam direction, $\hat{k}$. The projection of the micromotion can be on either the main Doppler cooling laser (397 nm for $^{40}$Ca$^+$) or the repumper laser (866 nm for $^{40}$Ca$^+$). Instead of recording the whole spectrum and extracting $\beta$ from the fit, one can instead fix the probe laser frequency to be at the micromotion sideband and minimize the fluorescence. By reducing the trap frequency, the sensitivity of this method can be increased until the trap becomes unstable.
Conclusions

In this work, we present an experimental scheme to perform direct spectroscopy on the $^2S_{1/2} \rightarrow ^2P_{1/2}$ and $^2D_{3/2} \rightarrow ^2P_{1/2}$ transitions for $^{40}\text{Ca}^+$. The method allows us to circumvent the usual limitations due to a lambda three-level energy structure present in many species of trapped ions and atoms. We apply this method to directly observe micromotion modulated spectra of the fluorescence for both transitions in $^{40}\text{Ca}^+$ and show that the dependence of the micromotion modulation index on the trap frequency agrees well with the model presented in [15]. Even in the presence of strong micromotion, we extract the centre frequency with statistical uncertainty on the order of 200 kHz, which is comparable to or better than recent measurements on dipole transitions of trapped ions [7–9, 16, 17]. We expect that the absolute transition frequencies of both transitions can be determined with a frequency comb to within the same order of uncertainty [7, 9]. Specifically, the absolute transition frequency for the $^2D_{3/2} \rightarrow ^2P_{1/2}$ of $^{40}\text{Ca}^+$ (866 nm) has not yet been determined to the level of $\sim 200$ kHz [18]. The method is easily adaptable to other species of trapped ions, and the number of lasers required is minimal.

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