Ultrafast coherent excitation of a $^{40}$Ca$^+$ ion

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Abstract

Fast entangling quantum gates can significantly enhance the performance of a trapped-ion quantum computer. In pursuit of implementing a fast two-qubit gate, we investigate the coherent excitation of a $^{40}$Ca$^+$ ion with a train of picosecond pulses resonant to the $4S_{1/2} \leftrightarrow 4P_{3/2}$ transition. The optical pulse train is derived from a mode-locked, stabilized optical frequency comb. We implement two techniques to characterize the pulse-ion interaction and show how all requirements can be met for an implementation of a fast phase gate operation.

1. Introduction

Trapped ions are a well-studied and promising system for the implementation of a scalable quantum computer [1–5]. Two-qubit entangling gate operations have been demonstrated [6–9] and combined with single-qubit gates to build an elementary quantum processor [10–12]. The entangling gate operations in these experiments have been carried out in an adiabatic regime in which they rely on coupling to spectroscopically-resolved motional sidebands of the qubit transition. The requirement to resolve sidebands introduces an intrinsic limit to the duration of a gate operation: it has to be longer than the period of motion of the ions in the trap (about 1 μs). Overcoming this limitation would advance the development of a scalable quantum computer as it would allow one to increase the number of gate operations (computational steps) that can be completed within the coherence time of the ion-qubits.

It has been proposed that two-qubit entangling gates with gate times of less than one trap period should be realizable by making the ions interact with counter-propagating ultrafast laser pulses [13]. Several groups are working on its realization [14, 15] but so far only single-qubit gate operations [16, 17] and single-ion spin-motion entanglement [18] have been reported on time scales shorter than the ion oscillation period. Creation of two-qubit entanglement by a train of ultrafast laser pulses within a few microseconds has been demonstrated in the ground-states of a pair of Yb$^+$ ions [19]. Recently, Schäfer et al reported a two-ion entangling gate faster than the motional period using amplitude-shaped laser pulses [20].

Our goal, beyond the scope of this work, is to implement an ultrafast two-qubit phase gate operation [13, 21] using picosecond laser pulses and to complete it in less than one trap period. The scheme uses resonant, counter-propagating pulse pairs for applying a state dependent, stimulated force [22] to a two-ion crystal [23]. By subjecting the ion crystal to a sequence of momentum kicks and times of free evolution of the crystal in the trap potential, the ions are forced to follow state-dependent, closed trajectories through phase-space. The area enclosed by these trajectories is proportional to the phase the state acquires during the pulse sequence [7]. When the relative phase between the state-pairs $|\downarrow\downarrow\rangle, |\uparrow\uparrow\rangle$ and $|\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle$ is $\pi/2$ and the pulse sequence returns motional modes $\{24\}$ of the ion crystal to the initial state, the operation will realize a maximally entangling geometric phase gate [13]. In general, the higher the repetition rate $f_{\text{rep}}$, the faster the gate operation can be completed.

A suitable pulsed laser system to implement such a fast gate operation should satisfy four requirements: (1) $f_{\text{rep}}$ should be much higher than the trap frequency of $\sim1$ MHz for fine-grained control over the timing of pulse sequences. (2) The pulse length has to be much shorter than the excited state lifetime to minimize spontaneous decay. (3) The spectrum should resonate with the atomic transition and (4) the laser intensity needs to be high.
enough to achieve a complete population transfer between the two levels by a single laser pulse. Alternatively to (3), non-resonant pulses can be used to apply state-dependent momentum kicks \cite{17}, where the laser is tuned in between the two fine structure components such that Stark shifts cancel, while limiting overlap of the laser’s optical spectrum with either of the two transition frequencies.

In this manuscript, we describe trapped-ion experiments with a laser system satisfying conditions (1)–(3). We use trains of equally spaced picosecond pulses to resonantly excite a $^{40}$Ca$^{+}$ ion on the dipole transition at 393 nm (see figure 1) and observe single laser pulses capable of flipping the state of the ion with a probability of 96(2)\%.

Furthermore, we report two ways to extract information on the Rabi frequency and compare them in terms of applicability and prerequisites. One method allows us to gain knowledge on pulse-to-pulse phase shifts, the other to observe the phase evolution of the $4S_{1/2}$ state due to the ion-laser interaction, with respect to the $3D_{5/2}$ state.

2. Generation of picosecond pulse trains

2.1. Picosecond-laser system

We use a picosecond laser system with repetition rates on the order of GHz and a center wavelength of 393.366 nm. The laser setup is shown in figure 2. It is derived from a commercial optical frequency comb that generates 70 fs pulses at a rate of $f_{\text{rep}} = 250$ MHz with a center wavelength of 1550 nm. The carrier envelope offset and $f_{\text{rep}}$ are locked to a frequency reference provided by a GPS-disciplined oven-controlled crystal oscillator with a fractional frequency instability of $\sim 2 \times 10^{-13}$.

The laser pulses travel through a stretcher fiber which adds dispersion and stretches the pulses to $\sim 50$ ps for chirped pulse amplification \cite{30}. Multiple semiconductor optical amplifiers (SOAs) and erbium doped fiber amplifiers (EDFAs) are used to compensate insertion losses at various stages in the set-up (see figure 2(B)). The pulse train then passes through a spectral filter which selects a new center wavelength of 1572 nm and reduces the spectral bandwidth to 8 nm. Two filter cavities, both with a free spectral range of 5 GHz, increase $f_{\text{rep}}$ from 250 MHz to 5 GHz to increase the resolution of the pulse timings. The cavities are locked to a CW-laser which in turn is locked to a mode of the frequency comb. The second cavity’s purpose is to increase the extinction ratio and therefore equalize the optical intensity of the output pulses. Furthermore, we use a second spectral filter to compensate amplifier-induced frequency shifts \cite{31} of up to 4 nm and further limit the bandwidth to 6.4 nm.

Next, a high power EDFA amplifies the pulses to a maximum average power of 2.8 W and subsequently a free-space third-order dispersion-compressor (TOD) reduces the pulse width to 680 fs (time bandwidth product 0.53). The compressed 1572 nm pulses are converted to 393 nm by frequency doubling the light twice in two separate nonlinear crystals as shown in figure 2(D). We achieve a maximum single-pass conversion efficiency (from 1572 to 393 nm) of 0.9\% (3.9\%) at $f_{\text{rep}} = 5$ GHz (1.25 GHz).

![Figure 1. Reduced energy level scheme of $^{40}$Ca$^{+}$, showing levels relevant to the experiment. Possible transitions are shown with their wavelengths and branching ratios \cite{25, 26}, excited electronic states with their lifetimes \cite{27–29}. The bell curve-shaped bars in the upper-left corner are a representation of the pulsed laser’s spectral modes. The qubit is encoded in two Zeeman substates of the $4S_{1/2}$ ($\downarrow |\rangle$) and $3D_{5/2}$ ($\uparrow |\rangle$) levels.](image-url)
2.2. Pulse picking
Laser pulse picking at a high repetition rate and high average power becomes a major challenge at ultraviolet wavelengths. To satisfy both requirements, we chose a twofold approach using a fast switching element before the high power EDFA (where the average laser power can be limited to 10 mW) and a slow element after the amplifier to create the desired pulse sequences (see panel C in figure 2).

The fast element is a pulse picker which is realized by a Mach–Zehnder interferometer with an electro-optic modulator of 7 GHz bandwidth. Since the maximum optical input power it can sustain is 10 mW, we install it before the high power EDFA. As the amplifier needs to be seeded continuously with a maximally allowed dark time of $\tau_{\text{dark}} \leq 20 \text{ ns}$, an additional switching element after the amplifier is required, which has a high damage threshold and a switching time of less than 20 ns. For this we use a Pockels cell that enables switching of the cell with a rise/fall time of 7 ns at a maximum repetition rate of 10 MHz and a measured optical extinction ratio of 30 dB. Both the pulse picker and the Pockels cell are controlled by an arbitrary waveform generator with a sample rate of 25 GS s$^{-1}$ which is synchronized with the frequency comb. A photodiode trace of arbitrary pulse patterns is shown in figure 3.

2.3. Pulse switching characteristics
The pulse picking scheme described above necessitates blocking the pulses with the fast pulse picker during the rise and fall time of the Pockels cell for typically 12 ns. This results in an equally long dark time of subsequent amplifiers. We examined the characteristics of picked pulses by measuring their intensities and relative phases. To characterize the phase shifts of the pulses, we interfere every picked pulse with its successive pulse in a Michelson interferometer, detailed in [32].

We vary $f_{\text{rep}}$ using the pulse picker and observe both repetition-rate-dependent intensity and phase changes. In the case of the highest possible $f_{\text{rep}}$ of 5 GHz, we observe that the first pulse after a dark time has a three times higher intensity and a phase difference of $\Delta\phi = 1.26 \pi$, both with respect to later pulses. However, at lower repetition rates, these anomalies weaken in effect and disappear at the lowest tested $f_{\text{rep}}$ of 1.25 GHz.

We have determined that both effects are caused by SOA2 (see figure 2), which serves as a preamplifier to the high power EDFA, and are due to the finite carrier lifetime of the SOA of $\sim 500$ ps. This causes the dynamical
behavior of the SOA to depend on the input signal of the past $\sim 500 \text{ ps}$ [33] and is known as the 'pattern effect' [34]. In future experiments, the SOA will be replaced by another fiber-based preamplifier.

3. Ultrafast coherent excitation of a $^{40}\text{Ca}^+$ ion with picosecond pulse trains

We present two different types of experiments in which a $^{40}\text{Ca}^+$ ion coherently interacts with trains of picosecond pulses. The experiments shed light on the interaction strength of the pulses with the ion as well as single pulse characteristics. In particular, measuring the excited state population at the end of the interaction enables us to infer the rotation angle per pulse $\theta = \int \Omega(t) \, dt$, where $\Omega(t)$ is the instantaneous Rabi frequency.

In the first approach, after laser cooling and preparing the ion in a Zeeman ground state, we excite the ion with pulse trains of various pulse numbers and measure the $4P_{3/2}$ state population via its decay to the metastable $3D_{5/2}$ state. In the second approach, we embed the pulsed excitation of the ion within a Ramsey experiment probing the coherence of the two qubit states. This approach enables us not only to monitor the population transfer induced by the short pulses, but also to track the change in the complex amplitude of the part of the wavefunction residing in the $4S_{1/2}$ ground state.

3.1. Experimental setup

Experiments are conducted in the linear Paul trap that was described in [35]. A 5 m long, polarization-maintaining, fused silica fiber guides the $\sim 2 \text{ ps}$ long pulses to the ion trap. After the fiber, the pulses are $\sigma_+$ circularly polarized and focused through holes in the trap’s endcap electrodes to a beam waist of $11.8(3) \text{ \mu m}$ at the position of the ion. As the quantization axis, which is defined by a bias magnetic field, is aligned with the trap axis, the laser couples only Zeeman states with magnetic quantum numbers differing by $\Delta m = \pm 1$.

Unless stated otherwise, all following measurements and experiments presented in this paper were conducted with a pump power (after TOD) of $\approx 600 \text{ mW}$. During times when the Pockels cell was off and the pulses directed into a beam dump, the high power EDFA was seeded with pulses at $f_{\text{rep}} = 1.25 \text{ GHz}$. Under these conditions, the average 393 nm power is about $9 \text{ mW}$ and the pulse energy $E_{\text{pulse}} \approx 7.2 \text{ pJ}$.

Figure 3. Pulse patterns generated by using either only the pulse picker (top), only the Pockels cell (middle) or both (bottom), measured by detecting the residual 786 nm light after the PPKTP crystal. The pulses labeled 'idle' (1.25 GHz) on both sides of the top panel are required to seed the high-power EDFA. The length of the dark time on either side of the 'payload' is determined by two factors: the rise and fall times of the Pockels cell of 7 ns and the minimum allowed time of 35 ns between switching the cell on and off — i.e., between the start of the rise time and the start of the fall time. The inset in the bottom panel shows the zoomed-in payload signal. Every grid line corresponds to the location of a pulse in the original 5 GHz pulse train. For the reason for the different pulse heights see section 2.3.
3.2. Coherent excitation using few pulses

Using few-pulse trains to excite the ion allows us to gain crucial insights into single pulse dynamics and characteristics. First, we initialize the ion by Doppler-cooling and optical pumping to the $4S_{1/2}$ state. We then send pulse trains of between 1 and 12 pulses to the ion. Afterwards, we collect ion fluorescence on the $4S_{1/2} \leftrightarrow 4P_{1/2}$ transition in order to determine whether the ion has decayed into the dark $3D_{5/2}$ state. We repeat the measurement 100 times to statistically determine the $3D_{5/2}$ state population. The probability of the $4P_{3/2}$ state to decay to the $3D_{5/2}$ state is $P_{D} = 0.0587(2)$ [25]. We amplify the signal by repeating the pulse train $m$ times as shown in the experimental sequence in figure 4 to accumulate population in the $3D_{5/2}$ state [25] and choose $m \approx 1/P_{D}$. Between repetitions, a waiting time $t_w = 20 \mu s$ much larger than the $4P_{3/2}$ state’s lifetime ensures that any population in that state has decayed. We then measure the $3D_{5/2}$ state population as a function of the laser detuning for different pulse train lengths.

Figure 5 shows experimental data and a simulation of the probability for being in the $3D_{5/2}$ state after $m = 20$ pulse trains. The model for the ion-laser interaction used for simulating our experiments is presented in the appendix. We vary the detuning $\Delta$ and the number of pulses for different repetition rates. The simulation parameters are obtained by a fit to the data. Free fit parameters are $\theta$, the phase offset of the first pulse $\Delta \phi$ and a detuning offset. Since we know that the first pulse in the case of $f_{\text{rep}} = 5 \text{ GHz}$ also has a different intensity, we use an additional fit parameter in that case: the rotation angle of the first pulse $\theta^{\text{fit}}$.

All fit values are within three standard deviations of those acquired previously with long pulse trains [32], as well as those acquired with the Michelson interferometer.

3.3. Single pulse with area $\pi$

In order to check if a single pulse can act as a $\pi$-pulse we repeat the previous experiment with only one pulse while varying the 393 nm light power ($n = 1$, $m = 15$). From the experimentally determined $3D_{5/2}$ state probability $P_D$ after the 15 repetitions of a single pulse we calculate the $4P_{3/2}$ state probability $P_P$ after only one single pulse using

$$P_P = \frac{1}{P_{D}^2}(1 - (1 - P_D)^2).$$

Due to measurement fluctuations, this can sometimes lead to unphysical values of $P_P > 1$ if we measure a $P_D$ that happens to be larger than the maximum expectation value of $P_{D,max} = 1 - (1 - P_{D}/2)^m \approx 0.60$.

The excitation probability is a function of the sine squared of a phase, which is proportional to the Rabi frequency $\Omega$. We hence plot $P_P$ versus the square root of the 393 nm pulse energy, which is also proportional to $\Omega$ (and $\theta$), in figure 6. The data points should therefore follow the curve

$$P_P = P_{P,max} \cdot \sin^2 (\pi \sqrt{E_{\text{pulse}}/\Omega}),$$

where $\xi$ is a proportionality factor and $P_{P,max}$ is the maximum probability of a single pulse exciting the ion to the $4P_{3/2}$ state. A fit of this curve to our data yields $\xi = 11.3(3) \sqrt{[\text{pJ}]}$ and $P_{P,max} = 96(2)\%$, showing how close we are to achieving a $\pi$-pulse. We found that the experimental value for $E_{\text{pulse}} = \frac{(\xi/2)^2}{2} = 32(3) \text{ pJ}$ is in agreement with the theoretical prediction of 30 pJ calculated using the transition’s well-known decay rate and wavelength, and our measured beam parameters [24].

![Figure 4. Experimental sequence of the coherent excitation with few-pulse trains. The ion is Doppler-cooled and prepared in the 4S_{1/2} state. A pulse train of $n$ pulses coherently drives the 4S_{1/2} \leftrightarrow 4P_{1/2} transition. The pulse train is repeated a total of $m$ times with a waiting time $t_w$ between each two repetitions. We choose $t_w = 20 \mu s \gg \tau$, where $\tau$ is the 4P_{3/2} state’s lifetime. Finally, we measure the 3D_{5/2} state probability.](image-url)
Figure 5. Probability to excite the ion to $3D_{5/2}$ with different numbers of pulses and detuning. Left column: experimental data; right column: simulation. 1st row: $f_{\text{rep}} = 5$ GHz, $\theta^{\text{meas.}} = 0.353(1) \pi$, $\theta = 0.195(18) \pi$, $\Delta \phi = 1.282(1) \pi$. 2nd row: $f_{\text{rep}} = 2.5$ GHz, $\theta = 0.312(10) \pi$, $\Delta \phi = 0.361(2) \pi$. 3rd row: $f_{\text{rep}} = 5/3$ GHz, $\theta = 0.339(7) \pi$, $\Delta \phi = 0.088(3) \pi$. 4th row: $f_{\text{rep}} = 1.25$ GHz, $\theta = 0.358(3) \pi$, $\Delta \phi = 0.051(1) \pi$. The asymmetry evident in the first two rows is due to the phase shift of the first pulse with respect to later pulses.

Figure 6. Probability to excite the ion to $4P_{3/2}$ with a single pulse as a function of 393 nm light power. $P_{P, \text{max}}$ is the maximum excitation probability to the $4P_{3/2}$ state and determined by a fit to the data. Measurement noise can cause unphysical values of $P_P > 1$ (see text).
The discrepancy is most likely due to an uncertainty in the nonlinear refractive index $n_2$ of fused silica fiber at 393 nm which we assumed to be $n_2 = 1.5 \times 10^{-20}$ m$^2$ W$^{-1}$ [36].

3.4. Ramsey contrast decay and revival

The coherent manipulation of a qubit entails a precise quantum control of the relative phase of the two qubit states’ amplitudes. The experiments of the previous subsections used only one of the two qubit states and the relative phase appeared only as an unobservable global phase. By designing experiments where the second qubit state serves as a phase reference, we obtain access to the desired information.

Towards this end, we let few-pulse trains interact with the ion during the waiting time of a Ramsey experiment probing the qubit coherence [16]. We monitor the phase coherence at the end of the Ramsey sequence by first creating a coherent superposition of the two qubit states by a $\pi$-pulse on the qubit transition with a 729 nm laser as illustrated in figure 7. Next, variable numbers of picosecond pulses are used to coherently drive the $4S_{1/2} \rightarrow 4P_{3/2}$ transition. If there is any population remaining in the $4P_{3/2}$ state after the pulses, this part of the population will undergo spontaneous decay and thus destroy the coherence. We finally analyze how much coherence remains by varying the phase of the second 729 nm laser pulse and measuring the $3D_{5/2}$ state probability, while keeping the Ramsey time $t_R = 10 \, \mu$s much shorter than the coherence time of the superposition (O(1 ns)).

Figure 7. Sequence of a Ramsey contrast decay and revival experiment. The ion is Doppler-cooled and prepared in the $4S_{1/2}$ state. The first 729 nm laser pulse creates a coherent superposition of the ion’s internal $4S_{1/2}$ and $3D_{5/2}$ states. A controlled and variable number of 393 nm laser pulses act on the remaining $4S_{1/2}$ state population, transferring population to the $4P_{3/2}$ state and possibly back, potentially reducing the S-D coherence. We finally analyze how much coherence remains by varying the phase of the second 729 nm laser pulse and measuring the $3D_{5/2}$ state probability, while keeping the Ramsey time $t_R = 10 \, \mu$s much shorter than the coherence time of the superposition (O(1 ns)).


\[
\begin{array}{c|c|c}
\text{power} & \theta = \pi/2 & \theta = \pi/2 \\
729 \text{ nm} & \phi = 0 & \phi' = 0-2\pi \\
393 \text{ nm} & \end{array}
\]

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{ramsey_sequence.png}
\caption{Sequence of a Ramsey contrast decay and revival experiment. The ion is Doppler-cooled and prepared in the $4S_{1/2}$ state. The first 729 nm laser pulse creates a coherent superposition of the ion’s internal $4S_{1/2}$ and $3D_{5/2}$ states. A controlled and variable number of 393 nm laser pulses act on the remaining $4S_{1/2}$ state population, transferring population to the $4P_{3/2}$ state and possibly back, potentially reducing the S-D coherence. We finally analyze how much coherence remains by varying the phase of the second 729 nm laser pulse and measuring the $3D_{5/2}$ state probability, while keeping the Ramsey time $t_R = 10 \, \mu$s much shorter than the coherence time of the superposition (O(1 ns)).}
\end{figure}

in order to extract the contrast $C$ and the phase $\Phi$ of the interference signal. There are two limiting cases: If the contrast is equal to one, the ion was still in a fully coherent superposition of the $4S_{1/2}$ and $3D_{5/2}$ states and there was no spontaneous decay of the $4P_{3/2}$ state. If the contrast is zero, all the initial $4S_{1/2}$ state population had been transferred to the $4P_{3/2}$ state and spontaneous decay has destroyed all coherence. For a resonant excitation of the $4S_{1/2} \rightarrow 4P_{3/2}$ transition, the phase is expected to undergo jumps of $\pi$ each time the ion’s state population is transferred to the $4P_{3/2}$ state and back again to the $4S_{1/2}$ state by the pulse train. This is due to the fact that the quantum states of a two-level system undergoing a $2\pi$ rotation pick up a minus sign, $|0\rangle \xrightarrow{2\pi} -|0\rangle$.

We repeat the measurement as a function of the number $n$ of 393 nm laser pulses and plot in figure 8 the remaining coherence and phase versus $n$. For a dataset taken with low pulse power (figure 8(a)), it took about 15 optical pulses at $f_{rep} = 5$ GHz to complete a $\pi$-pulse, which corresponds to $\theta = 0.065(1) \, \pi$. The fitted detuning of $\Delta/2\pi = 0.029(4) \, \text{GHz}$ of the nearest comb mode to the ion transition shows that the transition was resonantly excited. The data shows that we can drive the $4S_{1/2} \leftrightarrow 4P_{3/2}$ transition coherently and that each time we return to the $4S_{1/2}$ state, the data is consistent with the observation of phase jumps by $\pi$. Figure 8(b) shows another measurement which was carried out at a higher pulse power, that was the same as for the measurements presented in subsection 3.2. We can observe the same structure as in the first data set but we need only about 2.5 optical pulses to complete one $\pi$-pulse on the transition. The phase jumps are washed out, because the Bloch vector does not cross the poles of the $|S\rangle - |P\rangle$ Bloch sphere. A fit of the simulation of the ion-light interaction to the experimental data allows us to extract $\theta = 0.375(4) \, \pi$ and $\Delta/2\pi = 0.14(2) \, \text{GHz}$.

3.5. Discussion

Using few pulses to coherently excite the ion transition provides information about the pulse characteristics, such as pulse-to-pulse phase shifts and pulse intensity. The method does not require a laser resonant to the $4S_{1/2} \leftrightarrow 3D_{5/2}$, 1 Hz linewidth transition. Rather, only readily available lasers stabilized to a linewidth of $\lesssim 1$ MHz are needed. The only parameters required to fit the data are the number of pulses $n$ and the well-known decay rates...
In the following, we describe how we model the response of a trapped $^{40}\text{Ca}^+$ ion to a single optical pulse. We apply to measure the rotation angle per pulse. We model the response of a trapped $^{40}\text{Ca}^+$ ion with a pulsed laser field resonant to the ion’s $4S_{1/2} \leftrightarrow 4P_{3/2}$ transition. We assume a three-level system consisting of the $4S_{1/2}$ state ($\ket{1}$), the $4P_{3/2}$ state ($\ket{2}$) and the $3D_{5/2}$ state ($\ket{3}$), as indicated in figure 1. We consider spontaneous decay of state $\ket{1}$ with a rate of $\Gamma_{ps}$ and $\Gamma_{pd}$ from the fit, $\theta$, $\Delta \phi$ and a detuning offset of the laser pulses can be extracted. This allows us to tune the laser into resonance with the transition. The fit results are in agreement with the results of long pulse train measurements and of the experiments probing the coherence of consecutive pulses in a Michelson interferometer.

The Ramsey contrast technique is experimentally more challenging than the procedure described in the previous paragraph. It is necessary to create a coherent superposition of the $4S_{1/2}$ and $3D_{5/2}$ states which requires a few-kHz linewidth laser. However, the method allows tracking the phase of the $4S_{1/2}$ state with respect to the phase reference provided by the $3D_{5/2}$ qubit state. Furthermore, the signal does not need to be amplified and hence amplification noise is avoided.

For future experiments, it is crucial to mitigate intensity and phase fluctuations originated during pulses-picking, as both effects would undermine the fidelity of the quantum gate. Therefore, we intend to replace the responsible amplifier, as stated before. Beside the anomalies induced by the amplifier, the effect of inherent pulse-to-pulse intensity variations can be avoided with the addition of an optimum linear chirp to the pulses for adiabatic rapid passage, making the required population transfer robust against intensity fluctuations [37]. Chirping would also reduce the peak intensity in the fiber and thereby SPM. The latter can be further suppressed by using a large mode area fiber to increase the characteristic nonlinear length or by reducing the fiber length [38]. Splitting each pulse to create the counter-propagating pulse pairs makes the scheme insensitive to phase fluctuations that are slow compared to the gate time, if an equal number of kicks in both directions is applied. However, this requires higher pulse energy, such that adiabatic rapid passage is still possible after pulse splitting. Once we have sufficient power, we plan to investigate two-ion dynamics.

4. Conclusion

In summary, coherent manipulation of the trapped $^{40}\text{Ca}^+$ ion has been investigated using resonant picosecond pulses derived from a stabilized optical frequency comb. The desired laser wavelength (393 nm) is engineered via spectral filtering and cascaded second harmonic generation. Two different techniques have been developed and applied to measure the rotation angle per pulse. We find that the methods produce similar results within their respective error margins. We have shown the ability to create a $\pi$-pulse with a single optical pulse: a key-requisite to implementing a resonant, ultrafast, two-qubit phase gate operation.

Appendix. Pulse model for simulation and fits

In the following, we describe how we model the response of a trapped $^{40}\text{Ca}^+$ ion with a pulsed laser field resonant to the ion’s $4S_{1/2} \leftrightarrow 4P_{3/2}$ transition. We assume a three-level system consisting of the $4S_{1/2}$ state ($\ket{1}$), the $4P_{3/2}$ state ($\ket{2}$) and the $3D_{5/2}$ state ($\ket{3}$), as indicated in figure 1. We consider spontaneous decay of state $\ket{2}$ into state $\ket{3}$.
of 1.2 s [29] is much longer than the cycle time of our experiments of about 10 ns.

To model the time evolution of the quantum state over one pulse period of duration $\tau_{\text{pulse}} = 1/f_{\text{rep}}$, with $f_{\text{rep}}$ the repetition rate, we first account for the effect of a single picosecond pulse on the initial state by subjecting it to an $x$-rotation of the $|1\rangle - |2\rangle$ subsystem that transforms the initial density matrix $\rho_i$ into

$$
\rho' = \mathcal{U}_R \rho_i \mathcal{U}_R^\dagger,
$$

where

$$
\mathcal{U}_R = \exp \left( \frac{i}{2} \theta (|2\rangle \langle 1| + |1\rangle \langle 2|) \right),
$$

(A.4)

describes the $x$-rotation with rotation angle $\theta$.

Next, we take into account the possible detuning $\Delta$ of the laser light from the atomic transition and an additional light shift $\Delta'$ of the state $|3\rangle$ by a unitary $\mathcal{U}_Z$ inducing phase shifts. The unitary applies $z$-rotations on two of the state’s subsystems: one of the $|1\rangle - |2\rangle$ subsystem and another of the $|1\rangle - |3\rangle$ subsystem. The angles of rotation are proportional to $\tau_{\text{pulse}}$, $\Delta$ and $\Delta'$, respectively:

$$
\rho'' = \mathcal{U}_Z \rho' \mathcal{U}_Z^\dagger,
$$

(A.5)

with

$$
\mathcal{U}_Z = \exp \left( \frac{i}{2} \Delta (|1\rangle \langle 1| - |2\rangle \langle 2| + \Delta' |1\rangle \langle 1| - |3\rangle \langle 3|) \right) \tau_{\text{pulse}}.
$$

(A.6)

Finally, we account for spontaneous decay of state $|2\rangle$ during the pulse period by introducing the Kraus operators $\mathcal{N}, \mathcal{D}$ describing this process:

$$
\rho'' = \mathcal{N} \rho'' \mathcal{N}^\dagger + \mathcal{D} \rho'' \mathcal{D}^\dagger,
$$

(A.7)

with

$$
\mathcal{N} = |1\rangle \langle 1| + \sqrt{1-p-q} |2\rangle \langle 2| + |3\rangle \langle 3|,
$$

(A.8)

and

$$
\mathcal{D} = \sqrt{p} |1\rangle \langle 2| + \sqrt{q} |3\rangle \langle 2|,
$$

(A.9)

where $p = 1 - \exp (-\Gamma_{12} \tau_{\text{pulse}})$ and $q = 1 - \exp (-\Gamma_{23} \tau_{\text{pulse}})$. Calculating the decay of the excited state only after applying the $x$- and $z$-rotations operator is acceptable, since the pulse length of 2 ps is very short compared to the state’s lifetime of 6.9 ns.

To find the density operator $\rho_n$ after a train of $n$ pulses we iteratively apply these operators $n$ times

$$
\rho_n = \mathcal{N} \mathcal{U}_Z \mathcal{U}_R \rho_{n-1} \mathcal{U}_R^\dagger \mathcal{U}_Z^\dagger + \mathcal{D} \mathcal{U}_Z \mathcal{U}_R \rho_{n-1} \mathcal{U}_R^\dagger \mathcal{U}_Z^\dagger \mathcal{D}^\dagger
$$

(A.10)

and finish the calculation by letting state $|2\rangle$ decay completely. From the final density operator $\rho_n$ we can easily calculate experimentally accessible observables such as populations $\text{Tr}(|1\rangle \langle 1| \rho_n)$, $\text{Tr}(|3\rangle \langle 3| \rho_n)$ and coherences $\text{Tr}(|1\rangle \langle 3| \rho_n)$, $\text{Tr}(|3\rangle \langle 1| \rho_n)$. Contrast $C$ and relative phase $\Phi$, which were introduced in equation (3), can be calculated using

$$
C = \gamma \sqrt{\text{Tr}(\sigma_{x}^{13} \rho) + \text{Tr}(\sigma_{y}^{13} \rho)}
$$

(A.11)

and

$$
\Phi = \text{arg}[\text{Tr}(\sigma_{x}^{13} \rho) + i\text{Tr}(\sigma_{y}^{13} \rho)],
$$

(A.12)

with $\sigma_{x}^{13} = |3\rangle \langle 1| + |1\rangle \langle 3|$ and $\sigma_{y}^{13} = i(|3\rangle \langle 1| - |1\rangle \langle 3|)$, and where $\gamma$ is an additional scaling factor to account for a reduced contrast due to experimental imperfections.

In section 2.2.3, we observed that the first pulse in a pulse train possibly has a different (usually higher) peak power and a phase difference with respect to later pulses. To accommodate these observations, we allow for the first pulse to have a different rotation angle $\theta^{01}$ and phase $\Delta\phi$ and replace $\mathcal{U}_R$ with $\mathcal{U}_R^{01}$ in equation (A.4):
\[ \mathcal{U}_R \rightarrow \mathcal{U}_R^{\text{int}} = \exp \left( \frac{i}{2} \theta^{\text{int}} (e^{i\Delta \phi} |2\rangle \langle 1| + e^{-i\Delta \phi} |1\rangle \langle 2|) \right). \] (A.13)

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