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## QUANTUM INFORMATION PROCESSING AND RAMSEY SPECTROSCOPY WITH TRAPPED IONS

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High-resolution laser spectroscopy and quantum information processing have a great deal in common. For both applications, ions held in electromagnetic traps can be employed, the ions' quantum state being manipulated by lasers. Quantum superposition states play a key role, and information about the experiment is inferred from a quantum state measurement that projects the ions' superposition state onto one of the basis states. In this paper, we discuss applications of Ramsey spectroscopy for quantum information processing and show that techniques developed in the context of quantum information processing find useful applications in atomic precision spectroscopy.

*Keywords*: Trapped ions, quantum information processing, precision spectroscopy, Ramsey spectroscopy, entanglement

#### 1. Introduction

Single trapped and laser-cooled ions held in radio-frequency traps constitute a quantum system offering an outstanding degree of quantum control. The ions' internal as well as external quantum degrees of freedom can be controlled by coherent laser-atom interactions with high accuracy. At the same time, the ions are well isolated against detrimental influences of a decohering environment. The combination of these two properties have enabled spectacular ion trap experiments aiming at building better atomic clocks,<sup>1–4</sup> creating entangled states<sup>5,6</sup> and processing quantum information.<sup>7,8</sup>

At a first glance, the construction of a quantum computer and of an atomic clock might not seem to have much in common. However, there are close ties linking the two fields of research. The implementation of en $\mathbf{2}$ 

tangling quantum gates $^{9-13}$  with ultra-high fidelity necessitates a precise knowledge of the Hamiltonian governing the dynamics of the atomic system and its interaction with the laser beams applied for steering it. Equally important are the characterization of decohering or dephasing mechanisms arising from the interaction of the atoms with fluctuating electromagnetic fields. For this task, Ramsey spectroscopy turns out to be an extremely important tool. In an atomic clock, the transition frequency between two atomic levels is measured by exciting the atom with laser pulses. If the excitation is done in a Ramsey experiment, probing of the clock transition can be described in the language of quantum information processing as a phase estimation algorithm. For this purpose, the use of multi-particle entangled states has been shown to be of interest.<sup>14,15</sup> In addition, entangling interactions have found applications in atomic clock measurements for quantum state detection of a system that is otherwise difficult to measure  $^{16}$  and for the detection of small energy level shifts by preparing a system of two ions in a manifold of entangled states that are part of a decoherence-free subspace. $^{17}$ 

In the first part of this paper, generalized Ramsey experiments investigating ion-ion couplings which are important in the context of high-fidelity quantum gates will be presented. In its second part, experiments aiming at making quantum information processing more robust against environmental noise will be discussed. We will show how to apply (quantum mechanically) correlated states of two ions for precision measurements of atomic constants. These ion-trap experiments demonstrate high-precision spectroscopy in a decoherence-free subspace using a pair of calcium ions for a determination of energy level shifts and transition frequencies in the presence of phase noise. For the measurement, maximally entangled ions are advantageous for achieving a good signal to noise ratio. As the preparation of these states is more involved than single-ion superposition states, we explore the possibility of using classically correlated ions for achieving long coherence times.

## 2. Experimental setup

In our experiments, two <sup>40</sup>Ca<sup>+</sup> ions are confined in a linear Paul trap with radial trap frequencies of about  $\omega_{\perp}/2\pi = 4$  MHz. By varying the trap's tip voltages from 500 to 2000 V, the axial center-of-mass frequency  $\omega_z$  is changed from 860 kHz to 1720 kHz. The ions are Doppler-cooled on the  $S_{1/2} \leftrightarrow P_{1/2}$  transition. Sideband cooling on the  $S_{1/2} \leftrightarrow D_{5/2}$  quadrupole transition<sup>18</sup> prepares the stretch mode in the motional ground state  $|0\rangle_s$ . Simultaneous cooling of stretch and rocking modes is accomplished by alternating the frequency of the cooling laser exciting the quadrupole transition between the different red motional sidebands. Motional quantum states are coherently coupled by a laser pulse sequence exciting a single ion on the  $|S\rangle \equiv S_{1/2}(m = -1/2) \leftrightarrow |D\rangle \equiv D_{5/2}(m = -1/2)$  transition with a focused laser beam on the carrier and the blue sideband. Internal state superpositions  $(|S\rangle + e^{i\phi}|D\rangle)|0\rangle$  can be mapped to motional superpositions  $|D\rangle(|0\rangle + e^{i\phi}|1\rangle)$  by a  $\pi$  pulse on the blue motional sideband and vice versa. We discriminate between the quantum states  $S_{1/2}$  and  $D_{5/2}$  by scattering light on the  $S_{1/2} \leftrightarrow P_{1/2}$  dipole transition and detecting the presence or absence of resonance fluorescence of the individual ions with a CCD-camera. A more detailed account of the experimental setup is given in Ref.<sup>10,19</sup>

# 3. Ramsey spectroscopy techniques for quantum information processing

In trapped ion quantum computing, continuous quantum variables occur in the description of the joint vibrational modes of the ion string. The normal mode picture naturally appears when the ion trap potential is modelled as a harmonic (pseudo-)potential and the mutual Coulomb interaction between the ions is linearized around the ions' equilibrium positions.<sup>20</sup> In this way, the collective ion motion is described by a set of independent harmonic oscillators with characteristic normal mode frequencies. The normal modes are of vital importance for all entangling quantum gates as they can give rise to effective spin-spin couplings in laser-ion interactions.<sup>9</sup> All entangling ion trap quantum gates demonstrated so far use laser beams that intermittently entangle the internal states of the ion with a vibrational mode of the ion string. At the end of the interaction, the vibrational mode returns to its initial state and the propagator describing the entangling gate operations is an operator acting only on the ions' internal degrees of freedom. In most gate operations, the fidelity of the gate suffers if the vibrational state of the ion string couples to an environment that heats or dephases the ion motion.

In previous experiments investigating the coherence of the center-ofmass mode of a two-ion crystal, we had observed heating rates of about 100 ms/phonon and coherence times for superpositions  $|0\rangle + |1\rangle$  of vibrational states of about the same order of magnitude. For the coherence measurement, a Ramsey experiment was performed where first a carrier  $\pi/2$  pulse was applied to the ions in state  $|S\rangle|S\rangle|0\rangle$ , followed by a  $\pi$ -pulse on the blue sideband of the center-of mass mode in order to create the state  $|S\rangle|D\rangle(|0\rangle + |1\rangle)$ . After a variable delay  $\tau$ , the inverse pulse sequence mapped the state  $|S\rangle|D\rangle(|0\rangle + e^{i\phi}|1\rangle)$  onto a superposition  $|S\rangle(\cos(\phi - \phi_0)|S\rangle + \sin(\phi - \phi_0)|D\rangle)|0\rangle$ . A Ramsey fringe pattern was recorded by scanning the phase  $\phi_0$  which was achieved by switching either the phase of the second blue sideband pulse or the phase of the second carrier pulse with respect to the phase of the corresponding first pulse. Surprisingly, when this kind of Ramsey experiment was applied to investigate the coherence of the stretch mode of the two-ion crystal, the measured coherence time was found to be nearly two orders of magnitude shorter than for the center-of-mass mode. Fig. 1 (a) shows the contrast  $C(\tau)$  of the Ramsey fringe pattern as a function of the delay time. In this experiment, a coherence time of less than 2 ms was measured at a trap frequency  $\omega/(2\pi) = 1486$  kHz. We found that the loss of contrast could be attributed to the nonlinear terms in the Coulomb interaction between the ions giving rise to a cross-coupling between the normal modes.<sup>21</sup> For a two-ion crystal, this leads to a dispersive coupling between the stretch mode and the rocking mode where the ions oscillate out of phase in the transverse direction. As a result, the bare stretch mode frequency  $\nu_{str}^{(0)}$  is lowered slightly by an amount that is proportional to the number of rocking mode phonons so that  $\nu_{str} = \nu_{str}^{(0)} - \chi n_{rock}$ . After cooling the rocking modes to the ground state



Fig. 1. Experiments probing the coherence of the stretch mode. (a) Ramsey experiment. (b) Spin echo experiment.

before repeating the experiment shown in Fig. 1 (a), we observed coherence times similar to the ones found for the center-of-mass mode. The fact that the stretch mode frequency is varying from experiment to experiment for  $\langle n_{rock} \rangle \neq 0$  but constant within a single experiment is also revealed by a spin echo experiment probing the stretch mode coherence. For the experiment shown in Fig. 1 (b), a pulse sequence similar to the sequence for (a) is used, but with additional pulses in the middle of the sequence that swap the population of the two lowest quantum states of the stretch mode. This makes the experiment insensitive against small changes of the stretch mode frequency so that the contrast decays to half of its initial value only after 100 ms. To confirm that the observed spread in vibrational frequencies is indeed due to the postulated mechanism, we could even measure the shift induced by a single rocking mode phonon by performing a spin echo experiment and increasing the rocking phonon number by exactly one at the beginning of the second spin echo period by a blue sideband pulse on the rocking mode. The extra phonon shifts the Ramsey fringe pattern by an amount that can be related to the strength  $\chi$  of the cross mode coupling. In our experiments, we find frequency shifts of up to 20 Hz per phonon.<sup>21</sup> These shifts dramatically reduce the fidelity of Cirac-Zoller gate operations making use of the stretch mode as long as the rocking modes are cooled only to the Doppler limit.<sup>22</sup> For the realization of high-fidelity quantum gate operations, this observation points to the necessity of either cooling the rocking modes to the ground state or using the center-of-mass mode for mediating the ion-ion coupling.

# 4. Quantum information processing techniques for precision spectroscopy

In atomic high-resolution spectroscopy, dephasing is often the most important factor limiting the attainable spectral resolution. Possible sources of dephasing are fluctuating electromagnetic fields giving rise to random energy level shifts but also the finite spectral linewidth of probe lasers. Under these conditions, two atoms located in close proximity to each other are likely to experience the same kind of noise, i.e. they are subject to collective decoherence. The collective character of the decoherence has the important consequence that it does not affect the entangled two-atom state

$$\Psi_{+} = \frac{1}{\sqrt{2}} \left( |g\rangle|e\rangle + |e\rangle|g\rangle \right),\tag{1}$$

as both parts of the superposition are shifted by the same amount of energy by fluctuating fields. Here, for the sake of simplicity,  $|g\rangle$  and  $|e\rangle$  denote the ground and excited state of a two-level atom. Because of its immunity against collective decoherence, the entangled state  $\Psi_+$  is much more robust than a single-atom superposition state  $\frac{1}{\sqrt{2}}(|g\rangle + |e\rangle)$ . This properties makes states like  $\Psi_+$  interesting candidates for high-precision spectroscopy. In the following, we will first discuss how to use Bell states for the measurement

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of energy level shifts. Then, it will be shown that certain unentangled twoatom states can have similar advantages over single atom superposition states albeit at lower signal-to-noise ratio.

#### 4.1. Spectroscopy with entangled states

In a Ramsey experiment, spectroscopic information is inferred from a measurement of the relative phase  $\phi$  of the superposition state  $\frac{1}{\sqrt{2}}(|g\rangle + e^{i\phi}|e\rangle)$ . The phase is measured by mapping the states  $\frac{1}{\sqrt{2}}(|g\rangle \pm |e\rangle)$  to the measurement basis  $\{|g\rangle, |e\rangle\}$  by means of a  $\pi/2$  pulse. In close analogy, spectroscopy with entangled states is based on a measurement of the relative phase  $\phi$ of the Bell state  $\Psi_{\phi} = \frac{1}{\sqrt{2}}(|g\rangle|e\rangle + e^{i\phi}|e\rangle|g\rangle)$ . Here, the phase is determined by applying  $\pi/2$  pulses to both atoms followed by state detection.  $\pi/2$  pulses with the same laser phase on both atoms map the singlet state  $\frac{1}{\sqrt{2}}(|g\rangle|e\rangle - |e\rangle|g\rangle)$  to itself whereas the triplet state  $\frac{1}{\sqrt{2}}(|g\rangle|e\rangle + |e\rangle|g\rangle)$  is mapped to a state  $\frac{1}{\sqrt{2}}(|g\rangle|g\rangle + e^{i\alpha}|e\rangle|e\rangle)$  with different parity. Therefore, measurement of the parity operator  $\sigma_z^{(1)}\sigma_z^{(2)}$  yields information about the relative phase since  $\langle \sigma_z^{(1)}\sigma_z^{(2)} \rangle = \cos \phi$ . If the atomic transition frequencies are not exactly equal but differ by an amount  $\delta$ , the phase will evolve as a function of time  $\tau$  according to  $\phi(\tau) = \phi_0 + \delta \tau$ . Then, measurement of the phase evolution rate provides information about the difference frequency  $\delta$ . To keep the notation simple, it was assumed that in both atoms the same energy levels participated in the superposition state of eq. (1). In general, this does not need to be the case and the phase evolution is given by  $\phi(\tau) = ((\omega_{A_1} - \omega_{L_1}) \pm (\omega_{A_2} - \omega_{L_2}))\tau$ . Here,  $\omega_{A_{1,2}}$  denote the atomic transition frequencies of atom 1 and atom 2, and  $\omega_{L_{1,2}}$  are the laser frequencies used for exciting the corresponding transitions. The minus sign applies if in the Bell state the ground state of atom 1 is associated with an excited state of atom 2 and vice versa. If the Bell state is a superposition of both atoms being in the ground state or both in the excited state, the plus sign is appropriate.

### 4.2. Spectroscopy with unentangled states of two atoms

One may wonder whether entanglement is absolutely necessary for observing long coherence times in experiments with two atoms. In fact it turns out that the kind of measurement outlined above is applicable even to completely unentangled atoms.<sup>23</sup> If the atoms are initially prepared in the

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product state

$$\Psi_{p} = \frac{1}{2} (|g\rangle + |e\rangle) \otimes (|g\rangle + |e\rangle)$$

$$= \frac{1}{\sqrt{2}} \Psi_{+} + \frac{1}{2} |g\rangle |g\rangle + \frac{1}{2} |e\rangle |e\rangle,$$
(2)

this state will quickly dephase under the influence of collective phase noise. The resulting mixed state

$$\rho_p = \frac{1}{2} |\Psi_+\rangle \langle \Psi_+| + \frac{1}{4} |gg\rangle \langle gg| + \frac{1}{4} |ee\rangle \langle ee|$$
(3)

appears to be composed of the entangled state  $\Psi_+$  with a probability of 50% and the two states  $|gg\rangle$  and  $|ee\rangle$  with 25% probability each. If the state  $\Psi_+$  is replaced by the density operator  $\rho_p$  in the measurement procedure described in subsection 4.1, the resulting signal will be the same apart from a 50% loss of contrast. The states  $|g\rangle|g\rangle$  and  $|e\rangle|e\rangle$  do not contribute to the signal since they become equally distributed over all four basis basis states by the  $\pi/2$  pulses preceding the state detection. Their only effect is to reduce the signal-to-noise ratio by adding quantum projection noise since only half of the experiments effectively contribute to the signal.



Fig. 2. Parity oscillation caused by the interaction of a static electric field gradient with the quadrupole moment of the  $D_{5/2}$  state of  ${}^{40}\text{Ca}^+$ . The first data point significantly deviates from the fit since the quantum state has not yet decayed to a mixed quantum state.

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Fig. 3. Electric quadrupole shift measured with a pair of atoms in a product state. (a) The shift varies linearly with the applied electric field gradient. (b) Residuals of the electric quadrupole shift measurements. The plot shows deviations of the data points measured with unentangled ion (open circles) and entangled ions (filled circles) with respect to the fit obtained from the entangled state data.

## 4.3. Measurement of an electric quadrupole moment

We applied the method outlined in subsection 4.2 to a measurement of the quadrupole moment of the  $D_{5/2}$  state. For this, we prepared the state

$$\Psi_p = \frac{1}{2} (|-5/2\rangle + |-1/2\rangle) \otimes (|+3/2\rangle + |-1/2\rangle) \tag{4}$$

and let it decohere for a few milliseconds. Here,  $|m\rangle\equiv|D_{5/2},m\rangle$  denotes the Zeeman sub-level of  $D_{5/2}$  with magnetic quantum number m. After a waiting time ranging from 0.1 to 200 ms,  $\pi/2$  pulses were applied and a parity measurement performed. Fig. 2 shows the resulting parity oscillation whose contrast decays over a time interval orders of magnitude longer than any single atom coherence time in <sup>40</sup>Ca<sup>+</sup>. A sinusoidal fit to the data reveals an initial contrast of 48(6)% and an oscillation frequency  $\nu = 38.6(3)$  Hz. For the fit, the first data point at  $t=0.1\mu$ s is not taken into account. At this time, the quantum state cannot yet be described by a mixture similar to the one of eq. (3) as some of the coherences persist for a few milliseconds and thus affect the parity signal. The parity signal decays exponentially with a time constant  $\tau_d = 730(530)$  ms that is consistent with the assumption of spontaneous decay being the only source of decoherence (in this case, one would have  $\tau_d = \tau_{D_{5/2}}/2 \approx 580$  ms where  $\tau_{D_{5/2}}$  is the lifetime of the metastable state). The quadrupole moment is determined by measuring the quadrupole shift as a function of the electric field gradient E'. The latter is conveniently varied by changing the voltage applied to the axial trap electrodes. For a calibration of the gradient, the axial oscillation frequency of the ions is measured. Further details regarding the measurement procedure are provided in ref.<sup>17</sup> Fig. 3(a) shows the quadrupole shift  $\Delta \nu_{OS}$ as a function of the field gradient (the small offset at E' = 0 is caused by the second-order Zeeman effect). By fitting a straight line to the data, the quadrupole moment can be calculated provided that the angle between the orientation of the electric field gradient and the quantization axis is known. Setting  $\Delta \nu_{QS} = \alpha E'$ , the fit yields the proportionality constant  $\alpha = 2.977(11) \text{ Hz/(V/mm^2)}$ . The quadrupole shift had been previously measured using a pair of ions in an entangled state. Both measurement give consistent results and thus confirm the validity of the approach based on correlated, unentangled atoms.

#### 5. Conclusion

Techniques developed for atomic clock measurements turn out to be very useful for precisely characterizing quantum interactions in a system of trapped ions dedicated to quantum information processing. For the realization of ultra-high fidelity quantum gates even small effect like the crosscoupling between vibrational modes that is not predicted by the simple normal mode picture become important. Quantum gates based on nonresonant excitations of vibrational sidebands are less affected than those relying on a resonant excitation. Still, to approach the precision required 10

for fault-tolerant quantum operations might require cooling all modes to the ground state. On the other hand, precision spectroscopy itself can profit from concepts developed for processing of quantum information by making use of more advanced detection schemes.

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