Spontaneous Emission Lifetime of a Single Trapped Ca⁺ Ion in a High Finesse Cavity

A. Kreuter, C. Becher,* G. P.T. Lancaster, A. B. Mundt, C. Russo, H. Häffner, C. Roos, J. Eschner,† F. Schmidt-Kaler, and R. Blatt

Institut für Experimentalphysik, Universität Innsbruck, Technikerstraße 25, A-6020 Innsbruck, Austria (Received 22 December 2003; published 19 May 2004)

We investigate the spontaneous emission lifetime of a single trapped $^{40}\text{Ca}^+$ ion placed at different positions in the vacuum standing wave inside a high finesse cavity which is stabilized to the atomic transition. The lifetime is measured by quantum state detection after π -pulse excitation. The result for the natural lifetime of the $D_{5/2}$ metastable state of 1161(22) ms agrees, within 1 standard deviation, with the most precise published value. We observe a reduction of the spontaneous emission lifetime of $\approx 15\%$ in the node of the vacuum field.

DOI: 10.1103/PhysRevLett.92.203002 PACS numbers: 32.70.Cs, 03.67.Lx, 42.50.Pq

The rapid and promising development in the field of quantum information processing in recent years is based on the ability to control and manipulate single quantum systems. Among these, trapped ions have proven to constitute a model system for storing and processing quantum information. The transport of this information within distributed quantum networks [1] requires an interface between trapped ions and photons operating as moving quantum bits. Such an interface could be based on the deterministic coupling of a single atom or ion to a high finesse optical cavity [2,3], which requires the ability to precisely and stationary place the atom at a fixed position within the cavity field. So far, such deterministic coupling has been demonstrated only for intense light fields [2,3]. However, the transport of quantum information implies coupling of an atomic quantum bit to the cavity vacuum field, which in turn modifies the spontaneous emission properties of the atom. To demonstrate the feasibility of this approach, we investigate the stationary interaction of a single trapped Ca⁺ ion with the vacuum field inside an optical cavity by measuring the modification of the spontaneous emission lifetime of the metastable $D_{5/2}$ level (≈ 1 s) at various positions within the cavity. Because of the good localization of the ion we are able to map the standing-wave vacuum field.

The enhancement or inhibition of spontaneous emission due to the modification of the vacuum field by a resonator has long been predicted by Purcell [4] and Kleppner [5]. There have been experimental demonstrations with *ensembles of emitters* coupled to resonant structures [6,7] and with few or *single emitters* coupled to cavities: a single electron trapped in a microwave cavity [8], Rydberg atoms traversing microwave cavities [9,10], dilute atom beams traversing optical resonators [11,12], a single trapped ion interacting with a single mirror [13], and a single semiconductor quantum dot coupled to a microcavity [14,15]. Among these, only the experiments with ions and quantum dots have been carried out with one and the same single quantum emitter. In addition, besides the work involving a trapped ion [13],

these experiments lack either the stationary coupling or the deterministic control of the emitter position with respect to the resonator mode. In our experiment, the motional wave packet of a laser-cooled trapped ion is confined to a region much smaller than the optical wavelength λ (to approximately $\lambda/50$), and its position within a cavity standing-wave (SW) field is controlled with a precision of up to 7 nm [3].

The lifetimes of the metastable D levels of $^{40}\text{Ca}^+$ have been subject to several investigations, both theoretical and experimental, because of their high relevance to frequency standards [16] and atomic structure theory. All previous lifetime measurements of the $D_{5/2}$ level of single $^{40}\text{Ca}^+$ ions employed the quantum jump technique (see [17], and references therein). This technique is based on monitoring the fluorescence on the $S_{1/2}$ - $P_{1/2}$ dipole transition [see Fig. 1(a)], while at random times the ion is shelved to the metastable state where the fluorescence falls to the background level. Statistical analysis of these dark times yields the lifetime τ . The most precise measurement using this technique resulted in $\tau = 1168(7)$ ms [17].

Here, we introduce a new measurement technique based on coherent excitation and quantum state detection. The quantum jump method is not appropriate in our experiment as the 397 nm light that is used to monitor the fluorescence would broaden the $S_{1/2}$ ground state by dipole coupling to far beyond the linewidth of the cavity, and hence the vacuum effect would become negligible. Instead, we use an improved version of a technique that was used to measure the $D_{3/2}$ metastable level lifetime of single Ba⁺ ions [18]. The method is to first excite the ion deterministically with a π pulse and then measure the remaining excitation after a fixed waiting period, during which all lasers are shut off. The main advantage of this "state detection" method is that no residual light is present during the measurement which could affect the free decay of the atom. Thus we are able to measure the free-space lifetime with high precision. For the measurement of the cavity-modified lifetime, however, we cannot

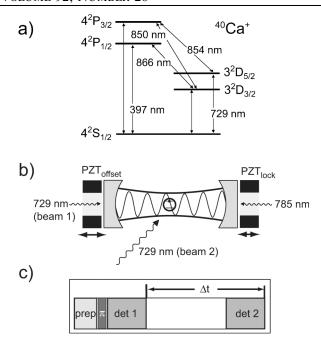


FIG. 1. (a) Ca⁺-level scheme with relevant transitions. (b) Sketch of the experimental setup with a single ion in the standing wave of a two-mirror near-confocal cavity at 729 nm. Laser beam 1 is used to measure the ion's position in the SW; beam 2 prepares the ion in the D state by a π pulse. (c) Schematic pulse sequence of one experimental cycle for the lifetime measurement (see text for details).

avoid residual light. The far off-resonant transfer laser light that is needed to keep the cavity frequency locked causes additional deshelving of the $D_{5/2}$ state by admixing a small fraction of the fast-decaying $P_{3/2}$ level. This effect has been carefully measured and is described below.

The experimental setup is shown schematically in Fig. 1 and is described in detail elsewhere [3,19]. The general experimental procedure starts with loading a single ⁴⁰Ca⁺ ion into a spherical Paul trap with radial and axial secular frequencies $(\omega_{\rm rl}, \omega_{\rm r2}, \omega_{\rm ax}) = 2\pi(1.9,$ 2.6, 4.5) MHz. The trap is located inside a near-confocal resonator with a finesse of $\mathcal{F} \approx 35000$ at 729 nm and a waist of 54 μ m. The mirrors are both mounted on piezoelectric translators (PZT) to allow for independent movement. The cavity is frequency stabilized using the Pound-Drever-Hall technique to a transfer laser (extended cavity diode laser at ≈ 785 nm) [19]. The transfer laser is frequency stabilized to the same ultrastable reference cavity as the Ti:Sapphire laser used to drive the 729 nm transitions. By an appropriate frequency tuning of the transfer laser with an acousto-optical modulator (AOM), the cavity is made resonant for both the transfer laser and the 729 nm laser [beam 1 in Fig. 1(b)]. This ensures that the cavity is resonant with the quadrupole transition without using resonant light.

The lifetime measurements described here consist of a repetition of a laser pulse sequence applied to the ion. The

sequence generally is composed of three steps [see Fig. 1(c)]: (i) state preparation and Doppler cooling consisting of 2 ms of Doppler cooling (397 and 866 nm light), repumping from the $D_{5/2}$ level (854 nm light), and optical pumping into the $S_{1/2}(m = -1/2)$ Zeeman sublevel (397 nm σ^+ polarized light), (ii) coherent excitation at 729 nm [beam 2 in Fig. 1(b)], with pulse length and intensity chosen to obtain near unity excitation (π pulse) to the $D_{5/2}(m = -5/2)$ Zeeman level, and (iii) state detection for 3.5 ms by recording the fluorescence on the $S_{1/2}$ - $P_{1/2}$ transition with a photomultiplier to discriminate between the states S and D. The state is measured before and after a fixed waiting period Δt between 10 and 500 ms to determine whether a decay of the excited state has occurred. This three-step cycle [steps (i)-(iii)] is repeated typically several thousand times to yield the decay probability p. For the calculation of the lifetime τ we use an exponential fit function 1-p= $A \exp\{-\Delta t/\tau\}$. For Δt we use the time interval between the ends of the two detection periods. Poissonian noise of the count rate or decay of the atom during the detection period can lead to a small error in the quantum state detection [20]. We model this error as a deviation of the fit parameter A from its ideal value of one.

As a first step, the natural free-space lifetime of the $D_{5/2}$ level is determined without the cavity influence to assure the accuracy of our measuring scheme and to exclude possible inconsistencies and systematic errors. The results for various waiting periods between 10 and 500 ms, based on several 10⁵ decay measurements each, are displayed in Fig. 2. The exponential fit function described above yields a lifetime of 1161(22) ms, in good agreement with earlier results [17] (the number in parentheses being the 1σ confidence level). The fit also verifies that the decay probability satisfies an exponential law. For the data in Fig. 2 we find A = 0.99939(50), indicating that the error in quantum state detection for the given length of detection periods is only $6(5) \times 10^{-4}$. We stress that this lifetime measurement is an independent check of earlier results since we used a different measurement technique.

We note that the lifetime measurement was found to be extremely sensitive to any background radiation at the repump wavelength of 854 nm, which can either originate from residual light of the 854 nm diode laser itself, background fluorescence of the 866 nm diode laser, or residual light from the laboratory environment. Great care was taken to avoid these sources of systematic error. The 854 nm light is switched by a mechanical shutter in the beam path (40 dB attenuation by an AOM in double pass configuration was found not to be sufficient) and the 866 nm beam is shut off by an AOM in single pass (attenuation >30 dB) during the waiting time. Other possible effects that result in a lifetime reduction include collisional effects, such as quenching and *J*-mixing [21,22]. Collisional quenching effects are neglected on

203002-2 203002-2

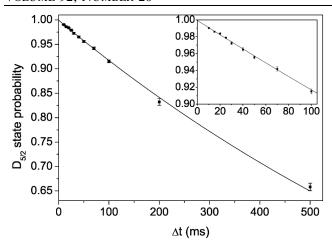


FIG. 2. Decay of the $D_{5/2}$ level as a function of waiting time. An exponential fit (solid line) yields the natural lifetime.

grounds of low pressures ($<3 \times 10^{-10}$ mbar). *J*-mixing is neglected as measurements using coherent excitation and deexcitation after 100 ms did not indicate any decoherence beyond the expected spontaneous decay.

As our state detection technique yields consistent lifetime results, we can now use it to investigate the influence of the cavity vacuum field on the atomic lifetime. For lifetime measurements with the frequency-stabilized cavity, the largest systematic error is due to the presence of transfer laser light at 785 nm. To investigate its influence, the $D_{5/2}$ decay rate was measured as a function of the ac Stark shift of the $D_{5/2}$ level induced by the transfer laser (Fig. 3). We determined the ac Stark shift by excitation spectroscopy on the $S_{1/2}$ - $D_{5/2}$ transition and use it as a measure for the intracavity power of the transfer laser since the latter is difficult to determine. The linear fit in Fig. 3 yields the additional deshelving rate of $0.0027 \, \mathrm{s}^{-1}/\mathrm{kHz}$ on the $D_{5/2}$ - $P_{3/2}$ transition due to the transfer laser light.

To measure the spatial dependence of the enhanced spontaneous emission, we frequency stabilize the cavity as described above, using the minimum possible transfer laser power (approximately 20 mW intracavity power) to keep the frequency lock stable. The SW pattern is measured by exciting the ion with short pulses at 729 nm (pulse length approximately equal to the $\pi/2$ time at the node) through the locked cavity [beam 1 in Fig. 1(b)] and varying the voltage of the offset PZT, resulting in the phase of the SW being shifted with respect to the spatially fixed ion (see inset of Fig. 4). To place the ion at a specific position in the SW, we then apply the corresponding offset voltage. This procedure is done before and after each lifetime measurement (a few 1000 sequences) to account for thermal drifts. Several measurements over around 15 h showed that the drift is linear over several hours and corresponds to a displacement by $\lambda/4$ in 3.9 h. Each measurement was limited to 19 min (2 \times 10⁴ single experiments), yielding a position uncertainty of $\approx \lambda/50$.

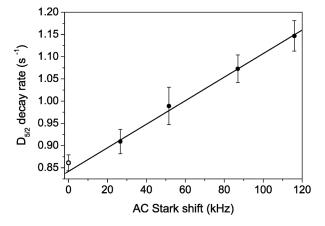


FIG. 3. Total decay rate from the $D_{5/2}$ state versus ac Stark shift on the $S_{1/2}$ - $D_{5/2}$ transition induced by the transfer laser. The open dot represents the decay rate in free space measured without transfer laser.

The measurements of the cavity-modified lifetime were performed with a fixed waiting time $\Delta t = 50$ ms. The raw data from many experimental runs are combined to yield the results of the lifetime measurement at five different points in the vacuum SW, shown in Fig. 4. The indicated lifetime errors are the 1σ statistical errors, while the errors in the phase result from the deviation of the SW phase in the individual measurements. Note that the drift during the time of the measurement (15 nm in 19 min) is not a systematic error but corresponds to an averaging over the sinusoidal variation and a loss of contrast. In addition, the spatial extension of the ion's wave packet (on the order of 20 nm, taking into account a heating rate of 0.1 ms⁻¹ during 50 ms) also leads to a loss of contrast [23]. The combined effect yields a visibility of the SW of $V \approx 90\%$. To exclude any residual

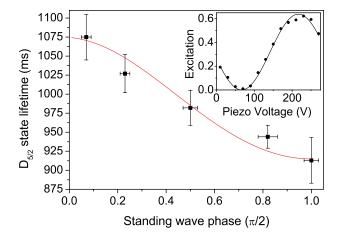


FIG. 4 (color online). $D_{5/2}$ state lifetime measured at various positions in the cavity vacuum standing-wave field. The solid line shows a simulation of the Purcell effect, assuming $\tau=1075$ ms, a Purcell factor F=1.175, and a visibility of V=0.9. The inset shows a measurement of an intense cavity standing wave by coherent excitation (c.f. [3,19]).

203002-3 203002-3

systematic errors (lifetime reduction due to effects other than the cavity field), the lifetime is also measured off resonance with the cavity shifted more than ten linewidths away from resonance by changing the frequency of the transfer laser by 2 MHz. This off-resonance lifetime was 1069(37) ms. The ac Stark shift for these measurements was maximally 16 kHz, which results in an additional deshelving rate of 0.04 s^{-1} . This appears as a <5% lifetime reduction, which varies sinusoidally with the transfer laser SW in the cavity. This transfer laser SW has, in general, a different phase than the vacuum SW, which was not determined independently for the cavity lifetime measurement. Thus each data point in Fig. 4 has an additional error of $_{-0}^{+5}\%$ due to the additional deshelving induced by the transfer laser. It is important to point out that the observed lifetime reduction is a genuine cavity QED effect, consistent with the measured phase of the vacuum SW, and can not be explained only by the deshelving effect of the transfer laser.

The observed maximum lifetime reduction in the node of the vacuum field is $15 \pm 5\%$. With our experimentally determined parameters, i.e., ion-field coupling constant $g = 2\pi \times 120$ Hz, cavity decay rate $\kappa = 2\pi \times 102$ kHz, and spontaneous emission rate $\gamma = 1/\tau = 2\pi \times$ 0.137 Hz, we calculate the cooperativity parameter [24] $C_0 = g^2/2\kappa\gamma = 0.52$ and a Purcell factor [24] F = $2C_0 + 1 = 2.04$, which should yield a 50% lifetime reduction [3]. However, there are several experimental imperfections that contribute to the reduction of the expected effect. First, disturbance (acoustical, etc.) of the cavity lock leads to an inhomogeneous broadening of the cavity linewidth. The resulting effective finesse found from scanning the 729 nm laser slowly over the locked cavity resonance is $\mathcal{F}_{\rm eff} \approx 22\,000$, which reduces the cooperativity to $0.62C_0$. Second, the coupling is reduced by a suboptimal lateral position of the ion in the waist of the cavity mode. This position has been optimized by moving the trap mount with micrometer screws and recording Rabi oscillation frequencies driven through the cavity field at every position. This positioning achieves an estimated precision of 20 μ m, resulting in a reduced cooperativity of $0.57C_0$. Taking into account the SW visibility V = 90%, as discussed above, the total effective cooperativity is $C_{\rm eff} = 0.62 \times 0.57 \times 0.9 \times$ $C_0 = 0.165$, corresponding to an expected 25% lifetime reduction. In the antinode of the cavity SW (SW phase = 0 in Fig. 4), the lifetime τ_a should be approximately equal to the free-space lifetime ($\tau_a = 0.98\tau = 1138 \text{ ms}$). However, in our experiment we measure a maximum lifetime of $\tau_a = 1075(30)$ ms in the cavity SW. Therefore we assume the following worst-case scenario: the transfer laser deshelving rate of 0.04 s⁻¹ leads to a reduction of the maximum observable lifetime in the SW antinode by $\approx 5\%$ (expected $\tau_a = 1089$ ms) and leaves the minimum lifetime in the SW node unchanged, yielding an expected 21% lifetime reduction.

In summary, we have demonstrated the deterministic coupling of a single ion to the vacuum field inside a high finesse cavity over an extended time. The spatial variation of the spontaneous emission rate has been investigated by measuring the $D_{5/2}$ state lifetime with a new method based on deterministic excitation and quantum state detection. As the position in the standing wave and the lifetime are measured independently, our experiment is a genuine demonstration of single-atom cavity QED.

This work is supported by the Austrian "Fonds zur Förderung der wissenschaftlichen Forschung" (SFB15), by the European Commission: IHP network "QUEST" (HPRN-CT-2000-00121), IST/FET Program "QUBITS" (IST-1999-13021), and the Marie-Curie-Program (H. H.), and by the "Institut für Quanteninformation GmbH." C. Russo acknowledges support by Fundação para a Ciência e a Tecnologia, Portugal, Grant No. SFRH/BD/6208/2001.

- *Electronic address: Christoph.Becher@uibk.ac.at †Present address: Institut for Photonic Sciences (ICFO), C/ Jordi Girona, 29, 08034, Barcelona, Spain.
- [1] J. I. Cirac et al., Phys. Rev. Lett. 78, 3221 (1997).
- [2] G. R. Guthöhrlein et al., Nature (London) 414, 49 (2001).
- [3] A. B. Mundt et al., Phys. Rev. Lett. 89, 103001 (2002).
- [4] E. M. Purcell, Phys. Rev. 69, 681 (1946).
- [5] D. Kleppner, Phys. Rev. Lett. 47, 233 (1981).
- [6] K. H. Drexhage, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1974), Vol. 12, p. 163.
- [7] F. DeMartini et al., Phys. Rev. Lett. **59**, 2955 (1987).
- [8] G. Gabrielse and H. Dehmelt, Phys. Rev. Lett. 55, 67 (1985).
- [9] P. Goy et al., Phys. Rev. Lett. 50, 1903 (1983).
- [10] R. G. Hulet, E. S. Hilfer, and D. Kleppner, Phys. Rev. Lett. 55, 2137 (1985).
- [11] W. Jhe et al., Phys. Rev. Lett. 58, 666 (1987).
- [12] D. J. Heinzen et al., Phys. Rev. Lett. 58, 1320 (1987).
- [13] J. Eschner et al., Nature (London) 413, 495 (2001).
- [14] A. Kiraz et al., Appl. Phys. Lett. 78, 3932 (2001).
- [15] G. S. Solomon, M. Pelton, and Y. Yamamoto, Phys. Rev. Lett. 86, 3903 (2001).
- [16] R. J. Rafac et al., Phys. Rev. Lett. 85, 2462 (2000).
- [17] P. A. Barton et al., Phys. Rev. A 62, 032503 (2000).
- [18] N. Yu, W. Nagourney, and H. Dehmelt, Phys. Rev. Lett. 78, 4898 (1997).
- [19] A. B. Mundt et al., Appl. Phys. B 76, 117 (2003).
- [20] C. Roos, Ph.D. thesis, University of Innsbruck, 2000.
- [21] M. Knoop, M. Vedel, and F. Vedel, Phys. Rev. A 52, 3763 (1995); 58, 264 (1998).
- [22] P. Staanum et al., Phys. Rev. A 69, 032503 (2004).
- [23] J. Eschner, Eur. Phys. J. D 22, 341 (2003).
- [24] Q. A. Turchette, R. J. Thompson, and H. J. Kimble, Appl. Phys. B 60, 1 (1995).

203002-4 203002-4