Magnetically induced electron shelving in a trapped Ca^+ ion

D. R. Crick, S. Donnellan, D. M. Segal,* and R. C. Thompson

Blackett Laboratory, Imperial College London, Prince Consort Road, London SW7 2AZ, United Kingdom

(Received 13 August 2009; published 10 May 2010)

Atomic states are perturbed by externally applied magnetic fields (Zeeman effect). As well as the usual Zeeman splittings, the magnetic field leads to mixing of states with different values of the J quantum number. We report on the direct experimental measurement of this effect using the electron shelving technique (employed to great effect in single-ion spectroscopy and quantum-information processing). Specifically we observe shelving to the metastable $(3p^6 3d)\ ^2D_{5/2}$ state in a single $^{40}Ca^+$ ion, via spontaneous decay on the strongly forbidden $4p\ ^2P_{1/2} \leftrightarrow 3d\ ^2D_{5/2}$ transition. The rate of this transition is shown to scale as the square of the magnetic-field strength. The scaling and magnitude of the effect is compared to the result derived from first-order perturbation theory. For applications in quantum-information processing the J-mixing effect causes a degradation of readout fidelity. We show that this degradation is at a tolerable level for Ca^+ and is much less problematic for other trapped ionic species.

DOI: 10.1103/PhysRevA.81.052503 PACS number(s): 31.15.xp, 32.60.+i, 37.10.Rs, 37.10.Ty

I. INTRODUCTION

It is well known that atoms are perturbed when subjected to an externally applied magnetic field, the principal result being the Zeeman energy shift of atomic m_J sublevels (or m_F sublevels in the case of an atom with hyperfine structure) [1]. However the magnetic field also leads to a mixing of levels with different values of J. This can lead to some forbidden transitions becoming weakly allowed. In this paper we demonstrate that, for a single trapped Ca^+ ion, the use of Dehmelt's "electron shelving" technique [2] renders this very small degree of mixing readily discernible.

This J-mixing effect is important in a wide range of experiments in atomic physics. It has been demonstrated that applying a magnetic field can enable spectroscopy of otherwise strictly forbidden transitions in clouds of neutral atoms, which has direct relevance to atomic clocks [3,4]. The strong magnetic field present in ion storage rings has been shown to cause significant effects on atomic decay rates [5]. The effect also has an immediate importance in the context of proposals for performing quantum simulation or quantum-information processing (QIP) using ions held in a Penning trap [6–8]. Laser cooled ions held in rf traps have been used to demonstrate many key processes in QIP [9]. Complications that arise in laser cooling in the Penning trap have meant that it has not, so far, been employed for QIP studies. Recent work has shown, however, that it is possible to improve laser cooling in this system—the controlled motion of small crystals of ions in a Penning trap has been demonstrated [10,11], as well as the imaging of individual ions in large planar crystals [12]. In the context of QIP in a Penning trap the J-state mixing caused by the large magnetic field required for the trap acts as a systematic effect compromising the readout fidelity.

We calculate the degree of J-mixing for our system and show that it is expected to depend quadratically on the magnetic-field strength B. A comparison with our experimental data confirms this B^2 dependence. Furthermore we show that despite the process in question having remarkably clearly visible consequences, the actual effect on readout fidelity in QIP applications will be small for $^{40}Ca^+$ qubits. Moreover, we show that the deleterious effect will be much smaller for some other candidate ion systems.

II. THEORY

The relevant energy levels for $^{40}Ca^+$ are shown in Fig. 1. Doppler laser cooling (and qubit readout) requires laser radiation at 397 nm and at 866 nm. Fluorescence at 397 nm is detected while the 866 nm light serves to repump population that falls into the $^2D_{3/2}$ state back into the $^2S_{1/2} \leftrightarrow {}^2P_{1/2}$ cooling cycle. The electric-dipole matrix element $M = \langle D_{5/2}|\mathbf{d}\cdot\mathbf{E}|P_{1/2}\rangle$ is identically zero so that, in the absence of any perturbation, decay from the excited $^2P_{1/2}$ state to the metastable $^2D_{5/2}$ state can only occur via a highly forbidden $M2/E3$ transition. As a result, during Doppler cooling, the $^2D_{5/2}$ state should not become populated. Any effect which would allow such population transfer would lead, for a single trapped ion, to dark periods in the fluorescence which would last on the order of the lifetime of the $^2D_{5/2}$ state (∼1 s). We show in the following that magnetic-field induced J-mixing provides just such an effect, opening a channel for shelving to the metastable state. The quantum jumps then give a macroscopic signal—an unambiguous indication of the presence of the J-mixing effect.

A magnetic field perturbs the orbital and spin angular momenta to different extents. Because of this, the states defined by the total angular momentum J are no longer pure eigenstates. Applying an external magnetic field causes some of the states to gain a small admixture of otherwise orthogonal states of the same m_J. After this perturbation, some small amplitude of the $^2P_{1/2}$ to $^2D_{5/2}$ transition becomes electric-dipole allowed. The transition probability $|M'|^2$ is then nonzero

$$\begin{aligned} |M'|^2 &= |\langle D'_{5/2}|\mathbf{d}\cdot\mathbf{E}|P'_{1/2}\rangle|^2 \\ &= |(\langle D_{5/2}| + \epsilon_D\langle D_{3/2}|)\mathbf{d}\cdot\mathbf{E}(|P_{1/2}\rangle + \epsilon_P|P_{3/2}\rangle)|^2 \\ &= |\epsilon_D\langle D_{3/2}|\mathbf{d}\cdot\mathbf{E}|P_{1/2}\rangle + \epsilon_P\langle D_{5/2}|\mathbf{d}\cdot\mathbf{E}|P_{3/2}\rangle|^2, \quad (1) \end{aligned}$$

ignoring the second-order term.

*d.segal@imperial.ac.uk

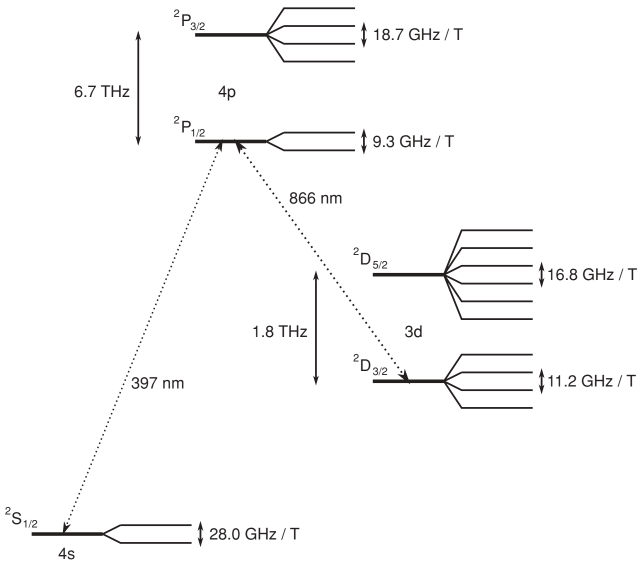

FIG. 1. Energy-level structure of $^{40}Ca^+$ in a magnetic field. The fine structure and Zeeman splittings have been exaggerated for clarity.

The coefficients ϵ_D and ϵ_P are, from first-order perturbation theory

$$\epsilon_D = \frac{\langle D_{3/2}|\boldsymbol{\mu}\cdot\mathbf{B}|D_{5/2}\rangle}{E_{D_{5/2}} - E_{D_{3/2}}} \quad \text{and} \quad \epsilon_P = \frac{\langle P_{3/2}|\boldsymbol{\mu}\cdot\mathbf{B}|P_{1/2}\rangle}{E_{P_{1/2}} - E_{P_{3/2}}}, \tag{2}$$

where E_ζ is the energy of state $|\zeta\rangle$ and the Hamiltonian $\boldsymbol{\mu}\cdot\mathbf{B} = \mu_B B(\hat{L}_z + 2\hat{S}_z)$ is the perturbation due to the magnetic field. These amplitudes can be computed by transforming the states into the $\{L,S,m_L,m_S\}$ basis, applying the $\boldsymbol{\mu}\cdot\mathbf{B}$ operator, and then transforming back into the $\{L,S,J,m_J\}$ basis. The coefficients are listed for each state in Table I.

We expect the J-mixing effect to manifest itself in our experiment as a change in the character of the fluorescence traces we see at different magnetic fields. We expect the ion to become dark ($D_{5/2}$ shelving) after emitting a (large) number n of 397 nm photons. Furthermore, we expect that the average number of blue photons scattered before shelving occurs is high at low magnetic fields but then decreases as B is increased. The mean value of n averaged over a large number of quantum jumps is the key number we extract from the experiment. This can be written as

$$n = \frac{\Gamma(P_{1/2},S_{1/2})}{\Gamma(P_{1/2},D_{5/2})} \to \infty \text{ at low } B, \tag{3}$$

TABLE I. Admixture coefficients for each state. ΔE is the fine-structure splitting.

Orbital	m_J	$\epsilon/\frac{\mu_B B}{\Delta E}$
P	$\pm\frac{1}{2}$	$\frac{\sqrt{2}}{3}$
	$\pm\frac{3}{2}$	0
D	$\pm\frac{1}{2}$	$\frac{\sqrt{6}}{5}$
	$\pm\frac{3}{2}$	$\frac{2}{5}$
	$\pm\frac{5}{2}$	0

where Γ is a transition rate. These rates can be written in terms of a reduced matrix element (which can in turn be expressed in terms of a normal matrix element and a Clebsch-Gordan coefficient through the Wigner-Eckart theorem [13])

$$\Gamma = \frac{4\omega^3}{3\hbar c^3}\frac{|\langle g,J\|\mathbf{d}\cdot\mathbf{E}\|e,J'\rangle|^2}{2J'+1}, \tag{4}$$

where $\mathbf{d}\cdot\mathbf{E}$ is the electric-dipole operator, g and e label the ground and excited states, and $\hbar\omega$ is the energy difference between the initial and final states. To arrive at a result for n, a number of reduced matrix elements for the relevant transitions in Ca^+ are required. Fortuitously these have been calculated using relativistic many-body theory by Guet and Johnson [14]. Using these calculated reduced matrix elements, we find the $^2P_{1/2}$ state in Ca^+ has a branching ratio between the $^2D_{5/2}$ and $^2S_{1/2}$ decay channels of $4.2\times10^{-7}B^2$ T^{-2}. Thus at 1 T, an average of $n = 2.4\times10^6$ photons at 397 nm will be emitted before the ion is shelved to the $^2D_{5/2}$ state.

Note that due to the fine-structure energy splittings, ϵ_P is negative while ϵ_D is positive, so there is a partial destructive interference between the two amplitudes contributing to the transition.

III. EXPERIMENT

Two grating-stabilized diode lasers tuned close to the $^2S_{1/2} \leftrightarrow {}^2P_{1/2}$ resonance transitions at 397 nm are used for Doppler cooling. Ions decay from $^2P_{1/2}$ to the metastable $^2D_{3/2}$ states with a branching ratio of 1 in 16 [14]. To avoid optical pumping into the $^2D_{3/2}$ levels, we use four diode lasers at 866 nm to repump ions back in to the cooling cycle. The 397 nm transitions chosen are both π ($\Delta m_J = 0$) polarized, while all 4 of the 866 nm transitions are σ ($\Delta m_J = \pm1$) polarized. To reveal the dependence of the J-mixing effect on magnetic field, the strength of this field is varied in our experiment. Each of the six laser wavelengths was manually retuned to optimize the fluorescence rate at each particular magnetic-field strength. Effects due to amplified spontaneous emission (ASE) in diode lasers have been seen in our system, and other similar systems [15]. The laser beams are therefore filtered to remove any unwanted light due to ASE at 393 and 850 nm, which could cause excitation to the $^2P_{3/2}$ state, and then decay to the $^2D_{5/2}$ state.

The trap depth of a Penning trap is dependent on the strength of the magnetic field, which is responsible for the radial confinement. We find empirically that to reliably trap Ca^+ in our Penning trap, we need $B \gtrsim 0.6$ T. Thus to probe a large range of B, the trap was operated as a combined rf-Penning trap. Such a trap can be operated with a magnetic field of arbitrary strength [16]. Ca^+ ions are generated inside the trap by electron-beam bombardment of a weak atomic beam.

IV. RESULTS

Fluorescence at 397 nm from a single Ca^+ ion is detected using a photon-counting photomultiplier tube (PMT). Typical signals at high and low B are shown in Fig. 2. At a given magnetic-field strength, the count rate was recorded for approximately 20 minutes. The mean number of photons detected before the ion turns dark (after subtracting a constant

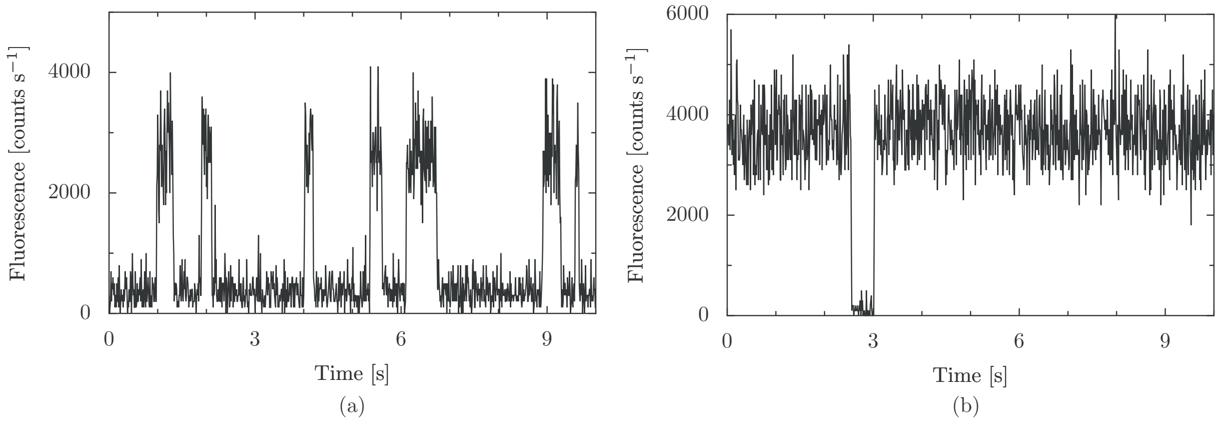

FIG. 2. Typical fluorescence at 397 nm from a single Ca^+ ion. (a): $B = 0.9$ T. (b): $B = 0.2$ T.

background level) n_{detected} is seen to decrease with increasing B. Experimental values for the inverse of this, predicted to scale as B^2, are shown in Fig. 3.[1] Also shown is the branching ratio given above ($4.2 \times 10^{-7} B^2\ \text{T}^{-2}$) divided by the detection efficiency of the system (estimated to be $0.04 \pm 0.01\%$). Although there is a large uncertainty in the detection efficiency, the B^2 trend can clearly be seen. This uncertainty in the detection efficiency is due largely to the relatively complicated imaging system required for our Penning trap, designed to ensure that the PMT is sufficiently far away from the vacuum chamber so that it is not affected by the magnetic field.

By analyzing the same data, a measurement of the lifetime of the $^2D_{5/2}$ state can also be extracted straightforwardly. A lifetime of 1.1 ± 0.1 s was found, which is consistent both with a recent calculation and with a variety of higher-precision measurements (see [17], and references therein).

V. DISCUSSION

Electron shelving, leading to the observation of quantum jumps in the fluorescence rate, is the key to high-fidelity readout in trapped-ion-based QIP. Thus far the only systems for which single ions and their associated quantum jumps have been seen in a Penning trap are Mg^+, Be^+, and Ca^+. Ions of Mg^+ and Be^+ differ from Ca^+ in that they do not have D levels lying below the lowest excited P levels. These systems are therefore amenable to Doppler laser cooling using a single laser frequency since no repumper is required. Electron shelving can be achieved using either Zeeman or hyperfine levels of the ground state. The lack of a metastable D state below the lowest excited P states means that the effect described previously will not be observed (unless higher-lying excited states are used). On the other hand, off-resonant radiation from the cooling laser drives ions both into a shelved level and brings ions back from that level into the cooling cycle [18]. This typically leads to shelving and repumping at a faster rate than would occur in Ca^+, using a metastable D state.

Considering the tiny degree of J-state mixing in Ca^+ provided by a relatively large laboratory field of 1 T, it is remarkable that the consequences are so stark. A branching ratio of only 4×10^{-7} leads to a $\sim$70% extinction of the fluorescence in our experiment. Given the severity of this effect on the time-averaged fluorescence rate the deleterious effect on readout fidelity is, in fact, surprisingly small. This is because typically the state detection period used in QIP experiments is kept as short as possible to speed up the rate of processing. A typical obstacle to efficient state readout in trapped ion qubits is spontaneous decay of the metastable state causing an ion initially in the dark state to turn bright during the measurement period. Due to the magnetic-field J-mixing effect in the Penning trap we have shown that it is also possible for an ion initially in the bright state to turn

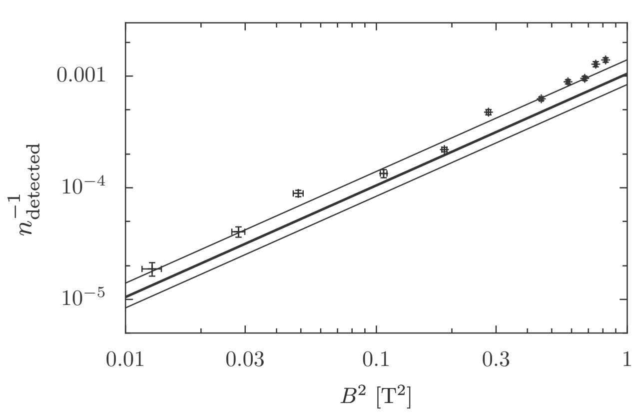

FIG. 3. Observed shelving rate per detected photon. The vertical error bars were estimated by bootstrapping the data at each value of B, while the horizontal error bars were estimated from the consistency of magnetic-field measurements at various electromagnet coil currents. The theoretical prediction (heavy line) using an estimated detection efficiency of $0.04 \pm 0.01\%$ is also shown (the lighter lines indicate the estimated uncertainty in the detection efficiency).

[1]The error on each value of n^{-1}_{detected} is estimated by "bootstrapping": For each B, a data set consists of a set of integers n_i, given by the number of photons detected between successive quantum jumps. The mean of n_i is n_{detected}. The bootstrap procedure involves creating a large number (10,000) of secondary data sets, each the same size as the original, by picking at random from the set of n_i. The mean values of the integers in each secondary set are scattered in a roughly Gaussian distribution around the mean value of the original set (n_{detected}). The standard deviation of this distribution is shown as the vertical error bars in the figure.

dark during a measurement period. For Ca^+ at 1 T, the decay rate into the $^2D_{5/2}$ state due to the J-mixing effect is similar to the the rate of spontaneous decay out of the $^2D_{5/2}$ state. So if the $^2D_{5/2} \to {}^2S_{1/2}$ decay rate is tolerable, then so too is the $^2P_{1/2} \to {}^2D_{5/2}$ decay. It has been shown that where readout fidelity is limited by decay of the metastable state, it can be improved by postanalysis of the fluorescence traces for each detection period [19]. A similar approach could also improve readout fidelity in the case of unwanted decays into the metastable state.

The rate of shelving caused by J-state mixing is inversely proportional to the energy splittings between the $^2P_{1/2}$ and $^2P_{3/2}$ states and between the $^2D_{3/2}$ and $^2D_{5/2}$ states. These splittings are due predominantly to the spin-orbit interaction. The analogous splittings in Ba^+ are ~10 times larger than they are for Ca^+, which leads to a J-mixing effect approximately 100 times smaller for that ion. The fine-structure splittings of various ions are shown in Table II. As can be seen by the relatively small splittings, the magnetically induced shelving effect is very much stronger in Ca^+ than any of the other ion species.

TABLE II. Comparison of ions with metastable D states. The final column shows approximately the magnetically induced transition rate at a fixed field relative to Ca^+.

Element	ΔE_D (meV)	ΔE_P (meV)	$\|\Delta E_D\|^{-2}$ (arb. units)
Ca^+	7.5	27.6	1
Sr^+	34.8	99.4	0.046
Ba^+	99.3	209.6	0.0057
Yb^+	170.1	412.9	0.0019
Hg^+	1864.8	1131.0	0.000016
Ra^+	205.6	602.3	0.0013

ACKNOWLEDGMENTS

The authors wish to acknowledge financial support from the UK EPSRC and from the EU through the integrated project SCALA. We also wish to thank S. Tokunaga and S. Ananthamurthy for useful discussions.

[1] P. Zeeman, Nature (London) **55**, 347 (1897).
[2] W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).
[3] A. V. Taichenachev, V. I. Yudin, C. W. Oates, C. W. Hoyt, Z. W. Barber, and L. Hollberg, Phys. Rev. Lett. **96**, 083001 (2006).
[4] Z. W. Barber, C. W. Hoyt, C. W. Oates, L. Hollberg, A. V. Taichenachev, and V. I. Yudin, Phys. Rev. Lett. **96**, 083002 (2006).
[5] P. Schef, P. Lundin, E. Biémont, A. Källberg, L. O. Norlin, P. Palmeri, P. Royen, A. Simonsson, and S. Mannervik, Phys. Rev. A **72**, 020501(R) (2005).
[6] D. Porras, J. I. Cirac, S. Kilina, S. Tretiak, and E. Einarsson, Phys. Rev. Lett. **96**, 250501 (2006).
[7] J. R. Castrejón-Pita, H. Ohadi, D. R. Crick, D. F. A. Winters, D. M. Segal, and R. C. Thompson, J. Mod. Opt. **54**, 1581 (2007).
[8] J. M. Taylor and T. Calarco, Phys. Rev. A **78**, 062331 (2008).
[9] H. Häffner, C. F. Roos, and R. Blatt, Physics Reports **469**, 155 (2008).
[10] D. R. Crick, H. Ohadi, I. Bhatti, R. C. Thompson, and D. M. Segal, Opt. Express **16**, 2351 (2008).
[11] D. R. Crick, S. Donnellan, S. Ananthamurthy, R. C. Thompson, and D. M. Segal, Rev. Sci. Instrum. **81**, 013111 (2010).
[12] T. B. Mitchell, J. J. Bollinger, D. H. E. Dubin, X. P. Huang, W. M. Itano, and R. H. Baughman, Science **282**, 1290 (1998).
[13] D. Budker, D. F. Kimball, and D. P. DeMille, *Atomic Physics* (Oxford University Press, Oxford, 2004).
[14] C. Guet and W. R. Johnson, Phys. Rev. A **44**, 1531 (1991).
[15] K. Toyoda, A. Miura, S. Urabe, K. Hayasaka, and M. Watanabe, Opt. Lett. **26**, 1897 (2001).
[16] D. J. Bate, K. Dholakia, R. C. Thompson, and D. C. Wilson, J. Mod. Opt. **39**, 305 (1992).
[17] B. K. Sahoo, M. R. Islam, B. P. Das, R. K. Chaudhuri, and D. Mukherjee, Phys. Rev. A **74**, 062504 (2006).
[18] H. F. Powell, M. A. van Eijkelenborg, W. Irvine, D. M. Segal, and R. C. Thompson, J. Phys. B: At. Mol. Opt. Phys. **35**, 205 (2002).
[19] A. H. Myerson, D. J. Szwer, S. C. Webster, D. T. C. Allcock, M. J. Curtis, G. Imreh, J. A. Sherman, D. N. Stacey, A. M. Steane, and D. M. Lucas, Phys. Rev. Lett. **100**, 200502 (2008).