High-Efficiency Detection of a Single Quantum of Angular Momentum by Suppression of Optical Pumping

M. J. McDonnell, J.-P. Stacey, S. C. Webster, J. P. Home, A. Ramos, D. M. Lucas, D. N. Stacey, and A. M. Steane

Centre for Quantum Computation, Department of Atomic and Laser Physics, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, England

(Received 18 November 2003; published 4 October 2004)

We propose and demonstrate experimentally the discrimination between two spin states of an atom purely on the basis of their angular momentum. The discrimination relies on angular momentum selection rules and does not require magnetic effects such as a magnetic dipole moment of the atom or an applied magnetic field. The central ingredient is to prevent by coherent population trapping an optical pumping process which would otherwise relax the spin state before a detectable signal could be obtained. We detected the presence or absence of a single quantum ($1\hbar$) of angular momentum in a trapped calcium ion in a single observation with success probability 0.86. As a practical technique, the method can be applied to read out some types of quantum computer.

DOI: 10.1103/PhysRevLett.93.153601 PACS numbers: 42.50.Gy, 03.67.Lx, 32.80.Bx

In this Letter, we present a method to detect, with high signal-to-noise ratio, the angular momentum state of a single atom. The method is nondestructive in the sense that the atom is not moved or lost but can then be re-prepared or used for some other purpose. The method relies on resonant excitation of a wanted transition using a weak probe laser beam, while suppressing excitation on a closely separated unwanted transition using an intense pump laser.

The essence of our method is to illuminate a single atom with a pair of laser beams in such a way that strong fluorescence is observed when the atom's angular momentum vector points along the laser beam propagation direction, but not when it has the opposite orientation. The discrimination arises from a combination of resonant behavior and angular momentum conservation; the idea which makes the experiment feasible is to use coherent population trapping (CPT) to suppress an optical pumping process which would otherwise relax the spin state before a detectable signal is obtained. The atom's component of angular momentum along the axis defined by the laser beams is $\pm\hbar/2$ for the two orientations involved. In our experiments when the atom is prepared in one of these two states, then in a single observation we correctly determine which one with probability 0.86. The method can achieve close to perfect discrimination when the atomic structure has the optimal form, which we describe below.

The pump laser beam determines the efficiency of the state discrimination by determining the width and depth of the CPT minimum on the unwanted transition. The method therefore measures the direction of the angular momentum of the atom with respect to the polarization state of the pump laser. This is in contrast to most measurements of "spatial quantisation" associated with the direction of an angular momentum vector, which show that the direction of a *magnet* (such as an atom or an electron) relative to the direction of the local magnetic field takes on discrete values. Atomic fine structure, and the original Stern-Gerlach experiment are examples of the latter type of experiment. The method presented here differs from these experiments in that the presence or absence of a magnetic dipole moment of the atom is irrelevant to the observed behavior. This means that spin states (e.g., $|M = \pm 1/2\rangle$) can be discriminated even when the energy separation (e.g., by a Zeeman effect) is small compared to all the line-broadening effects in an experiment, such as natural linewidths and laser linewidths. This feature both permits the use of low magnetic fields, and also greatly reduces the frequency stability constraints, which can represent a large saving in experimental complexity.

We first describe the principle of the spin measurement, and then present an experimental demonstration using a single atom (a trapped $^{40}Ca^{+}$ ion).

It is well known that spin states (i.e., those usually called Zeeman sublevels, though we will not need a Zeeman effect here) in atoms can be discriminated through polarization effects as well as by energy splittings. This fact is used, for example, in optical pumping with polarized light. However, whereas optical pumping allows a specific spin state to be prepared at zero magnetic field, it does not give a signal which allows a previously prepared spin state to be deduced. A typical situation is that by use of σ^{+}-polarized light on a $J = 1/2 \rightarrow J' = 1/2$ transition, for example, there is no excitation out of $|M = 1/2\rangle \equiv |+\rangle$ while there is excitation out of $|M = -1/2\rangle \equiv |-\rangle$. If the state was prepared in $|-\rangle$, the atom will scatter of order one photon before being optically pumped into $|+\rangle$; if it was prepared in $|+\rangle$ it will scatter none. If the single photon could be detected with high reliability, the spin measurement would be feasible, but this is not normally possible in practice because of the 4π steradians of solid angle into which

the photon is scattered, and the limitations of photon detectors.

One cannot take advantage of a cycling transition such as $|+\rangle \rightarrow |J' = 3/2, M' = 3/2\rangle$ to scatter more photons, because an atom prepared in $|-\rangle$ is quickly optically pumped to $|+\rangle$ by the incident radiation.

This problem is unavoidable in single-photon processes, whenever the excited state has a significant branching ratio for decay back to the $\{|-\rangle, |+\rangle\}$ manifold. If the main decay from the excited state is instead to a closed manifold which does not involve $|+\rangle$, then optical pumping could be used to move population from $|-\rangle$ to the other manifold while leaving $|+\rangle$ unaffected—this would be polarization-selective electron shelving. It can achieve the desired spin measurement, but the right combination of atomic levels and branching ratios is rare.

We use coherent population trapping (CPT) [1,2] to suppress the undesired optical pumping process. A suitable energy level structure is of the form $J' = J + 1$ and $J'' = J'$ or J, where the states $|\pm\rangle$ are in the ground or metastable level J, and it must be possible to drive the $J' \leftrightarrow J''$ transition with a Rabi frequency large compared with laser linewidths and the natural width of J''. $J' > J$ ensures a cycling transition or manifold is available which involves $|+\rangle$ but not $|-\rangle$; this need only be weakly driven. This energy level structure applies most straightforwardly, but not exclusively, to alkalis and alkali-like ions. For clarity, we will describe the method using a $S_{1/2}, P_{3/2}, D_{3/2}$ manifold which is the case for our experiments [see Fig. 1(a)]. The spin states $|\pm\rangle$ to be measured are the substates of the ground level $S_{1/2}$, and the notation $|L, M\rangle$ refers to the substate M of the level denoted by $L = S, P$ or D.

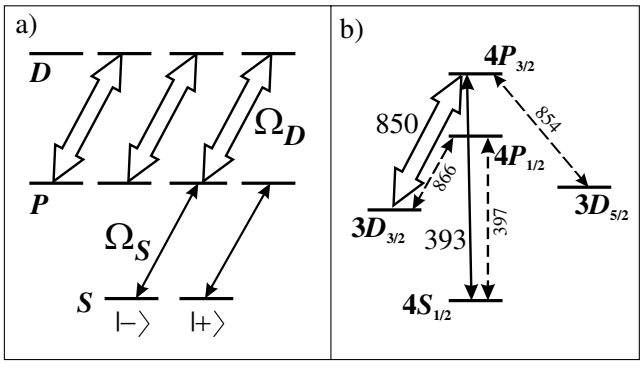

FIG. 1. (a) Basic concept of the spin measurement. A weak probe laser Ω_S causes fluorescence which is detected. A strong pump laser Ω_D selectively suppresses the excitation from $|-\rangle$ which would otherwise cause optical pumping from $|-\rangle$ to $|+\rangle$. (b) Low-lying energy levels of the Ca^+ ion used in the experiment, with laser wavelengths indicated in nm. The pump and probe transitions are $3D_{3/2} \leftrightarrow 4P_{3/2}$ (850 nm) and $4S_{1/2} \leftrightarrow 4P_{3/2}$ (393 nm), respectively.

An intense σ^+-polarized "pump" laser beam defines an axis in space and resonantly drives the $P \leftrightarrow D$ transition with Rabi frequency Ω_D. Owing to electric dipole selection rules, this laser does not couple to $|P, 3/2\rangle$ but it does couple to all the other sublevels of P. A weak σ^+-polarized "probe" laser resonantly drives the $S \leftrightarrow P$ transition with Rabi frequency $\Omega_S \ll \Omega_D$.

In this situation, if the atom is prepared in $|+\rangle$ it scatters many photons on the S–P transition at a rate R_+. Treating the cycling transition $|+\rangle \leftrightarrow |P, 3/2\rangle$ as a two-level system, we have

$$R_+ = \frac{C^2\Omega_S^2}{\Gamma_P^2 + 2C^2\Omega_S^2}\Gamma_P \tag{1}$$

in steady state, where $\Gamma_P = 1/\tau_P$ is the decay rate of P, and C is the ratio of the Clebsch-Gordan coefficients for the transitions out of $|+\rangle$ and $|-\rangle$ ($C = \sqrt{3}$ for the case $S_{1/2} \leftrightarrow P_{3/2}$.)

If the atom is prepared in $|-\rangle$, almost no excitation takes place because it is prevented by CPT: the atom remains in the dark state $(\Omega_D|-\rangle - \Omega_S|D, 3/2\rangle)(\Omega_D^2 + \Omega_S^2)^{-1/2}$ associated with the pump and probe lasers (the probe is turned on adiabatically to ensure this). In practice the nulling is limited by processes which destroy the coherence of the dark state. These are primarily laser phase fluctuations and spontaneous decay of D. An approximate model which captures the main features is to treat $\{|-\rangle, |P, 1/2\rangle, |D, 3/2\rangle\}$ as a closed three-level system. In steady state the scattering rate is

$$R_- = \frac{\Omega_S^2}{\Omega_D^2}\gamma \tag{2}$$

provided $\Omega_D \gg \Omega_S, \gamma$, where $\gamma = \Gamma_D + \gamma_{\text{pump}} + \gamma_{\text{probe}}$ is the sum of the natural linewidth of D and the linewidths of the pump and probe lasers. We thus achieve a ratio

$$\frac{R_+}{R_-} = \frac{C^2\Omega_D^2\Gamma_P}{(\Gamma_P^2 + 2C^2\Omega_S^2)\gamma} \simeq \frac{C^2\Omega_D^2}{\gamma\Gamma_P} \gg 1. \tag{3}$$

The scattering on $|+\rangle \leftrightarrow |P, 3/2\rangle$ (rate R_+) has to be detected before the undesired scattering on $|-\rangle \leftrightarrow |P, 1/2\rangle$ causes too much optical pumping from $|-\rangle$ to $|+\rangle$. The undesired optical pumping occurs at the rate αR_- where $\alpha = 2/3$ is the square of the Clebsch-Gordan coefficient for $|+\rangle \leftrightarrow |P, 1/2\rangle$. The probe laser is therefore pulsed on for a finite time, to optimize the difference between the numbers of fluorescence photons which are scattered when the initial state was $|+\rangle$ or $|-\rangle$, divided by the noise which is principally shot-noise in the detection. A simple rate-equation analysis suffices to find the optimum pulse duration. Fluorescence photons are counted and a threshold set to optimize the discrimination. Let $p_\pm$ be the probability that a prepared state $|\pm\rangle$ results in a signal above the threshold. Perfect behavior is

represented by $p_+ = 1$, $p_- = 0$. We define an "efficiency" $\epsilon = p_+ - p_-$.

For some atoms (such as many alkali-like ions) the energy level structure has a further property which can be exploited to amplify the signal. This is that the P state can decay to a metastable "shelf" level H. In this case any excitation to P has some probability of resulting in optical pumping to the shelf, and the lasers should then be operated for a time τ such that the probability of shelving from $|+\rangle$ is high. Rather than detect fluorescence while the pump and probe lasers are operating, one can check whether at the conclusion of the process the atom has reached the shelf or not. This can be done by driving the atom in a closed manifold of states including S but excluding H, and observing the presence or absence of fluorescence [3–5]. To find the optimum pulse time τ it is necessary to compare the rate αR_- with βR_+, where β is the branching ratio for decay from P to H compared with S. We find [6,7] $\tau = \ln(\beta R_+/\alpha R_-)/(\beta R_+ - \alpha R_-)$ and

$$\epsilon = \frac{1}{1-r}(r^{r/(1-r)} - r^{1/(1-r)}) \simeq 1 + r\ln r \qquad (4)$$

where $r = \alpha R_-/\beta R_+$ and the approximation applies when $r \ll 1$.

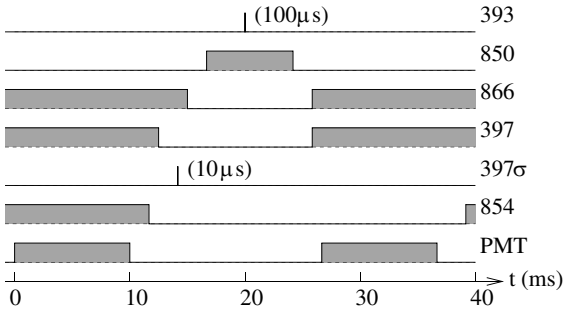

FIG. 2. Experimental sequence to prepare and measure the spin once. First the 397 and 866 nm beams cool the ion, and the PMT collects fluorescence (typical signal 100 counts), while the 854 nm beam serves to depopulate $D_{5/2}$, should the ion have been shelved in the previous sequence. Next a weak circularly polarized 397 nm beam is used to prepare the spin state by optical pumping. For σ^+ polarization the prepared state is a a mixture expected to be 99.9% $|+\rangle$, 0.1% $|-\rangle$, owing to the 3° between this beam and the quantisation axis set by the 850 nm beam. (We confirmed the preparation to 1% uncertainty in a separate experiment by observing the reduction in fluorescence as a waveplate was rotated.) All beams are then shut off, and the intense σ^+-polarized 850 nm pump beam is introduced. Then the σ^--polarized 393 probe beam, propagating at angle 6° to the 850 beam, is introduced for a precisely timed pulse of duration τ. The optimum pulse time was found experimentally to be $\tau = 150$ μs; this was consistent with our calculations discussed in the text. Finally the 850 nm beam is extinguished, and then the 397 nm and 866 nm beams are reintroduced and fluorescence detected during 10 ms. Absence of fluorescence indicates that the ion has been shelved.

With an optimum choice of experimental parameters, therefore, and the energy level systems described, efficiencies arbitrarily close to 1 can be achieved.

In our experimental demonstration, we used single ions of $^{40}Ca^+$. A diagram of the relevant energy levels and the laser wavelengths required is shown in Fig. 1(b). There is a shelving level (H is in fact $3D_{5/2}$); the experiment therefore consisted of preparing the ion in $|+\rangle$ or $|-\rangle$, illuminating it with lasers at 850 nm and 393 nm, and then checking to see whether it had been shelved by switching on the cooling lasers (397 nm and 866 nm). However, the fact that the $D = 3D_{3/2}$ level lies below $P = 4P_{3/2}$ reduces the efficiency: after excitation from $|+\rangle$ to P, the spontaneous decay to D can lead to optical pumping back to $|-\rangle$. This reduces the maximum possible value of p_+ in proportion to the branching ratio for decay from $|P, 3/2\rangle$ to D rather than H. This ratio is $\simeq 0.1$ and the maximum available ϵ was therefore $\simeq 0.9$ [8]. Another effect of the $D_{3/2}$ level being below the $P_{3/2}$ level is that the pump and probe lasers must have opposite circular polarizations.

The ions were loaded into a linear Paul trap in high vacuum. Many details of the apparatus are given in [7,9]. All the lasers were grating-stabilized diode systems, with all but that at 854 nm electronically locked to reference cavities. Switching was by acousto-optic modulator

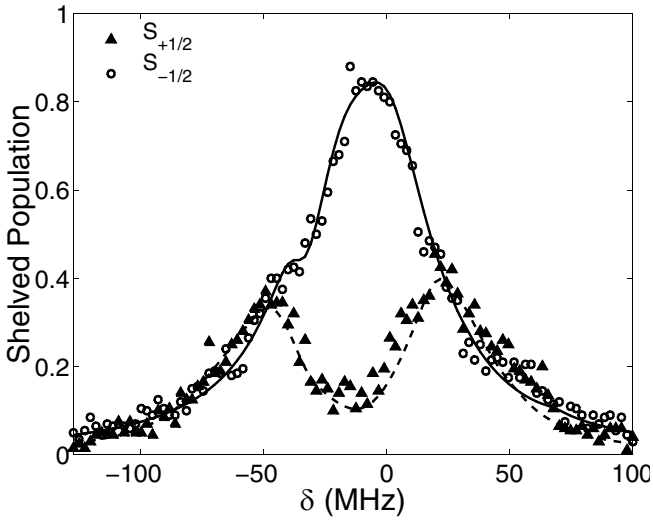

FIG. 3. Example data set. The proportion of times no fluorescence was observed in a set of 100 repetitions is shown versus detuning δ of the 393 nm laser. Circles: population prepared in $|+\rangle$; triangles: population prepared in $|-\rangle$. The lines show theoretical fits obtained from a full solution of the OBEs, with parameters $\Omega_S = 2\pi \times 0.82$MHz, $\Omega_D = 2\pi \times 85$MHz, $\gamma_{393} = \gamma_{850} = 2\pi \times 1.5$MHz, $B = 7$G. The fitted curve allows for 1% polarization impurity in the 850 nm beam. The angular momentum state is detected with $(1 + \epsilon)/2 = 86\%$ reliability in a single shot (for equal prior probabilities of $|-\rangle$, $|+\rangle$).

(AOM) for all beams except the intense 850 nm beam and the 854 nm beam, which were shuttered. To ensure good extinction ($\sim 10^{-9}$) of the light at 397 nm, two cascaded AOMs were used.

The experimental sequence to prepare and measure the spin state of the ion once is set out in Fig. 2. The probe beam at 393 nm had peak intensity 4 μW/mm^2 at the ion, hence $\Omega_S \simeq 2\pi \times 1$ MHz. The pump beam at 850 nm had peak intensity 800 mW/mm^2, hence $\Omega_D \simeq 2\pi \times 100$ MHz. A magnetic field, B, of a few gauss aligned along the 850 beam served to prevent population trapping during fluorescence detection [10]. Although this field also gave a small Zeeman splitting, that fact is irrelevant to the spin measurement being performed [11]. The sequence was repeated $n = 100$ times, and the number of times the ion was found to be shelved was recorded. This gives one data point in our data shown in Fig. 3. Next a parameter was changed and the whole sequence recommenced; this was continued to create a data set showing the shelved population as a function of the parameter. Then we reversed the polarization of the 397 nm preparation beam, and accumulated another data set in the same manner. The example data in Fig. 3 shows the effect of varying the detuning of the probe beam at 393 nm.

We studied the process by observing the effect of all the main parameters (laser intensities, detunings and polarizations) and compared the results with a numerical solution of the Optical Bloch Equations (OBEs). The calculations included all the ten levels of S, P, D plus a decay from P to H at the appropriate rate. Our data is fully consistent with the theory, assuming laser linewidths such that $\gamma = 2\pi \times 3$ MHz. The noise we observe on the locking error signals of the 393 nm and 850 nm lasers is consistent with this value. The fact that we observe good discrimination of the spin state with laser linewidths this large illustrates the relative insensitivity of the method to frequency instability. Note also that high discrimination is possible even with single-photon transition (S–P and D–P) linewidths as large as 22 MHz. We observe a maximum efficiency $\epsilon \simeq 0.72$. Using Eq. (4) with the prefactor 0.9, this implies $r = 0.0819$ and therefore $R_+/R_- = 154$ (since $\beta = 0.053$). This means the excitation out of $|-\rangle$ was suppressed by CPT by a factor $(1/3) \times 154 = 51$ [12].

Spin state measurements are of great interest for various quantum computing proposals, especially in solid state physics. The central ingredient of the CPT scheme we have proposed, that of modifying the system response by imposing a strong oscillating perturbation is expected to be widely applicable.

This work was supported by the EPSRC, the Royal Society, the Research Training and Development and Human Potential Programs of the European Union, the National Security Agency (NSA) and Advanced Research and Development Activity (ARDA) (P-43513-PH-QCO-02107-1).

[1] E. Arimondo, in *Progress in optics*, edited by E. Wolf (North-Holland, Amsterdam, 1996), Vol. XXXV, p. 257–354.

[2] M. Scully and M. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, England, 1997).

[3] W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).

[4] J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).

[5] T. Sauter, W. Neuhauser, R. Blatt, and P. E. Toschek, Phys. Rev. Lett. **57**, 1696 (1986).

[6] D. Stevens, J. Brochard, and A. M. Steane, Phys. Rev. A **58**, 2750 (1998).

[7] D. Lucas, in *Proceedings of the 1st International Conference on Experimental Implementation of Quantum Computation*, edited by R. Clark (Rinton Press, Princeton, New Jersey2001), p. 91–98.

[8] We note that to discover experimentally the relative weights $|a|^2$ and $|b|^2$ in some general state $a|+\rangle + b|-\rangle$, ϵ does not need to be very close to 1, since the signal-to noise ratio after n repeated repreparations and measurements is approximately $(n\epsilon)^{1/2}$. However, to detect correlations between the measured system and another may require $1 - \epsilon \ll 1$.

[9] P. Barton, C. Donald, D. Lucas, D. Stevens, A. Steane, and D. Stacey, Phys. Rev. A **62**, 032503 (2000).

[10] M. Boshier, G. Barwood, G. Huang, and H. Klein, Appl. Phys. B **71**, 51 (2000).

[11] If a smaller field were required, the population trapping could be prevented by modulating the 866 laser polarization. [10]

[12] With reduced laser linewidths, the OBEs predict a maximum efficiency 92% for this spin measurement in Ca^+.