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Ultrasensitive two-photon spectroscopy based on long spin-relaxation time in a dark optical trap

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Abstract. – We propose a new spectroscopic method for measuring weak transitions in cold and trapped atoms, which exploits the long spin relaxation times and tight spatial confinement offered by dark optical traps to achieve extremely high sensitivity. We demonstrate our scheme by measuring a $5S_{1/2} \to 5D_{5/2}$ two-photon transition in cold Rb atoms trapped in a new single-beam dark optical trap, using an extremely weak probe laser power of 25 μW. We were able to measure transitions with as small excitation rate as $< 0.1\,\mathrm{s}^{-1}$, using 10^7 fold quantum amplification of the transition rate due to spin shelving and normalized detection with a strong cycling transition.

The strong suppression of Doppler and time-of-flight broadenings due to the ultra low temperatures, and the possibility to obtain very long interaction times are obvious advantages of using free-falling cold atoms for precision spectroscopic measurements [1]. Even longer interaction times can be obtained for cold atoms trapped in optical dipole traps [2]. To obtain long atomic coherence times in these traps, spontaneous scattering of photons and energy level perturbations caused by the trapping laser are reduced by increasing the laser detuning from resonance [3]. To further reduce scattering, blue-detuned optical traps, where repulsive light forces confine atoms mostly in the dark (dark traps), have been developed [4]. The wide use of dark traps was limited by relatively complex setups that require multiple laser beams [5] or gravity assistance [6]. Recent developments of single-beam dark traps make them more attractive for precision spectroscopy [7,8].

Dark traps have an additional advantage that makes them especially useful for the spectroscopic measurements of extremely weak optical transitions. While preserving long atomic coherence times those traps can provide large spring constants and tight confinement of trapped atoms [7] to ensure good spatial overlap even with a tightly focused excitation laser beam. Therefore the atoms can be exposed to a much higher intensity of the excitation laser, yielding a further increase in sensitivity for very weak transitions.

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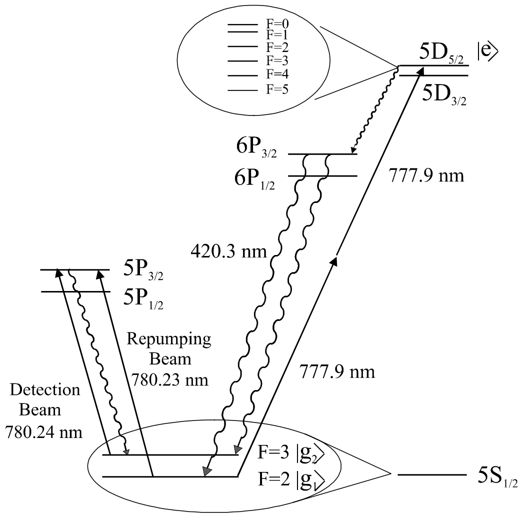

Fig. 1

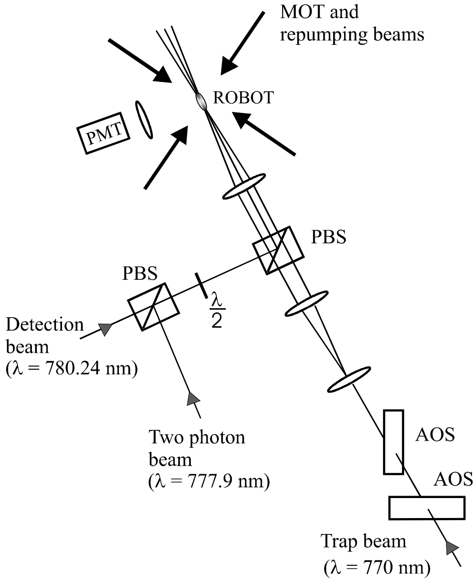

Fig. 2

Fig. 1 – Energy levels of ^{85}Rb and the transitions between them which are involved in the experiment. Spectroscopy of the $|g_1\rangle \rightarrow |e\rangle$ transition ($5S_{1/2}F = 2 \rightarrow 5D_{5/2}F'$ in the case of ^{85}Rb) is performed. Atoms which undergo the transition are shelved in the level $|g_2\rangle$ ($5S_{1/2}F = 3$ in ^{85}Rb), from which they are detected using a cycling transition (to $5P_{3/2}F = 4$).

Fig. 2 – Schematic diagram of the experimental setup. Two acousto-optic scanners (AOS) rotate a 10 nm blue-detuned laser beam that produces the ROBOT trap. The two-photon beam and the detection beam are co-aligned with the elongated axis of the trap.

In this letter we present a new and ultrasensitive method for measuring extremely weak transitions with cold atoms in a far-detuned single-beam dark optical trap, based on spin-shelving in an atomic Λ system. Atoms with two ground hyperfine levels ($|g_1\rangle, |g_2\rangle$) are stored in the trap in a level $|g_1\rangle$ that is coupled to the upper (excited) state, $|e\rangle$, by an extremely weak transition. An atom that undergoes the weak transition may be shelved by a spontaneous Raman transition in $|g_2\rangle$, that is uncoupled to the excited level by the weak transition. After waiting long enough, a significant fraction of the atoms will be shelved in $|g_2\rangle$. Similarly to the electron-shelving technique [9], the detection in our scheme benefits from multiply excited fluorescence of a strong cycling transition from the shelved level $|g_2\rangle$. Generally, our scheme utilizes the main advantage of the dark trap, namely the tight confinement combined with long spin-relaxation times of the trapped atoms to yield a very large (up to 10^7 fold) quantum amplification of the measured transition rate. Thanks to the use of a stable ground state as a "spin-shelve", the quantum amplification is limited only by spin relaxation processes which are strongly suppressed in our dark trap, whereas in electron-shelving spectroscopy the quantum amplification depends on the lifetime of a weakly coupled metastable excited state (the "electron shelve") [10]. Moreover, spin-shelving enables a normalized detection scheme that suppresses the atom-number noise and largely reduces the technical noise, as in radio-frequency (RF) atomic clocks [11].

We realized this scheme on a $5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition in cold and trapped

^{85}Rb atoms (see fig. 1 for the relevant energy levels) using an extremely weak ($25\,\mu$W) probe laser beam and we were able to measure transitions with an excitation rate as small as $0.09\,\mathrm{s}^{-1}$. The large quantum amplification due to spin shelving increases the sensitivity of our scheme far beyond the photon shot noise and technical noise encountered in the direct detection of two-photon induced fluorescence for this transition [12–15].

Our spectroscopic measurement was made on cold ^{85}Rb atoms trapped in a rotating-beam optical trap (ROBOT). The operation principles of the ROBOT are described elsewhere [16]. Briefly, a linearly polarized, tightly focused (16 μm $1/e^2$ radius) Gaussian laser beam is rapidly (100 kHz) rotated by two perpendicular acousto-optic scanners, as seen in fig. 2. This forms a dark volume which is completely surrounded by time-averaged dipole potential walls. The wavelength of the trapping laser was 770 nm (10 nm above the D_2 line) and its power was 380 mW. The initial radius of the rotation was optimized for efficient loading of the ROBOT from a magneto-optical trap (MOT). 700 ms of loading, 47 ms of compression and 3 ms of polarization gradient cooling produced a cloud of $\sim 3 \times 10^8$ atoms, with a temperature of 9 μK and a peak density of $1.5 \times 10^{11}\,\mathrm{cm}^{-3}$. On the last stage of the loading procedure, the atoms were optically pumped into the $F = 2$ ground state by shutting off the repumping laser 1 ms before shutting off the MOT beams.

After all laser beams were shut off (except for the ROBOT beam which was overlapping the center of the MOT), $\sim 3 \cdot 10^5$ atoms were typically loaded into the trap, with temperature and density comparable with those of the MOT. Next, we adiabatically compressed the trap by reducing the radius of rotation of the trapping beam from 70 μm to 29 μm so that the atoms will match the waist of the two-photon laser, to further increase the efficiency of the transition. The size of the final cloud in the radial direction was measured by absorption imaging and the temperature of the atoms was measured by time-of-flight fluorescence imaging. From these measurements and using our precise characterization of the trapping potential [16], the parameters of the final cloud are: radial size ($1/e^2$ radius) of 19 μm, axial size of 750 μm, rms temperatures of 55 μK [9 μK] in the radial [axial] direction, and a density of $7 \cdot 10^{11}\,\mathrm{atoms/cm}^{-3}$. The $1/e$ lifetime of atoms in the trap was measured to be 350 ms for both hyperfine ground states and was limited by collisions with background atoms. We measured the spin relaxation time of the trapped atoms to be > 1 s, by measuring spontaneous Raman scattering between the two ground-state levels [7, 17].

The spectroscopy was performed with an external-cavity diode laser which was tuned to the $5S_{1/2}F = 3 \rightarrow 5D_{5/2}F'$ two-photon transition (777.9 nm) and was split into two parts. The first part (10 mW) was used to frequency stabilize the laser using the 420 nm fluorescence signal from the two-photon excitation obtained from a 130 °C Rb vapor cell. The laser was focused into the cell to ~ 100 μm $1/e^2$ radius and reflected back to obtain Doppler-free spectra. We locked the laser to the atomic line either by Zeeman modulation technique [18] or directly to the side of the line. From the locking signal we estimated the peak-to-peak frequency noise of the laser to be ~ 3 MHz. The second part of the diode laser beam passed through an acousto-optic modulator ($\sim$1.5 GHz frequency shift) that shifted the laser frequency toward two-photon resonance with the $5S_{1/2}F = 2 \rightarrow 5D_{5/2}F'$ transition. The laser beam was then focused to a $26\,\mu$m ($1/e^2$ radius) spot size in the center of the vacuum chamber, in order to optimize the efficiency of the two-photon transition and was carefully aligned with the long (axial) axis of the ROBOT (see fig. 2).

We used a normalized detection scheme to measure the fraction of atoms transferred to $F = 3$ by the two-photon laser. To detect the total number of atoms in the trap we applied a strong 200 μs laser pulse, resonant with the $5S_{1/2}F = 3 \rightarrow 5P_{3/2}F = 4$ closed transition together with the repumping laser ($5S_{1/2}F = 2 \rightarrow 5P_{3/2}F = 2$) and imaged the fluorescent

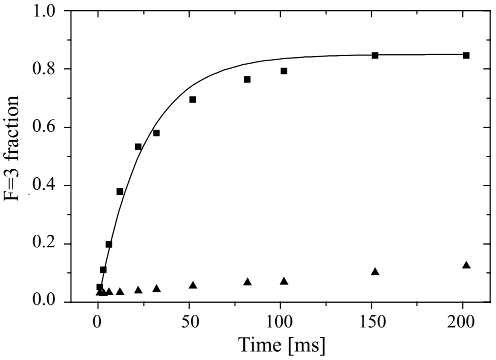

Fig. 3 – $F = 3$ normalized population fraction as a function of the interrogation time of the 170 μW two-photon laser tuned to resonance with the $5S_{1/2}F = 2 \rightarrow 5D_{5/2}F = 4$ line (■). The solid curve is a fit of the measurements by the function $N_{F=3}/N_{\text{total}} = A(1 - e^{-t/\tau_{4\text{p}}})$, resulting $A = 0.85$ as the steady-state population, and $\tau_{4\text{p}} = 25$ ms as the four-photon spontaneous Raman scattering time (see text). Spontaneous Raman scattering rate caused by the trapping laser is also given (▲).

signal on a photomultiplier tube (PMT). To measure only the $F = 3$ population we applied the detection pulse without the repumping laser. The $F = 3$ atoms were simultaneously accelerated and Doppler-shifted from resonance by the radiation pressure of the detection beam within the first 100 μs of the pulse. Then we could detect the $F = 2$ atoms by switching on the repumping laser that pumped the $F = 2$ population to the $F = 3$ state where atoms were measured by the second half of the detection pulse. This normalized detection scheme is insensitive to shot-to-shot fluctuations in atom number as well as fluctuations of the detection laser frequency and intensity.

After the adiabatic compression of the atoms in the ROBOT was completed, the two-photon laser on resonance with $5S_{1/2}F = 2 \rightarrow 5D_{5/2}F = 4$ was applied for various time intervals and the resulting $F = 3$ normalized population fraction was detected. The results for a 170 μW two-photon laser are presented in fig. 3. After 100 ms, $\sim 85\%$ of the atoms are pumped to the $F = 3$ state. This steady-state population is less than 100% since spontaneous Raman scattering from the trapping laser and from the two-photon laser (absorption of *one* photon followed by spontaneous emission) tends to equalize the populations of the two ground levels and therefore competes with the measured two-photon process. The characteristic $1/e$ time of the four-photon spontaneous Raman scattering process which is induced by the two-photon laser ($5S_{1/2}F = 2 \rightarrow 5D_{5/2}F' \rightarrow 6P_{3/2}F' \rightarrow 5S_{1/2}F = 3$, see fig. 1) is obtained from a fit to the data as $\tau_{4\text{p}} = 25$ ms. The corresponding (four-photon) rate is $\gamma_{4\text{p}} = 1/\tau_{4\text{p}} = 40\ \text{s}^{-1}$. Using the theoretical value of the two-photon cross-section of $\sigma = 0.57 \times 10^{-18}\ \text{cm}^4/\text{W}$ [19], the exact branching ratio (68%) for the two-photon excitation to decay to $F = 3$ [20], and our maximal excitation laser intensity of 16 W/cm^2, we calculate $\gamma_{4\text{p}} = 391\ \text{s}^{-1}$, a factor of ~ 10 larger than the measured rate. Using a measured value for the two-photon cross-section [15] yields a somewhat larger value of $\gamma_{4\text{p}} = 823\ \text{s}^{-1}$.

The main factor that reduced the measured excitation rate was the linewidth of the two-photon laser that was ~ 6 times larger than the 300 kHz natural linewidth of the two-photon transition [13]. The inhomogeneous broadening due to Stark shift was calculated for the compressed trapping potential to be ~ 400 kHz, which is smaller than the laser linewidth, hence it does not contribute to the reduced excitation rate. An additional reduction of the excitation rate may be caused by imperfect matching between the trapped atomic sample and

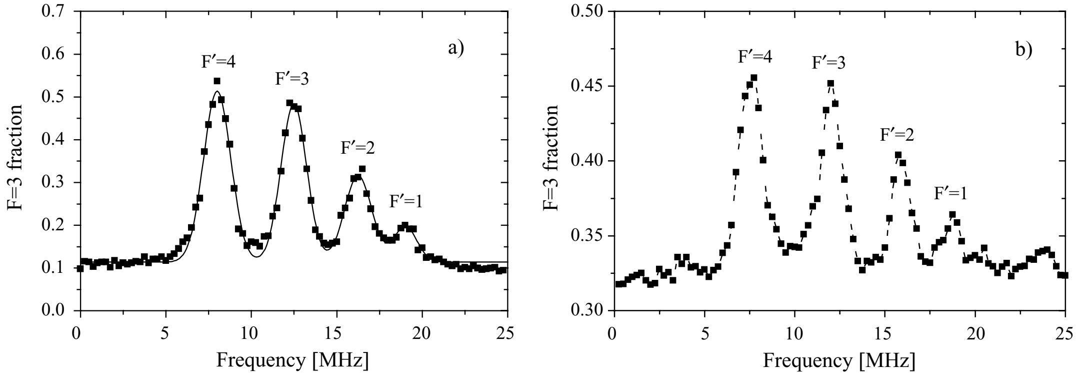

Fig. 4 – a) Frequency scan of the $5S_{1/2}F = 2 \to 5D_{5/2}F' = 4, 3, 2, 1$ line of the two-photon transition, after 50 ms exposure to a 170 μW two-photon laser. The solid curve is a fit to the data by a multi-peak Gaussian function (see text). b) The same frequency scan as in (a), after 500 ms exposure to a 25 μW two-photon laser. (The dashed curve connects the points and is given to guide the eye.)

the maximal intensity of the two-photon laser, so the overall agreement between the measured and the expected $\gamma_{4\mathrm{p}}$ is reasonable.

To measure the excitation spectrum of the $5S_{1/2}F = 2 \to 5D_{5/2}F'$ transition we scanned the frequency of the two-photon laser using the acousto-optic modulator. For each frequency point the whole experimental cycle was repeated, with 50 ms interrogation time of the two-photon laser. The $F = 3$ fraction of atoms as a function of the frequency of the two-photon laser is presented in fig. 4a. A 1.75 MHz linewidth (FWHM) of the atomic lines was determined by fitting the data with a multi-peak Gaussian function and is limited by the linewidth of the two-photon laser. This measurement agrees well with the frequency noise of the laser estimated from the locking signal. The distances between the lines obtained from this fit are 4.48 MHz, 3.76 MHz and 2.76 MHz, and are in excellent agreement with previously reported values of 4.50 MHz, 3.79 MHz and 2.74 MHz [13]. The height ratios between the lines obtained from the fit are $1 : 0.86 : 0.47 : 0.21$ for $F' = 4, 3, 2, 1$, respectively. The expected values were calculated using the strength of the two-photon transitions [13] together with the two-photon decay via the $6P_{3/2}$ level [20] to be $1 : 0.85 : 0.4 : 0.1$, in good agreement with the measured values, except for the weakest line. Note that although the two-photon transition $F = 2 \to F' = 0$ is allowed, a two-photon decay with $\Delta F = 3$ is forbidden and therefore this line is not detected.

Finally, we reduced the power of the two-photon laser to 25 μW, which reduced the transition rate by a factor of 46. Here, the interrogation time of the two-photon laser was 500 ms and the measured $F = 3$ population is shown in fig. 4b. A spectrum similar to that taken with the higher intensity is observed. A transition rate as small as 0.09 s^{-1} (for the $F = 3 \to F' = 1$ transition) is detected in this scan. The "quantum rate amplification" due to spin shelving (the ratio between the measured $\gamma_{4\mathrm{p}}$ transition rate and the rate of the one-photon cycling transition used for detecting the $F = 3$ population) is $\sim 10^7$ for this case.

In conclusion, we demonstrate a new and extremely sensitive scheme to measure weak transitions using cold trapped atoms. The key issues in our scheme are the long spin relaxation times combined with tight confinement of the atoms in a dark optical dipole trap, and the use of a shelving technique and normalized detection to enhance the signal-to-noise ratio. We demonstrated our scheme by measuring a two-photon transition $5S_{1/2} \to 5D_{5/2}$ for ^{85}Rb atoms trapped in a far-detuned rotating beam dark trap using only 25 μW laser power.

Our measurements may be improved in several ways. Improvements of the lifetime and spin relaxation time of atoms in the trap will allow much longer observation times and enable detection of much weaker transitions. This can be done by increasing the trapping laser detuning, where even longer spin-relaxation times are expected due to quantum interference between the two D lines [17]. Reduction of the linewidth of the two-photon laser will allow further improvements in the sensitivity of our scheme. It can also be combined with mode-locked laser spectroscopy [14] to obtain even larger sensitivities for a given time-averages power of the laser. Finally, our technique can be applied for other weak (forbidden) transitions such as optical clock transitions [21, 22] and parity-violating transitions where a much lower mixing with an allowed transition could be used.

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