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Full length article How dark is a grey state?

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Abstract

We present experimental results showing that a dark state with finite lifetime, a grey state, becomes darker the more the atom is exposed to on-resonant light. We discuss this phenomenon theoretically in the context of the complex eigenenergies and show that it can be understood qualitatively in terms of the quantum Zeno effect. The predicted dependence of the lifetime on the composition of the grey state and on the light intensity is experimentally confirmed with caesium atoms. This has implications for velocity-selective coherent population trapping and adiabatic transfer. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

The concept of dark states in atom optics [1] is well developed [2] and has led to subrecoil cooling schemes such as velocity selective coherent population trapping (VSCPT) [3] and adiabatic transfer [4]. Dark states are superpositions of internal atomic ground states with relative amplitudes and phases such that the excitation amplitudes of different ground states to the same excited state destructively interfere. Thus when an atom in such a state is exposed to near resonant light, it remains unexcited and cannot fluoresce. A perfect dark state can only exist if the constituent substates are degenerate in energy and thus do not change their relative phase with time. For 'dark' states set up by laser beams in more than one direction and comprising three or more degenerate atomic ground states, this cannot be fulfilled since the exchange of photon momenta lifts the degeneracy by an amount on the scale of the photon recoil energy. These states we call grey states expressing the feature that the dark state has a finite lifetime [5]. In this paper we will show that the lifetime of grey states increases with the light intensity. Thus the higher the intensity of on-resonant light, the less the grey state will couple to the excited state.

We will discuss this behaviour theoretically by analysing the eigenenergies of the Hamiltonian describing our system. The Hamiltonian is not unitary, since it involves the decay of the excited state to a non-detected state.

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Thus the eigenenergies will be complex, where the real part describes the phase evolution and the imaginary part describes the loss and thus the lifetime. Using these eigenenergies to determine the time evolution of the grey state, we find the velocity-selectivity as a function of light intensity and the form of the grey state.

The experiments were performed on caesium with orthogonally propagating circularly (σ^+) and linearly (π) polarised light beams on-resonance with an $F = 4 \rightarrow F' = 4$ transition. In this configuration a grey state exists and has been utilised for adiabatic transfer [6] and interferometry [7]. The free evolution lifetime of the grey state is measured and compared with the observed lifetime when the atoms are continuously exposed to light. A direct comparison between theory and experiment is given and shows good agreement.

2. Physical system

The level scheme for our grey state in caesium is shown in Fig. 1. It is a superposition of these magnetic sublevels with certain relative amplitudes and phases, such that the excitation amplitudes destructively interfere. This leads to the reduced excitation probability, characteristic of a grey state. Our main interest lies in the lifetime of these superposition states.

Qualitatively one can understand that these states can never be perfectly dark, since the exchange of photons between the laser beams involves a change of kinetic energy leading to a recoil shift of the constituent states (see Fig. 1). Thus, even if the state is dark at a particular instant, after a certain time the relative phases of the magnetic substates will have changed and the excitation amplitudes will no longer destructively interfere. In our experiments, there are two special cases where the grey state consists of only one magnetic sublevel and is therefore perfectly dark. These are for pure π or pure σ^+ light where the $m_F = 0$ and $m_F = 4$ states, respectively, are dark due to the dipole selection rules of an $F \rightarrow F' = F$ transition. For a given intensity ratio of π and σ^+ light a grey state can be found, which we will label with the fraction of the total light intensity which is in the σ^+ beam.

One picture of the grey state is that it is a state which is initially dark and is slowly evolving due to the non-degeneracy of its component magnetic ground states. The characteristic time for this evolution is given approximately by the reciprocal of the energy splittings between the ground magnetic substates. The question arises as to whether there is a mechanism to inhibit the evolution of the grey state so that it remains darker. One way is repeated projection of the grey state onto the dark state to stop its evolution. This is known as the quantum Zeno effect [8]. The projection is done on the time scale of the Rabi frequency between the magnetic ground and excited states. As the light intensity increases this time will become shorter. This leads to a higher repetition rate of the dark state projection, so that one expects that the lifetime of a grey state becomes longer the *higher* the on-resonant light intensity.

In the following we will show that the lifetime of a grey state increases with increasing on-resonance laser intensity. First we will derive and discuss the eigenenergies of the system, and after the description of the experimental details we will present our experimental results.



Fig. 1. The levels which form the basis states for a grey state in the presence of π and σ^+ light. The photon recoil shift is indicated but not to scale and is given for an $m_F = 0$ momentum of $-\sqrt{8}\hbar k$. For our experiments we used the D1 $(6^2S_{1/2} \rightarrow 6^2P_{1/2})$ $F = 4 \rightarrow F' = 4$ transition.

3. Theoretical description

For the remainder of this paper we will describe the situation of caesium in an orthogonal $\sigma^+ - \pi$ beam configuration of on-resonant D1 ($6^2S_{1/2} \rightarrow 6^2P_{1/2}$) $F = 4 \rightarrow F' = 4$ light. The Hamiltonian for this transition in the dressed state picture is [9]

$$H = \begin{pmatrix} E_{g}^{0} & \sqrt{10} q \hbar \Omega_{\sigma^{+}} & \\ \sqrt{10} q \hbar \Omega_{\sigma^{+}} & E_{e}^{1} & -q \hbar \Omega_{\pi} & \\ & -q \hbar \Omega_{\pi} & E_{g}^{1} & \sqrt{9} q \hbar \Omega_{\sigma^{+}} & \\ & & \sqrt{9} q \hbar \Omega_{\sigma^{+}} & E_{e}^{2} & -2 q \hbar \Omega_{\pi} & \\ & & & \sqrt{9} q \hbar \Omega_{\sigma^{+}} & E_{e}^{2} & \sqrt{7} q \hbar \Omega_{\sigma^{+}} & \\ & & & & \sqrt{7} q \hbar \Omega_{\sigma^{+}} & E_{e}^{3} & -3 q \hbar \Omega_{\pi} & \\ & & & & & -3 q \hbar \Omega_{\pi} & E_{g}^{3} & 2 q \hbar \Omega_{\sigma^{+}} & \\ & & & & & 2 q \hbar \Omega_{\sigma^{+}} & E_{e}^{4} & -4 q \hbar \Omega_{\pi} & \\ & & & & & -4 q \hbar \Omega_{\pi} & E_{g}^{4} & \end{pmatrix},$$
(1)

with $q = 1/(2\sqrt{48})$. The off-diagonal elements describe the coupling between the ground and excited magnetic sublevels with the appropriate Clebsch–Gordan coefficients. The intensity ratio of the two beams is the ratio of $\Omega_{\sigma^+}^2$ and Ω_{π}^2 . The diagonal elements describe the energy of the ground (g) and the excited states (e). The superscript indicates the magnetic sublevel. These energies are given by

$$E_{\rm g}^n = \frac{\left[p + \frac{2n\hbar k}{\sqrt{2}}\right]^2}{2m} - i\hbar\gamma_{\rm g}/2 \tag{2}$$

$$E_{\rm e}^{n} = \frac{\left[p + \frac{(2n+1)\hbar k}{\sqrt{2}}\right]^{2} + \frac{(\hbar k)^{2}}{2}}{2m} - i\hbar\gamma_{\rm e}/2$$
(3)

where the real terms correspond to the kinetic energy. The momentum for the $m_F = 0$ state is described by p and the momenta for the other magnetic sublevels can be found by the vector addition of the photon momenta exchanged. The spontaneous decay of the excited level is taken into account by γ_e , which is the inverse of the lifetime of the excited state. The coupling of the ground state to other nearby states is described by the loss rate γ_g which takes into account the off resonant excitation with subsequent spontaneous decay out of the system. In addition to this loss mechanism the coupling leads to different light shifts of the atomic magnetic ground states for our experimental parameters are smaller than the kinetic energy shifts due to initial velocity and acquired photon recoils.

4. Theoretical results

In the following we discuss our results from the numerical solution of the eigenvalue problem. The evolution of the eigenstates is given by the general phase factor $e^{iEt/\hbar}$, where *E* is the eigenenergy and *t* is the time. Thus the real part of the eigenenergies leads to a phase acquired during the interaction, while the imaginary part describes the population loss of the state and is the inverse of the lifetime.

In order to give an overview of the behaviour of the eigenenergies, we calculated them for different total intensities as a function of the fraction of the light intensity in the σ^+ beam. The initial momentum was chosen to be $-\sqrt{8}\hbar k$ for which the diagonal of the Hamiltonian becomes symmetric. Our results are shown in Fig. 2. Each column represents the eigenenergies for a given total light intensity indicated on the top of that column. In the case of caesium nine eigenstates exist. Their real energies are plotted in the upper row of the graphs, the lower graphs represent the lifetime of the longest living state, that is the grey state.

Below a total intensity of 3.3 I_{sat} the real eigenenergies show anti-crossings. For this situation a grey state still exists as can be seen in Fig. 2 in the bottom lefthand graph. For higher intensities the gaps between the eigenenergies become bigger, so that crossings do not occur. The lifetime of the grey state is found to increase with total intensity. Another feature is that the lifetime depends on the composition of the grey state. For total π and total σ^+ light an infinite lifetime is expected, since the corresponding grey state consists of only one internal state. There is also an increased lifetime near 0.5 σ^+ light intensity because the grey state mainly consists of $m_F = 1$, $m_F = 2$ and $m_F = 3$ where the $m_F = 2$ state has the minimum kinetic energy. This implies that the dephasing time is long for this superposition since these states have the smallest kinetic energy differences.



Fig. 2. Eigenenergies for different fractions of intensity in the σ^+ beam. Each column represents the eigenenergies for a given total light intensity I in units of the saturation intensity I_{sat} . The upper row shows the real part of the nine eigenenergies where the grey state energy is represented by the dashed line. The lower row gives the lifetime of the grey state. Note that the lifetime of the grey state increases with total light intensity.

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Fig. 3. Graph (a) shows the lifetime as a function of total intensity for different grey states labelled by the fraction of intensity in the σ^+ beam. Graph (b) shows the full width half maximum of the momentum distribution after 10 ms evolution in the light field assuming a $20\hbar k$ -wide initial momentum distribution.

The lifetimes of three different grey states as a function of the total intensity are given in graph (a) of Fig. 3. For all grey states the lifetime increases with the total light intensity and the lifetime is maximal for the 0.5 σ^+ light grey state. Another important aspect is the dependence of the lifetime on the initial momentum for different light intensities. The lifetime of the grey state is maximal for the initial momentum of $-\sqrt{8}\hbar k$ and is less for all other momenta due to a faster dephasing.

The graph (b) in Fig. 3 shows the full width half maximum of an initial $20\hbar k$ uniform momentum distribution after an evolution of 10 ms in the light field. One can see that the velocity selectivity becomes smaller the higher the intensity. This is important for both VSCPT and to understand the experimental results which will be presented.

5. Experiment description

The experiment was performed with caesium atoms cooled in a magneto optical trap and an optical molasses. The temperature achieved was 3 μ K. After cooling, the atoms were pumped with a short pulse of D2 $(6^2S_{1/2} \rightarrow 6^2P_{3/2})$ $F = 4 \rightarrow F' = 3$ light into the F = 3 ground state. A microwave π pulse (9.1 GHz) tuned to the $m_F = 0 \rightarrow m_F = 0$ of the $F = 3 \rightarrow F = 4$ ground state transition was used to prepare the atoms in the $m_F = 0$, F = 4 state. The effect of a magnetic field shifting the constituent magnetic substates of the caesium atom would lead to a shorter lifetime. In order to shield stray magnetic fields the interaction region was inside a three layer mu-metal box. A small homogeneous bias field of ≈ 5 mG was applied to allow for the selective $m_F = 0$ preparation with the microwaves.

The laser beams defining the grey states were realised with light from a Ti-Sapphire laser on resonance with the $F = 4 \rightarrow F' = 4$ D1 ($6^2S_{1/2} \rightarrow 6^2P_{1/2}$) transition. Great care was taken that the polarisation reaching the atomic ensemble was as pure as possible. The relative intensities of the linearly and circularly polarised beams were adjusted with an electro-optical modulator (EOM) changing the polarisation in front of a polarising cube (see Fig. 4). The total light intensity was controlled by an acousto-optical modulator (AOM) by changing the rf power driving the AOM.

A measurement of the time evolution of the grey state without light was accomplished using two 10 μ s pulses where the time between the pulses, T_f , was varied (see Fig. 4). The first pulse projected the initial $m_F = 0$ state to the grey state corresponding to the σ^+ light beam intensity. The second pulse acted as an

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Fig. 4. The experimental schematic: The light pulses were realised with an AOM and an EOM. The time lines for the free evolution and 'in light' evolution are shown.

analyser, leaving atoms which were in the grey state unaffected and pumping the others into a non-detectable state. Due to the short pulse length even some atoms which were not in the grey state survived both pulses, which led to the detected constant background.

In order to prepare the atoms efficiently in a definite grey state for the 'in light' lifetime measurements, adiabatic transfer was used [6]. The intensity time lines for the preparation are shown in Fig. 4. The light was adiabatically switched from pure linearly polarised light to the desired intensity ratio. This was accomplished within 45 μ s. A numerical calculation for these experimental settings showed that for the initial 3 μ K caesium ensemble the final momentum distribution had a full width half maximum of $7\hbar k$. This value was used for the direct comparison between theory and experiment.

For each parameter setting, the cooling, state preparation and light exposure were performed and the population of the F = 4 state was measured via the absorption of a probe beam 5 cm below the trap.

6. Experimental results

The experimental results concerning the free evolution are shown in Fig. 5(a). In the case of only π light, the corresponding grey state consists only of the $m_F = 0$ state and is perfectly dark. Thus the effect of a probe pulse does not depend on the time when it is applied. The observed small decay is due to off-resonant excitation to the F' = 3 state. For the 0.5 σ^+ grey state the free lifetime is as short as $8 \pm 2 \mu s$. This time scale can be understood in terms of the different phases accumulated by the different m_F states due to the photon recoil. The maximum difference in kinetic energy is between the ideal initial momentum $-\sqrt{8}\hbar k$ for $m_F = 0$ and $-\sqrt{2}\hbar k$ for $m_F = 1$ and corresponds to a $\pi/2$ dephasing time of 7 μs .

In the case where the light is left on one observes the behaviour shown in Fig. 5(b). Clearly the lifetime is about 100 times longer. The dependence on the grey state composition is shown with the different symbols. The

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Fig. 5. Experimental comparison of (a) free and (b) 'in light' evolution for different grey states. The number of atoms remaining in the F = 4 states normalised to the number after the preparation pulse is shown. In graph (a), the 0 σ^+ grey state is perfectly dark since it is the $m_F = 0$ state. For the 0.5 σ^+ state the dephasing of the magnetic substates due to the recoil energy shift leads to a decay time of $8 \pm 2 \mu s$. The lines are fitted exponential functions allowing for a background due to the survival of atoms for short pulses as discussed in the text. Graph (b) shows the result of the 'in light' lifetime for different grey states. Note the different time scale corresponding to lifetimes of $\approx 600 \mu s$. The solid lines are theory curves for the experimental $I = 3.2 I_{sat}$ including off-resonant excitation and initial momentum distribution.

fastest population loss is observed when 0.13 of the intensity is in the circularly polarised beam. The slowest decay is observed when this fraction is 0.87. From the simple lifetime calculations one would expect that the 0.53 fraction would live longest (see Fig. 3). However, taking into account the initial momentum distribution of $7\hbar k$ FWHM and the off-resonant excitation to the $6^2 P_{1/2} F' = 3$ state, one finds the theoretical solid curves shown in the graph which describe the observed decay very well. The total intensity was 3.2 times the saturation intensity.



Fig. 6. The observed time for decay to half the initial atom number for the 0.53 σ^+ grey state as a function of total light intensity. The solid line is a theoretical prediction with the same parameters as in Fig. 5. Clearly atoms survive longer with increasing light intensity.

The dependence of the lifetime on intensity is shown in Fig. 6. Clearly the survival time of the grey state increases with increasing light intensity, confirming the counter-intuitive behaviour. A grey state becomes darker the higher the light intensity. The solid line is the theoretically expected dependence with the same parameters as for the results shown in Fig. 5.

7. Conclusion

We have investigated the lifetime of grey states as a function of light intensity experimentally and theoretically. The final momentum distribution for a given light intensity was discussed theoretically and was necessary for the absolute comparison between theory and experiment. We find that a grey state becomes darker the higher the light intensity forming it but also that velocity selectivity decreases. These results characterise grey states which are a basis of many atom optical elements such as adiabatic transfer and VSCPT [10].

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