Multilevel Dark States: Coherent Population Trapping with Elliptically Polarized Incoherent Light

Valery Milner and Yehiam Prior
Weizmann Institute of Science, Rehovot 76100, Israel
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We present the first experimental proof for the existence of elliptical dark states, the multilevel analog of the well known three-level dark states. An ensemble of multilevel atoms, when prepared by elliptically polarized light, becomes transparent to probe light of the same polarization. The effect stems from laser-induced coherences between many ground-state magnetic sublevels which reflect the polarization of the light that had created the dark state. The novelty and essential character of the dark states are elucidated by the experimental demonstration of their creation by incoherent light. It is anticipated that elliptical dark states will play an important role in the laser cooling and manipulation of molecules. [S0031-9007(97)05201-0]

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The process of coherent population trapping (CPT) plays an important role in many elements of contemporary atomic and optical physics. CPT is being routinely used for laser cooling of atoms below the recoil limit [1,2], for trapping of atoms in optical lattices [3], and for the spatial manipulation of atoms by their adiabatic transfer between intensity-dependent dark states [4]. CPT has been shown (theoretically in [5] and experimentally in [6]) to lead to ionization suppression. The coherence between the atomic states participating in CPT gives rise to high efficiency in nonlinear and phase-matching-dependent processes such as frequency conversion [7], electromagnetically induced transparency [8], elimination of self-focusing [9], and lasing without inversion [10].

Dark states have been originally generated in closed three level Λ systems when two quantum interaction pathways interfere destructively [11,12]. In such systems, a coherent superposition of the two ground levels is generated, forming a dark state uncoupled to the radiation which had induced it. As long as the coherence persists, atoms in the dark state cannot be excited further, and once entering the state, cannot escape. This accumulation of population in the dark state is the corner stone in many optical cooling cycles.

However, since even simple atomic transitions have a magnetic sublevel structure which often cannot be ignored, coherent interactions in multilevel systems are receiving more attention [13,14]. Population trapping under *circularly* polarized excitation (well known as "optical pumping") in a degenerate two-level system is straightforward. If the number of the ground-state sublevels is greater than (or equal to) the number of the sublevels in the excited state, circularly polarized excitation preserves the population of the noninteracting "edge" magnetic sublevels (sublevels with the highest projection of angular momentum), where the population effectively accumulates. The existence of population trapping under the interaction with *elliptically* polarized light is not obvious, since all sublevels interact with the exciting light. A theoretical analysis of this case has been presented in [15], where the stationary solutions $\hat{V}\Psi_{\mathrm{CPT}} = 0$ of the time-dependent Schrödinger equation have been found analytically. These are coherent superpositions of the ground-state m sublevels, termed here as elliptical dark states or EDS, which are completely uncoupled to the applied elliptically polarized light. Ψ_{CPT} was shown to depend only on the light ellipticity and angular characteristics of a two-level system.

Although the interaction with elliptically polarized light is important in many experiments such as adiabatic momentum transfer [16,17], multipath atomic interferometry [18], and fabrication of arbitrary quantum cavity states [19], elliptical dark states have never been considered. In this Letter we present the first direct observation of multilevel EDS. We demonstrate experimentally that an open atomic system, when prepared by elliptically polarized light, does not interact with a probe light beam of the same ellipticity and becomes transparent to it. The atoms are not pumped out, as would have been naively expected for an open system, but rather a significant fraction of them survives in a coherent superposition of ground-state sublevels, the EDS. The character of the dark state is determined by the ellipticity of the light, eccentricity and orientation of the polarization, defined in our representation by the intensity ratio and phase difference between the two circular components of the polarized light, respectively. Unlike the case of coherent population trapping in a nondegenerate three-level scheme, in a degenerate system the quantum interference between the interaction pathways is created by the polarization components of the same laser field. We demonstrate experimentally (see below) that, because of this feature, the dark states in such systems may be induced by incoherent light, and therefore are much more robust than their three-level nondegenerate counterparts.

Consider first the D_1 $F = 2 \rightarrow F = 2$ transition in a sodium atom. The energy level scheme and all possible

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links of thistransition are shown in Fig. 1(a), where the quantization axis is taken to be directed along the wave vector of the applied radiation. The presence of the $F = 1$ ground state (not shown) makes this system open. Since the spontaneous decay rate to this state is equal to the decay rate back to the initial $F = 2$ level, under illumination by an incoherent combination of left and right circularly polarized light the atoms will be completely pumped out within a few lifetimes. The situation is different when the circular components are phased, as in the case of elliptically polarized light. The atoms will be trapped in a coherent superposition of the three ground-state sublevels ($m = -2, 0, +2$), connected to the two excited ones ($m = -1, +1$) by a double-Λ chain (solid lines). To show that a Λ structure provides the sufficient and necessary condition for CPT, Morris and Shore [13] considered a unitary transformation of the wave function basis into a new set of eigenstates, one of which is completely uncoupled to the exciting light. Depending on which state consists of more magnetic sublevels, the new uncoupled level belongs to either the ground state (in a Λ scheme), or to the excited state (in a V scheme). In the case of pure lifetime broadening, the lifetime of the uncoupled state scales as the population relaxation time. Thus, only a Λ-type chain is capable of producing a lasting coherent superposition between the sublevels (in the ground state). This is the reason why in the case of the sodium D_2 line, elliptically polarized dark states cannot build up within the $F = 2 \rightarrow F = 3$ transition which consists of the two V chains [Fig. 1(b)]. As was briefly discussed in our earlier work [20], the balance between the right circularly and left circularly polarized components, as defined by the light ellipticity, together with the distribution of the corresponding Clebsch-Gordan coefficients determine the dark state population distribution amongst the magnetic sublevels of the ground state. A full theoretical discussion will be given elsewhere [21].

The experiments were performed in a collimated atomic beam of sodium crossing two parallel laser beams separated by 2 mm (Fig. 2). The first elliptically polarized pump beam prepares the system, and the total fluorescence induced by the second (weak) probe is detected while the ellipticity of the probe beam is varied continuously between left and right circular by a photoelastic polarization modulator (Hinds, PEM-FS3). The polarization of the pump laser beam is accurately set by a Soleil-Babinet compensator to a well defined ellipticity and is maintained at a constant value throughout each experiment. The pump beam intensity corresponded to a Rabi frequency of 50 MHz (5 times larger than the natural linewidth), while the intensity of the probe was 2 orders of magnitude lower. The diameter of the pump beam is 1 mm (2 times larger than the size of the probe), which for the known atomic velocity ($v \approx 10^5$ cm/s) corresponds to an interaction time within the pump of more than 50 spontaneous lifetimes of the sodium $3^2P_{1/2}$ excited state ($\sim$16 nsec). The presence of the additional $F = 1$ level in the excited state 189 MHz apart from $F = 2$ (not shown in Fig. 1) can be safely neglected for all laser intensities in our experiments. The conditions in the vacuum chamber guarantee that there are no collisions within the interaction region and thus, the ground-state relaxation time is very long. The latter has been independently verified by measuring the linewidth of the laser-induced fluorescence spectrum at right angle to the sodium beam, after the atomic beam had been skimmed down to restrict the geometrical residual Doppler broadening to less than the natural linewidth. Three pairs of Helmholz coils were constructed around the interaction region so that a near zero magnetic field ($<$0.05 G) in all three dimensions was achieved, preventing the magnetic destruction of the laser-induced coherences.

A direct observation of the elliptical dark state is depicted in the left column of Fig. 3. Both pump and probe are resonant with the D_1 $F = 2 \rightarrow F = 2$ transition. The lasers were not locked to the atomic resonance, but the frequency drift during the data collection was verified to be much smaller than the natural linewidth. The normalized probe-induced-fluorescence intensity is measured

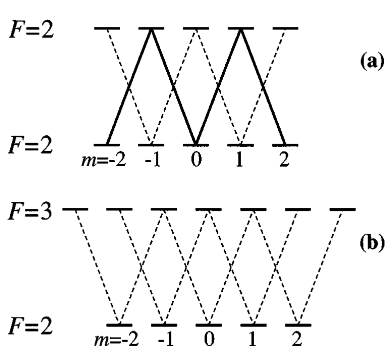

FIG. 1. Energy level and coupling scheme for the D_1 $F = 2 \rightarrow F = 2$ and the D_2 $F = 2 \rightarrow F = 3$ transitions (selection rules are $\Delta m = \pm 1$ according to the assumed quantization axis direction). Solid and dashed lines represent independent Λ and V chains.

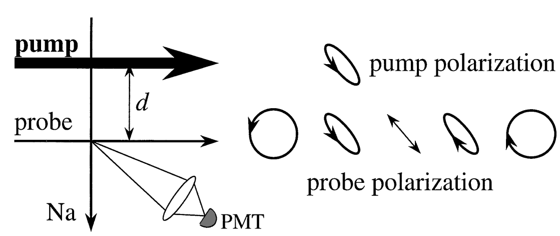

FIG. 2. Experimental arrangement for elliptical dark state observation. The pump polarization is constant and the probe ellipticity is continuously scanned from left to right circular. During the scan, only the eccentricity of the probe polarization is changed, while the major axis of the probe polarization ellipse is constant and coincides with that of the pump beam. The probe-induced fluorescence, extracted by spatial filtering out of the total fluorescence signal, is measured directly.

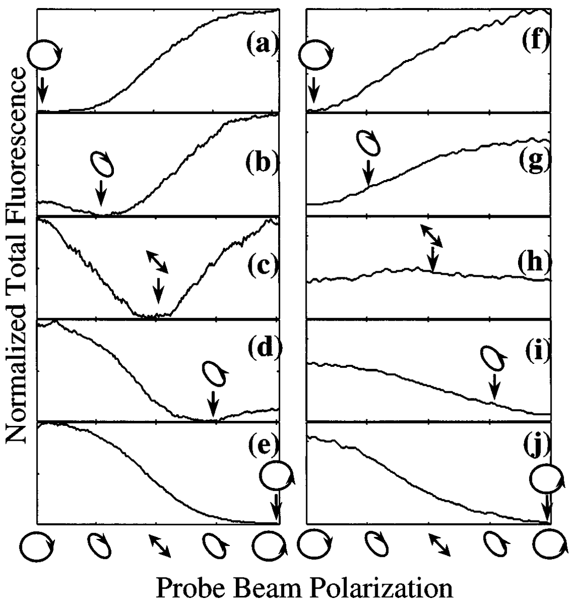

FIG. 3. Elliptical dark states and their dependence on the pump beam ellipticity. In each panel, the pump beam ellipticity is denoted by an arrow pointing to the horizontal ellipticity scale, and the probe-induced fluorescence signal is measured as a function of the probe beam ellipticity. (a)–(e) Pump beam is resonant with the D_1 $F = 2 \rightarrow F = 2$ transition; the fluorescence minima when the pump and probe polarizations coincide manifest the dark states, observed for each elliptical polarization. (f)–(j) Pump beam is resonant with the D_2 $F = 2 \rightarrow F = 3$ transition. No dark state is observed (see text).

while the ellipticity of the probe beam is scanned and the pump polarization (indicated by the arrows) is kept constant during each scan. When the probe polarization coincides with that of the pump, the probe beam does not interact with the atoms and the fluorescence signal drops to zero, manifesting the new dark state. Slight deviations of the probe ellipticity from the "dark" point result in nonzero fluorescence, proving that the $F = 2$ ground state had not been emptied out. For left (right) circular polarization [panels 3(a) and 3(e)], the population is optically pumped into the single uncoupled $m = -2(+2)$ sublevel and cannot be seen by a probe beam of the same polarization scanning the same transition. In panels 3(b)–3(d) we demonstrate that coherent population trapping exists for any elliptical pump polarization. Even though all magnetic sublevels of the double-Λ chain ($m = -2, 0, +2$) are coupled to the excited state, and there is population present in the ground-state manifold, the presence of the dark state is obvious and since the fluorescence minima coincide exactly with the pump ellipticity, its "polarization" reflects the polarization of light which had created it.

A strong test for the concept of elliptical dark states is provided when the pump is tuned to the D_2 $F = 2 \rightarrow F = 3$ transition (right column in Fig. 3). Under these conditions, the fluorescence minimum does not "follow" the pump beam ellipticity [panels 3(g)–3(i)], but rather remains at the edges of the polarization scale. As expected for V-type coupling, the atoms are all being pushed incoherently towards the $m = \pm 2$ ground sublevels, and accumulate mostly on $m = -2$, when the left circular component is stronger than the right one [left minima in the panels 3(f) and 3(g)], and on $m = +2$ [right minima in 3(i) and 3(j)] in the opposite case. Note that the atoms survive in the $F = 2$ ground state, because the $F = 2 \rightarrow F = 3$ transition is closed, and not due to coherent population trapping.

An additional test verifying the nature of the EDS was performed by observing their disappearance in a nonzero magnetic field. When a magnetic field of 1 G is applied along an arbitrary direction, the Zeeman splitting of 2.88 MHz between the adjacent $\Delta m = \pm 2$ sublevels destroys the laser-induced coherences [22]. Full discussion of the effect of magnetic field on the evolution of EDS will be presented elsewhere [21].

In the present experiment, the dark state is created among many ground-state sublevels by a single laser beam, unlike the "classical" three-level situation which involves two separate, possibly nondegenerate, transitions (and two light beams). In our model we decompose the elliptically polarized light into two circular components, which destructively interfere to create the EDS. Since these components are derived from the same laser, once the ellipticity is fixed, the amplitude ratio and the phase difference between them are constant. The corollary is that the total phase of the light is insignificant, and the dark state may also be produced by incoherent light. This prediction is important in order to ascertain the coherent superposition as a dark state, and indeed, the prediction is supported by a direct experiment.

As shown in our theoretical work [20], the pump fully establishes the dark state within several spontaneous lifetimes. This estimate provides a required coherence time scale for the dark state creation. The pump and probe beams were derived from two separate lasers, of ~2 MHz linewidth each, which are therefore coherent on this time scale. To prove that elliptical dark states may also be created by incoherent light, the experiment was repeated with laser light that was artificially spectrally broadened by a standard technique (see [23] and references therein). The pump beam was modulated by an acousto-optic modulator, driven by a carrier 200 MHz rf field, which was mixed with amplified and filtered thermal rf noise. The real-time trace of the noisy rf signal shows that the mean time between the phase jumps is less than the lifetime of the excited state, and therefore is much less than the dark state creation time (left inset in Fig. 4). The spectral broadening of the pump is depicted in Fig. 4 (right inset), where the linewidth of the D_1 $F = 2 \rightarrow F = 2$ laser-induced fluorescence signal in the weak field limit is plotted. The two results (in the time and frequency domain) verify that this light is incoherent on the relevant

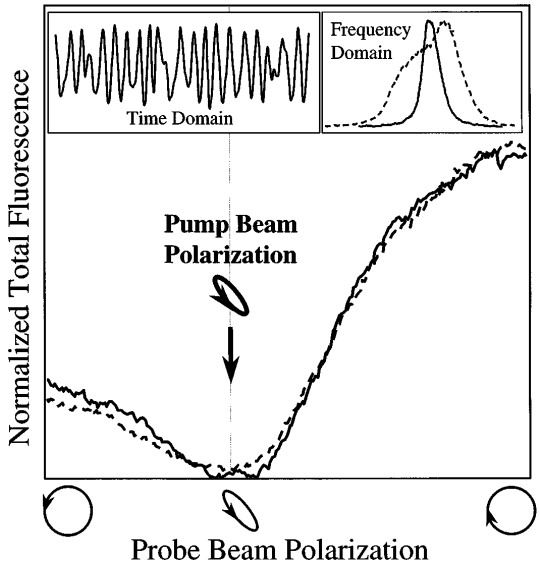

FIG. 4. Elliptical dark state, induced by coherent (solid line) and incoherent (dashed line) light (see text). Left inset: Real-time trace of the noisy rf field used to obtain the incoherent source. The full scale is 100 nsec. Right inset: Laser-induced fluorescence spectra of the D_1 $F = 2 \rightarrow F = 2$ line with (dashed line) and without (solid line) artificial spectral broadening of laser.

time scale. The solid line in Fig. 4 represents the dark state, induced by an elliptically polarized coherent pump, while the dashed line demonstrates the elliptical dark state created by incoherent (broadened) light. Within experimental error, there is no difference between the two, and the robustness of the dark state is very clear.

In conclusion, in this paper we have demonstrated for the first time the existence of dark states induced by elliptically polarized light in a degenerate open multilevel system. These atomic states are the multilevel analogs of the three level dark states, used very often in various studies in atomic physics. Each EDS reflects the ellipticity of the light that had created it, and is transparent to probe light of the same polarization. The elliptical dark states were shown to be insensitive to the coherence time of the exciting elliptical light, in complete agreement with our theoretical predictions. Unlike the adiabatic preparation of alignment by circularly polarized light, which starts from a single uncoupled m sublevel [17], the EDS formation need not be adiabatic and may be induced in any ensemble of atoms or molecules. The new multilevel EDS should prove important and useful for the preparation and manipulation of atoms and molecules by laser light, in analogy to the role played by the "standard" CPT in atomic physics in recent years.

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