Experimental observation of transient velocity-selective coherent population trapping in one dimension

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We report the observation of transient velocity-selective coherent population trapping (VSCPT) in a beam of metastable neon atoms. The atomic momentum distribution resulting from the interaction with counterpropagating $\sigma_+$ and $\sigma_-$ radiation which couples a $J_g = 2 \leftrightarrow J_e = 1$ transition is measured via the transversal beam profile. This transition exhibits a stable VSCPT dark state formed by the two $|J = 2, m = \pm 1 \rangle$ states, and a metastable dark state containing the $|J = 2, m = 0 \rangle$ states. The dynamics of the formation and decay of stable and metastable dark states is studied experimentally and numerically and the finite lifetime of the metastable dark state is experimentally observed. We compare the measured distribution with a numerical solution of the master equation.

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Velocity-selective coherent population trapping (VSCPT) occurs by optical pumping of atoms into states with well defined momenta, which are decoupled from the light field. In a $J_g = 1 \leftrightarrow J_e = 1$ transition a dark state exists, which is a trapping state for zero momentum, leading to efficient cooling below the one-photon recoil limit [1, 2], which may be assisted by polarization-gradient precooling [3]. For ground state angular momenta $J_g > 1$ several dark states may exist. These are not necessarily eigenstates of the kinetic energy Hamiltonian and thus they are transient, not leading to population trapping. The stability of those states may be recovered by introducing $m$-dependent shifts of the Zeeman sublevels by a dc stark field [3] or additional laser fields [4]. The extension of VSCPT on three dimensions has also been discussed [5]. The existence of transient dark states has for instance been used to demonstrate a multiple beam atomic interferometer in cesium [6]. Furthermore, for specific polarizations of the laser fields the existence of high-velocity dark states has been shown [7]. Recently studies of velocity-selective coherent population trapping and its relation to electromagnetically induced transparency (EIT) and atomic entanglement where reported [10].

For a $J_g = 2 \leftrightarrow J_e = 1$ transition coupled by counterpropagating $\sigma_+$ and $\sigma_-$ beams the existence of transient VSCPT has been shown theoretically [4], but to the best of our knowledge experimental momentum distributions have not been reported. In particular the metastability of some of the dark states has not been directly shown in the experiment. We present measurements that clearly show the signature of a stable and a transient dark state in the momentum distribution of a beam of neon atoms in the metastable state $^3P_2$.

The coupling scheme for the $J_g = 2 \leftrightarrow J_e = 1$ transition driven by $\sigma_+$ and $\sigma_-$ irradiation with equal frequency is shown in Figure I. The system is characterized by the Hamiltonian,

$$H = \frac{p^2}{2M} + i\hbar \omega_0 p_x + V_{AL},$$

where $p_x$ is the projector onto the excited states, $p$ the momentum operator and $M$ is the atomic mass. The interaction Hamiltonian $V_{AL} = V_L + V_{IW}$ consists of two parts. One describes the $\Lambda$-system \{g$_-1$, e$_0$, g$_{+1}$\}, the other the inverted-W configuration \{g$_{-2}$, e$_{-1}$, g$_0$, e$_{+1}$, g$_{+2}$\}. In the rotating wave approximation the two terms read

$$V_L = \sum_q \frac{\hbar}{2} \left[ \Omega_+ |e_q, q + h\rangle \langle g_{-1}, q - h| - \Omega_- |e_q, q + h\rangle \langle g_{-2}, q - 2h| \right] + \text{h.c.},$$

$$V_{IW} = \sum_q \frac{\hbar}{2} \left[ \sqrt{\frac{\Omega-}{\Omega+}} |e_{-1}, q - h\rangle \langle g_{-2}, q - 2h| + \sqrt{\frac{\Omega+}{\Omega-}} |e_{+1}, q + h\rangle \langle g_{+2}, q + 2h| \right] e^{-i\omega_L t} + \text{h.c.},$$

where $\Omega_+$ ($\Omega_-$) is the Rabi frequency of the coupling laser for the transition $|g_0\rangle \leftrightarrow |e_{+1}\rangle$ ($|g_0\rangle \leftrightarrow |e_{-1}\rangle$).

The interaction Hamiltonian $V_{AL}$ separates the atomic states into two sets of momentum families, which are coupled to each other only by spontaneous emission. These are

$$\mathcal{F}_L(q) = \{|e_0, q\rangle, |g_{-1}, q - h\rangle, |g_{+1}, q + h\rangle\}$$

$$\mathcal{F}_{IW}(q) = \{|g_{-2}, q - 2h\rangle, |e_{-1}, q - h\rangle, |g_0, q\rangle, |e_{+1}, q + h\rangle, |g_{+2}, q + 2h\rangle\}.$$
The $3^c\Lambda(q)$ family involves two states which have for $q = 0$ the same energy. Thus two-photon resonance within the $\Lambda$-system can be maintained using a single laser frequency and the dark state $|\Psi_{NC}^{\Lambda}(q = 0)\rangle$ formed by the members of this family will be stable. The $3^J_W(q)$ family involves states with different momenta. Thus two-photon resonance between the states $|g_{\pm 2}\rangle$ and $|g_0\rangle$ cannot be maintained with a single laser frequency. Furthermore the dark state $|\Psi_{NC}^{J_W}\rangle$ formed within this family is no eigenstate of the kinetic energy Hamiltonian $p^2/2M$. The lifetime $\tau_{J_W}$ of this state has been calculated perturbatively, assuming that all Clebsch-Gordan coefficients are equal $\frac{3}{4}$. The two dark states read

$$|\Psi_{NC}^\Lambda\rangle = \frac{1}{\sqrt{\Omega_+^2 + \Omega_-^2}} \times \left[ \Omega_- |g_{-1}, q - \hbar k\rangle - \Omega_+ |g_{+1}, q + \hbar k\rangle \right],$$

(6)

$$|\Psi_{NC}^J_W\rangle = \frac{1}{\sqrt{\Omega_+^4 + 6\Omega_+^2\Omega_-^2 + \Omega_-^4}} \times \left[ \Omega_+^2 |g_{-2}, q - 2\hbar k\rangle - \sqrt{6} \Omega_+ \Omega_- |g_0, q\rangle + \Omega_-^2 |g_{+2}, q + 2\hbar k\rangle \right].$$

(7)

For interaction times $\tau < \tau_{J_W}$ both dark states appear as trapping states for $q = 0$, giving rise to a momentum distribution with peaks at $-2\hbar k, -\hbar k, 0, \hbar k$ and $2\hbar k$. For interaction times $\tau > \tau_{J_W}$ the contribution of $|\Psi_{NC}^J_W\rangle$ vanishes and only two peaks at $\pm \hbar k$ remain in the momentum distribution.

In the experiment, a beam of neon atoms emerges from a liquid nitrogen cooled discharge nozzle source. A fraction of the order of $10^{-4}$ of the atoms is in the metastable states $3^2P_0$ or $3^2P_2$ of the $2p^53s$ electronic configuration. The flow velocity of the atoms is about $470 \text{ ms}^{-1}$ with a width of the velocity distribution of about $100 \text{ ms}^{-1}$ (FWHM). The beam is collimated by a $50 \mu\text{m}$ and a $10 \mu\text{m}$ slit positioned $144 \text{ cm}$ apart, corresponding to a width of the transversal velocity distribution of about $0.15\hbar k$. The population of the metastable $3^2P_0$ state is depleted by optical pumping before the atoms interact with the circular polarized laser beams in a region $20 \text{ cm}$ downstream of the second slit. The spatial distribution of the atoms in the states $3^2P_0$ and $3^2P_2$ is measured $120 \text{ cm}$ downstream of the interaction zone using a movable channeltron detector behind a entrance slit of $25 \mu\text{m}$ width, leading to a resolution of the transverse momentum of $\Delta p \approx 0.2\hbar k$. In the interaction region the magnetic field is actively compensated to less than $1 \mu\text{T}$, to assure the degeneracy of all Zeeman states to within better than $130 \text{ kHz}$. The laser beam passes through a polarizer, two quarter wave plates and optionally two cylindrical lenses before being retroreflected (Fig. 2), establishing a counterpropagating $\sigma_+ - \sigma_- - \text{configuration.}$ In order to realize different interaction times three different setups for the width of the laser beams were used. The first setup uses cylindrical lenses in a confocal arrangement with the atomic beam crossing the laser beam near the focus. The transit time of the atoms through the laser beam is estimated to be a few $100 \text{ ns}$, corresponding to $\Gamma t \approx 10$, where $\Gamma$ is the width of the transition between the $3^2P_1$ and $3^2P_2$ state. The laser beam profile at the position of the atomic beam is not measured directly but inferred from the dimensions of the optical setup. The laser beams are parallel to within $10^{-5}$ rad. The peak Rabi frequency is in the order of $500 \text{ MHz}$ and the lasers are tuned from the $|g_0\rangle \leftrightarrow |e_0\rangle$ resonance by $100 \text{ MHz}$ to reduce the influence of stray light from the windows. To increase the interaction between the atoms and the light field the cylindrical lenses can be removed, leading to an interaction time of $\Gamma t = 200$. Using a telescope in front of the polarizer the beam diameter can be increased to $8 \text{ mm}$, leading to an interaction time of $\Gamma t = 800$.

Figure 3 shows the initial momentum distribution (grey area) and the result for a short interaction time of $\Gamma t \approx 10$ (squares). Five peaks at $\pm 2\hbar k, \pm \hbar k$ and zero momentum are clearly resolved. The asymmetry of the momentum distribution is due to different intensities of the $\sigma_+$ and $\sigma_-$ beams: The retroreflected beam passes twice through a window and the (uncoated) cylindrical lens before crossing the atomic beam, resulting in a lower
FIG. 3: Transverse atomic momentum profile after a short interaction (Γt < 10). The grey area shows the initial momentum distribution (scaled down), the dashed lines are gaussian fits to the individual peaks. The full line is the sum of the gaussian fits.

intensity in the retroreflected beam. Thus the Rabi frequencies of the two laser beams are not equal, Ω+ ≠ Ω−, therefore we find an asymmetric population distribution within the dark states |g−> and |g0>. This is also confirmed by numerical simulations of the process. The peak at zero momentum also contains contributions from population in the 3P0 state, which is populated by spontaneous emission from the upper state 3P1 during the interaction. In order to do a supplementary test that we observe velocity-selective coherent population trapping the retroreflected beam was slightly tilted. A sufficient overlap of both beams was sustained but the retroreflected beam interacts with the atoms after the dark states have been populated. Due to optical pumping the population of the states |g−>, |g−1> and |g0> is depleted, and the height of the peaks at negative momenta decrease, since the internal and external states in the dark states are strongly correlated. The measurement with tilted lasers is shown in figure 4, which shows good agreement of experimental and calculated profiles.

When increasing the laser beam diameter with a telescope to 8 mm the interaction time is of the order Γt ≈ 800. We then observe the transversal beam profile shown in figure 5. The peaks at ±2ℏk are no longer seen and only the stable dark state |ΨΛNC⟩ survives, reflected by the peaks at the momenta ±ℏk. The peak at zero momentum appears because the state 3P0 is populated by spontaneous emission during the process of dark state preparation.

We compare the measured data to a solution of the generalized optical Bloch equations in the family momentum basis |ΨΛNC⟩, neglecting spontaneous decay to other levels outside the system. The Bloch equation reads

FIG. 4: Transversal momentum distribution after the interaction with a tilted retroreflected laser. The dashed lines are gaussian fits to the individual peaks, the solid line is their sum. The depopulation of the peaks with negative momentum is clearly visible.

$$\dot{\rho} = -\frac{i}{\hbar} [H, \rho] - \frac{\Gamma}{2} [(\Delta^+ \cdot \Delta^-) \rho + \rho (\Delta^- \cdot \Delta^+)] + \frac{3\Gamma}{8\pi} \int d^2\Omega \sum_{\epsilon \perp n} (\Delta^+ \cdot \epsilon) \exp(-i\epsilon \cdot \mathbf{R}) \rho \exp(i\epsilon \cdot \mathbf{R})$$

FIG. 5: Transverse atomic momentum profile after an interaction of 8 μs (Γt ≈ 800). The lines are gaussian fits to the peaks. The peaks at p = ±ℏk reflect the stable dark-state |ΨΛNC(q = 0)⟩.
into the solid angle around the direction 
ences by spontaneous emission of a fluorescence photon 
the feeding of the ground-state population and coher-
der the integral over all solid angles (third term) describes 
ences resulting from spontaneous emission; the term un-
the decrease of the excited-state populations and coher-
only on the external variables. The second term describes 
R is the position operator of the center of mass which acts 
duced dipol operator and 
where $\Delta q$ population is encoded by the greyscale given on the right.

For a short interaction time of $\Gamma t < 15$, the atoms ab-
sorb a photon from one of the laser beams, followed by stimulated emission into the other beam. This leads to a change in momentum by $\Delta p = \pm 2\hbar k$, and peaks in the momentum distribution at $p = \pm 2\hbar k$ appear. This

FIG. 6: Dynamics of the momentum distribution, derived from a numerical solution of the Bloch equations \[2\]. The population is encoded by the greyscale given on the right.

process of stimulated raman transitions causes an effi-
cient population transfer to the dark state $|\Psi_{NC}^{1W}(q = 0)\rangle$, characterized by a momentum distribution located at $p = \pm 2\hbar k$ and $p = 0$. Absorption of a photon fol-
lowed by spontaneous emission results in a random walk in momentum space due to the emission of photons in an arbitrary direction. This diffusive process successively 
populates the stable dark state $|\Psi_{NC}^{1W}(q = 0)\rangle$ which is characterized by the two peaks at momentum $p = \pm \hbar k$ in the momentum representation. Due to the statisti-
cal nature of this process the population of the dark state $|\Psi_{NC}^{1W}(q = 0)\rangle$ increases for longer interaction times, $\Gamma t > 200$.

The calculations show good agreement with the mea-
sured data. The calculations show a structure with five peaks at $-2\hbar k, ..., 2\hbar k$ for $\Gamma t \approx 100 - 200$, while the mea-
surements yield this structure for $\Gamma t \approx 10 - 20$. The values for the interaction time are not directly compar-
able, as the calculations where done for a constant Rabi 
frequency, while in the experiment the light fields have a gaussian shape. Furthermore the widths of the mea-
sured peaks is smaller than expected from the simula-
tions, which is due to neglecting spontaneous emission out of the system $\{^3P_1, ^3P_2\}$.

The measurements for a long interaction time of $\Gamma t = 800$ (Fig. 5) show good agreement with the numerical 
results. The population of the stable dark state (9) is rising, while the population of other states is decaying 
into this dark state via the diffusion in momentum space due to the spontaneous emission of photons.

In this work we have presented measurements which di-
rectly demonstrate the existence of transient dark states 
with a well defined momentum distribution in a $m$-state 
manifold of a $J_g = 2$-level coupled to a level with $J_e = 1$ 
by counterpropagating $\sigma_+$ and $\sigma_-$ radiation. The mea-
sured data show good agreement with quantum density 
matrix calculations for the velocity distribution for short 
as well as longer interaction times. For a quantitative 
analysis more detailed experiments as well as calculations 
including spontaneous emission into other states than the $^3P_2$-state are needed.

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