A multi-photon magneto-optical trap

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Abstract

We demonstrate a Magneto-Optical Trap (MOT) configuration which employs optical forces due to light scattering between electronically excited states of the atom. With the standard MOT laser beams propagating along the $x$- and $y$- directions, the laser beams along the $z$-direction are at a different wavelength that couples two sets of excited states. We demonstrate efficient cooling and trapping of cesium atoms in a vapor cell and sub-Doppler cooling on both the red and blue sides of the two-photon resonance. The technique demonstrated in this work may have applications in background-free detection of trapped atoms, and in assisting laser-cooling and trapping of certain atomic species that require cooling lasers at inconvenient wavelengths.

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The development of laser cooling and trapping techniques in the last three decades has greatly enhanced our ability to control atoms, impacting a range of fields from precision atomic measurements and atomic clocks to quantum degenerate gases and quantum information processing. To date, most laser cooling methods use the mechanical effect of single-photon transitions between ground states and electronically excited states. These include Doppler cooling, polarization gradient cooling, and velocity-selective coherent population trapping [1]. There are, however, a few theoretical and experimental studies involving laser cooling in three-level systems comprising a ground state and two electronically excited states. For example, ref. [2] showed an enhancement of radiation pressure by driving a 2-photon transition in a 3-level system. In other work, the effective linewidth for the cooling transition was controlled by dressing the excited state via a coupling to another excited state. This effect can either broaden [3, 4] or narrow [5] the effective single-photon cooling transition.

Exploiting the Doppler and Zeeman shifts of single-photon optical dipole transitions, the Magneto-Optical Trap (MOT) [6] has been the standard tool to cool and trap neutral atoms in 3D. The primary motivation of this work is to use Doppler and Zeeman shifts of multi-photon transitions to both cool and trap atoms. We demonstrate a trap geometry where the cooling and trapping of atoms along one axis of the 3D-trap is due entirely to optical forces from transitions between two electronically excited states [7]. Specifically, with the 852 nm cooling laser beams of a standard cesium(133Cs) MOT propagating along the x- and y- directions, we replace the laser beams along the z-direction with counter-propagating 795 nm laser beams that only couple the excited states of cesium (6P\textsubscript{3/2} F′\textsubscript{′}=5) to a third set of excited states (8S\textsubscript{1/2} F′′\textsubscript{′}=4) (see Fig. [1]). In this two-color MOT we find efficient cooling along the z-direction at both small and large two-photon detunings, while a magneto-optical restoring force was found when the helicities of the 6P-8S beams are opposite to those for the standard MOT. Remarkably, the two-color MOT can reach sub-Doppler temperatures at both positive and negative two-photon detunings.

The new feature of the two-color MOT sketched in Fig. [1] is in the cooling and trapping along the z-direction. Consider the low intensity regime where the rate of excited atoms leaving from both 6P\textsubscript{3/2} and 8S\textsubscript{1/2} states is dominated by spontaneous emission, with negligible contribution from stimulated processes. In this regime, the dominant radiation pressure along \( \hat{z} \) is due to 2-photon scattering, where the first photon is absorbed from the
FIG. 1: (Color online) (a): Schematic of the setup in this work; \( \sigma^\pm \) are specified with respect to the positive \( x, y \) and \( z \) axes. (b): Simplified level diagram and related transitions.

In-plane laser beams and the second is absorbed from the beams along \( \hat{z} \). In particular, we consider \( R_{ij}^{(2)} \), the rate of 2-photon scattering induced by a 6S-6P beam along \( \hat{i} \) and a 6P-8S beam along \( \hat{j} \). Here \( \hat{i} \in \{ \hat{x}, -\hat{x}, \hat{y}, -\hat{y} \} \) is one of the four directions of the 6S-6P beams, and \( \hat{j} \in \{ \hat{z}, -\hat{z} \} \) is one of the two directions of the 6P-8S beams. The scattering force along \( \hat{z} \) can be written as

\[
f^{(2)}_z = \hbar k_{ee'} \sum_{ij} R_{ij}^{(2)} \hat{j}.
\]

For an atom moving at velocity \( \mathbf{v} \), we have the 2-photon scattering rate in the low intensity limit:

\[
R_{ij}^{(2)} = \frac{\gamma |\Omega_{ge} \Omega_{ee'}|^2}{16|\Delta_1 - k_{ge} \hat{i} \cdot \mathbf{v}||\delta_2 - k_{ge} \hat{i} \cdot \mathbf{v} - k_{ee'} \hat{j} \cdot \mathbf{v}|^2}.
\]

Here \( \Omega_{ge} \) and \( \Omega_{ee'} \) are the Rabi frequencies of the laser induced couplings per beam; \( k_{ge} \) and \( k_{ee'} \) are the wavenumbers of the laser beams; \( \Delta_1 = \Delta_1 + i\Gamma/2 \) and \( \delta_2 = \delta_2 + i\gamma/2 \); \( \Delta_1 \) and \( \delta_2 \) are the 1-photon and 2-photon detunings for the 6S_1/2 \( F = 4 \) to 8S_1/2 \( F'' = 4 \) 2-photon excitation, with 6P_3/2 \( F' = 5 \) as the intermediate level (Fig. 1b); \( \Gamma/2\pi = 5.2 \) MHz and \( \gamma/2\pi = 1.5 \) MHz are the linewidths of the 6P_3/2 and 8S_1/2 states respectively.

Taylor-expanding Eq. (1) around \( v_z = \hat{z} \cdot \mathbf{v} = 0 \) gives

\[
f_z^{(2)} \approx -\alpha(2)v_z, \text{ with } \alpha(2) > 0 \text{ (damping)} \text{ for negative 2-photon detuning } \delta_2 < 0.
\]

This 2-photon version of the usual [1] Doppler cooling mechanism can be summarized with the level diagram in Fig. 2a: the Doppler effect enhances the absorption cross-section for the 6P-8S beam opposing the velocity. One qualitative difference from standard Doppler cooling is that the 2-photon transitions to the 8S states are not closed, so we expect that repumping light will be important to keep the population from pumping into the 6S_1/2 \( F = 3 \) ground states.

In addition to the velocity-dependent force, a position-dependent restoring force along the \( z \)-direction is essential for trapping. Figure 2b illustrates the basic principle of the trapping force. To simplify our discussion, we consider a hypothetical atom with angular momentum
FIG. 2. (Color online) (a): Schematic illustration of the velocity damping due to the Doppler effect for 2-photon scattering. Here $\Omega_{\pm e e'^{\pm}}$ represents the 6P-8S beam from the $\pm \hat{z}$ direction. (b): Schematic illustration of the trapping force along $z$ due to the Zeeman shift ($z$ quantization axis) of intermediate resonance in the 2-photon scattering in a linearly changing magnetic field. Only the excitation pathway enhanced by the Zeeman shift is shown. (c): Peak fluorescence of the two-color MOT vs 2-photon detuning $\delta_2$. Here $s_{ge} = 1$, $s_{ee'^{\prime}} = 15$. Inset gives a fluorescence image of the MOT at $\delta_2/2\pi = -3$ MHz.

$J=0$ ground state, $J'=1$ intermediate states and $J''=0$ excited state. As with the cooling force, the trapping force along $\hat{z}$ is due to the scattering of the 6P-8S light. The position dependence of this force is due to the spatially dependent Zeeman shift of the intermediate 6P$_{3/2}$ levels. Taking the quantization axis along $\hat{z}$, the 6S-6P beams in the $x-y$ plane provide both $\sigma$ and $\pi$ couplings between the ground state and the intermediate states. For a magnetic field along $+\hat{z}$, ($z > 0$: right side of Fig. 2b), the intermediate detuning of the 2-photon excitation is shifted toward resonance for the excitation pathway involving a $\sigma-$ transition to the intermediate state followed by a $\sigma+$ transition to the excited state. As a result, the atoms at $z > 0$ preferentially absorb the 6P-8S light propagating toward $-\hat{z}$, leading to a restoring force in a magnetic quadruple field. Unlike the damping force, this restoring force has the correct sign for both positive and negative $\delta_2$ when $\Delta_1 < 0$.

The above analysis is corroborated by our experimental observations. In particular, at moderate 6P-8S intensity the 2-photon detuning must be negative to achieve laser cooling along the $z$-direction of the trap. Surprisingly, at high intensities laser cooling and trapping behave differently. As detailed below, we found laser cooling on both the red and blue sides of the 2-photon resonance. We argue that this counter-intuitive effect is due to 3-photon and higher order scattering processes.

Our experiments capture, cool and trap atoms in a cesium vapor cell. The cooling light in the $x-y$ plane comprises the two pairs of counter-propagating 852 nm laser beams (6S-
6P beams) with 8 mm $1/e^2$ diameter. (See Fig. 1) The single photon detuning $\Delta_1/2\pi = -12.5$ MHz and the peak intensity of each beam is characterized by $s_{ge} \equiv \frac{2\Omega_{ge}^2}{\Gamma}$. The gradient of the magnetic quadruple field was 1.4 mT/cm along $\hat{z}$. The beams along $\hat{z}$ are a pair of 795 nm laser beams (6P-8S beams), and the peak intensity of each 6P-8S beam is characterized by the parameter $s_{ee'} \equiv \frac{2\Omega_{ee'}^2}{\gamma}$. We add two counter-propagating repump beams at 895 nm along $\hat{x}$, tuned to the 6S$_{1/2} F = 3$ to 6P$_{1/2} F' = 4$ transition to keep atoms in the $F = 4$ ground states.

With the 6P-8S beams at a moderate intensity of 20 mW/cm$^2$ ($s_{ee'} \approx 15$, $\Omega_{ee'}/2\pi \approx 4$ MHz), and guided by the 2-photon Doppler cooling picture (Fig. 2b), we set the 2-photon detuning $\delta_2$ to small negative values, comparable to the 8S linewidth $\gamma$. We observe trapped atoms in the two-color MOT when the helicities of the 6P-8S beams are set to be opposite to those of the 6S-6P beams in a standard MOT (Fig. 1b, Fig. 2b). As with a standard MOT [9, 10], we find that our trap tolerates wrong helicity components in the 6P-8S beams with up to $\approx 30\%$ in intensity. As expected, the two-color MOT is more sensitive to the repump efficiency than a standard MOT, and the counter-propagating beams need to be intensity-balanced to nullify the repump radiation pressure.

In Fig. 2, we plot the peak fluorescence of the two-color MOT vs $\delta_2$. At the optimal 2-photon detuning of $\delta_2/2\pi \approx -3$ MHz and with $s_{ge} \approx 4$, up to $8 \times 10^5$ atoms at a density of $5 \times 10^{10}$/cm$^3$ are accumulated in the two-color MOT from the pressure $P \approx 10^{-5}$ Pascal ($10^{-7}$ Torr) cesium vapor. Due to the weaker trapping and damping along $\hat{z}$, both the spatial and velocity distributions of the atomic sample are elongated along $\hat{z}$. The velocity spread of the atoms along $\hat{x}$ and $\hat{z}$ is characterized by effective temperatures $T_x \approx 70$ $\mu$K and $T_z \approx 700$ $\mu$K, both of which are reduced at smaller $s_{ge}$ (see below and Fig. 4). Typical $1/e^2$ widths of the atomic spatial distribution, fit to a Gaussian, are $w_x \approx 300$ $\mu$m and $w_z \approx 600$ $\mu$m. The number of trapped atoms is an order of magnitude smaller than that of a standard MOT under similar conditions, which is likely due to the reduced capture velocity and effective capture volume for the two-color MOT.

The 2-photon Doppler cooling picture (Fig. 2) fails dramatically at high 6P-8S beam intensities. As $s_{ee'}$ increases, the range of $\delta_2$ for MOT operation broadens and shifts to the red. When $s_{ee'}$ is larger than a threshold value of $s_{th} \approx 80$, the two-color MOT also works at positive $\delta_2 > \delta_{th} \approx 2\pi \times 10$ MHz (Fig. 3a). For $s_{ee'} \approx 1.8 \times 10^3$ (not shown in Fig. 3), the two-color MOT operates for $\delta_2$ spanning a range more than $2\pi \times 100$ MHz ($>> \gamma, \Gamma$) on
both the red and blue sides of the two-photon resonance. The maximum number of trapped atoms is similar to that achieved in the low 6P-8S beam intensity regimes, but with up to 50% increase of peak atom densities.

For high 6P-8S beam intensity and moderate 2-photon detuning, both the spatial and velocity distributions of the trapped atomic sample are more isotropic than those at low intensity. As $s_{ee'}$ increases, the ratio $w_z : w_x$ can reach or even go below unity at small positive $\delta_2$. The ratio $T_z : T_x$ decreases and approaches unity as $s_{ee'}$ increases, while a larger $s_{ee'}$ is needed for the same ratio to be reached at a larger $|\delta_2|$. The effective temperature $T_x$, and remarkably, also $T_z$, decrease linearly with $s_{ge}$ until the MOT stops working. For $s_{ge} < 1$, $T_z$ is well below the 125 $\mu$K D2 Doppler limit at both large $|\delta_2|$ as well as at small positive 2-photon detunings, as shown in Fig. 4. In addition, at large $|\delta_2|$ the MOT becomes less sensitive to the repump efficiency and intensity balance, as in a standard MOT.

The observation of laser cooling and trapping on the blue side of the 2-photon resonance is intriguing. Equation (1) indicates that for $\delta_2 > 0$, the Doppler effect leads to a velocity-dependent force that becomes anti-damping. At low intensity, this precludes operation of the MOT. However, the 2-photon force picture ignores higher order scattering processes, which can be important at high intensities. These include the 3-photon process sketched in Fig. 3b in which a 2-photon absorption is followed by a stimulated emission from 8S to 6P. These multi-photon processes can lead to efficient cooling along $\hat{z}$ in a manner similar to Doppleron cooling [11]. In the same way as for 2-photon force calculations, the 3-photon scattering force can be written as $f_z^{(3)} = 2\hbar k_{ee'} \sum_{i,j} R_{ij}^{(3)} \hat{z}_{ij}$, where, for atoms moving at velocity
FIG. 4: (Color online) (a, b): Temperature of the atoms vs. $s_{ge}$ for $s_{ee'} = 1.8 \times 10^3$, with $\delta_2/2\pi = -143$ MHz in (a) and $\delta_2/2\pi = 117$ MHz in (b). Notice the different temperature scales. (c): Temperature vs $\delta_2$ for atoms in the two-color MOT at various $s_{ee'}$ for $s_{ge} = 0.6$.

As in our treatment of Eq. (1), we Taylor-expand $f_z^{(3)}$ near $v_z = 0$ to find the 3-photon damping coefficient $\alpha^{(3)}$. For $\Delta_1 < 0$ and $\gamma^2 \ll \Gamma^2 + 4\Delta_1^2$, we find $\alpha^{(3)} > 0$ for either $\delta_2 < 0$, or $\delta_2 > -\Delta_1/2$. We note that $\alpha^{(3)}$ involves only the 3-photon process and ignores 2-photon processes, light shifts and higher order processes. The 3-photon cooling effect at $\delta_2 > 0$ can be understood qualitatively from the diagram in Fig. 3b: At large $|\delta_2|$, the Doppler sensitivity along $\hat{z}$ of the 6P-8S-6P Raman process becomes independent of $\delta_2$, but remains dependent on $\Delta_1$. The fact that $\alpha^{(3)}$ is positive is determined by the negative single-photon detuning $\Delta_1$. In addition, the decreased 8S population at large $|\delta_2|$ reduces the two-color contribution to unwanted optical pumping into the $F = 3$ ground states, which helps explain the decreased sensitivity on repump light.

There are at least two possible explanations for the sub-6P3/2-Doppler temperatures observed along $\hat{z}$ over the wide range of 2-photon detunings in Fig. 4. First, as with sub-Doppler cooling in standard optical molasses [12], there is an interplay between spatially dependent light shifts and optical pumping among the 6S Zeeman sublevels, leading directly
to sub-Doppler cooling for atoms moving along $\hat{z}$. This mechanism may be non-intuitive since the 852 nm light, which is the only light field that interacts with the 6S atoms, has no polarization gradient along $\hat{z}$. However, a $z-$dependent ground state spin polarization can be induced by multi-photon optical pumping processes: the 6P-8S coupling dresses the 6P$_{3/2}$ $F'=5$ Zeeman sublevels, shifting and mixing those sublevels in a $z$-dependent way. An atom excited to a 6P$_{3/2}$ $F'=5$ dressed state is thus spin polarized, and its $z$-dependent polarization is partially retained after the spontaneous decay to the 6S ground states. Combined with a light shift of the ground states due to 2-color processes which is not only $x$-, $y$-dependent but also $z$-dependent, sub-Doppler cooling can occur along $\hat{z}$. The inseparability of the two-color ground state light shift could also provide a second contribution to the low measured temperature along $\hat{z}$, by mixing the standard sub-Doppler-cooled motion along $\hat{x}$, $\hat{y}$ with the motion along $\hat{z}$. A quantitative analysis of this “two-color” polarization gradient cooling mechanism will appear in a future publication [13].

We have demonstrated a magneto-optical trap where cooling and trapping forces along its $z$-axis are provided entirely by photons associated with transitions between excited states. Up to $8 \times 10^5$ cesium atoms are trapped in a vapor cell, and the density of the trapped atoms reaches $8 \times 10^{10}$/cm$^3$ at optimal experimental parameters. Sub-Doppler cooling occurs over a wide range of positive and negative 2-photon detunings. Since we observe no density-dependent atom loss, we conclude that two-color-induced collisional loss processes are not particularly large. We believe that the number of atoms in the two-color MOT is lower than that in the standard MOT, because there is a reduced phase-space volume for capture from the room-temperature vapor. We have also observed atom cooling and trapping in a geometry complementary to the setup given by Fig. 1a, where the 852 nm beams are along $\hat{z}$, and the 795 nm beams are along $\hat{x}$ and $\hat{y}$, although this geometry traps even fewer atoms.

The two-color cooling and trapping demonstrated here may have practical applications. For instance, a high-numerical-aperture objective can be installed to collect 852 nm fluorescence along $\hat{z}$ in our setup, a direction along which the scattering of 6S-6P beams from the nearby optics is minimized; the 6P-8S beams at 795 nm wavelength can be easily filtered out. This would enable high-efficiency, near-background-free detection of trapped atoms. This or similar MOT arrangements may also allow completely background-free detection of fluorescence from atomic transitions driven by no laser beam. As another example, replacing regular cooling lasers with excited-state coupling lasers can be technically advantageous for
laser cooling of certain atomic species. For example, for atomic hydrogen or anti-hydrogen, the Lyman-\(\alpha\) cooling transition needs 121 nm coherent radiation, which is hard to generate and manipulate [14, 15]. Instead of setting up 3 pairs of Lyman-\(\alpha\) beams that couple 1S with 2P for a regular hydrogen MOT, two pairs of the beams may be replaced by laser beams that couple 2P and 3S excited states using the more readily available 656 nm light.

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