Two-Color Excitation of Metastable Helium Atoms for Efficient Laser Cooling

W. Rooijakkers, W. Hogervorst, and W. Vassen
Laser Centre Vrije Universiteit, Department of Physics and Astronomy, de Boelelaan 1081, 1081 HV Amsterdam, The Netherlands
(Received 28 November 1994)

Radiation pressure on metastable helium atoms can be increased by an order of magnitude using the combined action of two lasers exciting the $1s2s^2S_{1/2} - 1s2p^3P_{J=2} - 1s3d^3D_{J=1}$ cascade. This increase has been measured as a function of laser intensities and is explained in a model based on the optical Bloch equations, the coherences playing a crucial role. Possible applications for deceleration are discussed.

PACS numbers: 32.80.Pj, 42.50.Lc, 42.50.Vk

Ultrasound metastable helium atoms ($\text{He}^*$) are nowadays used in several experiments of fundamental interest in physics, such as atomic diffraction [1] and velocity-selective coherent population trapping in beams [2]. Recently two-dimensional velocity-selective coherent population trapping has been achieved using $\text{He}^*$ atoms, released from a magneto-optical trap (MOT) [3]. A study of ultracold collisions between $\text{He}^*$ atoms in a MOT [4] has demonstrated that Penning ionization severely limits the densities that can be obtained. A suppression of the decay in a trap by several orders of magnitude may be realized [5] using a magnetostatic spin-polarized trap, in which the ionization process is forbidden to first order. An efficient loading scheme for such a trap with cold atoms is of interest and may be of help to open a new field in the search for collective effects.

A sample of cold atoms (optical molasses) can be prepared starting from a thermal beam source and decelerating the atoms using the spontaneous scattering of photons. To compensate for the changing Doppler shift, Zeeman slowing or laser frequency chirping has to be applied. The choice of radiation pressure (i.e., a phase gradient) instead of other forces (based on intensity and/or polarization gradients) is favored by the fact that in this case friction can be made independent of the velocity of the atoms. Also a large velocity capture range is feasible.

Because of the rather long lifetime of the $2^3P$ state (98 ns) and the rather low recoil momentum (recoil velocity 9.2 cm/s) for the $2^3S_{1/2} - 2^3P_{1/2}$ transition used in laser cooling of metastable helium atoms, the minimum stopping distance $s$ using radiation pressure is unpractically long ($s = 4.3$ m for atoms with a typical initial velocity of 2000 m/s). To slow down a substantial fraction of the atoms this distance should even be increased in order to prevent atoms from being swept out of the locking range as a result of diffusion due to the stochastic nature of the interaction with the light. For this reason several groups [4,6] use cryogenically cooled sources as the stopping distance scales with the square of the initial velocity. But even in this case only atoms in the slow tail of the initial velocity distribution could be stopped.

In this Letter we describe a method to increase the radiation force by an order of magnitude, which can be applied to reduce the stopping distance by the same factor. The idea is to use the combined action of two lasers exciting the $2^3S_{1/2} - 2^3P_{1/2}$ (1083 nm, linewidth 1.62 MHz) as well as the $2^3P_{3/2} - 3^3D_{3/2}$ transition (588 nm, linewidth 11.2 MHz). Qualitatively this increased radiation force follows from the short lifetime of the $3^3D_3$ state (compared to that of the $3^3P_2$ state) allowing more absorption–spontaneous emission cycles per unit time. Also the momentum transferred by each yellow photon is larger.

The $2^3S_1 \rightarrow 2^3P_2 \rightarrow 3^3D_3$ three-level cascade is—to very good approximation—a closed level system. There are two minor complications. First there is a decay channel of the $3^3D_3$ state to the $3^3P_2$ state with a branching ratio of $2 \times 10^{-4}$ [7]. A second complication is the fine structure of the upper state: the $3^3D_3$ and $3^3D_2$ levels are separated by 76 MHz only so that $3^3D_2$ might be excited. In both cases there is no leak as all the atoms will eventually decay back to the $2^3S_1$ state. Using circularly polarized light with the same orientation for both lasers, the atom can be optically pumped to the magnetic sublevel with the highest $m$ value, and an almost ideal three-level system will be realized.

The average force $\langle F \rangle$ on a three-level atom subjected to two laser beams is calculated via $\langle F \rangle = \text{Tr}[ -\rho \nabla H ]$, where $H$ is the Hamiltonian describing the interaction of the atoms with the laser fields and $\rho$ is the density matrix. In the case of two overlapping traveling waves, only the phase gradient part is retained, resulting in

$$\langle F \rangle = \hbar k_2 \Gamma_2 \langle \pi_2 \rangle + \hbar k_3 \Gamma_3 \langle \pi_3 \rangle.$$

Here the subscripts denote the $J$ value of the level, $\hbar k_i$ the momentum transferred by the laser when exciting level $i$, $\Gamma_i/2\pi$ the linewidth of level $i$, and $\langle \pi_i \rangle$ the average level population in a steady state situation. Instead of one term as in the case of monochromatic two-level excitation, the force now has two contributions involving the fluorescence from the intermediate and the upper states. The origin of the increased radiation force in the case of metastable helium stems from the fact that the population of the $3^3D_3$ level is multiplied by a coefficient that is much larger than the corresponding one for the $3^3P_2$ level. The optimization of the force (1) is almost, though not entirely, equivalent to the optimization of the population in the upper level.
One might intuitively guess that at high saturation the populations would be equally distributed over the three levels. This, however, is not correct since in that case the coherences between the different levels are neglected. In a steady state situation with both transitions saturated \((\Omega_2 \gg \Gamma_2, \Omega_3 \gg \Gamma_3\), where \(\Omega_1\) is the Rabi frequency\) a dressed-atom model can be used [8]. Then it follows that for zero detuning of both lasers the maximum population of the upper level is given by

\[
\langle \pi_3 \rangle_{\text{max}} = \frac{\Gamma_2 + \Gamma_3}{(\sqrt{\Omega_2^2 + 2\Gamma_3^2})^2}.
\]  

(2)

This value is only reached when the Rabi frequencies \(\Omega_2\) and \(\Omega_3\) are related by

\[
\frac{\Omega_3}{\Omega_2} = \left(\frac{2\Gamma_3}{\Gamma_2}\right)^{1/4}.
\]  

(3)

The corresponding average population in the intermediate level then becomes

\[
\langle \pi_2 \rangle = \frac{\Gamma_3 \sqrt{(\Omega_2^2)(2\Gamma_3)}}{\Gamma_2 + 2\Gamma_3 \sqrt{\Omega_2^2/(2\Gamma_3)}}.
\]  

(4)

Substituting the data for our system we find \(\langle \pi_3 \rangle_{\text{max}} = 0.36\), \(\langle \pi_2 \rangle = 0.39\), and \(\langle \pi_1 \rangle = 0.25\). This results in a maximum increase of the radiation force by a factor of 9.9.

To calculate the force in the case of arbitrary intensities, with detunings of the lasers included, or in the case when a steady state is not achieved the optical Bloch equations (OBE's) for the three-level system have to be solved [9]. In Fig. 1 OBE calculations of the two-color average force are shown as a function of \(\Omega_3\) for different values of \(\Omega_2\). The force is normalized to the maximum Doppler-cooling force obtainable on the \(2^3S_1 \rightarrow 2^3P_2\) transition only \((F_{\text{max}} = h\kappa_2\Gamma_2/2)\). To get substantial increase of the force both transitions must be saturated strongly. The saturation parameter \(s_i\) for the excitation of level \(i\) is defined by \(s_i = I/I_{\text{sat},i} = 2\Omega_i^2/\Gamma_i^2\). The saturation intensity \(I_{\text{sat},2}\) for the infrared transition is 0.16 mW/cm\(^2\), so an intensity of 34 mW/cm\(^2\), as used in the experiment, corresponds to a saturation parameter \(s_2 = 213\) and a Rabi frequency \(\Omega_2 = 10.3\Gamma_2\); for the yellow transition \(I_{\text{sat},3}\) is 6.9 mW/cm\(^2\), so an intensity of 345 mW/cm\(^2\) corresponds to \(s_3 = 50\) and \(\Omega_3 = 5.0\Gamma_3\).

We have measured the radiation force in the two-color scheme using the setup shown in Fig. 2. A beam of metastable helium atoms was produced using a dc discharge \((-10^{12}\text{ metastable triplet atoms sr}^{-1}\text{ s}^{-1})\), maintained in an expanding helium gas through a capillary tube [10]. This beam was shaped by two 0.1 mm wide slits, separated by 15 cm. The beam was then crossed with overlapping yellow and infrared laser beams, both \(\sigma^+\) polarized, to excite the cascade. The waist of both laser beams were about 1 cm, their divergence less than 0.5 mrad, and the angle between them also less than 0.5 mrad. The laser beams were shaped by a \(4 \times 4\) mm\(^2\) aperture, about 10 cm from the interaction region, thus reducing effects of aperture diffraction. The laser power behind this aperture was measured using a calibrated power meter. The infrared light was produced by an Ar\(^{+}\)-laser-pumped Nd:LiMgAl\(_2\)O\(_4\) (lanthanum neodymium magnesium hexa-aluminate) laser (output 130 mW) which was locked to the lower He transition using a saturated absorption setup with a rf discharge cell. For the yellow light a Spectra Physics model 380D ring dye laser was used, locked to a temperature-stabilized reference étalon. The deflection of the atomic beam was monitored using a single multichannel plate and a phosphor screen. The fluorescence from the screen was read out by a camera and frame-grabber electronics. Details of the experimental setup will be published elsewhere.

A typical profile of the atomic beam, undeflected, deflected by infrared light only, and deflected by two-color excitation, is reproduced in Fig. 3. The broadening of the deflected beam is caused by the longitudinal velocity spread of the atoms, resulting in a varying transit time through the laser beams. In a separate time-of-flight experiment we have determined the center

![FIG. 1. Force enhancement for two-color excitation of metastable helium atoms as a function of the ratio of Rabi frequencies on the two transitions: (a) \(\Omega_3 = 2.5\Gamma_2\), (b) \(\Omega_3 = 5\Gamma_2\), (c) \(\Omega_3 = 10\Gamma_2\), and (d) \(\Omega_3 = 20\Gamma_2\).](image)

![FIG. 2. Experimental setup to measure the radiation pressure on the \(2^3S_1 \rightarrow 2^3P_2\) transition only (---) and using two-color excitation on the \(2^3S_1 \rightarrow 2^3P_2 \rightarrow 3^3D_3\) cascade (---).](image)
velocity to be 2250 m/s and the width of the distribution at half maximum to be 800 m/s. The width of the observed deflection profile can be fully explained from this measurement. The peak in the signal of deflected atoms corresponds to atoms with a longitudinal velocity of 2250 m/s in all laser configurations considered, allowing direct intercomparison of the measured profiles. There is a small fraction of undeflected singlet He* atoms and/or XUV photons from the source visible in the two-color deflection profile.

In the experiment the interaction length of 4 mm is sufficiently small to ensure that the Doppler shift during the deflection is much smaller than both Rabi frequencies. The deflection is therefore directly proportional to the force. Keeping the intensity of the infrared laser constant (\( \Omega_2 = 10.3 \Gamma_2 \), intensity 34 mW/cm\(^2\)), we have measured a deflection profile at different values of the intensity of the yellow laser (maximum intensity used was 345 mW/cm\(^2\)). The deflection was scaled to the deflection that corresponds to single infrared excitation at infinite saturation, calculated to be 0.47 mm. The results are plotted in Fig. 4, and compared with the model calculations. Optical pumping effects of the atom upon entering the light fields are not accounted for, but it is estimated that they have negligible influence on the deflection. At zero yellow light intensity (\( \Omega_3 = 0 \)) the force exerted by the intense infrared light agrees within 10% with the calculated maximum cooling force for the two-level case. As predicted by the model, we find a decrease of the force at high yellow intensities, emphasizing the importance of the coherences.

We have also checked the deflection at lower intensities of the infrared laser. In this regime the agreement with the model calculations is less good. This can be understood when a slow frequency drift of the yellow laser is considered as well as a greater sensitivity for small misalignments of the two overlapping laser beams. The effective detuning of the yellow laser can then no longer be neglected against the Rabi frequencies of both lasers.

In the near future we intend to decelerate metastable helium atoms using this two-color excitation scheme. Zeeman slowing [11] is not possible in a collinear geometry, since the Doppler shifts for the two transitions differ by a factor \( k_1/k_2 \). Slowing in a collinear geometry can be accomplished using chirp techniques [12]. In the simplest configuration two overlapping \( \sigma^+ \) polarized laser beams counterpropagating with the atomic beam are synchronously chirped in frequency to compensate for the changing Doppler shifts. Another promising option is to transversally excite the lower transition, using \( \pi \) polarized infrared light, and to decelerate with the \( \sigma^+ \) polarized, chirped yellow laser light. This has the added advantage (when a detuning of the infrared laser is used) that transverse heating of the atoms is counteracted. In this case the system consists of more than three levels, and an extended set of optical Bloch equations, including all the magnetic sublevels, should be used. A minimum stopping distance as small as 11 cm for atoms with an initial velocity of 1000 m/s (obtainable by cooling the helium discharge with liquid nitrogen) is calculated. Presently we are constructing a compact atomic beam apparatus for the trapping of metastable helium atoms using this two-color approach.

We thank Olivier Emile for many stimulating discussions and Jacques Bouma for his invaluable help with the beam apparatus. This work was funded by the Dutch Organization for Fundamental Research of Matter (FOM).

---


