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# Laser Cooling and Storage of Free Atoms\*

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Received February 2, 1987; accepted February 17, 1987

#### Abstract

Atomic beam cooling by counterpropagating laser light is shown to be a versatile tool in achieving low mean beam velocities and mK "beam temperatures". This cold beam or gas may be further cooled or manipulated by laser light for the needs of deflection, storage and density increase used for high precision spectroscopic experiments or other interesting future possibilities, which will be mentioned.

### 1. Introduction

Many high resolution spectroscopic experiments are ultimately limited by the random motion of atoms or velocity related effects. This motion causes, e.g., transit time effects or second order Doppler shifts and broadening of spectral lines, which are not canceled by most of the so called Doppler free spectroscopic techniques. Also atomic collision experiments suffer often from the broad velocity distribution and the angle spread of the collision partners.

A thorough solution of these general problems would be a direct velocity manipulation of free atoms aiming at a reduced average velocity and a strong reduction of the temperature respectively the width of the resulting velocity distribution.

This velocity modification can be achieved by the light pressure force from a resonant laser beam. The discussion of such radiation pressure forces has a long history [1]. But it remained nearly exclusively theoretical till the introduction of tuneable dye lasers in the atomic spectroscopy.

In the basic scheme of the light pressure force atoms absorb photons and their momentum  $\hbar k$  (k = wavevector of the photon) from a light beam. After a short time – typically 10 ns – the atoms reemit spontaneously a fluorescence photon. Because of the random direction of emission the momentum recoil from the emitted photons averages to zero whereas the atomic momentum change by the absorption of the photon momentum adds up constructively.

Therefore the velocity change of the atoms is  $n\hbar k/M$  (M = mass of the atom, n = number of absorption) on an average.

The resulting averaged "spontaneous" force f (atomic momentum change per time) reads:

$$f = hk \frac{1}{\tau} \frac{s}{1+2s}, \qquad (1)$$

where  $\tau$  is the natural lifetime of the "upper" cooling level and s is the saturation parameter.

The expression  $\tau(2 + 1/s)$  is the cycle-time, which depends

on the laser intensity and the detuning between the laser frequency and the Doppler shifted absorption frequency of the atoms. This force saturates with increasing saturation to  $f_{sat}$ :

$$f_{\rm sat} = \hbar k \, \frac{1}{2\tau}. \tag{2}$$

The corresponding decelerations a and  $a_{sat}$  read

$$a = \frac{\hbar k}{M} \frac{1}{\tau} \frac{s}{1+2s}$$
(3)  
$$a_{sat} = \frac{\hbar k}{M} \frac{1}{2\tau}$$

## 2. Atomic beam cooling

To avoid the problems, which existed in the first suggestion of atomic cooling in a gas by Hänsch and Schawlow [2], the theorists and experimentalists thought about atomic beam cooling. In an atomic beam almost only one degree of freedom – the longitudinal velocity distribution – has to be cooled and the atoms move in a high vacuum environment collision free without heating by walls or other molecules.

The basic scheme of atomic beam cooling consists of an atomic beam and a counter-propagating laser beam. This laser beam is tuned into resonance with a Doppler-shifted, i.e., high velocity, group in the atomic beam. Thus the atoms absorb momentum and are decelerated. For example, a sodium atom loses on an average 3 cm/s speed per absorption, when the laser is tuned to the sodium  $D_2$  line. This means, that a typical sodium atom with an average velocity of 600 m/s needs about 20 000 absorptions to be stopped. As the cycle-time has the order of magnitude of the lifetime  $\tau - \approx 16 \text{ ns} - \text{the time scale for complete deceleration is about 1 ms and the stopping distance is less than 1 m.$ 

The fundamental problems arising in such experiments are twofold:

(1) As atoms slow down, the changing Doppler shift moves them out of resonance with the laserfield and thus limits the cooling to a few homogeneous linewidths ( $\leq 100 \text{ m/s}$ ).

(2) The necessarily big number of absorption-emissioncycles ( $\approx 20\,000$  for sodium) demands a very pure two-level system for the cooling transition or counter-measures to avoid losses due to optical pumping.

Figure 1 shows the relevant energy levels of <sup>23</sup>Na, which is mostly used in atomic beam cooling experiments until now.

Early work on atom slowing by Letokhov and co-workers [3] showed interesting experimental curves, but the role of optical-pumping effects was not clarified.

<sup>\*</sup> This paper was originally presented at the 18th EGAS Conference, held in Marburg, 8–11 July, 1986, and will be reprinted in the proceedings of that conference (editors: M. Elbel and H. Hühnerman).

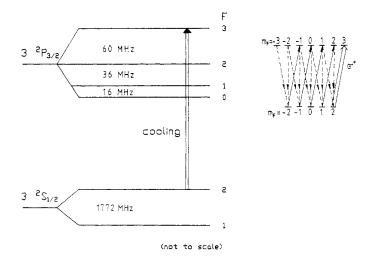


Fig. 1. Level scheme of Na with cooling transition.

In recent experiments [4, 5] two different experimental schemes were used to solve the basic problems in atomic beam-cooling experiments:

In the first successful cooling experiments, which produced really slow atoms in a sodium atomic beam [6], a longitudinal magnetic field with a decreasing field strength along the beam axis solved both problems. Using circularly polarized sodium  $D_2$  light the atoms are relatively fast optically pumped into the magnetic sub-level  ${}^{2}S_{1/2}$  (F = 2,  $m_{F}$  = 2) of the ground state, when they enter the magnetic field. This state is only excitable to the  ${}^{2}P_{3/2}$  ( $F = 3, m_{F} = 3$ ) level by the circularlypolarized cooling laser beam, forming a nearly ideal two-level system. When the magnetic field strength (e.g., a bias field) is strong enough, it can compensate for imperfect polarization of the light field and avoid resulting optical pumping. When the laser frequency is tuned into resonance with fast Dopplershifted and Zeeman-shifted atoms at the entrance of the magnetic field, they will stay in resonance during the slowing process in the magnet if the magnetic field strength decreases in a way, that the changing Zeeman shift compensates for the changing Doppler shift.

Assuming a constant deceleration a within the magnet the velocity v(z) changes like:

$$v(z) = (v_0^2 - 2az)^{1/2}$$
(4)

 $v_0$ : initial velocity entering the magnetic field

The corresponding change of the magnetic field strength B(z) to compensate the z-dependent Doppler shift by the changing Zeeman shift of the cooling transition reads then (for details see, e.g. [4, 6]):

$$\boldsymbol{B}(z) = \boldsymbol{B}_0 (1 - 2az/v_0^2)^{1/2} + \boldsymbol{B}_b$$
 (5)

where  $B_{\rm b}$  is the bias field.

The difference between the magnetic field strength at both ends of the field defines the magnitude of the velocity interval which will be slowed down and the laser detuning defines the final velocity of the decelerated atoms. Atoms outside this velocity interval – faster atoms and atoms slower than the final velocity – will not be affected (almost). As each location z within the magnet corresponds, as given in eq. (4), to a resonant velocity, every atom within the affected velocity interval will find its resonant point z and stay in resonance till the exit of the magnet. All atoms of the decelerated interval will thus have nearly the same velocity at the exit of the magnet. Therefore the resulting velocity distribution is highly compressed to some m/s (see below). The final velocity may be chosen over a broad range including zero. In order to get the atoms at rest outside the magnet, the laser frequency is tuned into resonance with a sufficiently slow velocity group at the exit of the magnet, allowing the atoms to slow down further, when they walk out of resonance [4].

In first experiments using this technique, the production of a steady flow of cooled sodium atoms with a temperature below 100 mK and a density of about 10<sup>5</sup> cm<sup>-3</sup> was demonstrated (for details see, e.g., Ref. [4]).

The second general cooling scheme uses fast frequency modulation techniques (without magnetic fields) to compensate optical pumping and to keep the resonance condition [6].

In alkali spectra (e.g., Na) the transition  ${}^{2}S_{1/2}$  (F = 2)– ${}^{2}P_{3/2}$  (F = 3) does not provide a completely ideal two-level system in zero magnetic field; because of the relatively small hyperfine splitting of the upper level and the limited perfection of the circular polarization of the laser light, the atoms can also make the transition  ${}^{2}S_{1/2}$  (F = 2)– ${}^{2}P_{3/2}$  (F = 2) and the upper level can decay to the level  ${}^{2}S_{1/2}$  (F = 1) which is out of resonance. To compensate for this optical pumping a second frequency in the cooling laser beam for the transition  ${}^{2}S_{1/2}$  (F = 1)– ${}^{2}P_{3/2}$  (F = 2) can repump the atoms into the level  ${}^{2}S_{1/2}$  (F = 2) via the transition  ${}^{2}P_{3/2}$  (F = 2)– ${}^{2}S_{1/2}$  (F = 2).

This second frequency can be provided as one of two sidebands of a frequency-modulated laser beam; in the case of the sodium  $D_2$  line the difference frequency would be 1712 MHz. For this purpose the laser beam is, for example, sent through an electro-optic phase modulator [5] that is driven at half the desired difference frequency. In case of a sufficient modulation index about 35% of the incoming intensity can be transferred to the first-order sideband.

The problem of maintaining the resonance condition for the decelerating atoms is solved in this scheme by fast tuning of the laser frequency synchronously with the rapidly changing Doppler shift. This again can be achieved by electro-optic modulation techniques, if the laser beam with the two frequencies is sent through a second electro-optic modulator, the driving frequency of which is chirped in the right way – in the sodium experiment, for example, from 5 MHz to 1000 MHz within about 1.5 ms. This will produce a pair of sidebands which stay in resonance with the decelerating atoms, if the carrier frequency is chosen correctly [5].

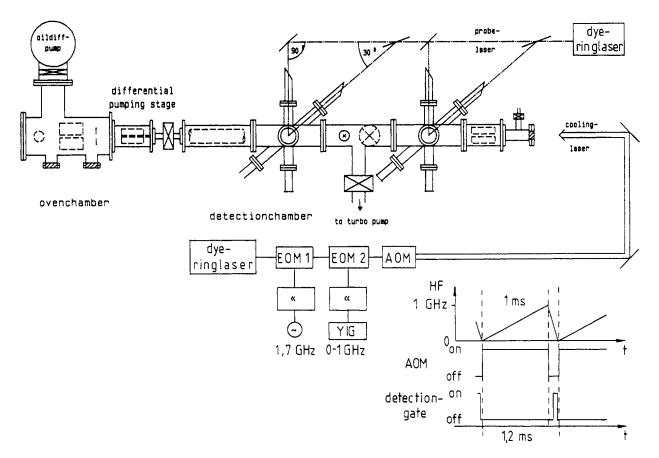
Figure 2 shows the experimental scheme for this experiment, the sideband spectrum is schematically shown in Fig. 3. As the cooling sideband and the repumping sideband differ only in 1.7 GHz both frequencies stay in resonance with the same velocity group as both frequencies are swept in the same way by the second modulator. In the scan method the deceleration a must match the scan speed  $\dot{v}_L$  of the laser frequency; assuming constant deceleration the scan speed is almost constant,

$$\dot{v}_{\rm L} = \frac{|a|}{\lambda} \tag{6}$$

and the frequency varies linearly in time, starting red shifted at the laser frequency  $v_s$ .

$$v_{\rm L}(t) = v_{\rm s}(1 + \alpha t).$$
 (7)

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*Fig. 2.* Schematic of atomic beam cooling experiment. The cooling laser output is fed through an acousto-optic shutter, a LiTaO<sub>3</sub> travelling-wave electro-optic modulator (EOM) which puts on the frequency-swept sidebands, and an additional EOM to provide the F = 1 atom recovery

sideband. The  $\sigma$ -polarized cooling laser beam is carefully "mode matched" to the weakly diverging atomic beam (~3 mrad full angle). The detection system for the fluorescence light is not shown.

This frequency  $v_s$  is in resonance with fast atoms  $v_s$  at the beginning of each cooling cycle. When the sideband is swept over a frequency interval  $\Delta v_L$  these fast atoms stay in resonance during the slowing process and slower atoms will get into resonance. Thus at the end of a cooling cycle the corresponding velocity interval

$$\Delta v = \Delta v_{\rm L} / \lambda \tag{8}$$

is compressed into a narrow velocity distribution (see below) at the final velocity  $v_s - \Delta v$ . In our experiments the scan interval was limited by our microwave equipment to ~1 GHz (see Fig. 4) what corresponds to a velocity interval of ~600 m/s. After each cooling scan, which takes about 1 ms, the swept sideband moves very fast within ~200  $\mu$ s back into

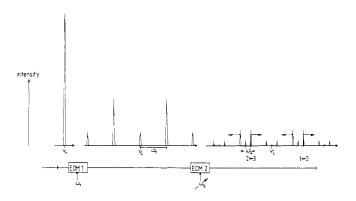


Fig. 3. Schematic frequency spectrum of the cooling laser beam behind the two electro-optic modulators (EOM). The cooling and repumping sidebands are marked.

the starting position beginning the next cooling cycle. During this time fresh incoming, uncooled atoms fly only  $\sim 1 \text{ m}$ before they get into resonance in the next cooling cycle. Thus all atoms slower than the starting velocity  $v_s$  will be cooled. As such cooling cycles are repeated periodically, the result will be a steady flow of cooled atoms similar to the previous scheme.

In the first experiments with this second scheme [5, 7] the resulting temperature within the cooled atomic beam was below 50 mK with a density of  $10^6$  atoms per cm<sup>3</sup>. As the cooled slow atoms move only a short distance during the cooling cycles, the resulting pile-up of slow atoms forms a nearly constant flow of cold atoms as in the previous scheme.

For probing the velocity distribution we used a second dye laser beam, which was split into a Doppler free beam – perpendicular to the atomic beam and a beam crossing the atomic beam with a small angle ( $\approx 30^{\circ}$ , see Fig. 4). The frequency of this probing laser was slowly tuned over the Doppler profile of the  ${}^{2}S_{1/2}$  (F = 2)– ${}^{2}P_{3/2}$  (F = 3) transition. The laser induced fluorescence signal was only periodically counted for a  $\sim 50 \,\mu$ s time interval during the flyback time of the cooling sideband. During this time the cooling laser beam was shut off by an acusto-optic modulator. Thus optical pumping and transient effects were avoided.

Because of the  $30^{\circ}$  crossing angle our velocity readout scale is compressed by a factor  $\cos 30^{\circ} = 0.866$ . Figure 4 shows our result. Because of the saturation in the probe beam being 2–3 and the Doppler free beam being  $\ll 1$  the whole width of the probe beam can be explained — in comparison to the width of the Doppler free width — by saturation effects

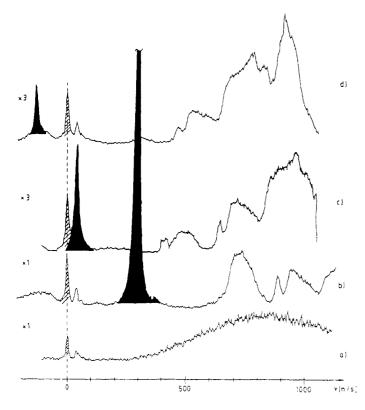


Fig. 4. Sodium-atomic-beam cooling using a frequency-chirped laser. Trace (a), cooling laser off. The  $D_2$  transition shows the velocity distribution of the F = 2 ground state as well as the frequency markers from the perpendicular probe beam. The vertical dashed line marks the position of the F = 3 resonance with zero velocity atoms. Traces (b)–(d), cooling laser sideband is swept  $\approx 1$  GHz (590 m/s), carrying atoms to lower velocities where they are left (blackened peaks) when the cooling laser is cut off and the velocity distribution is measured. The final velocity in trace (b) is  $\approx 290$  m/s. In trace (c)  $\approx 40$  m/s, and in trace (d)  $\approx -130$  m/s (!). The apparent weakening of the slow-atom peak is partly because fewer atoms are available, when the sweep starts below the velocity distribution maximum and for geometrical reasons. The figures also show "actions" by the other (unused) sidebands.

(within the momentary velocity resolution). As 10 MHz natural width corresponds to a Doppler broadening by 6 m/s, we can estimate a residual velocity width of less than 5 m/s (for the more complicated details see Refs. [6, 8]). This result is in good agreement with our Monte-Carlo-Simulation of the cooling process [9, 10] and the numerical integration of the Fokker-Planck-Equation [10].

## 3. Properties and handling of cooled atomic beams

As result of the numerous spontaneous emissions, some longitudinal momentum is transferred into transverse momentum. The resulting velocity width  $v_{\rm T}$  (rms) of this "transverse heating effect" by *n* spontaneous emissions reads

$$v_{\rm T}(\rm rms) = v_{\rm r}(n/3)^{1/2}$$
 (9)

$$v_{\rm r} = \hbar k/M \tag{10}$$

with

$$n = \frac{1}{v_{\rm r}} (2k_{\rm B}T/M)^{1.2}$$

where  $k_{\rm B}$  is the Boltzmann-constant, T is the evaporation temperature.

This transverse velocity spread becomes

$$v_{\rm T}(\rm rms) = \left(\frac{2k_{\rm B}T\hbar^2k^2}{9M^3}\right)^{1/4}.$$
 (11)

As the initial transverse beam velocity is generally reduced also by the converging cooling laser beam, the final transverse beam velocity corresponds to an initial collimation ratio  $\beta$  of

$$\beta = \left(\frac{\hbar^2 k^2}{18k_{\rm B}TM}\right)^{1/4}.$$
 (12)

The ratio  $\beta \approx 4 \times 10^{-3}$  for sodium and  $\beta \approx 2 \times 10^{-3}$  for caesium means, that the transverse heating generally compensates the transverse velocity reduction by the cooling beam.

For the further discussion it will be of special interest how to separate the cold atoms from the hot (fast) atoms, how to reduce the residual longitudinal and transverse velocity spread and how to increase the atomic-beam density.

One way of separation is simply to apply a second "twofrequency" transverse laser beam as shown in Fig. 5. If the intensity profile of this laser beam has the correct shape, only the slow atoms will be deflected and these atoms will show no further increase in velocity spread. The transverse laser beam providing the necessary spontaneous force to match the radial acceleration  $a_r = v_0^2/r$  ( $v_0$  is the average velocity of the slow atomic beam) has to have a saturation profile like

$$s(r) = \frac{1}{\frac{rv_{\rm r}}{\tau v_0^2} - 2}$$
(13)

or for  $s \ll 1$ 

9

$$s(r) = \frac{\tau v_0^2}{v_r} \frac{1}{r}.$$
 (14)

This means that a cylindrical lens gives the proper intensity profile for the purpose of bending an atomic beam of slow atoms. For a beam with 50 m/s average velocity and 5 mm diameter a lens with a focal length of 50 mm and a transverse laser beam of 20 mm waist only a few milliwatts are sufficient to bend the slow atomic beam  $\approx 40^{\circ}$  from the axis [8]. Additionally, the transverse velocity spread will be reduced in the bending plane as a by-product of the bending mechanism. Atoms which are less deflected than the average move into stronger saturation and are consequently more strongly deviated. The result of this bending scheme is thus a slow atomic beam, the direction of which is unconstrainedly selectable and which is unperturbed by fast atoms.

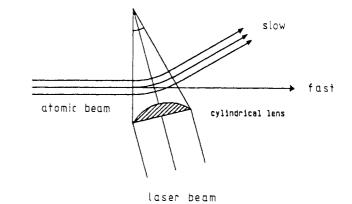


Fig. 5. Geometry for atomic beam deflection by a transverse laser beam.

The transverse velocity distribution of this beam may be further reduced by transverse laser beams, the frequency of which is tuned about one halfwidth to the red of the centre frequency. Thus the saturation of, and correspondingly the spontaneous force on, the slow atoms by the transverse laser beams increases for the atoms which are moving towards a transverse laser beam [11, 16]. The effect will be maximum when the red detuning  $\delta \omega$  is [8]

$$\delta\omega = \frac{\gamma}{2} \left( \frac{1+2s_0}{3} \right)^{1/2} \tag{15}$$

where  $\gamma$  is the natural linewidth (FWHM) and  $s_0$  is the saturation on centre frequency.

A similar velocity-dependent force will also reduce the longitudinal velocity distribution [5]: a counter-propagating laser beam, which is tuned slightly red of the absorption frequency of the slow atoms, will decelerate atoms, which remained faster than the average, more than slower ones, when they move out of resonance. This laser beam could be, for example, the carrier frequency of the cooling laser, if the chirping method is used for cooling. This frequency may be permanent in the cooling beam. This velocity-controlling effect is also responsible for the longitudinal velocity width just after cooling [5].

There are two principle ways to increase the density of cold atomic beams [8]:

(1) transverse laser beams with strong radial-positiondependent intensity distribution and with additional transverse cooling beams.

(2) the "dipole force" [1] in a strongly converging, redshifted longitudinal laser beam with additional transverse cooling beams.

In the first case, the position-dependent, ponderomotive forces will transfer momentum to the atoms, which do not move on axis, towards the axis. This can be realized, for example, by a rotationally-symmetric, nearly-parabolic mirror [8].

In the second case, for example, a parabolic mirror with the focus on axis could produce a strong focus on the atomic beam axis. If the frequency of this laser beam is red-shifted, the atoms will be accelerated in the direction of the intensity gradient by the so-called "dipole or gradient force" [1], i.e., towards the atomic axis. In both cases, the transverse cooling beams will damp the transverse motion. (It should be mentioned that for both position-dependent beams the Doppler shift is also position dependent.) The result will be a very dense beam in the converging region with diameters down to the  $\mu$ m-range. Behind this region, the width of the beam will increase depending on its transverse and longitudinal velocities. If a further confinement is desirable, one could use a collinear, so-called "doughnut-mode" laser beam. If this laser beam is blue-shifted, the atoms walk into the field-free region, that means again towards the atomicbeam axis (further details will be published elsewhere [8]).

The next question is: what are the cooling limits?

There are two limits (see, e.g., Ref. [12]):

(1) the motion  $v_r$  [eq. (10)] because of the photon recoil (2) the bad velocity resolution caused by the natural spectral width  $\delta v_n$  of the transition.

The laser effect results in a rms velocity  $v_{\rm rms}$  of

$$v_{\rm rms} = (2h\delta v_{\rm n}/M)^{1/2}.$$
 (16)

The corresponding temperatures are in the first case

$$T_{\rm r} = \frac{\hbar^2 k^2}{3k_{\rm B}M} \tag{17}$$

and in the second case

$$T_{\rm rms} = \frac{h\delta v_{\rm n}}{2k_{\rm B}}.$$
 (18)

Table I gives the temperatures and velocities for some interesting elements. It shows that for the cooling transitions  $v_{\rm rms}$ is generally the lower velocity limit. To reach lower velocities, one has to take, in a second cooling step, a "slower" transition. For transitions with a lifetime  $\tau$  of the upper level of

$$\tau \approx \frac{\lambda^2 M}{4h} \tag{19}$$

the two limits are nearly equal. It should be mentioned here that temperature in this context only means the relative motion of the atoms and not the average velocity in the laboratory frame.

Summarizing this section, one can say that slow and cold atomic beams can be produced with temperatures in the  $\mu$ K-range and corresponding relative velocities of some cm/s. The average velocity can be chosen over a wide range from negative up to very high velocities.

The slow atomic beams can be separated from the fast beam parts and they can be directed or bent quite simply with auxiliary laser beams.

## 4. Applications of cold atomic beams

One application of cold atomic beams is clearly its use for optical or microwave frequency standards.

Table I. Cooling limits for some elements which can be cooled by laser cooling;  $\lambda$  denotes the cooling wavelength and  $\tau$  denotes the natural lifetime of the upper level of the cooling transition; for  $v_r$  and  $v_{rms}$  see text,  $T_r$  and  $T_{rms}$  are the corresponding "temperatures"

Element	λ [nm]	$\tau$ [ns]	$v_{\rm r}  [{\rm cm/s}]$	$v_{ m rms}~[ m cm/s]$	$T_r$ [ $\mu$ K]	$T_{\rm rms}$ [ $\mu { m K}$ ]
Sodium	589	16	3	59	0.8	239
Rubidium	780	27	0.6	24	0.1	141
Caesium	852	32	0.35	17	0.07	119
Calcium	423	4.6	2.3	83	0.9	830
	657	$4 \times 10^5$	1.5	2.9	0.4	0.01
Magnesium	285	2	5.8	162	3.3	1910
	457	$2.3 \times 10^{6}$	3.6	0.15	1.3	0.002

The great interest in cold atoms for optical frequency standards is twofold: reduction of Doppler effects and prolonged interaction times. Whereas the first-order Doppler effect is still of the order of some hundreds of kHz to 1 MHz for optical transitions, the second-order Doppler effect is less than 10 mHz (for cold atoms). The interaction time of the light field with cold atoms may well exceed the interval of 1 s, offering spectral widths in the sub-Hz regime for selected transitions, Doppler-free schemes presupposed.

There are two main concepts for optical frequency standards using cold atomic beams. The first one uses the cold atomic beam mainly for filling atom-traps, which form the essential frequency discriminator of an optical frequency standard. The second one makes full use of the monochromatic velocity distribution of cold atomic beams.

The different concepts for atom traps are described, for example, in Refs. [12, 13]. Two of these concepts apply laser beams as trap walls, which cool the atoms within the traps in a kind of "optical molasses" [7] and form the trapping potential by the "dipole force" [13, 14]. It should be mentioned that atomic beam cooling is essential for atom traps, as all traps proposed are very shallow with well depths below one kelvin. Recently, the first observation of electromagnetically-confined neutral atoms was also reported [15], with trap times of about 1 s, limited by the background gas scattering.

The temperature, or low-velocity, limits which can be reached in those trapping schemes are generally the same as given above. In order to avoid disturbing interactions between the trapping mechanism and the clock transition, one has to shut down the trapping fields for the time the clock is running. When the clock is run with trapped atoms, an alternating periodic switching between cooling, trapping, and measuring is necessary, while the shutdown phase of the trapping fields should not exceed 1 ms, as discussed in Ref. [13]. This leads to spectral widths of the optical transition in the kHz range.

Whereas those transitions still have an attractive Q-value of about  $10^{12}$ , this restriction may be avoided when the clock transition takes place outside the trap: for this purpose the atoms are, for instance, collected and ultimately cooled within an atom trap, which they leave pushed by an auxiliary laser beam or, for example, an unbalanced laser beam of the optical trap. This should offer interaction times of 1 s or more, giving Q-values of about  $10^{15}$  or even larger. Interesting candidate atoms are given, for example, in Ref. [16].

The monochromatic atomic beam may also be deflected through 90° with a dipole magnet or a transverse laserbeam (see above). The resulting "Zacharias fountain" atomic beam would give an ideal opportunity for two-zone Ramsey excitation with very slow atoms going a second time through the same interaction region, when they are free falling down again [5, 17].

Other applications for cold atomic beams are clearly collision physics, surface physics, photon statistics, quantum effects (Bose condensation), polarized Targets or isotope separation.

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