Continuous-wave Doppler-cooling of hydrogen atoms with
two-photon transitions

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Abstract

We propose and analyze the possibility of performing two-photon continuous-
wave Doppler-cooling of hydrogen atoms using the $1S - 2S$ transition.
“Quenching” of the $2S$ level (by coupling with the $2P$ state) is used to increase
the cycling frequency, and to control the equilibrium temperature. Theoretical
and numerical studies of the heating effect due to Doppler-free two-photon
transitions evidence an increase of the temperature by a factor of two. The
equilibrium temperature decreases with the effective (quenching dependent)
width of the excited state and can thus be adjusted up to values close to the
recoil temperature.
Laser cooling of neutral atoms has been a most active research field for many years now, producing a great deal of new physics. Still, the hydrogen atom, whose “simple” structure has lead to fundamental steps in the understanding of quantum mechanics, has not yet been laser-cooled. The recent experimental demonstration of the Bose-Einstein condensation of H adds even more interest on laser cooling of hydrogen [1]. One of the main difficulties encountered in doing so is that all transitions starting from the ground state of H fall in the vacuum ultraviolet (VUV) range (121 nm for the $1S - 2P$ transition), a spectral domain where coherent radiation is difficult to generate. In 1993, M. Allegrini and E. Arimondo have suggested the laser cooling of hydrogen by two-photon $\pi$ pulses on the $1S - 3S$ transition (wavelength of 200 nm for two-photon transitions) [2]. Since then, methods for generation of CW, VUV, laser radiation have considerably improved, and have been extensively used in metrological experiments [3]. This technical progress allows one to realistically envisage the two-photon Doppler cooling (TPDC) of hydrogen in the continuous wave regime, in particular for the $1S - 2S$ two-photon transition.

Laser cooling relies on the ability of the atom to perform a great number of fluorescence cycles in which momentum is exchanged with the radiation field. It is well known that $2S$ is a long-lived metastable state, with a lifetime approaching one second. From this point of view, the $1S - 2S$ two-photon transition is not suitable for cooling. On the other hand, the minimum temperature achieved via Doppler cooling is proportional to the linewidth of the excited level involved on the process [4], a result that will be shown to be also valid for TPDC. From this point of view $2S$ is an interesting state.

In order to conciliate these antagonistic properties of the $1S - 2P$ transition, we consider in the present work the possibility of using the “quenching” of the $2S$ state to control the cycling frequency of the TPDC process. For the sake of simplicity, we work with a one-dimensional model. We write rate equations describing TPDC on the $1S - 2S$ transition in presence of quenching. The quenching ratio is considered as a free parameter, allowing control of the equilibrium temperature. The cooling method is then in principle limited only by photon recoil effects.
We also develop analytical approaches to the problem. A Fokker-Planck equation is derived, describing the dynamics of the process for temperatures well above the recoil temperature $T_r$ (corresponding to the kinetic energy acquired by an atom in emitting a photon). A numerical analysis of the dynamics of the cooling process completes our study.

Let us consider a hydrogen atom of mass $M$ and velocity $v$ parallel to the $z$-axis (Fig. 1) interacting with two counterpropagating waves of angular frequency $\omega_L$ with $2\omega_L = \omega_0 + \delta$, where $\omega_0/2\pi = 2.5 \times 10^{14}$ Hz is the frequency corresponding to the transition $1S \rightarrow 2S$, and also define the quantity $k \approx 2k_L = 2\omega_L/c$. The shift of velocity corresponding to the absorption of two-photons in the same laser wave is $\Delta = \hbar k/M = 3.1$ m/s. We will neglect the frequency separation between $2S$ and $2P$ states (the Lamb shift – which is of order of 1.04 GHz) and consider that the one-photon spontaneous desexcitation from the $2P$ states also shifts the atomic velocity of $\Delta$ randomly in the $+z$ or $-z$ direction. Note that $T_r = M\Delta^2/k_B \approx 1.2$ mK for the considered transition ($k_B$ is the Boltzmann constant). We neglect the photo-ionization process connecting the excited states to the continuum. This is justified by the $1/E$ decreasing of the continuum density of states as a function of their energy $E$ and by the fact that a monochromatic laser couples the excited levels only to a very small range of continuum levels.

The atom is subjected to a controllable quenching process that couples the $2S$ state to the $2P$ state (linewidth $\Gamma_{2P} = 6.3 \times 10^8$ s$^{-1}$). The adjustable quenching rate is $\Gamma_q$. Four two-photon absorption process are allowed: $i$) absorption of two photons from the $+z$-propagating wave (named wave “+” in what follows), with a rate $\Gamma_1$ and corresponding to the a total atomic velocity shift of $+\Delta$; $ii$) absorption of two photons from the $-z$-propagating wave (wave “−”), with a rate $\Gamma_{-1}$ and atomic velocity shift of $-\Delta$; $iii$) the absorption of a photon in the wave “+” followed by the absorption of a photon in the wave “−”, with no velocity shift and $iv$) the absorption of a photon in the wave “−” followed by the absorption of a photon in the wave “+”, with no velocity shift. The two latter process are indistinguishable, and the only relevant transition rate is that obtained by squaring the sum of the amplitudes of these process (called $\Gamma_0$). Also, these process are “Doppler-free”
(DF) as they are insensitive to the atomic velocity (to the first order in $v/c$) and do not shift the atomic velocity. Thus, they cannot contribute to the cooling process. As atoms excited by the DF process must eventually spontaneously decay to the ground state, this process heats the atoms. In the limit of low velocities, the transition amplitude for each of the four processes is the same. One thus expects the DF transitions to increase the equilibrium temperature by a factor of two.

We can easily account for the presence of the quenching by introducing an effective linewidth of the excited level (which, due to the quenching process, is a mixing of the $2S$ and $2P$ levels) given by

$$\Gamma_e = \Gamma_{2P} \frac{\Gamma_q}{\Gamma_q + \Gamma_{2P}} = g \Gamma_{2P} \quad (1)$$

with $g \equiv \Gamma_q / (\Gamma_q + \Gamma_{2P})$. This approximation is true as far as the quenching ratio is much greater than the width of the $2S$ state (note that this range is very large, as the width of the $2S$ state is about $10^{-8}$ times that of the $2P$ state).

The two-photon transition rates [6] are given by:

$$\Gamma_n = \Gamma_{2P} \frac{g}{2} \frac{(1 + 3\delta_n)I_0^2}{(\delta - nKV)^2 + g^2/4} \quad (2)$$

where $n = \{-1, 0, 1\}$ describes, respectively, the absorption from the “−” wave, DF transitions, and the absorption from the “+” wave. $\bar{I} \equiv I/I_s$ where $I_s$ is the two-photon saturation intensity, $\bar{\delta}$ is the two-photon detuning divided by $\Gamma_{2P}$, $K \equiv k\Delta/ \Gamma_{2P} = 0.26$ and $V \equiv v/\Delta$.

The rate equations describing the evolution of the velocity distribution $n(V, t)$ and $n^*(V, t)$ for, respectively, atoms in the ground and in the excited level are thus

$$\frac{\partial n(V, t)}{\partial t} = - [\Gamma_{-1}(V) + \Gamma_0 + \Gamma_1(V)] n(V, t) + \frac{\Gamma_e}{2} [n^*(V - 1) + n^*(V + 1)] \quad (3a)$$

$$\frac{\partial n^*(V, t)}{\partial t} = \Gamma_{-1}(V - 1)n(V - 1, t) + \Gamma_0 n(V, t) + \Gamma_1(V + 1)n(V + 1, t) - \Gamma_e n^*(V, t). \quad (3b)$$

The deduction of the above equations is quite straightforward (cf. Fig [4]). The first term in the right-hand side of Eq. (3a) describes the depopulation of the ground-state velocity
class $V$ by two-photon transitions, whereas the second term describes the repopulation of the same velocity class by spontaneous decay from the excited level. In the same way, the three first terms in the right-hand side of Eq. (3b) describe the repopulation of the excited state velocity class $V$ by two-photon transition, and the last term the depopulation of this velocity class by spontaneous transitions. For each term, we took into account the velocity shift ($V \rightarrow V \pm 1$) associated with each transition and supposed that spontaneous emission is symmetric under spatial inversion.

For moderate laser intensities, one can adiabatically eliminate the population of excited level. This is valid far from the saturation of the two-photon transitions and reduces the Eqs. (3a-3b) to one equation describing the evolution of the ground-state population:

$$\frac{dn(V, t)}{dt} = - \left[ \frac{\Gamma_0 + \Gamma_{-1}(V)}{2} + \frac{\Gamma_1(V)}{2} \right] n(V, t) + \frac{1}{2} \left\{ \Gamma_0 [n(V - 1, t) + n(V + 1, t)] + \Gamma_{-1}(V - 2)n(V - 2, t) + \Gamma_1(V + 2)n(V + 2, t) \right\}$$

(4)

Eq. (4) is in fact a set of linear ordinary differential equations coupling the populations of velocity classes separated by an integer: $V, V \pm 1, V \pm 2, \cdots$. This discretization exists only in the 1-D approach considered here, but it does not significantly affect the conclusions of our study, while greatly simplifying the numerical approach.

Eqs. (4) can be recast as $dn/dt = Cn$, where $C$ is a square matrix and $n$ is the vector $(\cdots n(-i, t), \cdots n(0, t), n(1, t), \cdots)$. Numerically, the equilibrium distribution is obtained in a simple way as the eigenvector $n_{eq}$ of $C$ with zero eigenvalue. In this way, the asymptotic temperature is obtained as:

$$\frac{T}{T_r} = \frac{\langle V^2 \rangle}{\sum_{i=-\infty}^{\infty} i^2 n_{eq}(i)}$$

(5)

Fig. 2 shows the equilibrium distribution obtained by numerical simulation for $\delta = -0.25$ and $g = 1/3$. The dotted curve corresponds to the distribution obtained by artificially suppressing DF transitions (i.e., by setting $\Gamma_0 = 0$). As we pointed out earlier, the DF transitions lead to a heating effect. Doppler cooling is efficient mainly for atoms distributed
on a range of $g/K$ around the velocity $V = \pm |\bar{\delta}|/K$ whereas Doppler-free transitions are independent of the velocity; all velocity classes are thus affected by the heating. As a consequence, DF transitions induce a deformation of the velocity profile, specially for small values of $g$ and $\bar{\delta}$, superimposing a sharp peak of cold atoms on a wide background of “hot” atoms. In what follows, all numerically calculated-temperatures are deduced form the width of the thin peak of cold atoms.

Eqs. (3a) and (3b) or Eq. (4) have no exact solution. However, using some reasonable hypothesis, it is possible to develop analytical approaches. The most usual of these approaches is to derive from the above equations a Fokker-Planck equation (FPE) describing the evolution of the velocity distribution. The derivation of the FPE for two-photon cooling follows the standard lines that can be found in the literature (see [7]). If $|V| \gg 1$ the coefficients in the resulting equation can be expanded up to second order in $1/|V|$ (this is the so-called hypothesis of small jumps). Moreover, if $K|V| \ll |\bar{\delta}|, g$ the resulting expression can be expanded up to the order $V$. The resulting FPE reads

$$\frac{\partial n}{\partial t} = 2\bar{\Gamma} \frac{\partial (Vn)}{\partial V} + \left(2\bar{\Gamma} + \frac{\Gamma_0}{2}\right) \frac{\partial^2 n}{\partial V^2}$$

where $\bar{\Gamma} \equiv \Gamma_\infty(0) = \Gamma_0/4$ and $\bar{\Gamma}'$ is the $V$-derivative of $\Gamma_\infty$ evaluated at $V = 0$. Multiplying this equation by $V^2$ and integrating over $V$ one easily obtains:

$$\frac{d\langle V^2 \rangle}{dt} = -4\bar{\Gamma}'\langle V^2 \rangle + \left(4\bar{\Gamma} + \Gamma_0\right)$$

As $\langle V^2 \rangle = T/T_r$, this equation shows that the characteristic relaxation time is $(4\bar{\Gamma}')^{-1} = (g\Gamma_2\bar{\delta}^2\bar{\Gamma}K)/(4\bar{\delta}^2 + g^2)$.

The equilibrium temperature is then given by

$$\frac{T}{T_r} = \frac{2\bar{\Gamma} + \Gamma_0/2}{2\bar{\Gamma}'} = \frac{\bar{\delta}^2 + g^2/4}{K|\bar{\delta}|}$$

This results confirms that the Doppler-free two-photon transitions, corresponding to the contribution $\Gamma_0/2 = 2\bar{\Gamma}$ in Eq. (8) increase the equilibrium temperature (at least in the range of validity of the FPE) by a factor 2. This fact can also be verified from the numerical
simulations, as shown in Fig. 3, where the dotted curve corresponds to the temperature obtained without DF transitions. As in one-photon Doppler-cooling, the equilibrium temperature is independent of the laser intensity (but the time need to achieve cooling obviously increases as the laser intensity diminishes).

Note that the range of validity of the FPE is $|V| \gg 1$. It thus fails when the temperature approaches the recoil temperature (or, in other words, $|V| \approx 1$). Fig. 4 shows the dependence of the equilibrium temperature as a function of the detuning for different values of parameter $g$. The minimum temperature is clearly reduced by the decreasing of $g$, up to values close to the recoil temperature $T_r$. Moreover, the figure shows that the minimum temperature generally agrees with the theoretical predictions: it is governed both by the effective linewidth $g$ of the excited state and by the detuning, the optimum value being $\bar{\delta} \approx -g/2$ (in the range of validity of the FPE). A reasonably good agreement between numerical data and the FPE prediction within its range of validity is also observed.

Let us finally note that an interesting practical possibility is to change the quenching parameter as the cooling process proceeds. One starts with a high value of $g$ in order to rapidly cool the atoms to a few recoil velocities. Then, the quenching parameter and the detuning are progressively decreased, achieving temperatures of order of the recoil temperature. A detail study of the procedure optimizing the final temperature is however out of the scope of the present paper.

In conclusion, we have suggested and analyzed, both analytically and numerically, the using of $1S - 2S$ two-photon transition together with the quenching of the $2S$-state to cool hydrogen atoms to velocities approaching the recoil limit. The quenching ratio gives an additional, dynamically controllable parameter.

Laboratoire de Physique des Lasers, Atomes et Molécules (PhLAM) is UMR 8523 du CNRS et de l’Université des Sciences et Technologies de Lille. Centre d’Etudes et Recherches Lasers et Applications (CERLA) is supported by Ministère de la Recherche, Région Nord-Pas de Calais and Fonds Européen de Développement Economique des Régions (FEDER).
FIG. 1. Hydrogen levels involved in the two-photon Doppler cooling in presence of quenching.

FIG. 2. Numerically calculated velocity distributions with $\bar{\delta} = -0.25$ and $g = 1/3$. The dotted curve corresponds to the distribution obtained by suppressing Doppler-free transitions (cf. text). Typically, the distribution exhibits two structures: a broad background due to the atoms heat by Doppler-free transitions and a sharp peak of cold atoms.
FIG. 3. Dependence of the temperature (log scale) on the detuning. The full curve takes into account all two-photon transitions, whereas in the dotted curve the Doppler-free transitions have been suppressed. The plot shows that the effect of the latter is to increase the temperature by a factor of two, in agreement with the FPE prediction.
FIG. 4. Dependence of the temperature (log scale) on the detuning for three values of $g$: 0.9 (full line) 0.5 (dashed line) and 0.09 (dotted line). The triangles correspond to the calculation based on Eq. (8) for $g = 0.5$ and show the breaking of the Fokker-Planck approach at temperatures close to $T_r$. The curve corresponding to $g = 0.09$ shows that the minimum temperature is very close to the recoil limit.
REFERENCES


[5] Quenching of the $2S$ state can be achieved by mixing the $2S$ and the $2P$ state. This can be done, e.g., by microwave radiation around 1.04 GHz (the spacing between the two levels) or by a static electric field of a few tenths of volts. For details, see W. E. Lamb and R. C. Retherford, Phys. Rev. 81, 222 (1951); F. Biraben, J. C. Garreau, L. Julien, and M. Allegrini, Rev. Sci. Instrum. 61, 1468 (1990).
