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

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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 2. 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.

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Laser cooling of neutral atoms has been a most active research field for many years now, producing a great deal of new physics. Hydrogen atoms are interesting because of their ''simple'' structure that has led to fundamental steps in the understanding of quantum mechanics. *Free* hydrogen atoms have not yet been laser-cooled, although optical resonance-assisted evaporative cooling has been reported [1]. The recent experimental demonstration of the Bose-Einstein condensation of H adds even more interest to laser cooling of free hydrogen atoms [2]. 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, Allegrini and Arimondo suggested the laser cooling of hydrogen by two-photon π pulses on the $1S$-$3S$ transition (wavelength of 200 nm for two-photon transitions) [3]. Since then, methods for generation of continuous wave (cw), vuv, laser radiation have considerably improved, and have been extensively used in metrological experiments [4]. This technical progress allows one to realistically envision 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 large 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 1 sec. From this point of view, the $1S$-$2S$ two-photon transition is not suitable for cooling. On the other hand, the minimum temperature that is achieved via Doppler cooling is proportional to the linewidth of the excited level involved on the process [5], 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 $1S$-$2S$ transition, we consider in the present work the possibility of using the ''quenching'' [6] 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 (1D). We write rate equations describing TPDC on the $1S$-$2S$ transition in the 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 an analytical approach to the problem. A Fokker-Planck equation is derived, describing the dynamics of the process for temperatures 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 ω_L with $2\omega_L=\omega_0+\delta$, where $\omega_0/2\pi=2.5\times10^{15}$ Hz is the frequency corresponding to the transition $1S$-$2S$, and also define the quantity $k\equiv 2k_L=2\omega_L/c$. The shift of velocity corresponding to the absorption of two photons in the *same* laser wave is $\Delta\equiv\hbar k/M\approx 3.1$ m/s. We will neglect the frequency separation between $2S$ and $2P$ states (the Lamb shift, which is of the order of 1.04 GHz) and consider that the one-photon spontaneous deexcitation from the $2P$ state also shifts the atomic velocity of Δ 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 photoionization process connecting the excited states to the continuum. A more complete discussion about this point will be given by the end of the paper.

The atom is subjected to a controllable quenching process that couples the $2S$ state to the $2P$ state (linewidth $\Gamma_{2P}=6.3\times10^8\ \mathrm{s}^{-1}$). The adjustable quenching rate is Γ_q. Four

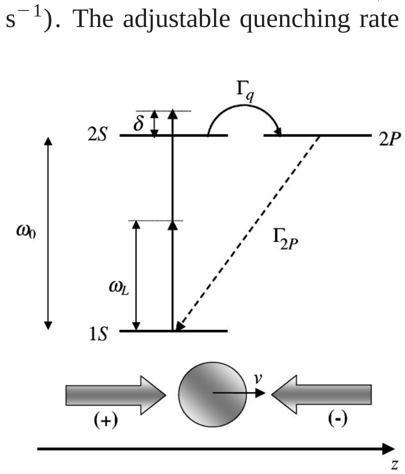

FIG. 1. Hydrogen levels involved in the two-photon Doppler cooling in the presence of quenching.

two-photon absorption processes are allowed: (i) absorption of two photons from the $+z$-propagating wave (named wave ''$+$'' in what follows), with a rate Γ_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 Γ_{-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 processes are indistinguishable, and the only relevant transition rate is that obtained by squaring the sum of the *amplitudes* of these processes (called Γ_0). Also, these processes 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 2.

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

$$\Gamma_e = \Gamma_{2P}\frac{\Gamma_q}{\Gamma_q+\Gamma_{2P}} = g\Gamma_{2P}, \tag{1}$$

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 [7] are given by

$$\Gamma_n = \Gamma_{2P}\frac{g}{2}\frac{(1+3\delta_{n0})\bar{I}^2}{(\bar{\delta}-nKV)^2+g^2/4}, \tag{2}$$

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 Γ_{2P}, $K \equiv k\Delta/\Gamma_{2P} \approx 0.26$ 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

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

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

The deduction of the above equations is quite straightforward (cf. Fig. 1). The first term on 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 first three terms on the right-hand side of Eq. (3b) describe the repopulation of the excited state velocity class V by the 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 ($V \to V \pm 1$) 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 the excited level. This is valid far from the saturation of the two-photon transitions and reduces Eqs. (3a) and (3b) to one equation describing the evolution of the ground-state population,

$$\frac{dn(V,t)}{dt} = -\left[\Gamma_0+\frac{\Gamma_{-1}(V)}{2}+\frac{\Gamma_1(V)}{2}\right]n(V,t) + \frac{1}{2}\{\Gamma_0[n(V-1,t)+n(V+1,t)] + \Gamma_{-1}(V-2)n(V-2,t)+\Gamma_1(V+2)n(V+2,t)\}. \tag{4}$$

Equation (4) is in fact a set of linear ordinary differential equations coupling the populations of velocity classes separated by an integer: V, $V\pm1$, $V\pm2$, ... This discretization exists only in the 1D approach considered here, but it does not significantly affect the conclusions of our study, while greatly simplifying the numerical approach.

Equation (4) can be recast as $d\mathbf{n}/dt = C\mathbf{n}$, where C is a square matrix and $\mathbf{n}$ is the vector $(\ldots,n(-i,t),\ldots,n(0,t),n(1,t),\ldots)$. Numerically, the equilibrium distribution is obtained simply as the eigenvector $\mathbf{n_{eq}}$ of C with zero eigenvalue. In this way, the asymptotic temperature is obtained as

$$\frac{T}{T_r} = \langle V^2\rangle = \frac{\sum_{i=-\infty}^{\infty} i^2 n_{eq}(i)}{\sum_{i=-\infty}^{\infty} n_{eq}(i)}. \tag{5}$$

Figure 2 shows the equilibrium distribution obtained by numerical simulation for $\bar{\delta}=-0.25$ and $g=1/3$. The dashed 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 mainly efficient for atoms distributed on a range of g/K around the velocity $V=\pm|\bar{\delta}|/K$ [5], whereas Doppler-free transitions are independent of the velocity; all velocity classes are thus are affected by the heating. As a consequence, DF transitions induce a deformation of the velocity profile, especially for small values of g and

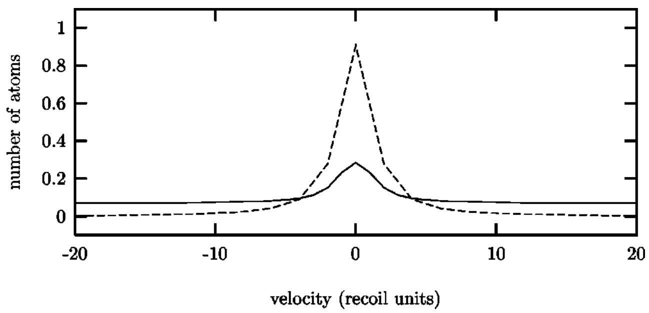

FIG. 2. Numerically calculated velocity distributions with $\overline{\delta}=-0.25$ and $g=1/3$. The dotted curve corresponds to the distribution obtained by suppressing Doppler-free transitions (cf. text). Typically, the distribution exhibits two structures: a broad background due to the atoms heat by Doppler-free transitions, and a sharp peak of cold atoms.

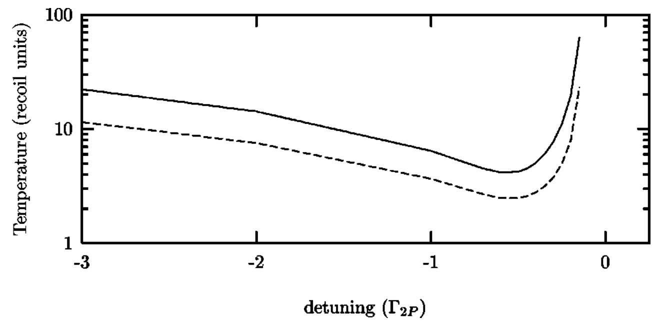

FIG. 3. Dependence of the temperature (log scale) on the detuning. The solid curve takes into account all two-photon transitions, whereas in the dashed curve the Doppler-free transitions have been suppressed. The plot shows that the effect of the latter is to increase the temperature by a factor of 2, in agreement with the FPE prediction.

$|\overline{\delta}|$, superimposing a sharp peak of cold atoms on a wide background of ''hot'' atoms. In what follows, all numerically calculated temperatures that are deduced from the width of the thin peak of cold atoms.

Equations (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 analytical approach 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 [8]). 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 $K|V| \ll |\overline{\delta}|, g$ the resulting expression can be expanded up to the order V. The resulting FPE reads

$$\frac{\partial n}{\partial t}=2\overline{\Gamma}'\frac{\partial (Vn)}{\partial V}+\left(2\overline{\Gamma}+\frac{\Gamma_0}{2}\right)\frac{\partial^2 n}{\partial V^2}, \tag{6}$$

where $\overline{\Gamma}\equiv\Gamma_{-1}(0)=\Gamma_0/4$ and $\overline{\Gamma}'$ is the V derivative of Γ_{-1} evaluated at $V=0$. Multiplying this equation by V^2 and integrating over V one easily obtains

$$\frac{d\langle V^2\rangle}{dt}=-4\overline{\Gamma}'\langle V^2\rangle+(4\overline{\Gamma}+\Gamma_0). \tag{7}$$

As $\langle V^2\rangle = T/T_r$, this equation shows that the characteristic relaxation time is $(4\overline{\Gamma}')^{-1}=(g\Gamma_{2P}\overline{I^2}\overline{\delta}K)/(4\overline{\delta}^2+g^2)$. The equilibrium temperature is then given by

$$\frac{T}{T_r}=\frac{2\overline{\Gamma}+\Gamma_0/2}{2\overline{\Gamma}'}=\frac{\overline{\delta}^2+g^2/4}{K|\overline{\delta}|}. \tag{8}$$

This result confirms that the Doppler-free two-photon transitions, corresponding to the contribution $\Gamma_0/2=2\overline{\Gamma}$ in Eq. (8) increase the equilibrium temperature (at least in the range of validity of the FPE) by a factor of 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 needed 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, $|V|\approx 1$). Figure 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 $\overline{\delta}\simeq -g/2$ (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.

Photoionization of the excited state by the laser radiation causes a decrease in the number of cooled atoms. In order to evaluate the impact of this effect on our problem, the important parameter to be considered is the *branching ratio* of the

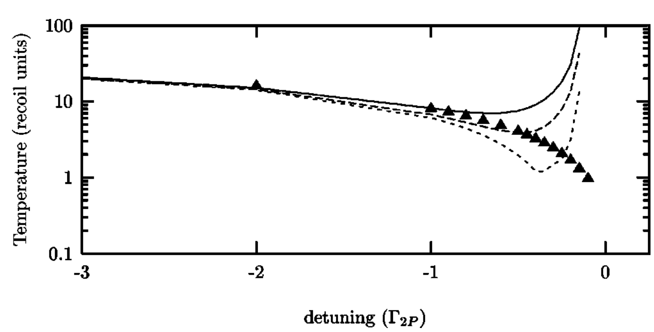

FIG. 4. Dependence of the temperature (log scale) on the detuning for three values of g: 0.9 (solid line), 0.5 (dashed line), and 0.09 (dotted line). The triangles correspond to the calculation based on Eq. (8) for $g=0.5$ and show the breaking of the Fokker-Planck approach at temperatures close to T_r. The curve corresponding to $g=0.09$ shows that the minimum temperature is very close to the recoil limit.

ionization process with respect to the spontaneous decay to the ground state, $\Gamma_{ion}/(\Gamma_{ion}+g\Gamma_{2P})$ (Γ_{ion} is the ionization ratio), that corresponds to the ionization probability per fluorescence cycle. Using the cross section for the ionization process reported in Ref. [9] one finds: $\Gamma_{ion}/\Gamma_{2P}=(1.8\times10^{-8}\ \mathrm{cm}^2\,\mathrm{W}^{-1})I$. Thus, for intensities of order of $I\approx10^3\ \mathrm{W\,cm}^{-2}$ ionization losses amount to 1.8×10^{-5} per fluorescence cycle; that is, about 18% for a full cooling cycle of typically 10^3 fluorescence cycles $(g=0.1)$.

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 detailed 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 use of the $1S$-$2S$ 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.

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