Nearly absolute one-dimensional cooling of helium atoms by two oppositely directed monochromatic plane waves

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An asymptotically exact velocity distribution is derived for atoms which are cooled by a resonant optical field in the method of coherent population trapping. A narrow structural feature which appears in the distribution function tends toward a δ-function with increasing duration of the interaction with the field.

Aspect *et al.*[1] have proposed and implemented a method for cooling helium atoms. The atoms, in the metastable 2^3S_1 state, interact on the 2^3S_1–2^3P_1 transition with oppositely directed waves with orthogonal circular polarizations. That method is fundamentally different from the scheme of Ref. 2 in terms of the depth of the cooling and also in that the efficiency of the cooling depends only weakly on the detuning from resonance. The numerical calculations carried out in Ref. 3 established general quantitative aspects of the mechanism, but they were of course incapable of precisely determining the limiting characteristics of the velocity distribution (or wave-vector distribution) of the atoms.

In the present letter we derive an exact analytic theory for this effect. We find a distribution function which is asymptotically exact (at long interaction times).

We denote the states by $2^3S_1(M_j = 1,\vec{p}) = |1,\vec{p}\rangle$, $2^3S_1(M_j = -1,\vec{p}) = |2,\vec{p}\rangle$, and $2^3P_1(M_j = 0,\vec{p}) = |0,\vec{p}\rangle$, where M_j is the projection of the magnetic moment, and $\vec{p}$ is the wave vector, which gives a quantum description of the motion of an atom as a whole.[4,5] We can then write a system of equations for the six density-matrix elements[3] $\sigma(1,\vec{p};1,\vec{p}')$, $\sigma(2,\vec{p};2,\vec{p}')$, $\sigma(1,\vec{p};2,\vec{p}')$, $\sigma(0,\vec{p};0,\vec{p}')$, $\sigma(0,\vec{p};1,\vec{p}')$, and $\sigma(0,\vec{p};2,\vec{p}')$, which describe the evolution of the state of the atom as it interacts with the field. Assuming $(dE/\hbar)^2(1/8\Gamma^2) = \kappa^2 \ll 1$, where $|d_{10}| = |d_{20}| = d$ is the dipole matrix element, E is the field amplitude, and 2Γ is the radiative width of the upper level, we can eliminate the three rapidly decaying elements $\sigma(0,\vec{p};0,\vec{p})$, $\sigma(0,\vec{p};1,\vec{p})$, and $\sigma(0,\vec{p};2,\vec{p})$ from this system of equations. The system of equations describing the evolution of the lower states then becomes

$$\frac{\partial\sigma_{11}(\vec{p})}{\partial t} = I[-M(\vec{p})\sigma_{11}(\vec{p}) + L(\vec{p})\sigma_{12}(\vec{p}) + L^*(\vec{p})\sigma_{12}^*(\vec{p}) + \frac{1}{2}A(\vec{p}+\vec{k})],$$

$$\frac{\partial\sigma_{22}(\vec{p})}{\partial t} = I[-M(-\vec{p})\sigma_{22}(\vec{p}) + L^*(-\vec{p})\sigma_{12}(\vec{p}) + L(-\vec{p})\sigma_{12}^*(\vec{p}) + \frac{1}{2}A(\vec{p}-\vec{k})], \quad (1)$$

$$\frac{\partial\sigma_{12}(\vec{p})}{\partial t} = -i2\frac{\hbar}{m}\vec{k}\vec{p}\sigma_{12}(\vec{p}) + I[L(\vec{p})\sigma_{11}(\vec{p}) + L^*(-\vec{p})\sigma_{22}(\vec{p}) - N(\vec{p})\sigma_{12}(\vec{p})].$$

Here $\sigma_{11}(\vec{p}) = \sigma(1,\vec{p}+\vec{k};1,\vec{p}+\vec{k})$, $\sigma_{22}(\vec{p}) = \sigma(2,\vec{p}-\vec{k};2,\vec{p}-\vec{k})$, $\sigma_{12}(\vec{p}) = \sigma(1,\vec{p}+\vec{k};2,\vec{p}-\vec{k})$; $k=\omega/c$ is the wave vector of the light; $I=(dE/\hbar)^2/4$; $L(\vec{p}) = [i(\omega-\omega_0-(\hbar/m)\vec{k}\vec{p}-\delta/2)-\Gamma]^{-1}$; ω_0 is the transition frequency; $M(\vec{p}) = -[L\vec{p}) + L^*(\vec{p})]$; $N(\vec{p}) = -[L(\vec{p}) + L^*(-\vec{p})]$; and $\delta = \hbar\vec{k}^2/m$ is the recoil shift. The integral incoming term A, which reflects the influence of spontaneous emission, is written in a form like that of Refs. 6 and 7. After $\sigma(0,\vec{p};0,\vec{p})$ is eliminated, this term is given by

$$A(\vec{p}) = \frac{1}{4\pi}\int dO_s[M(\vec{p}+\vec{s})\sigma_{11}(\vec{p}+\vec{s}) + M(-\vec{p}-\vec{s},)\sigma_{22}(\vec{p}+\vec{s})$$
$$+N(\vec{p}+\vec{s})\sigma_{12}(\vec{p}+\vec{s}) + N^*(\vec{p}+\vec{s})\sigma^*_{12}(\vec{p}+\vec{s})]; \quad |\vec{s}| = \omega/c.$$

At the time at which the field is applied, $t=0$, the density matrix is the equilibrium density matrix: $\sigma_{12}(\vec{p})=0$, $\sigma_{11}(\vec{p}) = W(\vec{p}+\vec{k})$, and $\sigma_{22}(\vec{p}) = W(\vec{p}-\vec{k})$, where $W(\vec{p})$ is the equilibrium wave-vector distribution. We assume that this distribution is the same for states 1 and 2.

It can be seen from Eqs. (1) that the values $\sigma_{11}(\vec{p}) = \sigma_{22}(\vec{p}) = -\sigma_{12}(\vec{p}) = D\delta(p)W(p\perp)$, where D is a constant, and p is the projection of $\vec{p}$ onto $\vec{k}$, cause the right sides of these equations to vanish. In other words, these values constitute the solution which corresponds to the coherent state mentioned in Ref. 1, which does not interact with the field. It is thus clear that in describing the solutions of (1) after a long but finite time we can seek the density matrix in the form $\sigma(\vec{p}) = W(p\perp)\rho(p)$. It is then convenient to take Fourier transforms in the p component of the wave vector and to also take Laplace time transforms:

$$F_{ik}(x,\lambda) = \int_0^\infty dt e^{-\lambda t}\int_{-\infty}^{\infty} dp e^{ixp}\rho_{ik}(p).$$

We also use the symmetry properties $\rho_{11}(p) = \rho_{22}(-p)$, $\rho_{12}(p) = \rho^*_{12}(-p)$, which follow from (1) and the initial conditions. From these properties we in turn find $F_{11}(x,\lambda) = F^*_{22}(x,\lambda)$, $F^*_{12}(x,\lambda) = F_{12}(x,\lambda)$. Assuming that the detuning from resonance is zero, i.e., $\omega-\omega_0-\delta/2=0$, we find

$$\frac{\partial f}{\partial z} = -\kappa^2[\hat{K}_-(u+f) + \hat{K}_+\varphi] - \frac{\lambda}{2\Gamma}\varphi; \quad \frac{\partial\varphi}{\partial z} = -\kappa^2[\hat{K}_+(u+f) + \hat{K}_-\varphi] - \frac{\lambda}{2\Gamma}f; \quad (2)$$
$$2\kappa^2(1-G)[\hat{K}_+(u+f) + \hat{K}_-\varphi] + \frac{\lambda}{\Gamma}u = \bar{W}(z)/\Gamma.$$

Here $z = k\Gamma x/\delta$; $f(z) = \frac{1}{2}[F_{12}(z,\lambda) + F_{12}(-z,\lambda)]$; $\varphi(z) = \frac{1}{2}[F_{12}(z,\lambda) - F_{12}(-z,\lambda)]$; $u(z) = \mathrm{Re}\,F_{11}(z,\lambda)$; $G(z) = \sin(\epsilon z)/\epsilon z$; and $\epsilon = 2\delta/\Gamma$. At large z, the imaginary part of $F_{11}(z,\lambda)$ falls off in proportion to $\sin^2(\epsilon z)/(\epsilon z)$ and plays no further role. The function $\bar{W}(x)$ is the Fourier transform of the distribution function at $t=0$:

$$\bar{W}(x) = \mathrm{Re}\int_{-\infty}^{\infty} e^{ixp}W(p+k)dp = \cos(kx)\int_{-\infty}^{\infty} W(q)e^{ixq}dq.$$

The operator $\hat{K}$ has the effect

$$\hat{K}_{\pm}\eta = \int_{-\infty}^{z} e^{-|z-z'|}\eta(z')dz' \pm \int_{z}^{+\infty} e^{-|z-z'|}\eta(z')dz'.$$

For long interaction times, a part of the density matrix ρ which is narrow with respect to p is determined by the behavior of the functions $F_{ik}(z,\lambda)$ or by the functions u, f, and φ defined in (2) at large values of z and small values of λ. The wave-vector distribution is related to the function $u(z,\lambda)$, which is given at large z by ($\overline{W}(z) \simeq 0$, $G(z) \simeq 0$)

$$u = -2\kappa^2\Gamma Q/\lambda, \quad Q = \hat{K}_+(u+f) + \hat{K}_-\varphi. \tag{3}$$

Using the relations $(\hat{K}_+\eta)' = -\hat{K}_-\eta$, $(\hat{K}_-\eta)' = -\hat{K}_+\eta + 2\eta$, we easily find from (2) equations for the functions $\psi = Q - f/\kappa^2$ and $g = Q + f/\kappa^2$:

$$\frac{d^2\psi}{dz^2} = a\psi + bg; \quad \frac{d^2 g}{dz^2} = c\psi + dg - 4\bar{W}/\lambda, \tag{4}$$

where

$$a = -\kappa^2 + \lambda\kappa^2/(2\Gamma) - \lambda/(4\Gamma) + \lambda^2/(8\Gamma^2),$$

$$b = -\kappa^2 - \lambda\kappa^2/(2\Gamma) - \lambda/(4\Gamma) - \lambda^2/(8\Gamma^2),$$

$$c = \frac{4\Gamma\kappa^2}{\lambda}(1-G) + 1 + \kappa^2 + \lambda/(4\Gamma) + \lambda\kappa^2/(2\Gamma) - \lambda^2/(8\Gamma^2),$$

$$d = \frac{4\Gamma\kappa^2}{\lambda}(1-G) + 1 - 3\kappa^2 + \lambda/(4\Gamma) - \lambda\kappa^2/(2\Gamma) + \lambda^2/(8\Gamma^2).$$

From (4) we find a fourth-order equation for ψ. At small values of λ, this equation is

$$-\lambda S(z)\psi^{(4)} + \psi^{(2)} - \frac{2\lambda\kappa^2}{\Gamma}(1 + 2G\kappa^2\Gamma S)\psi = -4\kappa^2 S\bar{W},$$

where $S = [\lambda + 4\Gamma\kappa^2(1-G)]^{-1}$. Since at small values of z we have $1 - G = \frac{2}{3}(\delta^2/\Gamma^2)z^2$, we see from an equivalent form of this equation,

$$\psi = -\frac{1}{2\sqrt{\beta}} \int_{-\infty}^{\infty} e^{-\sqrt{\beta}|z-z'|} S(z')[-4\kappa^2\bar{W}(z') + 4\lambda\kappa^4 G\psi(z') + \lambda\psi^{(4)}(z')]dz', \tag{5}$$

$$\beta = 2\lambda\kappa^2/\Gamma,$$

that the solution in the limits $\lambda \to 0$ and $z \to \infty$ is of the form $\psi = (C/\lambda)e^{-\sqrt{\beta}|z|}$. The constant C is found from (5) by setting $\lambda = 0$. It is [we recall that $\overline{W}(0) = 1$ and $G(0) = 1$] $C = [2\delta/\pi\sqrt{3\Gamma} + \kappa^2]^{-1}$. We then see from (4) that in the limit $z \to \infty$ the function g is $g(z) = C_1/\lambda e^{-\sqrt{\beta}|z|}$, where $C_1 + C = -(\lambda/\Gamma)C$, and we have

$Q = -(C/2\Gamma)e^{-\sqrt{\beta}|z|}$. Finally, for large z and small λ, we find

$$u(z,\lambda) = \frac{1}{\lambda} D \exp[-\sqrt{2\lambda\kappa^2/\Gamma}|z|], \quad D = \left[1 + \frac{2}{\pi\sqrt{3}} \frac{\delta}{\Gamma\kappa^2}\right]^{-1}. \tag{6}$$

Taking the inverse Fourier and Laplace transforms, we find the narrow part of the wave-vector distribution:

$$\rho_{11}(p) = \rho_{22}(p) = -\rho_{12}(p) = \frac{1}{2\pi i} D \int_{\xi - i\infty}^{\xi + i\infty} d\lambda \frac{e^{t\lambda}}{\sqrt{\lambda}} \frac{\frac{1}{\pi}\frac{\kappa}{\delta} k \sqrt{2\Gamma}}{p^2 + \lambda \frac{2\kappa^2\Gamma}{\delta^2} k^2}, \tag{7}$$

$$\mathrm{Re}\,\xi > 0, \qquad \int \rho_{11}(p) dp = D.$$

It is a simple matter to find the values of this function at small and large values of p. For $p \ll \Delta_0 = k\kappa/\delta\sqrt{(\pi/2)(\Gamma/t)}$, we find $\rho_{11}(p) = D/(\pi\Delta_0)$; for $p \gg \Delta_0$, we find $\rho_{11}(p) = (D/\pi)2\Delta_0/(\pi p^2)$. On the whole, this function is approximately Lorentzian, $\rho_{11}(p) \simeq (D/\pi)\Delta(p)/[p^2 + \Delta^2(p)]$, with a p-dependent width $\Delta(p)$ which varies from $\Delta = \Delta_0$ at $p = 0$ to $\Delta = (2/\pi)\Delta_0$ at $p \gg \Delta_0$. The mean value of these two quantities, $(\pi + 2/2\pi)\Delta_0$, characterizes the width of the narrow part of the distribution function highly accurately. This width is smaller by a factor of about 1.5 than the width derived on the basis of qualitative considerations in Ref. 3; it is essentially the same as the width found by a numerical method (Fig. 8a in Ref. 3).

Equations (6) and (7) embody all the principal features of the cooling mechanism. The width of the distribution in (7) is limited only by the interaction time, so the narrow part of the distribution function tends toward a δ-function as $t \to \infty$. In practice, this width is apparently limited only by the lifetime τ of the $^3S_1$ lower state (which is very long, $\tau \simeq 7000$ s; Ref. 8), by the classical description of the field, and (possibly) by the use of a nonrelativistic equation for the density matrix. The cooling efficiency (the fraction of the atoms which are brought into the process) is determined by the constant D, which is independent of the initial distribution (that at $t = 0$). The width of this distribution determines only (a) the time t at which the asymptotic behavior in (6) sets in and (b) the shape of the background, which is broad in comparison with (7), and whose integral is $1 - D$. Under the conditions of the numerical calculations of Ref. 3 (Fig. 11c), we would have $\kappa^2 = 0.05$. The ratio δ/Γ for the transition of interest would be $\delta/\Gamma = 10^{-1}$. From (6) we find the value $D \simeq 0.6$, which is 0.2 below the value found numerically in Ref. 3. It can be seen from (6) that the cooling efficiency D increases with increasing field intensity, i.e., with increasing κ^2. We should bear in mind, however, that this entire discussion has been conducted under the assumption $\kappa^2 \ll 1$, and it is obvious from qualitative considerations that the value of D could not be greater than the value given by (6) with $\kappa^2 = 1$.

An interesting aspect of distribution (7) is that all the moments diverge. In other words, it is impossible to determine, among other things, an average energy. Clearly, this stems from the fact that expression (6) is no longer applicable at small values of z. To find the function $u(z,\lambda)$ for small values of z will require an additional analysis. That analysis will be reported elsewhere.

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