How Large Photon Recoil Can Make Cold Atoms Lase

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We calculate the weak probe field absorption spectrum for ultracold three-level atoms in a Raman gain configuration in the limit of the recoil shift exceeding the natural linewidth and the Doppler broadening. Our results show that even in the limit of very large recoil gain persists with its maximum shifted toward the red of the atomic line by the recoil energy, as is the spontaneous emission line. Some preliminary calculations suggest the possibility of a cw deep uv laser, based on cooled metastable helium atoms.

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Our standard models for describing laser action in the infrared and visible frequency domain were established in the 1960s by Haken [1], Lax [2], and Lamb [3]. Albeit being based on several important simplifying assumptions, they have proved very successful in describing most of the lasing phenomena on the classical as well as the quantum level.

One particularly important assumption is to ignore the recoil imparted to an atom by emission or absorption of a laser photon, hence decoupling the internal and external degrees of freedom for the atomic time evolution. Although it has already been shown by Einstein [4] that a consistent thermodynamic description of atoms coupled to light requires the inclusion of photon momentum, this normally constitutes a fairly good approximation if the recoil-related changes in energy of the absorbed or emitted photons are much less than the natural linewidth of the lasing transition. This is the case for a typical optical transition. Hence the translational degrees of freedom enter the model only as external parameters in the form of Doppler shifts, i.e., they are manifested by an inhomogeneous broadening of the atomic lines. On the other hand, dramatic effects of mechanical light forces have recently been demonstrated in numerous theoretical and experimental works on laser cooling and trapping, optical gratings, etc. It was noted by Marcuse [5] as early as 1963 that *inversionless* maser action should be possible if the recoil shift exceeds the linewidth due to Doppler broadening because the emitted photons are redshifted and less likely to be absorbed by neighboring atoms.

Extending the theory of lasing into the uv and xuv domain, one has to think even more carefully about recoil-related effects. For shorter wavelengths the recoil energy is vastly increased according to the power law $E_R = (\hbar k)^2/2m$, with m being the atom's mass and $k = 2\pi/\lambda$ the wave vector. Because of higher mode densities in this frequency range also the spontaneous emission rates ($\propto k^3$) will be increased correspondingly, and thus we should in principle be allowed to use the same models.

However, in the ultraviolet domain incoherent optical pumping to achieve a steady state inversion is not efficient due to the presence of fast spontaneous decay. We realize that a viable gain scheme for uv radiation will need to provide a transition whose excited state does *not* decay rapidly. Consequently, the recoil shift is now likely to exceed the natural linewidth of the transition and will need to be accounted for. Hence the atomic absorption and emission line for an atom *initially at rest* will differ in frequency and a spontaneously emitted photon cannot be absorbed by a second atom also at rest. *But can there be stimulated emission at this frequency? And what may we expect in general for the frequency dependence of the stimulated emission probability?*

Despite some earlier work [5–7] only very recently experimental interest in the effects of photon recoil in spectroscopy [8,9] has been renewed. We propose utilizing a Raman gain scheme which allows the use of a metastable state to create atomic inversion with sufficiently large stimulated transition probability. In order to further benefit from the recoil-mediated reduction of reabsorption we will use cold atoms in a metastable state.

Let us now consider an ensemble of cold independent atoms interacting with two laser fields in a Raman configuration. By an external cooling mechanism (e.g., laser cooling) the velocity distribution of the atoms is narrowed. In a very simplified model of such a cooling process, we assume that level $|1\rangle$ is populated at a rate γ_t (owing to the number of fluctuations in the interaction region) with a narrow Gaussian momentum distribution. Loss from all levels is assumed to occur at the same rate irrespective of the momentum state of the atom.

A coherent optical light field with Rabi frequency Ω_p and detuning $\Delta_p = \omega_{12} - \omega_p$ drives the transition between levels $|1\rangle$ and $|2\rangle$ and acts as a pump inducing Raman–anti-Stokes processes toward the ground state $|0\rangle$, as shown in Fig. 1. Obviously, the corresponding anti-Stokes photons are redshifted by the recoil energy as required by conservation of energy. Spontaneous decay takes place on this transition at a rate $2\Gamma_p$. The uv transition $|0\rangle$-$|2\rangle$ is probed by a weak coherent field with Rabi frequency Ω_l and detuning $\Delta_l = \omega_{02} - \omega_l$. We

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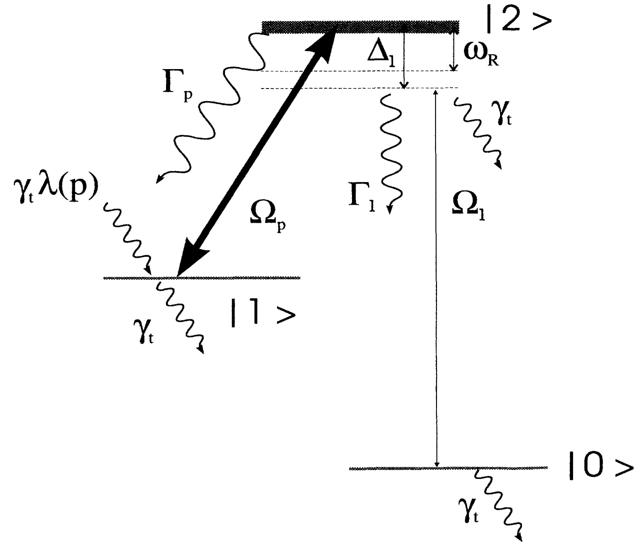

FIG. 1. Model of a three-level system interacting with a pump field Ω_p and a weak probe field Ω_s. The atoms enter the interaction region in state $|1\rangle$ with momentum distribution $\lambda(p)$.

assume the transition to be sufficiently weak so that the rate of spontaneous decay $2\Gamma_l$ may be assumed to be very small. The rates satisfy $\Gamma_l \ll \Gamma_p < \omega_R = \hbar k^2/2m$. A prototype for such a system is formed by the states 2^3S_1, 2^3P_1, and 1^1S_0 of He I [10].

The appreciable size of the recoil and the low temperature of the atomic ensemble command accounting for the atoms' translational degrees of freedom in the direction of propagation of the weak probe field. Because of the vastly different size of the decay rates on the transitions and of the k vectors associated with the two applied fields we assume the pumped transition to be unaffected by the atoms' motion, i.e., we neglect Doppler shifts and recoil effects on the 1-2 transition. A mathematical description of our system, as depicted in Fig. 1, is effected by a master equation for the atomic density operator $\rho(t)$. Considering the operator $\rho_I(t)$ in lieu of $\rho(t)$, we switch to a rotating frame (Schrödinger interaction picture) relative to H_0, where

$$H_0 = \omega_R \hat{p}^2 + \omega_l \sigma_{22} + (\omega_l - \omega_p)\sigma_{11}\,. \tag{1}$$

While the dimensionless momentum operator $\hat{p}$ (scaled in units of $\hbar k$) is time independent, the position operator $\hat{x}$ becomes explicitly time dependent in this rotating frame. Introducing $\gamma' = \Gamma_p + \Gamma_l$ and the distribution of spontaneous emission (normalized on $\{p|p \in [-1,1]\}$) into the propagation direction of the probe field [11] $N(p) = \frac{3}{8}(1 + p^2)$, we find

$$\begin{aligned}\dot{\rho}_I = &- i[H_I, \rho_I] - \gamma'\{\sigma_{22}, \rho_I\}_+ + 2\Gamma_p \sigma_{12}\rho_I\sigma_{21} \\ &+ 2\Gamma_l \int_{-\hbar k}^{\hbar k} N(q/\hbar k)\, \frac{dq}{\hbar k}\, S_q(t)\rho_I S_q(t)^\dagger \\ &- \gamma_t[\rho_I - \sigma_{11}\lambda(\hat{p})],\end{aligned} \tag{2}$$

where $S_q(t) = \exp[-iq\hat{x}(t)]\sigma_{02}$ and H_I describes the interaction of the atoms with the two applied fields,

$$\begin{aligned}H_I = &(\Delta_l - \Delta_p)\sigma_{11} + \Delta_l\sigma_{22} \\ &- i[\Omega_p\sigma_{12} + \Omega_l^* S_k(t) - \text{H.c.}]\,.\end{aligned} \tag{3}$$

For the momentum distribution of the cooled atoms we assume

$$\langle p|\lambda(\hat{p})|p'\rangle = \delta(p - p')\exp(-p^2/2\sigma_p)/\sqrt{2\pi\sigma_p}\,, \tag{4}$$

where $\sigma_p \approx k_B T/E_R \ll 1$ and p is given in units of $\hbar k$. Note that Eq. (4) implies a uniform distribution in position space. Denoting the density of the atomic medium by n we may establish the relation

$$\begin{aligned}\rho_{ij}(x,t) = n &\int dp\, dp' \\ &\times \rho_{ij}(p,p',t)e^{i[(p-p')kx-(p^2-p'^2)\omega_R t+\nu_{ij}t]},\end{aligned}$$

which we will use farther down in our calculation of the gain coefficient. Note that ν_{ij} follows from H_0.

A convenient way to treat a master equation is to formulate the problem in terms of momentum families of density matrix elements [11]. In the absence of spontaneous emission *with recoil* the family of states $\mathcal{F}(p) = \{|1,p\rangle, |2,p\rangle, |0,p-1\rangle\}$ is closed, cf. Fig. 2. It thus suffices to consider the matrix elements of a family density operator

$$\rho^{\mathcal{F}}(t,p) = \sum_{i,j=0}^{2} \rho_{ij}^{\mathcal{F}}(p)|i,p - \delta_{i0}\rangle\langle j,p - \delta_{j0}|\,. \tag{5}$$

We will refrain here from presenting the full set of equations for the matrix elements of $\rho^{\mathcal{F}}(t,p)$ as their derivation is straightforward from combining Eqs. (2) and (5).

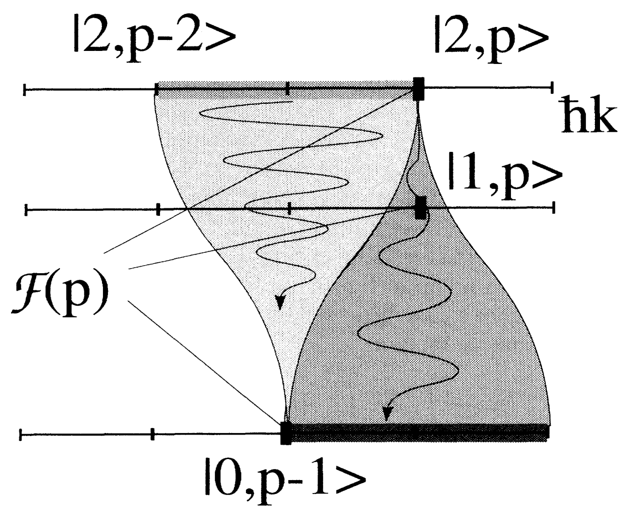

FIG. 2. Illustration of the way in which a continuum band of momentum families can couple to a family $\mathcal{F}(p)$ due to spontaneous decay on the 0-2 transition.

As a next step we now calculate the weak field absorption spectrum [12] on the uv transition which provides a measure of the gain to be expected:

$$W'(\omega_l) = \hbar\omega_l|\Omega_l|^2 \int_{-\infty}^{\infty} e^{i\omega_l t}\langle[S_k(t)^\dagger, S_k(0)]\rangle\, dt\,. \qquad (6)$$

Note that the brackets designate an average over all momentum families. To first approximation the effect of the weak probe field on the atomic system may be neglected which implies that there are only five nonzero matrix elements in any unperturbed family $\mathcal{F}(p)$. These quite naturally separate into two sets. One set of equations comprises only the matrix elements $\{\rho_{12}^{\mathcal{F}}, \rho_{21}^{\mathcal{F}}, \rho_{22}^{\mathcal{F}}, \rho_{11}^{\mathcal{F}}\}$, thus describing the driving of the atoms by an external pump field. The remaining nonzero element $\rho_{00}^{\mathcal{F}}$ of the family $\mathcal{F}(p)$ couples to the excited state matrix elements of a whole cluster of families $\mathcal{F}(p - p')$ (cf. Fig. 2) with $p' \in [0, 2]$ as required by the angular distribution of spontaneous emission $N(p')$ [Eq. (2)]. Dropping the superscript $\mathcal{F}$ we find the steady state relation

$$\rho_{00}(p) = \eta \int_{-1}^{1} dp\, N(p')\rho_{22}(p + p' - 1), \quad \text{with } \eta = \frac{2\Gamma_l}{\gamma t}\,. \qquad (7)$$

Using straightforward algebra we find the following steady state expressions of the unperturbed system (lowest order in $|\Omega_l|$):

$$\rho_{11}(p) = \frac{\Gamma + R}{R}\rho_{22}(p), \qquad \rho_{22}(p) = \frac{R\lambda(p)}{\Gamma + R(2 + \eta)}, \qquad (8a)$$

$$\rho_{00}(p) = \frac{\eta R}{\Gamma + R(2 + \eta)}\frac{3}{8}\left\{\frac{1}{2}[1 + (p - 1)^2 + \sigma_p] \times \left[\mathrm{erf}\left(\frac{p}{\sqrt{2\sigma_p}}\right) - \mathrm{erf}\left(\frac{p - 2}{\sqrt{2\sigma_p}}\right)\right] + \sigma_p[(p - 2)\lambda(p) - p\lambda(p - 2)]\right\}. \qquad (8b)$$

For the sake of compactness of notation we have set $\gamma = \gamma' + \gamma_t$, $R = \gamma|\Omega_p|^2/(\gamma^2 + \Delta_p^2)$, and $2\Gamma = \gamma' + \gamma$ in Eqs. (8). In Fig. 3 we depict the stationary momentum distribution $\Lambda(p) = \rho_{22}(p) + \rho_{11}(p) + \rho_{00}(p + 1)$ for three different ratios ω_R/Γ_p. Note that $\Lambda(p)$ approaches $\lambda(p)$ in the limit of negligible recoil.

We omit the rather uninspiring details of a recasting of the definition given in Eq. (6) in terms of the steady state solutions above and hence proceed with the result. For the spectrum we obtain

$$W'(\omega_l) = \hbar\omega_l|\Omega_l|^2 2\,\mathrm{Re}\int_{-\infty}^{\infty} dp\, \frac{1}{\gamma_{01}\gamma_{02} + |\Omega_p|^2} \times \left(\gamma_{01}[\rho_{22}(p) - \rho_{00}(p)] + \frac{\Gamma|\Omega_p|^2}{R\gamma_{12}^*}\rho_{22}(p)\right), \qquad (9)$$

where we have used the abbreviations $\gamma_{01} = \gamma_t + i(\delta_l - \Delta_p)$, $\gamma_{12} = \gamma + i\Delta_p$, $\gamma_{02} = \gamma + i\delta_l$, and $\delta_l = \Delta_l - \omega_R(1 - 2p)$. It is clear that in the general case the integral in Eq. (9) has to be evaluated numerically. The impact recoil has on the line shape of absorption can be visualized by considering the hypothetical case in which we may alter the wavelength of the probed transition while leaving all other parameters the same. Mathematically, this is achieved by altering the value of ω_R (in units of Γ_p) and σ_p. Figure 3 illustrates the effect this has on the stationary momentum distribution. For smaller recoil and thus larger σ_p the distribution grows wider on the scale given by the photon momentum $\hbar k$. But the broadening due to spontaneous emission remains the same and is therefore no longer well resolved.

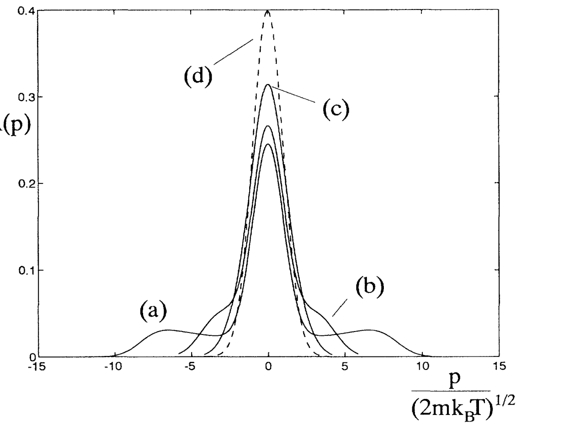

FIG. 3. Stationary momentum distribution of the atomic ensemble for a given initial distribution and $\omega_R/\Gamma_p = 15.571$ [curve (*a*)], $\omega_R/\Gamma_p = 4$ [curve (*b*)], and $\omega_R/\Gamma_p = 1$ [curve (*c*)]. Curve (*d*) depicts the initial momentum distribution $\lambda(p)$. The other parameters were chosen to be $\Omega_p/\Gamma_p = 0.5$, $\Gamma_p/\Gamma_l = 239$, $k_B T/\hbar\Gamma_p = 0.228$, and $\gamma_t/\Gamma_p = 1.7 \times 10^{-3}$.

Figure 4 illustrates the dependence of $W'(\Delta_l)$ on the ratio ω_R/Γ_p. The characteristic feature of a larger recoil shift is a more pronounced asymmetry in the absorption profile. Absorption takes on a maximum for laser detunings $\omega_l \approx \omega_{02} - \omega_R, \omega_{02} + 3\omega_R$. The qualitative behavior can be understood from simple energy and momentum conservation considerations. An atom that has emitted a photon into the direction of the probe light and is traveling with a momentum $p \approx -\hbar k$ is likely to reabsorb a photon with frequency $\omega_{02} - \omega_R$. Emission into the opposite direction speeds up the atom. Hence absorption is likely to occur at blueshifted frequencies $\omega_l \approx \omega_{02} + 3\omega_R$. Stimulated emission can be observed for $\omega_l \approx \omega_{02} - \omega_R$. The exact quantitative behavior is determined by averaging over the momentum distribution.

The above arguments hold if the width of the momentum distribution of the atoms is less than $\hbar k$ as is the case in curve (*a*). In this recoil-dominated limit the asymmetry is well resolved. Decreasing the size of the recoil shift ω_R while leaving the width of the momentum distribution the same results in a deformation of the line shape into

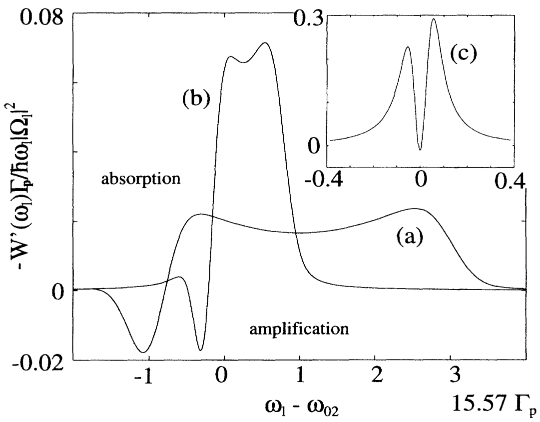

FIG. 4. $W'(\omega_l)/\hbar\omega_l|\Omega_l|^2$ as a function of $\omega_l - \omega_{02}$. The parameters for curves (a) and (b) are as for the corresponding curves in Fig. 3. In (c) we chose $\omega_R/\Gamma_p = 0.1$, $\sigma_p^{-1} = 0.44$. Note that for $\eta \geq 1$ there can be inversionless amplification.

something more symmetrical [curve (b)]. In the Doppler-shift dominated limit [curve (c)] we obtain a V-shaped curve exhibiting absorption for frequencies close to resonance with the Autler-Townes doublet and a narrow central gain region which is due to Raman–anti-Stokes processes.

The gain from a device consisting of an ensemble of cold atoms (forming a cloud or well collimated beam) inside a single-mode traveling wave ring resonator can be estimated by relating it to the absorption spectrum in Eq. (9). We find for the intensity gain per round-trip length [13] $G(\omega_l) \approx n(l_{\rm int}/L)2\Gamma_l\lambda^2\xi/8\pi g(\omega_l)$, where $l_{\rm int}$ denotes the length of the atomic sample and L is the round-trip length. Note that $g(\omega_l)^{-1} = W'(\omega_l)/\hbar\omega_l|\Omega_l|^2$, and $\xi = 3|\langle 0|x|2\rangle|^2/\sum_i |0\langle|x_i|2\rangle|^2$ is of the order of unity.

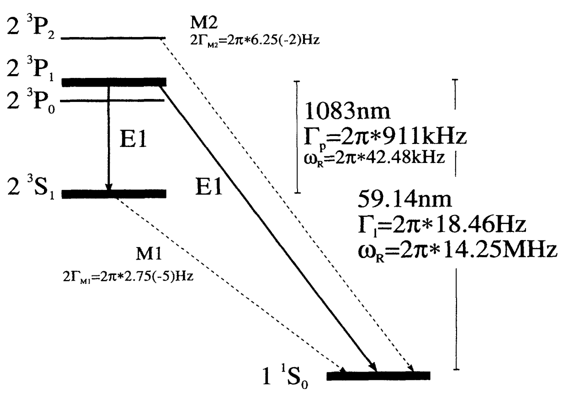

FIG. 5. Energy level scheme of He I.

As already indicated in the introduction, the states 2^3S_1, 2^3P_1, and 1^1S_0 of cold He I atoms form a real-life system that closely resembles our model. In Fig. 5 we depict the corresponding level scheme and specify the relevant spectroscopic data. The transition 2^3P_1-1^1S_0 is an intercombination transition (dipole-allowed but spin-forbidden) [14] with a small transition matrix element. The recoil shift on the 60 nm transition exceeds Γ_p by a factor of 15.571. Assuming a temperature of 10 μK we find $\sigma_p^{-1} \approx 68$. It is somewhat difficult to give a realistic estimate of the rate γ_t as this will depend strongly on the experimental configuration. For simplicity we set $\gamma_t = 2\pi(1.6 \times 10^3\mathrm{Hz})$. Eventually, we find for the gain $G(\omega_{2^3P_1\text{-}1^1S_0} - \omega_R) \approx (nl_{\rm int}/cL)5.56 \times 10^{-7}\ \mathrm{cm^3\,s^{-1}}$. Laser threshold is reached for gain exceeding the round-trip loss $\kappa \approx -\ln r/L$ with r the reflectivity of the in-out port. Assuming $l_{\rm int}/L \approx 0.01$ and $c\kappa = 1$ MHz densities in excess of $10^{16}\ \mathrm{cm^{-3}}$ are required.

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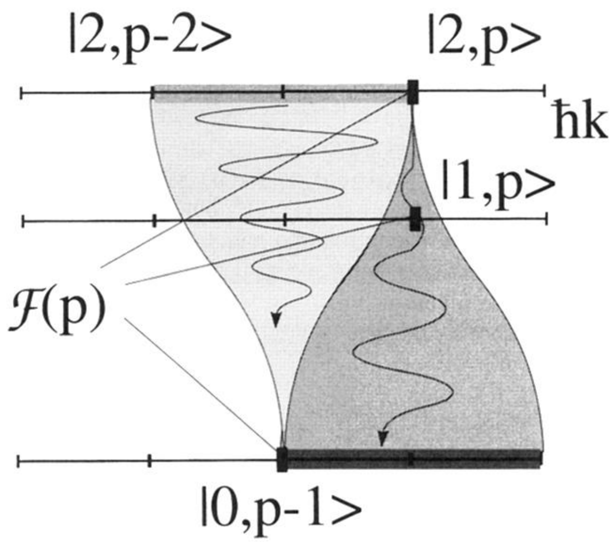

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