Gain without population inversion in two-level atoms

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For the case in which laser radiation interacts with two levels of an atom, it is shown that there can be gain, even when there is no population inversion, because of recoil effects. The effect of recoil on certain precision measurements is also shown.

For the case of a system in which laser radiation interacts with just two levels of an atom, as well as for more complicated systems, it is generally thought that the population of the upper level must be greater than that of the lower level in order for gain to exist, i.e., in order for amplification of the radiation to occur. The purpose of this paper is to show that, because of the effects of recoil, gain can be achieved with more atoms in the lower level than in the upper level. The effect is small, though significant for some precision measurements, for visible laser light impinging on ordinary atoms, but it can be large for light atoms and short-wavelength radiation, such as x rays. The effect is related to that underlying the free-electron laser.[1]

The problem under consideration is that of a wave of circular frequency ν traveling in the z direction and polarized in the x direction, impinging upon an atom with two states, a (upper) and b (lower). For simplicity we assume the atom to be hydrogen-like. Then the Hamiltonian describing the system of atom plus radiation is

$$H = H_0 + H' ,$$

where

$$H_0 = \frac{\vec{\eta}^2}{2M} + \frac{\vec{p}^2}{2\mu} + eV$$

and

$$H' = -\frac{e}{c}\vec{A}(\vec{R}) \cdot \frac{\vec{p}}{\mu} .$$

Here, $\vec{R}$ and $\vec{\eta}$ are the position and momentum of the center of mass, respectively, and $\vec{r}$ and $\vec{p}$ are the relative position and momentum. M is the total mass, and μ is the reduced mass. $\vec{A}(\vec{R})$ is the vector potential of the radiation field at the center of mass. [Exactly, $\vec{A}$ should be taken at the positions of each particle separately; using $\vec{A}(\vec{R})$ is equivalent to the dipole approximation.]

The laser beam is described by an electric field

$$\begin{aligned}\vec{\mathcal{E}}(z, t) &= \mathcal{E}(z, t)\hat{i}_x \\ &= E(z)\cos(\nu t - kz + \varphi)\hat{i}_x \\ &= -\frac{1}{c}\frac{\partial \vec{A}}{\partial t} .\end{aligned} \tag{1}$$

The eigenfunctions of H_0 satisfy the equation

$$H_0\phi_{nn'} = E_{nn'}\phi_{nn'} ,$$

where

$$\phi_{nn'} = \varphi_n u_{n'} ,$$

$$E_{nn'} = \mathfrak{N}_n^2/2M + W_{n'} .$$

Here,

$$(\vec{\eta}_n^{\,2}/2M)\varphi_n = (\vec{\eta}_n^{\,2}/2M)\varphi_n ,$$

$$\varphi_n = (1/\sqrt{V})\exp(i\vec{\eta}_n \cdot \vec{R}/\hbar) .$$

Also,

$$(\vec{p}^{\,2}/2\mu + eV)u_{n'} = W_{n'}u_{n'} ;$$

$u_{n'}$ is an atomic wave function. The kinetic-energy wave functions are normalized to a box of volume $\bar{V}$. Eventually, $\bar{V} \to \infty$.

We expand the wave function for the complete system in terms of the eigenfunctions of H_0:

$$\Psi = \sum_{nn'} a_{nn'}(t)\phi_{nn'}\exp(-iE_{nn'}t/\hbar) .$$

The time-dependent Schrödinger equation,

$$i\hbar\frac{\partial\Psi}{\partial t} = (H_0 + H')\Psi ,$$

results in the equation for the amplitudes,

$$i\hbar\dot{a}_{nn'} = \sum_{mm'} a_{mm'}H'_{nn';mm'}\exp[i(E_{nn'} - E_{mm'})t/\hbar] .$$

Here,

$$H'_{nn';mm'} = \frac{e}{c}\frac{cE}{2i\nu}\frac{1}{V}\times\int \exp\left(-i\frac{(\vec{\eta}_n-\vec{\eta}_m)\cdot\vec{R}}{\hbar}\right)u_n^*\times\left\{\exp[i(\nu t-KZ+\varphi)] -\exp[-i(\nu t-KZ+\varphi)]\right\}\frac{p_x}{\mu}u_{m'}d\vec{R}\,d\vec{r}\,. \quad (2)$$

The integration over $d\vec{R}$ is

$$\frac{1}{V}\int \exp\left[-i\frac{(\vec{\eta}_n-\vec{\eta}_m\pm\hbar K\hat{k})\cdot\vec{R}}{\hbar}\right]d\vec{R} = \delta(\vec{\eta}_n-\vec{\eta}_m\pm\hbar K\hat{k})\,.$$

Here, δ is the Kronecker delta, and the plus and minus signs go with the first and second terms in Eq. (2), respectively. The integration over $d\vec{r}$ is

$$\int u_{n'}\left(\frac{p_x}{\mu}\right)u_{m'}d\vec{r} = i w_{n'm'}\langle x\rangle_{n'm'}\,.$$

If

$$\mathcal{P}_{n'm'} = e\langle x\rangle_{n'n'}\,,$$

then

$$H'_{nn';mm'} = \frac{\mathcal{P}_{n'm'}E}{2}\frac{w_{n'm'}}{\nu}\times\{e^{i(\nu t+\varphi)}\delta(\vec{\eta}_n-\vec{\eta}_m+\hbar K\hat{k}) -e^{-i(\nu t+\varphi)}\delta(\vec{\eta}_n-\vec{\eta}_m-\hbar K\hat{k})\}\,. \quad (3)$$

Now suppose there are two atomic levels, a and b, and that $W_a > W_b$. a_n is the amplitude of the state for which the atom is in level a and has momentum $\vec{\eta}_n$; b_m is the amplitude of the state for which the atom is in level b and has momentum $\vec{\eta}_m$. Then

$$\dot{a}_n = \frac{-i}{\hbar}\sum_m b_m H'_{na;mb}e^{iw_{ab}t}\exp\left[\frac{i}{\hbar}\left(\frac{\eta_n^2-\eta_m^2}{2M}\right)t\right]. \quad (4)$$

In the rotating-wave approximation, the first term on the right-hand side of Eq. (3) can be neglected as nonresonant. Then Eq. (4) and a similar equation for $\dot{b}_m$ give

$$\dot{a}_n = i v b_m \exp[-i(\nu-w_{ab})t]\exp[i\nu(\beta_{nz}-\epsilon/2)t]\times\delta(\vec{\eta}_n-\vec{\eta}_m-\hbar K\hat{k})\,, \quad (5)$$

$$\dot{b}_m = i v^* a_n \exp[i(\nu-w_{ab})t]\exp[-i\nu(\beta_{mz}+\epsilon/2)t]\times\delta(\vec{\eta}_m-\vec{\eta}_n+\hbar K\hat{k})\,, \quad (6)$$

where

$$v\equiv\frac{\mathcal{P}_{ab}E}{2\hbar}\frac{w_{ab}}{\nu}e^{-i\varphi}\,,$$

$$\beta_{nz} = (\vec{\eta}_{nz}/Mc\,,$$

$$\epsilon = \hbar\nu/Mc^2\,.$$

Thus states a_n and b_m such that $\vec{\eta}_m = \vec{\eta}_n - \hbar K\hat{k}$ (or $\beta_{mx}=\beta_{nx}$, $\beta_{my}=\beta_{ny}$, $\beta_{mz}=\beta_{nz}-\epsilon$) are connected only to each other.

In order to account for spontaneous emission, we add phenomenological decay terms $-(\gamma_a/2)a_n$ and $-(\gamma_b/2)b_m$ to Eqs. (5) and (6), respectively, where γ_a and γ_b are reciprocal lifetimes.[2] To include excitation to levels a and b we use the density matrix method and add λ_{an} and λ_{bm} to $\dot{\rho}_{aa}^{nn}$ and $\dot{\rho}_{bb}^{mm}$, respectively.[3] Here λ_{an} and λ_{bm} are the number excited per second to states a_n and b_m, respectively; also, $\rho_{aa}^{nn}=a_n a_n^*$, $\rho_{bb}^{mm}=b_m b_m^*$, and $\rho_{ab}^{nm}=a_n b_m^*$. The resulting equations, derived from Eqs. (5) and (6) with the aforesaid additions, are

$$\dot{\rho}_{aa}^{nn} = \lambda_{an}-\gamma_a\rho_{aa}^{nn}-2\mathrm{Re}(iv^*e^{i\Omega_n t}\rho_{ab}^{nm})\,,$$

$$\dot{\rho}_{bb}^{mm} = \lambda_{bm}-\gamma_b\rho_{bb}^{mm}+2\mathrm{Re}(iv^*e^{i\Omega_n t}\rho_{ab}^{nm})\,,$$

$$\dot{\rho}_{ab}^{nm} = -\gamma_{ab}\rho_{ab}^{nm}-ive^{-i\Omega_n t}(\rho_{aa}^{nn}-\rho_{bb}^{mm})\,,$$

where

$$\Omega_n = \nu - w_{ab} - \nu(\beta_{nz}-\epsilon/2)$$

and

$$\gamma_{ab} = (\gamma_a+\gamma_b)/2\,.$$

These equations can be solved by assuming

$$\rho_{aa}^{nn}-\rho_{bb}^{mm} = \text{const.}$$

The result is

$$\rho_{aa}^{nn}-\rho_{bb}^{mm} = \frac{(\lambda_{an}/\gamma_a-\lambda_{bm}/\gamma_b)\delta(\vec{\beta}_m-\vec{\beta}_n+\epsilon\hat{k})}{1+8\alpha\gamma_{ab}^2/(\Omega_n^2+\gamma_{ab}^2)}\,,$$

$$\rho_{ab}^{nm} = \frac{ve^{-i\Omega_n t}(\lambda_{an}/\gamma_a-\lambda_{bm}/\gamma_b)\delta(\vec{\beta}_m-\vec{\beta}_n+\epsilon\hat{k})}{(\Omega_n+i\gamma_{ab})[1+8\alpha\gamma_{ab}^2/(\Omega_n^2+\gamma_{ab}^2)]}\,,$$

where

$$\alpha = |v|^2/2\gamma_a\gamma_b\,.$$

The polarization is given by

$$P = \int \psi^* ex\psi\,d\tau = 2\mathrm{Re}\left\{\sum_n \rho_{ab}^{nm}\frac{\mathcal{P}_{ab}^*}{V}\exp\left(i\frac{(\vec{\eta}_n-\vec{\eta}_m)\cdot\vec{R}}{\hbar}\right)\exp\left(\frac{-i(E_{na}-E_{mb})t}{\hbar}\right)\right\}$$

$$= 2\mathrm{Re}\left\{\sum_n \frac{\mathcal{P}_{ab}^* v(\lambda_{an}/\gamma_a V-\lambda_{bm}/\gamma_b V)\exp[-i(\nu t-kz)]}{(\Omega_n+i\gamma_{ab})[1+8\alpha\gamma_{ab}^2/(\Omega_n^2+\gamma_{ab}^2)]}\delta(\vec{\beta}_m-\vec{\beta}_n+\epsilon\hat{k})\right\}\,.$$

Here, $\lambda_{an}/\gamma_a \bar{V}$ is the number of atoms per cm³ in state a with β_{nz}, and $\lambda_{bm}/\gamma_b\bar{V}$ is the number per cm³ in b with $\beta_{mz}=\beta_{nz}-\epsilon$. Since states an and bm are not connected to states other than each other, we may say that one state an (with $\beta_{nz}=\beta_z$) and one state bm (with $\beta_{mz}=\beta_z-\epsilon$) are populated and integrate over β_z. We write $\lambda_{an}/\gamma_a\bar{V}\to n_a(\vec{v})d\vec{v}$, where $n_a(\vec{v})d\vec{v}$ is the number of atoms per cm³ in state a with velocity $\vec{v}\to\vec{v}+d\vec{v}$. Then

$$P=2\mathrm{Re}\left\{\frac{|\mathcal{P}_{ab}|^2E}{2\hbar}\frac{w_{ab}}{\nu}\exp[-i(\nu t-Kz+\varphi)]\int\frac{[n_a(\vec{v})-n_b(\vec{v}-c\epsilon\hat{k})]d\vec{v}}{(\Omega+i\gamma_{ab})[1+8\alpha\gamma_{ab}^2/(\Omega^2+\gamma_{ab}^2)]}\right\}$$
$$\equiv\mathrm{Re}\{(C+iS)\exp[-i(\nu t-Kz+\varphi)]\}. \tag{7}$$

Here, $\Omega=\nu(1-\beta_z+\epsilon/2)-w_{ab}$.

If relativistic effects are important, they can be included by solving the problem in the rest frame of an atom in state a traveling with velocity $c\vec{\beta}$. Then, after P is transformed back into the laboratory frame, Eq. (7) becomes

$$P=2\mathrm{Re}\left\{\frac{|\mathcal{P}_{ab}|^2E}{2\hbar}\frac{w_{ab}}{\nu}\exp[-i(\nu t-Kz+\varphi)]\int\frac{[n_a(\vec{v})-n_b(\vec{v}-c\epsilon'\hat{k})]d\vec{v}}{(\Omega'+i\gamma_{ab})[1+8\alpha\gamma_{ab}^2/(\Omega'^2+\gamma_{ab}^2)]}\right\},$$

where

$$\Omega'=\nu'(1+\epsilon'/2)-w_{ab},$$
$$\nu'=\frac{\nu(1-\beta z)}{(1-\beta^2)^{1/2}},$$
$$\epsilon'=h\nu'/Mc^2.$$

In order to calculate the gain from the polarization, we use Maxwell's equations to get the wave equation,[2] along with Eqs. (1) and (7), to find the gain

$$G=(-\nu/\epsilon_0c)(S/E).$$

G is defined so that

$$I=I_0e^{Gz},$$

where I is the intensity of the traveling wave. From Eq. (7) we see that

$$S=\frac{-|\mathcal{P}_{ab}|^2}{\hbar}E\frac{w_{ab}}{\nu}\gamma_{ab}\int\frac{[n_a(\vec{v})-n_b(\vec{v}-c\epsilon\hat{k})]d\vec{v}}{\Omega^2+\bar{\gamma}^2},$$

where

$$\bar{\gamma}\equiv\gamma_{ab}(1+8\alpha)^{1/2}.$$

For a Maxwellian velocity distribution,

$$n_a(\vec{v})=\frac{N_a}{\pi^{3/2}}\exp\left(-\frac{c^2\vec{\beta}^2}{u^2}\right)\frac{cd\vec{\beta}}{u},$$

where N_a is the total number of atoms per cm³ in state a, and u is the most probable speed, so that

$$G=\frac{\nu}{\epsilon_0c}\frac{|\mathcal{P}_{ab}|^2}{\hbar}\frac{w_{ab}}{\nu}\gamma_{ab}\left(\frac{N_a}{\sqrt{\pi}}\frac{c}{u}\int\frac{\exp[-(c\beta_z/u)^2]d\beta_z}{[\nu(1-\beta_z+\epsilon/2)-w_{ab}]^2+\bar{\gamma}^2}\;\frac{-N_b}{\sqrt{\pi}}\frac{c}{u}\int\frac{\exp\{-[c(\beta_z-\epsilon)/u]^2\}d\beta_z}{[\nu(1-\beta_z+\epsilon/2)-w_{ab}]^2+\bar{\gamma}^2}\right).$$

In general, the expression for G must be integrated numerically. However, in the Doppler limit, $\bar{\gamma}/Ku\ll 1$, we have

$$G\simeq\frac{|\mathcal{P}_{ab}|^2}{\epsilon_0\hbar}\frac{w_{ab}\gamma_{ab}}{\bar{\gamma}}\frac{\sqrt{\pi}}{u\nu}(N_a\exp\{-[(c/u\nu)(\nu-w_{ab}+\nu\epsilon/2)]^2\}-N_b\exp\{-[(c/u\nu)(\nu-w_{ab}-\nu\epsilon/2)]^2\})$$
$$\equiv\frac{|\mathcal{P}_{ab}|^2}{\epsilon_0\hbar}\frac{w_{ab}}{(1+8\alpha)^{1/2}}\frac{\sqrt{\pi}}{u\nu}\{N_a\exp[-(x+a)^2]-N_b\exp[-(x-a)^2]\}, \tag{8}$$

where

$$x\equiv(\nu-w_{ab})/Ku$$

and

$$a\equiv\nu\epsilon/2Ku.$$

Figure 1 shows the individual terms in the parentheses of Eq. (8) for $N_a=N_b$ and $a=0.1$. There is positive gain for all $x<0$.

For an atom of infinite mass, the two curves of Fig. 1 would be centered on $x=0$; the maximum of the gain due to the atoms in state a and the loss due to the atoms in state b would both occur at $\nu=w_{ab}$. For finite mass, the curves are displaced from $x=0$. The $N_b\exp[-(x-a)^2]$ curve is displaced to the right because the atom gains a small amount of kinetic energy at the absorption. This energy must be supplied by the incoming photon whose frequency is thus higher than ω_{ab}. The $N_a\exp[-(x+a)^2]$ curve is displaced to the left because the energy

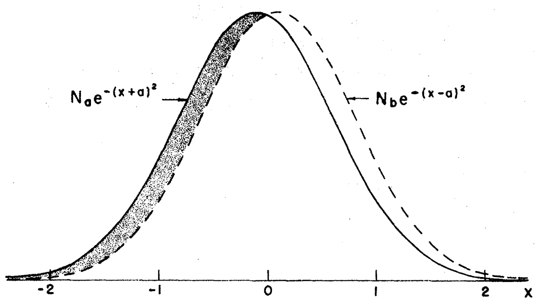

FIG. 1. $N_a \exp[-(x+a)^2]$ and $N_b \exp[-(x-a)^2]$ in arbitrary units for $a=0.1$, $N_a = N_b$. $G(x)$ is positive over the shaded region.

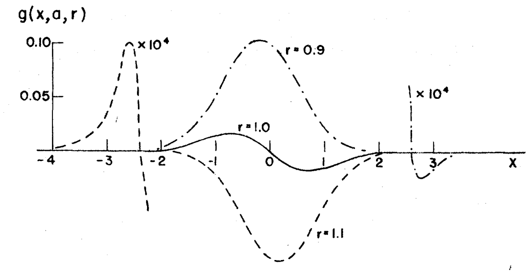

FIG. 3. $g(x,a,r)$ for $a=0.01$, $r=0.9$, 1.0, and 1.1. For r near 1, there is still substantial positive gain even for $a=0.01$. The curves are $r=0.9$ and 1.1 approaching the usual emission and absorption shapes, although their maxima are displaced from $x=0$. The positive gain part of the $r=1.1$ curve is much smaller than in Fig. 2, but is still present in the wing of the absorption curve.

of the stimulated photon plus the extra kinetic energy of the atom must add up to the transition energy, so that ν is less than w_{ab}.

Equation (8) can be written

$$G=\frac{|\mathcal{P}_{ab}|^2}{\epsilon_0 \hbar}\frac{w_{ab}}{u\nu}\frac{\sqrt{\pi}}{(1+8\alpha)^{1/2}}N_a g(x,a,r),$$

where

$$g(x,a,r)\equiv \exp[-(x+a)^2]-r\exp[-(x-a)^2],$$

$$r\equiv N_b/N_a .$$

If $r \geq 1$, there is no population inversion.

Figure 2 shows $g(x,a,r)$ as a function of x for $a=0.1$ and $r=0.9$, 1.0, and 1.1. Only the curve for $r=1.0$ is antisymmetric about both axes. As r grows above 1.0 (as N_b/N_a grows), the portion of the curve below the x axis, the portion for which absorption occurs, increases while the positive gain portion decreases. For $r<1 (N_a > N_b)$, the positive gain portion of the curve is greater than the absorption part. Here, for $a=0.1$ and $r=0.9$,

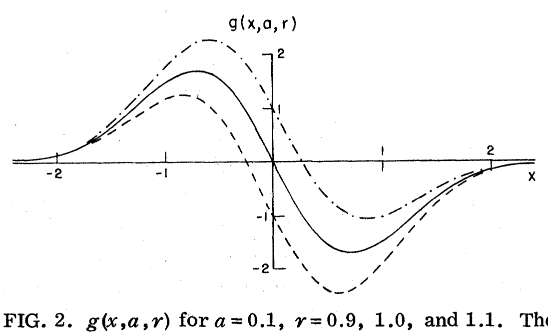

FIG. 2. $g(x,a,r)$ for $a=0.1$, $r=0.9$, 1.0, and 1.1. The curves are asymmetrical unless $r=1$. The region of positive g decreases as r increases (as the inversion decreases).

the gain part of the curve is quite shifted from $x=0$.

Figure 3 shows $g(x,a,r)$ for $a=0.01$, $r=0.9$, 1.0, and 1.1. Here, for a smaller value of a, the curves are approaching the usual emission and absorption curves except for r near 1.0. However, the curves are not exactly symmetrical about the ordinate and their maxima are shifted from $x=0$. For $r=1.1$, the positive gain portion of g is small. However, it is possible that the other factors in G could be made large enough for G to be substantial even for r as large as 1.1 and $a=0.01$.

The curves for $r=1$ for all a exhibit as much area above the x axis, of positive gain, as below. If a is smaller than about 0.1, the exponentials in $g(x,a,r)$ can be expanded, and one finds that

$$g(x,a,r)\simeq -4axe^{-x^2}.$$

Setting dg/dx equal to zero, we find the maximum for $r=1$ is always at $x_m = 1/\sqrt{2}$ and that $g(x_m, a, r=1) = 1.716a$. Thus, the size of the effect is directly proportional to a for $r=1$.

Another effect of recoil, one which is of importance to precision measurements, is the fact that the central resonance is shifted away from $x=0 (\nu = w_{ab})$ for $r\neq 1$. The shift can be found by taking the derivative of $g(x,a,r)$; for $r<1$, the maximum occurs at

$$x_m=\frac{1-r-[(1-r)^2+8a^2(1+r)^2]^{1/2}}{4a(1+r)}.$$

If

$$1-r \gg a,$$

$$x_m \simeq -a(1+r)/(1-r).$$

For a 6000 Å transition in H at room temperature,

a is about 2×10^{-4}, so that for $r=0.9$, $x_m=3.8\times 10^{-3}$; for $Ku/2\pi=3500$ MHz, $\nu-w_{ab}=1.3$ MHz. For $r\to 0$ $(N_a/N_b\to\infty)$, $x_m\to 2\times 10^{-4}$, and $\nu-w_{ab}\to 700$ KHz.

I have shown that there can be gain with zero or negative inversion density due to recoil effects and that recoil can affect precision frequency measurements in the specified way.

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