Quantum-Mechanical Fluctuations of the Resonance-Radiation Force

Richard J. Cook
University of California, Lawrence Livermore Laboratory, Livermore, California 94550
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The influence on atomic motion of quantum-mechanical fluctuations of the resonance-radiation force is investigated theoretically. Fluctuations due to both induced and spontaneous absorption-emission processes give rise to diffusion of atomic momentum, here described by a Fokker-Planck equation. It is shown that quantum-mechanical fluctuations of the radiation force place a lower bound on the temperature achievable by radiation cooling, inhibit cooling in a strong standing wave, and lead to finite, often short, confinement times for atoms in radiation traps.

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The importance of quantum-mechanical fluctuations of the radiation force in determining the motion of an atom in an electromagnetic wave was first emphasized by Einstein in 1917.[1] In this early work, Einstein showed that fluctuations due to both spontaneous and induced absorption-emission processes were necessary to account for the Maxwellian distribution of atomic velocity in thermal equilibrium. In recent years, a number of authors have proposed methods for trapping,[2,3] deflecting,[4,5] and cooling[3,6,7] atoms by use of the resonant light forces in tunable laser radiation. Although the fluctuations due to random recoils accompanying spontaneous emission (spontaneous fluctuations) have often been considered in these proposals, the fluctuations associated with induced absorption-emission processes (induced fluctuations) have usually been ignored. Only recently has it been suggested that induced fluctuations might be of some importance in cooling and trapping experiments.[8]

The purpose of this Letter is to point out that induced fluctuations can strongly influence atomic motion in resonant radiation and, in particular, that induced fluctuations place a lower bound on the temperature achievable by radiation cooling and lead to finite, often short, confinement times for atoms in radiation traps.

In strong coherent radiation, induced atomic processes are correctly described by interaction with a classical field. The Heisenberg equations for one-dimensional motion of an atom of mass M in a classically prescribed linearly polarized electromagnetic wave $\vec{E}(x,t)=\hat{\epsilon}E(x,t)$, in the dipole approximation, are

$$\dot{\hat{R}}=\hat{P}/M, \tag{1}$$

$$\dot{\hat{P}}=\hat{\mu}\partial E(\hat{R},t)/\partial x, \tag{2}$$

where $\hat{\mu}$ is the component of the electric dipole moment operator in direction $\hat{\epsilon}$. If the atomic wave packet is small compared to the distance over which $\partial E(x,t)/\partial x$ changes by a significant amount, the operator $\hat{R}$ in (2) may be replaced by its expectation value $R=\langle\hat{R}\rangle$, since any matrix element involving $\partial E(\hat{R},t)/\partial x$ is only negligibly affected by this replacement when the wave packet is small.

I consider the motion of a two-level atom with internal states $|1\rangle$ and $|2\rangle$ of energy E_1 and E_2, respectively, in a monochromatic field $E(x,t)=\mathcal{E}(x)\cos[\theta(x)+\omega t]$ with arbitrary amplitude $\mathcal{E}(x)$ and phase $\theta(x)$. In terms of atomic operators $\hat{S}=|1\rangle\langle 2|$ and $\hat{S}^\dagger=|2\rangle\langle 1|$, the dipole operator takes the form $\hat{\mu}=\mu(\hat{S}+\hat{S}^\dagger)$, where $\mu=\langle 1|\hat{\mu}|2\rangle$ is the transition dipole moment, taken here to be real. In the Heisenberg picture, operators $\hat{S}$ and $\hat{S}^\dagger$ are rapidly varying functions of time. It is more convenient to work with slowly varying operators $\hat{\sigma}$ and $\hat{\sigma}^\dagger$ defined by relations $\hat{S}=\hat{\sigma}\exp[-i(\theta+\omega t)]$ and $\hat{S}^\dagger=\hat{\sigma}^\dagger\exp[i(\theta+\omega t)]$, respectively. Upon substituting the above expressions for $E(x,t)$ and $\hat{\mu}$ into (2), and discarding inessential terms that oscillate at twice the optical frequency (rotating wave approximation), the equation for atomic momentum becomes

$$\dot{\hat{P}}=\frac{\hbar}{2}\left(\frac{\partial\Omega}{\partial x}\hat{\sigma}_1+\Omega\frac{\partial\theta}{\partial x}\hat{\sigma}_2\right), \tag{3}$$

where $\hat{\sigma}_1=\hat{\sigma}+\hat{\sigma}^\dagger$, $\hat{\sigma}_2=i(\hat{\sigma}-\hat{\sigma}^\dagger)$, and $\Omega=\mu\mathcal{E}/\hbar$ is the on-resonance Rabi flopping frequency of the two-level atom.

It is well known that the expectation values of operators $\hat{\sigma}_1$, $\hat{\sigma}_2$, and $\hat{\sigma}_3=\hat{\sigma}^\dagger\hat{\sigma}-\hat{\sigma}\hat{\sigma}^\dagger$, namely, $u=\langle\hat{\sigma}_1\rangle$, $v=\langle\hat{\sigma}_2\rangle$, and $w=\langle\hat{\sigma}_3\rangle$, are the components of the Bloch vector; and if the atom experiences relaxation due to spontaneous emission, these compo-

nents satisfy the optical Bloch equations[9]

$$\begin{aligned}\dot{u} &= (\Delta + \dot{\theta})v - \tfrac{1}{2}Au,\\ \dot{v} &= -(\Delta + \dot{\theta})u + \Omega w - \tfrac{1}{2}Av,\\ \dot{w} &= -\Omega v - A(w+1),\end{aligned} \tag{4}$$

where $\Delta = \omega - \omega_0$ is the detuning frequency [$\omega_0 = (E_2 - E_1)/\hbar$] and A is the Einstein spontaneous-emission coefficient. The expectation value of (3) is the mean radiation force acting on the atom,

$$F = \frac{\hbar}{2}\left(\frac{\partial\Omega}{\partial x}u + \Omega\frac{\partial\theta}{\partial x}v\right). \tag{5}$$

Equations (4) and (5) were recently derived with a slightly different approach,[10] and were applied to a number of problems of current interest.

Equation (3) indicates that the force $\hat{F} = \frac{1}{2}\hbar[(\partial\Omega/\partial x)\hat{\sigma}_1 + \Omega(\partial\theta/\partial x)\hat{\sigma}_2]$ should be regarded as an operator. With use of the fact that operators $\hat{\sigma}_1$ and $\hat{\sigma}_2$ satisfy the anticommutation relations $\hat{\sigma}_i\hat{\sigma}_j + \hat{\sigma}_j\hat{\sigma}_i = 2\delta_{ij}$, it is readily shown that the mean square force has the form

$$\langle\hat{F}^2\rangle = \frac{\hbar^2}{4}\left[\left(\frac{\partial\Omega}{\partial x}\right)^2 + \Omega^2\left(\frac{\partial\theta}{\partial x}\right)^2\right], \tag{6}$$

which is independent of the state of the atom. If we compare (6) with (5) and note that u and v are constrained only by the relation $u^2 + v^2 \leq 1$, it is easy to see that the rms fluctuation of the radiation force, $(\langle\hat{F}^2\rangle - F^2)^{1/2}$, can exceed the mean force F. This is a clear indication that quantum fluctuations of the radiation force will have a significant influence on atomic motion.

The motion of an atom under the influence of a fluctuating force is described by the Fokker-Planck equation. If in time Δt the force gives rise to a mean increment of momentum $\langle\Delta P\rangle$ and a mean square fluctuation of momentum about the mean increment $\langle(\Delta P - \langle\Delta P\rangle)^2\rangle$, if the limits

$$\lim_{\Delta t\to 0}\langle\Delta P\rangle/\Delta t = F \tag{7}$$

and

$$\lim_{\Delta t\to 0}\langle(\Delta P - \langle\Delta P\rangle)^2\rangle/\Delta t = 2D \tag{8}$$

exist, and if all other moments per unit time, e.g., $\langle(\Delta x - \langle\Delta x\rangle)(\Delta P - \langle\Delta P\rangle)\rangle/\Delta t$, vanish in this limit, the Fokker-Planck equation takes the form[11]

$$\frac{\partial f}{\partial t} = -\frac{P}{M}\frac{\partial f}{\partial x} - \frac{\partial}{\partial P}(Ff) + \frac{\partial^2}{\partial P^2}(Df), \tag{9}$$

where $f(x, P)$ is the distribution function in phase space.[12]

The limit (7) is clearly the mean radiation force of Eq. (5). To calculate D we proceed as follows. With $\hat{F}$ defined as the right-hand side of (3), the increment of momentum is

$$\Delta\hat{P} = \int_{t_0}^{t}\hat{F}(s)\,ds,$$

the mean increment is

$$\langle\Delta\hat{P}\rangle = \int_{t_0}^{t}F(s)\,ds,$$

and

$$\langle(\Delta\hat{P} - \langle\Delta\hat{P}\rangle)^2\rangle = \int_{t_0}^{t}\int_{t_0}^{t}ds_1\,ds_2[\langle\hat{F}(s_1)\hat{F}(s_2)\rangle - F(s_1)F(s_2)]. \tag{10}$$

The correlation time of fluctuations of the force $\hat{F}$ is essentially the correlation time of fluctuations of the dipole moment $\hat{\mu}$, which is known to be on the order of the natural lifetime $\tau_n = 1/A$. I consider the case in which the amplitude Ω and phase θ of the applied field (at the moving atom) are nearly constant over a natural lifetime. In this case, the field-dependent factors in the force (3) can be taken outside of the integrals in (10), and for $\Delta t = t - t_0$ somewhat larger than a correlation time, Eq. (10) yields

$$\begin{aligned}2D_I &\approx \frac{\langle(\Delta\hat{P} - \langle\Delta\hat{P}\rangle)^2\rangle}{\Delta t} \approx \int_{-\infty}^{\infty}ds[\langle\hat{F}(0)\hat{F}(s)\rangle - F(0)F(s)]\\ &= \frac{\hbar^2}{4}\left[\left(\frac{\partial\Omega}{\partial x}\right)^2\int_{-\infty}^{\infty}U_1(s)ds + \Omega^2\left(\frac{\partial\theta}{\partial x}\right)^2\int_{-\infty}^{\infty}V_2(s)ds + \Omega\frac{\partial\Omega}{\partial x}\frac{\partial\theta}{\partial x}\int_{-\infty}^{\infty}[V_1(s) + U_2(s)]ds\right],\end{aligned} \tag{11}$$

where

$$\begin{aligned}U_1(t) &= \langle\hat{\sigma}_1(0)\hat{\sigma}_1(t)\rangle - u(0)u(t),\\ V_1(t) &= \langle\hat{\sigma}_1(0)\hat{\sigma}_2(t)\rangle - u(0)v(t),\\ U_2(t) &= \langle\hat{\sigma}_2(0)\hat{\sigma}_1(t)\rangle - v(0)u(t),\\ V_2(t) &- \langle\hat{\sigma}_2(0)\hat{\sigma}_2(t)\rangle - v(0)v(t),\end{aligned} \tag{12}$$

I have set $t_0 = 0$ for simplicity, and the subscript on D_I indicates that this is the contribution from induced fluctuations.

The theory of dipole correlation functions, such as (12), is by now fairly standard, having been developed in connection with the problem of reso-

nance fluorescence by Mollow[13] and others.[14,15] According to this theory, if we introduce two additional correlation functions

$$W_1(t) = \langle \sigma_1(0)\sigma_3(t)\rangle - u(0)w(t),$$
$$W_2(t) = \langle \sigma_2(0)\sigma_3(t)\rangle - v(0)w(t), \tag{13}$$

then each of the sets of correlation functions U_i V_i, $W_i (i = 1, 2)$ satisfy homogeneous Bloch equations

$$\dot{U}_i = (\Delta + \dot{\theta})V_i - \tfrac{1}{2}AU_i,$$
$$\dot{V}_i = -(\Delta + \dot{\theta})U_i + \Omega W_i - \tfrac{1}{2}AV_i, \tag{14}$$
$$\dot{W}_i = -\Omega V_i - AW_i.$$

The remainder of the calculation is given in outline. First, it is clear from (11) that only the real parts of U_i, V_i, and W_i are significant. The real parts are even functions of t, and so the integrals in (11) are replaced by twice the integrals from 0 to ∞. Next, integrating Eqs. (14) from 0 to ∞, we obtain a set of algebraic equations for $\int_0^\infty U_i\,ds$, $\int_0^\infty V_i\,ds$, and $\int_0^\infty W_i\,ds$ involving initial conditions $U_i(0)$, $V_i(0)$, and $W_i(0)$. The initial conditions are obtained from (12) and (13) by use of $\hat{\sigma}_i\hat{\sigma}_j = \delta_{ij} + i\epsilon_{ijk}\hat{\sigma}_k$ and the steady-state solution of the Bloch equations (4). Finally, the algebraic equations for $\int_0^\infty U_i\,ds$, etc., are solved, and the results are used in (11) to obtain

$$D_I = \frac{\hbar^2}{2}\left[\left(\frac{\partial\Omega}{\partial x}\right)^2 \{A[(4\Delta_e^2 + A^2)^2 - 8\Delta_e^2\Omega^2] + 2A^{-1}\Omega^2(3A^4 + 6A^2\Omega^2 + 4\Omega^4)\}\right.$$
$$-\Omega\frac{\partial\Omega}{\partial x}\frac{\partial\theta}{\partial x}[16\Delta_e\Omega^2(2A^2 + \Omega^2)] + \Omega^2\left(\frac{\partial\theta}{\partial x}\right)^2\left.[2\Omega^2A(12\Delta_e^2 - A^2 + 2\Omega^2) + A(4\Delta_e^2 + A^2)^2]\right]$$
$$\times(4\Delta_e^2 + A^2 + 2\Omega^2)^{-3}, \tag{15}$$

where $\Delta_e = \Delta + \dot{\theta}$ is an effective detuning. In a weak field ($\Omega \to 0$), (15) becomes

$$D_I = \frac{\hbar^2 A}{2(4\Delta_e^2 + A^2)}\left[\left(\frac{\partial\Omega}{\partial x}\right)^2 + \Omega^2\left(\frac{\partial\theta}{\partial x}\right)^2\right], \tag{16}$$

and in a strong field ($\Omega \to \infty$)

$$D_I = \frac{\hbar^2}{2A}\left[\left(\frac{\partial\Omega}{\partial x}\right)^2 + \frac{A^2}{2}\left(\frac{\partial\theta}{\partial x}\right)^2\right], \tag{17}$$

where the second term in (17) is usually negligible when $\partial\Omega/\partial x \neq 0$.

The term containing D in the Fokker-Planck equation represents diffusion of momentum. In addition to the coefficient of induced diffusion D_I, there is a well-known contribution D_S to the total diffusion coefficient $D = D_I + D_S$, resulting from random recoils associated with spontaneous emission. If the atom's dipole moment is transverse to the x direction, it is readily shown that the coefficient of spontaneous diffusion is

$$D_S = \tfrac{1}{5}A(\hbar\omega_0/c)^2P_2 = \tfrac{1}{10}A(\hbar\omega_0/c)^2(w+1)$$
$$= A(\hbar\omega_0\Omega)^2/5c^2(4\Delta_e^2 + A^2 + 2\Omega^2),$$

where $P_2 = \frac{1}{2}(w+1)$ is the probability that the upper atomic level is occupied, and I have taken the steady-state value of w to obtain the final form. In a strong field D_S saturates to the value $D_S = \frac{1}{10}A(\hbar\omega_0/c)^2$.

I now consider a few simple examples to illustrate the above theory. In a strong resonant traveling wave $E(x, t) = \mathcal{E}\cos(kx - \omega_0 t)$, the saturated radiation force[10] is $F = \frac{1}{2}A\hbar k$ ($k = \omega_0/c$); the coefficient of induced diffusion, Eq. (17), is $D_I = \frac{1}{4}A(\hbar k)^2$; the spontaneous coefficient is $D_S = \frac{1}{10}A(\hbar k)^2$; and the total momentum-diffusion coefficient $D = 7A(\hbar k)^2/20$ is 3.5 times that which might have been expected on the basis of spontaneous recoils alone.

In a strong standing wave $E(x, t) = 2\mathcal{E}_0\cos kx \times\cos\omega t$ ($\Omega = 2\Omega_0\cos kx$, $\Omega_0 = \mu\mathcal{E}_0/\hbar$, $\theta = 0$), the dipole force acting on a slowly moving atom[10] is $F = (-\hbar\Delta\partial\Omega^2/\partial x)/(4\Delta^2 + A^2 + 2\Omega^2)$, the induced diffusion coefficient, Eq. (17), is $D_I = \frac{1}{2}\hbar^2(\partial\Omega/\partial x)^2/A \sim \Omega_0^2(\hbar k)^2/A$, and D_S is negligible compared to D_I. On resonance ($\Delta = 0$) the dipole force vanishes and atomic motion is dominated by induced momentum diffusion. In a standing wave, induced diffusion results from a splitting of the atomic trajectory after each spontaneous transition to the ground state, and is closely related to the optical Stern-Gerlach effect.[16,17]

The cooling or heating of an atomic vapor by resonant radiation is calculated as follows. Generally the radiation force consists of a part $F' = -\partial V(x)/\partial x$ derivable from a potential and a part F'' not derivable from a potential. Multiplying the Fokker-Planck equation, (9), by $P^2/2M + V(x)$, integrating over phase space, and performing some integrations by parts (assuming $f = 0$ at $|x| = \infty$ and $|P| = \infty$), we obtain the relation

$$\dot{E} = M^{-1}\iint (D + PF'')f\,dx\,dP \tag{18}$$

for the rate of change of the mean translational

energy $E = \int\int (P^2/M + V) f\, dx\, dp$ of the atom. For example, in a weak ($\Omega \ll A$) standing wave tuned below resonance by the amount $\Delta = -\frac{1}{2}A$, the total diffusion coefficient is $D \approx 7\Omega_0^2(\hbar k)^2/10A$, the radiation damping force,[10] for small atomic velocity, is $F'' = -(2\Omega_0^2\hbar k^2/A^2)v$, and (18) states that the energy E dissipates until $\langle D + PF'' \rangle_{av} = 0$ or $\frac{1}{2}M\langle v^2 \rangle_{av} \approx 7\hbar A/40$. For $A = 10^8$ s^{-1} this corresponds to a temperature $T \approx 10^{-4}$ K. However, as the intensity increases, D increases without bound, because of induced diffusion, while the strength of the dissipative force F'' is certainly bounded by $\frac{1}{2}A\hbar k$. Therefore induced fluctuations inhibit cooling in a strong standing wave.

Finally, consider radiation trapping of atoms by the dipole force in a strong field ($\Omega \gg A$) of amplitude $\Omega(x) = \Omega_0 \exp(-x^2/w_0)$, e.g., transverse trapping in a Gaussian laser beam. The potential energy of the dipole force,[10] $V = \frac{1}{2}\hbar\Delta Ln(1 + \Omega^2/2\Delta^2)$, assumes its minimum value $V_{min} = -0.28\hbar\Omega_0$ for $\Delta = -0.35\Omega_0$. Since no dissipative force acts on the atom, (18) states that $\dot{E} = \overline{D}/M$ and (17) yields the estimate $D = D_I + D_S \approx \frac{1}{10}\hbar^2\Omega_0^2/Aw_0^2 + \frac{1}{10}A(\hbar k)^2$ for the average D in the well. A trapped atom gains energy from fluctuations and escapes from the well in a time of order

$$\Delta t = -V_{min}/\dot{E} = 2.8\hbar\Omega_0 M/(A\hbar^2k^2 + \hbar^2\Omega_0^2/Aw_0^2).$$

For a sodium atom in a Gaussian beam of radius $w_0 = 10$ μm and power 50 mW, tuned to the $3^2S_{1/2} \rightarrow 3^2P_{3/2}$ transition, the confinement time is $\Delta t \approx 10^{-4}$ s. This is about two orders of magnitude less than the value obtained if only spontaneous fluctuations are considered.

The above examples indicate that quantum-mechanical fluctuations of the resonance-radiation force will be an important consideration in the design of experiments to trap or cool atoms and molecules.

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