Forces on three-level atoms including coherent population trapping

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We present a calculation of the force on a stationary three-level atom excited by a nearly resonant Raman light field, which may be composed of an arbitrary combination of standing- and traveling-wave fields. The effects of the ground-state coherences are explicitly included and are shown to play a crucial role in the nature of the force on the atom. We show that the force contains terms that vary on length scales both shorter and longer than the optical wavelength and that the magnitude of these terms can be made arbitrarily large.

Recently there has been considerable interest in the force due to the interaction between three-level atoms and nearly resonant optical fields.[1-4] It has been suggested that three-level forces may explain some of the differences between observations on real trapped atoms and predictions for two-level atoms[1,5] (TLA) or incoherent processes in multilevel atoms. The three-level Λ system is of particular interest because of the effects of coherent population trapping. Recently, velocity-selective coherent population trapping has been used to cool atoms below the single-photon recoil limit.[6] In this Letter we calculate the net average force F on a stationary atom in a Λ configuration excited by two fields $\mathbf{E}_1$ and $\mathbf{E}_2$ each in an arbitrary combination of standing and traveling waves. We calculate F by solving the optical Bloch equations (OBE's) in the steady-state limit. For pure traveling waves, F is spatially invariant, and the direction of the spontaneous force F_{sp} depends only on the wave vectors of $\mathbf{E}_1$ and $\mathbf{E}_2$. For pure standing waves, the solutions predict that the stimulated force can have spatial components that vary on scales both shorter and longer than the optical wavelength (λ_{opt}).[7] For experimentally attainable parameters, the size of these force terms can be substantially larger than the maximum F_{sp} on a TLA. We interpret our results in a dressed atom picture and show that many important aspects of F can be attributed to the effects of the ground-state coherences.

We consider the Λ system shown in Fig. 1 (left), which interacts with two fields $\mathbf{E}_1 = |E_1| \times \exp[i(\omega_1 t + \phi_1)]$ and $\mathbf{E}_2 = |E_2|\exp[i(\omega_2 t + \phi_2)]$ that have frequencies ω_1 and ω_2, where the Λ system is closed and $\Gamma = 2\gamma_{ea} = 2\gamma_{eb}$. We confine ourselves to the case where $\mathbf{E}_1$ ($\mathbf{E}_2$) interacts only with the $|a\rangle \rightarrow |e\rangle$ ($|b\rangle \rightarrow |e\rangle$) transition. We derive F using the Lorentz expression $F_j = \mathbf{P} \cdot \nabla_j \mathbf{E}$, where $j = x, y, z$. $\mathbf{P} = \mathbf{P}(\mathbf{E}_1, \mathbf{E}_2) = tr(\hat{\rho}\mu)$ is the polarization induced in the atom, where $\hat{\rho}$ and μ are the density-matrix and vector dipole operators, respectively. Thus we solve for F by solving the OBE's[8] for the off-diagonal elements of $\hat{\rho}$ in the steady-state limit by using the rotating-wave approximation. However, the physical significance of F may be more easily interpreted in terms of states $|-\rangle$, $|+\rangle$, and $|e\rangle$ derived from $|a\rangle$, $|b\rangle$, and $|e\rangle$ by a unitary transformation R, where

$$R = \begin{bmatrix} \cos(\theta) & -\sin(\theta) & 0 \\ \sin(\theta) & \cos(\theta) & 0 \\ 0 & 0 & 1 \end{bmatrix}. \qquad (1)$$

Here θ is a measure of the relative strengths of $\mathbf{E}_1$ and $\mathbf{E}_2$, given by $g_1 = |(\mu_{ea} \cdot \mathbf{E}_1)/\hbar| = g_1(x) = g \sin\theta$, and $g_2 = |(\mu_{eb} \cdot \mathbf{E}_2)/\hbar| = g_2(x) = g\cos\theta$ are the Rabi frequencies, with $g = (g_1^2 + g_2^2)^{1/2}$.

The $|+\rangle$ and $|-\rangle$ states are the eigenstates of the atom field system in the absence of spontaneous emission and are sometimes referred to as the dressed states.[9] The OBE's can then be derived by applying R to all the matrices that describe the time evolution of the $|a\rangle$, $|b\rangle$, $|e\rangle$ system. In the dressed-state basis the Hamiltonian for the OBE's,[8] H_{TD}, is

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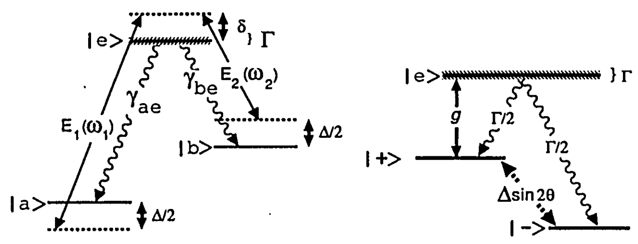

Fig. 1. Left: three-level Λ system in Raman excitation field. Right: coupling between the states of the Λ system in the dressed-state basis [see Eq. (2)].

$$H_{\mathrm{TD}} = \frac{\hbar}{2}\begin{bmatrix} \Delta\cos(2\theta) & \Delta\sin(2\theta) & 0 \\ \Delta\sin(2\theta) & -\Delta\cos(2\theta) & -g \\ 0 & -g & -2\delta \end{bmatrix}, \qquad (2)$$

where ω_a (ω_b) is the resonant frequency of the $|a\rangle \to |e\rangle$ ($|b\rangle \to |e\rangle$) transition. $\Delta = (\omega_1 - \omega_a) - (\omega_2 - \omega_b)$ is the differential detuning, and $\delta = 1/2[(\omega_1 - \omega_a) + (\omega_2 - \omega_b)]$ is the common detuning (see Fig. 1). The dressed-state source and decay matrices[8] can be obtained by a similar transformation.

Since the only important components of F are $P_{ea}(\mathbf{E}_1,\mathbf{E}_2) \cdot \nabla\mathbf{E}_1$ and $P_{eb}(\mathbf{E}_1,\mathbf{E}_2) \cdot \nabla\mathbf{E}_2$, F can be expressed in terms of the field gradients $\alpha_j = (1/g_j)(\delta g_j/\delta x)$, $\beta_j = (\delta\phi_j/\delta x)$,

$$F = [C_{\mathrm{sum}}(\alpha_1 + \alpha_2) + C_{\mathrm{dif}}(\alpha_1 - \alpha_2) + C_{\mathrm{sp}}(\beta_1 + \beta_2)], \qquad (3)$$

where $C_{\mathrm{sum}} = -4\Delta^2\delta C_o$, $C_{\mathrm{dif}} = -\Delta(g^2 - 2\Delta^2)C_o$, and $C_{\mathrm{sp}} = 2\Delta^2\Gamma C_o$. Here $C_o = 4\hbar g_1{}^2 g_2{}^2/D$ and $D = g^6 + 8\delta\Delta[(g_2{}^4 - g_1{}^4) + 2\Delta^2(g_1{}^2 - g_2{}^2)] + 4\Delta^2[g^2(4\delta^2 + \Gamma^2 + \Delta^2) - (g_1{}^4 - 4g_1{}^2g_2{}^2 + g_2{}^4)]$.[10] The α's are normalized gradients of the field amplitudes. The α terms are associated with stimulated processes[11] and are not proportional to Γ. C_{sum} and C_{dif} are derived from the different contributions of $\hat{\rho}$ to $\mathbf{P}$. C_{dif} is derived from P_{-e}, which is a weighted average of the components of P_{ae} that are 180° out of phase with P_{be}. This term is associated with ρ_{-e} and thus is related to the correlation between the populations in $|-\rangle$ and $|e\rangle$. In contrast, C_{sum}, which is associated only with ρ_{+e}, is derived from P_{+e}. P_{+e} is a weighted average of the components of P_{ae} that are in phase with P_{be},[9,12] and it is related to the correlation between the populations in $|+\rangle$ and $|e\rangle$. The $|+\rangle$ population also contributes a force term proportional to $(\beta_1 + \beta_2)$, which can be associated with spontaneous processes.[11] There is no $\beta_1 - \beta_2$ term.

We can gain some physical insight into F by considering C_{sum}, C_{dif}, and C_{sp}, which are determined by the correlations between the steady-state population distributions among the dressed states. We can estimate these correlation by considering the coupling (i.e., the population transfer rate) between the $|+\rangle$, $|-\rangle$, and $|e\rangle$ states.

Consider first the coupling between $|+\rangle$ and the other states. The Rabi flopping rate between the $|+\rangle$ and $|e\rangle$ states is given by g, and $\Gamma/2$ is the rate at which spontaneous emissions returns atoms from $|e\rangle$ to $|+\rangle$ (see Fig. 1, right). This is analogous to the coupling between the TLA states. The force on a TLA is $F_{\mathrm{TLA}} \sim \rho_{ee}[\Gamma\beta_o - 2(\omega - \omega_o)\alpha_o]$, where ω_o is the TLA resonant frequency. Similarly, the contribution to F associated with $|+\rangle$ can be written as $F_{\mathrm{sum}} = \hbar\rho_{ee}[\Gamma(\beta_1 + \beta_2) - 2\delta(\alpha_1 + \alpha_2)]$. Thus, just as there is no semiclassical stimulated force on a TLA when $\omega - \omega_o = 0$, in the Λ system, $C_{\mathrm{sum}} = 0$ if $\delta = 0$. Note that the C_{sum} force component will be zero if E_1 and E_2 have opposite gradients.

In contrast, the contribution to F associated with $|-\rangle$ does not have a simple TLA analogy. It is not directly proportional to the excited-state population ρ_{ee}, and it will be zero if the two fields have the same gradient. Consider the $|-\rangle$ and $|+\rangle$ to $|e\rangle$ couplings. Unlike $|+\rangle$, which is directly coupled to $|e\rangle$, $|-\rangle$ is never coupled directly to $|e\rangle$. Since $\gamma_{ea} = \gamma_{eb}$, if $\Delta = 0$, the only coupling is a source term that transfers population from ρ_{ee} to ρ_{--} at a rate $\Gamma/2$. Thus, independent of any other parameter, if $\Delta = 0$, an atom will be optically pumped into $|-\rangle$ and will remain there forever. This is why $|-\rangle$ is often referred to as the trapped or dark-resonance state.[13] In the steady state, then, there is no population in $|e\rangle$; therefore the off-diagonal matrix elements are all zero (i.e., $\rho_{-e} = 0 = \rho_{+e}$). Thus $F = 0$ independent of the field gradients (i.e., α and β) and of δ, g_1, and g_2.

If $\Delta \neq 0$, then there is a coupling between $|-\rangle$ and $|+\rangle$, given by an effective Rabi flopping rate $\Delta\sin(2\theta)$. The $|-\rangle$ state is no longer a trapped state, since atoms in $|-\rangle$ can precess into $|+\rangle$, which is in turn coupled to $|e\rangle$ by g. Thus $\rho_{ee} \neq 0$ and there can be a force associated with the population in the $|-\rangle$ state. In fact, F can be dominated by the C_{dif} term. For example, this occurs if $\delta = 0$.

If both fields are traveling waves, then $\alpha_j = 0$ and $F = C_{\mathrm{sp}}(\beta_1 + \beta_2)$, a purely spontaneous force. If the fields are counterpropagating, $F = \hbar(k_1 - k_2)g_1{}^2g_2{}^2\Delta^2\Gamma/D$. Surprisingly, the direction of F is determined entirely by $(k_1 - k_2)$. The atom is not necessarily pushed in the direction of propagation of the stronger field or the field nearer resonance. If $|k_1| = |k_2|$, then $F = 0$ is independent of Δ, δ, g_1, and g_2. This result can be understood by noting that when one of the transitions (say $|a\rangle \to |e\rangle$) is much more strongly driven than the other ($|E_1| \gg |E_2|$), almost all of the population accumulates in the ground state of the weakly driven transition, $|b\rangle$.

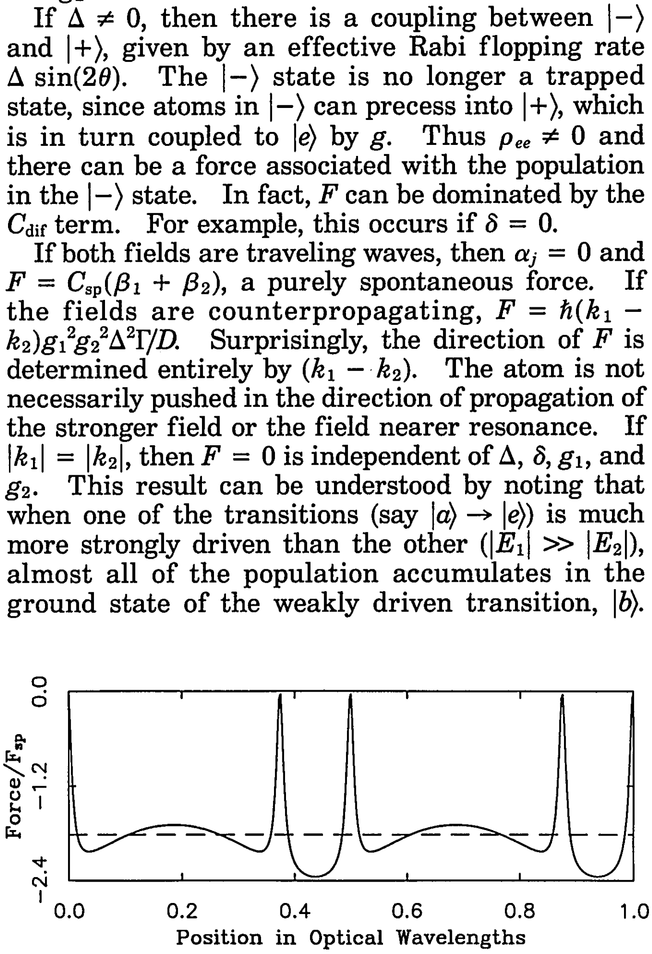

Fig. 2. Stimulated force for $\delta = 0$, $\Delta = g_o/2 = 4\Gamma$, and $\chi = \pi/4$. $F_{\mathrm{sp}} = \hbar\Gamma/\lambda_{\mathrm{opt}}$. The dashed line is the spatially averaged force. Note that the force is completely rectified (unipolar). For other choices of parameters, the force is not completely rectified, and there are potential minima associated with the sharp features.

Hence the $P_{ea} \to 0$ such that $P_{ea}\nabla \mathbf{E}_1 = -P_{eb}\nabla \mathbf{E}_2$ and $F = 0$. If g is increased while Δ is held fixed, then $C_{sp} \to 0$ since all the population will accumulate in $|-\rangle$. This is in striking contrast to F_{sp} for a TLA, which approaches $\hbar k\Gamma/2$.

Now consider the case where E_1 and E_2 are standing waves ($\beta_1 = 0 = \beta_2$) with equal maximum Rabi frequencies g_o. If $|k_1 - k_2| \ll |k_1 + k_2|$, then let $k = k_1$ and $k_2 x = (k_2 - k_1)x + k_1 x \approx \chi + kx$. Thus $g_1 = g_o \cos(kx)$ and $g_2 = g_o \cos(kx + \chi)$. Then

$$F = \frac{2\hbar k g_0^4}{D\{x,\chi\}}[\cos(\chi) + \cos(2kx + \chi)](F_{\mathrm{dif}} + F_{\mathrm{sum}}), \tag{4}$$

where $F_{\mathrm{sum}} = 4\Delta^2\delta \sin(2kx + \chi)$, $F_{\mathrm{dif}} = -\Delta \sin(\chi)(g^2 - 2\Delta^2)$, and χ is assumed constant over several λ_{opt}. Note that F can have a substantial nonzero average over λ_{opt}. The contribution to F from the out-of-phase component of $\mathbf{P}$, F_{dif}, is associated with $|-\rangle$ and has terms that do not change sign over λ_{opt}. For $\Delta^2 = g_0^2 \sin^2(\chi)/2$ this component is completely rectified. If $\delta = 0$, F is completely rectified (i.e., $|F|$ is positive definite[14]; see Fig. 2). The term associated with the real, in-phase components of $\mathbf{P}$, F_{sum}, is not completely rectified but can still have a nonzero spatial average over λ_{opt}. Thus both F_{dif} and F_{sum} have unbounded stimulated components that vary in space with a period $2/|k_1 - k_2|$.

It is possible to integrate Eq. (3) to form an expression for a pseudopotential that appears to become infinitely deep as $|k_1 - k_2| \to 0$. However, since F is approximately constant over a long distance, an atom will simply accelerate to a velocity for which these equations are no longer valid. In order to predict the motion of an atom accurately, the effects of nonconservative forces[15–17] as well as force fluctuations due to the interaction with the vacuum field[11,18] must be included.

Another remarkable feature of Eq. (3) is that F can have components that vary on a scale much shorter than λ_{opt}, even in the absence of saturation. The spacing between the features can be controlled by varying χ. Although Eq. (3) does not predict any limit for the narrowness of the features in F, the motion of an atom may not necessarily be accurately predicted. Eventually, motion due to F will produce a velocity that is not consistent either with the zero-velocity approximation or the assumption that the internal state of the atom is in equilibrium at a particular point. We also note that for sufficiently low velocities the atom will have a large de Broglie wavelength, and a fully quantum-mechanical treatment of atomic coordinates will be needed. Still, the possibility of such narrow resonances in the force may merit further investigation.

In sum, we have shown that for a stationary atom[19] a steady-state solution to the OBE's predicts that there can indeed be a finite force associated with the population in the antisymmetric ($|-\rangle$) ground state and that, moreover, this force component can dominate the total force on the atom.

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References

1. J. Javanainen, Phys. Rev. Lett. **64,** 519 (1990).
2. F. Mauri and E. Arimondo, in *Abstracts of 12th International Conference on Atomic Physics* (University Microfilms, Ann Arbor, Mich., 1990), p. IV 21.
3. S. Chang, B. M. Garraway, and V. G. Minogin, Opt. Commun. **77,** 19 (1991).
4. V. G. Minogin, M. A. Olshany, and S. U. Shulga, J. Opt. Soc. Am. B **6,** 2108 (1989).
5. N. P. Bigelow and M. G. Prentiss, Opt. Lett. **15,** 1479 (1991).
6. A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, Phys. Rev. Lett. **61,** 826 (1988).
7. M. S. Shahriar, P. R. Hemmer, N. P. Bigelow, and M. G. Prentiss, in *Quantum Electronics and Laser Science,* Vol. 11 of 1991 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1991), p. 118.
8. See, e.g., M. Sargent, M. Scully, and W. Lamb, eds., *Laser Physics* (Addison-Wesley, Reading, Mass., 1974), Chap. 7.
9. We note that a correspondence has been established between the dressed states and the normal modes of a coupled pendulum system. See P. R. Hemmer and M. G. Prentiss, J. Opt. Soc. Am. B **5,** 1613 (1988).
10. Note that D can depend on position, but it is always positive definite; consequently, the sign of F is determined entirely by the numerator.
11. J. P. Gordon and A. Ashkin, Phys. Rev. A **21,** 1606 (1980).
12. We note that ρ_{+e} contributes to both C_{dif} and C_{sum}, whereas ρ_{-e} contributes only to C_{dif}, and that it may be possible to obtain an increased appreciation of this correspondence by considering the analogy between the Raman system and a system of three-coupled pendulums, which is discussed in detail in Ref. 9.
13. H. J. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3,** 218 (1978).
14. We note the introduction of the term rectified in the context of the TLA. See A. P. Kazantsev and I. V. Krasov, JETP Lett. **46,** 420 (1987); V. S. Voitsekhovich, D. M. Danileiko, A. M. Negrijko, V. I. Romanenko, and L. P. Yatsenko, Zh. Tekh. Fiz. **58,** 1174 (1988); also recent TLA experiments of R. Grimm, Yu. B. Ovchinnikov, A. I. Sidorov, and V. S. Lethokov, Phys. Rev. Lett. **65,** 1415 (1990). See also Ref. 1.
15. A. Aspect, J. Dalibard, A. Heidmann, C. Salomon, and C. Cohen-Tannoudji, Phys. Rev. Lett. **57,** 1688 (1986).
16. J. Dalibard, C. Salomon, A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, in *Proceedings of Eleventh International Conference on Atomic Physics* (World Scientific, Singapore, 1989), p. 199.
17. S. Chu, D. S. Weiss, Y. Shevy, and P. J. Ungar, in *Proceedings of Eleventh International Conference on Atomic Physics* (World Scientific, Singapore, 1989), p. 636.
18. R. J. Cook, Phys. Rev. A **20,** 224 (1979).
19. We have calculated the first-order velocity dependence of F and find that the atomic motion can be strongly damped.