Nonlinear Mixing of Light-Pressure Forces in a Three-State Atom

Juha Javanainen
Department of Physics, University of Connecticut, Storrs, Connecticut 06269-3046
(Received 10 October 1989)

Nonlinear mixing of wave vectors present in the field driving a three-state atom may generate long-wavelength optical forces whose effect on the motion of the atom exceeds by orders of magnitude the influence of the forces familiar from two-state atomic models.

PACS numbers: 32.80.Pj, 42.50.Vk

Temperatures below the Doppler limit[1] predicted by two-state atomic models, supermolasses[2] which holds atoms much longer than the ordinary molasses, and inhomogeneities in the distribution of atoms in optical molasses[3] constitute prime examples of unexpected results obtained in recent experiments on laser cooling of atoms. A consensus seems to be forming that the abnormally low temperatures reflect the interplay between the multistate level structure of atoms and the spatially varying polarization direction of light,[4] but the spatial properties of the atom clouds remain unexplained.

To this end I would like to draw attention to the work of Kazantsev and Krasnov.[5] They investigate light-pressure forces of a bichromatic field acting on a two-state atom. Kazantsev and Krasnov point out that the nonlinear mixing of the forces corresponding to each individual light field may create long-wavelength components in the optical force that command a large influence on the motion of the atom. The present Letter is based on my realizing that *multistate systems* analogous to those abounding in the experiments[1-3] exhibit a similar "rectification." By analyzing a three-state model atom in various standing-wave fields, I present examples in which the variation of the kinetic energy of the atom traversing the interference pattern of the fields far exceeds the predictions from a two-state atomic model.

I consider a three-state atom with the Λ configuration of states (Fig. 1). Its two arms are driven by low-intensity fields $\mathbf{E}_{1,2}(\mathbf{r})e^{i\omega_{1,2}t}$+c.c. with the same linear polarization but possibly different detunings $\Delta_{1,2}$, defined positive if the laser is tuned above the respective resonance. For simplicity I assume that the dipole moment matrix element d and the spontaneous decay rate γ are the same for both arms of the Λ system, so that the linewidth (half width at half maximum) of both transitions $1\rightarrow 3$ and $2\rightarrow 3$ is taken to be γ as well. I omit the velocity dependence of the forces, and hence laser cooling. Furthermore, I work at low enough intensities that saturation of the excited state 3 is negligible, and leave out of my considerations the nonabsorbing two-photon resonance.

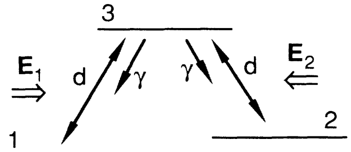

FIG. 1. Scheme of the three-state atom.

Under these conditions optical pumping first establishes a steady-state distribution of population among the states 1 and 2, and subsequently the dipole forces[6,7] of the fields acting on the transitions $1\rightarrow 3$ and $2\rightarrow 3$ simply add. Taking the detunings to be large enough that only the stimulated component of the force has to be retained, I obtain the force on the atom

$$\mathbf{F}=-\frac{\hbar(\Delta_1 R_2\nabla R_1+\Delta_2 R_1\nabla R_2)}{2\gamma(R_1+R_2)}. \tag{1}$$

Here

$$R_i(\mathbf{r})=\frac{2d^2|\mathbf{E}_i|^2(\mathbf{r})\gamma}{\hbar^2(\Delta_i^2+\gamma^2)} \tag{2}$$

are the light-induced rates for the transitions $i\rightarrow 3$. It should be recognized that, while the nonlinearity of (1) with respect to field intensity superficially resembles saturation of a two-state system, here the nonlinearity arises from the equilibrium populations of states 1 and 2 as dictated by optical pumping, $R_2/(R_1+R_2)$ and $R_1/(R_1+R_2)$.

I study Eq. (1) in the case when the fields acting on the atom are both standing waves, $\mathbf{E}_i(\mathbf{r})=\mathbf{e}\cos(\mathbf{k}_i\cdot\mathbf{r})$. To facilitate easy appreciation of the results I introduce the scales of force and potential energy,

$$F_0=\frac{\hbar|\Delta_1 k_1+\Delta_2 k_2|R_1(0)R_2(0)}{2\gamma[R_1(0)+R_2(0)]}, \tag{3}$$

$$U_0=\frac{2F_0}{k_1+k_2}. \tag{4}$$

If the light beams are parallel and have the same wavelength, $\mathbf{k}_1=\mathbf{k}_2\equiv\mathbf{k}$, then the force is in the direction of $\pm\mathbf{k}$ and its amplitude as a function of position is $F_0\sin(2\mathbf{k}\cdot\mathbf{r})$. By further setting $R_1(0)=R_2(0)$, $\Delta_1=\Delta_2\equiv\Delta$ in (3), $F_0\sin(2\mathbf{k}\cdot\mathbf{r})$ also gives a good approximation of the force on the two-state atom obtained by removing one leg of the Λ.[6,7] The force varies as a function of position with the period of half the wavelength, suggestive of trapping of atoms to the nodes or antinodes of the field. However, at least within the two-state theory no trapping takes place because the corresponding maximum variation of the potential energy, given by U_0, turns out to be smaller than or at most comparable to the minimum kinetic energy of the atoms attainable with laser cooling.

I now assume that the wave vectors of the fields are parallel to, say, the z axis, but that their absolute values are different; $\delta k = k_2 - k_1 \neq 0$. The force has Fourier components at $\pm 2k_{1,2}$ because the field intensity has. But, as a result of the nonlinear mixing, the force may be expected to have Fourier components also at the wave numbers $\pm 2\delta k$.

To demonstrate the size of such components I plot in Fig. 2 the potential energy of the force field in units of U_0 as a function of the dimensionless variable $|\delta k| z/\pi$. k_1 and k_2 are chosen commensurate in such a way that the range of z in Fig. 2 spans 64 spatial periods of field 1 and 63 periods of field 2. I set $\Delta_1 = \Delta_2 > 0$,[8] and the ratio of the pumping rates is $r \equiv R_2(0)/R_1(0) = 2$. By its very definition, one unit of the vertical axis represents the largest peak-to-peak variation of the potential energy attainable with $\delta k = 0$. The variation in the figure is much larger.

The expansion of (1) into a power series of the variable $1-r$ and a subsequent numerical Fourier analysis show that for $\Delta_1 = \Delta_2$, $r \rightarrow 1$, and in the limit when the difference of the wave vectors δk is small compared to their average $k \equiv (k_1 + k_2)/2$, the long-wavelength component in the force is given by

$$F_L(z) = 0.09296\,\mathrm{sgn}(\Delta_1)(1-r)\sin(2\delta k\, z) F_0 . \quad (5)$$

In this asymptotic limit the amplitude of F_L is independent of δk, so that the peak-to-peak amplitude of the corresponding potential is inversely proportional to the difference of the wave numbers k_1 and k_2:

$$\phi_{\max} - \phi_{\min} = 0.09296 \left| \frac{k(1-r)}{\delta k} \right| U_0 . \quad (6)$$

A small difference δk implies a deep confining potential well for the atoms in the z direction. The price one has to pay is that the confinement is soft, over a long spatial scale. The force at the spatial frequency $2\delta k$ may be 10 times smaller than the force acting on the scale of a wavelength, but it may still yield a much larger value of work on the atom because it points in the same direction over a longer distance.

The long-wavelength force of the origin I have described may already have been seen in experiments in which a beam of sodium atoms was deflected by two standing waves.[9] The authors discuss their results in terms of bichromatic excitation of a two-state system,[5] but the fact that the frequency difference between the two waves was close to the hyperfine splitting of the sodium ground state suggests that optical pumping may have played a role. Unfortunately, the authors do not provide enough data for a quantitative comparison between two- and three-state theories.

Also in the optical-molasses experiments[1-3] on sodium there are two frequencies present to prevent optical pumping to one or the other of the hyperfine levels $F=1$ and $F=2$. The frequency difference is of the order of 2 GHz, though, and the associated length scale $\pi/\delta k \sim 10$ cm is much larger than the size of the molasses. In search of a model that might simulate structure within the molasses, I therefore turn to the case of two misaligned beams.

I write the wave vectors of the two fields as

$$\mathbf{k}_1 = k\mathbf{e}_x, \quad \mathbf{k}_2 = k(\cos\varphi\,\mathbf{e}_x + \sin\varphi\,\mathbf{e}_y) . \quad (7)$$

In the example of Fig. 3 the ensuing force field is plotted for an angle φ such that the x direction accommodates 512 spatial periods of field 1 and 511 half periods of field 2, and the y direction $\frac{1}{2}$ period of field 2. This area, which in both the x and the y direction houses precisely one period of the force components at $\pm 2(\mathbf{k}_1 - \mathbf{k}_2)$ generated by the mixing, has been scaled to a square, although in reality the region shown is about 23 times longer in the x direction than in the y direction. A Fourier-transform low-pass filter has been applied to re-

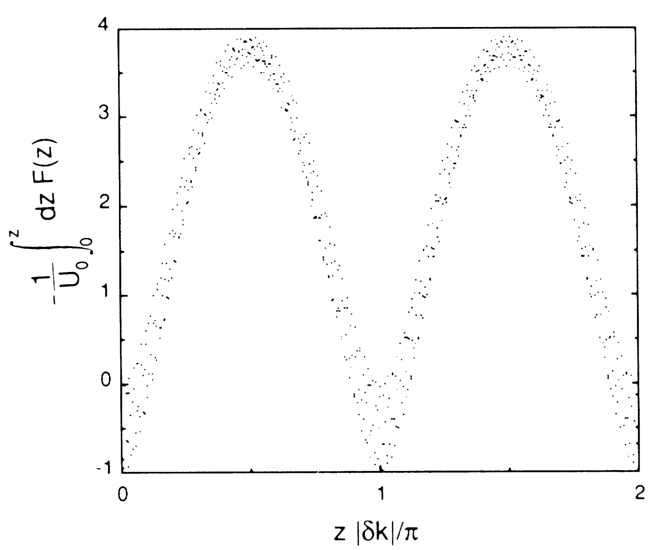

FIG. 2. Potential energy of the force field as a function of distance for two fields whose propagation vectors are both in the z direction but whose wave numbers are different, $\delta k = k_1 - k_2 \neq 0$, for the parameters $r=2$ and $\Delta_1 = \Delta_2 > 0$. The individual wave numbers k_1 and k_2 are chosen such that the regon of z shown in the figure covers 64 periods of field 1, 63 periods of field 2, and 2 periods of the nonlinear-mixing force with wave number $2\delta k$. The potential is plotted at equidistant points, and the spread of the points gives an impression of the variation of the potential on the scale of a wavelength $\lambda \approx 2\pi/k_1 \approx 2\pi/k_2$.

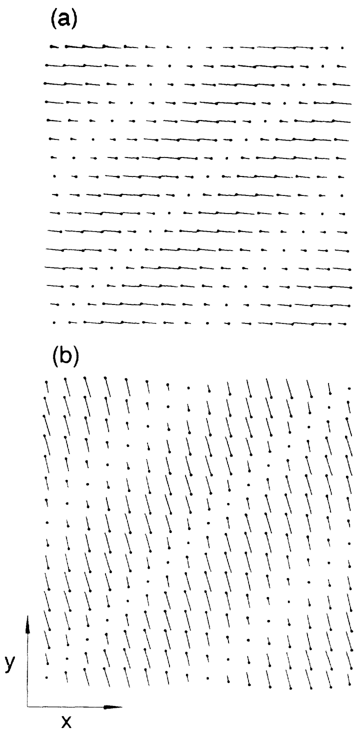

FIG. 3. Long-wavelength component of the force field in the x-y plane for the case of two misaligned beams with the angle between them being $\varphi = \arccos(1023/1024)$, for $r = 2$ and $\Delta_1 = \Delta_2 > 0$. The drawings cover one period of the force, i.e., the next column to the right would repeat the first column and similarly for the rows. The figures have been scaled to a square even though the range of the x coordinate in reality is about 23 times the range of the y coordinate. In (a) the force is shown unscaled, while in (b) the Cartesian components of the force have been scaled in the same ratio as the spatial x and y coordinates. The length of the pins in the picture is proportional to the value of the force, and the force points away from the round head.

move force components with $|\mathbf{k}\cdot\mathbf{e}_x| > 1.5k$. Figure 3(a) displays the true direction of the force, while in Fig. 3(b) the components of the force have been scaled by the same factor as the spatial coordinates. Under the latter scaling, flow patterns such as vortices might have been revealed more easily, but there seem to be none. The force parameters are $r = R_2(0)/R_1(0) = 2$, and $\Delta_1 = \Delta_2 > 0$.[8]

In one set of numerical experiments Newton's equations of motion of an atom were integrated in force fields such as in Fig. 3, starting at $x = 0$, $y = 0$ with velocity at an angle $-\varphi$ and kinetic energy equal to U_0. Thanks to the periodicity of the force, these initial conditions correspond to sending the atom along the direction of the forces down from the upper-left corner of Fig. 3(a) or 3(b). Large-scale oscillations in the kinetic energy were found. In the limit $|\varphi| \ll |1-r| \ll 1$ and for $\Delta_1 = \Delta_2 > 0$ the maximum kinetic energy was empirically given by

$$E_{k,\max} \approx 0.5 \left| \frac{1-r}{\varphi} \right|^{3/2} U_0 . \tag{8}$$

The nonlinear mixing may again create long-wavelength forces whose effect on the kinetic energy of the atom enormously exceeds the effects of the simple force field with $\mathbf{k}_1 = \mathbf{k}_2$.

If the detunings and hence the sign of the force are reversed (tuning below resonance), Figs. 3 show that the force tends to restore the atoms to the diagonal, to the maximum of the interference pattern of the two driving fields. As the force is not conservative and cannot be described in terms of a potential, I have not so far found a quantitative description of this "trapping." It is clear, though, that the light field can guide atoms along its interference patterns.

To demonstrate the difference between the two- and three-state models I note that in a pure two-state system driven by a monochromatic field with an arbitrary spatial field distribution $\mathbf{E}(\mathbf{r})$, the stimulated-emission force is conservative. When saturation is allowed for, the dependence of the potential energy on the intensity is of the form[7]

$$V(\mathbf{r}) = A \ln[1 + B\mathbf{E}^2(\mathbf{r})] , \quad B > 0 . \tag{9}$$

While the absolute value of the potential increases with increasing intensity, the potential obtained by superimposing two fields never exceeds twice the sum of the potentials of the two fields taken separately:

$$|V_{\mathbf{E}_1+\mathbf{E}_2}| = |A \ln[1 + B(\mathbf{E}_1 + \mathbf{E}_2)^2]| \le |A \ln[1 + 2B(\mathbf{E}_1^2 + \mathbf{E}_2^2)]| \le 2|V_{\mathbf{E}_1} + V_{\mathbf{E}_2}| . \tag{10}$$

The valleys and hills of the potential in Fig. 2 coincide with the maxima and minima of the interference pattern of the two driving fields, but the potential is not large or small simply because the intensity is high or low. It is easy to prove that the nonlinearity of a saturated two-state system could not generate the result of Fig. 2; the optical-pumping nonlinearity of a three-state system obviously can.

In summary, the nonlinear mixing of wave vectors of the driving field taking place in a multistate atom may create long-wavelength components in the force. Such forces may be comparable in strength to the ordinary induced forces known from two-state theories; hence their effect on the atomic motion accumulated over a multiwavelength spatial scale may vastly exceed the net acceleration or deceleration due to the same field configuration acting on a two-state atom. An experiment to demonstrate this prediction may have been carried out already,[9] although the authors themselves interpret their findings in terms of the action of a bichromatic field on a two-state atom. At any rate, Ref. 9 serves as a template for a possible experiment.

The Λ system does not support any mixing of the forces at all if both fields consist of traveling waves (instead of standing waves), and an immediate application of my model to optical molasses falls through. However, in a realistic optical molasses three orthogonal and orthogonally polarized pairs of counterpropagating waves all drive basically the same transitions between degenerate atomic levels. Opportunities for wave-vector mixing are far more numerous than in analytically treatable few-state models, and a numerical theory that incorporates both the atomic-state structure and the fully three-dimensional light field is called for. That is an *extremely* ambitious undertaking, but might pay off. I speculate that we presently see just the tip of the iceberg; that the new type of force mixing becomes the rule rather than an exception as the level structure of the atom and/or the light-field configuration get more complicated; and that with an improved understanding, a whole field of atom trapping with mixing forces may open up.

I thank Phillip Gould for initially bringing the work of Kazantsev and Krasnov to my attention, as well as for a number of discussions. I am also grateful to Paul Lett for his pointing out Ref. 9 to me. This work is partially supported by the National Science Foundation, Grant No. PHY-8902659.

[1]P. D. Lett, R. N. Watts, C. I. Westbrook, W. D. Phillips, P. L. Gould, and H. J. Metcalf, Phys. Rev. Lett. **61**, 169 (1988).

[2]S. Chu, M. G. Prentiss, A. E. Cable, and J. E. Bjorkholm, in *Laser Spectroscopy VIII*, edited by S. Svanberg and W. Persson (Springer-Verlag, Berlin, 1987), p. 58.

[3]M. Prentiss and A. Cable, in *Proceedings of the Conference on Quantum Electronics and Laser Science, 1989*, Technical Digest Series Vol. 12 (Optical Society of America, Washwavelength $\lambda \approx 2\pi/k_1 \approx 2\pi/k_2$.

[4]J. Dalibard and C. Cohen-Tannoudij, J. Opt. Soc. Am. B **6**, 2023 (1989); P. J. Ungar, D. S. Weiss, E. Riis, and S. Chu, J. Opt. Soc. Am. B **6**, 2072 (1989).

[5]A. P. Kazantsev and I. V. Krasnov, Pis'ma Zh. Eksp. Teor. Fiz. **46**, 333 (1987) [JETP Lett. **46**, 420 (1987)]; Zh. Eksp. Teor. Fiz. **95**, 104 (1989) [Sov. Phys. JETP **68**, 59 (1989)].

[6]A good account of the two-state theories may be found, e.g., in S. Stenholm, Rev. Mod. Phys. **56**, 699 (1986).

[7]A. P. Kazantsev, Zh. Eksp. Teor. Fiz. **66**, 1599 (1974) [Sov. Phys. JETP **39**, 784 (1974)]; A. Ashkin, Phys. Rev. Lett. **40**, 729 (1978).

[8]$\Delta_1 = \Delta_2$ is also the condition when the nonabsorbing two-photon resonance is most severe, and in general cannot be ignored. However, in the low-intensity limit the width of the two-photon resonance is determined by experimental artifacts such as laser linewidths, and the resonance may be very narrow compared to the detuning dependence of the force (1). If that is the case, the detunings may be chosen in such a way that the force closely approximates the case $\Delta_1 = \Delta_2$, yet the two-photon resonance is only a minor perturbation.

[9]V. S. Voitsekhovich, D. V. Danileiko, A. M. Negrijko, V. I. Romanenko, and L. P. Yatsenko, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 138 (1989) [JETP Lett. **49**, 161 (1989)].