Quantum jumps via spontaneous Raman scattering

Randall G. Hulet and D. J. Wineland
Time and Frequency Division, National Bureau of Standards, Boulder, Colorado 80303
(Received 27 April 1987)

A single laser, which is used to induce and detect spontaneous Raman transitions, can be used to observe quantum jumps in a single atom. The population dynamics of a particular system, consisting of two $^2S_{1/2}$ ground-state levels and four $^2P_{3/2}$ excited-state levels split by a magnetic field, is analyzed for a laser tuned near a particular transition. We find that the statistics of the fluorescence emitted by this system are described by the same formalism developed for the three-level V configuration irradiated by two light sources. Over a wide range of observation times, the fluorescence intensity will be two valued, either off or on, as has been verified for the V configuration. Some surprising and elegant features of this new system are described.

The fluorescence emitted from a single atom can exhibit phenomena which are unobservable in a collection of many atomic emitters. For example, the fluorescence emitted by a driven two-level system reveals a quantum phenomenon known as "photon antibunching."[1,2] Antibunching is demonstrated by measuring the time correlation between fluorescence photons emitted by a single atom. It is found that the probability of detecting a photon instantaneously after detecting the previous one is zero, that is, they are antibunched. This is a purely quantum effect with no classical analog.

A three-level manifestation of quantum photon emission was first analyzed by Cook and Kimble[3] and is based on Dehmelt's "electron shelving" scheme.[4] Figure 1 shows a three-level system in the V configuration. The ground state (0) is strongly coupled to an excited state (1) which has a spontaneous decay rate A_1, and is weakly coupled to another excited state (2) which decays at a rate A_2 ($A_1 \gg A_2$). Two radiation sources separately drive the $0 \leftrightarrow 1$ and $0 \leftrightarrow 2$ transitions while only the fluorescence from level 1 is monitored. On a time scale which is long compared to A_1^{-1}, but less than or on the order of A_2^{-1}, the fluorescence is expected to display periods of constant intensity interrupted by periods of zero intensity while the atomic electron is "shelved" in level 2. The on-off switching of the fluorescence indicates a discontinuous jump, or "quantum jump," between level 2 and the strongly fluorescing system consisting of level 0 and level 1.[3,5–12] Recently, three groups reported the observation of this phenomenon in a single confined ion[13–15] and a fourth group is able to infer the effect from the time correlation of fluorescence from a weak atomic beam.[16]

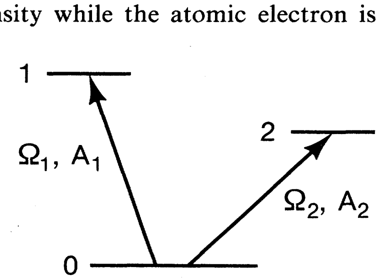

FIG. 1. The V configuration of energy levels. Two excited states are coupled to the ground state with two different lasers. The spontaneous decay rate of level i is denoted by A_i. Ω_i denotes the Rabi frequency of the $0 \leftrightarrow i$ transition. It is assumed that $A_1 \gg A_2$. Quantum jumps in the intensity of the fluorescence emitted from level 1 have been observed under these conditions (Refs. 13–16).

Kimble *et al.* have shown that the excitation of the weak transition ($0 \leftrightarrow 2$) is a rate process for the V level configuration.[11] The population dynamics of the three levels can be reduced to those of an effective two-level system for times which are long compared to A_1^{-1}. R_+ is defined as the rate of excitation out of the strongly fluorescing level-0–level-1 system and into level 2, while R_- is the rate back out of level 2. The rate equation analysis yields the mean fluorescence on-time $\langle T_{\text{on}} \rangle$, off-time $\langle T_{\text{off}} \rangle$, and the statistical distribution of the fluorescence as a function of the two Rabi frequencies, spontaneous decay rates, and laser detunings.

In this paper we analyze the population dynamics of a single atom in a magnetic field, when a *single* laser is tuned near one of the principal transition resonances. We assume that the ground state has a $^2S_{1/2}$ configuration and the excited state is $^2P_{3/2}$. Additionally, the effects of hyperfine structure are assumed to be negligible. This configuration is realized by some singly ionized alkaline-earth atoms, $^{24}Mg^+$ or $^{26}Mg^+$, for example. We find that for the time scale of interest, the evolution of the six $^2S_{1/2}$ and $^2P_{3/2}$ levels is also described by a two-level rate equation and therefore, obeys the same statistical functions derived for the three-level V configuration. Thus this system should also exhibit quantum switching. We find that the rates R_+ and R_- for this level configuration, in contrast to the V configuration, are quite simple and, as we will show, possess a surprising and elegant feature.[17]

Figure 2 schematically shows the Zeeman levels of the $^2S_{1/2}$ and $^2P_{3/2}$ states. It is assumed that the field is sufficiently weak that the levels are well described by the LS-coupling scheme. For convenience, the levels are labeled from 1 to 6, starting with the lowest ground level. The energy difference between levels i and j is denoted by $\hbar\omega_{ij} \equiv \hbar(\omega_i - \omega_j)$. A laser of frequency ω is assumed tuned near ω_{31}, the ${}^2S_{1/2}$, $m_J = -\frac{1}{2} \leftrightarrow {}^2P_{3/2}$, $m_J = -\frac{3}{2}$ ($1 \leftrightarrow 3$) resonance frequency. If Δ denotes the detuning from resonance then $\omega = \omega_{31} + \Delta$. $\hbar\Delta$ is assumed small compared to the energy separation between Zeeman levels, which is in turn assumed to be small compared to the ${}^2P_{3/2}$ and ${}^2S_{1/2}$ energy difference. The laser light is assumed to be linearly polarized with polarization perpendicular to the magnetic field axis. The only dipole decay allowed for level 3 is back to level 1. Therefore, the fluorescence from level 3 continues until an off-resonance transition from level 1 to level 5 is induced. There is a small probability of a nonresonant excitation to this level due to the finite linewidth of the transition. If level 5 becomes populated, the atom may decay to either ground state: back to level 1 (with $\frac{1}{3}$ probability), where the $1 \leftrightarrow 3$ cycling continues, or to level 2 (with $\frac{2}{3}$ probability), where the electron remains until another transition, again far from resonance, may remove it. The dipole selection rules allow transitions from level 2 to either level 4 or to level 6. Once in level 4, the atom may decay back to level 1 (with $\frac{2}{3}$ probability) to resume the strong fluorescence cycling. From level 6, the atom may decay only to level 2. The other Zeeman ground level, level 2, is the "shelf" level which removes the electron from the strongly fluorescing system. Thus, as in the three-level V configuration, which uses two light sources, the fluorescence from this six-level, one-laser system is expected to alternate between periods of darkness and light. As opposed to other realizations of quantum jumps, the shelf level in this case "decays" via spontaneous Raman transitions since spontaneous radiative decay from level 2 to level 1 is negligible.

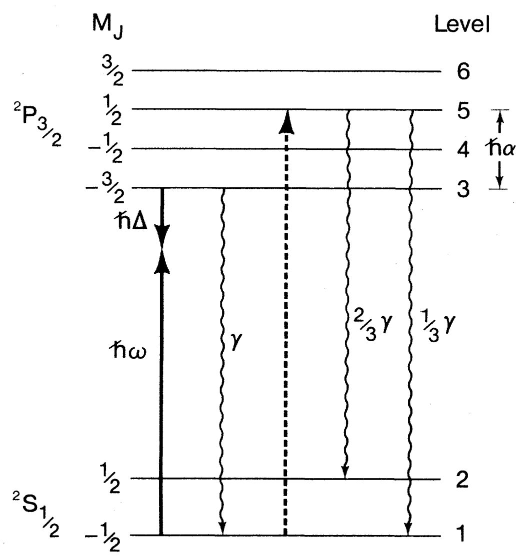

FIG. 2. The energy-level structure of the ${}^2S_{1/2}$ and the ${}^2P_{3/2}$ states of an atom in a magnetic field. A laser of frequency ω is assumed to be tuned near the level 1 to level 3 transition frequency which gives rise to a (on resonance) Rabi frequency of Ω. The laser polarization is assumed to allow only $\Delta m_J = \mp 1$ transitions. The detuning from resonance is denoted by Δ. The total spontaneous decay rate of each excited level is γ and the energy separation between excited levels is given by $\hbar\alpha/2$. The dashed arrow indicates off-resonance excitation of the $1 \rightarrow 5$ transition by the laser. The wavy arrows denote the dipole-allowed spontaneous decays from levels 3 and 5. Off-resonance excitation of the $2 \rightarrow 4$ and $2 \rightarrow 6$ transitions and spontaneous decay from levels 4 and 6 are not shown in the figure. It is assumed that $\alpha \gg \Delta, \gamma, \Omega$.

We use the density-matrix formulation to determine the steady-state level populations and to show that the two-state rate approximation is valid during the observation time of interest. In the rotating-wave approximation, the density-matrix equations are

$$
\begin{aligned}
\dot\rho_{11} &= \Omega(\mathrm{Im}\sigma_{13} + \mathrm{Im}\sigma_{15}/\sqrt{3}) + \gamma(\rho_{33} + 2\rho_{44}/3 + \rho_{55}/3) , \\
\dot\rho_{22} &= \Omega(\mathrm{Im}\sigma_{24}/\sqrt{3} + \mathrm{Im}\sigma_{26}) + \gamma(\rho_{44}/3 + 2\rho_{55}/3 + \rho_{66}) , \\
\dot\rho_{33} &= -\Omega \mathrm{Im}\sigma_{13} - \gamma\rho_{33} , \\
\dot\rho_{44} &= -\Omega \mathrm{Im}\sigma_{24}/\sqrt{3} - \gamma\rho_{44} , \\
\dot\rho_{55} &= -\Omega \mathrm{Im}\sigma_{15}/\sqrt{3} - \gamma\rho_{55} , \\
\dot\rho_{66} &= -\Omega \mathrm{Im}\sigma_{26} - \gamma\rho_{66} , \\
\dot\sigma_{13} &= [-i(\omega - \omega_{31}) - \tfrac{1}{2}\gamma]\sigma_{13} + \tfrac{1}{2}i\Omega(\rho_{33} - \rho_{11} + \rho_{35}^*/\sqrt{3}) , \\
\dot\sigma_{15} &= [-i(\omega - \omega_{51}) - \tfrac{1}{2}\gamma]\sigma_{15} + \tfrac{1}{2}i\Omega[(\rho_{55} - \rho_{11})/\sqrt{3} + \rho_{35}], \\
\dot\sigma_{24} &= [-i(\omega - \omega_{42}) - \tfrac{1}{2}\gamma]\sigma_{24} + \tfrac{1}{2}i\Omega[(\rho_{44} - \rho_{22})/\sqrt{3} + \rho_{46}^*], \\
\dot\sigma_{26} &= [-i(\omega - \omega_{62}) - \tfrac{1}{2}\gamma]\sigma_{26} + \tfrac{1}{2}i\Omega(\rho_{66} - \rho_{22} + \rho_{46}/\sqrt{3}) , \\
\dot\rho_{35} &= (i\omega_{53} - \gamma)\rho_{35} + \tfrac{1}{2}i\Omega(\sigma_{15} - \sigma_{13}^*/\sqrt{3}) , \\
\dot\rho_{46} &= (i\omega_{64} - \gamma)\rho_{46} + \tfrac{1}{2}i\Omega(\sigma_{26}/\sqrt{3} - \sigma_{24}^*) ,
\end{aligned}
\tag{1}
$$

where $\Omega \equiv \Omega_{13} = E_0 d_{13}/\hbar$ is the Rabi frequency of the $1 \leftrightarrow 3$ transition, γ is the total spontaneous decay rate of each excited level, $\mathrm{Im}\sigma_{ij}$ denotes the imaginary part of a coherence and the asterisk denotes complex conjugation. The other Rabi frequencies are determined by the relative size of the dipole-matrix elements for light linearly polarized perpendicular to the quantization axis: $d_{15} = d_{13}/\sqrt{3}$, $d_{24} = d_{26}/\sqrt{3}$, and $d_{26} = d_{13}$.

There are only six nonzero coherences for our choice of laser polarization. The four σ_{ij} are due to direct laser coupling of a ground and an excited level, while the two ρ_{ij} represent the stimulated Raman coherences between pairs of excited levels coupled by the laser via a ground level.

It is most compact to express the detunings, $\omega - \omega_{ij}$ in terms of Δ ($\Delta = \omega - \omega_{31}$) and the magnetic field splitting of the levels. In the LS-coupling scheme, the Lande' g factors are $g({}^2S_{1/2}) = 2$ and $g({}^2P_{3/2}) = \frac{4}{3}$ (we take the electron g factor equal to 2 for this calculation). The detunings become

$$\omega-\omega_{51}=\Delta-\alpha\ ,$$
$$\omega-\omega_{42}=\Delta+\alpha/4\ ,$$
$$\omega-\omega_{62}=\Delta-3\alpha/4\ .$$

In the above expressions $\omega_{53}=\omega_{64}=\alpha$, where $\hbar\alpha\equiv 8\mu_B B/3$ is twice the Zeeman energy splitting between adjacent ${}^2P_{3/2}$ levels due to the magnetic field B and μ_B is the Bohr magneton. We will assume hereafter that $\alpha \gg \Delta,\Omega,\gamma$.

Solving the set of algebraic equation $\dot\rho_{ii}=\dot\sigma_{ij}=\dot\rho_{ij}=0$, and using the conservation equation $\sum_i \rho_{ii}=1$ yields the steady-state values of the populations $\bar\rho_{ii}$. Thus to the first nonzero order of γ/α or Ω/α,

$$\begin{aligned}
\bar\rho_{11}&\approx\frac{16}{17}\frac{\gamma^2+4\Delta^2+\Omega^2}{\gamma^2+4\Delta^2+2\Omega^2}\ ,\\
\bar\rho_{22}&\approx\tfrac{1}{17}\ ,\\
\bar\rho_{33}&\approx\frac{16}{17}\frac{\Omega^2}{\gamma^2+4\Delta^2+2\Omega^2}\ ,\\
\bar\rho_{44}&\approx\frac{4\Omega^2}{51\alpha^2}\ ,\\
\bar\rho_{55}&\approx\frac{4\Omega^2}{51\alpha^2}\ ,\\
\bar\rho_{66}&\approx\frac{4\Omega^2}{153\alpha^2}\ .
\end{aligned}\tag{2}$$

Terms of order (Δ/α) have been neglected.

We follow Cook *et al.*[3] by defining new population variables

$$\rho_-\equiv\rho_{11}+\rho_{33}+\rho_{55}\ ,$$
$$\rho_+\equiv\rho_{22}+\rho_{44}+\rho_{66}=1-\rho_-\ .$$

Therefore, in steady state,

$$\bar\rho_-\approx\tfrac{16}{17}+O(\Delta/\alpha)\ ,$$
$$\bar\rho_+\approx\tfrac{1}{17}+O(\Delta/\alpha)\ .$$

The ratio $\bar\rho_-/\bar\rho_+=16+O(\Delta/\alpha)$ is the ratio of the mean duration of the fluorescence on periods to the off periods. A simple rate equation analysis confirms that the ratio of the ground-state level populations is 16 in the low-intensity limit ($\Omega\ll\gamma$) and for $\Delta=0$.[18] However, we find that this ratio, unlike that for the three-level V configuration, is largely independent of laser intensity and is only weakly dependent on detuning (for $\gamma,\Omega,\Delta\ll\alpha$). The surprising feature here is that this ratio is independent of saturation and power broadening effects. As Ω is increased from zero, $\bar\rho_{11}$ decreases as population is transferred to level 3. We might expect, therefore, that the probability of transferring population from level 1 into the shelving level should diminish. However, Eqs. (2) indicate that the population of level 5, from which the shelving level (level 2) is populated via spontaneous decay, increases as Ω^2 with no effect due to saturation of the $1\leftrightarrow3$ transition. The apparent explanation of this phenomenon is that population is transferred from level 3 to level 5 via the Raman coherence ρ_{35}. The increase in ρ_{35} with increasing Ω exactly compensates for the effect of the decreasing population in level 1.

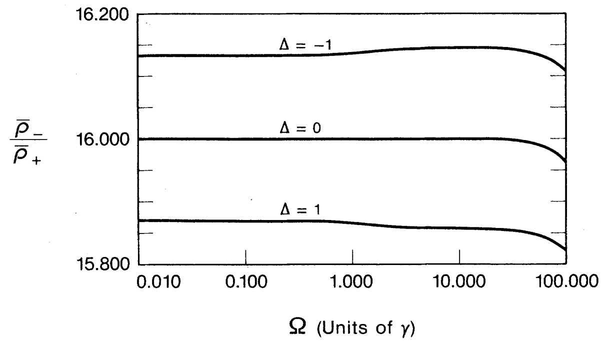

FIG. 3. The ratio of the steady-state population in levels 1, 3, and 5 (ρ_- system) to the steady population in levels 2, 4, and 6 (ρ_+ system) is plotted vs Ω for various detunings, Δ, and for $\alpha=1215$ (all in units of γ). $\bar\rho_-/\bar\rho_+$ is equal to the ratio of the mean duration of the fluorescence on periods to the off periods. There is little effect due to saturation of the $1\leftrightarrow3$ transition and only a slight deviation from the value of 16. The downturn in $\bar\rho_-/\bar\rho_+$ for very large Ω is due to the power-broadened linewidth of the transition becoming comparable to the separation between excited levels.

We have used a computer to numerically solve Eqs. (1) for the exact steady-state solutions using various values of the parameters Δ and Ω. Figure 3 is a plot of $\bar\rho_-/\bar\rho_+$ versus Ω for several values of Δ. The ratio remains remarkably close to the value of 16 for $\Delta=0$ and $\Omega\ll\alpha$, and even for $\Omega\gg\gamma$. The ratio is offset from the value 16 when $\Delta\neq0$ by approximately $-160(\Delta/\alpha)$.

Figure 4 demonstrates the importance of including coherence effects in the calculation. Figure 4 is a plot of $\bar\rho_-/\bar\rho_+$ versus Ω when the coherences between excited states ρ_{35} and ρ_{46} are neglected. ρ_{35} and ρ_{46} are due to stimulated, off-resonant Raman coupling. Since this

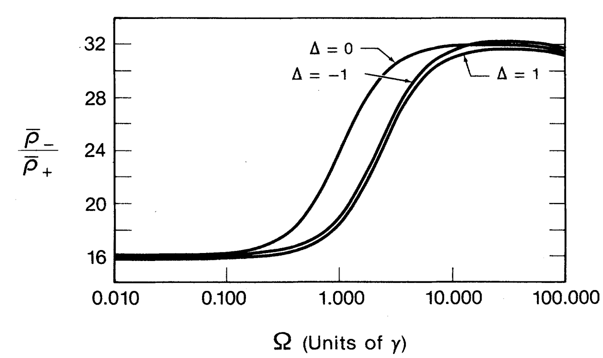

FIG. 4. Same as Fig. 3, except that the stimulated Raman coherences ρ_{35} and ρ_{46} have been neglected in the calculation. Saturation of the $1\leftrightarrow3$ transition causes $\bar\rho_-/\bar\rho_+$ to deviate from 16 when $\Omega\gtrsim\gamma$. This demonstrates that the Raman coherences are responsible for the lack of intensity dependence of $\bar\rho_-/\bar\rho_+$.

coupling is small for $\Omega \ll \gamma$, $\bar{\rho}_-/\bar{\rho}_+$ remains close to 16 in the low-intensity regime. However, $\bar{\rho}_-/\bar{\rho}_+$ rapidly deviates from 16 when Ω is increased beyond γ, indicating the increasing role the Raman coherences assume in correctly describing the population evolution as Ω increases.

The dynamics of the population evolution is governed by rates from two widely differing time domains: (1) a "short" time of order γ^{-1} and (2) a much longer time in which population is transferred between the ρ_- and ρ_+ systems. An examination of the $\dot{\sigma}_{ij}$ equations of Eqs. (1) reveals that the short-time behavior is exponentially damped in a time γ^{-1}. Therefore, for times of interest which are greater than γ^{-1} we can ignore the short-time behavior. We would like to show that the evolution of population between the ρ_- and ρ_+ levels of this system is similar to that of the V configuration. (The population dynamics also determine the statistics of the emitted photons.) To do so, we must cast the dynamical equations in the form of a two-state rate equation. If we identify R_+ as the rate out of ρ_-, the strongly fluorescing ($-$) system, and R_- as the rate back into it, the two-state rate equations are

$$\dot{\rho}_- = -R_+\rho_- + R_-\rho_+ \ ,$$

$$\dot{\rho}_+ = R_+\rho_- - R_-\rho_+ \ .$$

Population is transferred between the two systems via spontaneous radiative decay. To switch from the $-$ system to the $+$ system the atom must decay from level 5 to level 2. The branching ratio for this transition is $\frac{2}{3}$. Similarly, the reverse process requires a decay from level 4 to level 1, also with $\frac{2}{3}$ probability. Therefore,

$$R_+\bar{\rho}_- = (\tfrac{2}{3})\gamma\bar{\rho}_{55}$$

and

$$R_-\bar{\rho}_+ = (\tfrac{2}{3})\gamma\bar{\rho}_{44} \ .$$

Using the expressions [Eqs. (2)] for the $\bar{\rho}_{ii}$ gives

$$R_+ = \frac{\Omega^2\gamma}{18\alpha^2} + O(\Delta/\alpha)$$

and

$$R_- = \frac{8\Omega^2\gamma}{9\alpha^2} + O(\Delta/\alpha) \ .$$

The correctness of these values for R_- and R_+ has been checked by numerically integrating Eqs. (1) and verifying that the evolution towards steady state is indeed governed by these rates. The decay of the short-time behavior in a time $t \approx \gamma^{-1}$ was also verified.

The calculations which we have described assume that a laser is tuned near the $1 \leftrightarrow 3$ transition. However, we note that an entirely analogous set of equations and results are obtained by tuning the laser near the $2 \leftrightarrow 6$ transition.

The observation of quantum jumps in this system provides an excellent opportunity for comparison with theory. The very weak dependence of $\bar{\rho}_-/\bar{\rho}_+$ $(=R_-/R_+)$ on all of the parameters which can fluctuate in an experiment means that the fluorescence statistics may be very accurately measured and compared with calculation. Additionally, the switching rate can be carefully normalized by observing a portion of the laser beam transmitted through the apparatus. An experiment is now underway at the National Bureau of Standards (NBS) to observe these effects. A single $^{24}Mg^+$ ion has been confined by an electromagnetic (Penning) ion trap while a nearly resonant laser drives the $3\,^2S_{1/2}, m_J = -\frac{1}{2} \leftrightarrow 3\,^2P_{3/2}, m_J = -\frac{3}{2}$ $(1 \leftrightarrow 3)$ transition. This new apparatus has substantially better sensitivity than a previous experiment which was able to detect a single Mg^+ ion but which did not have the sensitivity to detect quantum jumps.[19] For a magnetic field of 1.4 T, the Zeeman splitting of the excited states is $\frac{1}{2}\alpha \approx \frac{1}{2}(1215\gamma) = (2\pi)26.1$ GHz for $^{24}Mg^+$, where $\gamma = (2\pi)43$ MHz. Therefore, the regime of $\alpha \gg \gamma, \Omega, \Delta$ is quite readily achieved experimentally.

The authors gratefully acknowledge the support of the U.S. Air Force Office of Scientific Research and the U.S. Office of Naval Research. One of us (R.G.H.) acknowledges the support of the National Research Council. We thank R. J. Cook and L. Hollberg for their helpful suggestions concerning the manuscript.

[1]H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).
[2]See, for example, J. D. Cresser, J. Häger, G. Leuchs, M. Rateike, and H. Walther, in *Dissipative Systems in Quantum Optics,* Vol. 27 of *Topics in Current Physics,* edited by R. Bonifacio (Springer-Verlag, Berlin, 1982) and references therein; F. Diedrich and H. Walther, Phys. Rev. Lett. **58**, 203 (1987).
[3]R. J. Cook and H. J. Kimble, Phys. Rev. Lett. **54**, 1023 (1985).
[4]H. G. Dehmelt, J. Phys. (Paris) **42**, C8-299 (1981).
[5]T. Erber and S. Putterman, Nature (London) **318**, 41 (1985).
[6]J. Javanainen, Phys. Rev. A **33**, 2121 (1986).
[7]A. Schenzle, R. G. DeVoe, and R. G. Brewer, Phys. Rev. A **33**, 2127 (1986).
[8]C. Cohen-Tannoudji and J. Dalibard, Europhys. Lett. **1**, 441 (1986).
[9]D. T. Pegg, R. Loudon, and P. L. Knight, Phys. Rev. A **33**, 4085 (1986).
[10]A. Schenzle and R. G. Brewer, Phys. Rev. A **34**, 3127 (1986).
[11]H. J. Kimble, R. J. Cook, and A. L. Wells, Phys. Rev. A **34**, 3190 (1986).
[12]P. Zoller, M. Marte, and D. F. Walls, Phys. Rev. A **35**, 198 (1987).
[13]W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).
[14]J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wine-

land, Phys. Rev. Lett. **57**, 1699 (1986).
[15]Th. Sauter, W. Neuhauser, R. Blatt, and P. E. Toschek, Phys. Rev. Lett. **57**, 1696 (1986).
[16]M. A. Finn, G. W. Greenlees, and D. A. Lewis, Opt. Commun. **60**, 149 (1986).
[17]R. G. Hulet and D. J. Wineland, Bull. Am. Phys. Soc. **31**, 938 (1986).
[18]D. J. Wineland, J. C. Bergquist, W. M. Itano, and R. E. Drullinger, Opt. Lett. **5**, 245 (1980).
[19]D. J. Wineland and W. M. Itano, Phys. Lett. **82A**, 75 (1981).