Threshold and Linewidth of a Mirrorless Parametric Oscillator

M. Fleischhauer,[1,2] M. D. Lukin,[1] A. B. Matsko,[3,4] and M. O. Scully[3,4]
[1]*Institute for Theoretical Atomic and Molecular Physics, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts 02138*
[2]*Sektion Physik, Universität München, D-80333 München, Germany*
[3]*Department of Physics, Texas A&M University, College Station, Texas 77843-4242*
[4]*Max-Plank-Institut für Quantenoptik, Garching, D-85748, Germany*
(Received 9 July 1999)

We analyze the above-threshold behavior of a mirrorless parametric oscillator based on resonantly enhanced four-wave mixing in a dense atomic vapor. It is shown that, in the ideal limit, an arbitrary small flux of pump photons is sufficient to reach the oscillator threshold. We demonstrate that, due to the large group velocity delays associated with electromagnetically induced transparency, an extremely narrow oscillator linewidth is possible, making a narrow-band source of nonclassical radiation feasible.

PACS numbers: 42.50.Gy, 42.65.Hw

Stable and low-noise sources of coherent and nonclassical radiation are of interest in many areas of laser physics and quantum optics. Such sources have a wide range of applications such as frequency standards, optical magnetometry, gravitational wave detection, and high-precision spectroscopy.

The present theoretical work is motivated by recent experiments demonstrating a phase transition to mirrorless oscillation of counterpropagating Stokes and anti-Stokes fields in resonant, double-Λ Raman media [1]. In contrast to earlier studies involving instabilities in alkali vapors [2–5], this oscillation could be achieved with pump fields of μW power (nanojoule pulse energy) and is accompanied by a dramatic narrowing of the beat signals between driving and generated fields. Oscillations of this type are clear manifestations of atomic coherence and interference effects, which have recently led to many exciting developments in resonant nonlinear optics [6–9]. In particular, the unusual efficiency of the present processes is expected to lead to a new regime of quantum nonlinear optics in which interactions at a level of a few light quanta are feasible. Furthermore, the photon pairs generated can possess nearly ideal quantum correlations, resulting in almost complete squeezing of quantum fluctuations [10].

Here we study theoretically the quantum dynamics of the mirrorless oscillator above threshold. We show that for an infinitely long-lived atomic dark state an arbitrary small stationary flux of pump photons is sufficient to maintain the oscillation. We furthermore analyze frequency locking and linewidth narrowing of the beat note between oscillation and pump frequencies. In particular, we show that the beat-note linewidth is given by an expression similar to the Schawlow-Townes formula for lasers, where the cavity storage time is replaced by the group-delay time $\tau_{\rm gr}$ in the medium. Because of the large linear dispersion associated with electromagnetically induced transparency (EIT) in optically thick media, the group delay can be extremely large [11–13], leading to a very small beat-note linewidth. This effect is analogous to the line narrowing in intracavity EIT [14,15]. Since only very small pump powers are needed to reach threshold, ac-Stark shifts and the associated systematic effects on the beat-note frequency can be made very small. The combination of line narrowing and small pump-power requirements makes the mirrorless parametric oscillator an interesting novel source of stable and narrow-linewidth nonclassical radiation. Possible applications include frequency standards, optical magnetometry, and few-photon nonlinear optics.

Consider the propagation of four nearly resonant plane waves, parallel or antiparallel to the z axis, in a medium consisting of double-Λ atoms (see Fig. 1). These include two counterpropagating driving fields with equal frequencies ν_d and (complex) Rabi frequencies E_f and E_b, and two generated fields (anti-Stokes and Stokes) with carrier frequencies ν_1 and ν_2 obeying $\nu_1 + \nu_2 = 2\nu_d$. The fields interact via the long-living coherence (decay rate γ_0) on the transition between b_1 and b_2 with frequency splitting $\omega_0 = \omega_{b2} - \omega_{b1}$.

Because of resonantly enhanced four-wave mixing, the coherent pump fields generate counterpropagating anti-Stokes and Stokes fields (here described by the complex Rabi frequencies E_1 and E_2). For a sufficiently large density-length product of the medium and for a certain

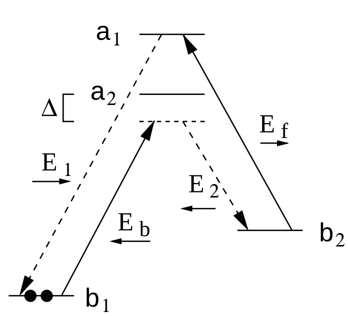

FIG. 1. Atoms in double Λ configuration interacting with two classical driving fields in forward (E_f) and backward directions (E_b) and two quantum fields ($E_{1,2}$). All optical transitions are assumed to be radiatively broadened.

 0031-9007/00/84(16)/3558(4)$15.00

pump-field intensity, the system shows a phase transition to self-oscillations [1]. The feedback mechanism required for oscillations is provided here by the gain medium itself: A spontaneously generated Stokes photon stimulates "downstream" a Raman process. As a result an anti-Stokes photon is generated with a fixed relative phase. This photon propagates in the opposite direction and stimulates another scattering event "upstream." If the phase matching condition is fulfilled, this causes a second Stokes emission in phase with the first one closing the feedback cycle. A crucial condition for the coherence of this mechanism is a sufficiently long-lived Raman coherence.

We now discuss the transition to self-oscillation and the classical and quantum dynamics above threshold in detail. To simplify the analysis we ignore inhomogeneous broadening and assume equal coupling strength of all fields as well as equal radiative decays. Furthermore we assume that the forward driving field E_f is in resonance with the $b_2 \to a_1$ transition, whereas the backward driving field E_b has a detuning $\Delta \gg |E_b|$ from the $b_1 \to a_2$ transition. In this case linear losses of the fields are minimized.

In order to calculate the medium response we solve the single-atom density matrix equations in third order of the Stokes and anti-Stokes fields and assume $|\Delta| \gg \gamma$, $|E_{f,b}| \gg \gamma_0, \delta$, where $\delta = \nu_d + \omega_0 - \nu_1$ is the two-photon detuning. In a frame rotating with the carrier frequencies, the propagation of the classical fields can then be described by the following equations for the slowly varying complex Rabi frequencies:

$$\frac{d}{dz}\tilde{E}_1 = i\kappa \frac{\tilde{E}_1^2 E_2 E_f^* \tilde{E}_b^* + E_f \tilde{E}_b E_2^* (2|\tilde{E}_1|^2 - |E_f|^2)}{\Delta |E_f|^4} + i\left[\kappa \frac{i\gamma_0 - \delta}{|E_f|^2} - \kappa \frac{|\tilde{E}_b|^2 - |E_f|^2}{\Delta|E_f|^2} - \Delta k\right]\tilde{E}_1, \quad (1)$$

$$\frac{d}{dz} E_2^* = i\kappa \frac{(|\tilde{E}_1|^2 - |E_f|^2) E_f^* \tilde{E}_b^* \tilde{E}_1}{\Delta |E_f|^4}, \quad (2)$$

$$\frac{d}{dz} E_f = i\kappa \frac{\tilde{E}_1^* E_2^* \tilde{E}_b E_f^2}{\Delta |E_f|^4}, \quad (3)$$

$$\frac{d}{dz} \tilde{E}_b^* = -i\kappa \frac{\tilde{E}_1^* E_2^* E_f |E_f|^2}{\Delta |E_f|^4}. \quad (4)$$

In these equations we have kept ac-Stark induced phase terms only in lowest order of the generated fields, since we are interested in the case $|E_{f,b}| \gg |E_{1,2}|$. $\kappa = (3/8\pi) \times N\lambda^2\gamma_a$ is the equal coupling constant of all fields with N being the atom density, λ the average wavelength of the fields, and γ_a the common population decay rate out of the excited states. $E_1 = \tilde{E}_1 e^{i(\Delta k - \kappa/\Delta)z}$, $E_b = \tilde{E}_b e^{i\kappa z/\Delta}$, with $\Delta k = k_2 - k_1$ being the phase mismatch.

We note an important feature of the nonlinear coupling in Eqs. (1) and (2): In contrast to the usual $\chi^{(3)}$ media, the lowest-order cross-coupling terms are proportional to the ratio of the pump fields rather than the product;

$$\frac{d}{dz} E_1 \sim -i\chi^{(3)} E_f E_b E_2^* \to -i\,\frac{\kappa}{\Delta}\,\frac{E_b}{E_f^*}\,E_2^*. \quad (5)$$

Thus for $|E_f| = |E_b|$ these terms are independent of the pump-field amplitudes. We will see later that this leads to a rather unusual threshold behavior.

In the present system a transition to spontaneous oscillations is possible [1], if the phase matching condition

$$\kappa \frac{\delta}{|E_f|^2} + \kappa \frac{|E_b|^2 - |E_f|^2}{\Delta|E_f|^2} + \Delta k = 0 \quad (6)$$

is fulfilled. For large values of κ, Eq. (6) describes a pulling of the frequency differences between generated fields and driving fields towards the ac-Stark shifted frequency of the Raman transition

$$\nu_1 - \nu_{d1} = \nu_{d2} - \nu_2 = \frac{\eta[\omega_0 + (|E_b|^2 - |E_f|^2)/\Delta]}{1 + \eta}. \quad (7)$$

This equation shows a close analogy with intracavity EIT. $\eta = c\kappa/2|E_f|^2$ is a frequency stabilization factor [14]. This factor also governs the group velocity of the eigenmodes of the system $v_{\rm gr} = c/(1 + \eta)$ and can be rather large. For conditions close to the experiments of Ref. [1], a reduction factor of $\eta \sim 5 \times 10^6$ was measured [13]. In the limit of large η the beat notes between generated and pump fields lock tightly to the Raman-transition frequency of the medium.

We next consider the classical steady state solution of the propagation problem. In the ideal limit ($\gamma_0 = 0$), Eqs. (1)–(4) have four constants of motion: the total intensity of the generated and pump fields $|E_1|^2 + |E_2|^2$ and $|E_f|^2 + |E_b|^2$, as well as $\mathrm{Re}[E_f^* E_b^* E_1 E_2]$ which has a similar structure to the cubic expression conserved in three-wave mixing [16]. There is also the somewhat unusual constant of motion, $|E_f|^2 \exp(|E_1|^2/|E_f|^2)$. If we take into account, however, that Eqs. (1)–(4) hold only to third order in the generated fields, this constant is equivalent to $|E_f|^2 + |E_1|^2$. With this, Eqs. (1)–(4) can be solved analytically, if the phase matching condition is approximately fulfilled. Assuming equal input intensities of the driving fields $|E_f(0)| = |E_b(L)|$ at $z = 0$ and $z = L$, respectively (L being the cell length), and disregarding linear losses due to the finite lifetime of the ground-state coherence, one finds, in second order of the generated fields,

$$|E_1(z)| = E\sin\vartheta(z), \qquad |E_2(z)| = E\cos\vartheta(z), \quad (8)$$

$$|E_f(z)| = [|E_f(0)|^2 - E^2\sin^2\vartheta(z)]^{1/2}, \quad (9)$$

where $\vartheta(z) = \kappa z/\Delta[1 - E^2/2|E_f(0)|^2]$. For $\kappa L/\Delta < \pi/2$, $E \equiv 0$. For values of $\kappa L/\Delta$ larger than the critical value $\pi/2$ there are two solutions,

$$E = \sqrt{2}\,|E_f(0)| \begin{cases} 0 \\ \sqrt{1 - \frac{\pi}{2}\frac{\Delta}{\kappa L}} \end{cases} \quad \text{for} \quad \frac{\kappa L}{\Delta} \geq \frac{\pi}{2}, \tag{10}$$

with $E = 0$ being unstable. It should be noted that, in contrast to degenerate four-wave mixing in usual $\chi^{(3)}$ media [17], the threshold condition does not contain the amplitude of the pump fields. Thus under the ideal conditions assumed here, i.e., for an infinitely long-lived dark state, an arbitrarily small stationary pump intensity is sufficient to reach the oscillation threshold. Figure 2 shows the above-threshold behavior of E as a function of $\kappa L/\Delta$ and the field amplitudes normalized to $|E_f(0)|$ inside the cell. If the system oscillates not too far above threshold, the depletion of the pump fields is small and we may assume, in the following, constant driving field amplitudes, $|E_f(z)| = |E_b(z)| = E_d$.

To calculate the linewidth of Stokes and anti-Stokes fields relative to the drive field above threshold, we assume that the generated fields can be represented as a sum of the classical stationary solutions and a time-dependent fluctuation $[\hat{E}_{1,2}(z,t) = E_{1,2}(z) + \delta E_{1,2}(z,t)]$. We utilize a standard linearized c-number Langevin approach in which collective atomic variables and fields are described by time- and position-dependent stochastic differential equations with δ-correlated Langevin forces [18]. The diffusion coefficients or noise correlations are derived using the fluctuation-dissipation theorem and generalized Einstein relations. We obtain for the Fourier components of the Stokes and anti-Stokes fluctuations $\delta E_{1,2}(\omega) \equiv 1/\sqrt{2\pi}\int dt\, \delta E_{1,2}(t) e^{-i\omega t}$,

$$\frac{d}{dz}\begin{bmatrix} \delta E_1^* \\ \delta E_2 \end{bmatrix} = i\begin{bmatrix} -\kappa\omega/E_d^2 - \omega/c & \kappa/\Delta \\ \kappa/\Delta & \omega/c \end{bmatrix} \times \begin{bmatrix} \delta E_1^* \\ \delta E_2 \end{bmatrix} + \begin{bmatrix} f_1^* \\ f_2 \end{bmatrix}, \tag{11}$$

where small fluctuation frequencies ($\omega \ll E_d$) and a constant phase of the pump field have been assumed. Following the procedure of Ref. [18], we find for the dominant noise correlation

$$\langle f_1(z,\omega) f_2(z',\omega')\rangle \simeq \frac{\kappa^2 L}{\mathcal{N}}\frac{i}{\Delta}\,\delta(z - z')\delta(\omega + \omega'), \tag{12}$$

where $\mathcal{N}$ is the number of atoms in the cell, and we have identified the quantization length defined in [18] with the length of the cell L.

Solving the inhomogeneous boundary problem for the Fourier components of the field fluctuations with $\delta E_1^*(0,\omega) = 0$ and $\delta E_2(L,\omega) = 0$, one eventually finds the phase fluctuation of, e.g., δE_1. In phase-diffusion approximation, the linewidth $\Delta\nu_1$ of E_1 relative to the pump field is given by $\langle \delta\phi_1(L,\omega)\delta\phi_1(L,\omega')\rangle = \Delta\nu_1 \delta(\omega + \omega')/\omega^2$. Using Eq. (12), we arrive at

$$\Delta\nu_1 = \frac{2E_d^4}{\Delta^2}\frac{\hbar\nu}{P_{\text{out}}}, \tag{13}$$

where P_{out} is the output power of the mode.

Equation (13) can be represented in a very instructive form, if the group-delay time $\tau_{\text{gr}} = L/c(1 + \eta)$ is introduced. In the appropriate limit, $\eta \gg 1$, and near threshold such that $\kappa L/\Delta \approx \pi/2$ the linewidth can be written as

$$\Delta\nu_1 = \frac{\pi^2}{8}\tau_{\text{gr}}^{-2}\frac{\hbar\nu}{P_{\text{out}}} \approx \tau_{\text{gr}}^{-2}\frac{\hbar\nu}{P_{\text{out}}}. \tag{14}$$

Equation (14) is formally identical to that of an ideal laser with the cavity decay time replaced by the group-delay time. In usual four-wave mixing, based on nonresonant Kerr nonlinearities [2–5], the group velocity is essentially equal to the vacuum speed of light. In the present scheme, however, it can be substantially reduced due to EIT.

It is important to emphasize that the photon pairs generated by the oscillation process near threshold are in quantum correlated states. This results in a dramatic suppression of intrinsic quantum fluctuations in a quadrature of the combined mode [10].

In the discussion above we have neglected the relaxation rate of the ground-state coherence γ_0. If this decay is taken into account, one finds the modified threshold condition: $\cos(\xi L) + (\gamma_0\Delta)/(2E_d^2)\sin(\xi L) = 0$, with $\xi = \kappa\sqrt{1/\Delta^2 - \gamma_0^2/4E_d^4}$. In particular, oscillation can be achieved only if $E_d^2 \geq \gamma_0|\Delta|/2$. This can be translated into a condition for the photon flux Φ, i.e., the number of pump photons traversing the cell per unit time. One finds that the threshold photon flux in each pump beam is equal to the number $\mathcal{N}$ of atoms in the ensemble decaying out of the dark state per unit time:

$$\Phi_{th} = f\mathcal{N}\gamma_0, \tag{15}$$

where f is a numerical prefactor of order unity. Since by using buffer gases or coated cells very small values of γ_0 can be achieved, a threshold flux corresponding to only a few photons in the cell is feasible, leading to an interesting new regime of nonlinear optics.

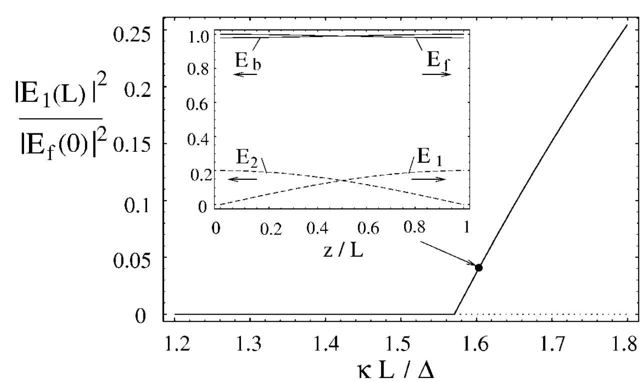

FIG. 2. Phase transition to mirrorless parametric oscillations. Analytic solution for amplitude of generated field for $\gamma_0 = 0$. Inset: normalized field amplitudes inside medium for $E/|E_f(0)| = 0.2$.

Furthermore, the nonvanishing linear losses resulting from the decay of the ground-state coherence lead to an additional noise contribution to the linewidth

$$\Delta\nu_1 = \frac{\pi^2}{8}\,\tau_{\rm gr}^{-1}(\tau_{\rm gr}^{-1} + 2\gamma_0)\,\frac{\hbar\nu}{P_{\rm out}}\,. \qquad (16)$$

This result can easily be interpreted. $P_{\rm out}\tau_{\rm gr}/\hbar\nu$ is equal to twice the number of Stokes or anti-Stokes photons in the cell. As in a usual laser, photon correlations are maintained over a time equal to the number of photons multiplied by the time a single photon stays in the system [19]. The latter time is here given by the group-delay time (if γ_0 is sufficiently small). If the lifetime of the dark state becomes shorter than the group delay, the phase information carried by a photon is lost faster and $\tau_{\rm gr}^{-1}$ is dominated by $2\gamma_0$. Thus the minimum linewidth is ultimately determined by the lower-level coherence decay.

Similar to the case discussed in Ref. [14] for the intracavity system, the present results for the frequency locking [Eq. (7)] and the linewidth [Eqs. (13) and (16)] are a consequence of the large atomic dispersion associated with two-photon resonances in phase coherent media. In the limit of long-lived ground-state coherences, the beat-note linewidth can be extremely narrow. At the same time the resonantly enhanced nonlinearity makes it possible to achieve oscillation with very low pump powers.

In order to see, whether the small intrinsic linewidth can indeed be exploited, we now estimate the influence of systematic effects. The most serious limitations arise from the ac-Stark shifts, as indicated by Eq. (7). At large values of pump intensities these shifts are large and hence fluctuations in pump powers and frequencies will result in associated broadening of the oscillator linewidth. However, the resonantly enhanced nonlinearity already makes oscillation possible when $E_d^2 \geq \gamma_0\Delta$, i.e., when the near-resonant ac-Stark shift E_d^2/Δ exceeds the ground-state coherence decay γ_0. Thus, with stabilized pump frequencies and intensities, technical fluctuations of the beat frequency due to ac-Stark shifts could be several orders of magnitude smaller than γ_0. In the experiment of Ref. [1], for instance, short-term linewidth values below 100 Hz have been measured even though the transient time broadening of a Raman transition was about 50 kHz. It is clear that observation of quantum-limited linewidth of the oscillator is most likely in the regime of ultralow pump intensities. It is, however, this regime which is of main interest for quantum control and manipulation of quantum properties of few photon fields [10].

In conclusion, we have demonstrated that resonant nonlinear interactions involving atomic coherence can be used for the efficient generation of nonclassical photon fields with a stable and narrow beat-note linewidth and small pump requirements. We expect these features to be of interest in many areas of quantum and nonlinear optics.

The authors gratefully acknowledge useful discussions with P. Hemmer, V. Sautenkov, and A. Zibrov, and the support from the National Science Foundation, the Office of Naval Research, and the Welch Foundation. M. F. would like to thank the Institute for Theoretical Atomic and Molecular Physics at the Harvard-Smithsonian Institution for their hospitality during his stay.

[1] A. S. Zibrov, M. D. Lukin, and M. O. Scully, Phys. Rev. Lett. **83**, 4049 (1999).
[2] A. Yariv and D. Pepper, Opt. Lett. **1**, 16 (1977).
[3] Y. Silberger and I. Bar-Joseph, J. Opt. Soc. Am. B **1**, 662 (1984); I. Bar-Joseph and Y. Silberger, Phys. Rev. A **36**, 1731 (1987).
[4] G. Khitrova *et al.*, Phys. Rev. Lett. **60**, 1126 (1988).
[5] D. Gauthier *et al.*, Phys. Rev. Lett. **61**, 1827 (1988); **64**, 1721 (1990).
[6] P. R. Hemmer, D. P. Katz, J. Donoghue, M. Cronin-Golomb, M. S. Shahriar, and P. Kumar, Opt. Lett. **20**, 982 (1995).
[7] S. E. Harris, Phys. Today **50**, No. 7, 36 (1997).
[8] M. Jain, H. Xia, G. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996).
[9] M. D. Lukin, P. R. Hemmer, and M. O. Scully, in *Advances in Atomic, Molecular and Optical Physics,* edited by B. Bederson and H. Walther (Academic Press, Boston, 1999), Vol. 42B, p. 347.
[10] M. D. Lukin, A. B. Matsko, M. Fleischhauer, and M. O. Scully, Phys. Rev. Lett. **82**, 1847 (1999).
[11] L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature (London) **397**, 594 (1999).
[12] O. Schmidt, R. Wynands, Z. Hussein, and D. Meschede, Phys. Rev. A **53**, R27 (1996).
[13] M. M. Kash *et al.*, Phys. Rev. Lett. **82**, 5229 (1999).
[14] M. D. Lukin, M. Fleischhauer, M. O. Scully, and V. L. Velichansky, Opt. Lett. **23**, 295 (1998).
[15] See also M. Fleischhauer, U. Rathe, and M. O. Scully, Phys. Rev. A **50**, 1748 (1994).
[16] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).
[17] H. P. Yuen and J. H. Shapiro, Opt. Lett. **4**, 334 (1979).
[18] M. Fleischhauer and Th. Richter, Phys. Rev. A **51**, 2430 (1995).
[19] This is also valid for so-called bad-cavity lasers; see S. J. M. Kuppens, M. P. van Exter, and J. P. Woerdman, Phys. Rev. Lett. **72**, 3815 (1994).