Nondegenerate Parametric Self-Oscillation via Multiwave Mixing in Coherent Atomic Media

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We demonstrate an efficient nonlinear process in which Stokes and anti-Stokes components are generated spontaneously in a multilevel medium coherently prepared by resonant, counterpropagating fields. A medium of this kind combines large nonlinear gain and efficient intrinsic feedback, thereby allowing for very weak optical fields to induce nonlinear processes. Mirrorless self-oscillation induced by microwatts of light power (nanojoules of pulse energy) is observed.

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The possibility of mirrorless oscillation in a nonlinear system involving counterpropagating fields was suggested by Harris over 30 years ago [1]. We report here an experiment in which such spontaneous parametric oscillation is induced by a pair of very weak fields in optically dense resonant multilevel media. Such generation does not involve an optical cavity and manifests itself as Stokes and anti-Stokes components generated with a frequency shift corresponding to that of the two-photon transition. As the oscillator goes over threshold, dramatic intensity increase and frequency narrowing of the generated light takes place. These results show that a combination of a steep dispersion and resonantly large nonlinearities associated with electromagnetically induced transparency can cause striking nonlinear processes to be induced by a light field corresponding to μW of power and nJ of pulse energy.

The present work builds on recent theoretical and experimental studies demonstrating that the optical properties of dense ensembles of coherently prepared multistate atoms can be drastically altered. Examples include making an opaque medium transparent over the distances corresponding to several thousand absorption lengths [2], reduction of group velocities to few tens of meters per second [3,4], lasing without population inversion [5], and a large enhancement of nonlinear optical effects [2,6–9].

Processes of the kind demonstrated in the present Letter open the door for a new regime of quantum nonlinear optics, which can potentially be used to study interactions of very low-energy light fields without the need of high-Q cavities [10]. Recent theoretical work indicates that this regime can be used for few photon quantum control [11], switching [12], and quantum noise correlation [13]. Likewise, narrow-linewidth signals may be of interest for optical magnetometry and frequency standards.

Before proceeding we emphasize that the notion that a nonlinear system can self-oscillate is not new. Although the original proposal [1] based on nondegenerate frequency mixing has not been realized up to now due to small values of nonlinearities in available materials and, especially, difficulties in achieving phase matching [14], a number of other relevant experiments have been carried out. In fact, it is much easier to achieve mirrorless oscillation in degenerate four-wave mixing. The possibility of self-oscillation in such interactions has been predicted in [15], and it has later been identified as a generic source for instabilities arising in interactions of powerful laser beams with alkali vapor [16]. A number of such nearly degenerate instabilities have been observed in a vapor driven by very strong (at a level of hundreds of mW), off-resonant fields [17]. Workers in the field have also noted the importance of two-photon nonlinearities in studies of polarization instabilities [18]. In this work instabilities were seen with tens of mW laser power.

As compared to this earlier work the present results utilize atomic coherence effects in a resonant, strong coupling regime [2,6–9] in which nonlinearities cannot be derived from a usual perturbation expansion. In this regime quantum interference allows one to eliminate resonant losses and drastically increase the relevant nonlinearity. Similar to the earlier experiments, oscillation occurs due to effective feedback created by two counterpropagating driving fields. In a medium with a very slow group velocity, such a feedback corresponds to an effective "cavity" with a lifetime corresponding to a group delay time τ_g [19], which exceeds the free-space delay time by a factor $\eta \sim 10^6$ [4]. Such a steep dispersion allows for automatic phase matching in an intrinsically phase-mismatched configuration. The resulting oscillation frequency is governed by a frequency pulling equation and is tightly locked to a light-shifted two-photon transition frequency.

Because of the combination of resonant nonlinearities and steep dispersion, laser power required to observe oscillation was several orders of magnitude lower than in any of the experiments reported so far. Furthermore, oscillation here occurs in a nondegenerate configuration, which is intrinsically phase mismatched. Unlike the important earlier work [18] it does not involve polarization instabilities and also does not, in general, occur in the form of a cone. Furthermore, the present work demonstrates a dramatic narrowing of the generated signals at

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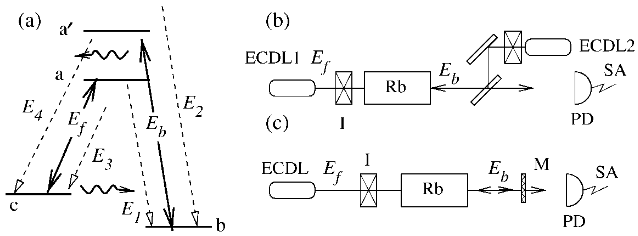

FIG. 1. (a) A prototype four-level model for self-oscillations. In general, we assume that each driving field couples both of the ground states. The upper levels of this double-Λ system can represent some manifolds of states. In the experiment states c and b are hyperfine sublevels of the Rb ground state $5S_{1/2}$. (b),(c) Experimental setups (schematic).

oscillation threshold (without a cavity) as well as tight frequency locking to the two-photon resonance.

We first observed the oscillation as the result of simple Fresnel reflection from the rear window of the Rb cell. Figure 1b shows the experimental arrangement used in most experiments, and Fig. 1c shows the simplest experimental configuration that produced oscillation. In the setup of Fig. 1b a 794 nm beam from an extended-cavity diode laser [tuned to the vicinity of $F = 2(1) \to F' = 3(2)$ transition of $\mathrm{Rb}^{85(87)}$] passes successively through an optical isolator (I), a focusing lens, a heated 5 cm long Rb cell, and onto a fast photodiode (PD). A beam from another extended-cavity diode laser (794 or 780 nm) propagates in the opposite direction. The signal from the photodiode is detected using a microwave spectrum analyzer (SA). With proper tuning of laser frequency, the backward beam induces the oscillation in the system. When the self-oscillation occurs, the detected Raman beat note signal at a frequency of hyperfine splitting (ω_{hfs}) increases in amplitude by as much as 60 dB and its linewidth narrows from 200 kHz to less than 300 Hz (Fig. 2a). Under appropriate conditions the beat note linewidths as narrow as 100 Hz FWHM were observed (Fig. 3a). This is much narrower than the usual broadening mechanisms for two-photon transitions under the present conditions (primarily, transit broadening $\gamma_{bc} \sim 50$ kHz and power broadening $\sim$500 kHz). The oscillation occurs without any cavity enclosing the cell. We have been careful to eliminate possible extraneous sources of feedback to lasers or other optical and electronic elements.

We found that the oscillation occurs readily if the forward and backward fields are tuned to the different ground state hyperfine level as diagramed in Fig. 1a. It is much more difficult to make the system oscillate if the backward beam is tuned to the same frequency as the forward beam. If tuned to different ground state hyperfine levels, the oscillation was observed with the backward beam coupling either the same ($P_{1/2}$) or different ($P_{3/2}$) upper-state fine-structure levels as the forward beam. In our two-laser experiments it was easy to see oscillation for both ^{85}Rb and ^{87}Rb isotopes. When oscillating, the Rb vapor can convert as much as 4% of the total input power into the frequency shifted Stokes and anti-Stokes sidebands. In the two-laser experiments oscillation was observed for forward laser powers 300 μW–15 mW, spot sizes 0.3–2 mm, and cell temperatures 65–110 °C. When both of the driving lasers were close to respective single-photon resonances of D_1 and D_2 lines and the power of the forward (795 nm) laser was about 5 mW, oscillation was observed with backward (780 nm) power as low as a few μW (Fig. 3b). Temperature for this observation was $\sim$110 °C. Oscillation has also been seen when the intensity of such a weak backward beam was turned on and off with a chopper at a rate of up to a few kHz. This indicates that backward pulse energies of order of nJ are sufficient to induce oscillation.

The oscillation frequency shift (ω_0) does change somewhat with laser tuning (typically $\sim$30 Hz/MHz of laser tuning) and with an angle between the forward and backward beams [20]. However, the oscillation frequency always remains within the bandwidth (few hundred kHz) of the power broadened and shifted transparency window [9]. In general, oscillation occurred readily when two beams had identical circular polarizations, and generated

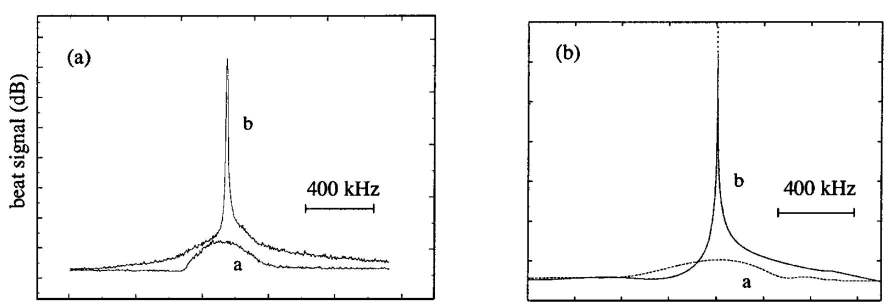

FIG. 2. (a) A typical signal recorded by a fast photodiode. Curve a is recorded with only forward driving beam present. Self-oscillations occur in the presence of the forward and backward driving fields (curve b). Parameters are as follows: cell temperature 92 °C; forward driving beam with power 10 mW and spot size 1.5 mm is detuned by 80 MHz to the red side of the $F = 3 \to F' = 3'$ transition of the D_1 line; backward driving beam with power 2.5 mW and spot size 1.5 mm is detuned by 800 MHz to the blue side of the $F = 2 \to F'' = 3''$ transition of the D_2 line. Doppler broadening is $\sim$500 MHz. (b) Calculated signals corresponding to the experimental conditions of (a). (At the point corresponding to parametric oscillation linear theory predicts infinite growth.)

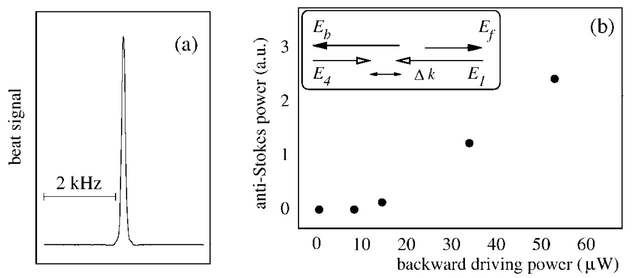

FIG. 3. (a) A typical beat signal at 3.034 GHz recorded when the frequency of the driving laser is locked to a reference cavity. (b) A typical dependence of the generated anti-Stokes forward power as a function of the backward driving power in a vicinity of oscillation threshold. Inset illustrates wave vector mismatch. If all fields are propagating along the z axes $[k_0^F + k_0^B - k_1 - k_4]_z = 2\omega_0/c$.

beams had polarizations identical to those of driving beams. It was possible, but more difficult to see oscillations with other polarization configurations. Polarization instabilities have not been observed. We have also analyzed spatial properties of the generated beams. In the vicinity of oscillation threshold the shapes of generated beams were similar to Gaussian, and the directions of their propagation coincided with those of copropagating driving beams. Far above threshold, significantly richer spatiotemporal dynamics was observed [20].

We have analyzed the characteristics of the forward and backward beams by making beat notes with independently tuned laser sources, and by using optical cavities to analyze the spectra. We found that the field components at frequencies of the forward and backward driving fields (ν_f or ν_b) are surrounded by generated first order Stokes and anti-Stokes fields at frequencies $\nu_{f,b} \pm \omega_0$. In certain cases second order components have been seen as well. The generated components produce, in general, an asymmetric spectrum. In particular, in cases when forward driving field is tuned to, e.g., upper ground state hyperfine sublevel and backward driving beam is tuned to the lower hyperfine sublevel, the anti-Stokes component observed in a forward direction is much more ($\sim$10–20 dB) intense than the Stokes one.

Motivated by experimental results, we consider a theoretical model in which atoms in a double Λ-type configuration are interacting with six optical fields (Fig. 1a). These include two counterpropagating driving fields with frequencies ν_F, ν_B and complex slowly varying amplitudes $\mathcal{E}_F$ and $\mathcal{E}_B$; anti-Stokes and Stokes components with frequencies $\nu_{1,3} = \nu_F \pm \omega_0$ propagating in the forward direction ($\mathcal{E}_1, \mathcal{E}_3$), and corresponding components with frequencies $\nu_{2,4} = \nu_B \pm \omega_0$ propagating in the backward direction ($\mathcal{E}_2, \mathcal{E}_4$). Below we focus on the linear theory describing the oscillation threshold. Hence, all generated components are treated to first order only and saturation effects are disregarded. The driving fields, however, are treated to all orders, in contrast to the usual χ^3 theories. For the present problem the relevant linear and nonlinear polarizations were calculated from density matrix equations for each velocity group and averaged over Maxwellian velocity distribution. In the present calculations we consider fields interacting in a slab of medium of the length L. Assuming that all fields are in single spatial modes, we restrict ourselves to an effective 1D problem and numerically integrate the appropriate Maxwell equations. The boundary conditions are taken to include a weak "seed" input ($\mathcal{E}$) at anti-Stokes frequencies (corresponding to, e.g., spontaneous emission, or vacuum field).

We first illustrate the origin of the oscillation. To this end, let us assume that absorption of the driving fields is negligible, and there is no inhomogeneous broadening. Furthermore, we consider a special case in which forward driving field is resonant with optical transition $c \rightarrow a$ and the backward driving field is close to resonance with transition $b \rightarrow a'$ (Δ_B is a single photon detuning) and disregard the coupling of the driving fields with all other transitions. In such a situation only forward anti-Stokes ($\mathcal{E}_1$) and backward Stokes ($\mathcal{E}_4$) fields are involved in nonlinear interaction as shown in Fig. 1a. In this case weak fields evolve according to $c\partial\bar{\mathcal{E}}_i/\partial z = a_{ij}\bar{\mathcal{E}}_j$, with $\{i,j\} = \{1,4\}$ and $a_{11} = -\eta[\gamma_{bc} + i(\omega_0 - \omega_{\mathrm{hfs}}) + i|\Omega_B|^2/\Delta_B] - i\Delta k$, $a_{14} = i\eta[\Omega_B\Omega_F/(\Delta_B)]$, $a_{41} = i\eta 4[\Omega_B^*\Omega_F^*/(\Delta_B)]$, $a_{44} = i\eta|\Omega_F|^2)/\Delta_B$. Here we assumed equal dipole elements and radiative decay rates γ_{rad} on all transitions. $\eta = 3/(4\pi^2)N(\lambda)^2\gamma_{\mathrm{rad}}c/|\Omega_F|^2$, where λ is optical wavelength, N is atom density, and $\Omega_{F,B}$ are Rabi frequencies. In the relevant limit $\eta \gg 1$, this quantity corresponds to the ratio of c to the group velocity. $\Delta k = [\vec{k}_1 + \vec{k}_4 - \vec{k}_F - \vec{k}_B]_z$ is a wave vector mismatch. Above equations contain cross-coupling terms between two fields resulting in four-wave mixing gain [21], and the loss term, which is proportional to the dephasing rate of ground state coherence γ_{bc}. The latter can be made extremely small for dipole-forbidden transition.

When the phase matching condition is satisfied [Im$(\delta a) = 0$, $\delta a \equiv (a_{11} - a_{44})/2$], we find

$$\mathcal{E}_1(L) \sim \mathcal{E}_4(0)^* \sim \frac{\mathcal{E}}{\delta a \sin(sL/c) - s\cos(sL/c)}, \quad (1)$$

where $s = \sqrt{a_{14}a_{41} - (\delta a)^2}$, and the unimportant proportionality constants have absolute values of the order of unity. These solutions diverge if $\tan(sL/c) = s/(\delta a)$, which indicates the onset of mirrorless oscillations. Note that the latter condition can be satisfied if $|\Omega_F\Omega_B| > \gamma_{bc}|\Delta_B|$, which is identical to a strong coupling condition of Refs. [2,6–9,11–13]. When dephasing of ground state coherence can be disregarded, $\gamma_{bc} \rightarrow 0$, the oscillation condition becomes

$$\frac{|\Omega_B|\,|\Omega_F|}{\Delta} = \frac{\pi}{2}\,\frac{c}{\eta L} \equiv \frac{\pi}{2}\,\frac{1}{\tau_g}, \quad (2)$$

i.e., in the limit of large group delay τ_g and small detuning Δ a tiny laser power is sufficient to induce oscillation. As shown in [10,13] the above condition corresponds to only a few photons per atomic cross section in (at least) one of the beams. We now examine the phase matching condition. Close to the two-photon resonance we have

$$\eta/2[\omega_0 - \omega_{\mathrm{hfs}} - (|\Omega_F|^2 - |\Omega_B|^2)/\Delta_B] + c\Delta k = 0 . \tag{3}$$

It is interesting that this equation resembles closely the frequency pulling equation of the usual laser theory, with frequency stabilization coefficient $\eta/2$. The first term in the left-hand side corresponds to atomic dispersion, and the second describes the geometrical phase mismatch. This contribution is proportional to the Raman transition frequency (e.g., for fields propagating along the same direction $\Delta k = 2\omega_0/c$, Fig. 3) and also depends on relative angles between driving beams. Hence it plays a role analogous to the cavity shift. Note, however, that under the typical oscillation conditions stabilization coefficient $\eta \gg 1$ and the oscillation frequency is tightly locked to the light-shifted Raman transitions frequency.

In the process considered above scattering of Stokes and anti-Stokes fields into each other results in an effective feedback, and this scattering is also accompanied by parametric amplification. Hence self-sustained oscillation can occur. Other nonlinear processes can also contribute to oscillation. When only a forward driving field is present nonlinear interaction results in the coupling between copropagating Stokes and anti-Stokes fields leading to coherent Raman scattering and amplification of the copropagating pair of fields in the vicinity of two-photon resonance [7,8]. Oscillation is not possible in this case, since no effective feedback is present. However, when coherent Raman scattering exists in addition to the coupling between counterpropagating Stokes and anti-Stokes components, it can result in lowering the oscillation threshold. There exists also a process leading to the scattering of the counterpropagating anti-Stokes (or Stokes) waves into each other, which does not change the total photon number in generated fields. Consequently, it alone can never lead to the oscillations. However, oscillations can emerge if in addition to the parametric energy exchange additional amplification mechanisms (e.g., coherent Raman scattering) are present.

In general, for the detailed comparison of the theory and experiment all such processes should be taken into account. They give rise to simultaneous generation of all components in both directions. To make a comparison we have solved the full system of propagation equations numerically, taking into account Doppler broadening, and propagation of all fields. The results (Fig. 2b) show good qualitative agreement with experiments.

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[20] When the gain is high it is possible to have a comb of oscillations with spacings between modes that could be tuned from tens to hundreds of kHz. This subsidiary comb structure depends strongly on the angle of the return beam and the laser frequency, and is correlated with spatial mode changes in the generated light.
[21] We note that phase insensitive Stokes gain is negligible in a resonant situation.