Realization of a Continuous-Wave, Two-Photon Optical Laser

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(Received 15 October 1991)

We report the first observation of continuous-wave two-photon lasing in the optical regime, and demonstrate that its initiation requires the injection of a trigger pulse into the laser resonator. Successful operation of the two-photon laser relies on the use of a novel gain medium consisting of laser-driven, two-level atoms and the use of a high-finesse optical cavity to isolate the two-photon gain from competing processes. Threshold conditions for laser action are in good agreement with recent theoretical predictions.

PACS numbers: 42.50.Hz, 42.55.Hq, 42.65.Dr, 42.65.Pc

Many of the unique properties of lasers derive from those of the stimulated emission (SE) process on which they are based. Virtually all existing lasers are based on the one-photon SE process—a SE event results in the creation of one new photon. Early on in the laser era, it was suggested [1] that lasers based on higher-order SE processes, wherein each SE event results in the creation of two or more photons, may be possible. It was predicted [2,3] that lasers based on higher-order SE processes possess operational characteristics qualitatively different from those found in normal (one-photon-SE-based) lasers. Unfortunately, tests of these predictions have not been possible, since efforts to realize lasers based on higher-order SE have themselves met with limited success [4]. A primary obstacle to the realization of higher-order-SE-based lasers is the tendency of higher-order SE processes to be weak both in absolute terms and in comparison to one-photon SE processes. In the microwave regime, the use of a unique gain medium and an extremely high-Q resonator has allowed researchers to achieve continuous-wave (cw), two-photon masing [5]. While masers are interesting in their own right, many of the intriguing predictions concerning two-photon lasers cannot be tested with them because their low-frequency photons are difficult to detect.

Recently, it has been demonstrated [6] that strongly driven two-level atoms display two-photon gain, and it has been predicted [7] that this gain can, under reasonable experimental conditions, be useful in the realization of a two-photon laser. We report in this Letter on the use of a driven-atom gain medium to provide the first demonstration of cw two-photon lasing in the optical regime. The two-photon laser operates in the degenerate mode (both photons generated in the SE process have the same frequency) and displays dynamics that are dramatically different from those found in the case of normal one-photon-gain-based lasers.

As discussed in detail elsewhere [6,7], the two-photon gain that arises in the driven two-level-atom system can be understood simply using the dressed-atom picture [8]. The dressed-atom energy eigenstates are shown in Fig. 1(a), where it is seen that the dressed-state doublets are separated in energy by $\hbar\omega_d$, where ω_d is the driving-field frequency, and are split by $\hbar\Omega'_d$, where Ω'_d is the generalized Rabi frequency of the pump field. As usual, $\Omega'^2_d = \Omega^2_d + \Delta^2_d$, where Ω_d is the resonant Rabi frequency and $\Delta_d = \omega_d - \omega_0$ is the detuning of the driving field from the atomic transition frequency ω_0.

For a nonzero atom–driving-field detuning, the populations among the dressed states are imbalanced giving rise to n-photon inversions ($n = 1, 2, \ldots$) on various transitions. In particular, for $\Delta_d < 0$, the populations are imbalanced as indicated in Fig. 1(a), and degenerate two-photon gain occurs at $\omega_d - \Omega'_d/2$. Since this frequency is removed from spectral regions of strong one-photon gain [9], it is possible to selectively enhance two-photon gain using a high-finesse optical cavity. Note that the periodic structure of the dressed-atom energy spectrum guarantees relatively strong, near resonantly enhanced, degenerate two-photon transitions. These factors, together with a paucity of competing one-photon processes, make the driven-atom gain medium unusual in comparison to previously investigated two-photon gain media. We note that a gain medium (Rydberg atoms) of similar character [5] was used in the first realization of two-photon maser operation.

A schematic diagram of our apparatus is shown in Fig. 1(b). The two-photon laser consists of a spherical-mirror confocal cavity of length $L_c = 5$ cm through whose center passes a collimated ≈ 1.5-mm-diam atomic beam of barium. The barium atoms are driven by a traveling-wave laser beam as they pass through the center of the cavity. The atomic-beam axis, the cavity axis, and the pump-beam axis are mutually orthogonal, thereby providing for nearly Doppler-free atom-pump and atom-cavity

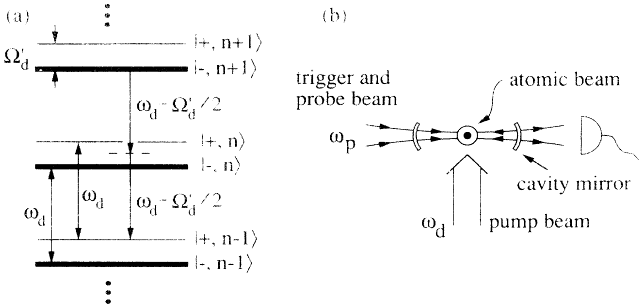

FIG. 1. (a) Dressed-atom doublets represented by lines whose thicknesses indicate the relative populations. Degenerate two-photon gain occurs at $\omega_d - \Omega'_d/2$. (b) Schematic of the two-photon laser.

interactions and ensuring that nonlinear-wave-mixing-type processes are not phase matched for emission into the cavity modes. The lasing processes of interest here do not exhibit a phase-matching requirement.

The pump laser, a ring dye laser, is actively stabilized (linewidth ≈ 1.5 MHz), locked to the 553.5-nm ${}^1S_0 \rightarrow {}^1P_1$ transition (natural linewidth $\gamma/\pi = 19$ MHz) of a barium reference cell, and has a full width at half maximum (FWHM) beam diameter of ≈ 2 mm as it passes through the center of the cavity. Typically, $\Omega_d/2\pi \approx 410$ MHz at the center of the beam. A second actively stabilized dye laser is employed to produce a probe beam that propagates along the cavity axis as needed. All laser fields are linearly polarized along the atomic-beam propagation direction.

The atomic-beam source nozzle (collimating aperture) has a 1.6-mm (1-mm) diameter and is located 28.5 cm (1.5 cm) away from the cavity axis. The source nozzle (barium reservoir) has a maximum operating temperature of 1180 °C (1060 °C). At these temperatures, the atomic beam displays a residual Doppler width of ≈ 48 MHz and an unsaturated single-pass linear absorption of $(60 \pm 5)\%$ at the central frequency of the ^{138}Ba (78% natural abundance) transition. We estimate that the maximum ^{138}Ba atomic-beam density η is 1.0×10^{10} atoms cm^{-3} using $\eta = -(\gamma_D/\gamma)(2\pi/3\lambda^2)\ln(1-A)/d_a$, where $2\gamma_D$ is the residual Doppler width, λ is the transition wavelength, A is the measured absorption, and d_a is the atomic-beam diameter at the cavity.

Cavity-decay-time measurements indicate a fundamental-mode linewidth of $\Gamma_c/\pi = 1.7 \pm 0.1$ MHz (FWHM), implying a finesse of 1800 (using $c/2L_c$ as the fundamental-mode free spectral range). An aperture of 650 μm (1.6 mm) diameter is placed on one (the other) cavity mirror to lower the finesse of nearly degenerate, high-order transverse cavity modes [10]. With excitation of all cavity modes permitted by the apertures, the cavity displays a linewidth of 4 MHz (FWHM). The cavity mirror spacing (and hence resonant frequency) can be piezoelectrically scanned. The frequency interval between adjacent longitudinal cavity modes is sufficiently large ($c/4L_c = 1.5$ GHz) compared to other relevant frequency intervals that all save one can be ignored. Only those driven atoms located within a cylindrical volume of length ≈ 1.5 mm (the atomic-beam diameter) and diameter $2w_0 = 120$ μm (the cavity-mode diameter) couple significantly to the cavity. The number of atoms in this volume is $\approx 2.0 \times 10^5$.

Before describing our experimental results, we provide a simple motivation for the dramatic differences between the switch-on behavior of one- and two-photon lasers. In general, lasing will commence when the round-trip gain equals the round-trip loss. Applying this condition to the one-photon laser implies that it will turn on when a uniquely defined minimum number of atoms, determined by various system parameters, are present in the cavity. The situation becomes more complicated when the same condition is applied to the two-photon laser because the two-photon gain increases not only with the number of active-gain atoms N, but also with the intracavity light intensity (until saturation occurs). A threshold number of atoms N_{th} can be defined as the number of atoms needed to satisfy the gain-equals-loss condition with an intracavity intensity just sufficient to saturate the two-photon gain. However, even if one starts with $N > N_{\text{th}}$ atoms in the active volume, gain will not exceed losses until some critical intracavity light intensity is reached. If the laser is initially off, it cannot turn on unless a fluctuation brings the intracavity light intensity above the necessary critical value [11]. Alternatively, a field may be injected into the cavity to start the two-photon laser [3].

Specific predictions for the threshold conditions for dressed-state two-photon lasers have been made in theoretical studies [7] of a model system resembling our experimental one. In the model system, the gain medium consists of a collection of naturally broadened two-level atoms contained in a single-mode cavity characterized by an atom-cavity-mode coupling coefficient g that is essentially constant over the active gain volume, e.g., a unidirectional ring cavity. The driving field is assumed to be of constant amplitude and possess a resonant Rabi frequency much larger than the cavity linewidth $2\Gamma_c$ and spontaneous emission linewidth 2γ. The theory thus neglects the spatial variations in g actually present in our standing-wave cavity, the spatial inhomogeneity of the driving field, Doppler broadening, and the presence of nearly degenerate transverse cavity modes. Despite this fact, comparison of the theoretical and experimental results serves to test our basic interpretation of the observations.

The two-photon laser calculations indicate that N_{th} is minimized for a given Ω_d when $\Delta_d \approx -0.4\Omega_d$. At the optimum detuning

$$N_{\text{th}}^{\text{min}} \approx 25.4\,\Omega_d\Gamma_c/g^2 . \tag{1}$$

Also, the number of intracavity photons expected with the two-photon laser operating slightly above threshold with the pump at its optimum detuning is given by

$$n_{\text{th}}^{\text{min}} \approx 4.7\,\Omega_d\gamma/g^2 . \tag{2}$$

We estimate that the atom-cavity coupling constant, spatially averaged over the standing-wave pattern, is equal to 4.0×10^6 rad/sec, where we have used the relation $g = (6\gamma\lambda^2 c/\pi V_{\text{eff}})^{1/2}$ and the effective fundamental-mode volume $V_{\text{eff}} = \pi w_0^2 L_c$. Applying conditions (1) and (2) to our experimental configuration, we find that $N_{\text{th}}^{\text{min}} \approx 2.2 \times 10^4$ and $n_{\text{th}}^{\text{min}} \approx 4.5 \times 10^4$. The possibility of obtaining two-photon laser action is indicated by the fact that values of N in excess of $N_{\text{th}}^{\text{min}}$ are available in our apparatus.

Figure 2(a) shows the total optical power emitted out one end of the laser cavity as a function of the cavity-pump-laser detuning $\Delta_c = \omega_c - \omega_d$, where ω_c is the empty-cavity resonance frequency. In this case, $\Omega_d/2\pi$

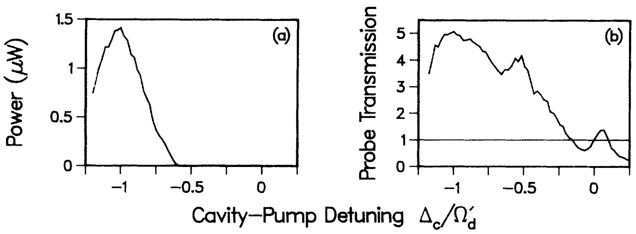

FIG. 2. (a) Power emitted out one end of the cavity as a function of the cavity-pump-laser detuning Δ_c for $\Omega_d/2\pi \approx 410$ MHz, $\Delta_d/2\pi \approx -160$ MHz, and $\Omega_d'/2\pi \approx 440$ MHz. The observed signal is due to one-photon-based lasing. Note that there is no power emitted for $\Delta_c = -0.5\Omega_d'$. (b) Maximum value of the power emitted from the cavity as a saturating probe beam (Rabi frequency ≈ 240 MHz) is scanned through the cavity resonance for various values of the cavity frequency. Other parameters are the same as in (a). The pronounced, probe-dependent feature at $\Delta_c = -0.5\Omega_d'$ is due to two-photon gain.

≈ 410 MHz, $\Delta_d/2\pi \approx -160$ MHz, $\Omega_d'/2\pi \approx 440$ MHz, and N is maximized. The output power observed in this measurement arises from one-photon-gain-based laser oscillation [9], as is seen from the fact that the maximum output power occurs for $\Delta_c \approx -\Omega_d'$, corresponding to the peak of driven-atom one-photon gain. In addition, we have found that the laser threshold behavior is consistent with that expected for one-photon laser oscillation. Importantly, one-photon lasing is not observed for $\Delta_c = -\Omega_d'/2$, the frequency at which we expect maximum two-photon gain, despite the fact that the one-photon laser is far above threshold for $\Delta_c \approx -\Omega_d'$.

To make two-photon as well as one-photon gain visible, a cw probe laser (approximately matched to the fundamental cavity mode and resonant with it) is injected into one end of the cavity. The power emitted out the opposite end of the cavity is plotted versus Δ_c in Fig. 2(b). The intracavity resonant Rabi frequency of the probe beam is set to ≈ 240 MHz and other experimental parameters are the same as those used in Fig. 2(a). One-photon lasing occurs over a portion of the one-photon gain region. This signal is also present in the absence of a probe, as shown in Fig. 2(a). The vertical scale is calibrated so that unit transmission represents the empty-cavity probe transmission. It is seen that the output power has a pronounced peak near $\Delta_c = -\Omega_d'/2$. We attribute this probe-dependent feature to two-photon gain.

To determine whether the two-photon gain depicted in Fig. 2(b) is large enough to support two-photon lasing, the cavity resonance frequency is tuned near the peak of the two-photon gain feature ($\Delta_c/2\pi \approx -210$ MHz) and a trigger pulse, acousto-optically gated from the probe laser, with a duration of 1.4 μsec, transition times of 18 nsec, and a carrier frequency coincident with the cavity resonance frequency, is injected into the cavity. The time-resolved cavity output just prior to and following the trigger pulse is recorded. The measurement is repeated for various trigger-pulse powers. In preparation for the next trigger pulse, the pump field is briefly blocked using a mechanical shutter to quell any preexisting two-photon lasing.

The time-resolved cavity output powers are shown in Figs. 3(a)–3(c) for various trigger-pulse powers. In Fig. 3(a), the trigger-pulse power is apparently insufficient to boost the intracavity photon number to the critical level [see condition (2)] needed to initiate two-photon lasing. As a result, the cavity output power decays to zero after the trigger pulse is turned off. Spiking behavior, apparently unaccounted for in published theoretical treatments of two-photon lasing, is clearly evident. We estimate that the photon number n in the cavity after the initial spiking is approximately 3.4×10^4 photons using the relation in $n = P_{\text{out}}L_c/cT\hbar\omega_c$, where P_{out} is the observed output power and $T = 1.6\times10^{-3}$ is the transmission of the output mirror. In Fig. 3(b), the effect of a somewhat higher-power trigger pulse, providing for $\approx 5.4\times10^4$ intracavity photons, is shown. After the trigger pulse, the cavity output power remains high. The trigger-induced transition to a state of nonzero cavity output power is entirely consistent with the expected switch-on behavior of a two-photon laser [3]. We thus conclude that the signal to the right of the trigger pulse in Fig. 3(b) results from two-photon laser operation. Following switch-on, the two-photon laser is found to remain on with an essentially constant intensity until the pump laser is turned off momentarily or the system experiences some other external perturbation. Figure 3(c) is a longer-time-scale

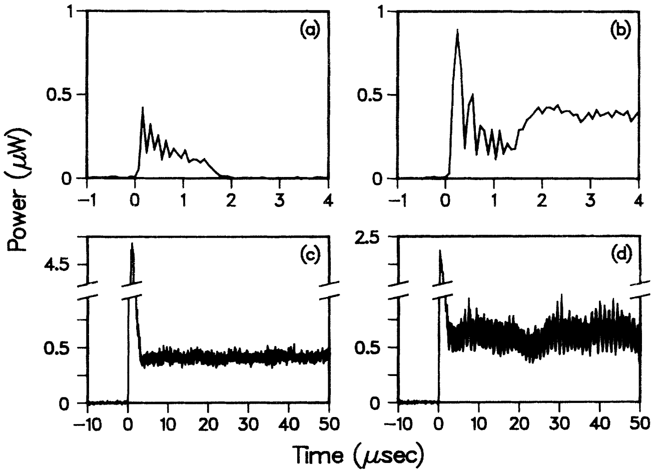

FIG. 3. Time-resolved power emitted from the cavity before and after a 1.4-μsec trigger pulse is injected into the cavity for various values of the injected power. In (a)–(c), $\Delta_c \approx -210$ MHz and the other parameters are the same as in Fig. 2. (a) Number of intracavity photons after the initial gain spiking is approximately 3.4×10^4. (b) The two-photon laser switches on; $n \approx 5.4\times10^4$. (c) $n \approx 1.3\times10^6$. (d) Oscillatory behavior of the two-photon laser for $\Delta_d \approx -140$ MHz, $\Delta_c \approx -197$ MHz. Note the change in scale of the horizontal axis and the break in the vertical axis for (c) and (d). The sampling interval is 80 nsec.

recording of cavity output power in the case of high-power ($\approx 1.3\times 10^6$ intracavity photons) trigger pulse, where it is seen that there is little spiking during the switch-on.

It should be noted that one-photon gain at the frequency of the two-photon gain feature in Fig. 2(b) contributes to the total gain in the cavity ($\approx 35\%$). This nonideality could be reduced under other, but in our particular case unachievable, experimental conditions. The operational characteristics of the laser appear to be relatively unaffected by the presence of a small amount of one-photon gain. Careful inspection of the intensity data in Fig. 3(c) reveals a small oscillation with a frequency of ≈ 3.8 MHz. For other experimental parameters even more dramatic oscillatory behavior is observed. For example, in Fig. 3(d) where the pump laser is tuned closer to resonance ($\Delta_d/2\pi \approx -140$ MHz) and the cavity is tuned near the peak of the two-photon gain ($\Delta_c/2\pi \approx -197$ MHz), we find well-defined oscillatory components at 1.1 MHz, 2.2 MHz, and a broad set of Fourier components centered at 3 MHz. At this time, the significance of the oscillatory behavior is unclear. It may simply represent beating of distinct transverse cavity modes, or it may arise from an instability driven by competition between the two-photon gain process and the off-resonant one-photon gain process. Future experiments using single-mode cavities are underway to elucidate this behavior.

For pump-laser detunings that are closer to resonance, the separation between the one- and two-photon gain features becomes smaller and the region of one-photon lasing can be made to overlap the region of two-photon gain. Under some conditions, we find that the intensity of the one-photon laser is sufficient to satisfy condition (2). Hence, in this case, the two-photon laser can be continuously self-started by the one-photon laser. Details of these results will be presented elsewhere.

We would like to thank M. Lewenstein and J. Zakrzewski for their theoretical support, Yifu Zhu for his contributions to earlier phases of this work, H. Carmichael and M. Raymer for useful discussions, and P. Sellin for laboratory assistance. This work was supported by the U.S. Army Research Office under Contract No. DAAL03-91-G-0313.

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