Polarization instabilities in a two-photon laser

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We describe the operating characteristics of a new type of quantum oscillator that is based on a two-photon stimulated emission process. This two-photon laser consists of spin-polarized and laser-driven $^{39}$K atoms placed in a high-finesse transverse-mode-degenerate optical resonator, and produces a beam with a power of $\sim 0.2 \, \mu W$ at a wavelength of 770 nm. We observe complex dynamical instabilities of the state of polarization of the two-photon laser, which are made possible by the atomic Zeeman degeneracy. We conjecture that the laser could emit polarization-entangled twin beams if this degeneracy is lifted.

The high degree of temporal coherence of laser light arises from a complex interplay between the fundamental light-matter interactions of absorption, spontaneous emission, and stimulated emission. The coherence properties of the generated light, characterized by the temporal coherence function, can be altered significantly and often in a surprising manner when quantum or nonlinear optical effects are paramount. For example, the threshold condition for all lasers is that the round-trip gain must equal the round-trip loss. For one-photon lasers this yields the well known result that lasing will commence when a uniquely defined minimum inversion density (proportional to the gain) is attained via sufficient pumping. The situation is more complicated for the two-photon laser because the unsaturated gain increases with increasing inversion density and with increasing cavity photon number. This results in a threshold condition specified by a uniquely defined minimum inversion density and cavity photon number so that it cannot turn on unless quantum fluctuations or an injected field brings the intracavity light above the critical value. In addition, once the minimum photon number exists in the cavity, the photon number undergoes a run-away process, growing rapidly until the two-photon transition is saturated. Therefore, the two-photon laser operates in the saturated regime (a source of optical nonlinearity) even at the laser threshold, giving rise to the possibility that the laser will display dynamical instabilities.

The primary purpose of this paper is to present experimental observations of degenerate two-photon optical lasing that is based on laser-driven potassium atoms contained in a very high finesse optical resonator. We verify that the laser displays the expected turn-on behavior and observe that it displays polarization instabilities. Such experiments provide a window into the often conflicting predictions concerning two-photon lasers and will help identify generic two-photon laser properties. In addition, we suggest that this two-photon laser could emit polarization-entangled light states of interest for quantum information applications.

Achieving two-photon lasing is hampered by the smallness of typical two-photon stimulated emission rates and high saturation intensities, and competing nonlinear optical processes often swamp or prevent the occurrence of two-photon lasing. To overcome these difficulties, we follow the general approach of using laser-driven interactions to generate the two-photon gain, and high-quality laser mirrors (developed for cavity quantum electrodynamics experiments) for the optical resonator, as described below.

The two-photon gain medium we use in the experiments consists of a dense, collimated, effusive beam of laser-driven $^{39}$K atoms that passes through the center of the optical resonator orthogonal to the resonator axis, as shown schematically in Fig. 1(b). We use the $D_1$ tran-
sition of $^{39}$K (6 MHz natural linewidth) whose Zeeman hyperfine structure is displayed in Fig. 2. The atomic states are denoted by $|\alpha F_a M_a\rangle$, with $\alpha = g$ for $4^2 S_{1/2}$ and $\alpha = e$ for $4^2 P_{1/2}$ levels. Quantum numbers $F$ and $M$ denote the total angular momentum and its projection on the quantization axis $\hat{z}$, defined by a weak magnetic field ($\leq 1$ Gauss) produced by a small Helmholtz coil set around the cavity. The two-photon stimulated emission rate is enhanced by the smallness of the ground state hyperfine splitting for $^{39}$K of $\Delta g/2\pi = 462$ MHz. To create a population inversion for multiphoton transitions starting from the state $|g22\rangle$, the atoms are continuously optically pumped into this state by two $\sigma_\uparrow$-polarized fields whose frequencies are set close to $\omega_{21}$ and $\omega_{22}$ (Bohr frequencies are denoted by $\omega_{F_e,F_g}$). We note that the homogeneous linewidth of the Raman two-photon transition is $\approx 6$ MHz due to lifetime broadening of the states by the optical pumping beams. The maximum atomic number density in the resonator is $\approx 2 \times 10^{11}$ atoms/cm$^3$, the atomic beam diameter is 2.5 mm, and the measured residual Doppler absorption width is 30 MHz.

The two-photon laser resonator is placed in a vacuum chamber and consists of two high-reflectivity ultra-low-loss mirrors of radius of curvature $R=5$ cm set close together at a distance of $L = 1.464$ cm in a linear (standing-wave) transverse-mode-degenerate sub-confocal configuration. This configuration suppresses normal one-photon lasing that can arise from spectrally nearby gain features [10]. The laser mirrors are mounted on a super-invar ceramic for fine scanning $L$. The mirrors have a transmissivity of $T = 2 \times 10^{-4}$ as measured by the manufacturer (Research Electro-Optics). We find that the cavity finesse $\mathcal{F} = 15, 400 \pm 150 \simeq \pi/(T + A)$ using the ring-down technique, yielding a loss per mirror $A = (4 \pm 2) \times 10^{-6}$.

We use a short cavity length so that the free spectral range ($FSR = c/2L = 10.24$ GHz) is larger than the frequency difference between the two-photon gain feature and all other nearby one-photon gain features, which are within 400 MHz of the two-photon frequency. We set the cavity length precisely to a sub-confocal condition $L = R[1 \pm \cos(\pi/p)]$ with $p = 4$ so that the transverse modes group into degenerate clusters, spaced by $c/2pL = 2.56$ GHz and centered on the longitudinal mode frequencies. Finally, to suppress one-photon lasing on high-order transverse modes (which are no longer frequency degenerate with the lower-order modes due to spherical aberrations [12]), we accurately position under vacuum 400 $\mu$m pinholes in front of each cavity mirror and along the cavity axis using picomotor-actuated translation stages. With the pinholes in place, $\mathcal{F} = 15,140$, resulting in a longitudinal mode linewidth $FSR/\mathcal{F} = 680$ kHz. Within the cavity mode volume, we estimate that there are a maximum of $7 \times 10^{10}$ atoms.

Two-photon stimulated emission into the laser cavity mode (frequency $\omega$) occurs when the optically pumped atoms are illuminated by an intense $\sigma_\uparrow$-polarized Raman pump beam of frequency $\omega_{d}$, and $\omega$ is adjusted to the two-photon transition frequency ($\approx \omega_{d} + \Delta g/2$). Photons are added two at a time to the cavity field via stimulated emission when two photons are annihilated from the $\sigma_\uparrow$-polarized Raman pump field and the atom undergoes a multi-photon transition from the state $|g22\rangle$ to any one of the states $|g1M_g\rangle$, as shown in Fig. 2. Note that such a hyper-Raman transition can be mapped onto the idealized two-photon stimulated emission process shown in Fig. 2(a) by working in a basis in which the multi-level atomic structure is dressed by the Raman pump field. The Raman pump beam has a maximum power of 300 mW and is focused to an elliptical spot size (1/e field radius) of 3 mm along the cavity axis and 250 $\mu$m along the atomic beam axis, achieving a maximum intensity of approximately 25 W/cm$^2$. Throughout the experiments, the pump beam frequency is adjusted so that it is tuned to the blue side of the $\omega_{22}$ transition by 512 MHz. The mutually orthogonal geometry of our experiment suppresses competing phase-matched processes, ensuring that we are constructing a true two-photon laser and not a parametric oscillator.

Our experimental system is very rich because there are 4 quantum pathways, depicted in Fig. 2, that connect the initial state $|g22\rangle$ and the final states $|g1M_g\rangle$, differ only by the state of polarization of the cavity field, and are frequency degenerate at zero magnetic field. The two-photon resonator can support any state of polarization (it is measured to be highly isotropic) hence all pathways of Fig. 2 must be considered.

Experimental evidence for the initiation of two-photon lasing is presented in Fig. 3, obtained using the following procedure. The atomic, optical pumping, and Raman pump beam are turned on and a weak tunable continuous-wave $\sigma_\uparrow$-polarized laser beam, produced by an auxiliary laser, is injected into the two-photon laser resonator. The frequency of the injected beam is scanned repeatedly through the cavity resonance while monitoring the power emanating from the cavity, and the cavity frequency is manually adjusted around the vicinity of the expected two-photon transition frequency ($\omega \approx \omega_{d} + \Delta g/2$). We observe that the emitted power increases when the cavity resonance is set to the two-photon transition frequency and, for sufficiently high injected photon number, two-photon lasing is initiated as indicated by the emitted power remaining high even after the frequency of the injected light has swept through the cavity resonance.

To perform more detailed measurements of the initiation process, we block the auxiliary laser beam, set the cavity frequency to the peak of the observed two-photon transition frequency, momentarily block the Raman pump beam to quell any pre-existing two-photon lasing, and measure the power of the light emitted from the two-photon laser resonator using a avalanche photo-
diode (Hamamatsu C5460, 20 MHz bandwidth). In this situation, there is essentially no light emitted from the resonator, indicating that quantum fluctuations do not provide a sufficient number of photons in the cavity to satisfy both lasing criteria. In an attempt to force the laser to the ‘on’ state, we typically inject a 1.2 µs-long pulse of light into the resonator, which is gated from the auxiliary laser beam using an acousto-optic modulator. Figure 3(a) shows the case when a below-threshold pulse is injected into the resonator. The dashed vertical lines indicate the duration of the injected pulse and we estimate that \( n_{\text{inj}} = 1.1 \times 10^5 \) photons are injected into the cavity. For slightly higher trigger pulse powers [Fig. 3(b)], \( n_{\text{inj}} = 2.2 \times 10^5 \), the cavity photon number grows substantially and remains high for a few microseconds, but is still insufficient to initiate lasing. The fact that the photon number stays high for a duration much longer than the pulse width indicates that we are very close to satisfying both two-photon laser threshold criteria. For \( n_{\text{inj}} = 3.3 \times 10^5 \) [Fig. 3(c)], the power emitted by the cavity rises to approximately 0.2 µW and remains at this value, corresponding to an average cavity photon number of \( 2.2 \times 10^6 \) and an intracavity intensity of 7.3 W/cm² at the cavity waist. We observe that the power emitted from the cavity remains high for up to a few seconds until the frequency of the Raman pump or the cavity drifts off the two-photon resonance or the Raman pump beam is blocked momentarily. Figure 3(d) shows the power emitted from the resonator on a longer time scale for a larger injected number of photons and during a different experimental run but under essentially identical conditions. The experimental data of Fig. 3 gives convincing evidence that we have indeed observed two-photon lasing.

As mentioned earlier, the degenerate hyperfine structure of \( ^{39} \)K opens up the possibility that the laser can operate on different states of polarization. Recall that we trigger the laser to the ‘on’ state using a \( \hat{z} \)-polarized trigger field. To gain some understanding of the subsequent state of polarization, we place a linear polarizer oriented in the \( \hat{z} \)-direction in the beam and before the detector. We find that the two-photon laser displays polarization instabilities even though the total emitted power remains nearly constant on the time scale of the instability. It is seen in Fig. 3(a) that the state of polarization undergoes very regular oscillations of period 0.11 µs, with a 50% depth of modulation. Similar dynamical behavior is observed for all polarizer orientations, suggesting that the state of polarization is elliptical with an ellipticity of 0.5 and a rotating major axis. Additional experiments using multiple detectors to simultaneously record the power emitted in different states of polarization are needed to accurately quantify the laser’s polarization state.

We find that the laser dynamics is quite sensitive to the applied magnetic field: an increase by as little as 0.5 Gauss is sufficient to generate a much more complicated pattern. Figure 3(b) shows the complex, possibly chaotic behavior observed for a magnetic field strength of 2.0 G. An analysis of the data shown in Fig. 3(b) during the interval after which the injected pulse is turned off reveals that the irregular oscillations are characterized by a broad power spectrum and a correlation function that drops to near zero within 100 ns: necessary, but not sufficient conditions for the existence of chaotic oscillations. Determining whether the oscillations are chaotic will require a much longer data set analyzed using nonlinear dynamics methods [13].

The observation of instabilities in the Raman two-photon laser is surprising. Theoretical models of generic two-photon lasers, based on simplified energy level schemes, predict that the laser should be stable in the so-called ‘good cavity limit’ [14]. Our experiment is carried out in this regime, where the cavity linewidth is narrower than, but comparable to the population and atomic coherence linewidths. We suggest three possible mechanisms that may be responsible for the observed instability. One mechanism arises from the multiple frequency-degenerate final lasing states. As the laser turns on and begins to saturate the pathway shown in Fig. 3(a) (because we inject a \( \hat{z} \)-polarized trigger field), an \( \hat{x} \)-polarized field can begin to grow on the unsaturated pathways shown in Fig. 3(b) and (c) (that is enhanced by the existing \( \hat{z} \)-polarized cavity field). We note the one-photon lasers with a continuously injected signal [14] or with vector degrees-of-freedom can exhibit instabilities even in the ‘good cavity limit’ [15].

Another possible mechanism is that the lasing takes place on multiple non-frequency-degenerate transverse modes arising from cavity mirror birefringence, for example. We do not believe that this is likely because: we observe that a linearly polarized laser beam passing through the cavity in the absence of atoms remains linear to within our measurement sensitivity (one part in \( 10^6 \)) for various input states of polarization, the transverse profile of the laser beam appears to be a lowest order TEM\(_{00}\) mode as measured by a video camera, and the pinholes placed in the cavity suppress all but the lowest-order transverse modes (we estimate the diffraction loss of the TEM\(_{01}\) mode of the empty cavity due to the pinholes is about a factor of 10 greater than the loss of the TEM\(_{00}\) mode [16]). Additional high-speed measurements of the spatial-temporal behavior of the beam profile will be needed to completely rule out the presence of transverse-mode instabilities.

Finally, the presence of standing waves may induce the instabilities; it is well known that counterpropagating laser beams in conjunction with a tensor nonlinear optical interaction can give rise to polarization instabilities with a reduced instability threshold [17]. Distinguishing between the first and the third mechanisms and possibly suppressing the instabilities should be possible using a stronger magnetic field to lift the degeneracy of the different quantum pathways.

Lifting the degeneracy between the pathways also has the promise of yielding polarization entanglement. Consider, for simplicity, a single emission event. When the Zeeman levels are degenerate, as in Fig. 3, the
superposition of all possible amplitudes gives a photon pair $(1,2)$ in the state $|\alpha_1\hat{z}_{1}\rangle + \beta_1|\hat{x}_{1}\rangle |\alpha_2\hat{z}_{2}\rangle + \beta_2|\hat{x}_{2}\rangle$ with $|\alpha_1^2| + |\beta_2^2|=1$, where $|\hat{e}_i\rangle$ ($\hat{e}=x,y$) is the polarization-labeled Fock state of the $i^{th}$ photon of the emitted pair. In a strong magnetic field, the $\hat{x}\hat{z}$ and $\hat{z}\hat{x}$ pairs become off-resonant and should stop lasing, creating the polarization-entangled state $\alpha_1\alpha_2|\hat{z}\rangle_1|\hat{x}\rangle_2 + \beta_1\beta_2|\hat{x}\rangle_1|\hat{z}\rangle_2$. Maximal entanglement $(\alpha_1\alpha_2=\beta_1\beta_2)$ can be achieved by varying the resonant enhancement of a pathway, e.g., varying the Raman pump's frequency and power, and the magnetic field.

In conclusion, we have described the design and characteristics of the first Raman two-photon laser. The multi-level nature of the two-photon transition allows interesting effects such as polarization instabilities and possible photon entanglement. This opens up exciting lines of study in nonlinear laser dynamics and quantum optics.

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