Two-photon stimulated emission in laser-driven alkali-metal atoms using an orthogonal pump-probe geometry

Olivier Pfister, William J. Brown, Michael D. Stenner, and Daniel J. Gauthier

Department of Physics and Center for Nonlinear and Complex Systems, Duke University, Durham, North Carolina 27708-0305

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We observe and analyze a two-photon continuous-wave optical gain mechanism designed for building a two-photon laser. The two-photon stimulated emission is spectrally isolated and resonantly enhanced using the multilevel structure of $^{39}$K, in conjunction with an alternative interaction geometry involving orthogonal beams and polarizations. The observed two-photon laser beam amplification increases linearly with low input laser beam intensity, as expected, and saturates at a gain of $2 \times 10^{-4}$ at high intensity. A theoretical analysis of the observations is outlined. [S1050-2947(99)50812-3]

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The two-photon laser [1] is a novel quantum oscillator based on the two-photon stimulated emission process whereby two photons incident on a population-inverted atom induce a transition and four photons are scattered coherently. Due to the unusual nature of this process, the behavior of the laser is expected to be considerably different from its one-photon counterpart. For example, the two-photon laser may display complex dynamical instabilities because it operates in the highly nonlinear regime, even at threshold [2]. In addition, photon correlations arising from the stimulated emission process may lead to the generation of highly nonclassical light by the laser [3]. While there have been experimental realizations of a two-photon maser [4] and laser [5,6], many of the often conflicting predictions concerning two-photon oscillators remained untested. To address this issue, two-photon lasers need to be developed whose parameters can be well controlled, and different gain media need to be developed to identify the generic properties of two-photon lasers.

As a step toward achieving these goals, we demonstrate continuous-wave, two-photon optical laser beam amplification by a collection of laser-driven potassium atoms. Laser beam amplification occurs when the two photons participating in the two-photon stimulated emission process, together with two drive-laser photons, induce a hyper-Raman transition between the magnetic hyperfine sublevels of the $4S_{1/2}$ state of $^{39}$K, as shown in Fig. 1(a). When unsaturated, the observed amplification exhibits a linear dependence on the probe beam power, as expected for the two-photon emission mechanism.

The multilevel structure of potassium provides a rich experimental system for studying two-photon lasers. For example, laser beams with different states of polarization can be amplified in this gain medium, opening up the possibility of a hybrid one- and two-photon laser. Such a laser is interesting to study because one- and two-photon lasers have fundamentally different threshold behaviors, analogous to second- and first-order phase transitions, respectively [8].

The gain medium we use in the experiments consists of a dense, collimated, effusive beam of laser-driven $^{39}$K atoms. We use the $D_1$ transition of $^{39}$K (6-MHz natural linewidth) whose Zeeman hyperfine structure is displayed in Fig. 1(a). The atomic states are denoted by $|\alpha F_g M_g\rangle$, where $\alpha=g$ for $4^2S_{1/2}$ and $\alpha=e$ for $4^2P_{1/2}$ levels, and $F$ and $M$ are the quantum numbers for the total angular momentum and its projection on the quantization axis $\hat{z}$.

The atoms are first optically pumped into the initial state $|g_{22}\rangle$ by two $\sigma_+\!$-polarized fields, whose frequencies are set close to $\omega_{21}$ and $\omega_{22}$ [9] (Bohr frequencies are denoted by $\omega_{F_F^G}$). The optical pumping creates a population inversion for multiphoton transitions starting from $|g_{22}\rangle$. Note that the atoms are decoupled from the optical pumping fields when they are in this state.

The atoms then undergo a two-photon emission process by way of the hyper-Raman transition shown, in lowest order of perturbation, in Fig. 1(a). It is seen that two photons are absorbed from an intense off-resonant $\hat{\sigma}_-\!$ pump beam of frequency $\omega_{d}$, and the emission of two $\hat{z}$-polarized photons with the two-photon gain feature, thereby opening up the possibility of a hybrid one- and two-photon laser. Such a laser is interesting to study because one- and two-photon lasers have fundamentally different threshold behaviors, analogous to second- and first-order phase transitions, respectively [8].

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FIG. 1. (a) Raman two-photon emission in the driven $D_1$ transition of $^{39}$K where $\Delta_p=462$ MHz and $\Delta_d=58$ MHz. The pump frequency is given by $\omega_{d}=\omega_{d}+\Delta_{d}$, where the pump detuning $\Delta_{d}>0$ (typically 20 to 300 MHz) in the experiments. The probe frequency is given by $\omega_{p}=\omega_{d}+\Delta_{d}/2$. (b) The interaction geometry and states of polarization of the laser beams.

*Present address: Department of Physics, University of Virginia, 382 McCormick Rd., Charlottesville, VA 22903.
†Author to whom correspondence should be addressed.
at $\omega_p$ is stimulated by an incoming probe beam. As shown in Fig. 1(b), the pump and probe beams are perpendicular to the atomic beam to suppress Doppler broadening, and to each other to suppress all phase-matched wave-mixing processes that could compete with the two-photon gain [5]. This geometry and the angular-momentum quantum numbers restrict the states of polarization used in the experiment. Note that there exists in this system analogous $n$-photon Raman processes where $n$ is dictated by the total angular momentum of the atom. For $^{39}$K and our experimental configuration, $n = 1, 2, 3$, but no higher.

Previous observations of stimulated two-photon Raman emission have used an optical lattice of cold atoms [10] and a vapor cell [11]. In these experiments, where the pump and probe beams are nearly collinear, $n$-photon Raman gain is possible with $n=1,2,3,\ldots$, which have been interpreted as subharmonic resonance [11,12]. As far as their extension to the realization of a two-photon laser is concerned, both gain media present difficulties because of the presence of stronger adjacent one-photon gain and consequent off-resonant one-photon emission that can overwhelm or prevent two-photon lasing, and the possibility of phase-matched wave-mixing processes. In the optical lattice [10], the different gain features arise from Raman transitions between the vibrational levels of the confined atoms. Because these levels are spaced only by 165 kHz, the tails of the one- and two-photon gain features overlap, where the one-photon gain is one order of magnitude stronger than the two-photon gain. In the cell experiment [11], even though the one- and two-photon gain are completely resolved, the one-photon gain is at least 1000 times stronger and therefore still a problem.

Our experimental setup for observing the two-photon stimulated emission of Fig. 2(a) is based on a high-flux atomic beam. A 2-mm-wide collimating aperture is placed 20 cm downstream from the 3-mm-wide oven’s nozzle. From this geometry, the half-angle divergence of the atomic beam should be 10 mrad and the diameter should be 2.5 mm in the interaction region (the intersection of the atomic beam with the laser beams, 5 cm after the collimating aperture). The maximum atomic number density obtained in the beam at the interaction region is $2 \times 10^{11}$ atoms/cm$^3$ for an oven temperature of 250 $^\circ$C. At this temperature, we measure a residual Doppler absorption width of 30 MHz full width at half maximum, implying a half-angle divergence of 30 mrad, which is larger than expected from the aperture geometry. We attribute this discrepancy to collisions at the nozzle, since the mean free path is of the order of the nozzle size at this temperature.

The optical pump and Raman pump beams are produced by a commercial Ti:sapphire laser (Coherent 899) whose frequency is shifted by acousto-optical modulators. The $\hat{\sigma}_+$ optical pumping beams have a waist of 4.5 mm (1/e radius of the field at the focal plane) and a power of 5 mW (16 mW/cm$^2$ intensity). The $\hat{\varsigma}$ probe beam, provided by an extended-cavity diode laser (EOSI 2010), has a power up to 6 mW focused to a waist of 90 $\mu$m in the atomic beam (47 W/cm$^2$ maximum intensity). The $\hat{\sigma}_-$ Raman pump beam has a maximum power of 300 mW, focused to an elliptic spot of size 3 mm along the probe beam (covering the diameter of the atomic beam) and 250 $\mu$m along the atomic beam. The latter size is kept small to increase the beam’s intensity (25 W/cm$^2$ maximum), but is larger than the probe waist to minimize the inhomogeneous broadening caused by spatial variations of pump-beam-induced light shifts. A weak uniform magnetic field ($B_z \approx 10^{-4}$ T) is applied in the interaction region to overwhelm stray magnetic fields.

We measure the two-photon amplification by scanning the probe frequency and measuring the transmission through the pump-driven atoms. By first monitoring the absorption of a weak (<1 $\mu$W) probe beam with and without optical pumping, we check that the atomic state preparation is carried out to a satisfactory level (>95% of atoms in $|g22\rangle$). The Raman pump beam is then turned on and the probe power increased to several milliwatts to observe two-photon stimulated emission. This experiment is challenging because the two-photon signal can be as small as a few $10^{-5}$, while the probe power is high enough to saturate the detector’s electronic gain stage or photodiode. We solve these problems using high-power photodiodes (Hamamatsu S3994, linear response up to several tens of milliwatts) in a difference-

![FIG. 2. Probe transmission spectrum of the Raman-pumped atomic beam. The atomic number density is $10^{11}$ cm$^{-3}$, $\Delta_\omega=60$ MHz, the pump power is 300 mW, and the probe power is 2.1 mW. (a) Transmission spectrum for a large probe frequency range. Vertical lines indicate the four hyperfine absorption resonances (I corresponds to the $|g2M_g\rangle\rightarrow|e1M_e\rangle$ transition; II to $|g2M_g\rangle\rightarrow|e2M_e\rangle$; III to $|g1M_g\rangle\rightarrow|e1M_e\rangle$; IV to $|g1M_g\rangle\rightarrow|e2M_e\rangle$). (b) High-resolution transmission spectrum. The gain peaks are identified by level diagrams as in Fig. 1(a). In (e) the doublet arises from the hyperfine splitting of the excited state.](image-url)
detection setup. The probe beam is split into two beams of equal power, one of which (the signal beam) is sent through the vacuum chamber and the atoms. The other (reference) beam is sent directly to its detector. The two photocurrents are subtracted and converted to a voltage that is acquired and averaged by a digital oscilloscope. At maximum sensitivity, we can detect probe intensity changes of \(10^{-5}\).

At this level of sensitivity, etalon interference fringes from the uncoated glass windows of the vacuum chamber complicate our experiment. We reduce the effect of the fringes to a level of \(10^{-3}\) of the probe beam power by sending the signal beam through the vacuum chamber at the most oblique angle of incidence possible in our configuration (15°). The residual fringes are removed by fitting a low-finesse Airy transmission function to the signal’s baseline and subtracting this baseline from the data.

A typical probe laser transmission spectrum is displayed in Fig. 2. Besides the \(D_1\) hyperfine absorption lines, we observe several stimulated emission features that disappear when the optical pumping beams are blocked. We identify the mechanisms responsible for each gain and absorption feature by their location and probe-power dependence. The lowest-order contributions to these processes are diagrammed as insets to Fig. 2(b), as done in Fig. 1(a). The two strongest gain peaks are the two Raman one-photon peaks: \((\alpha)\) at \(\omega_p = \omega_d\) and \((\delta)\) at \(\omega_p = \omega_d + \Delta g\). The Raman two-photon peak \((\gamma)\) is sitting about halfway between \((\alpha)\) and \((\delta)\) (\(\omega_p = \omega_d + \Delta g/2\) in lowest order). The doublet \((\epsilon)\) is “pump-dressed” one-photon gain. The weaker peaks are one-photon-emitting transitions originating from ground states much less populated than \(|g22\rangle\), as well as gain and absorption lines arising from \(|41K\rangle\) (6.7% abundance).

The weak-field assignment of the gain spectrum neglects strong-field effects like pump-induced light shifts that can modify the resonance frequencies. To check the validity of our assignment in the case of high pump beam power, we have analyzed the interaction in the basis of the atom dressed by the pump field [13]. This calculation of the frequencies of all possible one- and two-photon transitions between dressed states confirms the weak-field assignment shown in Fig. 2(b).

To demonstrate that the peak denoted by \((\gamma)\) is a genuine two-photon process, we measure the dependence of its size on the intensity of the probe beam. This intensity dependence is different for one- and two-photon amplification processes. This can be understood simply from an analysis of the amplification process, considering a plane wave propagating through a homogeneously broadened thin medium of length \(\ell\). The gain experienced by a probe beam (obtained through solution of the Maxwell-Bloch equations) and its resonant unsaturated limit is given by

\[
G_1 = \frac{I}{I_0} - 1 = \frac{g_1/}{1 + \Delta_1^2/\Gamma_1^2 + I_0/I_{1s}} \rightarrow g_1/1, \quad (1)
\]

\[
G_2 = \frac{I}{I_0} - 1 = \frac{g_2/}{1 + \Delta_2^2/\Gamma_2^2 + I_0/I_{2s}} \rightarrow g_2/1, \quad (2)
\]

for the one- and two-photon cases, respectively. In these equations, \(I\) and \(I_0\) are the output and input probe intensities, \(g_1\) and \(g_2\) unsaturated gains, \(I_{1s}\) and \(I_{2s}\) saturation intensities, \(\Gamma_1\) and \(\Gamma_2\) homogeneous widths, and \(\Delta_1\) and \(\Delta_2\) detunings from resonance that include the light shifts of the transitions induced by the probe beam. Equations (1) and (2) show that the unsaturated one-photon gain is constant in \(I_0\), whereas unsaturated two-photon gain is proportional to \(I_0\).

Figure 3 displays the height of peak \((\gamma)\) versus the input probe power. A clear linear increase of the gain is observed for powers up to about 3 mW (24 W/cm² intensity), for which a saturation plateau occurs at a maximum peak height of \(4 \times 10^{-5}\). This observed linear dependence below saturation is a central result of this paper: it can be considered as a direct check that photons are emitted two at a time (in addition to the fact that the two-photon peak is at the expected frequency). Note that the height of peak \((\gamma)\) is not the net two-photon gain because it overlaps with the wing of the large absorption feature in Fig. 2(a). Based on our measurements, we estimate the maximum net gain to be about 50% of the peak height. The height of the peak was determined by approximating a zero-absorption baseline at the two-photon peak, using the fact that this peak is narrow and well resolved.

As a preliminary analysis, we perform a least-squares fit of the data shown in Fig. 3, using the estimated interaction length (2.5 mm) and keeping \(g_2\) a free parameter. The fit of the unsaturated linear region (dashed line) gives \(g_2=(0.78 \pm 0.06) \times 10^{-4}\) cm/W. The fit with the full expression of \(G_2\) (solid line) gives \(g_2=(0.90 \pm 0.06) \times 10^{-4}\) cm/W, and \(I_{2s} = 36 \pm 3\) W/cm². The two fits are plotted in Fig. 3 and seem to give reasonable agreement with the experiment, with the caveat that this analysis is simplified and leaves out a few essential physical effects.

A more realistic theoretical description of the gain medium must take into account these physical effects, which we merely mention since such an analysis is beyond the scope of this paper. The two-photon gain is reduced because the population inversion is reduced somewhat by the Raman pump field. Moreover, three inhomogeneous broadening mechanisms reduce the amplification. One is the residual Doppler broadening in the atomic beam. The other two are due to the spatial variation of the probe beam intensity over its Gaussian transverse profile. This intensity variation re-
As an aside, we illustrate with one example the wealth of possibilities offered by a multilevel gain medium for the realization of a two-photon laser. From the weak-field description of the gain features in Fig. 2(b), it is straightforward to determine how their resonance frequencies depend on the Raman pump beam frequency $\omega_d$: Raman processes (one- and two-photon) tune as $\omega_d$, whereas pump-dressed processes tune as $2\omega_d$. In particular, peak ($\beta$), a weak pump-dressed one-photon gain process, can be tuned in and out of coincidence with the two-photon Raman gain ($\gamma$) without altering significantly the rest of the spectrum (keeping all strong gain peaks at bay). This will enable us to realize a two-photon laser capable of sustaining either pure two-photon or simultaneous one- and two-photon lasing.

In conclusion, we have observed well-resolved net two-photon Raman stimulated emission in laser-driven $^{39}\text{K}$ using an orthogonal pump-probe geometry. The potentially overwhelming effects of power-broadened absorption and one-photon gain should not be a hindrance to bringing cavity-resonant two-photon emission above lasing threshold, which we are now pursing.

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References:

[9] The two optical pumping frequencies are slightly shifted away from these values, in opposite directions, so as to avoid coherent population trapping as discussed in E. Arimondo, Progress in Optics XXXV, edited by E. Wolf (Elsevier, Amsterdam, 1996).