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Observation of large continuous-wave two-photon optical amplification

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We observe 30% two-photon optical amplification of a probe laser-field propagating through a laser-pumped potassium vapor. This amplification is spectrally isolated and substantially larger than that of previously reported continuous-wave two-photon amplifiers. The combination of large amplification and spectral isolation of the two-photon gain feature will greatly facilitate precise studies of the photon statistics of this highly nonlinear quantum amplifier and the development and characterization of a two-photon laser based on this gain medium. We also observe spectrally-distinct three-photon amplification ($\sim 5\%$) in the same system under different experimental conditions. We present a simple model of the interaction that gives qualitative agreement with our observations and explains the dependence of the two-photon gain on the various system parameters. This model predicts that the size of the two-photon gain is quite sensitive to an interference between two different quantum pathways. [S1050-2947(97)00208-4]

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The study of light-matter interactions in the regime where the coupling between the atoms and the radiation field is highly nonlinear is a subject of fundamental importance in quantum optics. Devices that operate exclusively in this regime, such as two-photon amplifiers and two-photon lasers [1–3], have intrigued researchers for years because their dynamical behavior [4], photon statistics, and coherences [5] are predicted to be very different from their one-photon counterparts. Despite the fundamental and practical interest in these devices, there have been few experimental tests of the numerous, often conflicting, predictions regarding their behavior. The primary limitation on these tests has been the difficulty in realizing practical two-photon lasers and amplifiers in the laboratory.

In this work, we demonstrate a new two-photon optical gain medium that amplifies a beam of light by 30%. The observed two-photon gain is approximately 300 times larger than that obtained previously in continuous-wave two-photon optical amplifiers [6,7]. This large gain is obtained using a relatively simple apparatus: a laser-driven potassium vapor contained in a glass cell. Furthermore, the observed two-photon gain is spectrally isolated from other competing processes, facilitating precise studies of the characteristics of two-photon amplifiers and lasers. This amplifier operates in the degenerate mode, where both photons generated in the stimulated emission process have the same frequency.

The two-photon gain in this system arises from a stimulated emission process that we call two-photon stimulated Raman scattering, shown schematically in Fig. 1(a) [8]. Intense pump (solid) and probe (dashed) fields stimulate the atom to make a transition between the initial state $|g\rangle$ and the final state $|g'\rangle$ by absorbing two photons from the pump field (frequency ω_d) and adding two new photons to the probe field (frequency ω_p) via virtual intermediate states. Energy conservation requires that $\omega_p = \omega_d - \Delta_{gg'}/2$, where $\hbar\Delta_{gg'}$ is the energy separation between $|g\rangle$ and $|g'\rangle$. We stress that this scattering process is a pure gain process based on the stimulated emission of two probe photons; that is, the stimulated transition rate does not depend on the relative phase of the fields. Hence, we expect this gain medium to display all of the properties associated with a generic, phase-insensitive, two-photon amplifier. This is in contrast to any parametric wave-mixing process which might also result in the addition of photons to the probe field when $\omega_p = \omega_d - \Delta_{gg'}/2$. To obtain continuous-wave two-photon gain based on this stimulated Raman process, a steady-state population imbalance must exist between states $|g\rangle$ and $|g'\rangle$ such that the population of state $|g\rangle$ is larger than that of state $|g'\rangle$. In this system, this imbalance is maintained via optical pumping of atoms by the intense pump field.

We note that n-photon Raman scattering processes can occur in this system for probe-beam frequencies $\omega_p = \omega_d - \Delta_{gg'}/n$, for $n = 1,2,3,\ldots$. Kumar and coworkers [9] have studied extensively the one-photon Raman process ($n = 1$) in a laser-driven sodium vapor, while Hemmerich *et al.* [10] have observed multiphoton Raman scattering in cooled rubidium atoms trapped in the potential wells of a three-dimensional optical lattice. Trebino, Rahn, and

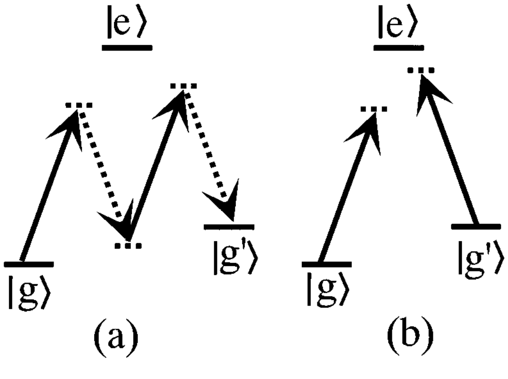

FIG. 1. (a) The two-photon stimulated Raman transition between long-lived atomic states $|g\rangle$ and $|g'\rangle$ is resonantly enhanced by the excited state $|e\rangle$. The process involves the annihilation of two pump photons of frequency ω_d (solid) and the creation of two probe photons of frequency ω_p (dashed). (b) The strong pump beam, which interacts on both the $|g\rangle \rightarrow |e\rangle$ and $|g'\rangle \rightarrow |e\rangle$ transitions, creates a population imbalance in $|g\rangle$ and $|g'\rangle$ via optical pumping.

Lucht [11] and Agarwal [12] have also investigated a related multiphoton parametric wave-mixing process in laser-driven sodium atoms. To our knowledge, however, the current work is the only one to investigate the usefulness of the two-photon stimulated Raman scattering process as a source of gain in a two-photon amplifier.

In order to observe two-photon gain in this system, it is necessary that both the pump and probe beams interact on both the $|g\rangle \rightarrow |e\rangle$ and $|g'\rangle \rightarrow |e\rangle$ transitions. Because of this and the fact that the intensity of each of these beams substantially exceeds the one-photon saturation intensity, a complete theoretical model of the interaction is rather complicated. While the development of such a model is beyond the scope of the current work, much can be said about the two-photon gain mechanism by considering the two-photon transition probability in the presence of the applied fields using time-dependent perturbation theory. To proceed, we take the form of the applied fields to be

$$\mathbf{E}^{d,p}(t) = \mathbf{E}_0^{d,p} e^{-i\omega_{d,p}t} + \text{c.c.}, \tag{1}$$

where the superscripts denote the pump and probe fields, respectively. The interaction strengths of the fields $\mathbf{E}^{d,p}(t)$ with the two transitions $|g\rangle \rightarrow |e\rangle$ and $|g'\rangle \rightarrow |e\rangle$ are given by the Rabi frequencies as

$$\Omega_{g(g')}^{d,p} = \frac{2\boldsymbol{\mu}_{g(g')e} \cdot \mathbf{E}_0}{\hbar}. \tag{2}$$

In terms of these one-photon Rabi frequencies, we find that the effective two-photon Raman Rabi frequency is given by

$$\Omega_{2\gamma} = \frac{\Omega_g^d \Omega_{g'}^{p*}}{8\Delta(\Delta_{gg'}/2)(\Delta + \Delta_{gg'}/2)} [\Omega_{g'}^{p*} \Omega_{g'}^d - \Omega_g^d \Omega_g^{p*}], \tag{3}$$

where $\Delta = \omega_{eg} - \omega_d$ is the detuning of the pump beam from the $|g\rangle \rightarrow |e\rangle$ transition and we have taken the probe-pump detuning to be $\omega_p - \omega_d = -\Delta_{gg'}/2$, the resonant condition for the two-photon transition.

Equation (3) reveals several important relationships between the two-photon gain expected in this system and the various experimental parameters. First, the two-photon transition rate is seen to scale quadratically with the intensity of the probe field, until the effects of saturation become important. As a result, the probe beam is not amplified when it is weak and the two-photon gain *increases linearly* as a function of probe-beam intensity, attaining its maximum when the probe beam intensity is approximately equal to the two-photon saturation intensity [13]. Saturation of the two-photon gain occurs when the two-photon Rabi frequency becomes approximately equal to the optical pumping rate between the ground states. This dependence of the two-photon gain on the probe intensity differs from optical amplifiers based on the one-photon stimulated emission process. In one-photon amplifiers, the stimulated emission rate scales linearly with the intensity of the input field, resulting in a one-photon gain that is intensity independent for weak input fields and which decreases as the intensity approaches the one-photon saturation intensity.

Because of the need for the pump and probe fields to interact strongly with both the $|g\rangle \rightarrow |e\rangle$ and $|g' \rightarrow |e\rangle$ transitions, a general condition for observing large two-photon Raman gain is $\Omega_{g,g'}^{d,p} \approx \Delta_{gg'}$. For continuous-wave pump and probe fields, this limits the possible systems to those for which $\Delta_{gg'}/2\pi \lesssim 1$ GHz. This requirement, combined with the appearance of $\Delta_{gg'}$ in the denominator of Eq. (3) above, suggests that optimum systems for demonstrating large two-photon Raman gain would be those for which $\Delta_{gg'}$ is relatively small. However, one must bear in mind that decreasing the value of $\Delta_{gg'}$ also decreases the spectral separation between the two-photon and one-photon Raman gain features. Thus the two-photon and one-photon gain features may not be resolved for small values of $\Delta_{gg'}$, making it difficult to conduct precise studies of the properties of the two-photon amplifier. For this work, we have chosen a system in which the ground-state splitting is large enough so that the two-photon gain is spectrally isolated, but small enough to permit large two-photon gains at relatively modest pump and probe intensities. This is in contrast to the work of Hemmerich *et al.* in which two-photon gain was observed via stimulated Raman transitions between trapped atomic vibrational states [10]. In that work, the value of the ground-state splitting ($\sim$165 KHz) was comparable to the observed width of the saturated one- and two-photon Raman transitions, resulting in a two-photon gain feature that appeared as a small peak on the side of a much larger one-photon gain feature.

One very interesting feature of the two-photon Raman transition rate given in Eq. (3) is the possibility that the term in brackets vanishes when $\Omega_g^p = \Omega_{g'}^p$ and $\Omega_g^d = \Omega_{g'}^d$. Thus the two-photon transition rate is nearly zero whenever each of the pump and probe beams interacts equally on both transitions. This surprising effect occurs as a result of a quantum interference between two indistinguishable pathways from the initial state $|g\rangle$ to the final state $|g'\rangle$, as illustrated in Fig. 2. For our system the $|g\rangle \rightarrow |e\rangle$ and $|g'\rangle \rightarrow |e\rangle$ transitions have different strengths so that the destructive interference between the two pathways is not complete, resulting in a two-photon transition rate that is reduced from that of either pathway considered alone by only a factor of ~ 2. Furthermore, it may be possible to eliminate the interference altogether by making use of the Zeeman sublevels associated with $|g\rangle$ and $|g'\rangle$ and careful choice of the pump- and probe-beam polarizations.

As discussed above, our two-photon amplifier uses a composite (atom plus field) gain medium created by driving a potassium vapor with an intense pump laser field. The potassium vapor is contained in a 7-cm-long evacuated pyrex cell with uncoated, near normal incidence optical windows. The cell is heated to a temperature of 150 °C, producing a number density of approximately 10^{13} atoms/cm^3. The states $|g\rangle$, $|g'\rangle$, and $|e\rangle$ of our idealized level scheme [Fig. 1(a)] correspond to the $4S_{1/2}(F=1)$, $4S_{1/2}(F=2)$, and $4P_{1/2}$ states of ^{39}K, respectively, where the ground-state hyperfine splitting is $\Delta_{gg'}/2\pi = 462$ MHz. Because we use natural-abundance potassium, we also observe gain and absorption features due to scattering from ^{41}K where $\Delta_{gg'}/2\pi = 254$ MHz.

The pump laser field is generated by an actively stabilized Ti:sapphire ring laser. It is linearly polarized, collimated to a diameter of 150 μm [intensity full width at half maximum (FWHM)] as it passes through the cell, and is tuned approxi-

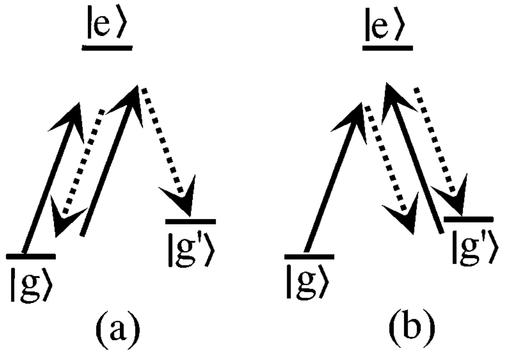

FIG. 2. The two possible quantum pathways for the atom to undergo a transition from $|g\rangle$ to $|g'\rangle$ via virtual intermediate states. Both involve the absorption of two pump photons (solid) and the subsequent emission of two probe photons (dashed). Because the two paths are indistinguishable, they interfere, giving an overall two-photon transition rate which may differ significantly from that due to either pathway considered alone. (a) Annihilation of a pump and creation of a probe photon via interaction with the $|g\rangle \rightarrow |e\rangle$ transition induces a second-order amplitude for the state $|g\rangle$. Subsequent annihilation of a pump photon via the $|g\rangle \rightarrow |e\rangle$ transition and creation of a probe photon via the $|g'\rangle \rightarrow |e\rangle$ transition leads to a fourth-order amplitude for the state $|g'\rangle$. (b) Annihilation of a pump via interaction with the $|g\rangle \rightarrow |e\rangle$ transitions and creation of a probe photon via the $|g'\rangle \rightarrow |e\rangle$ transition induces a second-order amplitude for the state $|g'\rangle$. Subsequent annihilation of a pump photon and creation of a probe photon via the $|g'\rangle \rightarrow |e\rangle$ transition leads to a fourth-order amplitude for the state $|g'\rangle$.

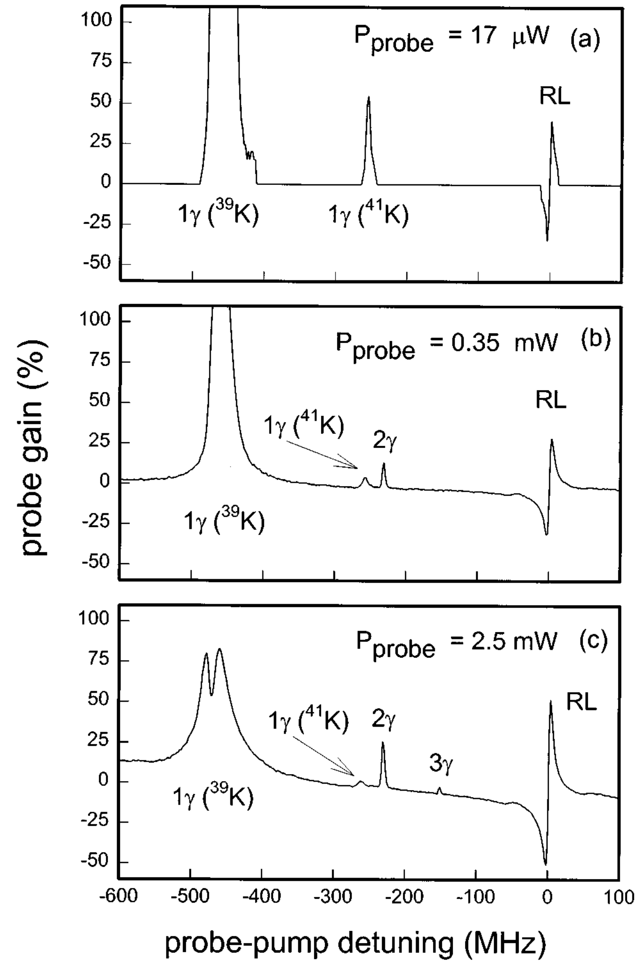

FIG. 3. (a) Normalized probe transmission spectrum at low probe power P_{probe}. The curve has been smoothed to reduce the digitization error. (b) As the probe intensity is increased a new gain feature appears at $\omega_p - \omega_d = -\Delta_{gg'}/2 \approx 2\pi(-230\ \mathrm{MHz})$ corresponding to two-photon gain. (c) Near the two-photon saturation intensity, the two-photon gain reaches its maximum value of 30%. Note the appearance of a three-photon gain peak at $\omega_p - \omega_d = -\Delta_{gg'}/3 \approx 2\pi(-150\ \mathrm{MHz})$.

mately 2.4 GHz to the low-frequency side of the $D1$ transition $[4S_{1/2}(F=2) \rightarrow 4P_{1/2}(F=1)]$ occurring near $\lambda = 769.9$ nm. Noticeable self-defocusing is apparent under these experimental conditions; the divergence angle of the beam increases approximately by a factor of 3 and it no longer has a lowest-order Gaussian profile after passing through the cell. We observe significant beam breakup due to self-focusing when the pump laser is tuned to the high-frequency side of the transition; hence our choice of red detuning for the pump beam. The total power in the pump field at the entrance to the cell is approximately 850 mW and serves a dual purpose: it provides the photons of frequency ω_d needed to drive the two-photon Raman scattering process shown in Fig. 1(a) and it maintains the necessary population imbalance between $|g\rangle$ and $|g'\rangle$. This latter effect is accomplished via preferential optical pumping of the state $|g'\rangle$, since the pump field drives the $|g'\rangle \rightarrow |e\rangle$ transition more strongly than the $|g\rangle \rightarrow |e\rangle$ transition [Fig. 1(b)].

To characterize the properties of the composite gain medium, we measure the transmission of a probe beam through the cell as a function of its frequency for several different probe beam powers. The probe beam is generated by a grating-stabilized diode laser and has a maximum power of 10 mW. We set the probe laser field polarization orthogonal to that of the pump field, since we found that this orientation resulted in the maximum two-photon gain. The probe beam, which is collimated to a diameter of 65 μm (intensity FWHM) in the cell, overlaps and is nearly copropagating with the pump beam. In this geometry, the two-photon Raman scattering process is nearly Doppler-free. We use a small crossing angle of 12 mrad to allow maximum overlap of the pump and probe beams in the cell while minimizing the effects of parametric wave mixing which can potentially compete with the two-photon gain process. The total transmitted power of the probe beam is measured by focusing the output probe beam onto a photodiode. A polarizer in front of the detector blocks pump scatter from the cell windows.

The probe-beam transmission spectra are quite rich, displaying many spectrally distinct gain and absorption features. To identify the microscopic origin of these features, we record high-resolution transmission spectra as a function of the probe-pump detuning. Figure 3 shows several such spectra as a function of the probe-beam power. We specify the probe-beam power rather than its intensity since the effects of self-defocusing make it difficult to infer the exact spatial profile of the probe beam in the cell from the known spatial profile outside the cell. For each of these spectra, we subtract the residual pump scatter and the loss incurred by the windows. This allows us to interpret the spectra directly in terms of gain and loss experienced by the probe beam due to the laser-driven atomic vapor. For a weak probe beam,

Fig. 3(a), we observe three narrow spectral features in the frequency region of interest. The first, labeled $1\gamma(^{39}\mathrm{K})$, is due to one-photon stimulated Raman scattering (Stokes scattering) from optically pumped $^{39}\mathrm{K}$ and occurs when $\omega_p-\omega_d\approx-\Delta_{gg'}=2\pi(-462\ \mathrm{MHz})$ [9]. In this case, the maximum observed one-photon Raman gain is $\sim$5000% (off scale), corresponding to an output probe power that is 50 times larger than the input power. We have observed 15 000% amplification due to this process for very low input probe powers ($\ll 1\ \mu\mathrm{W}$). The gain feature labeled $1\gamma(^{41}\mathrm{K})$ in Fig. 3(a) corresponds to a similar one-photon Raman process occurring in $^{41}\mathrm{K}$ where $\Delta_{gg'}(^{41}\mathrm{K})=2\pi(254\ \mathrm{MHz})$. This feature is much smaller than that of $^{39}\mathrm{K}$ because the natural abundance of $^{41}\mathrm{K}$ is only 6.7%. The dispersive-shaped feature (RL) occurring near $\omega_p-\omega_d\approx 0$ is due to stimulated Rayleigh gain (and absorption) arising from population oscillations between the magnetic sublevels of each of the ground states [14].

For higher probe-beam intensities, Fig. 3(b), the one-photon gain features saturate significantly and a new, intensity-dependent gain feature (2γ) appears at $\omega_p-\omega_d\approx-\Delta_{gg'}(^{39}\mathrm{K})/2=2\pi(-231\ \mathrm{MHz})$. We attribute this new feature to two-photon optical amplification because it occurs at the expected frequency and it is not present for low probe-beam intensities, as expected for the two-photon stimulated emission process. In addition, we verify that the two-photon gain feature is not associated with a four-wave-mixing process by analyzing the spectral content of the radiation emanating from the cell. Note that the two-photon Raman gain from $^{39}\mathrm{K}$ and the one-photon Raman gain from the $^{41}\mathrm{K}$ isotope are spectrally resolved, even though they are separated by only 23 MHz.

For still higher probe intensities, Fig. 3(c), we observe the onset of saturation in the two-photon gain, leading to a maximum in the two-photon gain of approximately 30%. For this probe intensity, we estimate our effective saturated two-photon Rabi frequency [Eq. (3)] to be $2\pi(1.4\ \mathrm{MHz})$, in reasonable agreement with the expected value of $2\pi(1.2\ \mathrm{MHz})$ imposed by the optical pumping rate from $|g'\rangle$ to $|g\rangle$. Even at these intensities, the two-photon Raman gain feature remains spectrally isolated from the other gain features. This is in contrast to the previously reported two-photon dressed-state gain medium where the two-photon amplification was accompanied by a significant amount of one-photon amplification [6,10]. For each of the three intensities shown in Fig. 3, we note that the application of a small axial magnetic field does, in some cases, cause the two-photon gain to increase substantially, possibly due to a modification in the destructive interference discussed above. In addition to the two-photon Raman gain feature, we also observe 5% three-photon Raman amplification, labeled 3γ in Fig. 3(c), at $\omega_p-\omega_d\approx-\Delta_{gg'}(^{39}\mathrm{K})/3=2\pi(-150\ \mathrm{MHz})$. For still higher probe-beam intensities, the two-photon amplification decreases and the three-photon amplification increases.

In conclusion, we have presented results demonstrating large two-photon gain in a dense vapor of atomic potassium driven by a strong pumping field. The observed gain of 30% is approximately 300 times larger than that previously obtained and is spectrally isolated from competing gain processes. This should greatly facilitate the development of practical two-photon amplifiers and lasers based on this new gain medium. The observed gain is explained in terms of a simple model based on a time-dependent perturbation theory treatment of two-photon Raman scattering. Future experiments will explore in greater detail the scaling of the n-photon amplification processes with experimental parameters, including the effects of the ground-state sublevels and laser beam polarizations on the quantum interference that determines the overall n-photon gain.

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