Observation of a Two-Photon Gain Feature in the Strong-Probe Absorption Spectrum of Driven Two-Level Atoms

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A feature associated with continuous-wave two-photon optical gain has been observed in the absorption spectrum of an ensemble of barium atoms driven by a strong near-resonant optical field. In the dressed-atom picture, the observed gain is attributable to inverted two-photon transitions with nearly resonant intermediate states. A cw optical two-photon laser utilizing this gain appears feasible.

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The interaction of two-level atoms (TLA's) with strong resonant or nearly resonant optical radiation has been analyzed from a number of perspectives.[1,2] Of interest here, it has been shown that TLA's driven by a near-resonant driving field (pump) can act to amplify a weak-probe laser appropriately tuned relative to the atomic and pump frequencies.[3-8] In fact, single-photon lasers based on driven TLA gain have been constructed.[9-12] Driven TLA gain occurs in the absence of inversion between the ground and excited states. The largest weak-probe gain feature, which has been referred to as Raman gain,[4] can be understood in terms of a stimulated hyper-Raman-scattering process or in terms of population inversions on transitions between dressed atom-field states. Raman gain can be viewed as a single-photon gain process. At higher probe intensities, additional gain features, corresponding to multiphoton analogs of the Raman gain process and involving two- or more-photon gain,[13,14] become important. In this paper, we describe an experimental study of the interaction of a strong probe with a driven TLA and provide the first demonstration of cw two-photon gain in the optical regime.

Consider an ensemble of stationary TLA's having a transition frequency ν_a, an upper-state radiative lifetime T_1, and a homogeneous dephasing time $T_2=2T_1$. The atoms are driven by a monochromatic pump field of frequency ν_0, atom-pump detuning $\Delta \equiv \nu_a - \nu_0$, and resonant Rabi frequency Ω_0. We study the gain and absorption spectrum of a probe field interacting with the pump-driven TLA's. The probe is assumed to have a frequency ν_1, probe-pump detuning $\delta = \nu_1 - \nu_0$, and resonant Rabi frequency Ω_1. This spectrum can be evaluated using the expressions for nonlinear susceptibility derived by Agarwal and Nayak[15] using a continued-fractions method. This approach has also recently been employed by Gruneisen *et al.*[16] in calculations related to the energy transfer between propagating beams using stimulated Rayleigh scattering.

Using the method of Agarwal and Nayak, we have calculated the probe gain and absorption as a function of probe-pump detuning for various probe intensities. The results are presented in Fig. 1. Throughout Fig. 1 we have set $\Omega_0/\Delta=3$ and $2\pi\Delta T_2=10$. In the weak-probe limit ($\Omega_1/\Delta=0.1$), the probe gain and absorption spectrum reduces [see Fig. 1(a)] as expected to the probe spectrum studied by Mollow and co-workers.[3,6] The dominant features in this spectrum correspond to single-photon gain (positive peak at $\delta \simeq -\Omega'$) and single-photon absorption (negative peak at $\delta \simeq \Omega'$), where $\Omega' \equiv (\Omega_0^2+\Delta^2)^{1/2}$. For $\Omega_1/\Delta=0.6$ [Fig. 1(b)], new features appear in the probe gain and absorption spectrum. The peak at $\delta \simeq -\Omega'/2$ ($\delta=\Omega'/2$) corresponds to

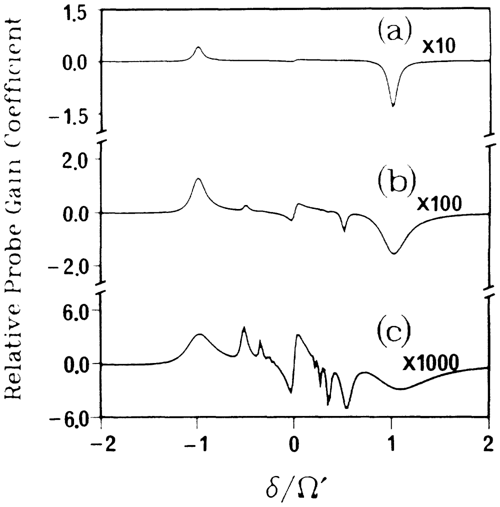

FIG. 1. Calculated probe gain coefficient as a function of probe-pump detuning δ/Ω' for $\Omega_0/\Delta=3$, $2\pi\Delta T_2=10$, and $\Omega' \equiv (\Omega_0^2+\Delta^2)^{1/2}=\sqrt{10}\Delta$. The vertical scale corresponds to the probe gain coefficient normalized so as to be -1 at the center of the weak-probe absorption profile measured in the absence of a pump laser. In the figure, the calculated values have been multiplied by (a) 10 times, (b) 100 times, and (c) 1000 times. (a) Weak-probe laser, $\Omega_1/\Delta=0.1$. One-photon gain (absorption) features exist at $\delta \simeq -\Omega'$ ($\delta \simeq \Omega'$), a Rayleigh-scattering feature at $\delta \simeq 0$. (b) For $\Omega_1/\Delta=0.6$, two-photon gain (absorption) appears at $\delta \simeq -\Omega'/2$ ($\delta=\Omega'/2$). (c) For $\Omega_1/\Delta=1.4$, two-photon gain is the largest gain feature. Generally, n-photon gain occurs at $\delta \simeq -\Omega'/n$, while n-photon absorption occurs at $\delta \simeq \Omega'/n$.

two-photon gain (absorption). For $\Omega_1/\Delta = 1.4$ [Fig. 1(c)], still more features appear with n-photon gain (absorption) occurring at $\delta \simeq -\Omega'/n$ ($\delta \simeq \Omega'/n$). In Fig. 1(c), the two-photon gain feature is actually larger than the one-photon gain feature. Reversing the sign of Δ simply reverses all the gain and absorption features of the spectra about $\delta = 0$. The existence of probe gain and absorption features at probe-pump detunings given by subharmonics of the pump Rabi frequency has been discussed in several contexts[17-21] including cases involving nonzero Δ,[16] but the connection of these features with multiphoton gain has apparently been overlooked. As in the case of the single-photon Raman gain, the n-photon gain can be understood in terms of a high-order stimulated hyper-Raman effect or in terms of n-photon transitions between dressed states in nonadjacent doublets (see Fig. 2).

To avoid unessential complexity, the spectra of Fig. 1 were calculated assuming Doppler-free conditions. To realize this condition experimentally, we have employed a collimated atomic beam of natural barium. The use of an atomic beam limited the achievable atomic densities, interaction lengths, and hence single-pass gain and absorption to relatively small values. To compensate, the probe gain and absorption was measured in an intracavity mode; i.e., the probe power was measured after transmission through a confocal optical cavity whose mirrors were positioned symmetrically about the atomic beam. The cavity performed two functions. First, it increased the effective optical thickness of the atomic beam. Second, it increased the achievable probe intensity for a given probe input power. High probe intensity is essential to bring the nonlinear gain into operation, but low probe power is essential to avoid saturation of the gain medium. The atomic beam was chopped at 191 Hz. Probe gain and absorption spectra represent measurements of probe power transmitted through the cavity during atomic-beam-on cycles normalized by measurements of probe power transmitted during atomic-beam-off cycles. Standard frequency-modulation techniques, active while the atomic beam was blocked, were employed to keep the empty-cavity resonance frequency locked to the probe frequency. Probe gain and absorption measurements, obtained as described above using an enhancement cavity, are complicated by the fact that atomic dispersion can cause shifts in the cavity resonance frequency and thereby affect probe transmission through the cavity. With our scheme of locking the empty-cavity resonance frequency to the probe frequency, dispersive effects always act to decrease atomic-beam-on probe transmission and therefore mimic absorption rather than gain.

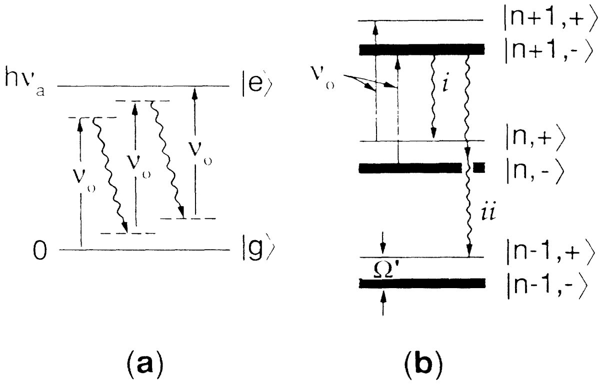

FIG. 2. Alternative pictures of the two-photon gain process. (a) Stimulated hyper-Raman scattering. Three photons from the pump laser are scattered, leading to the emission of two photons (wavy lines). (b) Dressed-state picture. Two-photon gain results from the two-photon transition (indicated by ii) between the inverted dressed levels $|n+1,-\rangle$ and $|n-1,+\rangle$. Here i indicates the well-known single-photon gain process (thick lines represent the more heavily populated states). Analogous pictures can be drawn for n-photon gain processes.

Our experimental apparatus is depicted in Fig. 3. The atomic beam was 800 μm in diameter as it passed through the center of a 1-cm-long, 200 finesse, confocal enhancement cavity. The linearly polarized pump and probe lasers were tuned near the ^{138}Ba 553.5-nm ${}^1S_0 \rightarrow {}^1P_1$ transition which has a natural linewidth of 19 MHz. Since ^{138}Ba has zero nuclear spin, the ${}^1S_0$-${}^1P_0$ transition closely approximates a two-level quantum system. The experiment is complicated, however, by the presence of other barium isotopes (about 22% total abundance) some of which have nonzero nuclear spin. The axis of the enhancement cavity, the Ba atomic beam, and the propagation direction of the traveling-wave pump laser were mutually orthogonal. The probe laser propagated along the cavity axis. In our experimental configuration, nonlinear-wave-mixing-type gain processes are not phase matched. The Rabi frequency Ω_0 of the pump laser was determined by measuring the

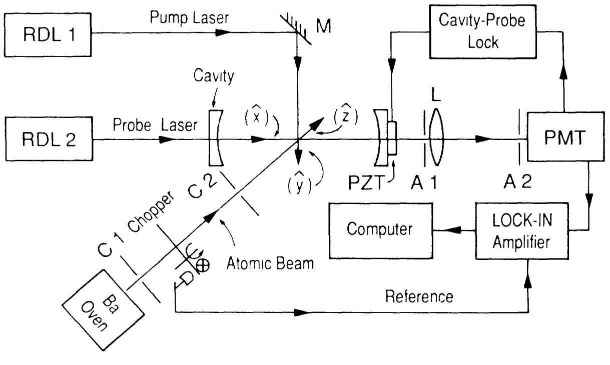

FIG. 3. Experimental schematic. RDL1, pump laser; RDL2, probe laser. Both are single-mode cw ring dye lasers. PZT, piezoelectric transducer; M, mirror; PMT, photomultiplier tube; A1 and A2, apertures; L, lens; C1 and C2, atomic-beam collimators; x, y, and z, three perpendicular spatial axes.

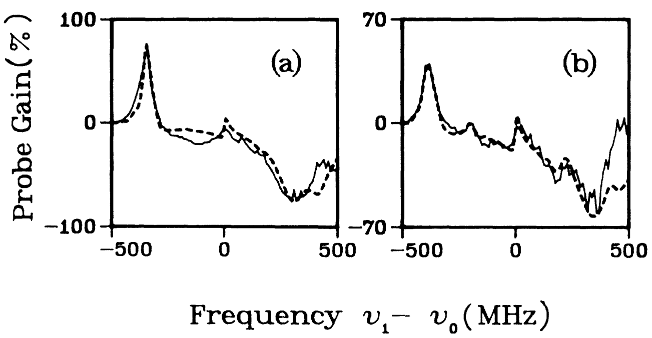

FIG. 4. Measured gain in probe power transmitted through the cavity as a function of probe-pump detuning ($\delta=\nu_1-\nu_0$). Solid curves represent experimental data. Dashed curves represent numerical calculations including effects of the dispersion (pseudoabsorption), pump-field inhomogeneity, the standing-wave character of the probe field, and Ba isotopes. The vertical scales represent 100×[(atomic-beam-on probe power transmission) − (atomic-beam-off probe power transmission)]/(atomic-beam-off probe power transmission). This quantity is positive for gain. (a) $\Omega_0\simeq340$ MHz, $\Omega_1\simeq44$ MHz, and $\Delta\simeq100$ MHz; features observed are one-photon gain (absorption) at $\delta\simeq-\Omega_0$ ($\delta\simeq\Omega_0$), and Rayleigh scattering at $\delta\simeq0$. (b) $\Omega_0\simeq390$ MHz, $\Omega_1\simeq140$ MHz, and $\Delta\simeq100$ MHz; the peak at $\delta=-\Omega'/2$ is the two-photon gain feature.

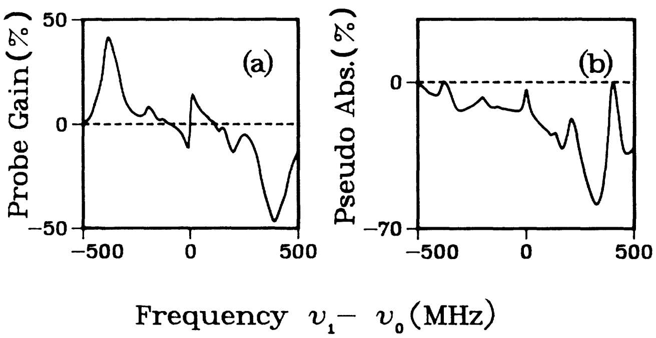

FIG. 5. (a) Calculated probe gain (positive) of Fig. 4(b) excluding pseudoabsorption effects. (b) Calculated probe absorption resulting exclusively from pseudoabsorption (dispersive) effects. The vertical scale in both parts of the figure represents 100×[(transmitted probe power) − (incident probe power)]/(incident probe power). Parameters are the same as those in Fig. 4(b). Dashed lines are zero-gain (-absorption) base lines.

splitting of the Mollow triplet resonance fluorescence spectrum produced by the pump with no probe. This spectrum was observed by using the enhancement cavity as a Fabry-Pérot interferometer. The Rabi frequency Ω_1 of the probe laser was deduced by measuring the power-broadened width of the probe absorption spectrum while the pump laser was shut off.

A weak-probe gain and absorption spectrum is shown in Fig. 4(a) for the case where $\Omega_0\simeq340$ MHz, $\Omega_1\simeq44$ MHz, and $\Delta\simeq100$ MHz. It is seen that there are only single-photon gain (absorption) features at $\delta=-\Omega'$ ($\delta=\Omega'$), and a Rayleigh-scattering feature at $\delta\simeq0$. The absorption feature is complicated primarily because of the effects of the other Ba isotopes which are all distributed to the blue-frequency side of the ^{138}Ba resonance. As the probe-laser Rabi frequency was increased to $\Omega_1\simeq140$ MHz (with $\Omega_0\simeq390$ MHz, $\Delta\simeq100$ MHz), a two-photon gain (absorption) feature appeared [see Fig. 4(b)] as a small peak at $\delta\simeq-\Omega'/2$ ($\delta\simeq\Omega'/2$). While these peaks are small and comparable to the experimental noise level, they are observed to appear consistently in many separate measurements of the probe spectrum. It will be noticed that the peak of the two-photon gain feature is near zero, and hence the peak constitutes a decrease in absorption rather than an actual gain. This occurs, as discussed above, because of dispersion-mediated shifts in the cavity resonance frequency and concomitant decreases in the atomic-beam-on probe transmission. Since our detection scheme compares the transmitted probe-laser power with and without the Ba atomic beam, and since the empty cavity was actively locked to the probe-laser frequency, dispersion-mediated cavity frequency shifts look like absorption. We refer to this effect as pseudoabsorption. The dashed lines shown in Fig. 4 are calculated probe gain and absorption spectra. Except for the extreme blue end, the calculated and observed spectra are in excellent agreement, indicating that we have indeed observed cw two-photon gain. The discrepancies between the experiment and theory at the blue end of the spectra are due to a persistent partial loss of the cavity-probe lock at the end of the locking range. In Fig. 5(a), we plot the probe gain calculated as in Fig. 4(b) except that the effect of pseudoabsorption has not been included. Note that in this case the two-photon gain peak is positive and represents what our observations would have shown if we had been able to eliminate dispersive (pseudoabsorption) effects. In Fig. 5(b), probe pseudoabsorption calculated using the same parameters as in Fig. 5(a) is plotted. This spectrum shows how much the transmitted probe power was reduced due exclusively to the effect of pseudoabsorption. In the absence of pseudoabsorption, we conclude that the experimentally observed probe transmission would actually have increased by 8% at the center of the measured two-photon gain peak.

In conclusion, we have experimentally demonstrated that strongly driven two-level atoms can be utilized as a two-photon gain medium and may therefore be useful in the realization of a two-photon laser.[22] We point out that this simple system is also a potential gain medium for three- or more-photon amplification.

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