PHYSICAL REVIEW A VOLUME 49, NUMBER 6 JUNE 1994

Observation of gain due to coherence effects in a potassium-helium mixture

J. A. Kleinfeld and A. D. Streater
Physics Department, Lehigh University, Bethlehem, Pennsylvania 18015
(Received 22 February 1994)

We report steady-state gain due to coherence effects in a potassium-helium mixture, for strong pumping near the D_2 line and probing near the the D_1 line. The absorption-gain spectra are in good agreement with a four-level Raman-driven lasing without inversion model, where collisional transfer populates the probed excited state. Using parameters corresponding to the experimental conditions, the model predicts gain without bare-state population inversion by a large margin.

PACS number(s): 42.50.Gy, 42.50.Hz, 34.90.+q, 32.70.Jz

Coherence effects in atomic systems provide a rich variety of phenomena for scientific study and application. In recent years there have been many intriguing theoretical proposals for manipulating some of these coherence effects to achieve amplification of a weak probe beam in a gain medium without requiring a traditional population inversion among the bare atomic states of the medium. These proposals, several of which are discussed in Refs. [1] and [2], have been broadly classified as "lasing without inversion" (LWI), and have created a great deal of interest because of their potential for generating laser light at regions of the electromagnetic spectrum where it is currently difficult or impossible to do so. There have also been several interesting experiments designed to explore some of these mechanisms [3–6], all in a time-dependent manner. The purpose of this Rapid Communication is to describe our steady-state investigation of a Raman-driven four-level system for LWI, similar to that studied theoretically by Narducci and co-workers [7–9].

The Raman-driven four-level system is described in detail in Refs. [7–9]. The system is depicted in Fig. 1. A strong coupling of levels $|1\rangle$, $|2\rangle$, and $|4\rangle$, together with a suitable choice of other parameters (decay rates, pumping rates, etc.), can cause the ρ_{12} and ρ_{34} coherences (viewed in the bare-state basis) to develop in such a way that can lead to amplification without inversion of a probe beam tuned near the ω_3 resonance, provided that a small incoherent population has been placed in level $|3\rangle$. More precisely, the ρ_{12} coherence can give rise to a population trapping mechanism that radiatively decouples the ground-state population from the probe beam, thereby reducing absorption, and the ρ_{34} coherence can be associated with a Raman scattering of the pump beam [9]. These two coherences contribute to varying degrees, depending on the laser intensities, frequencies, and other conditions of the experiment. For the trapping mechanism, we note that it is particularly important that the ground-state relaxation (mixing) rate W_2 be small compared to the excited-state relaxation rate W_4, and that the doublet splitting be small compared to the Rabi frequency due to the strong pump beam [8]. The theoretical papers [7–9] do not specify a pumping mechanism to populate level $|3\rangle$, but it is assumed to be small enough so as not to create a population inversion.

We have chosen potassium as our four-level Raman-driven gain medium. The two ground states are then the hyperfine $F = 1$ and $F = 2$ states, and the pump and probe states are the fine-structure $4P(J = \frac{3}{2})$ and $4P(J = \frac{1}{2})$ levels, respectively. A buffer gas is added to achieve the necessary population in level $|3\rangle$ via collisional transfer from level $|4\rangle$. Potassium is a good candidate for LWI since the ground-state hyperfine splitting (462 MHz) is smaller than that of other alkali metals. Additionally, the ground-state hyperfine collisional mixing cross sections (related to W_2) are typically small enough [10] that transit relaxation, an effective mixing due to the finite duration that an atom remains in the beam, is the dominant relaxation mechanism is the lowest two levels. The buffer gas is useful since it not only allows us to control the transfer rates between levels $|3\rangle$ and $|4\rangle$ by adjusting the pressure, but it also allows us to model the experiment with a closed four-level system.

Our own theoretical analysis of this system uses four-level optical Bloch equations, derived under appropriate rotating-wave approximations, and includes both dephasing and inelastic collision rates. We also assume that W_2 is dominated by transit relaxation. The Bloch equations are then solved in steady state. The probe beam absorption coefficient is proportional to the sum of the off-diagonal matrix elements $\rho_{13}+\rho_{23}$, and is plotted as a function of the probe detuning. The resulting curves can be generated for various pump and probe intensities, col-

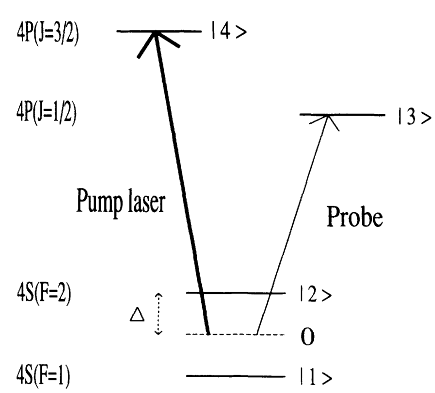

FIG. 1. The Raman-driven four-level system [7]. Level $|3\rangle$ has energy $\hbar\omega_3$. Level $|4\rangle$ decays at rate W_4, and levels $|1\rangle$ and $|2\rangle$ mix at rate W_2. Corresponding potassium energy levels are indicated on the far left. Note that "zero" detuning is directly between the ground-state doublet.

1050-2947/94/49(6)/4301(4)/$06.00

lisional mixing rates, etc. These probe absorption-gain profiles do not include Doppler averaging. However, our experience modeling two-level systems has shown that for generalized Rabi frequencies greater than the Doppler width, the averaged curve shapes remain qualitatively the same in those cases, and we expect that this is true here as well. To within the validity of the Bloch equations, the solution has no restriction on the intensity of either the pump or probe beam.

Computer experiments reveal several interesting aspects of this system. (1) Gain without inversion is predicted. (2) For a detuned pump, the gain feature and central absorption are separated by approximately the generalized Rabi frequency. (3) The central absorption is dramatically narrowed and shifted, particularly for close detunings. (4) For off-resonance pumping, the gain feature can display a Fano-like profile. Such profiles are often associated with coherence effects. (5) The magnitude of the probe gain peak is often maximum for pump detunings somewhat off resonance. (6) Gain is only realized for cases where the inelastic transfer rate $|4\rangle \rightarrow |3\rangle$ is greater than the rate $|3\rangle \rightarrow |4\rangle$. When this condition does not hold, only an extra absorption feature is present for various detunings (as might be expected from Ref. [11]). (7) Dephasing collisions hinder gain.

We also note that it is possible to increase the pressure high enough such that a population inversion can exist for large pump strengths and transfer rates $|4\rangle \rightarrow |3\rangle$ greater than $|3\rangle \rightarrow |4\rangle$. In these cases, gain seems to occur on resonance (as well as on the coherent sideband) even for off-resonant pumping.

Based on these observations, especially point (7) above, we chose helium as the buffer gas, since the ratio of the fine-structure mixing cross section to the dephasing cross section is relatively large [12,13]. Theoretical models for a helium pressure of 8 torr at $T = 458$ K and pump and probe Rabi frequencies of 2.4 GHz and 0.15 MHz, respectively, are shown in Fig. 2. For these cases,

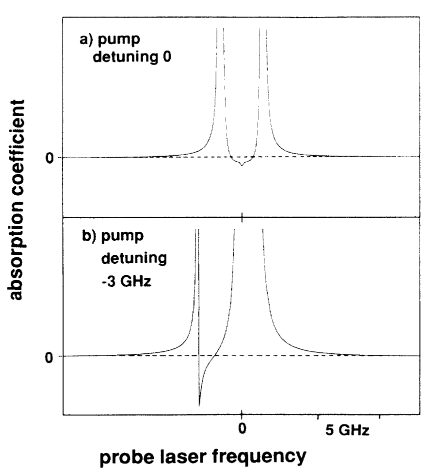

FIG. 2. Theoretical calculations of the absorption coefficient (arbitrary units) vs probe detuning for parameters roughly modeling 8-torr helium, $T = 458$ K, and a pump Rabi frequency of 2.4 GHz. (a) Zero pump detuning; (b) -3.0 GHz pump detuning. A negative value indicates gain.

there is no population inversion by a significant margin according to the model. For example, the populations for Fig. 2(b) are $\rho_{11}, \rho_{22}, \rho_{33}, \rho_{44} = 0.278, 0.418, 0.145, 0.160$, respectively.

Our experimental setup is relatively straightforward. The strong driving radiation is provided by a Kr^+-pumped Coherent 699-29 cw single-mode dye laser (linewidth ≈ 1 MHz). The beam is focused into a specially designed short path length (5 mm) borosilicate glass cell containing potassium vapor and buffer gas. The estimated pump beam waist in the cell is 53 μm with intensities typically ~ 2 kW/cm^2. The potassium number density ($\sim 10^{14}$ cm^{-3}) is controlled by controlling the temperature of an oven in which the cell is placed. A weaker probe beam from a Coherent 899-29 cw single-mode Ti:sapphire laser (linewidth ≈ 1 MHz) is then focused and copropagated with the pump beam. The estimated probe beam waist in the cell is 24 μm with a resulting intensity of ~ 60 mW/cm^2. Both beams are linearly polarized in the same direction. Since the D_1 ($J = \frac{1}{2}$) and D_2 ($J = \frac{3}{2}$) transitions differ in wavelength by roughly 3 nm, the copropagating beams can be separated by a diffraction grating, and the isolated 770-nm radiation is detected with phase-sensitive detection by a photodiode.

The probe beam is technically not weak, in that the probe Rabi frequency is on the order of the natural width. However, the model predicts that, for the conditions of the experiment, the absorption coefficient for a probe beam of this power is very similar to the weak-probe-beam limit. Indeed, decreasing the probe power by a factor of 3 does not significantly alter the experimental results, except for a decrease in the signal-to-noise ratio.

The measurements yield a plot of transmitted probe intensity vs detuning from the D_1 resonance. Experimental results for 8-torr helium at $T = 473$ K are shown in Fig. 3. Gain is clearly evident. The agreement between these cases and our simplistic Bloch equation model in many of its general features is encouraging. In particular, the height of the experimental gain peak does go through a maximum as the pump detuning is increased, and, for pump detunings of a few gigahertz, the gain feature exhibits an asymmetric profile, particularly evident in Fig. 3(d). Also, the central absorption is notably narrowed and shifted in Figs. 3(b) and 3(d).

Figure 4(a) shows the results of switching the pump and probe laser frequencies so that we are pumping the D_1 line and probing the D_2 line, for conditions equal to those in Fig. 3. Since we do not expect the collisional transfer rates for $\frac{3}{2} \rightarrow \frac{1}{2}$ to equal those for $\frac{1}{2} \rightarrow \frac{3}{2}$, we should find gain on only one transition. This is certainly the case: the D_2 line displays the extra absorption feature discussed above. An interesting point here is that by direct application of the mixing rates from Ciurylo and Krause [12], which include degeneracies, we would expect the amplification to occur for pumping D_1 and probing D_2, and this is indeed where we first looked for it. However, ignoring theses degeneracies, as does our model, the ratio of these rates should be determined by the Boltzmann factor from detailed balance, and this leads to amplification on the D_1 line.

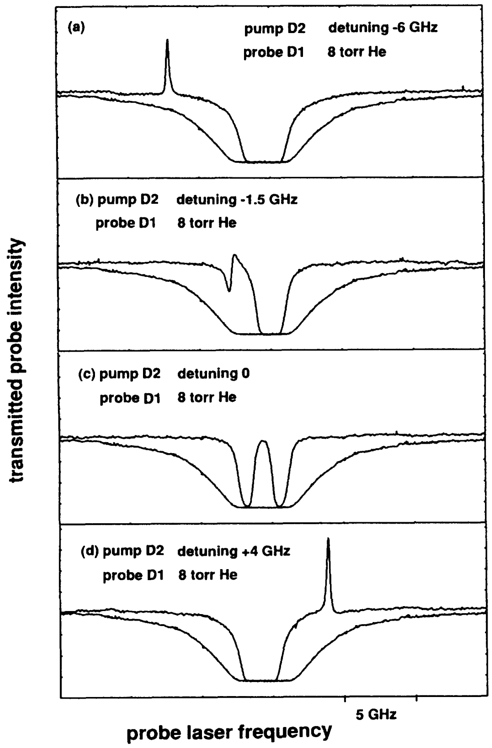

FIG. 3. Experimental results of the D_1 probe beam transmitted intensity vs detuning for 8 torr helium and $T = 473$ K. Pump beam detunings from the D_2 resonance are indicated. Estimated pump intensity is 3.2 kW/cm^2 (Rabi frequency $\sim$ 2.9 GHz). Background scans with the pump beam blocked are superimposed. Detunings are from the fluorescence maximum using a vacuum reference cell.

Figure 4(b) shows the result of reducing the helium pressure to 0.4 torr while again pumping D_2 and probing D_1, as in Fig. 3. In this case there is very little collisional transfer, and the transit relaxation rate is higher. The D_1 transmission spectrum displays only the extra absorption feature. This agrees with the model, which indicates that in the low-pressure limit there is no difference between D_1 and D_2 pumping. This is also the limit studied by Pinard and Grynberg [11] in a three-level system, where only the extra absorption feature is predicted.

Figure 5 demonstrates line center gain for a pressure of 16 torr helium, and shows the power dependence of the gain. The curve is strikingly similar to Fig. 2 of Narducci *et al.* [7]. The splitting of the double humped gain increases with pump power, as expected, as the dressed energy levels move apart. This splitting is related to the Rabi frequency of the pump and is roughly in agreement with what we would expect based on the pump intensity. We have observed that the absorption-gain profiles from our theoretical model are very sensitive to the input parameters—which we feel we know only marginally well, as discussed below. The model predicts only a double humped gain (no absorption dips just outside the gain peaks) for our best estimates of appropriate model parameters for the experimental conditions of Fig. 5(a). We feel that the uncertainties of the parameters and the crudeness of the model itself account for this disagreement. If the collisional rates are lowered slightly, the model also gives results similar to Fig. 5(a) with still no population inversion.

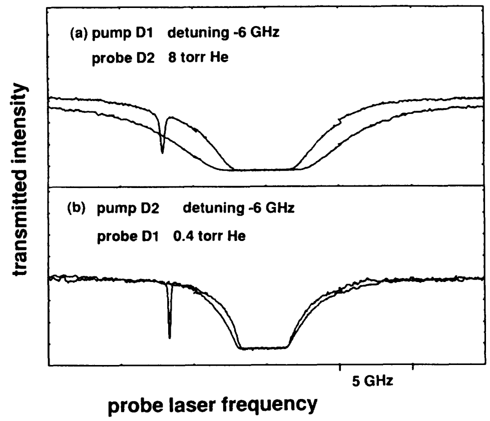

FIG. 4. Probe beam transmitted intensity vs detuning. (a) shows a scan of the D_2 line with D_1 pumping for 8 torr helium, $T = 473$ K. (b) shows a D_1 scan with D_2 pumping for 0.4-torr helium. In both cases only the extra absorption feature discussed in the text is present.

It is important to point out that many of these same features can be obtained theoretically using a three-level V system with collisional transfer. Both systems produce similar results in the wings, as might be expected for pump detunings much greater than Δ, but yield different calculated probe absorption spectra for pump detunings near line center. From this we are led to conclude that the upper-state coherence is the dominant amplifi-

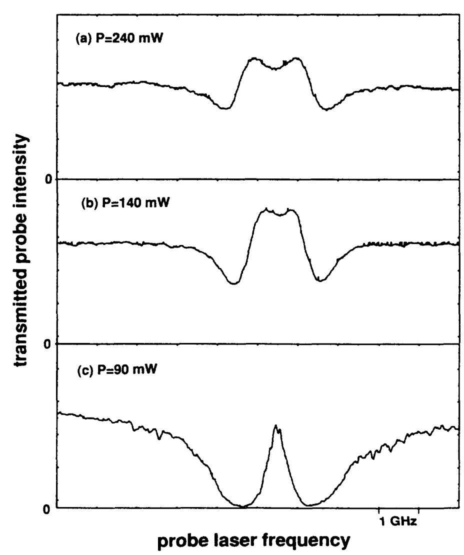

FIG. 5. Dependence of the D_1 probe beam intensity vs detuning for various pump beam powers for 16 torr helium in the cell. Pump tuned to the D_2 fluorescence maximum from a vacuum cell. Note that the dressed energy levels move apart with increased pumping power. Temperatures vary somewhat for the cases shown, ranging from 438–483 K.

cation mechanism for large detunings. At line center, however, the four-level model shows significantly more gain, presumably due to the population trapping mechanism associated with the ρ_{12} coherence. At this point we cannot determine the absolute gain coefficient accurately enough to conclude that the ρ_{12} coherence effects are dominant for line center pumping.

There are several experimental realities that either challenge a direct comparison with the four- (or three-) level model, or obscure the theoretical values that should be used. First, our real system (potassium) possesses both hyperfine structure with magnetic sublevels in the upper states, and several magnetic sublevels in the lower states. Each allowed transition has its own line strength. These degeneracies are not as easily introduced in Bloch equations as they are in rate equations, so potassium is clearly not a simple four-level system. However, such simplifying assumptions as ours have been effectively employed in the past on similar systems, most notably the two-level case [14]. Next, radiation trapping, focusing effects, precise beam overlap, and Gaussian beam behavior all obscure the choice of appropriate theoretical values relating to lifetimes, Rabi frequencies, etc.

We feel that our work here is complementary to that of Gao *et al.* [3], but lends itself more readily to a closed-system, steady-state analysis. While we have not directly monitored the populations and therefore have not proven inversionless amplification, we believe that our results compare favorably with the Raman-driven four-level theory in many details: the asymmetric and sometimes Fano-like gain feature, gain behavior with respect to pump detuning and intensity, the additional absorption feature at low pressures and for interchanging pump and probe transitions, the narrowed and shifted central absorption, and the intensity-dependent splitting of the symmetric gain profile for line center pumping. These features are a compelling demonstration of steady-state gain arising from atomic coherence effects. Additionally, they support a model which also predicts LWI. For parameters which approximate the conditions of the experiment, the model predicts no population inversion by a significant margin.

Experimentally, proving no inversion in the steady-state Raman-driven four-level case is an interesting problem, since in theory the ρ_{12} coherence creates a trapping state for ground-state atoms that does not lend itself to interrogation with dipole-allowed radiation. Also, given the optical depth of potassium in the cell and the resulting large degree of radiation trapping, we believe fluorescence measurements would be difficult and unreliable. However, we feel that a combination of time-dependent ground-state absorption measurements (similar to Ref. [6]) and/or steady-state $4P$ probing to higher levels could prove fruitful in dealing with the question of inversion. We are currently making preparations to conduct such measurements.

This experiment indicates that the collisional transfer mechanism does not unduly damp the coherences to the point of ineffectiveness. We find this suggestive, and think that one extension of this collisional transfer mechanism might be to simply attempt the same experiment using collisional pooling and multiphoton processes to populate higher levels. These processes are clearly occurring in our cell, as evidenced by purple fluorescence, probably from the $5P$ states. They might also provide enough of a populating mechanism to find gain on higher-energy transitions without the necessity of a buffer gas, and perhaps without as much undesirable dephasing due to collisions.

We gratefully acknowledge helpful discussions with L. M. Narducci, E. D'Angelo, J. R. Tredicci, M. S. Malcuit, and J. P. Huennekens. We also thank J.P.H. for generous use of laboratory space and equipment. We gratefully acknowledge financial support from both USDE and Lehigh University.

[1] O. Kocharovskaya, Phys. Rep. **219**, 175 (1992).
[2] M. O. Scully, Phys. Rep. **219**, 191 (1992).
[3] J. Gao, C. Guo, X. Guo, G. Jin, P. Wang, J. Zhao, H. Zhang, Y. Jiang, D. Wang, and D. Jiang, Opt. Commun. **93**, 323 (1992).
[4] A Nottelman, C. Peters, and W. Lange, Phys. Rev. Lett. **70**, 1783 (1993).
[5] W. E. van der Veer, R. J. J. van Diest, A. Donszelmann, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **70**, 3243 (1993).
[6] E. Fry, X. Li, D. Nikonov, G. G. Padmabandu, M. O. Scully, A. V. Smith, F. K. Tittel, C. Wang, S. R. Wilkinson, and S. Y. Zhu, Phys. Rev. Lett. **70**, 3235 (1993).
[7] L. M. Narducci, H. M. Doss, P. Ru, M. O. Scully, S. Y. Zhu, and C. Keitel, Opt. Commun. **81**, 379 (1991).
[8] L. M. Narducci, M. O. Scully, C. H. Keitel, S. Y. Zhu, and H. M. Doss, Opt. Commun. **86**, 324 (1991).
[9] C. H. Keitel, O. A. Kocharovskaya, L. M. Narducci, M. O. Scully, S. Y. Zhu, and H. M. Doss, Phys. Rev. A **48**, 3196 (1993).
[10] M. A. Bouchiat, J. Brossel, and L. C. Pottier, J. Chem. Phys. **56**, 3703 (1972).
[11] M. Pinard and G. Grynberg, J. Phys. (Paris) **49**, 2027 (1988).
[12] J. Ciurylo and L. Krause, J. Quant. Spectrosc. Radiat. Transfer **28**, 457 (1982).
[13] E. L. Lewis, Phys. Rep. **58**, 1 (1980).
[14] M. T. Gruneisen, K. R. MacDonald, and R. W. Boyd, J. Opt. Soc. Am. B **5**, 123 (1988).