Experimental Demonstration of Laser Oscillation without Population Inversion via Quantum Interference in Rb

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Laser oscillation without population inversion is demonstrated experimentally in a V-type atomic configuration within the D_1 and D_2 lines of Rb vapor. It is shown that the effect is due to the atomic interference. The experimental results, as first predicted by careful theoretical analysis, are in a good agreement with detailed calculations.

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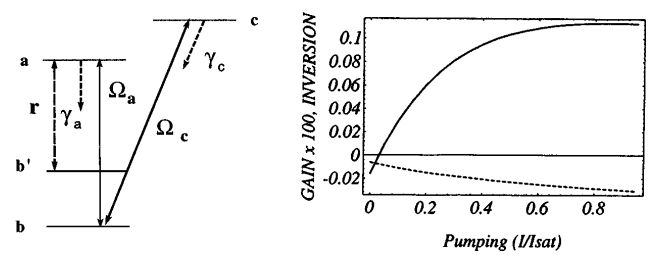
In the present work, laser oscillation without population inversion (LWI) [1] is reported for the first time. Our demonstration makes use of noninversion gain in a V-type [2] cw amplifier. In order to obtain a clear proof-of-principle demonstration of LWI oscillation, we carried out a set of experiments which include the following: the observation of inversionless amplification of a weak probe light by a coherently prepared atomic medium, experimental verification of the absence of inversion, and, finally, observation of noninversion laser oscillation in a cavity.

Several recent experiments [4,5] have already demonstrated the possibility of noninversion amplification. These experiments were based on observation of amplification in Λ-type systems [6] which involved coherent population trapping and can therefore be viewed as amplification with inversion in a dressed state basis.

V-type inversionless amplification is itself conceptually interesting in that the physics of resonant V-type LWI is the cancellation of absorption via quantum interference. And there is, in general, no dressed state basis in which there is a "hidden" inversion. Furthermore, the present work is a technical advance, in that it is the first observation of gain without inversion using low power cw diode lasers.

The present experiment was suggested and modeled by a detailed theoretical analysis, the results of which may be understood by considering the four-level model of Fig. 1(a), although the actual calculations were carried out using a realistic model of Rb^{87} with all hyperfine and Zeeman sublevels included. Two sublevels of the ground state are coupled to a pair of excited states via three fields. A strong driving field with Rabi frequency Ω_c and a weak probe field with Rabi frequency Ω_a are assumed to be quasimonochromatic. These fields have linewidths $\Delta\nu_c$ and $\Delta\nu_a$, both of which are much less than atomic radiative decay rates. The third (pump) field is taken to be incoherent, i.e., it has a very broad linewidth ($\Delta\nu_{\text{pump}} \gg \gamma_a, \gamma_c$) and is represented by an incoherent pumping rate r [see Fig. 1(a)].

In the absence of the incoherent pump field, almost all the population is optically pumped into the state b' by the strong driving field. The incoherent pump destroys this optical pumping by populating the upper state a, and thus state b, via spontaneous decay. Hence the population difference $\rho_{aa} - \rho_{bb}$ is determined by the rates with



(a) (b)

FIG. 1. (a) Simplified four-level model for lasing without inversion. In our realization of the model in Rb^{87} $|a\rangle$ corresponds to sublevels $|P_{1/2},\ F=2\rangle$, $|c\rangle = |P_{3/2},\ F=2\rangle$ and the two sublevels of ground state $|b\rangle = |S_{1/2},\ F=1\rangle$ and $|b'\rangle = |S_{1/2},\ F=2\rangle$. Driving field Ω_c is tuned to the D_2 resonance from the $F=1$ ground state to $F=2$ of excited state, probe field Ω_a couples the same sublevels of the ground state with levels having $F=2$ of the $P_{1/2}$ excited state. Polarizations of the fields are described in the text. (b) Results of numerical modeling for the realistic 32-level scheme of Rb. Gain $\times\ 8\pi/N\lambda^2 L$ (solid line) and inversion per atom (dashed line) as a function of the incoherent $\Delta\nu = 50$ MHz pump laser intensity; parameters are $|\Omega_c| = 30$ MHz, $H_z = 10^{-4}$ T. All fields are in the exact resonance with corresponding transitions in the center of the Doppler profile. Inversion is defined as the sum of populations of levels $P_{1/2}$, $F = 2$, $M_F = 0, \pm 1$ minus populations of ground state sublevels having $F = 1$ with corresponding M_F. Incoherent pumping rate is expressed in terms of Rabi frequency and a linewidth of a pumping laser as $r = 2|\Omega_{\text{pump}}|^2/(\Delta\nu + \gamma_a/2)\gamma_a$.

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which atoms leave and decay into the states a and b. Note that in the limit of a weak probe field, atoms can leave state b only via state c. Thus the population difference above is determined by the ratio of decay rates from level a to level b and from level c to level b'. If level c decays more slowly than level a, an inversion on the transition coupled by the weak probe field cannot be created.

Amplification of a weak probe field on the a to b transition is, however, possible, due to the presence of atomic coherence between upper levels a and c. The physical origin of this mechanism is to be interpreted as a quantum interference cancellation of absorption. Alternatively, reduction of absorption and enhanced gain can be viewed as a result of a Fano-type interference between the dressed states [2].

The gain (absorption) coefficient for a weak probe field on the a to b transition is proportional to the imaginary part of the off-diagonal element of the atomic density matrix ρ_{ab}. In the weak probe field limit, and under conditions of exact resonance, we obtain the gain

$$G = \frac{3\lambda^2 NL\gamma_a}{4\pi}\frac{\mathrm{Im}\rho_{ab}}{\Omega_a} = \frac{3\lambda^2 NL\gamma_a}{4\pi} \times \frac{(\rho_{aa}^0 - \rho_{bb}^0) + |\Omega_c|^2/\gamma_{ac}\gamma_{bc}(\rho_{bb}^0 - \rho_{cc}^0)}{\gamma_{ab} + |\Omega_c|^2/\gamma_{ac}}, \quad (1)$$

where ρ_{ii}^0 is the population of level i calculated to the zeroth order in the probe field, γ_{ij} is the relaxation rate of the density matrix element ρ_{ij}, N is the density of atoms, L is cell length, and λ is the probe laser wavelength.

From the second equality in Eq. (1) we see that there are two contributions of the upper-level coherence which modify the usual gain or absorption. The first is the dynamical Stark effect [7], studied theoretically and experimentally in a V-type configuration in [3]; it is responsible for the $|\Omega_c|^2$ term in the denominator.

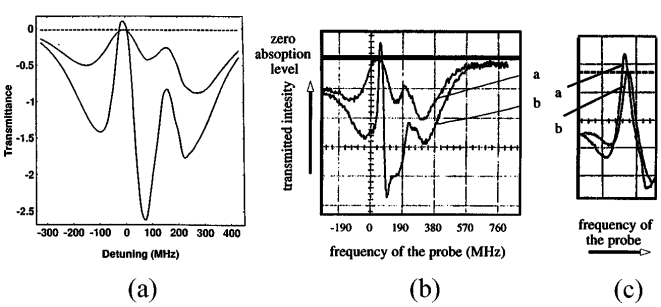

FIG. 2. (a) Calculated absorption (gain) coefficient for a weak probe field as a function of its frequency in the vicinity of $1 \rightarrow 2'$ resonance of the D_1 line. Parameters and scaling correspond to those of Fig. 1(b). Detuning is from the center of the absorption line. Upper curve, without incoherent pump; lower curve, $r = 0.4$. (b) Experimentally measured transmission of the probe laser as a function of its frequency. Curve a, without the incoherent laser; curve b, with incoherent laser. Estimated parameters for the experiment are $\Omega_c \sim 30$ MHz, $\Omega_a \sim 0.1$ MHz, $r \sim 0.4$. (c) Experimental results for transmission of the probe field showing the influence of a probe laser linewidth on its transmission spectrum. Curve a, without modulation of current; curve b, with modulation of current ($\Delta\nu_p \sim 10$ MHz).

There is, however, another important effect which modifies the properties of the V scheme. This contribution is represented by the term proportional to the $|\Omega_c|^2$ in the numerator of Eq. (1) and is precisely due to the quantum interference of absorption mentioned above. It follows from Eq. (1) that absorption interferes destructively if $\rho_{bb} - \rho_{cc} > 0$. This leads to a reduction of absorption and enhances the gain. As a result the probe field can be amplified even if $\rho_{aa} - \rho_{bb} < 0$. We would like to underscore the fact that quantum interference is the only reason for inversionless gain in the V scheme, i.e., there is no inversion in any dressed state picture (provided the drive and the probe fields are equally detuned from resonance).

Motivated by these simple considerations, we choose for the present experiment in Rb^{87} the following combination of the fields. The coherent drive is $\hat{x}$ polarized and couples sublevel 1 [8] of the $S_{1/2}$ state with sublevels of the $P_{3/2}$ state (the power ranged from 1 to 20 mW in 0.9 mm spot). The probe laser is $\hat{z}$ polarized and couples sublevels $1 \rightarrow 2'$ (the probe power ranged from 0.5 to 50 μW in a 0.7 mm spot). The third beam (incoherent pump) comes from a solitary diode laser that had a broad variable linewidth of 50–200 MHz, an output power 10 mW, and a diameter in the cell of 8 mm, which encompassed both the probe and pump beams. All fields propagate in the same direction, hence, for sufficiently strong drive field the amplification peak is essentially Doppler free.

To obtain amplification without inversion in our scheme, a weak magnetic field in the z direction is required. This field destroys Zeeman coherences within the $F = 2$ ground state manifold created by an $\hat{x}$ polarized incoherent pumping field. Without the magnetic field population tends to collect in certain coherent superpositions of sublevels of this manifold. We found that the optimum range (for the given linewidth of the incoherent field) of the magnetic field H_z is of order 10^{-4} T.

To investigate the possibility of LWI in this scheme we have numerically solved density matrix equations for the 32-level scheme of Rb, taking into account all relevant Clebsch-Gordan and Racah coefficients, field polarizations, Zeeman sublevels, and Doppler broadening. The results of the modeling are presented in Fig. 1(b). The results, for this particular field configuration, turn out to be very similar to those obtained from the simple four-level model of Fig. 1(a). As shown in Fig. 1(b) the gain increases as a function of the incoherent pump intensity, while inversion is negative and decreases within incoherent pump intensity.

A remarkable feature of the amplification peak in Figs. 2(a)–2(c) is its narrow width. This is determined in our case by the Rabi frequency of the driving laser. In contrast, the optical pumping peaks are broader, since their width is determined by the optical pumping rate

of the driving field and the slow ground state relaxation rate. The calculated spectrum for the probe transmission is presented in the Fig. 2(a). Agreement between theory and experiment is excellent.

In the experiment we use a 4 cm long Brewster cell containing natural Rb heated to ~60 °C. The driving and probe lasers are tunable extended cavity diode lasers, having short-term linewidths on the order of 100 kHz. The drive and probe beams were separated after the cell with a diffraction grating.

In the first set of experiments the transmission of the probe field was measured as a function of its frequency. The frequency of the driving laser was tuned within the Doppler profile of the D_2 absorption line, while the frequency of the probe laser was swept through the D_1 line. The results of the transmission measurements are shown in the Fig. 2(b). This figure shows the portion of the transmission spectrum which corresponds to the coupling of the probe field to the $1 \rightarrow 2'$ transition within the D_1 line.

The first experiment was done without the incoherent pump laser [curve a in Fig. 2(b)]. On the broad Doppler profile apparent in the transmission spectrum of the probe beam we see two transmission peaks. They are due to the optical pumping of specific velocity groups by the drive laser. There are three such groups within the velocity distribution; for one of these groups the drive field is resonant with $1 \rightarrow 2''$ transition [left peak in Fig. 2(b)] and for others it couples levels 1 with $1''$ and 1 with $0''$. Since the frequency separation between the last two transitions is small, both of these groups contribute to the right optical pumping peak.

In the second experiment [curve b in Fig. 2(b)] the incoherent pump was added and its frequency was tuned such that it couples $2 \rightarrow 2'$ transition. We clearly observe an increase in the transmission of the probe corresponding to amplification of the weak probe field in the vicinity of $1 \rightarrow 2''$ optical pumping peak. The amplification occurs in spite of the fact that the results of optical pumping are destroyed by the incoherent laser. Because of the reverse optical pumping effect, the absorption of the probe increases in all other parts of the spectrum. We also notice that the amplification peak has a much narrower width [~30 MHz in Fig. 2(b)] than the pure optical pumping peaks of curve a. With present experimental conditions we observe approximately 8% to 16% gain per pass.

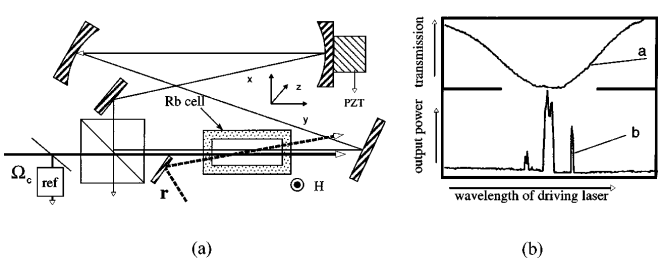

FIG. 3. (a) Schematic of the ring resonator closed by a polarization beam splitter cube for the LWI oscillator experiment; ref is a reference cell with Rb; output from the cavity is analyzed using a monochromator. (b) Dependence of the driving field absorption (curve "a") in the reference cell and the corresponding dependence of the cavity output power at 794 nm (curve "b") as a function of the frequency of a driving field. Three peaks of the curve a correspond to the three longitudinal modes of the cavity (with frequency separation ~170 MHz).

To verify the absence of a population inversion we carry out an experiment with different linewidths for the probe laser. Since the probe field is weak, it does not affect the population distribution in our system. However, its phase fluctuations do affect the coherent contribution to the gain coefficient [9]. This is due to fact that phases of atomic coherences cannot follow fast phase fluctuations of the fields and therefore, on average, coherent contributions are reduced. Indeed, the probe laser linewidth adds to the decay rate of the upper-level coherence. Consequently, if $\Delta\nu_a \geq \gamma_{a,b}$, the linewidth contribution dominates the coherence decay rate, and therefore reduces the contribution due to atomic interference in the gain coefficient. In the limit $\Delta\nu_a \gg \gamma_{a,b}$ the interference contribution vanishes, and therefore amplification without inversion should disappear.

In our original experiment the linewidths of probe and driving lasers are relatively narrow ($\Delta\nu \sim 100$ kHz $\ll \gamma_a = 5.4$ MHz, $\gamma_b = 5.6$ MHz). However, we can change these linewidths from 100 kHz to 50 MHz by modulating the diode laser's injection current with a noise signal. Figure 2(c) shows the result of such a modulation of the current to the probe laser. In this experiment all three lasers are present in the cell, and therefore without the additional modulation we see the typical amplification peak [curve a in Fig. 2(c)]. In the presence of the current modulation the linewidth of the probe laser was ~10 MHz, and we observe that the amplification disappears (curve b). Since the populations of the levels are not changed, this proves that the observed amplification is not due to population inversion, i.e., it is LWI due to quantum interference effects.

Having observed the amplification of a weak probe field and having verified the absence of a population inversion, we are able to demonstrate the buildup of laser oscillations in a cavity at the frequency corresponding to the $1 \rightarrow 2'$ transition of the D_1 line. To this end the Rb cell was placed inside the ring cavity [Fig. 3(a)]. One mirror of this cavity was formed by a polarization cube in order to allow for a substantial transmission of $\hat{x}$-polarized driving field, and, at the same time, high reflectance for a $\hat{z}$-polarized lasing field. The laser radiation at 794 nm was selected at the cavity output by using a monochromator. The frequencies of driving and incoherent pump lasers were controlled by using additional reference cells with Rb. The typical dependence of the output power from the cavity at 794 nm and corresponding absorption of the driving field in the reference cell is presented in the Fig. 3(b) as a function of driving field frequency. When driving laser (780 nm) is tuned near the center

of the D_2 absorption line ($1 \rightarrow 2''$ transition), the laser oscillation at 794 nm appears in the ring cavity. This laser field oscillates at the frequency corresponding to the $1 \rightarrow 2'$ transition of the D_1 line as was verified by making a beat note with an independent laser. The beat note measurement shows a direct one-to-one correspondence of the tuning of 795 nm laser with the frequency of the driving laser, which again confirms that the observed lasing is caused by coherent interaction of the drive laser with atoms. This tuning, along with the fact that the incoherent pump is spectrally broad, rules out the possibility that the lasing is due to Raman gain. Under conditions similar to that of the pump-probe experiment except with no probe present we observed approximately 30 μW of lasing power at 794 nm.

In conclusion, we have experimentally shown inversionless amplification and lasing in a Rb gas cell coherently prepared by a cw diode laser. To prove that the gain is "without inversion" we performed measurements with various linewidths of the probe laser, showing that when the linewidth of the probe laser exceeds the radiative decay rate, the amplification peak disappears. This establishes the LWI nature of the observed amplification and the absence of population inversion. And, finally, self-generated laser oscillation was observed when the inversionless gain medium was placed inside a laser cavity that was resonant with the appropriate transition of the D_1 line.

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[1] For recent theoretical work on LWI in multilevel schemes see, e.g., O. A. Kocharovskaya and Ya. I. Khanin, Pis'ma Zh. Exp. Teor. Fiz. **48**, 581 (1988) [JETP Lett. **48**, 630 (1988)]; S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989); M. O. Scully, S.-Y. Zhu, and A. Gavrielides, *ibid.* **62**, 2813 (1989); A. Imamoğlu, J. E. Field, and S. E. Harris, *ibid.* **66**, 1154 (1991). For a review of the subject, see O. Kocharovskaya, Phys. Rep. **219**, 175 (1992)

[2] Interaction of optical fields in a V-type atomic configuration was studied long ago [3] but recently attracted renewed attention and was investigated theoretically in the context of LWI, see, e.g., O. Kocharovskaya and P. Mandel, Opt. Commun. **84**, 179 (1991); M. Fleischhauer *et al.*, Phys. Rev. A **46**, 1468 (1992); W. Tan, W. Lu, and R. G. Harrison, *ibid.* **46**, 3613 (1992); G. A. Wilson, K. K. Meduri, P. B. Sellin, and T. W. Mossberg, *ibid.* **50**, 3394 (1994); M. Fleischhauer, T. McIllrath, and M. O. Scully, Appl. Phys. B (to be published).

[3] I. M. Beterov and V. P. Chebotaev, Sov. Phys. JETP Lett. **9**, 127 (1969); T. Hänsch *et al.*, Z. Phys. **226**, 293 (1969); T. Hänsch and P. Toschek, *ibid.* **236**, 213 (1970); M. S. Feld and A. Javan, Phys. Rev. **177**, 540 (1969).

[4] Xingfu Li *et al.*, in *Proceedings of the International Conference on Lasers '92, Houston, 1992* (STS Press, McLean, VA 1992), p. 446; A. Nottelmann, C. Peters, and W. Lange, Phys. Rev. Lett. **70**, 1783 (1993); E. S. Fry *et al.*, *ibid.* **70**, 3235 (1993); W. E. van der Veer *et al.*, *ibid.* **70**, 3243 (1993).

[5] Some interesting experimental studies of LWI continue to be a source of discussion. For example, in the interesting experiments of J. Gao *et al.* [Optics Commun. **93**, 323 (1992)] and Gao *et al.* [*ibid.* **110**, 590 (1994)] it is difficult to assess experimental evidence for the absence of inversion. Moreover, theoretical modeling which takes into account Zeeman sublevels shows that simple LWI of the type studied theoretically by L. Narducci *et al.* [Opt. Commun. **81**, 379 (1991); **86**, 324 (1991)] cannot, in fact, be observed under the conditions of these experiments, see, e.g., G. M. Meyer *et al.*, Quantum Opt. **6**, 231 (1994). Interpretation of some of the results in the recent experiment of J. A. Kleinfeld and D. A. Streater, Phys. Rev. A **49**, 4301 (1994), are also complicated from the theoretical point of view for a similar reason. Nevertheless, coherence effects were observed in these experiments. Further theoretical considerations along these lines will be published elsewhere.

[6] G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento B **36**, 5 (1976); E. Arimondo and G. Orriols, Nuovo Cimento Lett. **17**, 333 (1976); H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978).

[7] S H. Autler and C. H. Townes, Phys. Rev. **100**, 707 (1955).

[8] In the present paper we use the following notations to distinguish different sublevels of Rb^{87}. 1 and 2 denote the $F = 1$ and $F = 2$ hyperfine components of the $S_{1/2}$ ground state, $1'$ and $2'$ are hyperfine components of the first excited state $P_{1/2}$, and $0''$, $1''$, $2''$, and $3''$ are sublevels of the $P_{3/2}$ state with corresponding F.

[9] G. S. Agarwal, Phys. Rev. A **18**, 1490 (1978); B. J. Dalton and P. L. Knight, Optics Commun. **42**, 411 (1982); M. Fleischhauer *et al.*, *ibid.* **110**, 351 (1994).