Laser Oscillation without Population Inversion in a Sodium Atomic Beam

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Continuous wave (cw) amplification and laser oscillation without population inversion have been observed for the first time in a Λ scheme within the sodium D_1 line. This is also the first demonstration in which the lasing medium was an atomic beam; this is an approach which, in addition to elucidating the physics, lays a foundation for extensions into the ultraviolet. Calculations using realistic atomic structure were critical to the choice of experimental approach. Observations agree with full density-matrix calculations and clearly show there was no population inversion.

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Lasing without inversion (LWI) has attracted attention in recent years [1], as it opens new perspectives for laser physics via the effects of atomic coherence and interference [2]. The Λ and V schemes are frequently discussed techniques for LWI in atomic systems. In a Λ scheme [3] the fields have a common upper level, and LWI is insured by the coherence between two lower levels; whereas, in a V scheme [4] the fields share a lower level and LWI is made possible by coherence between upper levels. All previous LWI experiments employing a Λ scheme observed only transient gain (less than a few nsec) or gain on a probe laser pulse [5]. Electromagnetically induced transparency (EIT) based on these schemes has also been observed [6]; and related experiments involving a V scheme have been reported [7].

The first cw laser oscillator without inversion was recently reported by Zibrov *et al.* in a V configuration [8]. Here we describe a very different experimental demonstration of a cw laser oscillator without population inversion. It is based on the Λ scheme and is closest to the concept proposed by Imamoğlu, Field, and Harris [9]. The active medium is a sodium atomic beam and the transitions are within the D_1 line. By using a weak probe laser, we first demonstrate complete transparency and then inversionless gain. Next, a laser cavity is installed and aligned. With the probe blocked, we find that the laser starts spontaneously from vacuum fluctuations.

A generic Λ scheme for our LWI experiments is shown in Fig. 1. The atomic system consists of an upper level a connected by a dipole-allowed transition to two lower levels: to c via a strong "driving" field with Rabi frequency Ω_D , and to b via the "lasing" transition with Rabi frequency Ω_L . In preliminary studies a very weak "probe" field with Rabi frequency Ω_P was tuned through the region of the $a \leftrightarrow b$ transition frequency. To observe gain and lasing, some population is transferred into state a using an incoherent pump field $(b \leftrightarrow a)$ with a transition rate r_I . Level a decays radiatively to levels c and b at rates γ_c and γ_b , respectively; its radiative lifetime is $1/(\gamma_c + \gamma_b)$.

Since the active medium is an atomic beam, fresh atoms enter the probe interaction region in state b, and we model the exit via a decay of all states at a relatively slow rate γ' .

Significant insight into this generic Λ configuration is obtained by considering analytic forms for the steady state solutions of the density matrix equations [10]. At resonance and to first order in the laser field Ω_L , the gain is proportional to

$$\frac{\rho_{ab}}{-i\Omega_L} = \frac{\{(\rho_{aa}^0 - \rho_{bb}^0)\gamma_{bc} + (\rho_{cc}^0 - \rho_{aa}^0)|\Omega_D|^2/\gamma_{ac}\}}{\gamma_{ab}\gamma_{bc} + |\Omega_D|^2},$$
(1)

where ρ_{ab} is the off-diagonal element of the density matrix to the first order in Ω_L ; and $\rho_{aa}^0, \rho_{bb}^0, \rho_{cc}^0$ are the populations to zero order in Ω_L . The second term in the braces is a result of quantum interference; if $\rho_{cc}^0 - \rho_{aa}^0 > 0$, it can lead to gain even when $\rho_{aa}^0 - \rho_{bb}^0 < 0$, corresponding to no population inversion. The dephasing rates are $\gamma_{ab} = \gamma' + r_I + \gamma/2$, $\gamma_{ac} = \gamma' + (r_I + \gamma)/2$, and $\gamma_{bc} = \gamma' + r_I/2$, where $\gamma = \gamma_b + \gamma_c$.

We consider the limits $\gamma' \ll r_I$, $\gamma' \ll K$ where $K = r_I$

We consider the limits $\gamma' \ll r_I$, $\gamma' \ll K$ where $K = 4|\Omega_D|^2/(r_I + \gamma)$ is a pumping rate. From the analytic model, it can then be shown that the populations to zero order in Ω_L are $\rho_{aa}^0 = Kr_I/D$, $\rho_{bb}^0 = K(r_I + \gamma_b)/D$, $\rho_{cc}^0 = K(r_I + \gamma_b)/D$

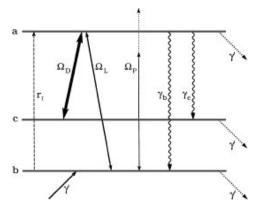


FIG. 1. The generic Λ scheme for our LWI experiments.

 $r_I(\gamma_c + K)/D$, where $D = K(3r_I + \gamma_b) + r_I\gamma_c$. These results together with Eq. (1) show that a necessary condition for gain (G > 0) on the lasing transition is $\gamma_c > \gamma_b$; i.e., radiative decay on the drive transition must be faster than on the lasing transition.

The zero order populations show that a population inversion $\rho_{bb}^0 < \rho_{aa}^0$ can never occur. The strongest condition for population inversion $\rho_{bb}^0 < \rho_{aa}^0 + \rho_{cc}^0$ only occurs if $r_I > \gamma_b/(1 + \gamma_c/K) \approx \gamma_b$; the approximation assumes an intense drive so that $K \gg \gamma_c$. Thus the strongest condition is met only if the incoherent pump rate exceeds the spontaneous decay rate of b. Specifically, for pump rates r_I less than γ_b , populations in any linear combination of states a and c must be less than in b, and inversion cannot exist in any basis of states [11]. Finally, the present system cannot be a Raman laser; that requires two photon inversion, $\rho_{bb}^0 < \rho_{cc}^0$, which only occurs for very high pump rates $r_I > (\gamma_b/\gamma_c)K$.

The energy level scheme for Na is shown in Fig. 2; levels are identified by their F value, with a prime for $3^2P_{1/2}$ states and no prime for $3^2S_{1/2}$ states. The 1 \leftrightarrow 1' transition was chosen for the laser oscillator since it has the slowest decay rate (as required by the gain condition $\gamma_c > \gamma_b$). This leaves the 2 \leftrightarrow 1' transition for the driving field. The interactions of polarized fields with all 16 states of the sodium D_1 line, including appropriate coupling constants between states with various angular momenta, were analyzed with full density-matrix calculations; Doppler broadening due to transverse components of velocity in the atomic beam was included. The vacuum wavelength is 589.76 nm and the upper levels lifetime is 16.1 nsec (natural linewidth $\gamma/2\pi = 9.9$ MHz). The drive and laser oscillator beams have perpendicular polarization; theoretical analysis shows this is essential for gain. The incoherent pumping field is on the $1 \leftrightarrow 1'$ transition; results are qualitatively similar regardless of its polarization.

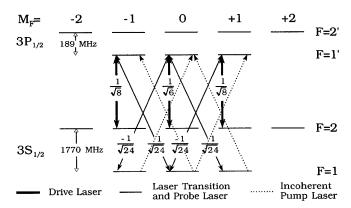


FIG. 2. Relevant energy levels of Na. Each transition is labeled with its angular momentum coupling constant. For comparison, the levels a, b, and c of the generic Λ scheme would each correspond to an appropriate level of F=1', F=1, and F=2, respectively.

The experimental setup is shown in Fig. 3. A sodium atomic beam was prepared by thermal effusion from a 0.5 nm diameter aperture in an oven at \approx 280–320 °C. It propagates along the Z axis and is collimated with slits to a 0.6×1 cm² cross section. All laser beams are perpendicular to the Z axis and intersect the atomic beam 15 cm from the oven aperture. The residual Doppler width is \approx 60 MHz. Three mutually perpendicular pairs of Helmholtz coils reduce the magnetic field throughout the interaction region to <5 mG.

The output of a frequency-stabilized cw ring dye laser (Coherent model 899, linewidth ~1 MHz) provides the drive beam. A polarizing beam splitter separates a small fraction of it which passes through an acousto-optic frequency shifter (AO) and is used as the probe beam. The AO provides a shift of \approx 1772 MHz and sweeps the probe beam frequency for the transparency and gain measurements. The probe laser intensity is stabilized using a commercial laser noise reduction unit. A linear polarizer in each beam sets its final polarization, another is rotated to vary its intensity. The probe laser lies on the Y axis and is X polarized. The fraction of the probe beam transmitted by the sodium atomic beam passes through a linear polarizer to a photodiode. The drive laser is in the X-Y plane at 2.0° to the Y axis and is Z polarized; the small angle was required to separate the two beams at the detector and to clear the mirror for the oscillator cavity.

The output of a second frequency-stabilized cw ring dye laser (Coherent model 699, linewidth ~ 1 MHz) passes through another AO which destroys its phase coherence and produces an incoherent pump beam. The AO was driven by an amplified white noise generator with a 40 MHz bandwidth centered at a frequency shift of 70 MHz. Since the linewidth of the laser is increased to ≈ 40 MHz ($>\gamma/2\pi$), this beam serves as an incoherent source [12]. Its bandwidth was verified by heterodyning with the unshifted laser. The incoherent pump beam

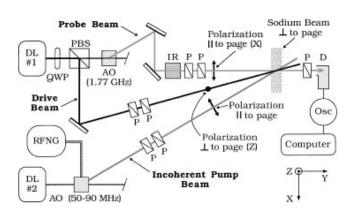


FIG. 3. Schematic of the experiment to observe inversionless gain and cw laser oscillation. DL, dye laser, QWP, $\lambda/4$ plate; PBS, polarizing beam splitter; AO, acousto-optic frequency shifter; RFNG, rf noise generator/amplifier; P, polarizer; IR, intensity regulator; D, detector.

propagates in the X-Y plane at 10.6° to the Y axis; results are relatively insensitive to this angle.

Figure 4(a) gives probe transmission through the Na atomic beam as a function of frequency in the neighborhood of the $1 \leftrightarrow 1'$ transition. Curve P is the transmission when only the probe laser is present. Curve D is the transmission in the presence of the strong drive laser; it shows that, at resonance, nearly complete EIT is achieved. Off resonance, the probe transmission is reduced in the presence of the strong drive due to optical pumping. Specifically, the drive beam has a larger diameter than the probe, and incoming atoms are first optically pumped by it. Atoms entering the probe interaction volume are completely pumped out of the important F = 2, $m_F = 0, \pm 1$ states (zero population). They are optically pumped into the F=2, $m_F=\pm 2$ states (which are out of play and can be disregarded) and into the F = 1 states (leading to greater off resonance absorption of the probe). Curve D&I shows the gain in the presence of the incoherent pump. Figure 4(b) shows a full density matrix calculation of the experimental results; the agreement is excellent.

The observed gain depends on the intensity of the incoherent pump laser, i.e., on the number of atoms pumped into the excited state. Up to 10% gain has been obtained using an atomic beam producing over 16% absorption in the absence of the drive. For the data shown, these parameters were 2% and 6%, respectively; the probe laser power was 0.3 μ W in a diameter of \approx 1 mm; the drive laser power was 10 mW in a diameter of \approx 3.5 mm, and the incoherent pump laser power was 1 mW in a diameter of \approx 3.5 mm. All diameters are $1/e^2$ intensity points. The corresponding reduced matrix element Rabi frequen-

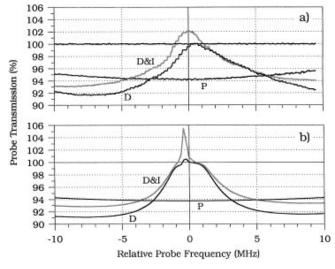


FIG. 4. Frequency dependence of probe laser transmission near the center of the $1 \leftrightarrow 1'$ transition: (a) experiment, (b) theory. The relatively smooth horizontal line is the transmission in the absence of sodium atoms and provides normalization; P, probe laser only; D, with drive laser, D&I, with the drive and incoherent pump lasers.

cies are $\tilde{\Omega}_P/2\pi=0.38$ MHz, $\tilde{\Omega}_D/2\pi=20$ MHz, and $\tilde{\Omega}_I/2\pi=6.4$ MHz, where we take $\tilde{\Omega}=\wp E/2\hbar$, E is the amplitude of the electric field, and the reduced matrix element is $\wp=\langle\alpha',J'=1/2\parallel\mu\parallel J=1/2,\alpha\rangle=3.01\times10^{-29}$ C m. The actual Rabi frequency for each transition is obtained by multiplying by the corresponding coupling constant from Fig. 2.

Other relevant parameters are the total decay rates $\gamma_{1'\to 1}/2\pi=1.65$ MHz and $\gamma_{1'\to 2}/2\pi=8.24$ MHz (calculations actually utilize individual decay rates between each pair of magnetic sublevels). The rate γ' at which atoms enter the interaction region (in the F=1 and F=2, $m_F=\pm 2$ states) and leave (via all states) is estimated as the average velocity of an atom in the beam divided by the probe laser diameter $\gamma'/2\pi=0.14$ MHz. Finally, we assume the incoherent pump has constant spectral intensity over a bandwidth $\Delta\omega\gg\gamma$; taking $\Delta\omega/2\pi=40$ MHz and using the square of the coupling constant from Fig. 2 (which is the same for all the incoherent pump transitions), we have $r_I=\pi\tilde{\Omega}_I^2/12\Delta\omega$ or $r_I/2\pi\approx0.3$ MHz for each transition.

The data of Fig. 4(a) also show there is no population inversion. When the drive and incoherent pump lasers are off, the ground state population is isotropic (uniformly distributed among all Zeeman sublevels of F = 1, 2). A measure of the fraction in the lower lasing level F = 1is given by the probe transmission (curve P). Since the drive beam has a larger diameter than the probe, incoming atoms are first optically pumped by it. Thus atoms entering the probe interaction volume are completely pumped out of the three important F=2, $m_F=0,\pm 1$ states, and into the F=1 and F=2, $m_F=\pm 2$ states (Fig. 2 basis of states). Atoms optically pumped into the latter are out of play, but now contain greater than isotropic populations. The increased population in the lower lasing level F = 1 is due to the optical pumping and leads to the observed reduction of probe transmission in the wings of the line, curve D. With the incoherent pump, the probe transmission in the wings of the line increases slightly (curve D&I), but is still less than curve P; i.e., the total population in the lower laser level is greater than isotropic, actually greater than the total population in all other states except perhaps the inactive F=2, $m_F=\pm 2$ states. This optical pumping by the drive leads to an absence of Raman inversion according to both our numerical simulations and auxiliary measurements of absorption from these levels.

After obtaining net LWI gain, a ring cavity was installed to observe cw laser oscillation. Figure 5 shows the cavity schematic; its free spectral range was 630 MHz (cavity length = 47.5 cm). First, the probe laser was coupled into the cavity through the curved mirror (CM). This facilitated precise alignment of the laser cavity and provided a verification of the gain condition. The probe beam was then blocked and the resonance frequency of the ring cavity was scanned via the PZT mounted

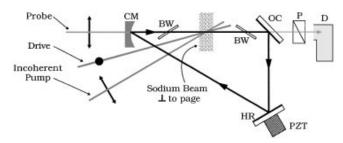


FIG. 5. Ring laser cavity. CM, curved mirror, R=99.5%; OC, output coupler, R=99.0%; HR, high reflector, $R\sim100\%$; PZT, piezotranslator; BW, Brewster window on the vacuum chamber.

high reflector (HR). Well-collimated outputs (transmitted through the output coupler) from the cavity were observed at $\approx\!600$ MHz intervals of the cavity scan; there was no observable output otherwise; see Fig. 6. When laser oscillation occurred, it tended to flicker on and off due to acoustic vibrations of the cavity length.

We confirm several predictions of inversionless lasers in a Λ scheme: (1) If pump and drive lasers have different detunings from line center, the gain and the laser output disappear. The probe and drive beams must be precisely aligned in the X-Y plane and must have orthogonal linear polarization in order to achieve $\approx 100\%$ probe transparency (EIT). (2) No gain could be observed and EIT was reduced if a coherent pump (noise source for AO turned off) was used; i.e., a coherent pump produces other atomic coherences that lead to population trapping and no excitation to the upper lasing level. (3) The drive and probe lasers must copropagate to produce 100% transparency, and laser oscillation is only observed around the ring cavity in a direction coinciding with that of the drive beam in the one leg of the ring cavity.

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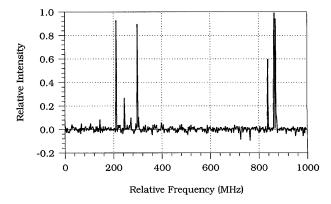


FIG. 6. Output of the LWI oscillator as the resonant frequency of the laser cavity is scanned. Observation parameters are peak gain of 5% corresponding to 9% linear absorption.

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- [1] O. Kocharovskaya and Y. I. Khanin, JETP Lett. 48, 585 (1988); S. E. Harris, Phys. Rev. Lett. 62, 1033 (1989); for a review, see O. Kocharovskaya, Phys. Rep. 219, 175 (1992). Early papers suggesting the possibility of LWI are T. Hänsch and P. Toschek, Z. Phys. 236, 213 (1970); T. Y. Popova and A. K. Popov, Sov. Phys. J. Appl. Spectrosc. 12, 734 (1973); V. G. Arkhipkin and Yu. I. Heller, Phys. Lett. 98A, 12 (1983). We also call the reader's attention to works by D. Grandclement, G. Grynberg, and M. Pinard, Phys. Rev. Lett. 59, 40 (1987), and by the group of T. W. Mossberg, *ibid.* 68, 464 (1992).
- [2] G. Alzetta *et al.*, Nuovo Cimento Soc. Ital. Fis. **36B**, 5 (1976); R.M. Whitley and C.R. Stroud, Jr., Phys. Rev. A **14**, 1498 (1976).
- [3] S. E. Harris, J. E. Field, and A. Imamoğlu, Phys. Rev. Lett. 64, 1107 (1990); A. Imamoğlu and S. E. Harris, Opt. Lett. 14, 1344 (1989).
- [4] M. Scully, in Proceedings of the NATO Advanced Research Workshop on Noise and Chaos in Nonlinear Dynamical Systems, Turin, Italy, 1989 (Plenum, New York, 1990); O. Kocharovskaya and P. Mandel, Opt. Commun. 84, 179 (1990); L. M. Narducci et al., ibid. 86, 324 (1991).
- [5] X. Li et al., in Proceedings of the International Conference on Lasers, Houston, Texas, 1992 (STS Press, McLean, VA, 1993), 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).
- [6] K.-J. Boller, A. Imamoğlu, and S.E. Harris, Phys. Rev. Lett. 66, 2593 (1991).
- [7] J. Gao *et al.*, Opt. Commun. **93**, 323 (1992); J. A. Kleinfeld and A. D. Streater, Phys. Rev. A **49**, R4301 (1994).
- [8] A. S. Zibrov et al., Phys. Rev. Lett. 75, 1499 (1995).
- [9] A. Imamoğlu, J. E. Field, and S. E. Harris, Phys. Rev. Lett. 66, 1154 (1991).
- [10] M. O. Scully, Quantum Opt. B 6, 203 (1994); G. M. Meyer et al., ibid. 6, 231 (1994); D. E. Nikonov et al., ibid. 6, 245 (1994).
- [11] In other words, the present LWI experimental results cannot be explained solely on the basis of a hidden inversion; atomic decay and pumping processes (e.g., r_I) lead to a partial contravention of population trapping. The deeper quantum interference and coherence physics that is essential to understanding the present experiment is contained in our expression for the gain.
- [12] M. Fleischhauer et al., Opt. Commun. 110, 351 (1994).