Atomic Coherence Effects within the Sodium D_1 Line: Lasing without Inversion via Population Trapping

Edward S. Fry,[(1),(2)] Xingfu Li,[(2)] Dmitri Nikonov,[(2)] G. G. Padmabandu,[(1),(2)] Marlan O. Scully,[(1),(2),(4)] Arlie V. Smith,[(6)] Frank K. Tittel,[(1),(5)] Ching Wang,[(1),(2)] Steve R. Wilkinson,[(3),(6)] and Shi-Yao Zhu[(2),(3)]

[(1)]*Texas Laser Laboratory, HARC, 4800 Research Forest Drive, The Woodlands, Texas 77381*
[(2)]*Department of Physics, Texas A&M University, College Station, Texas 77843*
[(3)]*Center for Advanced Study and Department of Physics, University of New Mexico, Albuquerque, New Mexico 87131*
[(4)]*Max-Planck-Institut für Quantenoptik, W-8046 Garching, Germany*
[(5)]*Department of Electrical Engineering, Rice University, Houston, Texas 77251*
[(6)]*Sandia National Laboratory, Department 1128, Albuquerque, New Mexico 87185*
(Received 16 February 1993)

Atomic coherence effects within the sodium D_1 line are shown to lead to the suppression of optical pumping, to the switching of light on and off when the coherence effects are turned on and off, and especially to lasing without inversion.

PACS numbers: 42.50.−p, 42.55.−f

Atomic coherence effects [1–4] are the basis of the most commonly discussed schemes for lasing without inversion (LWI) [5–11]. These schemes typically envision an intense beam preparing the medium so as to allow LWI on a weak probe. In the present paper, we present an experimental and theoretical demonstration of LWI and related coherence effects using a different type of system. In this system two strong beams prepare the atomic coherence as in Figs. 1(a) and 1(b) and LWI is observed as an increased intensity on these same strong beams.

To understand the basic idea behind these new experiments, consider the case of optical pumping within the states associated with the Na D_1 line as in Fig. 1(b). The field and intensity driving transitions $b' \to a$ are labeled E_1 and I_1 (frequency ν_1), respectively; similarly, for $b \to a$ they are E_2 and I_2 (frequency ν_2); see Fig. 1(b). From a naive rate equation perspective, one would expect that in the presence of both E_1 and E_2 all the atoms would be optically pumped into the b_2 state, i.e., at steady state $\rho_{b_2b_2}=1$ in the absence of collisions. However, due to the presence of coherent fields at ν_1 and ν_2, atomic coherences $\rho_{bb'}$ are generated between the pairs of states (b_0,b'_0), (b_{+1},b'_{+1}), and (b_{-1},b'_{-1}); thus population is trapped in these pairs of states and their population percentages are shown in Fig. 1(b).

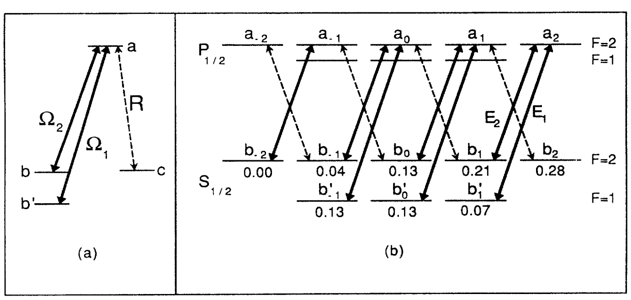

FIG. 1. (a) Two strong beams prepare a coherent superposition of b and b'. This leads to LWI on the same two transitions when a is populated via a weak pump at rate R from level c (four-level system). (b) The structure of the $3^2S_{1/2}$ and $3^2P_{1/2}$ levels of sodium. Shown above each sublevel is our designation for it; the subscript on a level designation is the m_F value for the level. Shown below the sublevels are their fractional steady state populations under the action of two strong RCP fields (heavy lines) of equal intensity; initially isotropic populations are assumed. The weak LCP field that provides an upper level population is shown as dashed lines.

The atomic coherence between and trapping within these paired states modifies the optical pumping of the ground states. It is manifested in the phenomenon of "coherence switching" which is reported here. Having demonstrated (via coherence switching) the presence of ground-state coherences and population trapping, we are in a position to observe LWI by exciting a small fraction of the atoms to the a state [Fig. 1(a)] or the a_0 and a_1 states [Fig. 1(b)].

In order to most simply understand the physics of the present coherence switching and LWI, we consider the Na atomic structure in Fig. 1(b). The essence of this structure, as it applies to the present problem, is explained by two four-level groupings: $a_0,(b_{-1},b'_{-1}),b_1$ and $a_1,(b_0,b'_0),b_2$, each of which is equivalent to the simpler level scheme of Fig. 1(a). It is easy to see that there will be populations coherently trapped in the (b,b') pair of Fig. 1(a). Consider the case of resonance and equal Rabi frequencies Ω_1 and Ω_2. Then the symmetric state $|s\rangle=[|b\rangle+|b'\rangle]/\sqrt{2}$ is coupled to the fields, and the antisymmetric state $|\bar{s}\rangle=[|b\rangle-|b'\rangle]/\sqrt{2}$ is uncoupled from the fields [2]. Thus, if we start with b, b', and c equally populated, then after many optical pumping cycles there will be a steady state in which the $|s\rangle$ state will be depleted while the $|\bar{s}\rangle$ and $|c\rangle$ states will each have 50% of the population (assuming equal decay rates). In the b, b', c basis this corresponds to $\rho_{b,b}=\frac{1}{4}$, $\rho_{b'b'}=\frac{1}{4}$, and $\rho_{cc}=\frac{1}{2}$. In other words, half of the atoms are trapped in the (b,b') pair; this is a manifestation of atomic coherence $\rho_{b,b'}$.

Now if we turn off one of the fields, say E_2, and thereby negate the effects of atomic coherence, the E_1 field will be strongly absorbed. Thus turning off the effects of atomic coherence (by turning off E_2) effectively "switches off" E_1, since there is a large population of sodium atoms that can now absorb it. This is the coherent switching effect shown theoretically in Fig. 2(a) and experimentally in Fig. 2(b).

On the other hand, if in the presence of the atomic coherence we move a small fraction of the atoms into state a, such that $\rho_{aa}<\rho_{bb}$ and $\rho_{b'b'}$, then we would expect LWI. This is indeed found to be the case, as is shown theoretically in Fig. 3(a) and experimentally in Fig. 3(b).

These are the main results of the present paper. In order to clearly demonstrate and understand the physics behind these effects, we present the theoretical basis for Figs. 2(a) and 3(a), and then discuss the experimental arrangement leading to the results given in Figs. 2(b) and 3(b).

For clarity, we will first consider the simple four-level system of Fig. 1(a). The Hamiltonian in the interaction picture is $V=\wp_1E_1|a\rangle\langle b'|+\wp_2E_2|a\rangle\langle b|+\wp_3E_3|a\rangle\langle c|+\text{H.c.}$, where E_i $(i=1,2,3)$ are the electric field amplitudes and $\wp_i$ represents the electric dipole matrix element. The dynamics of the system is described by the master equation for the density matrix ρ:

$$\dot{\rho}=(1/\hbar)[V,\rho]+L\rho+R\rho\,, \qquad (1)$$

where operators L and R correspond to relaxation processes and incoherent pumping, respectively, and $\Omega_i=\wp_iE_i/\hbar$.

We note that the polarization equation $\rho_{ab'}$ has interference terms such as $i\Omega_1\rho_{bb'}$ in addition to the usual gain and loss terms, $i\Omega_2(\rho_{aa}-\rho_{b'b'})$. This interference term, determined by atomic coherence $\rho_{bb'}$, can thus lead to absorption cancellation for field E_2. Notice this cancellation can only occur in the presence of E_1. Without E_1, the atomic coherence $\rho_{bb'}$ does not play a role and we have no cancellation of absorption.

In order to calculate the fields E_1 and E_2, we first solve for the atomic polarizations $P_1=\wp N\rho_{ab'}+\text{H.c.}$ and $P_2=\wp N\rho_{ab}+\text{H.c.}$, where N is the atomic number density. These polarizations then act as driving terms in Maxwell's equations, which in the slowly varying amplitude and phase approximation yield [12,13]

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathcal{E}_i(z,t)=-\frac{\nu}{2c\varepsilon_0}\mathcal{P}_i(z,t)\quad(i=1,2)\,, \qquad (2)$$

where $\mathcal{E}_i(z,t)$ is the slowly varying complex amplitude and $\mathcal{P}_i(z,t)$ is the imaginary part of the slowly varying polarization. In this way we calculate the evolution of the field E_1 (or E_2) after switching off field E_2 (or E_1).

In an analogous way we solve the full density matrix equations appropriate to sodium, i.e., Fig. 1(b). In Fig. 2(a) we plot the results for the output intensity of field E_1 as a function of time after E_2 is switched off. It is clear that E_1 experiences a sudden absorption after E_2, and thus the coherence effects are turned off. That is, after E_2 is switched off, the ground-state fractions $\rho_{b'_{-1},b'_{-1}}$

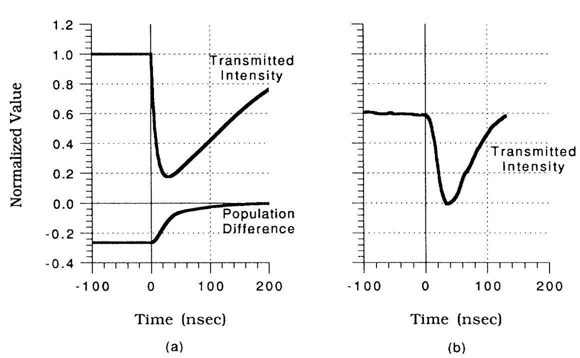

FIG. 2. (a) Theoretical prediction for the normalized intensity of the strong field E_1 that is transmitted by a sodium gas as a function of time after the other strong field E_2 is switched off. The complete Na structure of Fig. 1(b) is used in these calculations. Initially the system is in a steady state under the action of both strong RCP fields E_1 and E_2. The population difference $(\rho_{aa}-\rho_{b'b'})$ is shown at the bottom. (b) Experimental measurement of the transmitted intensity (arbitrary units) corresponding to the theoretical prediction for (a). The drop in the transmission of E_1 is a consequence of the coherently trapped population that is released when E_2 is turned off.

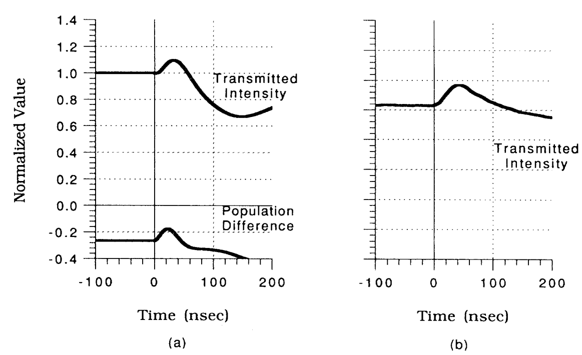

FIG. 3. (a) Theoretical prediction for the normalized intensity of the strong field E_1 that is transmitted by a sodium gas as a function of time after a weak LCP excitation field E_3 is switched on. The complete Na structure of Fig. 1(b) is used. Initially the system is in a steady state under the action of both strong RCP fields E_1 and E_2 and these fields are not changed. Also shown is the population difference $(\rho_{aa}-\rho_{b'b'})$; clearly, there is no population inversion. (b) Experimental measurement of the transmitted intensity (arbitrary units) corresponding to the theoretical prediction for (a). The increase in the transmission of E_1 is a consequence of stimulated emission and demonstrates LWI.

$=0.13$, $\rho_{b'_0,b'_0}=0.13$, and $\rho_{b'_1,b'_1}=0.07$, as shown in Fig. 1(b), are free to absorb field E_1, and the intensity I_1 drops immediately. However, after a few pumping cycles the b'_i states are depleted, and E_1 is again transmitted; this accounts for the increase in the intensity I_1 after it reaches the minimum shown in Fig. 2(a).

Figure 4 illustrates the generic experimental system used for these studies. The Na cell was constructed of Pyrex, typically 19 mm diameter×40 mm long with flat windows on each end. Appropriate sodium vapor densities were obtained by maintaining the cell at various temperatures in the 150 °C to 200 °C range using electric heating tapes. The oven was placed at the center of three mutually orthogonal pairs of Helmholtz coils which reduced the transverse (X,Y) components of the Earth's magnetic field to less than 3 mG over the volume occupied by the oven. The Z component (parallel to the direction of the laser beam) of the magnetic field was 300 mG.

The laser is a frequency stabilized cw dye laser tuned to frequency ν_2 [i.e., the frequency of the $3S_{1/2}(F=2)\to 3P_{1/2}(F=2)$ component of the Na D_1 line] [14]. The laser linewidth is ≈ 30 MHz. This dye laser beam passes through an acousto-optic frequency shifter (Brimrose model GPF 177-50) to produce a second beam shifted in frequency by $\Omega=1.77$ GHz, corresponding to the hyperfine splitting of the Na ground level. This second beam, frequency ν_1, drives the $3S_{1/2}(F=1)\to 3P_{1/2}(F=2)$ component of the Na D_1 line. Both beams are right circularly polarized (RCP) by using a quarter waveplate.

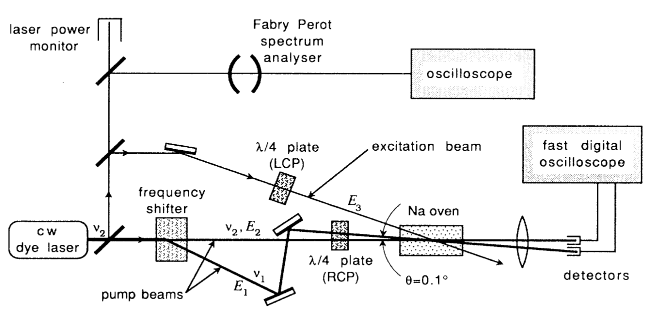

FIG. 4. Schematic of the experimental apparatus. A single frequency cw dye laser is used as the source. Its output is split and is also passed through an acousto-optic frequency shifter to obtain three beams: two strong pump beams E_1 and E_2 with frequencies $\omega+\Omega$ and ω, respectively, and a weak excitation beam E_3 with frequency ω. Here, ω is the resonant frequency of the $3S_{1/2}(F=2)\to 3P_{1/2}(F=2)$ transition, and Ω is the frequency of the ground-state hyperfine splitting. The two strong pump beams cross and completely overlap in the Na cell; the angle between them is 0.1° and they both pass through the same quarter wave plate to produce RCP. A lens following the Na cell focuses the two beams onto separate detectors whose outputs are monitored with a fast digitizing oscilloscope. Laser power is monitored for normalization, and the laser frequency is monitored using a Fabry-Pérot spectrum analyzer.

The angle between the beams is $\theta=0.1°$, and they completely overlap in the Na cell. A lens of focal length f following the cell focuses the beams to two spots with a separation θf: The transmission of the beams through the cell is separately monitored with two fast photodiodes and a 125 MHz digitizing oscilloscope. The initial intensity of each beam is ≈ 20 mW, and the beam diameters are 6 mm. A beam splitter in front of the frequency shifter samples the direct dye laser output and provides beams for monitoring laser power and laser frequency as well as providing a third weak excitation beam that is left circularly polarized (LCP) and has frequency ν_2. The field of this third beam will be denoted by E_3. Not shown in Fig. 4 are Pockels cells that can be inserted in any of the beams and used to turn them on or off in any combination with approximately a 1 ns rise time.

If both the E_1 and E_2 fields are applied, then the atoms will be optically pumped and coherently prepared. Ideally, in the absence of collisions, both beams would be completely transmitted. The theoretical predictions for the fractions of the atoms trapped in the various sublevels due to coherences are shown in Fig. 1(b) (assuming isotropic initial populations). If we now switch off the E_2 field, then the coherences no longer play a role and the field E_1 will be absorbed until the levels b'_{-1},b'_0,b'_1 are emptied, at which time the E_1 field will be transmitted again. The time-dependent experimental results for the transmitted field E_1 when the field E_2 is switched off are given in Fig. 2(b); the minimum transmissivity is 25% less than that with both fields on. This experimental result proves that the population is not totally pumped to the $m=2$ sublevel b_2, and some population is coherently trapped in the $(b_0b'_0)$, $(b_{-1}b'_{-1})$, and $(b_1b'_1)$ pairs; the experiment directly demonstrates this trapped population.

Having set the stage proving that population is coherently "locked" in the $F=1$ and 2 manifolds, we can utilize this configuration to demonstrate LWI. First let us look at the four-level system, Fig. 1(a). As soon as the coherence $\rho_{bb'}$ is established, lasing without inversion can be achieved by moving a small population from c to a. That can be accomplished by using an incoherent or coherent field E_3. The atoms are initially in the steady state under the action of E_1 and E_2; this establishes the coherent trapping in b and b'. After switching on the LCP field E_3, the transmitted field E_1 initially increases due to stimulated emission. The theoretical result is shown in Fig. 3(a) and the corresponding experimental result in Fig. 3(b). The dotted line in Fig. 3(a) is the population difference $(\rho_{aa}-\rho_{b'b'})$; since it is always negative, there is no inversion. In Fig. 3(a) we note that although the field initially rises, it begins to decrease after a few radiative lifetimes. This occurs because the population of atoms that can be moved to the excited state is depleted and absorption begins to dominate. During this latter absorption phase, population in excess of the trapping fraction accumulates in state b', and the transmission falls below the $t=0$ value. The increase (10% maximum) in the transmitted intensity I in Fig. 3(b) provides explicit experimental confirmation of LWI [15,16].

For further verification that there is no population inversion, a direct experimental test was made of the necessity for the coherence produced by E_1 and E_2. Specifically, the above experiment demonstrating LWI was repeated except at the time the LCP excitation pulse (field E_3) was turned on, the RCP field E_2 was turned off. The coherence effects are lost when E_2 is turned off, and the experimental observation of the transmitted intensity I_1 shows only absorption. This is in agreement with the theoretical predictions and is strong confirmation that the present result is LWI.

Absorption cancellation due to atomic coherences has been demonstrated. Previous experiments indirectly proved this population trapping by observing fluorescence from the upper level; here it has been directly shown by observing their absorption. Finally, gain due to LWI has been demonstrated; it is a direct consequence of the ability of a phase coherent atomic ensemble to cancel absorption.

We gratefully acknowledge support from the Office of Naval Research under Grant No. N00014-93-1-0155, from the Welch Foundation under Grant No. A-1218, and from the General Dynamics Corporation. We also wish to thank E. Arimondo, C. Benkert, F. B. Dunning, M. Fink, M. Fleischhauer, W. Gawlik, M. Graf, T. Hänsch, S. Harris, D. Heinzen, G. Herling, J. Keto, T. Kunz, P. Lambropoulos, G. M. Meyer, H. Pilloff, U. Rathe, C. Su, R. G. Torti, and H. Walther for helpful and stimulating discussions, and Electro-Scientific Industries for the loan of an Ar^+ laser.

[1] The photon echo experiments of I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. **141**, 391 (1966), provide a dramatic example of atomic coherence in quantum optics.

[2] For early work involving ground-state ("pair") coherence and population trapping, see G. Alzetta, L. Moi, and G. Orriols, Nuovo Cimento **52B**, 209 (1979); and H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978).

[3] For the application of atomic coherences to laser cooling, see an excellent review of the subject, C. Cohen-Tannoudji and W. Phillips, Phys. Today **43** (10), 33 (1990).

[4] A laser physics application of excited state "pair" coherence is provided by the correlated emission laser; see M. Scully, Phys. Rev. Lett. **55**, 2802 (1985); and M. Winters, J. Hall, and P. Toschek, Phys. Rev. Lett. **65**, 3116 (1990).

[5] S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989); A. Imamoğlu and S. E. Harris, Opt. Lett. **14**, 1344 (1989).

[6] O. Kocharovskaya and Ya. I. Khanin, Pis'ma Zh. Eksp. Teor. Fiz. **48**, 581 (1988) [JETP Lett. **48**, 630 (1988)]; O. Kocharovskaya and P. Mandel, Phys. Rev. A **42**, 523 (1990).

[7] M. Scully, S. Y. Zhu, and A. Gavrielides, Phys. Rev. Lett. **62**, 2813 (1989); M. Scully, in *Proceedings of the NATO Advanced Research Workshop on Noise and Choas in Nonlinear Dynamical Systems, Torino, Italy, 7–11 March 1989* (Plenum, New York, 1990).

[8] A. Lyras, X. Tang, P. Lambropoulos, and J. Zhang, Phys. Rev. A **40**, 4131 (1989).

[9] A. Karawajczyk, J. Zakrzewski, and W. Gawlik, Phys. Rev. A **45**, 420 (1992).

[10] 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). They present an interesting experiment designed to show LWI; the extent to which it does so continues to be a source of discussion. We wish to thank B. Wellegehausen for sending us his data and thoughts on this problem.

[11] L. M. Narducci, H. M. Doss, P. Ru, M. O. Scully, S. Y. Zhu, and C. Keitel, Opt. Commun. **81**, 379 (1991).

[12] See, for example, M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, MA, 1974).

[13] A. Yariv, *Quantum Electronics* (Wiley, New York, 1967).

[14] We note that in the actual experiment, it is hard to differentiate between the $3P_{1/2}$, $F=1$, and $F=2$ manifolds. In practice, the frequencies are simply adjusted to maximize the signal. However, the physics of the $F=1$ and $F=2$ cases are similar and the discussion is couched in terms of the $3P_{1/2}$, $F=2$ levels as the upper manifold.

[15] S. Y. Zhu, M. O. Scully, and E. E. Fill, *Coherence and Quantum Optics VI*, edited by J. H. Eberly, L. Mandel, and E. Wolf (Plenum, New York, 1989).

[16] After the submission of this work, an interesting LWI paper by A. Nottelmann, C. Peters, and W. Lange [Phys. Rev. Lett. **70**, 1783 (1993)] appeared. We also wish to call the reader's attention to the works of D. J. Gauthier, Q. Wu, S. E. Morin, and T. W. Mossberg [Phys. Rev. Lett. **68**, 464 (1992)] and D. Grandclement, G. Grynberg, and M. Pinard [Phys. Rev. Lett. **59**, 40 (1987)], which bear on the present studies.