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

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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.

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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 $\nu_1$), respectively; similarly, for $b\to a$ they are $E_2$ and $I_2$ (frequency $\nu_2$); 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 $\rho_{b_2}=1$ in the absence of collisions. However, due to the presence of coherent fields at $\nu_1$ and $\nu_2$, atomic coherences $\rho_{bb'}$ are generated between the pairs of states $(b_0,b_0'), (b_1,b_2'), (b_0,b_2'),$ and $(b_0,b_2)$; thus population is trapped in these pairs of states and their population percentages are shown in Fig. 1(b).

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: $a_0, (b_0,b_0'),(b_1$ and $a_1, (b_0,b_0'), h_2$, 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 $\Omega_1$ and $\Omega_2$. Then the symmetric state $|s\rangle\equiv(|b\rangle+|b'\rangle)/\sqrt{2}$ is coupled to the fields, and the antisymmetric state $|c\rangle\equiv(|b\rangle-|b'\rangle)/\sqrt{2}$ 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 $|s\rangle$ and $|c\rangle$ states will each have 50% of the population (assuming equal decay rates).
the $b$, $b'$, $c$ basis this corresponds to $\rho_{b,b} = \frac{1}{4}$, $\rho_{b',b'} = \frac{1}{4}$, and $\rho_{c,c} = \frac{1}{2}$. 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 $\rho_{bb'}$, 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

$$H = \hbar E_1 |a\rangle\langle b| + \frac{1}{2} \hbar E_2 |a\rangle\langle b| + \frac{1}{2} \hbar E_3 |a\rangle\langle c| + \text{H.c.},$$

where $E_i$ $(i=1,2,3)$ are the electric field amplitudes and $\rho_i$ represents the electric dipole matrix element. The dynamics of the system is described by the master equation for the density matrix $\rho$:

$$\dot{\rho} = \frac{\hbar}{i} [V, \rho] + L \rho + R \rho,$$

where operators $L$ and $R$ correspond to relaxation processes and incoherent pumping, respectively, and $\Omega_i = \frac{\rho_i}{\hbar}$. We note that the polarization equation $\rho_{ab}$ has interference terms such as $i \Omega_1 \rho_{ab}$ in addition to the usual gain and loss terms, $i \Omega_2 (\rho_{aa} - \rho_{bb})$. 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 $P_1 = \rho N \rho_{ab} + \text{H.c.}$ and $P_2 = \rho N \rho_{bb} + \text{H.c.}$, 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]

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

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 $\rho_{b',b',1}$

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 $(\rho_{aa} - \rho_{bb'})$ 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 $(\rho_{aa} - \rho_{bb'})$; 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.
\[ \rho_{bb'} = 0.13, \quad \rho_{bb''} = 0.13, \quad \text{and} \quad \rho_{b'1} = 0.07, \] 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 \( b'1 \) 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 \times 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\(^\circ\)C to 200\(^\circ\)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 \( v_2 \) [i.e., the frequency of the \( 3S_{1/2}(F=2) \rightarrow 3P_{1/2}(F=2) \) component of the Na \( D_1 \) line] [14]. The laser linewidth is \( \approx 30 \) 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 \( v_1 \), drives the \( 3S_{1/2}(F=1) \rightarrow 3P_{1/2}(F=2) \) component of the Na \( D_1 \) line. Both beams are right circularly polarized (RCP) by using a quarter waveplate.

The angle between the beams is \( \theta = 0.1^\circ \), 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 \( \theta f \). 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 \( \approx 20 \) 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 \( v_2 \). 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 \( b'_{-1}, b', b_{1} \) 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 \( (b_0 b_0'), (b_{-1} b'_{-1}), \) and \( (b_1 b_1') \) 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, losing 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 \( \langle \rho_{aa} - \rho_{bb'} \rangle \); 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% max-

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 \( \omega \), respectively, and a weak excitation beam \( E_3 \) with frequency \( \omega \). Here, \( \omega \) is the resonant frequency of the \( 3S_{1/2}(F=2) \rightarrow 3P_{1/2}(F=2) \) transition, and \( \Omega \) 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\(^\circ\) 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 with a fast digitizing oscilloscope. Laser frequency is monitored using a Fabry-Pérot spectrum analyzer.
imum) 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 $E_1$) 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 $I$ 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.

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[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.
