Experimental Demonstration of Light Amplification without Population Inversion

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We report the observation of light amplification without population inversion. A linear superposition of an $m = 1$ and an $m = -1$ magnetic substate is populated coherently. Absorption from this superposition to a higher state is canceled by interference of channels, a so-called dark resonance. An inverse transition is used for amplification of light. The temporal behavior of the amplified pulse and the gain are studied as a function of the energy splitting due to a magnetic field. In a vapor of cadmium-112 gain factors of up to 4.3 are observed.

The subject of lasers without inversion is intriguing and has recently drawn a great deal of attention. Many theoretical articles have been published, which were reviewed recently by Kocharovskaya [1] and by Scully [2]. The observation of light amplification without population inversion has been claimed previously by Gao et al. [3]. However, we like to emphasize that apart from the observation of amplification also the lack of inversion should be established. In the experiment mentioned, the determination of this lack of inversion is not clear.

In this Letter we present the experimental observation of both amplification and the absence of population inversion. We would like to make a closer examination of two types of inversionless lasers: lasing in a two-level inversionless system and lasing in a three-level inversionless system. In the latter case a V or a Λ system is considered.

In the two-level case, evidently, the energy difference between the upper and the lower level $E_U - E_L$ is the same as the photon energy $\hbar\omega$ of the incoming light. If the two-level atom is represented by a harmonic oscillator, the energy transfer between field and atom resembles the textbook case of a forced oscillation. There is gain, independent of the (nonzero) population of the two levels, if the phase difference between the oscillator and field is between 0 and π. An example is the scheme introduced by Harris [4], but also more familiar situations as Rabi oscillations [5–7]. However, the required phase relation between field and atom precludes a noncoherent population of the upper state.

The nature of the three-level inversionless laser is different [8–10]. Here, a statistical population of the upper level is allowed. There is no phase relation between the carrier frequency of the light and the difference frequency of the upper and lower levels. However, either the upper level or the lower level is split, although this splitting is much smaller than that between upper and lower states. If these states are coherently populated, transitions to other higher levels will be periodically prohibited, due to destructive interference between the

contributions of the two levels [11]. This cancellation of absorption is generally known as dark resonance. The periodicity can be associated with the energy difference between the two levels E_1 and E_2 [Fig. 1(a)]. Although the absorption is canceled, stimulated emission is still possible to linear combinations of the lower states other than the prohibited combination. Under these conditions it is possible to convert population of the upper level into gain, irrespective of the initial population of the lower levels. As mentioned, the presence of a dark resonance is only temporary. Therefore, in the case of pulsed operation the duration of the seed pulse has to be shorter than $h/(E_2 - E_1)$, and in the case of continuous operation the beam has to be amplitude modulated with the frequency $(E_2 - E_1)/h$.

In many systems discussed in literature the coherence between the lower states is introduced by a rf or microwave field [10,12]. The relative amount of atoms in the coherent superposition depends on the power of this field. In the experiment discussed below all the atoms are excited into a coherent superposition.

We would like to stress that in our system the gain is introduced by coherence. The light fields driving this sys-
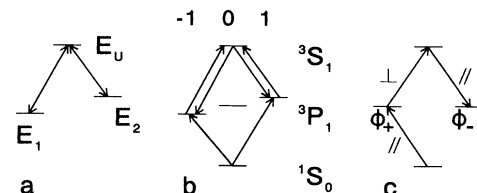

FIG. 1. (a) Level scheme for inversionless amplification in the three-level system. (b) The $m = 1$ and $m = -1$ states are populated coherently. (c) The $^3P_1$ level is populated by a light pulse with a $\parallel$ polarization, resulting in a ϕ_+ state. A second pulse (polarization $\perp$) populates the $^3S_1$ level. A probe pulse (polarization $\parallel$) stimulates the transition to the ϕ_- state. This arrangement of polarization yields a maximum amplification at $B = 0$.

tem have a relatively small intensity; therefore Rabi oscillations are not important to this scheme.

In the experiment cadmium atoms are placed in a magnetic field. A linearly polarized light pulse (pump 1) excites ground state atoms $(5s^2\, ^1S_0)$ to a coherent superposition of the $|m=1\rangle$ and the $|m=-1\rangle$ magnetic substates of the $5s5p\, ^3P_1$ state [Fig. 1(b)]. Let us suppose that this superposition is the $\phi_+ = (|m=1\rangle + |m=-1\rangle)/\sqrt{2}$ state; then the $\phi_- = (|m=1\rangle - |m=-1\rangle)/\sqrt{2}$ state is still empty [Fig. 1(c)].

A second light pulse, pump 2, excites a part of these atoms to the $5s6s\, ^3S_1$ state. The polarization of this pulse is perpendicular to that of pump 1, ensuring a maximum population of the $5s6s\, ^3S_1$ state at $B=0$. A weak seed beam, polarized parallel to the polarization of pump 1, stimulates the transition from the $^3S_1$ to the ϕ_- state. The energy splitting due to magnetic field causes a temporal evolution of the coherence between the ϕ_+ and ϕ_- states; the population is periodically transferred between these two states. Therefore amplification can only be observed when the population has returned to ϕ_+.

At larger magnetic field strengths this oscillation time becomes short compared to the duration of the laser pulse. Atoms excited at different moments in the laser pulse have a different phase factor. The level splitting due to the magnetic field therefore enables an adjustable dephasing of the ensemble of atoms. Under these conditions ϕ_+ and ϕ_- cannot be populated separately and the gain is lost. This is used to prove experimentally that *no population inversion is present* in the atomic system. Apart from the Zeeman splitting the $^3P_1$ state may also experience hyperfine splitting. In cadmium this occurs in isotopes 111 and 113. This can cause a disturbance of the induced coherence [11,13]. To prevent this, we use a sample of highly enriched ^{112}Cd isotopes.

The experimental apparatus consists of two dye lasers driven by a Q-switched Nd:YAG laser, a heated cell with

cadmium vapor, and light detectors with the associated electronics (see Fig. 2).

The first dye laser consists of a Hänsch type dye oscillator, a transversely pumped amplifier and a longitudinally pumped amplifier. The organic dye is DCM dissolved in methanol. The generated light is doubled to the wavelength of 326 nm by a KDP crystal. The beam is linearly polarized.

The light for the second excitation step is provided by dye laser 2. It consists also of a Hänsch type dye oscillator and a transversally pumped amplifier. The cavity of this oscillator is fitted with an etalon, which limits the bandwidth to approximately 1.3 GHz. This is of the same magnitude as the Doppler width of the excited transitions: ≈ 1 GHz. The dye is coumarin 102 dissolved in methanol. The light pulse generated by this laser can be delayed with respect to the pulse from dye laser 1. With the polarizers P_2 and P_3 this beam is polarized perpendicular to the polarization of the first laser beam. The energy of both the pump pulses is ≈ 1 μJ; the duration is ≈ 6 ns.

A small part of the light from laser 2 is delayed and the intensity is reduced further by placing the polarizer P_4 almost perpendicular to P_5. The resulting seed pulse is directed via beam splitter BS_1 into the cell. The polarization is parallel to that of pump 1. After passing through the cell the seed pulse is separated from the pump beams by the polarizing beam splitter PB and directed into the photomultiplier PM_2. The polarizer P_6 and the UV absorption filter f_2 are used to reject stray light originating from the pump beams.

The cell consists of an evacuated fused silica envelope, which contains 1 mg cadmium 112 enriched to 96%. The diameter of the cell is 1 cm; the volume is 3 cm^3. To maintain the sample free from contaminations, a gas-sorption getter (SAES St 707) is sealed in the cell. The cell is mounted in an oven heated by a resistance wire, wound bifilarly to prevent stray magnetic fields. For the same reason no ferromagnetic materials were used in the vicinity of the cell. A well defined homogeneous magnetic field, parallel to the pump and seed beams, is supplied by a set of magnetic field coils.

The cell is heated to a temperature of 220°C. This is the maximum temperature at which the coherence in the $^3P_1$ state is not measurably affected by interatomic collisions. The vapor pressure is 1.3×10^{-3} mbar (1.9×10^{13} atoms/cm^3). The diameter of the pump and seed beams is 2 mm; the number of atoms in the interaction region is therefore 6×10^{11}.

The wavelength of the dye lasers is adjusted by observing the fluorescent radiation from the cell. To tune dye laser 1 a UV transmission filter f_1 is placed in front of photomultiplier PM_1, which enables the observation of the fluorescent radiation emitted in the decay from the $5s5p\, ^3P_1$ state to the $5s^2\, ^1S_0$ ground state. Replacing f_1 by an interference filter transparent at 508 nm $(5s6s\, ^3S_1 - 5s5p\, ^3P_2)$ enables the observation of the popu-
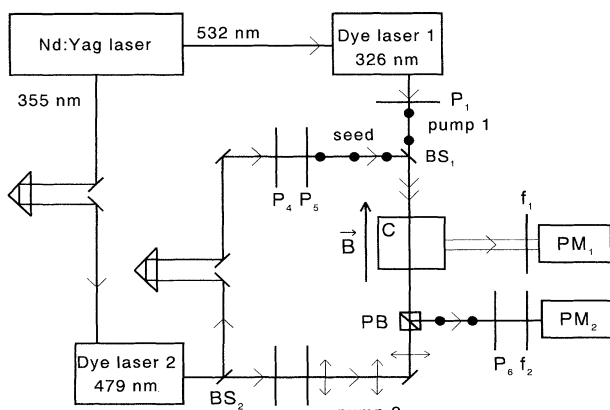
FIG. 2. The experimental setup. $P_{1...6}$ polarizers, $BS_{1,2}$ beam splitters, PB polarizing beam splitter, $f_{1,2}$ filters, $PM_{1,2}$ photomultipliers, and C cell with cadmium vapor.
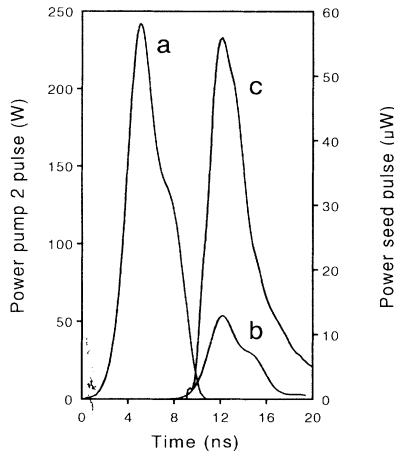
FIG. 3. The temporal behavior of curve a, the pump 2 pulse (left scale); curve b, the seed pulse (right scale); curve c, the amplified pulse (right scale).

FIG. 4. Fluorescent intensity (a) of the $^3S_1$-$^3P_2$ transition and gain (b) as a function of the magnetic field strength. The pump 2 pulse is delayed 30 ns with respect to the pump 1 pulse.

lation of the $5s\,6s\,^3S_1$ state while eliminating stray light from the pump pulses.

The temporal behavior of the (amplified) seed pulse or the fluorescent signals are observed with PM_2 or PM_1, respectively, and a fast transient digitizer. The signals can also be averaged over the duration of the pulse with a gated integrator. Scanning the current through the magnetic field coils with a programmable power supply allows the measurement of the gain or the intensity of the fluorescent radiation as a function of the magnetic field strength.

The amplification of the seed pulse is observed under various timing and magnetic field conditions. In Fig. 3 the temporal behavior of the pump 2 pulse (curve a), the seed pulse (curve b), and the amplified seed pulse (curve c) are shown. The delay of the pump 2 pulse with respect to the pump 1 pulse is 30 ns. The seed pulse is delayed 7 ns with respect to the pump 2 pulse. When the gain factor is defined as the integral of the amplified pulse divided by the integral of the seed pulse, we observe a gain of 4.3 for a seed pulse with a power 5×10^{-14} J (10^5 photons). Note that the intensity of the amplified pulse rises more steeply than that of the seed pulse. While the pulse is amplified, the coherence in the $^3P_1$ state is affected and the gain is reduced.

During the interval between the pump 1 and pump 2 pulses the coherence in the $^3P_1$ state can evolve, which causes a modulation of the probability of excitation to the $^3S_1$ state. In Fig. 4(a) the intensity of the fluorescent radiation is given as a function of the magnetic field strength (see also [11]). The maximum at $B = 0$ indicates that excitation of the $^3S_1$ state is favorable if the coherence does not evolve. The two other maxima correspond to those magnetic field strengths at which the associated rate of evolution causes the coherence to return to its original state in about 30 ns. Measurements of the
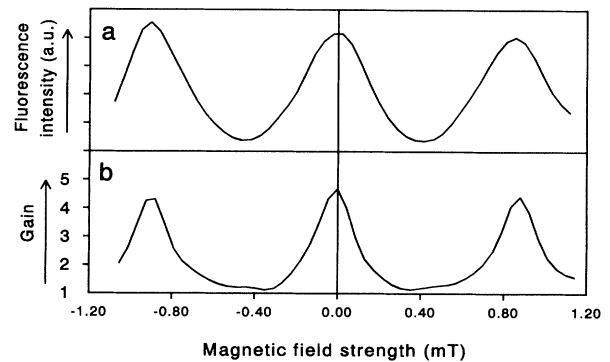
gain as a function of the magnetic field strength reveal the same periodicity. However, the gain drops more sharply than the population of the $^3S_1$ level. This indicates that the observed amplification effect is indeed sensitive to both the population in the upper state and the phase of the superposition in the lower state.

When the delay between pump 1 and pump 2 is eliminated, no dependence of the probability of excitation to the $^3S_1$ state on the magnetic field strength is observed. The coherence in the $^3P_1$ state is lost at higher field strengths because the evolution time of the coherence becomes short compared to the duration of the laser pulse and the precession of the atoms is no longer synchronous (similar to the Hanle effect [14]). For instance at a magnetic field strength of 8 mT the evolution time is 3 ns, considerably shorter than the duration of the pump pulses (6 ns). The measurement shown in Fig. 5 demonstrates this loss of gain at higher field strengths while the population of the upper state remains the same. This indicates that indeed no population inversion is present between the $^3S_1$ and $^3P_1$ levels.
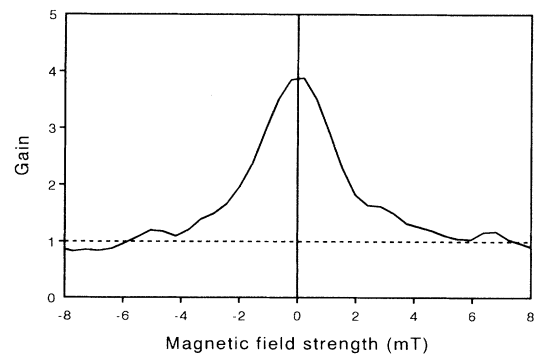

FIG. 5. Gain as a function of the magnetic field strength. The loss of gain for larger field strengths indicates the absence of population inversion.

In conclusion, we would like to stress that, despite the observed asymmetry between absorption and emission, there is no difference between the Einstein B_{12} and B_{21} coefficients. Therefore, in the three-level case, the term "with hidden inversion" may be more appropriate than "without inversion" as noted by Kocharovskaya [1]. In terms of the ϕ_+ and ϕ_- states used to describe the experiment, the population of the lower state is "hidden" in the ϕ_+ state, yielding a population inversion between the upper state and ϕ_-. Because of the energy difference between the coherently excited states, this inversion is only periodically accessible.

Note added.— After submission of this manuscript an experiment, aiming for the same goal but by different means, was reported by Nottelmann *et al.* [15]. In that experiment use is made of a train of picosecond pulses to induce Zeeman coherences.

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