Inversionless Amplification of Picosecond Pulses due to Zeeman Coherence

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We extend the basic idea of inversionless amplification to a picosecond test pulse in the absence of initial optical coherences. The crucial role of population trapping by lower level Zeeman coherence in atomic samarium vapor ($J = 1 \leftrightarrow J' = 0$) is demonstrated experimentally and theoretically in a time separated preparation-test setup.

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Amplification without population inversion has recently attracted considerable attention [1–6]. To achieve the reduction of upper state population in the atomic transition under consideration, some proposals involve the coupling of excited atomic states [2]. Related experiments have already demonstrated electromagnetically induced transparency [7–9]. On the other hand, most of the proposals and theoretical analyses of inversionless amplification deal with the preparation of macroscopic coherence between some of the low-lying atomic states. In this paper, we propose a new scheme for an experimental realization of this general idea and present experimental evidence of amplification without inversion (AWI). Our approach makes use of picosecond pulses (PSP's) and has the virtue of clearly separating in time the creation of coherence between low-lying states, the preparation of population in the upper state, and the amplification of a probe beam. Insofar as we use short pulses the experiment is related to [1,4]. We found it essential, however, to use pulses short compared to all relaxation times of the medium.

It follows from the Bloch vector description of a two-level system that PSP's can be amplified in the absence of inversion if coherence between the upper and the lower level, optical coherence, is initially present in the system [10]. In this paper, however, we discuss AWI, which is entirely due to coherence between low-lying states. Since in our experiment this coherence can be controlled by an external parameter, we can arbitrarily switch off the AWI and even switch to increased absorption, thus demonstrating the crucial role of the lower states' coherence.

In most of the cited publications, the Λ-type system is discussed as a modified laser transition. Beyond that, we demonstrate that AWI can be realized with the more complicated $J = 1 \leftrightarrow J' = 0$ transition. We therefore prepare a Zeeman coherence in a $J = 1 \leftrightarrow J' = 0$ transition of atomic samarium (Sm) in the presence of a magnetic field without having population inversion at any time ($\rho_{ii} < \rho_{44}$, $i = 1, 2, 3$; see Fig. 1). The Zeeman coherence is described by the off-diagonal component ρ_{13} of the density matrix and is created in the lower state of the 570.68-nm line by applying an equidistant train of optically resonant PSP's. This pulse train has an rf period T_p and its linear polarization is perpendicular to the static magnetic field so that the σ^+ and σ^- transitions labeled in Fig. 1 are driven.

After each PSP, ρ_{13} oscillates with the double Larmor frequency (2Ω). To always prepare the atoms in the same way and to achieve the largest values of $|\rho_{13}|$, in this paper we only use discrete values of the magnetic field such that the ρ_{13} oscillation period is rf resonant with the pulse train, $\pi/\Omega = T_p/n$ with $n = 1, 2$, or 4. Our idea can be demonstrated with n being the only free parameter of the experiment.

A similarly polarized test PSP follows the last PSP of the train with a delay time $T_p/4$. Depending on the value of n, the test PSP arrives at different phases of the ρ_{13} oscillation (see Fig. 2). Energy transfer is coupled only to $\mathrm{Re}\rho_{13}$, not to $\mathrm{Im}\rho_{13}$. In the case $n = 1$, the test PSP arrives when $\mathrm{Re}\rho_{13} = 0$ and, because there is no inversion, it is attenuated. If $n = 2$, $\mathrm{Re}\rho_{13}$ is maximum with a positive sign leading to even stronger attenuation. For $n = 4$, inversionless amplification is maximized.

During the time interval $T_p/4$, the optical coherences relax almost completely. Argon buffer gas with a pressure of 90 hPa is used to match the relaxation times. In Sm, a part of the upper state population relaxes as well. For that reason, an orthogonally polarized pump PSP driving the π transition in Fig. 1 reestablishes the up-

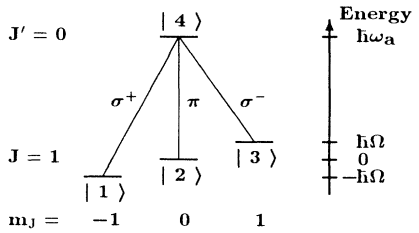

FIG. 1. Energy scheme of the $J = 1 \leftrightarrow J' = 0$ transition in the representation of the unperturbed atom in a magnetic field, B. $\omega_a = |\omega_2 - \omega_4|$ is the atomic frequency; $\Omega = |\omega_1 - \omega_2| = g\mu_B B/\hbar$ is the Larmor frequency. In Sm, $\omega_a = 2\pi c/(570.68\,\mathrm{nm})$ and $g = 1.498$.

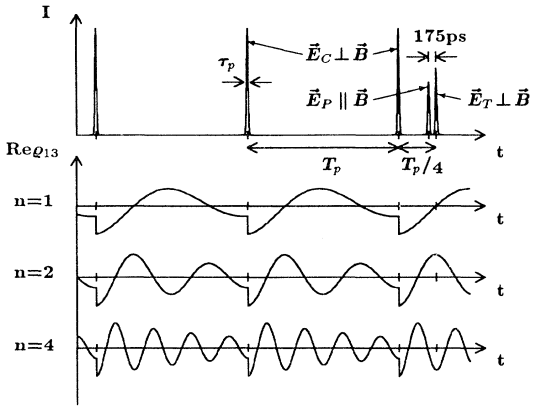

FIG. 2. Time sequence of the last PSP's of the pulse train, $\mathbf{E}_C$, pump PSP, $\mathbf{E}_P$, and test PSP, $\mathbf{E}_T$. The oscillation of Reρ_{13} is displayed for the rf-resonant magnetic fields.

per state population 175 ps before the arrival of the test PSP. Because of orthogonality, the optical coherence thus produced does not influence the test PSP.

For a quantitative description of the experiment, we write the Hamiltonian

$$H = H_0 + H_B + H_C(t) + H_P(t) + H_T(t). \tag{1}$$

H_0 is the Hamiltonian of the unperturbed atom, H_B that of the static magnetic field, and H_C, H_P, and H_T are those of the coherence creating pulse train, the pump PSP, and the test PSP, respectively. Most of the time, the electromagnetic fields are not present and the system is described by $H = H_0 + H_B$. We choose the quantization axis to be parallel to the magnetic field and to the z axis, so $H_0 + H_B$ becomes diagonal with the elements of the energy axis in Fig. 1.

We solve the Liouville equation for the density operator:

$$i\hbar \frac{\partial \rho}{\partial t} = [H, \rho] + R(\rho). \tag{2}$$

The term $R(\rho)$ phenomenologically describes the relaxations to the thermal equilibrium represented by $\rho_{11}^{\mathrm{eq}} = \rho_{22}^{\mathrm{eq}} = \rho_{33}^{\mathrm{eq}} = \frac{1}{3}$, with all other elements zero. The relaxations can conveniently be written for the irreducible matrix components. Here only upper state population ρ_{44}, optical coherences ρ_{i4}, $i = 1, 2, 3$, and alignment components $\rho_0^2 = (\rho_{11} - 2\rho_{22} + \rho_{33})/\sqrt{6}$ and $\rho_2^2 = \rho_{13}$ have to be considered:

$$\frac{\partial \rho_{44}}{\partial t} = -\frac{1}{T_1}\rho_{44}, \quad T_1 = 8.87\,\mathrm{ns}, \tag{3}$$

$$\frac{\partial \rho_{i4}}{\partial t} = -\frac{1}{T_2}\rho_{i4}, \quad T_2 = 1.04\,\mathrm{ns}, \tag{4}$$

$$\frac{\partial \rho_{0,2}^2}{\partial t} = -\frac{1}{\tau_a}\rho_{0,2}^2, \quad \tau_a = 14.9\,\mathrm{ns}, \tag{5}$$

for 90 hPa argon buffer gas pressure and a temperature of 1050 K [11]. During the relaxation phases, the ground-state coherence oscillates:

$$\frac{\partial \rho_{13}}{\partial t} = 2i\Omega\rho_{13} - \frac{\rho_{13}}{\tau_a}. \tag{6}$$

The dipole interaction of all PSP's ($\mathbf{k} = k\mathbf{e}_x$) is described by $H_l = -\boldsymbol{\mu} \cdot \mathbf{E}_l$ with the dipole operator $\boldsymbol{\mu}$ and

$$\mathbf{E}_{C,T} = \left(\frac{\mathcal{E}_{C,T}}{2} e^{i(kx-\omega t)} + \mathrm{c.c.}\right)\mathbf{e}_y, \tag{7}$$

$$\mathbf{E}_P = \left(\frac{\mathcal{E}_P}{2} e^{i(kx-\omega t)} + \mathrm{c.c.}\right)\mathbf{e}_z. \tag{8}$$

$\mathcal{E}_l(t)\mathcal{E}_m(t) \propto \delta_{lm}$, $l, m = C, P, T$, and ω is the center frequency of the pulse spectrum. The envelopes $\mathcal{E}_l$ are slowly varying in time t and propagation direction x, and describe the incoming Gaussian fields. The nonvanishing dipole matrix elements are $(H_{C,T})_{14} = (H_{C,T})_{34} = -i\boldsymbol{\mu} \cdot \mathbf{E}_{C,T}/\sqrt{2}$ and $(H_P)_{24} = -\boldsymbol{\mu} \cdot \mathbf{E}_P$.

The equations of motion for the pulses' interaction with the atoms are obtained by use of the rotating wave approximation with the usual definition $\rho_{i4} = \sigma_{i4} e^{i(kx-\omega t)}$ and noting that for the pulse duration τ_p and the whole Doppler spectrum: $\tau_p \ll 2\pi/\Omega$ and $\tau_p \ll \sqrt{\pi}/\Delta_D < T_2 < T_1 < T_p < \tau_a$ (τ_p is the full width of half maximum of the intensity, Δ_D the half $1/e$ width of the Doppler spectrum). The phase of the reduced dipole matrix element $\mu_r = \sqrt{3}\mu$ is chosen such that $\mu\mathcal{E}_l$ is real:

$$v = -\sqrt{2}\,\mathrm{Re}\,(\sigma_{14} + \sigma_{34}), \tag{9}$$

$$w = \rho_{44} - \rho_{11}/2 - \rho_{33}/2 - \mathrm{Re}\rho_{13}, \tag{10}$$

$$\frac{\partial v}{\partial t} = \frac{\mu\mathcal{E}_{C,T}}{\hbar} w, \quad \frac{\partial w}{\partial t} = -\frac{\mu\mathcal{E}_{C,T}}{\hbar} v, \tag{11}$$

$$\bar{v} = \mathrm{Im}\sigma_{24}, \tag{12}$$

$$\bar{w} = \rho_{44} - \rho_{22}, \tag{13}$$

$$\frac{\partial \bar{v}}{\partial t} = \frac{\mu\mathcal{E}_P}{\hbar} \bar{w}, \quad \frac{\partial \bar{w}}{\partial t} = -\frac{\mu\mathcal{E}_P}{\hbar} \bar{v}. \tag{14}$$

All other matrix elements remain unchanged. In particular, because of the symmetry of pulse and Doppler spectra and because of the relaxation of the optical coherences, no orientation is created, i.e., $\rho_{11} = \rho_{33}$ and $\rho_{12} = \rho_{23} = 0$. Furthermore, all matrix elements except the optical coherences can be treated independently of atomic velocity.

Equations (11) and (14) describe the rotation of Bloch vectors by an angle Θ_l:

$$\Theta_l = \frac{\mu}{\hbar} \int_{\text{one PSP}} dt\, \mathcal{E}_l(t). \tag{15}$$

Via Eqs. (10) and (11) Zeeman coherence is created, i.e., population trapping occurs. In principle, this is the mechanism proposed for AWI of a pulse in [1,4], but, in that treatment, a Λ-type system, homogeneous broadening, and $T_2 \ll \tau_p$ were assumed.

For the pulse train, we calculate the self-consistent solutions of all matrix elements for alternating PSP's and

relaxations. To ensure that no inversion is created, the pulse area of each PSP of the train has to be smaller than 0.76π. The train prepares ρ_{13}, a population ρ_{44}, and also an enhanced population ρ_{22} which relaxes slowly with the time constant τ_a. Therefore, the pulse area of the delayed pump PSP has to be smaller than only 0.46π. We find that macroscopic values of $|\mathrm{Re}\rho_{13}|$ up to 0.14 can be created. Because of the relationship between the actual Sm dipole matrix element $\mu = 4.39 \times 10^{-30}/\sqrt{3}$ C m and the power of our laser system (maximum $P \approx 120$ mW), the three beams have to be focused and in order to achieve the specified pulse areas must be individually adjusted.

Figure 3 shows the transverse preparation of population difference and AWI at the beginning of the vapor, where all of the three Gaussian beams are focused. Here on axis, the largest values of upper state population ρ_{44} in our process are created; it can be seen that no inversion occurs. The test pulse area is chosen to be maximum 0.77π, which is not a small perturbation and destroys $\mathrm{Re}\rho_{13}$ significantly, but assures a sufficient signal-to-noise ratio in our experiment. We define a transverse integrated relative amplification factor as the ratio of energy change by amplification, ΔW_{AWI}, and by attenuation without any preparing field, ΔW_a. In the first layer of the vapor it should be $\Delta W_{\mathrm{AWI}}/\Delta W_a = 0.22$. This amplification is weak because of the transverse structure of the preparation.

Taking into account the focusing, during the propagation through the vapor all pulse areas, Θ_l, are attenuated by both dispersion [10] and diffraction. Although the test pulse area decreases during propagation, the test pulse energy can be amplified. An exact analysis of preparation and test pulse shaping during the propagation described by Maxwell equations will be given in a forthcoming publication.

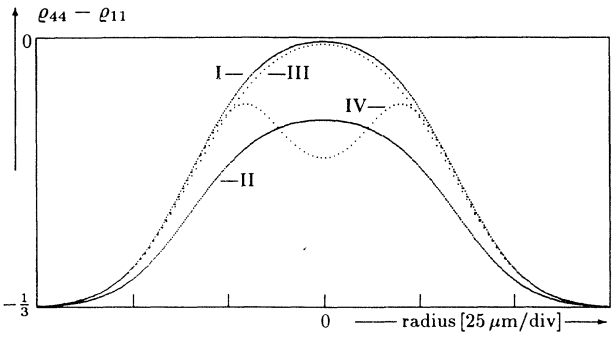

FIG. 3. Transverse profiles of the population difference at the beginning of the vapor at different times: I is exactly after the last pulse of the pulse train, II is before the orthogonal pump pulse, III is before, and IV after, the test pulse. Not even in the center of the beam does inversion occur: $\rho_{11} > \rho_{44}$ ($\rho_{22} > \rho_{11}$). Beam waists are 42, 45, and 25 μm and maximum pulse areas are 0.76π, 0.46π, and 0.77π for pulse train, pump, and test PSP's, respectively.

AWI can be realized if the pulse areas are only slightly decreased. Because of our laser equipment, focusing cannot be avoided and therefore only small particle densities are allowed. In this case, the propagation is determined by diffraction and the beams remain approximately Gaussian in the vapor. Transverse and longitudinal integration (see experimental parameters below) then lead to a relative amplification factor of $\Delta W_{\mathrm{AWI}}/\Delta W_a \approx 0.16$.

In our experiment (see Fig. 4), we use a pulse train from a synchronously pumped dye laser resonantly tuned to the Sm line with a pulse length of $\tau_p = 30$ ps measured by optical autocorrelation. This corresponds to a spectral width of ≈ 15 GHz which is small enough to assure that the PSP's do not interact with the adjacent Sm lines 45 and 50 GHz apart. The pulse separation is $T_p = 13.214$ ns. With a period of $64T_p$, a single PSP is separated from the pulse train using a laser triggered high voltage pulse of length $\approx T_p$ applied to an electro-optic modulator. This single PSP is divided into the test PSP and an orthogonally polarized pump PSP and all PSP's are made collinear again.

The focusing is achieved by the same lens for pulse train, pump, and test PSP's with resulting Gaussian beam waists of 42, 45, and 25 μm, respectively, all exactly at the beginning of the vapor. In the chosen geometry, the pumping beams overlap the test beam for the whole interaction region of 5 mm. It is assured by optical autocorrelation that pump and test PSP remain separated in time during the propagation through the vapor.

The (unsaturated) low density Sm vapor is confined between two apertures of tantalum 5 mm apart which are placed in an electrically heated quartz tube (1050 K). For reasons of purity, the cell is evacuated to $< 10^{-5}$ hPa and heated before introducing the Sm. The whole construction including the heating is antimagnetic. With two coils, the magnetic field of $n \times (1.805\,\mathrm{mT})$, $n = 1, 2, 4$, transverse to the polarization of the test field is applied. After passing through the cell, the pump PSP is eliminated by a polarizer and the test pulse is detected with a rapid photodiode and a sampling oscilloscope.

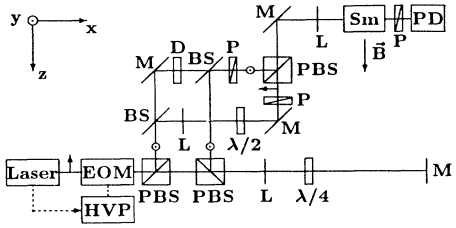

FIG. 4. Schematic experimental setup. EOM: electro-optic modulator; HVP: high voltage pulse generator; PBS: polarizing beam splitter; M: mirror; L: lens; BS: beam splitter; D: delay; PD: photodetector; arrows mark the direction of the beam polarization.

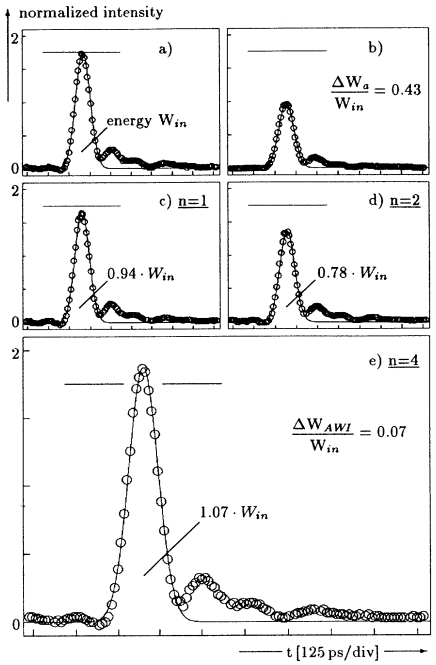

FIG. 5. Experimental energy attenuation and inversionless amplification displayed by time resolved test pulse intensities as described in the text. The relative gain of $\Delta W_{\mathrm{AWI}}/\Delta W_a \approx 0.16$ is in good agreement with the theory.

We observe the time resolved intensity of the transverse integrated test pulse convolved with the electronic response function of photodiode and sampling oscilloscope (full width at half maximum of 93.9 ps). Figure 5 (b) displays the absorption of the resonant test pulse without preparing fields. In Fig. 5(a), the frequency has been changed to observe the freely propagating test PSP. In Figs. 5 (c), 5(d), and 5(e) the test PSP is resonant and all fields are present, but the magnetic field is 1.805 mT, 3.61 mT, and 7.22 mT, respectively. Each curve is an average of 16 single curves, each of which has been sampled over 5500 test pulses. In this way, all discrete fluctuations of the pump laser have been eliminated.

For the sake of completeness, it should be mentioned that an experiment on amplification of a test field in sodium vapor has been recently reported using the theory of inversionless amplification [12]. This interpretation is problematic because of the unfavorable transient dynamics in the complex level structure of sodium and because of the well-known fine-structure state-changing collisions [13].

In conclusion, we demonstrate for Zeeman coherences the macroscopic effect of population trapping by lower level coherence on the amplification of a picosecond pulse, an effect that does not exist in the conventional laser process. In the representation of the unperturbed atoms in the static magnetic field, this amplification is inversionless with respect to all participating Zeeman states which is a rather restrictive definition. Of course, with another choice of representation, for example, rotation of the quantization axis, the process is not inversionless. However, the important result is that sublevel dynamics may play a determining role in new amplification schemes. Presently, we are extending our experiment by adding a second atomic transition to demonstrate frequency up-conversion.

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