Coherent population trapping in an optically dense medium

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The phenomenon of coherent population trapping in an optically dense medium is analyzed through a self-consistent solution of the system of Maxwell-Bloch equations. When this trapping occurs, there is a transparency window for a laser beam. The width of this window decreases with increasing optical thickness. Possible practical applications of this phenomenon are discussed.

1. INTRODUCTION

The phenomenon of coherent population trapping in three-level media which are interacting with two electromagnetic waves has recently been the subject of active theoretical[1–6] and experimental[7,8] research. The primary reason for this interest is that this trapping is a fundamental physical phenomena which occurs in diverse media and over broad ranges of the frequencies of the exciting fields. It is manifested during the excitation of a medium by either optical[1–8] or rf fields.[5] It is seen during both cw[4] and pulsed excitation.[6] It has also attracted interest because of a variety of applications: for frequency stabilization,[4,8] for spectroscopy free of not only Doppler broadening but also homogeneous broadening of spectral lines,[6] in optical bistability systems,[9,10] and elsewhere.[11,12]

The concept of a three-level medium is itself quite general, since it means that each element of the medium has a three-level excitation scheme. In this sense we could speak in terms of a three-level atomic medium, a medium consisting of three-level nuclei, color centers, etc. For all such (qualitatively different) three-level media one can observe coherent population trapping; the conditions for the occurrence of this trapping depend only on a relation among the parameters of the exciting fields.

Coherent population trapping is known to occur most obviously in the interaction of two-frequency laser light with a three-level medium having a Λ-shaped level configuration (a "Λ medium").[13] Coherent population trapping can be summarized as follows: Under the condition for a two-photon resonance for optical fields propagating in the same direction, i.e., under the condition

$$(\omega_1-\omega_{31})-(\omega_2-\omega_{32})\equiv\Omega_1-\Omega_2=0, \qquad (1.1)$$

where $\omega_{1,2}$ are the field frequencies, and ω_{31} and ω_{32} are the frequencies of the 3–1 and 3–2 transitions, coherent superposition of the states $|1\rangle$ and $|2\rangle$ arises between the low-lying levels in the Λ system, and the system is not excited to state $|3\rangle$ even in resonant fields. In other words, the steady-state population of the upper level is approximately zero, while the probability for finding the system in each of the lower levels is approximately 1/2, if the fields are equal in intensity. This assertion is the physical essence of coherent population trapping.

As a result, a medium in which coherent population trapping is occurring can neither absorb nor emit light, and a narrow gap (a "dark line") appears in its fluorescence spectrum. The width of this dark line can be much smaller than the homogeneous linewidth corresponding to the 3–1 and 3–2 transitions.[7]

In this paper we analyze the propagation of cw laser light through optically dense three-level media under conditions corresponding to coherent population trapping. A problem of this sort was first taken up by Kocharovskaya and Khanin,[6] for the case of pulsed excitation with a pulse width τ_p whose reciprocal is much larger than the frequency separation ω_{21} of the low-lying levels: $\tau_p^{-1}\gg\omega_{21}$. In this case the light interacts with the two resonant transitions simultaneously in the Λ system, and bleaching of the medium occurs if the pulse repetition frequency T^{-1} is a multiple of ω_{21}. Because of this bleaching, we can say that coherent bleaching of the Λ medium occurs when ultrashort light pulses are applied to it. The physical reason for this bleaching is the imposition of low-frequency coherence between the low-lying levels during the pulse.

We would like to stress that this bleaching of a medium is qualitatively different from both the bleaching which results from optical pumping in a three-level system and the bleaching caused by light so intense that it saturates a resonant transition in a two-level system. The primary reason for these qualitative differences is that in the case of coherent population trapping there are no reradiated photons in the optically dense medium.

Similar bleaching should occur in the case of cw two-frequency laser light if the widths of the frequency spectra of the exciting waves, $\Delta\omega_{1,2}$, are smaller than the distance between levels $|1\rangle$ and $|2\rangle$ ($\Delta\omega_{1,2}\ll\omega_{21}$). In this case each resonant transition in the Λ system is excited by only a single wave. If the conditions for coherent population trapping are satisfied by virtue of the appearance of a coherent superposition of states $|1\rangle$ and $|2\rangle$, the three-level medium will not be excited to state $|3\rangle$, so bleaching of the medium will again occur.

In the present paper we analyze the propagation of cw laser light through an optically dense Λ medium. We show that during coherent population trapping bleaching occurs in the medium, and the propagation of the light in the medium is qualitatively different from that described by the Bougher-Lambert law. We find the conditions which correspond to the lowest laser light intensity at which the medium absorbs only weakly. We also find that as the frequencies ω_1 and ω_2 are scanned there is a transparency window due to the coherent population trapping. Outside this window, the light is attenuated exponentially in the medium. We show that the width of this transparency window decreases with

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increasing optical thickness and can reach a value on the order of the homogeneous linewidth of the 2–1 transition. We discuss some possible practical applications of the effects found here.

The paper is organized in the following way. In Sec. 2 we write self-cosistent Maxwell-Bloch equations for an optically dense medium. They describe both the dynamics of the excitation of the medium and the propagation of the laser light in it. These equations are used in Sec. 3 to derive analytic solutions for several cases of practical importance. On the basis of these solutions, one can draw conclusions regarding the nature of the light propagation in the medium. In Sec. 4 we discuss some possible pratical applications, and we report some numerical estimates of the effects corresponding to realistic experimental situations.

2. BASIC EQUATIONS

To solve our problem, we specify a light field consisting of two traveling plane light waves with amplitudes E_μ, frequencies ω_μ, wave vectors $\mathbf{k}_\mu$, and unit polarization vectors $\mathbf{e}_\mu$:

$$\mathbf{E}(z,t)=\sum_{\mu=1,2} E_\mu \mathbf{e}_\mu \exp[i(k_\mu z-\omega_\mu t)]. \quad (2.1)$$

We also assume that the light waves propagate parallel to the z axis of the Cartesian coordinate system. We stress that for light waves which are propagating in the same direction the coherent population trapping occurs regardless of the velocity of an atom [condition (1.1)], so we will be ignoring the thermal motion of the atoms in the Λ medium. If this motion is taken into account in the final results, the homogeneous optical absorption line is replaced by an inhomogeneous Doppler line. The properties of the dark line, on the other hand, are not affected by the thermal motion of the atoms in this case.

Let us examine the interaction of the field (2.1) with a three-level Λ medium. We assume that a dipole transition between levels 1 and 2 is forbidden and that the partial probabilities for decay from optical level 3 to levels 1 and 2 are equal. We furthermore assume that each of the light waves acts on "its own" resonant transition (ω_1 is in resonance with the 1–3 transition, and ω_2 with the 2–3 transition). The quantum-mechanical kinetic equations describing the dynamics of the density matrix of the active atoms under these conditions are given in Ref. 14. For the problem at hand, however, that system of equations is not complete. It must be supplemented with Maxwell's wave equations. We take the customary approach[15,16] of writing simplified wave equations for the slowly varying amplitudes E_μ of waves which are propagating along the z axis ($\mu=1,2$):

$$\frac{\partial E_\mu}{\partial z}+\frac{1}{c}\frac{\partial E_\mu}{\partial t}=i\frac{2\pi N\omega_\mu}{c}d_{\mu3}F_{3\mu}, \quad (2.2)$$

where $d_{\mu3}$ is the dipole matrix element for the μ–3 transition, N is the density of active atoms, and $F_{3\mu}=\rho_{3\mu}\exp[i(\omega_\mu t-k_\mu z)]$ is the optical coherent.

We are interested in a steady-state solution of this system of equations or, more specifically, in the propagation of laser light in such a medium. For convenience we introduce the dimensionless optical length τ and the dimensionless field intensity $J_\mu(\tau)$ ($\omega\equiv\omega_1\approx\omega_2$, $d\equiv d_{31}\approx d_{32}$):

$$\tau=\frac{4\pi N\omega d^2}{c\hbar A}z,\quad J_\mu(\tau)=\frac{|E_\mu(\tau)|^2}{|E_\mu(0)|^2}=\frac{I_\mu(\tau)}{I_\mu(0)}. \quad (2.3)$$

We also introduce the quantities α_μ, β, δ_μ, and W_μ, which are defined by

$$\alpha_\mu=\frac{|E_\mu(0)|^2}{|E_n|^2},\quad \beta=\frac{|E_2(0)|^2}{|E_1(0)|^2},\quad \delta_\mu=\frac{1}{2}+i\frac{\Omega_\mu}{A},$$
$$W_\mu=\frac{d^2}{\hbar^2}|E_\mu(0)|^2 A/(\Omega_\mu^2+A^2/4), \quad (2.4)$$

where A is the rate of the spontaneous decay of state $|3\rangle$, $E_n=\hbar A/2d$ is the field amplitude which saturates the optical transition, W_μ represents the rate of the coherent optical pumping by field μ, and $I_\mu(0)$ is the intensity of the light for field μ at the entrance to the medium.

Using the rotating-wave approximation, and eliminating the off-diagonal matrix elements (coherences), we find the following system of equations after some lengthy but straightforward manipulations:

$$\frac{d}{d\tau}J_1=-\alpha_1^{-1}\left[\rho_{33}+\frac{\gamma}{A}(\rho_{22}-\rho_{11})\right],\quad J_1(0)=1,$$
$$\frac{d}{d\tau}J_2=-\alpha_2^{-1}\left[\rho_{33}-\frac{\gamma}{A}(\rho_{22}-\rho_{11})\right],\quad J_2(0)=1,$$
$$A\rho_{33}+\gamma(\rho_{22}-\rho_{11})-W_1J_1\Big\{2(\rho_{11}-\rho_{33})-\alpha_2J_2$$
$$\times\operatorname{Re}\left[\frac{A}{\Gamma+i\Delta}\left(\frac{\delta_1^*}{\delta_1}(\rho_{11}-\rho_{33})+\frac{\delta_1^*}{\delta_2^*}(\rho_{22}-\rho_{33})\right)\right]\Big\}=0, \quad (2.5)$$
$$A\rho_{33}-\gamma(\rho_{22}-\rho_{11})-W_2J_2\Big\{2(\rho_{22}-\rho_{33})-\alpha_1J_1$$
$$\times\operatorname{Re}\left[\frac{A}{\Gamma-i\Delta}\left(\frac{\delta_2^*}{\delta_2}(\rho_{22}-\rho_{33})+\frac{\delta_2^*}{\delta_1^*}(\rho_{11}-\rho_{33})\right)\right]\Big\}=0,$$
$$\rho_{11}+\rho_{22}+\rho_{33}=1,$$

where

$$\Gamma\pm i\Delta=\Gamma+\beta W_1J_2/2+W_2J_1/2\beta$$
$$\pm i[\Omega_1(1-\beta W_1J_2/A)-\Omega_2(1-W_2J_1/A\beta)],$$

ρ_{ii} is the population of state $|i\rangle$ and γ and Γ are the rates of the longitudinal and transverse relaxation between levels 1 and 2.

Equation (2.5) are a self-consistent system of nonlinear equations which can be solved in the general case only by numerical methods. However, there are several particular cases of practical interest in which it is possible to derive some fairly simple analytic solutions by making use of the symmetry properties of Eqs. (2.5).

3. PROPAGATION OF LASER LIGHT; THE TRANSPARENCY WINDOW

Let us examine some analytic solutions of system (2.5).

A. Case of a two-photon resonance. Let us examine the conditions which are most favorable for the manifest of this coherent population trapping. We know that these conditions are that the deviations of the exciting fields from the resonant frequencies be equal and that the amplitudes of these fields also be equal:

$$\Omega_1=\Omega_2=\Omega,\quad E_1(\tau=0)=E_2(\tau=0). \quad (3.1)$$

From (2.4) under condition (3.1) we find

$$\alpha_1=\alpha_2,\ \beta=1,\ \delta_1=\delta_2,\ W_1=W_2=W. \tag{3.2}$$

Using (3.2), and introducing the resultant intensity $J=(J_1+J_2)/2$, we find a nonlinear differential equation which describes the propagation of laser light in the case of a two-photon resonance:

$$\frac{d}{d\tau}J=-L(\Omega)J\left(1-\frac{WJ}{\Gamma+WJ}\right),\quad J(0)=1, \tag{3.3}$$

where $L(\Omega)=A^2/4(A^2/4+\Omega^2)^{-1}$ is Lorentzian.

Equation (3.3) has the solution

$$\frac{W}{\Gamma}(1-J)-\ln J=L(\Omega)\tau, \tag{3.4}$$

from which we see that the propagation law for the light depends on the intensity of the laser light as it enters the medium [the initial intensity $I(0)=I(\tau=0)$], on the optical length τ, and on the frequency deviation Ω. A characteristic parameter of this problem, as in Ref. 6, is the coherent intensity

$$I_c=\frac{\Gamma}{A}I_s,\ \text{where}\ I_s=\frac{c}{8\pi}\hbar^2A^2/4d^2 \tag{3.5}$$

is the intensity which saturates the optical transition. We know, for example, that optical transitions of alkali metal atoms can usually be saturated by a fairly modest intensity $I_s\approx0.1\ \text{W/cm}^2$. In this case the coherent intensity is $I_c\approx10\ \mu\text{W/cm}^2$ for the values $\Gamma\approx10^3\ \text{s}^{-1}$ and $A=10^7\ \text{s}^{-1}$.

At an initial $I(0)\gg I_c$ ($W\gg\Gamma$) it follows from (3.4) that the propagation law for the light in the medium is linear:

$$J(\tau)=1-L(\Omega)\frac{\Gamma}{W}\tau. \tag{3.6}$$

If, however, an increase in τ is accompanied by a decrease in the intensity $I(\tau)$ to the extent that the relation $I(\tau)\ll I_c$ becomes satisfied, the propagation law becomes exponential (the Bougher-Lambert law), while for $I(0)\ll I_c$ the light attenuation is exponential for arbitrary τ (Fig. 1a).

The physical meaning of the coherent intensity I_c is that for $I\gtrsim I_c$ coherent population trapping, in which the atoms or molecules are trapped in a superposition state between the low-lying levels, arises in the medium. In this case the coupling with optical level $|3\rangle$ is broken, and the medium absorbs the optical radiation only weakly.

Note that coherent population trapping would be possible only under fairly restrictive conditions on $\Delta\omega_{1,2}$ (the spectral widths of the exciting waves). According to Ref. 4, this trapping occurs for $\Delta\omega_{1,2}\lesssim W$; in the opposite case, the coherence ρ_{12} is disrupted, and the coherent population trapping disappears.

Figure 1b shows a spectral plot of $J(\tau,\Omega)$ versus the frequency difference Ω for various values of τ. We see that the medium becomes progressively more transparent to the light as $|\Omega|$ increases. The reason is that a nonresonant bleaching mechanism comes into play along with the bleaching due to the coherent population trapping.

B. Case of mirror-image frequency differences. In this case we have

$$\Omega_1=-\Omega_2=\Omega,\ E_1(\tau=0)=E_2(\tau=0)=E(0). \tag{3.7}$$

In place of (3.2) we have

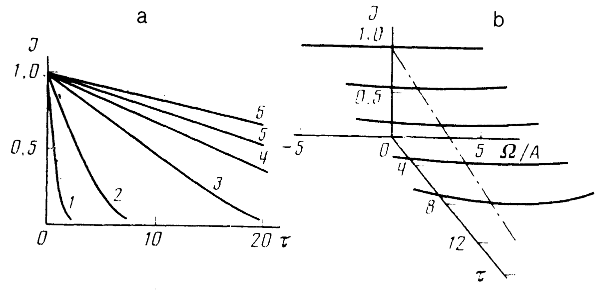

FIG. 1. Propagation of laser light under condition of coherent population trapping ($\Omega_1=\Omega_2\equiv\Omega$; $A=3.8\cdot10^7\ \text{s}^{-1}$; $\gamma=\Gamma=100\ \text{S}^{-1}$). a: $\Omega=0$. 1—$V=10^4\ \text{s}^{-1}$; 2—$V=10^5\ \text{s}^{-1}$; 3—$V=2B\ 10^5\ \text{s}^{-1}$; 4—$V=3B\ 10^5\ \text{s}^{-1}$; 5—$V=4B\ 10^5\ \text{s}^{-1}$; 6—$V=5B\ 10^5\ \text{s}^{-1}$. b: Spectral plot of the signal, for $V=3B\ 10^5\ \text{s}^{-1}$.

$$\alpha_1=\alpha_2,\ \beta=1,\ \delta_1=\delta_2^*,\ W_1=W_2=W.$$

For the resultant intensity J we then find the differential equation

$$\frac{d}{d\tau}J=-L(\Omega)J\left[1-\frac{WJ(\Gamma+WJ)}{(WJ+\Gamma)^2+4\Omega^2}\right],\quad J(0)=1, \tag{3.8}$$

whose solution is

$$\frac{W}{\Gamma}(1-J)-\ln J-\frac{4\Omega^2}{\Gamma^2}\ln\frac{\Gamma^2+4\Omega^2+\Gamma W}{\Gamma^2+4\Omega^2+\Gamma WJ}=L(\Omega)\tau. \tag{3.9}$$

In deriving (3.8) we used the condition $W\ll A$, which implies a limit on the intensity of the laser light as it enters the medium: $I(0)\ll I_n$.

At $\Omega=0$, the two cases are the same, and (3.9) becomes (3.4). However, there is a substantial distinctive feature in the $J(\tau,\Omega)$ spectrum in this version of the problem (Fig. 2): At frequency differences $\Gamma_0\ll|\Omega|\ll A$ the decay of the light with increasing τ is exponential, while for $|\Omega|\lesssim\Gamma_0$ it is linear. As a result, as the field frequency is scanned a transparency window arises in the medium due to coherent population trapping. Interestingly, the width of this window, Γ_0, depends on the optical length, and it decreases with increasing τ (Fig. 2b). The analytic expression found for Γ_0 through an analysis of the numerical calculations on the basis of (3.9) is

$$\Gamma_0\approx\frac{\Gamma}{2}+\frac{2V^2}{A}J(\tau), \tag{3.10}$$

where $V=d|E(0)|/\hbar$ is the Rabi frequency for the field $E(0)$. The large intensity in the transparency window, $I(\tau)$,

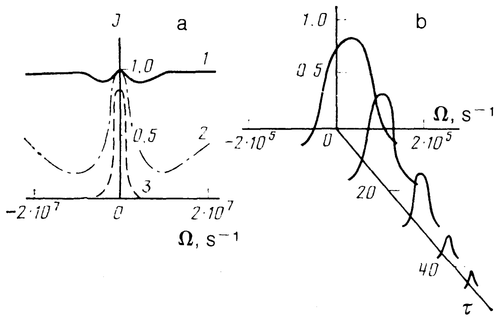

FIG. 2. A transparency window. $\Omega_1=-\Omega_2\equiv\Omega$, $V=3\cdot10^5\ \text{s}^{-1}$. a: $|\Omega|\lesssim A$. 1—$\tau=0.1$; 2—$\tau=1$; 3—$\tau=4$. b: Central part of the line. The values of the other parameters are the same as in Fig. 1.

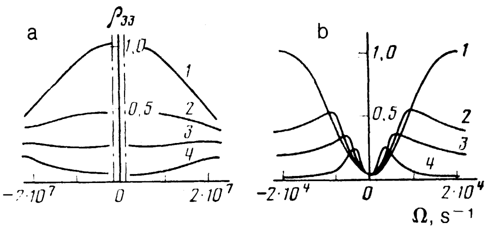

FIG. 3. Spectrum of the resonant fluorescence. The parameter values are the same as in Fig. 2 a: $|\Omega| \lesssim A$. b: Central part of the signal (the "dark line"). 1—$\tau = 0.1$; 2—$\tau = 0.5$; 3—$\tau = 1.4$; 4—$\tau = 2$.

falls off linearly. However, the decay becomes exponential at values of τ for which the intensity satisfies $I(\tau) \ll I_c$. At large frequency differences, $|\Omega| \gtrsim A$, the medium absorbs only weakly (Fig. 2a), by a nonresonant mechanism.

Finally, we find the spectrum of the upper level, ρ_{33}, which determines the dark line in the fluorescence spectrum during coherent population trapping for this case.

Using (3.8), we find from (2.5)

$$\rho_{33} = \frac{W}{A} J\left[1 - WJ\frac{WJ+\Gamma}{(WJ+\Gamma)^2+4\Omega^2}\right], \tag{3.11}$$

where $J(\tau)$ is given by (3.9). Figure 3 shows curves of $\rho_{33}(\tau, \Omega)$. At the centers of these curves there are clearly defined dips (this is the dark line), with a width which is given quite accurately by (3.10).

4. MODULATION OF OPTICAL RADIATION

The analysis above the propagation of light in a Λ medium provides a solid basis for the practical use of coherent trapping in optically dense media. For definiteness, we take as the Λ medium a gas-filled cell holding the vapor of an alkali metal, e.g., rubidium. The Λ arrangement which we need can be formed by two sublevels of the hyperfine structure of the $5S$ state and the upper $5P_{1/2}$ level. As was shown in Sec. 3, under conditions of coherent population trapping, even in the case of fairly intense fields, $I(0) > I_c$, there is a narrow transparency window in the Λ medium in which the light decays linearly in the medium; i.e., bleaching of the medium occurs. Outside this transparency window, the light is rapidly (exponentially) attenuated in the medium. These circumstances can be utilized to convert frequency modulation of a light beam into amplitude modulation. Let us consider the case in which the frequency of one of the light waves, e.g., that of the wave with the frequency ω_1, is modulated by square pulses of length t_0 and repetition frequency T^{-1} in such a manner that we have

$$\omega_1^m = \omega_1 + \Delta \quad \text{for} \quad t_1 < t < t_2, \tag{4.1}$$
$$\omega_1^m = \omega_1 \quad \text{for} \quad t_2 < t < t_3.$$

We choose the optical frequencies ω_1 and ω_2 to satisfy condition (1.1). In this case t_0 and T must be larger than A^{-1}:

$$t_0,\ T > A^{-1}, \tag{4.2}$$

where A^{-1} is the time scale of the spontaneous decay of the $5P_{1/2}$ level ($A \approx 4\cdot 10^7$ s^{-1}), and the modulation depth Δ must exceed the width of the transparency window, Γ_0:

$$\Delta > \Gamma_0. \tag{4.3}$$

For example, if we use a laser beam intensity $I \approx 0.1$ mW/cm^2 and the $5S$–$5P$ transition of Rb atoms excited from two low-lying states of the hyperfine structure, the width of the windows is $\Gamma_0 \approx 10^4$ s^{-1}. The reason for the restriction (4.2) is that the coherent population trapping is established and destroyed in a time of order A^{-1} in a Λ system.[2] For the conditions specified above, the medium is bleached at $t_2 < t < t_3$, while at $t_1 < t < t_2$ we observe a pronounced absorption of light by the medium. As a result, the frequency modulation of the light wave with a carrier of optical frequency ω_1 controls the transmission of the light by the gas-filled cell, so the frequency modulation is converted into an amplitude modulation. The extent of the conversion of one type of modulation into the other depends in this case only on the magnitude of the optical absorption in the medium. It can reach 100% by virtue of a simple increase in the optical density of the medium. Since the optical length is given by $\tau \approx N\lambda^2 z$ [see (2.3)], the attenuation of the light by a factor of e outside the transparency window occurs over a distance $l \approx (N\lambda^2)^{-1}$. In the case of a cell holding a vapor of Rb atoms whose $5S$–$5P$ transition ($\lambda \approx 10^{-5}$ cm) is to be used, and for an atomic density $N = 10^{10}$ cm^{-3}, we find an absorption length $l \approx 1$ cm. Consequently, a gas-filled cell only 1 cm in size would have an optical thickness sufficient for a complete conversion of one type of modulation into another.

Converting frequency modulation into amplitude modulation through the use of coherent population trapping requires optical fields with very stable frequencies ω_1 and ω_2. The degree of this stabilization, i.e., the spectral widths $\Delta\omega_{1,2}$ of these fields, must be smaller than the rate of the coherent optical pumping:

$$\Delta\omega_{1,2} < W. \tag{4.4}$$

Under the condition $\Delta\omega_{1,2} \gtrsim W$, there is a complete decay of the coherent superposition of states of the low-lying levels, and the atom is excited into the upper state.

We would like to stress two other possible applications of a converter of this sort. First, a cell filled with three-level atoms could work as part of a signal coincidence circuit, with both of the optical fields (ω_1 and ω_2) being modulated. As before, the modulation is by rf pulses, and in general it is random. The cell would then transmit light only in the case in which the frequency differences $\Omega_{1,2}$ satisfying condition (1.1) are temporally coincident. The second possibility involves the simultaneous operation of a converter of this sort with a large spatially separate signals of modulated light beams. Since light beams can be focused to a transverse dimension $r_0 \approx \lambda$, up to $n \sim S/\lambda^2 \sim 10^7$ data channels could be packed on the surface of a converter with an area $S \approx 1$ mm^2. Because of its simplicity and high conversion quality, such a modulator could be developed into readout devices for optical-fiber communications and optoelectronics.

Finally, there is the possibility of using magnetic sublevels of one of the hyperfine states as the low-lying levels of the Λ system. In this case, however, it would be necessary to use light of definite polarization, and the choice of this polarization would depend on the quantum numbers of the magnetic sublevels. Coherent population trapping would occur even in a zero magnetic field, and even in the case of the level crossing, with $\omega_{21} \to 0$. Here there is a unique opportunity to observe a coherent population trapping excited by a single laser beam, provided that this beam acts on both of the degenerate components of the low-lying level.

CONCLUSION

In summary, this study has established that substantial bleaching of a medium occurs under conditions of coherent population trapping in the case of continuous excitation of an optically dense Λ medium by light at two frequencies. The light propagation law in the medium is linear and differs from the familiar Bougher-Lambert exponential attenuation. We have found the minimum light intensity at which bleaching of the medium is possible. We have detected a transparency window and determined its properties. We have discussed a new practical application of coherent population trapping, for optical modulation, and we have estimated the characteristics of a corresponding modulator (the extent of the conversion, the speed, etc.).

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