Coherent population trapping in nonmonochromatic laser light

I. E. Mazets and B. G. Matisov

Leningrad State Technical University

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The coherent trapping of population which occurs in a medium upon the application of a single laser beam with a finite spectral width is analyzed. An analytic solution is derived for a nonlinear equation describing the transport of optical radiation at the frequencies of spectral lines for arbitrary spectra of the light and the absorption. The conditions under which coherent population trapping is observed are determined. During coherent population trapping, the decay of the overall intensity of the laser light in the medium is linear and independent of the spectra of the incident light and the absorption. The propagation of each spectral component is determined by the set of all other components (there is a "frequency memory").

1. INTRODUCTION

The excitation of multilevel quantum systems by coherent fields has revealed some new effects of both conceptual and practical importance. One is coherent population trapping (CPT), which has recently become the subject of very active research. It is the topic of a large number of theoretical and experimental papers (see, for example, the bibliographies in Refs. 1 and 2).

To a large extent, this interest stems from the very wide range of possible applications of this effect: ultrahigh-resolution spectroscopy, free of not only Doppler broadening but also of homogeneous broadening of spectral lines, in both the optical[3] and rf[4] frequency ranges; frequency stabilization;[5] ultradeep cooling of atoms,[6] in which temperatures on the order of 10^{-6} K can be reached with the help of CPT; passive laser mode locking;[7] the conversion of a frequency modulation of optical radiation into an amplitude modulation;[8] the development of atomic interferometers for measuring the phase and amplitude of a radiation field;[2] the development of lasers which work without a population inversion;[9-13] and others.

Coherent population trapping can occur in atomic, molecular, or any other quantum systems which are interacting with fields, with either discrete or continuous spectra of states.[14] It is seen most vividly in the interaction of two-frequency laser light with a three-level medium having a Λ configuration of levels (a "Λ system"). Usually, two of the levels would be sublevels (1 and 2) of the ground state or low-lying metastable levels, while the upper level, 3, would be an optical level. Both transitions from the upper level to the lower levels are allowed. The effect can be summarized by saying that when the condition for a two-photon resonance is satisfied for optical fields propagating in the same direction, i.e., under the condition

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

where $\omega_{1,2}$ are the frequencies of the fields, and ω_{31} and ω_{32} are the frequencies of the 3–1 and 3–2 transitions, a coherent superposition of states $|1\rangle$ and $|2\rangle$ between the lower levels arises in the medium. The atoms "get stuck" (are trapped) in this superposition of states, and they are not excited into the $|3\rangle$ state, even if resonant fields are present. As a result, the medium can neither absorb nor emit light during CPT. A narrow coherent dip (a "black line") appears near the two-photon resonance. This dip has been observed experimentally in several places.[15-17]

The most convenient way, and at the same time the most valid way, to describe CPT is by a method based on a quantum kinetic equation for the density matrix of the atoms. In the case of a Λ system, the CPT can be associated with the excitation of a low-frequency coherence ρ_{12}—an off-diagonal element of the density matrix—under condition (1.1). Two-photon resonance (1.1) is a necessary condition for the observation of CPT, so the nonmonochromatic nature of real laser light sources, which implies nonresonant components of the spectrum, should lead to a disruption of the coherence ρ_{12} and thus a CPT. This problem, however, is not so obvious.

An effect of fluctuations of laser fields on CPT has been studied in several papers[18-20] for the case of an optically thin medium. The basic results of this research show that when two uncorrelated laser fields with spectral widths Δ_1 and Δ_2 interact with a Λ system the CPT is disrupted, and the coherent narrow dip is washed out of the fluorescence spectrum. If the fields are instead mutually correlated, with $\Delta_{12}\neq 0$ (Δ_{12} is the cross spectral width), the CPT reappears to the extent to which the relaxation rate Γ' of the coherence ρ_{12} is restored (reduced):

$$\Gamma'=\Gamma+\Delta_1+\Delta_2-2\Delta_{12},\qquad |\Delta_{12}|\leqslant(\Delta_1\Delta_2)^{1/2}, \tag{1.2}$$

where Γ is the "dark" relaxation rate of ρ_{12}. This coherence can be achieved quite easily in an experiment, through the excitation of two fields from an original fluctuating field by acoustooptic modulation or frequency multiplication. At a critical value of the mutual correlation ($\Delta_{12}=(\Delta_1\Delta_2)^{1/2}$, $\Delta_1=\Delta_2$), the CPT is completely restored in nonmonochromatic fields. These conclusions are supported by experiments[21] which used an acoustooptic modulation method.

Coherent population trapping gives rise to important features in the propagation of laser light in optically dense media.[7,8] So far, only two cases have been studied: the interaction of two-frequency cw laser light with an optically dense medium[8] and the case in which the light is a periodic

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train of ultrashort pulses with a width which spans the two lower levels of the Λ system.[7] It was shown in those studies that the decay of the laser light with increasing optical path length is linear, in contrast with the classical Bouguer-Lambert law, which is exponential. In the latter case, a bleaching of the medium occurs if the frequency of the splitting of the lower levels is a multiple of the pulse repetition frequency. On the other hand, the propagation of nonmonochromatic cw laser light with a spectrum spanning the two lower levels remains an open problem, although it is important to numerous physical applications. For example, there has recently been active research on systems in which the frequency distance between the lower levels is extremely small[11,12] (these levels might be, for example, Zeeman sublevels of the ground state), or in which the atomic levels are degenerate in angular-momentum projection.[22,23] Under these conditions the actual spectrum of the laser light spans the sublevels, and a CPT state may be excited in the quantum system.

We thus see the need for a detailed theoretical study of CPT in an optically dense medium in the case of a nonmonochromatic laser field.

In this paper we analyze the transport of laser light at the frequencies of spectral lines in an optically nonlinear CPT medium. We derive an analytic solution of the transport equation for the light for arbitrary spectra of the light and the absorption. We find that the propagation of each spectral component is determined by the set of all others; i.e., there is a "frequency memory." We show that the medium is bleached during CPT. The change in the overall intensity is linear and independent of the incident and absorbing spectra. At large optical thicknesses, however, at which the CPT fades away, the wings of the incident line strongly influence the decay of the overall light intensity.

2. EQUATION FOR THE DENSITY MATRIX

As we mentioned above, for laser sources with a real output spectrum, CPT is seen most vividly when the two laser fields are completely correlated. Experimentally, it is more convenient to work with a closely related case:[24,25] the simultaneous interaction of a broad-spectrum laser beam with the two transitions 1–3 and 2–3.

To solve this problem, we specify the optical field to be a traveling plane wave with a carrier frequency ω_L, a wave vector $\mathbf{k}$, and a unit polarization vector $\mathbf{e}$:

$$\mathbf{E}(z,\ t)=\mathbf{e}E(z,\ t)\exp[i(kz-\omega_L t)]+\text{c.c.} \tag{2.1}$$

Here $E(z,t)$ is the complex field amplitude, which has a regular component as well as possible frequency, phase, and amplitude fluctuations.

Let us consider the interaction of field (2.1) with a Λ medium. We assume that levels 1 and 2 are, for example, Zeeman sublevels of the ground state, while the third level (3) is an optical level. We assume that the partial probabilities for decays from level 3 to levels 1 and 2 are equal. For the case under consideration here (a single laser beam), condition (1.1) holds regardless of the velocity of the atom. We will accordingly ignore the thermal motion of the atoms below. When this motion is taken into account, the final results are changed in that the homogeneous optical absorption lineshape is replaced by an inhomogeneous lineshape.

Under these conditions, and in the approximation of a rotating wave, the system of kinetic equations for the density matrix of the atoms is

$$\begin{gathered}\dot{f}_{\mu\mu}=\frac{A}{2}f_{33}+\frac{\gamma}{2}(f_{\nu\nu}-f_{\mu\mu})+2\mathrm{Re}(iVf_{\mu3}),\\ f_{11}+f_{22}+f_{33}=1\\ \dot{f}_{\mu3}=-(A/2+i\Omega_\mu)f_{\mu3}+iV^*(f_{\mu\mu}-f_{33})+iV^*f_{\mu\nu},\\ \dot{f}_{12}=-(\Gamma+i\omega_{21})f_{12}+iVf_{13}-iV^*f_{32},\end{gathered} \tag{2.2}$$

where f_{ik} are the elements of the (unaveraged) density matrix, γ is the rate of longitudinal relaxation between levels 1 and 2, $V(z,t)$ is the Rabi frequency for the field $E(z,t)$, A is the rate of the spontaneous decay of the $|3\rangle$ state, $\Omega_\mu=\omega_L-\omega_{3\mu}$, and ω_{ik} is the frequency of the i–k transition. For simplicity we have assumed that V is the same for the two optical transitions (3–1 and 3–2).

We integrate the last three equations in (2.2), and we substitute the expressions for the optical coherences f_{13} and f_{23} into the integral for the low-frequency coherence f_{12}:

$$\begin{aligned}f_{12}=&-\int_{-\infty}^{t}dt'\exp[(i\omega_{21}-\Gamma)(t-t')]\\ &\times\int_{-\infty}^{t'}dt''\left\{\exp\left[-\left(i\Omega_1+\frac{A}{2}\right)(t'-t'')\right]\right.\\ &\times V(z,t')V^*(z,t'')(f_{11}-f_{33}+f_{12})|_{t''}\\ &+\exp\left[\left(i\Omega_2-\frac{A}{2}\right)(t'-t'')\right]V^*(z,t')V(z,t'')\\ &\left.\times(f_{22}-f_{33}+f_{12})|_{t''}\right\}.\end{aligned} \tag{2.3}$$

We average this expression over the possible fluctuations of the random field, in order to go over to the components of the average density matrix of the atoms, $\hat{\rho}$. Under the assumption that the state of the atomic system does not affect the subsequent values of $E(z,t)$ at the same point, and under the assumption that the inequality $t'\geqslant t''$ holds in (2.3), we can "uncouple" the ternary correlation functions which arise in the course of the averaging. For example, we can write

$$\begin{aligned}&\langle V(z,\ t')V^*(z,\ t'')f_{12}(z,\ t'')\rangle\\ &=\langle V(z,\ t')V^*(z,\ t'')\rangle\langle f_{12}(z,\ t'')\rangle\\ &=\langle V(z,\ t')V^*(z,\ t'')\rangle\rho_{12}.\end{aligned} \tag{2.4}$$

The correlation function describing the steady-state process is determined by the spectral intensity $J(\omega,z)$ of the laser field, according to the Wiener-Khinchin theorem. This correlation function can be written in the form (Ref. 26, for example)

$$\langle V(z,t')V^*(z,t'')\rangle=B\int d\omega\exp[-i(\omega-\omega_L)(t'-t'')]J(\omega,z), \tag{2.5}$$

where B is the Einstein coefficient for the stimulated emission. Integrating over t'' and t' in (2.3), and focusing on the steady-state solution, we then find the following expression for the low-frequency coherence:

$$\rho_{12}=-\frac{1}{2}\frac{\tilde{W}_1(\rho_{11}-\rho_{33})+\tilde{W}_2^*(\rho_{22}-\rho_{33})}{\Gamma+{}^1/_2\tilde{W}_1+{}^1/_2\tilde{W}_2^*-i\omega_{21}}, \quad (2.6)$$

where

$$\tilde{W}_\mu=2B\int\limits_{-\infty}^{\infty}\frac{J(\omega,z)\,d\omega}{A/2+i(\omega-\omega_{3\mu})}, \quad \mu=1,2. \quad (2.7)$$

Taking the average of the equations for the populations f_{ii} of system (2.2) in the corresponding way, and setting $\dot{\rho}_{ii}=0$ for the steady state, we find the following system of equations ($\mu,\nu=1,2;\mu\neq\nu$):

$$A\rho_{33}+\gamma(\rho_{\nu\nu}-\rho_{\mu\mu})-2W_\mu(\rho_{\mu\mu}-\rho_{33})$$

$$+\mathrm{Re}\left\{\frac{\tilde{W}_\mu}{G_{\mu\nu}}[\tilde{W}_\mu(\rho_{\mu\mu}-\rho_{33})+\tilde{W}_\nu^*(\rho_{\nu\nu}-\rho_{33})]\right\}=0, \quad (2.8)$$

$$\rho_{11}+\rho_{22}+\rho_{33}=1.$$

Here $G_{\mu\nu}=\Gamma+\tilde{W}_\mu/2+\tilde{W}_\nu^*/2+i\omega_{\mu\nu}$, where $W_\mu=\mathrm{Re}\,\tilde{W}_\mu$.

Equations (2.8) are sufficient for describing CPT in the field of nonmonochromatic laser light for an optically thin medium. [If we are instead interested in the propagation of the light through an optically dense medium, we need to supplement this system of equations with a transport equation to describe the change in the spectral intensity $J(\omega,z)$ of the light.]

Let us analyze the case of an optically thin medium. We assume that the laser light spectrum is symmetric with respect to the carrier frequency ω_L and that the frequency spacing between the lower levels, ω_{21}, is much smaller than the spectral width of the incident light, Δ: $\omega_{21}\ll\Delta$. Under these conditions we have $\tilde{W}_1=\tilde{W}_2^*$, $W_1=W_2\equiv W$, and $\rho_{11}=\rho_{22}$, and ρ_{33} from (2.8) is

$$\rho_{33}=\frac{W}{A}\left[1-\frac{W(\Gamma+W)}{(\Gamma+W)^2+\omega_{21}^2}\right]. \quad (2.9)$$

In deriving (2.9) we used the condition $W\ll A$; here W means the rate of optical excitation.

From (2.9) we see that under the conditions $W\gg\Gamma,\omega_{21}$ the population ρ_{33} satisfies $\rho_{33}=\Gamma/A$ and does not depend on the laser intensity. This result is evidence of population trapping in the lower levels. If the condition $\omega_{21}\gg W$ holds instead, then there is no coherent trapping even if $W\gg\Gamma$, and the upper level is filled in proportion to the light intensity: $\rho_{33}\propto W$. These conclusions are supported by the experiments of Ref. 25, where the frequency spacing (ω_{21}) of the Zeeman sublevels of the ground state of cesium atoms was varied by varying the strength of a static magnetic field.

We thus conclude that CPT can be observed even when an atomic system is excited by only a single laser beam, but in this case the spacing of the lower levels, ω_{21}, must not exceed the rate of optical excitation, W: $\omega_{21}<W$.

3. OPTICALLY DENSE MEDIUM

We turn now to the case of an optically dense medium. For this purpose, Eqs. (2.8) must be supplemented with an equation for the propagation of the light. To find this equation, we average a simplified Maxwell's wave equation:[8]

$$\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=i\frac{2\pi N\omega_L}{c}(d_{13}f_{31}+d_{23}f_{32}), \quad (3.1)$$

where $d_{\mu 3}$ is the matrix element of the dipole moment for the μ–3 transition, and N is the density of atoms.

Since the spectral intensity of the light, $J(\omega,z)$, is related to the field by

$$J(\omega,z)=\frac{1}{2\pi}\mathrm{Re}\int\limits_{-\infty}^{\infty}\left\langle E(z,t)E^*(z,t')\frac{c}{8\pi}\right\rangle$$

$$\times\exp[i(\omega-\omega_L)(t-t')]\,dt,$$

by carrying out the appropriate manipulations with (3.1) and setting $d_{31}\approx d_{32}\equiv d$, we find

$$\frac{\partial J(\omega,z)}{\partial z}$$

$$=-2\hbar\omega_L N\sum_{\mu=1,2}\frac{1}{2\pi}\mathrm{Re}\int\limits_{-\infty}^{\infty}\int\limits_{-\infty}^{t'}\exp\left[\left(\frac{A}{2}+i\Omega_\mu\right)(t''-t')\right.$$

$$\left.+i(\omega-\omega_L)(t-t')\right]\langle V(z,t)V^*(z,t'')(f_{\mu\mu}-f_{33}+f_{\mu\nu})|_{t'}\rangle\,dt''\,dt, \quad (3.2)$$

where $\nu=1,2$, $\nu\neq\mu$. Breaking up the ternary correlation functions in the same manner as above, and integrating over t'', we finally find

$$\frac{\partial J(\omega,\tau)}{\partial\tau}$$

$$=-J(\omega,\tau)\sum_{\mu=1,2}\mathrm{Re}\left[\frac{A}{A/2+i(\omega-\omega_{3\mu})}(\rho_{\mu\mu}-\rho_{33}+\rho_{\mu\nu})\right]. \quad (3.3)$$

For convenience we have introduced here a dimensionless optical length τ, which is given by

$$\tau=\frac{4\pi N\omega_L d^2}{c\hbar A}z.$$

Equations (2.6), (2.8), and (3.3) constitute a self-consistent system of nonlinear integrodifferential equations. These equations can be used to find a correct description of the propagation of laser light, with the real spectrum, under the conditions corresponding to CPT.

Solving this system of equations in the general case would require numerical calculations, but there is one case of practical importance in which an analytic solution can be found. This case characterizes radiation transport in a medium in detail.

We assume the same conditions as in the derivation of (2.9). Equation (3.3) can then be written

$$\frac{\partial J(\omega,\tau)}{\partial\tau}=-J(\omega,\tau)[\beta_1(\omega)+\beta_2(\omega)]R(W), \quad (3.4)$$

with the boundary condition $J(\omega,0)=J_0(\omega)$. Here $\beta_\mu(\omega)$, the profile of the absorption line corresponding to the μ–3 transition ($\mu=1,2$), is given by

$$\beta_\mu(\omega)=\frac{A^2/4}{A^2/4+(\omega-\omega_{3\mu})^2},$$

where $R(W)=R[W(\tau)]$ is an ω-independent function of the optical length, which is determined by the population of the upper level:

$$R(W)=\frac{A}{W}\rho_{33}, \tag{3.5}$$

$$W(\tau)=\sigma\int\beta_\mu(\omega)J(\omega,\tau)\,d\omega, \tag{3.6}$$

and σ is the cross section for absorption of light at the center of the line.

We seek a solution of Eq. (3.4) in the form

$$J(\omega,\tau)=J_0(\omega)\exp\{-[\beta_1(\omega)+\beta_2(\omega)]\psi(\tau)\}, \tag{3.7}$$

where $\psi(\tau)$ is a nonlinear optical length. Substituting (3.7) into (3.4), we find that ψ satisfies the equation

$$\frac{d\psi}{d\tau}=R(\psi),\qquad \psi(0)=0, \tag{3.8}$$

which can be integrated easily:

$$\tau=\int_0^\psi\frac{d\varphi}{R(\varphi)}. \tag{3.9}$$

For given lineshapes of the absorption, $\beta_\mu(\omega)$, and of the optical excitation, $J_0(\omega)$, the function $R=R(\psi)$ depends explicitly on ψ, and (3.9) is simply a transcendental equation for $\psi=\psi(\tau)$. To demonstrate this point, we substitute (3.5) and (2.9) into (3.9). The latter then takes the form

$$\tau=\psi+\int_0^\psi\frac{W(\Gamma+W)}{\Gamma(\Gamma+W)+\omega_{21}^2}\,d\varphi, \tag{3.10}$$

where W is given by (3.6) and is a function of ψ. If $\omega_{21}\ll A$, for example, i.e., if the absorption lines $\beta_1(\omega)$ and $\beta_2(\omega)$ overlap strongly, then by approximating them by a single Lorentzian line and assuming that the optical excitation line $J_0(\omega)$ is also Lorentzian, we find from (3.6)

$$W=W_0\exp(-\psi/2)\,[I_0(\psi/2)-I_1(\psi/2)], \tag{3.11}$$

where I_0 and I_1 are modified Bessel functions, and $W_0\equiv W(\tau=0)$.

Let us find the law describing the decay of the integral intensity of the optical radiation, which we define by

$$U(\tau)=\int J(\omega,\tau)\,d\omega. \tag{3.12}$$

We note that U and W are related by

$$W=-\frac{\sigma}{2}\frac{dU}{d\psi}. \tag{3.13}$$

In several cases, expression (3.10) can be simplified. We set $\omega_{21}^2\ll\Gamma(\Gamma+W)$. Using (3.13), we then find

$$\tau=\psi+\frac{W_0}{\Gamma}\left[1-\frac{U(\psi)}{U_0}\right], \tag{3.14}$$

where $U_0\equiv U(\tau=0)$ is the integral intensity of the laser light at the entrance to the medium. A simple analytic solution of (3.14), valid for arbitrary lineshapes of the incident light, $J_0(\omega)$, and of the absorption lines, $\beta_\mu(\omega)$, shows that the propagation of each spectral component $J(\omega,\tau)$ in (3.7) is determined by the set of all other components. In other words, ψ depends on the integral intensity U.

It has been established[7,8] that coherent population trapping arises only if the intensity of the laser light exceeds a certain threshold, the so-called coherent intensity:

$$U(\tau)\gg U_c=\frac{\Gamma}{A}U_n, \tag{3.15}$$

where U_n is the saturation intensity of the optical transition. In (3.14), condition (3.15) corresponds to the case $W_0\gg\Gamma$. Expanding $U(\psi)$ in a power series in ψ, and using (3.14), we find

$$U(\tau)=U_0\left(1-\frac{\Gamma}{W_0}\tau\right). \tag{3.16}$$

In other words, the decay of the integral intensity in the medium is linear, regardless of the lineshapes of the incident light and the absorption. This is the behavior until $U(\tau)$ becomes comparable to the intensity U_c.

The nature of the decay at large optical thicknesses τ under the condition $U(\tau)\ll U_c$, or at arbitrary τ if the condition $U_0\ll U_c$, holds, depends strongly on the $J_0(\omega)$ and $\beta_\mu(\omega)$ lineshapes. It is determined completely by the wings of these lines.

Under the condition $U(\tau)\ll U_c$ ($W\ll\Gamma$) we find from (3.10)

$$\tau=\psi+\frac{W_0\Gamma}{\Gamma^2+\omega_{21}^2}\left(1-\frac{U}{U_0}\right). \tag{3.17}$$

Since we have $W_0\ll\Gamma$, we thus have $\psi\simeq\tau$. According to (3.7), the spectral intensity $J(\omega,\tau)$ thus falls off exponentially. The behavior of the integral intensity, in contrast, depends on the combinations of $J_0(\omega)$ and $\beta_\mu(\omega)$. For Lorentzian lineshapes of both the incident light and the absorption, with identical widths, we find from (3.12)

$$U(\tau)=U_0\exp(-\tau/2)I_0(\tau/2). \tag{3.18}$$

At large optical thicknesses we have $U(\tau)\propto\tau^{-1/2}$. This slow decay of $U(\tau)$ is due entirely to the wings of the emission line. It demonstrates that the approximation of the spectrum of the incident light by a Lorentzian line is a rather crude approximation. If the laser light has an approximately Gaussian spectrum, we would have

$$U(\tau)\propto\exp(-\lambda\tau^{1/2}), \tag{3.19}$$

where the parameter λ depends on the ratio of the widths of the incident line and the absorption line.[27] These results remain in force in the case $\omega_{21}\gg W_0$, even if $U\gg U_c$.

Finally, we wish to call attention to an important result of this section of the paper: A coherent bleaching of a medium (a linear decay law) occurs for laser light with a spectrum spanning the two lower sublevels in the cw regime, and not only for ultrashort pulses with a repetition frequency which is a multiple of ω_{21} (Ref. 7).

4. CONCLUSION

In this paper we have examined the coherent population trapping which arises in a medium to which a single laser beam, with a finite spectral width, is applied. We have shown that if the spectrum of the light spans the magnetic sublevels of the ground state, and if the frequency distance between these sublevels is smaller than the rate of optical excitation, then the atoms will be trapped in these sublevels, and the medium will be bleached. The decay of the integral intensity of the laser light in the medium is linear in this case and does not depend on the lineshape of the incident light or that of the absorbed. The propagation of each spectral component $J(\omega,\tau)$, on the other hand, is determined by the set of all other components (there is a "frequency memory"). At large optical thicknesses, at which there is no CPT, the nature of the decay of the integral intensity depends strongly on the wings of the line of the incident light.

Since this effect is sensitive to changes in the strength of the static external magnetic field, it might prove useful for controlling the propagation of light through a medium and also for developing CPT quantum magnetometers.

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