Multicolored Coherent Population Trapping in a Λ System Using Phase Modulated Fields

Harshawardhan Wanare*

Department of Physics and Center for Laser Technology, Indian Institute of Technology, Kanpur 208 016, India

(Received 2 January 2006; published 11 May 2006)

We demonstrate the simultaneous occurrence of coherent population trapping at a series of frequencies separated by modulation frequency of phase-modulated fields. The two arms of the Λ system are coupled to two phase-modulated fields and the response of the atomic system to such fields is calculated nonperturbatively. A judicious choice of modulation characteristics allows one to selectively switch on or off the occurrence of coherent population trapping at specific frequencies. A new technique is developed to compute two-dimensional tridiagonal matrix equations. This generalized technique provides the vital methodology needed to calculate the response of such systems in the strong modulation regime and for arbitrary field strengths.

DOI: 10.1103/PhysRevLett.96.183601 PACS numbers: 42.50.Gy, 42.50.Hz, 42.65.−k

One of the most significant and macroscopic demonstrations of atomic coherence is the disappearance of fluorescence due to coherent population trapping (CPT), also known as dark resonance. The CPT state has been used in a variety of applications like atomic cooling [1], electromagnetically induced transparency (EIT) [2], lasing without inversion [3], and adiabatic population transfer [4]. Such coherent atomic interference phenomenon is usually associated with excitation by a monochromatic field with well-controlled phase stability. In this Letter, we demonstrate that even on excitation with *multicolored* phase-modulated (PM) fields, CPT occurs at a series of frequencies.

Multicolored CPT provides a promising avenue towards a variety of applications. For example, in atomic cooling substantial improvement is envisaged, wherein it allows for addressing atoms over a large velocity range and capturing them into lower momentum states at a series of velocity selective CPT states. These states would result in significantly reduced spontaneous emission reheating and could lead to the capture of considerably larger fraction of atoms. In the context of quantum information science, EIT based on multicolored CPT provides a platform for multipartite entanglement across many distinct frequencies which can be addressed individually. The multicolored atomic coherence can be utilized to generate giant refractive index accompanied with negligible absorption at a *series* of frequencies and could lead to novel pulse dynamics and pulse shape engineering. Many more such effects, which are an interplay of reduced absorption and enhanced atomic coherence at a *series* of distinct frequencies opens up a platform for multicolored coherent control. The limitation is the trade off between the number of frequencies where CPT occurs and the magnitude of atomic coherence occurring at these frequencies.

Modulated fields are widely used and provide powerful spectroscopic tool. In particular, phase modulation spectroscopy has become a routine technique, wherein the spectral content of a PM probe field is used to extract resonances at the modulation frequency. The strength of the technique lies in its ability to simultaneously address resonances at disparate frequencies with precise control of the modulation characteristics [5]. In this Letter, the comb of frequencies of two PM fields are matched across atomic transitions leading to CPT phenomenon. Furthermore, selective tailoring of spectral content of the comb of frequencies allows one to control occurrence of CPT at specific frequencies. In a related context, extremely narrow optical resonances have been observed with unprecedented resolution using the femtosecond-laser frequency combs [6], although the motivation there is quite different.

The PM field coupled to atomic system has been dealt with; in particular, the two-level atomic system driven by a PM field has been studied nonperturbatively [7], and later has been shown to exhibit population trapping [8]. This trapping is distinctly different from the CPT exhibited in three-level Λ systems. In the three-level atomic system suppression of a series of Autler-Townes resonances is shown to occur when driven by a strong PM field and probed by a weak monochromatic field [9]. In a three-level ladder system the PM drive also offers control over population transfer and trapping [10]. We have earlier shown the possibility of lasing without inversion [11] in a three-level V system driven by a single PM field. To our knowledge there has been no study involving *two arbitrary* PM fields coupling strongly to a three-level atom. It should be noted that this regime involving a large index of modulation coupled with strong fields has not been explored sufficiently due to obvious experimental and theoretical difficulties. In this Letter we briefly outline the methodology we have developed to calculate *nonperturbatively* the above response, the details of which will be published elsewhere.

A three-level atom in the Λ configuration is coupled to two distinct PM fields

$$\vec{E} = \vec{E}_1 e^{-i[\omega_1 t + \Phi_1(t)]} + \vec{E}_2 e^{-i[\omega_2 t + \Phi_2(t)]},$$
$$\Phi_1(t) = M_1 \sin(\Omega_1 t), \qquad \Phi_2(t) = M_2 \sin(\Omega_2 t), \tag{1}$$

where $\vec{E}_{1,2}$ are the amplitudes of the fields coupling the two arms of the Λ system and other parameters are as shown in Fig. 1.

The total Hamiltonian of the system is

$$H = \hbar\omega_{13}|1\rangle\langle 1| + \hbar\omega_{23}|2\rangle\langle 2| - \vec{d}\cdot\vec{E}, \tag{2}$$

where the dipole moment operator $\vec{d} = \vec{d}_{13}|1\rangle\langle 3| + \vec{d}_{12}|1\rangle\langle 2| + \text{c.c.}$ The first two terms in the Hamiltonian correspond to the unperturbed atomic system where the energies are measured from the ground state $|3\rangle$, and the last term is the interaction term in the dipole approximation. The semiclassical density matrix equation governing the atom-field dynamics is

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho] + \mathcal{L}\rho, \tag{3}$$

where, the Liouvillian operator $\mathcal{L}$ contains all the incoherent processes, such as spontaneous emission from the level $|1\rangle$ to $|2\rangle$ ($|3\rangle$) with rate $2\gamma_1$ ($2\gamma_2$), and the relaxation of the ground state coherence Γ between the lower states $|2\rangle$ and $|3\rangle$ [12]. We transform the equation of motion (3) into a frame rotating with the instantaneous frequency of the field and undertake the rotating-wave approximation. This involves neglecting the counterrotating terms at nearly twice the optical frequency in comparison to slowly varying terms at difference frequencies for near resonant excitation. This approximation is quite valid at optical frequencies and also for the modulated field because $|d\Phi_{1,2}(t)/dt| \ll \omega_{1,2}$ for typical phase modulation in the optical regime.

Because of the coupling of the atom to the PM fields, the slowly varying density matrix equations involve time-dependent detuning factors which go as $d\Phi_{1,2}(t)/dt$. We use the two-dimensional Fourier decomposition

$$\rho_{ij} = \sum_{p,q=-\infty}^{\infty} \rho_{ij}^{(p,q)} e^{-i(p\Omega_1 t + q\Omega_2 t)}, \tag{4}$$

which involves integral harmonics of the modulation frequencies $\Omega_{1,2}$ and there are a set of nine equations for ρ_{ij}, where i, j label the atomic levels 1, 2, and 3. By equating various powers of $\Omega_{1,2}$, we obtain an infinite set of coupled first order differential equations for $\rho_{ij}^{(p,q)}$, where p and q are integers that independently vary from $-\infty$ to ∞ and denote the various harmonics. Furthermore, we also note that the set of equations for (p, q) are coupled to the set for $(p \pm 1, q \pm 1)$, because phase modulations $\Phi_{1,2}(t)$ are circular functions. The closure of the above system requires that $\rho_{11}^{(p,q)} + \rho_{22}^{(p,q)} + \rho_{33}^{(p,q)} = \delta_{p,0}\delta_{q,0}$. This population conservation condition is used to eliminate one of the nine equations in each set of (p, q) equations, giving rise to following two-dimensional tridiagonal equation

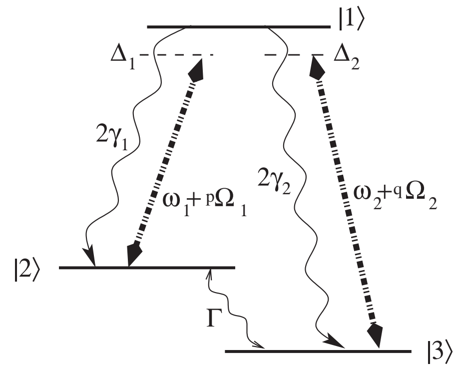

FIG. 1. Three-level Λ system driven by PM optical fields at the central frequency ω_1 (ω_2) and modulation frequency Ω_1 (Ω_2) couple the transition $|1\rangle \leftrightarrow \langle 2|$ ($|1\rangle \leftrightarrow \langle 3|$), where p (q) take positive and negative integer values corresponding to the various harmonics of the PM fields.

$$\frac{d}{dt}\mathbf{\Psi}^{p,q} = \hat{\mathbf{A}}^{p,q}\mathbf{\Psi}^{p,q} + \hat{\mathbf{B}}\mathbf{\Psi}^{p-1,q} + \hat{\mathbf{C}}\mathbf{\Psi}^{p+1,q} + \hat{\mathbf{D}}\mathbf{\Psi}^{p,q-1} + \hat{\mathbf{E}}\mathbf{\Psi}^{p,q+1} + \mathbf{R}^{o}\delta_{p,0}\delta_{q,0}, \tag{5}$$

where, $\mathbf{\Psi}^{p,q}$ is an 8×1 column vector with elements $\rho_{ij}^{(p,q)}$ in the given order $\mathbf{\Psi}^{p,q} = [\rho_{11}, \rho_{12}, \rho_{13}, \rho_{21}, \rho_{22}, \rho_{23}, \rho_{31}, \rho_{32}]^T$, and $\hat{\mathbf{A}}\ldots\hat{\mathbf{E}}$ represent 8×8 matrices with time-independent coefficients. $\mathbf{R}^o$ is also an 8×1 column vector with constant entries. The $\mathbf{R}^o$ term arises due to population conservation in the closed three-level system considered here. The nonzero elements of the matrices in Eq. (5) are given below:

$$\begin{gathered}
A_{11}^{p,q} = -2(\gamma_1+\gamma_2) + i(p\Omega_1 + q\Omega_2), \qquad A_{22}^{p,q} = -(\gamma_1+\gamma_2+i\Delta_1) + i(p\Omega_1 + q\Omega_2),\\
A_{33}^{p,q} = -(\gamma_1+\gamma_2+i\Delta_2) + i(p\Omega_1 + q\Omega_2), \qquad A_{44}^{p,q} = -(\gamma_1+\gamma_2-i\Delta_1) + i(p\Omega_1 + q\Omega_2),\\
A_{55}^{p,q} = i(p\Omega_1 + q\Omega_2), \qquad A_{66}^{p,q} = -[\Gamma - i(\Delta_1-\Delta_2)] + i(p\Omega_1 + q\Omega_2),\\
A_{77}^{p,q} = -(\gamma_1+\gamma_2-i\Delta_2) + i(p\Omega_1 + q\Omega_2), \qquad A_{88}^{p,q} = -[\Gamma + i(\Delta_1-\Delta_2)] + i(p\Omega_1 + q\Omega_2),\\
A_{12}^{p,q*} = A_{14}^{p,q} = -A_{21}^{p,q} = A_{25}^{p,q} = A_{36}^{p,q} = -A_{41}^{p,q*} = A_{45}^{p,q*} = -A_{52}^{p,q*} = -A_{54}^{p,q} = -A_{63}^{p,q*} = A_{78}^{p,q*} = -A_{87}^{p,q} = iG_1,\\
A_{13}^{p,q*} = A_{17}^{p,q} = A_{28}^{p,q} = -A_{35}^{p,q} = A_{46}^{p,q*} = -A_{64}^{p,q} = -A_{75}^{p,q*} = -A_{82}^{p,q*} = iG_2, \qquad A_{31}^{p,q} = A_{71}^{p,q*} = -i2G_2,\\
A_{51}^{p,q} = 2\gamma_1, \qquad B_{22} = -B_{44} = -B_{66} = B_{88} = C_{22} = -C_{44} = -C_{66} = C_{88} = iM_1\Omega_1/2,\\
D_{33} = D_{66} = -D_{77} = -D_{88} = E_{33} = E_{66} = -E_{77} = -E_{88} = iM_2\Omega_2/2, \qquad R_3^o = R_7^{o*} = iG_2 \times \delta_{p,0}\delta_{q,0}.
\end{gathered} \tag{6}$$

In order to obtain the steady-state solutions, we set the time derivatives to zero and recast the above equation to obtain tridiagonal matrix recurrence relation in p of the following form:

$$\begin{bmatrix} \vdots & \vdots & \vdots \\ \hat{\mathbf{A}}^{p,q-1} & \hat{\mathbf{E}} & \mathbf{0} \\ \hat{\mathbf{D}} & \hat{\mathbf{A}}^{p,q} & \hat{\mathbf{E}} \\ \mathbf{0} & \hat{\mathbf{D}} & \hat{\mathbf{A}}^{p,q+1} \\ \vdots & \vdots & \vdots \end{bmatrix} \begin{bmatrix} \vdots \\ \mathbf{\Psi}^{p,q-1} \\ \mathbf{\Psi}^{p,q} \\ \mathbf{\Psi}^{p,q+1} \\ \vdots \end{bmatrix} + \begin{bmatrix} \vdots & \vdots & \vdots \\ \hat{\mathbf{B}} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \hat{\mathbf{B}} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \hat{\mathbf{B}} \\ \vdots & \vdots & \vdots \end{bmatrix} \begin{bmatrix} \vdots \\ \mathbf{\Psi}^{p+1,q-1} \\ \mathbf{\Psi}^{p+1,q-1} \\ \mathbf{\Psi}^{p+1,q+1} \\ \vdots \end{bmatrix} + \begin{bmatrix} \vdots & \vdots & \vdots \\ \hat{\mathbf{C}} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \hat{\mathbf{C}} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \hat{\mathbf{C}} \\ \vdots & \vdots & \vdots \end{bmatrix} \begin{bmatrix} \vdots \\ \mathbf{\Psi}^{p-1,q-1} \\ \mathbf{\Psi}^{p-1,q} \\ \mathbf{\Psi}^{p-1,q+1} \\ \vdots \end{bmatrix} + \begin{bmatrix} \vdots \\ 0 \\ \mathbf{R}^{o} \\ 0 \\ \vdots \end{bmatrix} = 0, \quad (7)$$

where, if p varies from $-N$ to N, and q from $-M$ to M, then the size of square matrices in Eq. (7) would be $(8[2M+1]) \times (8[2M+1])$ and the size of the vectors would be $(8[2M+1]) \times 1$. In order to obtain $\mathbf{\Psi}$, the matrix continued fraction technique is used to solve this two-dimensional tridiagonal relations in p and q. The matrix continued fraction technique is as described in Refs. [7,13], however, the usual technique involves tridiagonal relations involving just one index, say p. Here, we need to accommodate two-dimensional tridiagonal relations, and recasting Eq. (5) in the form of Eq. (7) results in tridiagonal relations involving only one index p. Although, the matrices in Eq. (7) are much larger because each matrix itself spans over all possible values of q, yet, it continues to involve the tridiagonal matrix relation involving p and $p \pm 1$. This system of equations can be solved to the desired accuracy by taking sufficient number of terms, in the matrix size, as well as in the continued fraction. The limits N and M depend on the choice of G_i and M_i, and as is well known, $J_n(M_i)$ is a decreasing function of n when $n > M_i$; one needs to take at least as many terms. Needless to say, we have checked the numerical convergence of the results by increasing the limits N and M, which results in more terms in the continued fraction and larger matrices, respectively.

In this Letter, we present exact nonperturbative numerical computation because, the Rabi frequency of the fields, the values of the modulation frequency $\Omega_{1,2}$, and the index of modulation $M_{1,2}$ are large. One observes CPT occurring simultaneously at a series of frequencies, when $\Omega_1 = \Omega_2$ for $\Delta_1 = \Delta_2 \pm k\Omega$, see Fig. 2. Here the two-photon resonant Raman condition is satisfied not just for the central frequencies but also for each sideband of the PM fields. Moreover, choosing $G_1 = G_2$ and $M_1 = M_2$ ensures that the strength of each pair of fields involved in the Raman transition given by $G_i J_k(M_i)$ is the same. The CPT effect can be dominant whenever the comb of frequencies on one arm of the Λ system matches with the comb of frequencies on the other arm, albeit with different weights. For different values of the detuning Δ_2, the comb of frequencies $\omega_2 + k\Omega_2$ keeps shifting and wherever the two-photon Raman condition is satisfied, which occurs simultaneously for a series of frequencies in the comb, the CPT effect is significant. We show the excited state population $\rho_{11}^{(0,0)}$ and the extent of coherence between the ground states $\mathrm{Re}\{\rho_{23}^{(0,0)}\}$ in Fig. 2. All the frequency units throughout this Letter are scaled in the units of γ_1. The dips in the excited state population due to CPT are accompanied by increased atomic coherence at a series of equidistant frequencies. One can obtain significantly larger values for the atomic coherence by decreasing the index of modulation, which in turn limits the number of sidebands of the PM field and thus the occurrence of CPT to those few sidebands. It appears as though the maximum atomic coher-

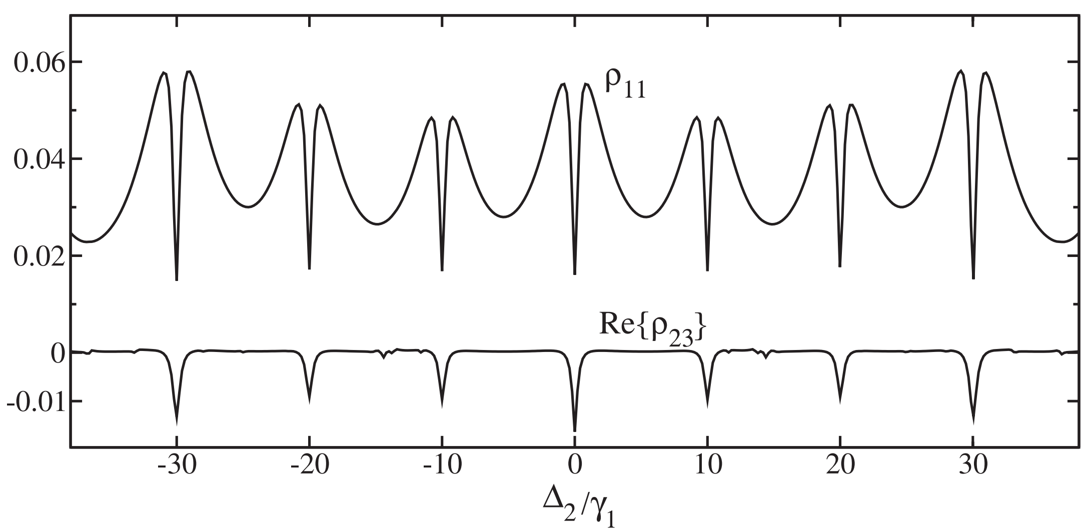

FIG. 2. Multicolored CPT occurs at a series of frequencies separated by integer multiples of the modulation frequency. The upper curve is the excited state population $\rho_{11}^{(0,0)}$ and the lower curve is the measure of atomic coherence $\mathrm{Re}\{\rho_{23}^{(0,0)}\}$. The parameters are chosen to be the same along the two arms of the Λ system, $\Omega_1 = \Omega_2 = 10\gamma_1$, $M_1 = M_2 = 14.0$, $G_1 = G_2 = 3\gamma_1$, $\gamma_2 = \gamma_1$, $\Gamma = 0.01\gamma_1$, and the detuning $\Delta_1 = 0$.

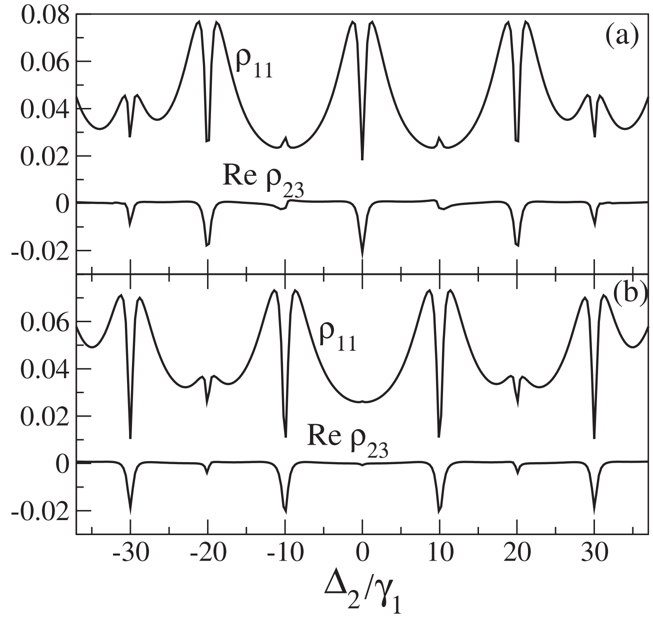

FIG. 3. The CPT feature is selectively switched off by careful choice of the modulation characteristics, (a) the CPT at $\Delta_2 = \pm 10\gamma_1$ are suppressed with $M_1 = M_2 = 9.4$ and $\Omega_1 = \Omega_2 = 10\gamma_1$; (b) the CPT features at $\Delta_2 = 0$ and $\pm 20\gamma_1$ are suppressed with $M_1 = 9.4$, $M_2 = 8.6537$, $\Omega_1 = 20\gamma_1$, and $\Omega_2 = 10\gamma_1$. The other parameters common to (a) and (b) are $G_1 = G_2 = 3\gamma_1$, $\Delta_1 = 0$, and $\Gamma = 0$.

ence obtained in the usual CPT case (with monochromatic fields) is *shared* across a comb of frequencies in the multicolored case [14]. Furthermore, the Bessel function $[J_k(M)]$ for the coupling strength provides significant control over the response, for example, a choice of M such that $J_k(M) = 0$ eliminates the $\omega \pm k\Omega$ sideband. Such control can be used to selectively shut off CPT at particular frequencies, as is shown in Fig. 3.

We further illustrate the issue of control of CPT at specific frequencies. We choose $M_1 = M_2 = 9.4$ this eliminates the CPT $\omega_1 \pm \Omega$ frequency, as seen in Fig. 3(a). Even though the choice of M_i does not eliminate any specific frequencies of the comb, the nonlinear coupling is such that the CPT effect is eliminated at some specific frequencies. It is worthwhile to note that the coupling being highly nonlinear, a simplistic matching of comb of frequencies need not always result in CPT. Although a judicious choice of modulation parameters does provide sufficient control over CPT. We choose $M_2 = 8.6537$ such that $J_0(M_2) = 0$ this eliminates the central ω_2 frequency and also choose the modulation frequencies such that $\Omega_1 = 20\gamma_1$ and $\Omega_2 = 10\gamma_1$. As the detuning Δ_2 is varied, one observes that the CPT has disappeared for the zero detuning case. By eliminating the central frequency of one of the fields, and ensuring that no matching of frequencies occurs even at the neighboring frequencies, complete elimination of CPT at the line center is obtained, see Fig. 3(b). On similar grounds the CPT feature at $\Delta_2 = \pm 20\gamma_1$ is nearly nonexistent.

We would like to emphasize that the contribution of all the neighboring frequencies is as important as the central frequency itself. It is also noted that the CPT dip seems sharper in the multicolored field case than the monochromatic one. This is because the power broadening effects are less prevalent here as the same field strength G_i is being shared by series of frequencies with weights $J_k(M_i)$.

In conclusion, we have shown the simultaneous occurrence of CPT at a series of frequencies when driven by PM fields along the two arms of the Λ system. This would facilitate the realization of a variety of a coherent optical phenomenon [15] *simultaneously* at a series of frequencies. A proper choice of parameters can coherently switch on or off the CPT structure selectively. We have developed a generalization of the matrix continued fraction technique to deal with two-dimensional tridiagonal relations. This technique being nonperturbative and exact provides a useful tool to model a family of strongly coupled systems.

*Electronic address: hwanare@iitk.ac.in

[1] See E. Arimondo, in Progress in Optics, edited by E. Wolf (Elsevier, Amsterdam, 1996), Vol. 35, p. 257 for an overview of CPT phenomenon and its applications.
[2] S. E. Harris, Phys. Today **50**, No. 7, 36 (1997).
[3] E. S. Fry, X. Li, D. Nikonov, G. G. Padmabandu, M. O. Scully, A. V. Smith, F. K. Tittel, C. Wang, S. R. Wilkinson, and S. Y. Zhu, Phys. Rev. Lett. **70**, 3235 (1993).
[4] S. Schiemann, A. Kuhn, S. Steuerwald, and K. Bergmann, Phys. Rev. Lett. **71**, 3637 (1993).
[5] A. Schenzle, R. G. DeVoe, and R. G. Brewer, Phys. Rev. A **25**, 2606 (1982).
[6] See Th. Udem, R. Holzwarth, and T. W. Hänsch, Nature (London) **416**, 233 (2002), and the references therein.
[7] N. Nayak and G. S. Agarwal, Phys. Rev. A **31**, 3175 (1985).
[8] G. S. Agarwal and W. Harshawardhan, Phys. Rev. A **50**, R4465 (1994).
[9] V. N. Smelyanskiy, G. W. Ford, and R. S. Conti, Phys. Rev. A **53**, 2598 (1996); V. N. Smelyanskiy, R. S. Conti, and G. W. Ford, *ibid.* **55**, 2186 (1997).
[10] W. Harshawardhan and G. S. Agarwal, Phys. Rev. A **55**, 2165 (1997).
[11] Harshawardhan Wanare, Phys. Rev. A **65**, 033417 (2002).
[12] G. Orriols, Nuovo Cimento B **53**, 1 (1979); S. Swain, J. Phys. B **15**, 3405 (1982).
[13] H. Risken, *The Fokker-Planck Equation* (Springer-Verlag, Berlin, 1984), p. 196.
[14] The CPT occurs at more than 20 frequencies for the parameters in Fig. 2 (not shown fully). For smaller index of modulation $M_1 = M_2 = 1.0$ one obtains $|\mathrm{Re}\{\rho_{23}\}| \approx 0.3$ at the line center and ≈ 0.2 at the sidebands, and the occurrence of CPT is now restricted to just these three frequencies. The value of atomic coherence is comparable to the usual monochromatic CPT case, where $|\mathrm{Re}\{\rho_{23}\}| = 0.5$.
[15] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, England, 1997).