# Microscopic approach to coherent population trapping state and its relaxation in a dense medium

**G.S. Agarwal**

Physical Research Laboratory, Ahmedabad-380009, India
and
Max-Planck Institut für Quantenoptik, D-85748 Garching, Germany

*gsa@prl.ernet.in*

**Abstract:** Using a master equation with cooperative interaction of radiative nature included, we demonstrate the generation and relaxation characteristics of the coherent population trapping state. We also show how the microscopic master equation in the mean field approximation leads to density matrix equations obtained from local field considerations.

**OCIS codes:** (270.0270) Quantum optics; (020.0020) Atomic and molecular physics

## References

1. For a recent review on coherent population trapping see E. Arimondo in *Progress in Optics*, Vol. XXXV, ed. E. Wolf (North-Holland, Amsterdam, 1996) p. 257 and references therein.
2. I.V. Jyotsna and G.S. Agarwal, *Phys. Rev.* **A 53**, 1690 (1996).
3. C.M. Bowden, A.S. Manka, J.P. Dowling and M. Fleischhauer, in *Coherence and Quantum Optics*, eds. J.H. Eberly, L. Mandel and E. Wolf (Plenum, NewYork, 1996) p. 271.
4. G.S. Agarwal, *Quantum Optics* (Springer-Verlag, Berlin, 1974) Sec. 6.
5. M. Born and E. Wolf, *Principles of Optics*, 6th ed. (Pergamon, Oxford, 1980) Chap. 2.
6. R. Friedberg, S.R. Hartmann, and Jamal T. Manassah, *Phys. Rev.* **A 40**, 2446 (1989); *Phys. Rev.* **A 42**, 494 (1990).
7. J.J. Maki, M.S. Malcuit, J.E. Sipe, and R.W. Boyd, *Phys. Rev. Lett.* **67**, 972 (1991).
8. Nonlinear density matrix equations have been used earlier in quantum optics, see e.g. [2,3] and G.S. Agarwal, *Phys. Rev.* **A 4**, 1791 (1971); K. Molmer and Y. Castin, Ref. 3, p. 193.

In this letter we present a microscopic approach to the phenomena of coherent population trapping[1] in a dense medium[2,3] . In a dense medium we have the possibility of cooperative interactions and thus one can transfer excitation from one atom to the other via the dipole-dipole interaction[3,4]. Besides one also has the possibility of decay of an atom due to the presence of the other atoms. One obviously has to understand the effect of such cooperative interactions on the phenomena of coherent population trapping. Starting from a master equation[4] for the collective system of N lambda systems we demonstrate the existence of the coherent population trapping state even in presence of cooperative effects. However, the relaxation behavior is quite sensitive to such cooperative interactions.

Consider a system of N identical lambda systems (Fig. 1) located at the positions $\vec{r}_i$. The system is driven by two coherent fields $\vec{\varepsilon}_1$ and $\vec{\varepsilon}_2$ of frequencies $\omega_1$ and $\omega_2$. The fields interact with different transitions. The levels $|2\rangle$ and $|3\rangle$ do not decay. We will assume that the energy separation $\hbar\omega_R$ between $|2\rangle$ and $|3\rangle$ is very large compared to various detunings and Rabi frequencies. The master equation describing cooperative behavior is known from earlier works[4] which we adopt now. The master equation for our present system in the frame in which optical frequencies have been eliminated is

$$\frac{\partial \rho}{\partial t} = -i[H,\rho] + \Lambda\rho, \tag{1}$$

where H represents interaction with coherent fields and the liouville operator $\Lambda$ contains the effects of spontaneous emission and cooperative effects. Let $A_{\alpha\beta j}$ be the operators of the form $|\alpha\rangle\langle\beta|$ for $j^{th}$ atom, clearly $A_{13j}$, $A_{31j}$ are respectively the raising and lowering operators for the transition $|1\rangle \rightarrow \langle 3|$ in the $j^{th}$ atom. The mean values of the operators like $A_{11j}$ give the populations. Then H can be written as

$$\begin{aligned} H &= \sum_j H_j, \\ H_j &= \hbar\Delta_1 A_{11j} + \hbar(\Delta_1 - \Delta_2)A_{22j} \\ &\quad -\hbar(G_{1j}A_{13j} + G_{2j}A_{12j} + H.C.), \end{aligned} \tag{2}$$

where $\Delta' s$ are detunings as shown in the Fig. 1 and $G_{1j}(\equiv \frac{\vec{d}_{13}\cdot\vec{\varepsilon}_{1j}}{\hbar}), G_{2j}(\equiv \frac{\vec{d}_{12}\cdot\vec{\varepsilon}_{2j}}{\hbar})$ are the Rabi frequencies of the fields at the position of $j^{th}$ atom.

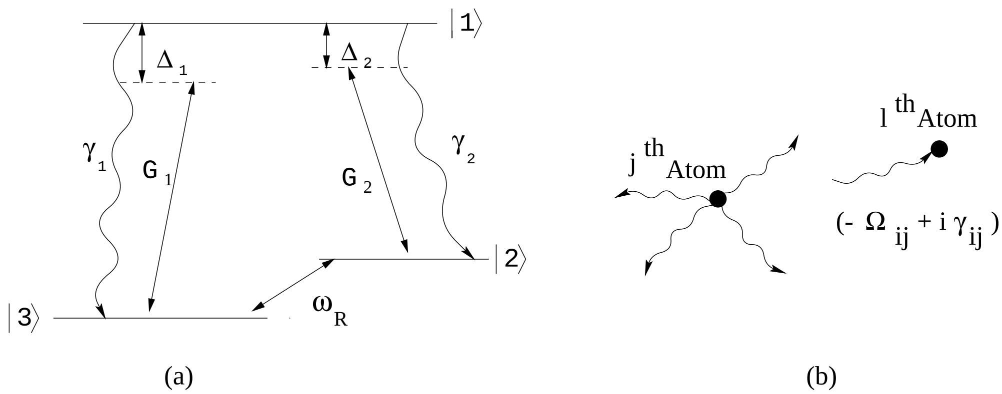


Fig. 1. Schematic diagram (a) of the system and its energy level and (b) of the interaction with the dipole field.

The operator $\Lambda$ has the structure

$$\begin{aligned} \Lambda\rho \equiv &-\sum_{jl} \gamma_{jl}^{(1)}(A_{13j}A_{31l}\rho - 2A_{31l}\rho A_{13j} + \rho A_{13j}A_{31l}) \\ &-\sum_{jl} \gamma_{jl}^{(2)}(A_{12j}A_{21l}\rho - 2A_{21l}\rho A_{12j} + \rho A_{12j}A_{21l}) \\ &-i\sum_{j\neq l} \Omega_{jl}^{(1)}[A_{13j}A_{31l},\rho] - i\sum_{j\neq l}\Omega_{jl}^{(2)}[A_{12j}A_{21l},\rho]. \end{aligned} \tag{3}$$

The superscripts 1 and 2 refer to the transitions $|1\rangle \rightarrow |3\rangle$ and $|1\rangle \rightarrow |2\rangle$. Here $2\gamma_{jj}^{(1)}$, $2\gamma_{jj}^{(2)}$ are the usual decay rates for $|1\rangle \rightarrow |3\rangle$, $|1\rangle \rightarrow |2\rangle$. The cross term $\gamma_{jl}$ represents cooperative decay and $\Omega_{jl}$ represents the retarded dipole-dipole interaction. These parameters are related to the field produced by an excited dipole, i.e., the field at the point $r_j$ produced by an excited dipole at the point $r_l$

$$-\Omega_{jl}^{(1)} + i\gamma_{jl}^{(1)} = \frac{1}{\hbar}\sum_{\kappa\mu}(\vec{d}_{13})_\kappa(\vec{d}_{13}^*)_\mu \chi_{\kappa\mu}(\vec{r}_j,\vec{r}_l,\omega_{13}), \tag{4}$$


Received June 27, 1997; Revised July 18, 1997

where

$$\chi_{\kappa\mu}(\vec{r}_1, \vec{r}_2, \omega) = (\frac{\omega^2}{c^2}\delta_{\kappa\mu} + \frac{\partial^2}{\partial r_{1\kappa}\partial r_{2\mu}})\frac{e^{i\frac{\omega}{c}|\vec{r}_1 - \vec{r}_2|}}{|\vec{r}_1 - \vec{r}_2|}. \tag{5}$$

It may be noted that Eq. (5) is the dyadic propagator for the electromagnetic field. The cooperative parameters are similarly defined for the other transition.

The previous work essentially considers a dilute system of atoms so that all $j \neq l$ terms in (3) can be ignored. We would now like to understand the effect of such terms on coherent population trapping phenomena. We search for a steady state solution of (1) under the condition $\Delta_1 = \Delta_2$. Note the very important stucture of cooperative interactions - these always have a combination of raising and lowering operators corresponding to the transition say $|1\rangle \rightarrow |3\rangle$ in $j^{th}$ atom and $|3\rangle \rightarrow |1\rangle$ in $l^{th}$ atom. Using this fact and some algebra we find that the steady state is

$$\rho_s = \prod_j |\psi\rangle_{jj}\langle\psi|, \tag{6}$$

where

$$|\psi\rangle_j = \frac{G_{2j}|3\rangle_j - G_{1j}|2\rangle_j}{(|G_{ij}|^2 + |G_{2j}|^2)^{1/2}}. \tag{7}$$

We thus find the remarkable result that the CPT state is independent of the cooperative effects. It should be borne in mind that we include only cooperative effects of *radiative* origin. We do not include collisions etc.

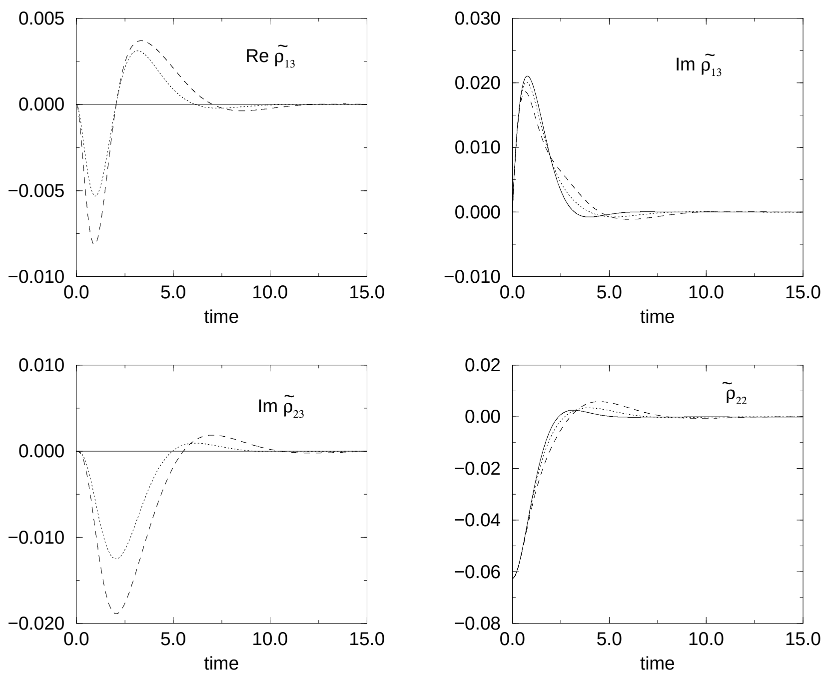


Fig. 2. Relaxation of various elements of the density matrix. We plot deviation $\tilde{\rho}$ from steady state ($\tilde{\rho} \equiv \rho - \rho_s$) for $G_1 = G_2 = \gamma_1 = \gamma_2 = 1$, $\Delta_1 = \Delta_2 = 0$, $\alpha_1 = \alpha_2$=0 (solid), 0.9 (dot) and 1.5 (dashed).

We next examine the dynamical behavior of the system. Note that we have a many body problem involving not only coherent interactions but also dissipative ones.

#2019 - $10.00 US
Received June 27, 1997; Revised July 18, 1997

In order to proceed further we will make *mean field approximation* as is very commonly done. We thus write

$$\rho \approx \prod_j \rho^{(j)}, \tag{8}$$

where $\rho^{(j)}$ is a single atom density matrix but does include the effects of other atoms in a mean field way. We substitute (8) in (1) and trace over all the atoms except say $j^{th}$ atom. We find the following eqution for $\rho^{(j)}$:

$$\begin{aligned}\frac{\partial \rho^{(j)}}{\partial t} &= -i[\tilde{H}^{(j)}, \rho^{(j)}] \\ &\quad -\gamma_{jj}^{(1)}(A_{13j}A_{31j}\rho - A_{31j}\rho A_{13j} + H.C.) \\ &\quad -\gamma_{jj}^{(2)}(A_{12j}A_{21j}\rho - A_{21j}\rho A_{12j} + H.C.),\end{aligned} \tag{9}$$

where $\tilde{H}^{(j)}$ is obtained from $H^{(j)}$ by using the replacements:

$$\begin{aligned} G_{1j} &\Rightarrow G_{1j} + \sum_{l\neq j}[-\Omega_{jl}^{(1)} + i\gamma_{jl}^{(1)}]\langle A_{31l}\rangle, \\ G_{2j} &\Rightarrow G_{2j} + \sum_{l\neq j}[-\Omega_{jl}^{(2)} + i\gamma_{jl}^{(2)}]\langle A_{21l}\rangle. \end{aligned} \tag{10}$$

Thus starting from our quantum master equation we show that within the mean field approximation the cooperative effects can be included by replacing the external field on the atom $j$ by the field consisting of the external field and the field radiated by all the other atoms. Recall that $-\Omega^{(1)} + i\gamma^{(1)}$ is essentially the propagater for the electromagnetic field. The evaluation of (10) is the standard problem of "local field effects"[5–7] in dense media. One here has the standard Lorentz result[5] that $E \to E + \frac{4\pi}{3}P$. In the mean field approximation one recovers from (9) *nonlinear* density matrix equations[8] (the index $j$ is now dropped)

$$\begin{aligned}
\dot{\rho}_{11} &= -2(\gamma_1+\gamma_2)\rho_{11} + iG_1\rho_{31} + iG_2\rho_{21} + c.c., \\
\dot{\rho}_{12} &= -\{\gamma_1+\gamma_2 - i[-\Delta_2 + \alpha_2(\rho_{22}-\rho_{11})]\}\rho_{12} + iG_1\rho_{32} \\
&\quad + iG_2(\rho_{22}-\rho_{11}) + i\alpha_1\rho_{13}\rho_{32}, \\
\dot{\rho}_{13} &= -\{\gamma_1+\gamma_2 - i[-\Delta_1 + \alpha_1(1-2\rho_{11}-\rho_{22})]\}\rho_{13} + iG_2\rho_{23} \\
&\quad + iG_1(1-2\rho_{11}-\rho_{22}) + i\alpha_2\rho_{12}\rho_{23}, \\
\dot{\rho}_{22} &= 2\gamma_2\rho_{11} - iG_2\rho_{21} + c.c., \\
\dot{\rho}_{23} &= i(-\Delta_1+\Delta_2)\rho_{23} - iG_1\rho_{21} + iG_2^* + i(\alpha_2-\alpha_1)\rho_{13}\rho_{21}.
\end{aligned} \tag{11}$$

where $\alpha' s$ are given in terms of the density $n$ of atoms

$$\alpha_1 = \frac{4\pi n|d_{13}|^2}{3\hbar},\ \alpha_2 = \frac{4\pi n|d_{12}|^2}{3\hbar}. \tag{12}$$

One can now study the relaxation characteristics of the system by using the usual method of linearization around the coherent population trapping state. For $G_1 = G_2$ we use replacements $\alpha_1\rho_{13}\rho_{32} \to -\frac{\alpha_1}{2}\rho_{13},\ \alpha_2\rho_{12}\rho_{23} \to -\frac{\alpha_2}{2}\rho_{12},\ \alpha_2(\rho_{22}-\rho_{11})\rho_{12} \to \frac{\alpha_2}{2}\rho_{12},\ \alpha_1(1-2\rho_{11}-\rho_{22})\rho_{13} \to \frac{\alpha_1}{2}\rho_{13},\ (\alpha_2-\alpha_1)\rho_{13}\rho_{21} \to 0$ and examine the eigenvalues of the resulting system of equations. The eigenvalues show interesting changes as the local field parameter $\alpha$ changes. The degeneracy of eigenvalues is lifted. In particular the degenerate eigenvalues -1+i for $\alpha_1 = \alpha_2 = 0,\ G_1 = G_2 = \gamma_1 = \gamma_2 = 1,\ \Delta_1 = \Delta_2 = 0$ go over to $(a) - 1.38 + 1.61i,\ -0.61 + 0.71i$ for $\alpha_1 = \alpha_2 = 0.9;\ (b) - 1.54 + 2.11i,\ -$

$0.45 + 0.61i$ for $\alpha_1 = \alpha_2 = 1.5$. For larger Rabi frequencies $G \gg \alpha$, the changes in the eigenvalues are negligible. This is in agreement with an earlier conclusion[6] in connection with two level systems that the local field effects are most pronounced in moderate fields $G \sim \gamma \sim \alpha$. The effect of a small perturbation on the time evolution is shown in Fig. 2. Only some elements of $\rho$ are shown. This figure clearly shows that the relaxation to coherent population trapping state is sensitive to cooperative interactions.

Thus in conclusion we have shown the generation of a coherent population trapping state even in the presence of cooperative interactions of radiative nature. The relaxation of the system is however sensitive to the cooperative interactions. We show the emergence of the local field effects from our master equation in the mean field approximation. Finally, it may be noted that our approach can in principle be used to examine effects beyond mean field approximation.

The author thanks R.P. Singh for computational help.