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HAL Id: jpa-00222589
https://hal.archives-ouvertes.fr/jpa-00222589
Submitted on 1 Jan 1983

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RESONANT INTRACAVITY PHASE CONJUGATION IN TWO AND THREE-LEVEL SYSTEMS

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Abstract: A general theory of intracavity phase conjugation, applicable to any resonance situation, is developed. It is applied to discuss the cases of two-level and A-type three-level systems. The phase-conjugate reflectivity exhibits bistability and hysteresis in both cases.

INTRODUCTION

Phase conjugation through resonant degenerate four-wave mixing (DFWM) is a subject of considerable interest /1-11/. Among other applications, the use of DFWM in high-resolution spectroscopy is now well recognized /3,4/. In the past attention was mainly paid to one-photon-resonant media /1-5/, modeled in terms of a two-level system /2/. More recently, two-photon processes have been considered /6-10/ since they allow extraction of spectroscopic information on dipole-forbidden transitions /6/. In the two-photon case the nonlinear medium is modeled in terms of either a two-level or a three-level system /11/ depending upon whether an intermediate energy level is far from or near to a resonance situation. Three-level systems, although they have attracted less attention /11-13/, may be useful for phase conjugation in view of their doubly resonant nature.

Recently, the intracavity DFWM configuration has been considered since it leads to critical behavior in phase conjugation /5,8,9/. Here the counter-propagating pump beams required for DFWM are obtained by placing the nonlinear medium inside an externally driven Fabry-Perot cavity. Under appropriate conditions, the phase conjugate reflectivity displays bistability and hysteresis as the incident beam intensity or the laser frequency is varied in a continuous manner. Bistability in DFWM can also arise without an external cavity /14-17/. We shall however restrict our discussion to the intracavity case only.

This paper presents a general formalism which treats resonant phase conjugation in a unified manner. It is developed without assuming a particular form of the medium response and is therefore applicable to any resonance situation, involving single-photon or multi-photon transitions, for which a steady-state nonlinear susceptibility has been found. Furthermore, intracavity phase conjugation can be readily treated since pump absorption is included within the mean-field approximation /5/. In contrast to the previous work /5,8,9/, the present analysis fully incorporates the standing-wave or spatial hole-burning effect arising from interference of the counter-propagating pump waves. In view of the potential spectroscopic applications, particular attention is paid to the intracavity DFWM spectrum. The results are presented for the case of a two-level as well as a three-level system.
In the intracavity DFWM geometry shown in Fig. 1 the counter propagating pump waves $E_1$ and $E_2$ are obtained by placing the nonlinear medium inside a Fabry-Perot cavity which is driven by an external field $E_0$. The probe wave $E_3$ and the conjugate wave $E_4$ propagate in opposite directions along the $z'$ axis which is assumed to make a small angle with the cavity axis. In the continuous-wave case the nonlinear interaction among the four waves is governed by the susceptibility $\chi$ obtained using the steady-state solution of the appropriate density-matrix equations. The particular form of $\chi$ depends on the details of matter-radiation interaction and for time being we leave it unspecified. Assuming that all waves have the optical frequency $\omega$ and are linearly polarized along the same direction, the total cavity field $E_c = \text{Re}[E \exp(-i\omega t)]$ satisfies the wave equation

$$\left(\nabla^2 + k^2\right)E = -k^2 \chi(E)E,$$

where $k = \omega/c$, $E = E_p + E'$ and

$$E_p = E_1 + E_2 = A_1 e^{ikz} + A_2 e^{-ikz},$$

$$E' = E_3 + E_4 = A_3 e^{ik'z} + A_4 e^{-ik'z}.$$  \hfill (2)

For arbitrary pump and probe intensities the nonlinear-propagation problem has to be solved numerically. Further analytical progress is possible if we assume intense pump beams and neglect their depletion. Assuming $|E'| \ll |E_p|$, we expand $\chi(E)$ around $E_p$ and retain the terms linear in $E_p$. Using Eq. (1) we obtain

$$\left(\nabla^2 + k^2\right)E_p = -k^2 \chi(E_p)E_p,$$

$$\left(\nabla^2 + k^2\right)E' = -k^2 \left[ \chi(E_p) (\partial \chi/\partial E_p) \right] E' + E_p (\partial \chi/\partial E_p) E'.$$  \hfill (4)

We now substitute Eqs. (2) and (3) in Eqs. (4) and (5) respectively, and make the paraxial and the plane-wave approximations. Expanding $\chi(E_p)$ and $\partial \chi/\partial E_p$ in a Fourier series and collecting the appropriate Fourier coefficients, we obtain

$$\frac{dA_1}{dz} = \frac{ik}{2} \left( \chi' + 4 \chi \right) A_1,$$

$$\frac{dA_2}{dz} = \frac{-ik}{2} \left( \chi' + 4 \chi \right) A_2,$$

$$\frac{dA_3}{dz} = -\alpha A_3 + ik \alpha^* A_1,$$

$$\frac{dA_4}{dz} = \alpha^* A_4 + i \kappa A_3,$$

where the saturated absorption coefficient $\alpha$ and the nonlinear coupling coefficient $\kappa$ are given by

$$\alpha = -\frac{ik}{2} (\chi_0 + \chi'),$$

$$\kappa = \frac{k}{2} \chi'' e^{i(\phi_1 + \phi_2)}.$$  \hfill (10)

$\phi_j$ being the phase of $A_j$. The Fourier components
\[ \chi_o = \langle \chi(E_p) \rangle, \chi_1 = \langle \chi(E_p) \cos \theta \rangle, \chi^* = \langle |E_p|^2 (\partial \chi / \partial E_p^2) \rangle, \tag{11} \]

where \( \theta = 2kz + (\phi_1 - \phi_2) \) is the relative phase between the pump waves. The angular brackets \( \langle \ldots \rangle \) denote spatial averaging over the standing-wave pattern arising from interference of the counter-propagating pump waves. In particular, \( \chi_1 \) accounts for the contribution of the population grating produced by the phenomenon of spatial hole-burning. In the previous work on intracavity phase conjugation\(^5,8,9\)/ the standing-wave effects were ignored by assuming an average pump intensity.

The set of Eqs.\((6)-(9)\) is solved subject to the boundary conditions given by

\[ A_1(o) = \sqrt{R_m} A_2(o) + \sqrt{1-R_m^t} E_0 \tag{12a} \]
\[ A_2(L_C) = \sqrt{R_m} A_1(L_C) \exp(2ikL_C) \tag{12b} \]
\[ A_3(z^* = 0) = E_3, \quad A_4(L^*) = 0, \tag{13} \]

where \( L_C \) is the cavity length, \( R_m \) is the mirror reflectivity, and \( L \) is the length of the nonlinear medium along the \( z^* \) axis. The objective is to obtain the phase-conjugate reflectivity

\[ R = |A_1(o)/A_2(o)|^2 \tag{14} \]
as a function of the incident field \( E_0 \) and other input parameters. Equations \((6)-(14)\) solve formally the problem of intracavity phase conjugation. Although pump depletion is neglected, they include pump absorption. It is important to realize that in the intracavity case pump absorption cannot be ignored because the same mechanism which gives rise to DFW also induces bistability in the pump fields through their nonlinear absorption. For a strongly absorbing medium, Eqs.\((6)-(9)\) have to be solved numerically. However, an analytical solution of them can be obtained for a weakly absorbing nonlinear medium placed inside a high finesse cavity.

**INTRACAVITY PUMP INTENSITY**

The nonlinear pump equations \((6)\) and \((7)\) subject to the boundary conditions \((12)\) can be readily solved if we assume that \( kL|x| \ll 1 \) and \((1-R_m) \ll 1 \). Under these conditions the forward and the backward pump intensities are approximately equal, \( |A_1|^2 = |A_2|^2 \), and do not vary significantly along the cavity length. Since the algebra is straightforward and the details can be found elsewhere \(/18/\), we write down the final result. Equations \((6)\) and \((7)\) lead to the optical-bistability state equation given by

\[ |E_o|^2 = T_m |A_1|^2 \left\{ \left[ 1 + (kL/T_m) \Im(X_0 + X_1) \right]^2 + \left[ \phi - (kL/T_m) \Re(X_0 + X_1) \right]^2 \right\}, \tag{15} \]

where \( T_m = (1 - R_m) \) is the mirror transmittivity and \( \phi \) is a measure of the cavity detuning in units of the cavity line width, i.e.,

\[ \phi = 2 (\omega_m - kL_C)/T_m = (\omega_C - \omega)/\gamma_C, \tag{16} \]

where \( \omega_C \) is the cavity resonance frequency closest to \( \omega \) and \( \gamma_C = cT_m/2L_C \) is the cavity line width, \( L_C \) being the cavity length. Note that Eq.\((15)\) represents a nonlinear algebraic equation for \(|A_1|^2\) since \( X_0 \) and \( X_1 \) themselves depend on the intracavity pump intensity \(|A_1|^2\). Under suitable conditions it can have multiple solutions for a given input intensity \(|E_0|^2\) and lead to optical bistability \(/18/\). Each solution should be examined for its stability.

**PHASE-CONJUGATE REFLECTIVITY**

The phase-conjugate reflectivity \( R = |A_1(o)/A_2(o)|^2 \) is obtained by solving Eqs.\((8)\) and \((9)\) together with the boundary conditions \((13)\). In the general case a numerical approach is required since the coefficients \( \alpha \) and \( \kappa \) vary with the distance through their dependence on the intracavity pump intensity. However, as mentioned earlier, when \( kL|x| \ll 1 \) and \( T_m \ll 1 \) the pump intensity is approximately constant. Equations \((8)\) and \((9)\) are then readily solved and the phase-conjugate reflectivity is given
Our general description of intracavity phase conjugation is now complete. For a
given functional form of \( \chi \) three spatial integrations are performed to obtain \( x_0, x_1 \) and \( x_2 \) given by Eq.(11). Equation (15) is then used to obtain the intracavity pump intensity \( |A_1|^2 \) through which \( \alpha \) and \( \kappa \) are known using Eq.(10). The use of \( \alpha \) and \( \kappa \) in
Eq.(17) yields finally the phase-conjugate reflectivity \( R \). We now apply this general
scheme to study phase conjugation in a two-level and a three-level system. We may
note that although the formalism is developed for the intracavity case, it is readily
applicable for the usual DFWM geometry. In the absence of the cavity Eq.(15) is
replaced by the trivial relation \( |A_1|^2 = |A_2|^2 = |E_0|^2 \) if the pump absorption is
neglected /2/.

TWO-LEVEL SYSTEM

Assuming that the nonlinear medium can be modeled in terms of a homogeneously broad-
dened two-level system, the nonlinear susceptibility under one-photon resonance
condition is given by /2,5/

\[
\chi(E) = \frac{2\alpha_0}{k} \left( \frac{1 + \Delta^2 + |E|^2}{1 + \Delta^2 + |E|^2} \right)
\]

where \( \alpha_0 = \frac{\alpha}{2} \) is the resonant small-signal field-absorption coefficient,
\( \Delta = (\omega_0 - \omega)T_i \) is the atomic detuning parameter, \( I_s = \hbar^2/\left(\mu^2 T_1 T_2\right) \) is the saturation intensity. The
atomic parameters \( \mu, \omega_0, T_1 \) and \( T_2 \) represent, respectively, the dipole moment, the
transition frequency and the longitudinal and transverse relaxation times. The spatial
integrals in Eq.(11) can be analytically performed. Using Eqs.(10) and (15) we obtain

\[
2I_0 = T_m I_p \left( \frac{(1+2C/I_p)(1-S)}{1+\Delta^2} \right)^2 \left( \frac{\phi-(2C\Delta/I_p)(1-S)}{1+\Delta^2} \right)^2
\]

\[
\alpha = \frac{\alpha_0}{1+\Delta^2} \left( \frac{1 + \Delta^2 + I_p}{1 + \Delta^2} \right) S^3
\]

\[
\kappa = \frac{\alpha_0}{1+\Delta^2} \left( \frac{I_p}{1+\Delta^2} \right) S^3
\]

where \( C = \alpha_0 L/T_m \) is the bistability parameter /18/ and

\[
S = \left[ 1 + 2I_p/(1 + \Delta^2) \right]^{-1/2}
\]

Furthermore, the input intensity \( I_0 = |E_0|^2/I_s \) and the total intracavity pump
intensity \( I_p = 2 |A_1|^2/I_s \) are measured in units of the saturation intensity \( I_s \).
The state equation (19) has been studied earlier in the context of optical bistabi-
lity /18/ and was found to have two stable solutions for \( I_p \) in a limited domain of
the multidimensional parameter space covered by \( C, \Delta, \phi \) and \( I_p \). Since the phase-
conjugate reflectivity \( R \) is a unique function of \( I_p \), it also exhibits bistability
and hysteresis in the same domain of the parameter space. Some of the qualitative
features of such a bistable behavior were studied /5/ after neglecting the standing-
wave effects. In order to compare with this previous work, Fig. 2 shows the reflec-
tivity behavior with and without inclusion of the standing-wave effects for a given
set of input parameters. The effect of spatial hole-burning is to decrease the
reflectivity, the hysteresis width, and the magnitudes of the switching intensities.
Although the simplified theory of Ref.5 captures the essential qualitative features,
the standing-wave effects should be included when comparing theory and experiment.
In view of the potential spectroscopic applications, it is of interest to investiga-
te how the intracavity nature of DFWM affects the reflectivity spectrum. In Fig. 3
the DFWM spectrum is shown for the cavity detuning \( \phi = 2 \). The spectral line exhibits
hysteresis whose width is only a fraction of the line width. Furthermore, it can be
controlled by varying the cavity length. An important point to note is that the bis-
table behavior occurs only in a detuned cavity. For \( \phi = 0 \) the bistable branch, al-
though it exists, can not be physically accessed, resulting in a symmetric single-
peak spectrum.

The present analysis can readily be extended to the case of a two-photon-resonant
medium. If the intermediate level is assumed to be far from resonance it can be
modeled in terms of an effective two-level system /19/. A new feature is the inten-
sity dependence of the ac Stark shift. The spatial integrals appearing in Eq. (11) can be analytically performed for this case as well. Since the qualitative features have been studied previously /8,9/ after neglecting the standing-wave effects, we refer to that work for further details.

THREE-LEVEL SYSTEM

We now consider intracavity phase conjugation in a nonlinear medium whose resonance behavior requires the use of a three-level system. Specifically, a doubly-degenerate lower level is assumed in exact one-photon resonance ($\omega = \omega_0$) with a common upper level. The degeneracy can be removed through the Stark or Zeeman splitting of the lower level. The resonance behavior is then modeled in terms of a $\Lambda$-type three-level system with the additional simplification that the laser frequency lies exactly in the middle of the two sublevels. Such a system has been shown to exhibit bistability /20/. A study of phase conjugation in this system /11/ reveals that significant values of the phase-conjugate reflectivity can be obtained at pump intensities well below saturation. Assuming that the sublevel-coherence decay time is very long, the nonlinear susceptibility is approximated by /11/

$$X(E) = \frac{21\alpha_0/k}{(1 + \delta^2 + \frac{3}{2} |E|^2/I_S)^{-1}},$$

where

$$\delta = \frac{\Omega_{21}T_2}{2},$$

and $\Omega_{21}$ is the sublevel-splitting frequency and other symbols have the meaning identical to the two-level case. Equation (23) shows that $X(E)$ is purely imaginary, a consequence of our assumption that the dipole moments for the two one-photon reso-

Fig. 2 - Illustrating the effects of spatial hole-burning on the phase-conjugate reflectivity: (a) before and (b) after including the standing-wave effects. The parameters used are $\Delta = 0$, $\phi = 0$, $\alpha_0L = 0.1$, and $R_m = 0.99$. The dashed portion of each curve is experimentally inaccessible.

Fig. 3 - The phase-conjugate spectrum for a two-level system using $I_0 = 1$, $\phi = 2$, $\alpha_0L = 0.2$, and $R_m = 0.99$. Hysteresis arises because the dashed portion is experimentally inaccessible.
nances are identical. The coupling between the two transitions is manifested through the intensity dependence of the effective detuning $\delta$. In particular $X$ vanishes when $\delta = 0$. This feature arises from a phenomenon called coherent population trapping and is sometimes also referred to as transverse optical pumping. It is a consequence of the two-photon-induced atomic coherence between the two sublevels. When the allowance is made for the finite decay time of this atomic coherence, $X$ no longer vanishes when $\delta = 0$, but nonetheless shows a dip whose magnitude and width is intensity dependent. The important point to note is that this nonlinear feature occurs well below the intensities required for the saturation of one-photon transitions.

After using Eq. (23) in (11) the three spatial integrals can be performed analytically even for a three-level system /11/. The resulting expressions for the bistability state equation and the coefficients $\alpha$ and $K$ are too cumbersome to reproduce here and can be found elsewhere /21/. Figure 4 shows the reflectivity behavior as a function of the splitting frequency at a fixed incident intensity $I_0 = 0.1$ in a tuned cavity. The reflectivity curve exhibits hysteresis on both sides of the $\delta = 0$ line. An interesting aspect is that when $\delta$ is varied in a given direction the line shape will appear asymmetric. However, the mirror image is obtained when $\delta$ is varied in the opposite direction.

Fig. 4 - The phase conjugate spectrum for a three-level system using $I_0 = 0.1, \phi = 0, \alpha_0 L = 0.1, R_m = 0.99,$ and $T_2/T_1 = 2.$

CONCLUSIONS

A general theory is developed to treat intracavity phase conjugation in a resonant nonlinear medium. It is then applied to discuss the cases of a two-level and a $\Lambda$-type three-level system. In both cases the phase-conjugate reflectivity exhibits bistability and hysteresis. Important differences between the two cases, however, exist in view of the different nonlinear mechanism which give rise to DFWM. Particular attention is paid to the reflectivity spectrum which, under appropriate input conditions, exhibits hysteresis whose width is only a fraction of the line width and can be externally controlled. The features discussed here should be easily observable. In a recent experiment on a $\Lambda$-type three-level system /22/ bistability was observed on the $D_1$ absorption line of Sodium using a Zeeman level of the ground state. It is expected that the bistable behavior shown in Fig. 4 can be observed in this system.

ACKNOWLEDGMENTS

The author wishes to thank M. Lax and C. Flytzanis for their interest in this work. The work at Quantel was supported by the Direction des Recherches, Etudes et Techniques (DRET). The research at the City College was partially supported by the U.S. Army Research Office.
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