

## Oscillator models for multiphoton optical bistability and phase conjugacy

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**Abstract.** Nonlinear oscillator models are constructed to treat the bistability in situations involving elementary excitations in solids. Such models are shown to be useful not only in describing single photon but also multiphoton bistability. The resulting bistability both with and without cavity is considered. The two-photon excitonic bistability in CuCl is in detail. The effect of local field corrections can also be incorporated, in a simple manner, in such models.

**Keywords.** Oscillator models; optical bistability; phase conjugacy.

### 1. Introduction

Oscillator models for explaining the various physical processes in solids have been quite successful because of their simplicity and the ease with which experimental data can be correlated with theoretical formulae (Barker and Loudon 1972; Garrett 1968). Besides it is possible to relate the parameters in the oscillator models to those in the more realistic microscopic theories. Some years back Barker and Loudon (1972) reviewed the most attractive features of such oscillator models. Loudon and coworkers also showed how various nonlinear interactions in solids such as Raman scattering and two-photon absorption by polaritons (Boggett and Loudon 1973) can be studied with great ease. In the last few years, one has discovered the remarkable phenomena of optical bistability first in atomic systems (Gibbs *et al* 1976) and later in semiconductors (Gibbs *et al* 1979; Miller *et al* 1979). Currently available theories are microscopic (Hanamura 1981, 1983; Koch and Haug 1981; Haug *et al* 1982; Sarid *et al* 1983; Steyn-Ross and Gardiner 1983) in nature and hence each case is to be examined separately. Therefore it would be desirable to investigate the phenomenological models based on the anharmonic oscillators. This would enable one to examine the optical bistability in a number of situations, say those involving molecular vibrations (*cf.* Flytzanis and Tang 1980) excitons, biexcitons, polaritons, plasmons etc. Moreover one can study the optical bistability not only when a single-photon transition is involved but also possible bistability in nonlinear processes such as Raman scattering and two-photon absorption (Agrawal and Flytzanis 1980; Agarwal 1980; Agarwal and Singh 1984).

In the present paper we consider how the multiphoton bistability can be discussed in terms of the coupled oscillator models. We also describe the related problem of phase conjugation (for reviews see Fisher 1982; Agarwal 1983). The organisation of this paper is as follows. In §2 we show how the standard bistability characteristics of an anharmonic oscillator are modified if such a system of anharmonic oscillator is contained in a ring cavity. Section 3 considers the case of a quadratic nonlinearity in a

single oscillator and in a system of two coupled oscillators. Such a system exhibits two-photon bistability, details of which are investigated in the context of excitonic bistability in CuCl. Section 4 deals with the bistability in the coupled oscillator model if one of the oscillator frequencies corresponds to three-photon resonance  $\omega_0 \approx 3\omega$ . The generation of phase conjugate signals is considered in §5. The general features of such a generation, which are independent of the specific model are given. Finally the effect of local field corrections (*cf.* Bedeaux and Bloembergen 1973; Bowden and Sung 1984) is briefly treated in the appendix.

## 2. Single-photon bistability in anharmonic oscillators contained in a ring cavity

In this section we consider the behaviour of anharmonic oscillators contained in a ring cavity. Consider for simplicity, a one-dimensional anharmonic oscillator interacting with the field  $\varepsilon \exp(-i\omega t) + \text{c.c.}$  The equation of motion is

$$\ddot{x} + \Gamma \dot{x} + \omega_0^2 x + gx^3 = \frac{e}{m} [\varepsilon \exp(-i\omega t) + \varepsilon^* \exp(i\omega t)]. \quad (1)$$

We assume that the applied field frequency  $\omega$  is close to the natural frequency  $\omega_0$ . From the theory of nonlinear oscillations, it is well known (see for example Morse and Ingard (1968) for the classical case and Drummond and Walls (1980) for certain quantum aspects) that the displacement  $x$  has a bistable behaviour for certain range of parameters *i.e.* in the long time limit one has

$$x(t) \simeq x \exp(-i\omega t) + \text{c.c.}, \quad (2)$$

$$x[1 + 3gD(\omega)|x|^2] = \frac{e}{m} \varepsilon D(\omega), \quad D(\omega) = (\omega_0^2 - \omega^2 - i\omega\Gamma)^{-1}. \quad (3)$$

Here  $x$  vs  $\varepsilon$  or  $x$  vs  $(\omega - \omega_0)$  exhibits bistable behaviour provided that  $2(\omega - \omega_0)/\Gamma > \sqrt{3}$ . Flytzanis and Tang (1980) have discussed this bistability explicitly for molecular vibrations. A question which we investigate is: what happens if such a system of anharmonic oscillators is contained in a ring cavity (Bonifacio *et al* 1981)? In that case the bistable characteristics will also depend on the nature of the cavity. In the slowly varying envelop approximation  $E = \varepsilon(z) \exp(ik\sqrt{\tilde{\varepsilon}}z - i\omega t) + \text{c.c.}$ , one can show that in the steady state

$$\frac{\partial \varepsilon}{\partial z} = \frac{2\pi i k n x}{\sqrt{\varepsilon}} + \frac{ik}{2\sqrt{\varepsilon}} (1 - \varepsilon) \varepsilon; \quad k = \omega/c, \quad (4)$$

where  $n$  is the density of anharmonic oscillators (number of excitations in the medium) and  $x$  is given by (3)  $\tilde{\varepsilon}$  denotes the linear dielectric function of the medium. Due to the feed back provided by the ring cavity, we have the boundary condition (Bonifacio *et al* 1981)

$$\varepsilon(0) = \sqrt{T} \varepsilon^{(i)} + R\varepsilon(l) \exp(-i\theta T), \quad (5)$$

where  $T = 1 - R$  and  $R$  is the reflectivity of the mirrors,  $\theta$  is the cavity detuning. In order to simplify further, we replace the field  $\varepsilon$  by its average, which is the mean field approximation of Bonifacio and Lugiato (1976). The mean field approximation is expected to be a good approximation since the sample size for semiconductor bistability is in micron range. Using (4) and (5), we find  $\varepsilon$  in terms of  $\varepsilon^{(i)}$  and  $x$  which is

then combined with (3). The resulting equation for  $x$  then becomes

$$x[1 + 3g D\mu |x|^2] = \frac{e}{m} \varepsilon^{(i)} \sqrt{T} D(\omega) [1 - R \exp(-i\theta T)]^{-1}, \quad (6)$$

where

$$\mu = 1 - \frac{i\omega l}{2c\sqrt{\varepsilon}} (1 - \varepsilon) [1 - R \exp(-i\theta T)]^{-1}. \quad (7)$$

The equation for  $x$  has the same form as in the absence of the cavity with the modifications

$$\begin{aligned} g &\rightarrow g\mu \\ \varepsilon^{(i)} &\rightarrow \sqrt{T} \varepsilon^{(i)} [1 - R \exp(-i\theta T)]^{-1}. \end{aligned} \quad (8)$$

Thus the nonlinearity coefficient  $g$  is changed to a new (complex) coefficient. This change could be quite significant depending on the nature of  $\varepsilon$ . This, one would expect due to the feedback provided by the cavity. The additional changes are because the model approximately takes into account the action of the medium on the field. The field transmitted  $\varepsilon^{(T)}$  by the cavity will be given by

$$\varepsilon^{(T)} = (\mu)^{-1} \sqrt{T} [1 - R \exp(-i\theta T)]^{-1} \left( \sqrt{T} \varepsilon^{(i)} + \frac{2\pi i \omega \varepsilon l x}{c\sqrt{\varepsilon}} \right). \quad (9)$$

We will present an explicit result in §3, which shows the effect of the cavity.

### 3. Two-photon bistability in coupled anharmonic oscillators

We will now show how anharmonic interactions could lead to two-photon bistability. For this purpose, we consider quadratic nonlinearity and write the equation of motion as

$$\ddot{x} + \Gamma \dot{x} + \omega_0^2 x + gx^2 = \frac{e}{m} 2\varepsilon \cos \omega t. \quad (10)$$

We look for steady-state solution of the form

$$x(t) = x \exp(-i\omega t) + z \exp(-2i\omega t) + \text{c.c.} \quad (11)$$

In this case coupled equations for  $x$  and  $z$  are

$$D^{-1}(2\omega)z + gx^2 = 0, \quad D^{-1}(\omega)x + 2gx^*z = \frac{e}{m} \varepsilon \quad (12)$$

and hence

$$x[1 - 2g^2|x|^2 D(\omega)D(2\omega)] = \frac{e}{m} \varepsilon D(\omega). \quad (13)$$

Thus the basic equation has the same structure as (3), except that now we have the two-photon resonant character. Keeping this resonant character in view, we can write approximately

$$x \left( 1 + \frac{2g^2|x|^2}{(\omega_0^2 - \omega^2) 2\omega_0 \Gamma(i - \delta)} \right) = \frac{e\varepsilon}{m(\omega_0^2 - \omega^2)}, \quad (14)$$

$$\delta = \frac{2}{\Gamma} (\omega_0 - 2\omega). \quad (15)$$

The response  $x$  as a function of  $\varepsilon$  will show a bistable character provided that  $-\delta > \sqrt{3}$  (Agarwal and Singh 1984).

In the above we have considered a very simple situation. In order to describe a more realistic situation, one may have to consider a two-oscillator model—such two-oscillator models have been previously used in connection with Raman scattering, two-photon absorption etc. Recently considerable work has been done on the two-photon bistability in CuCl (Hanamura 1981, 1983; Koch and Haug 1981; Haug *et al* 1982; Levy *et al* 1983; Sarid *et al* 1983; Peyghambarian *et al* 1983). In CuCl, the system absorbs photons, excitons are created and then in the second stage, excitons combine to form exciton molecules (Hanamura 1973). One can also resonantly enhance such a two-photon absorption process by tuning the incident frequency close to the excitonic frequencies. In order to describe such a situation adequately, we have to introduce a two-oscillator model with the two oscillators corresponding to excitons and excitonic molecules. Let us represent the exciton molecular oscillator by  $Q$  and excitons by  $x$ . The two are taken to interact by the potential  $gx^2Q$ . The dynamical equations for the two-oscillator model are

$$\ddot{x} + \Gamma_e \dot{x} + \omega_e^2 x = (q_e E/m_e) - (2g/m_e) x Q, \quad (16)$$

$$\ddot{Q} + \Gamma_b \dot{Q} + \omega_b^2 Q = \frac{q_b E}{m_b} - (g/m_b) x^2. \quad (17)$$

Assuming that the applied field frequency  $\omega$  is near the frequency  $\omega_b/2$ , the above equations can be approximately solved by writing

$$Q = Q_+ \exp(-2i\omega t) + \text{c.c.} + \text{other terms}, \quad (18)$$

$$x = x_+ \exp(-i\omega t) + \text{c.c.} + \text{other terms}. \quad (19)$$

The final equation for  $x$  is found to be

$$x_+ \left( 1 - \frac{2g^2}{m_e m_b} D_b(2\omega) D_e(\omega) |x_+|^2 \right) = \frac{q_e E}{m_e} D_e(\omega), \quad (20)$$

which has the same structure as (13) except now that we can account for the resonant enhancement of the two-photon absorption by choosing  $\omega \sim \omega_e$ . Within the framework of this model the induced polarization becomes

$$P = \chi^{(1)} \varepsilon + \chi^{(3)} \varepsilon |\varepsilon|^2 + \dots, \quad (21)$$

with

$$\begin{aligned} \chi^{(1)} &= (nq_e^2/m_e) D_e(\omega), \\ \chi^{(3)} &= (2g^2 q_e^4 n/m_e^4 m_b) D_e^2(\omega) D_b(2\omega) |D_e(\omega)|^2, \end{aligned} \quad (22)$$

where  $n$  is the density of excitons. The linear dielectric function of the medium will be  $\bar{\varepsilon} = \bar{\varepsilon}_\infty + 4\pi\chi^{(1)}$ , where  $\bar{\varepsilon}_\infty$  represents the non-resonant contribution to  $\varepsilon$ . A more exact expression for the intensity-dependent dielectric function can be obtained by writing (20) as

$$\begin{aligned} x_+ &= \frac{q_e E}{m_e} D_e(\omega) \left( 1 - \frac{2g^2}{m_e m_b} D_b(2\omega) D_e(\omega) |x_+|^2 \right)^{-1} \\ &\approx \frac{q_e E}{m_e} D_e(\omega) \left( 1 - \frac{2g^2 q_e^2 |\varepsilon|^2}{m_e^3 m_b} D_b(2\omega) D_e(\omega) |D_e(\omega)|^2 \right)^{-1}. \end{aligned} \quad (23)$$

We next examine what happens if such a system is contained in a ring cavity. We have in

place of (4):

$$\frac{\partial \varepsilon}{\partial z} = \frac{2\pi i k q_e n x_+}{\sqrt{\varepsilon}} + \frac{i k}{2\sqrt{\varepsilon}} (\tilde{\varepsilon}_\infty - \tilde{\varepsilon}) \varepsilon. \quad (24)$$

We solve (24) in the mean field approximation and use the boundary condition (5). This procedure leads to

$$\varepsilon = \left[ 1 - R \exp(-i\theta T) - \frac{i k}{2\sqrt{\varepsilon}} (\tilde{\varepsilon}_\infty - \tilde{\varepsilon}) l \right]^{-1} \left( \sqrt{T} \varepsilon^{(i)} + \frac{2\pi i k q_e n x_+ l}{\sqrt{\varepsilon}} \right), \quad (25)$$

$$\begin{aligned} x_+ & \left\{ 1 - \frac{2g^2}{m_e m_b} D_b(2\omega) D_e(\omega) |x_+|^2 \left[ 1 - \frac{i k l}{2\sqrt{\varepsilon}} (\tilde{\varepsilon}_\infty - \tilde{\varepsilon}) \right. \right. \\ & \left. \left. \times (1 - R \exp(-i\theta T))^{-1} \right] \right\} \\ & = \frac{q_e}{m_e} \sqrt{T} D_e(\omega) \varepsilon^{(i)} (1 - R \exp(-i\theta T))^{-1}. \end{aligned} \quad (26)$$

On making a series of scale transformations,

$$\chi^{(3)}/\chi^{(1)} = B(1 + i\eta), \quad \eta = \frac{\text{Im} \chi^{(3)}}{\text{Re} \chi^{(3)}}, \quad \beta = cT/\sqrt{\tilde{\varepsilon} l},$$

$$\psi = \sqrt{B} \frac{m_e}{q_e} D_e^{-1}(\omega) x_+, \quad T \ll 1, \quad (27)$$

we can reduce the above set to

$$\psi - (1 + i\eta) \psi |\psi|^2 \left[ 1 + \frac{i\omega_c}{2(1 + i\theta)\beta} \left( 1 - \frac{\tilde{\varepsilon}_\infty}{\tilde{\varepsilon}} \right) \right] = \frac{\varepsilon^{(i)} \sqrt{B}}{\sqrt{T} (1 + i\theta)}, \quad (28)$$

$$\begin{aligned} \sqrt{B} \varepsilon & = \left[ 1 + \frac{i\omega_c}{2\beta(1 + i\theta)} \left( 1 - \frac{\tilde{\varepsilon}_\infty}{\tilde{\varepsilon}} \right) \right]^{-1} \left\{ \frac{\varepsilon^{(i)} \sqrt{B}}{\sqrt{T} (1 + i\theta)} \right. \\ & \left. + \frac{i\omega_c}{2\beta(1 + i\theta)} \left( 1 - \frac{\tilde{\varepsilon}_\infty}{\tilde{\varepsilon}} \right) \psi \right\} \equiv \mathcal{F}, \quad |\mathcal{F}^{(i)}|^2 = \frac{|\varepsilon^{(i)}|^2 B}{T}, \end{aligned} \quad (29)$$

where  $\omega_c$  is the cavity frequency. Notice that if we had approximated polarization  $P$  by (21), then in place of (28) and (29) we would have obtained

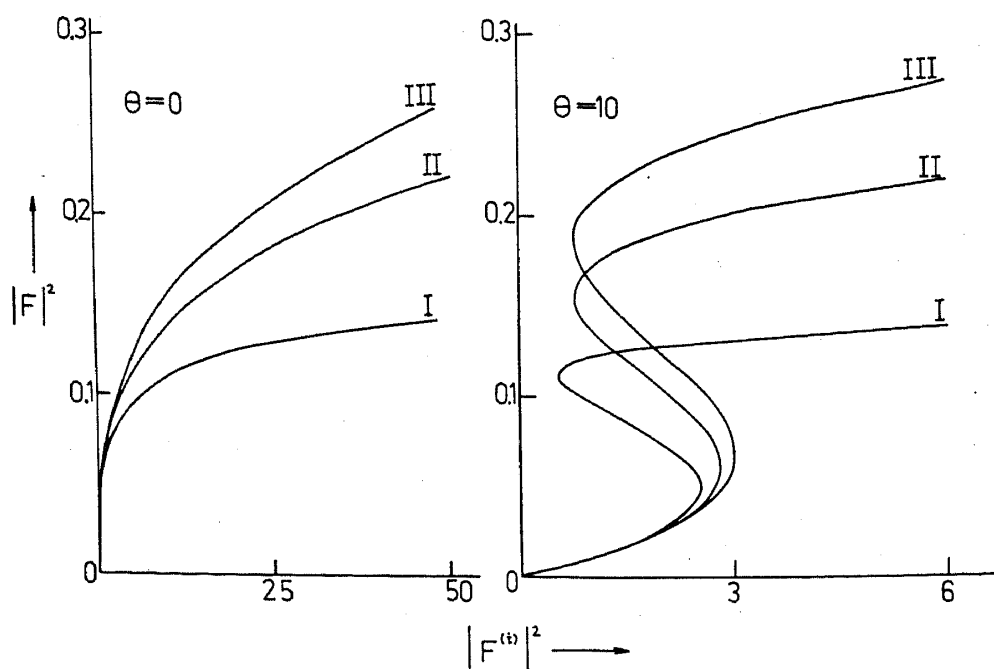
$$\mathcal{F} = \frac{\varepsilon^{(i)} \sqrt{B}}{\sqrt{T} (1 + i\theta)} + \frac{i\omega_c}{2\beta(1 + i\theta)} \left( 1 - \frac{\tilde{\varepsilon}_\infty}{\tilde{\varepsilon}} \right) (1 + i\eta) \mathcal{F} |\mathcal{F}|^2, \quad (30)$$

which is the form used by Haug *et al* (1982). A more exact model which is analogous to (23) of Haug *et al* (1982) leads, in place of (30), to the following state equation

$$\mathcal{F} = \frac{\varepsilon^{(i)} \sqrt{B}}{\sqrt{T} (1 + i\theta)} + \frac{i\omega_c}{2\beta(1 + i\theta)} \left( 1 - \frac{\tilde{\varepsilon}_\infty}{\tilde{\varepsilon}} \right) \frac{(1 + i\eta) \mathcal{F} |\mathcal{F}|^2}{(1 - (1 + i\eta) |\mathcal{F}|^2)}. \quad (31)$$

We next solve (27) to (30) numerically using the parameters appropriate to CuCl

$$\tilde{\varepsilon} = \tilde{\varepsilon}_\infty + \frac{(\tilde{\varepsilon} - \tilde{\varepsilon}_\infty) \omega_e^2}{\omega_e^2 - \omega^2}, \quad (32)$$



**Figure 1.** Two-photon optical bistability in the framework of two-oscillator model with parameters appropriate to the two photon absorption in CuCl. The curves I-III refer to the exact model (equation (29)) and the approximate models (equations (31) and (30)). The other parameters have been chosen as  $T = 0.1$ ;  $\theta = 10$ .

$\hbar\omega_e = 3.2027$  eV,  $\bar{\epsilon}_\infty = 5$ ,  $\bar{\epsilon}_0 = 1.003437 \epsilon_\infty$ , and choosing  $\hbar\omega = 3.18591$  eV. Note that the parameter

$$\eta = \frac{\Gamma_b}{2(\omega_b - 2\omega)}, \quad \hbar\omega_b = 6.3725 \text{ eV}$$

and thus  $\eta$  depends on the damping. Take  $\eta = 0.1$ , we get the results shown in figure 1. For  $\theta = 0$ , the output  $\mathcal{F}$  vs  $\mathcal{F}^{(i)}$  is a monotonic function. The system becomes bistable if the cavity is detuned sufficiently. The exact and the approximate models have qualitatively similar behaviour except that the bistability thresholds (maxima and minima) are shifted which suggests that for a given  $\mathcal{F}^{(i)}$  the dynamic properties will be quite different in various models. The transmission is considerably less on the upper branch in the exact model for a given value of the input field.

#### 4. Three-photon bistability in a system of anharmonic oscillators

In this section we demonstrate how the response  $x(t)$  at the applied frequency  $\omega$  of a system of coupled oscillators can have a three-photon resonance  $\omega_0 \sim 3\omega$  and how such a response can exhibit bistability under suitable conditions on the parameters. For this purpose consider, as in §3, two coupled oscillators with amplitude  $x$  and  $Q$  with anharmonic interaction  $V = (g_x/4)x^4 + g_Q x^3 Q$ . Assume that the frequencies  $\omega_Q$  and  $\omega_x$  of the oscillators are such that  $\omega_x \sim \omega$ ,  $\omega_Q \sim 3\omega$ . Keeping the resonant approximation in view, we can write

$$\ddot{x} + \Gamma_x \dot{x} + \omega_x^2 x + g_x x^3 + 3g_Q x^2 Q = \frac{eE}{m} : \ddot{Q} + \Gamma_Q \dot{Q} + \omega_Q^2 Q + g_Q x^3 \approx 0. \quad (33)$$

Writing further

$$x(t) = x \exp(-i\omega t) + \text{c.c.} + \dots, \quad Q(t) = Q \exp(-3i\omega t) + \text{c.c.} + \dots \quad (34)$$

and concentrating on the steady-state response we get

$$\begin{aligned} D_x^{-1}(\omega) x + 3g_x |x|^2 x + 3g_Q x^2 Q &= e\varepsilon/m, \\ D_Q^{-1}(3\omega) Q + g_Q x^3 &= 0, \end{aligned} \quad (35)$$

which on simplification leads to

$$x [1 + 3g_x D_x(\omega) |x|^2 - 3g_Q^2 |x|^4 D_x(\omega) D_Q(3\omega)] = \frac{e}{m} D_x(\omega) \varepsilon. \quad (36)$$

Note that (36) gives the following Taylor series expansion for the oscillator polarization

$$ex = \alpha_1 \varepsilon + \alpha_3 |\varepsilon|^2 \varepsilon + \alpha_5 |\varepsilon|^4 \varepsilon + \dots, \quad (37)$$

$$\alpha_1 = \frac{e^2}{m} D_x(\omega), \quad \alpha_3 = \frac{-3g_x D_x(\omega)}{e^2} |\alpha_1|^2 \alpha_1,$$

$$\alpha_5 = \frac{-3\alpha_1}{e^4} |\alpha_1|^4 D_x [g_x^2 (2D_x(\omega) + D_x^*(\omega)) + g_Q^2 D_Q(3\omega)]. \quad (38)$$

Note that the three-photon resonance is contained in the fifth order polarizability  $\alpha_5$ . We now discuss the bistability character that follows from (36). For three-photon resonance  $\omega_0 \sim 3\omega$  and hence we assume  $D_x(\omega)$  to be real. In terms of the scaled variable  $\mathcal{F}$

$$|\mathcal{F}|^2 = 3g_x D_x(\omega) |x|^2, \quad \mathcal{F}^{(i)} = \frac{e\varepsilon}{m} D_x^{3/2}(\omega) (3g_x)^{1/2}, \quad (39)$$

we can write (36) as

$$\mathcal{F} [1 + |\mathcal{F}|^2 - (p_1 + ip_2) |\mathcal{F}|^4] = \mathcal{F}^{(i)} \quad (40)$$

$$p_1 + ip_2 = (g_Q^2/3g_x^2) D_Q(3\omega)/D_x^*(\omega),$$

$$\delta = 2(\omega_0 - 3\omega)/\Gamma. \quad (41)$$

In order to see the resulting behaviour of  $\mathcal{F}$  as a function of  $\mathcal{F}^{(i)}$ , we rewrite (40) as

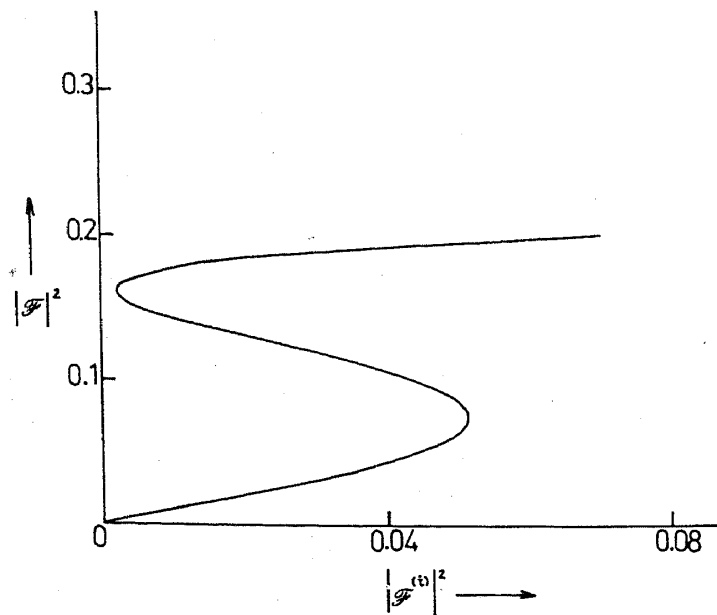
$$|\mathcal{F}^{(i)}|^2 = \phi \{ (1 + \phi - p_1 \phi^2)^2 + p_2^2 \phi^4 \}, \quad \phi = |\mathcal{F}|^2 \quad (42)$$

and look for the maxima and minima of  $|\mathcal{F}^{(i)}|^2$  as a function of  $\phi$ :  $\partial |\mathcal{F}^{(i)}|^2 / \partial \phi = 0$ . These extrema are given by

$$5(p_1^2 + p_2^2) \phi^4 - 8p_1 \phi^3 + 3(1 - 2p_1) \phi^2 + 4\phi + 1 = 0. \quad (43)$$

The roots of (43) can be investigated graphically by plotting  $4\phi + 1$  and  $5(p_1^2 + p_2^2) \phi^4 - 8p_1 \phi^3 + 3(1 - 2p_1) \phi^2$ . Using this one can now show that the conditions for the existence of bistability in the model (40) are

$$5(p_1^2 + p_2^2) \psi^4 - 8p_1 \psi^3 + 3(1 - 2p_1) \psi^2 + 4\psi + 1 < 0,$$



**Figure 2.** Three-photon bistability exhibited by the coupled oscillator system *i.e.*, the excitation amplitude  $|E|$  (equation (40)) at the fundamental frequency  $\omega$  as a function of the drive. The parameters are chosen so that the conditions (44) are satisfied.

$$\psi = \frac{1}{5(p_1^2 + p_2^2)} [3p_1 + \{9p_1^2 - \frac{15}{2}(1 - 2p_1)(p_1^2 + p_2^2)\}^{1/2}], p_1 > 0. \quad (44)$$

In figure 2 we display the bistability that follows from (40). The parameters have been chosen so as to satisfy (44).

### 5. Multiphoton bistability and phase conjugacy

We now discuss the generation of the phase conjugate signals in the context of the multiphoton bistability in nonlinear oscillators. It is known (*cf* Fisher 1982) that the systems possessing non-zero  $\chi^{(3)}$  give rise to the phase conjugate signals, for example a system interacting with two pump fields  $\exp(ik \cdot r - i\omega t)$ ,  $\exp(-ik \cdot r - i\omega t)$  and a signal field  $\exp(ik^1 \cdot r - i\omega t)$  gives rise to the conjugate field  $\exp(-ik^1 \cdot r - i\omega t)$ . Flytzanis and Tang (1980) discussed many interesting features of the conjugate fields under the conditions that the pump fields were strong enough so that the system had bistable behaviour. In what follows we concentrate our attention on the degenerate four-wave mixing. We thus investigate the change in  $x$  due to a change  $\delta\varepsilon$  in  $\varepsilon$ . In the foregoing sections we have shown that the steady-state behaviour of the system irrespective of the nature of the underlying multiphoton absorption process has the form

$$xf(|x|^2) = \beta\varepsilon, \quad (45)$$

where  $f$  is a complex function. The applied field  $\varepsilon$  is also complex. Using (45) and systematic Taylor series expansion, we find

$$\begin{pmatrix} \delta x \\ \delta x^* \end{pmatrix} = \beta [ |f|^2 + |x|^2 (ff'^* + f^*f') ]^{-1} \\ \times \begin{pmatrix} f^* + |x|^2 f'^* & -x^2 f' \\ -x^{*2} f'^* & f + |x|^2 f' \end{pmatrix} \begin{pmatrix} \delta\varepsilon \\ \delta\varepsilon^* \end{pmatrix}. \quad (46)$$



The coefficient  $S_c$  responsible for the generation of the phase conjugate signal is

$$S_c = \delta x / \delta \varepsilon^* = -\beta x^2 f' / \lambda_1 \lambda_2, \quad (47)$$

$$\lambda_1 \lambda_2 = [ |f|^2 + |x|^2 (ff'^* + f^*f') ], \quad (48)$$

where  $\lambda_1$  and  $\lambda_2$  are the eigenvalues of the so-called relaxation matrix. It is very generally known (Bonifacio *et al* 1981; Agarwal and Shenoy 1982) that near the bistability threshold (turning points in figures 1, 2) one of the eigenvalue, say,  $\lambda_1 \rightarrow 0$  as  $(|\varepsilon|^2 - |\varepsilon_c|^2)^{1/2}$ . Hence the phase conjugate signal for pump intensities close to such critical values corresponding to the bistability threshold will be very large. These conclusions are independent of the underlying multiphoton process.

We have so far presented the results for the steady-state behaviour though the questions regarding the transients, switching times etc can also be examined in the context of such models. We have also not discussed the stability of such states. Such questions obviously need to be examined at length. We however expect that only the middle portion of the *s*-shaped bistability curve to be unstable.

Thus in conclusion we have demonstrated how nonlinearly-coupled oscillator models can be used to treat the multiphoton bistability in a number of situations involving elementary excitations in solids.

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### Appendix

#### Comments on local field corrections

In this appendix we briefly comment on the effect of the local field corrections. We will show how the local field corrections lead to a renormalization of the various parameters. The effect of such corrections on the optical bistability in CuCl has been discussed recently (Bowden and Sung 1984). On writing the local field  $\varepsilon_l$  as

$$\varepsilon_l = \varepsilon + sP, \quad (A1)$$

the new susceptibilities  $\tilde{\chi}^{(3)}$ ,  $\tilde{\chi}^{(1)}$  are known to be (cf Bedeaux and Bloembergen 1973)

$$\tilde{\chi}^{(1)} = (1 - ns \alpha^{(1)})^{-1} n \alpha^{(1)}, \quad (A2)$$

$$\tilde{\chi}^{(3)} = (1 - ns \alpha^{(1)})^{-4} n \alpha^{(3)}. \quad (A3)$$

For the two-oscillator model discussed in §4, these renormalized susceptibilities—which incorporate the effects of local field corrections are given by (we choose for simplicity  $\tilde{\varepsilon}_\infty = 1$ )

$$\tilde{\chi}^{(1)} = q_e^2 \frac{n}{m_e} \tilde{D}_e(\omega) \quad (A4)$$

$$\tilde{\chi}^{(3)} = \frac{2g^2 q_e^4 n}{m_e^4 m_b} D_b(2\omega) \tilde{D}_e^2(\omega) |\tilde{D}_e(\omega)|^2, \quad (A5)$$

where

$$\tilde{D}_e^{-1}(\omega) = D_e^{-1} - (nsq_e^2/m_e). \quad (\text{A6})$$

Thus the effect of the local field corrections is to change the frequency  $\omega_e$  of the excitonic oscillator:  $\omega_e^2 \rightarrow \omega_e^2 - (nsq_e^2/m_e)$ , oscillator strength and the nonlinearity  $g$ . Hence it is evident that if in the calculations, we use the experimentally observed parameters such as  $\omega_e$ , then the local field corrections are automatically included and there is no further need to account for these.

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