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Transient Resonant Spiking in Degenerate Four Wave Mixing in Saturable Absorbers

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Abstract.

We show that under certain conditions the time evolution of the conjugate reflectivity for degenerate four wave mixing in saturable absorbers can exhibit a resonant spike after a time equal to the molecular relaxation time of the system.

Silverberg and Bar-Joseph have developed a model for transient degenerate four wave mixing (DFWM) in saturable absorbers [1]. The model is an extension of the steady state model of Abrams and Lind [2] and it gives the conjugate reflectivity as a function of time when the pump beams consists of pulses of width much longer than the molecular decay time of the system. The model has recently being extended to the case where the pump beams consist of pulses of arbitrary width, in particular comparable to or much shorter than the molecular decay time [3]. The model is in good agreement with experiment [1]. It predicts an enhanced transient reflectivity which has been observed experimentally [1,4]. We now show that this model also predicts a transient resonant spiking in the time evolution of the conjugate reflectivity. The difficulties that would be faced in observing this transient spiking experimentally are also pointed out.

We consider degenerate four wave mixing in a three level saturable absorber. The saturable absorber has a fast 2-3 transition and a relatively long lifetime τ of level 3. This includes most of the organic dye saturable absorbers and many solid state systems [5]. If n_0 is the total population, then during the pulse the gound state population, n, evolves according to [1]

$$\frac{dn}{dt} = -\frac{\sigma n I}{\hbar\omega} + \frac{(n_0 - n)}{\tau} \tag{1}$$

where I is the intensity and ω is the frequency of the radiation and σ the absorptive cross section for the transition $1 \rightarrow 2$.

With the usual DFWM notation we consider four plane waves of the form [1,3]

$$E_i(\underset{\sim}{r}_i, t) = \frac{1}{2} A_i(\underset{\sim}{r}_i) \exp[i(\omega t - \underset{\sim}{k}_i \cdot \underset{\sim}{r}_i)] + \text{c.c.} \tag{2}$$

where E_1 and E_2 are two strong counter-propagating pump fields and E_3 and E_4 are two weak fields also counter-propagating but in a different nck chi

direction from E_1 and E_2. These fields interact through the non-linear susceptibility χ of the medium. In the case where the pump beams consist of rectangular pulses of width τ_p, Silverberg and Bar-Joseph have shown that, in the non-depleted pump approximation and for $\tau_p >> \tau$, the conjugate reflectivity is given by [1]

$$R = \frac{|A_3|^2}{|A_4|^2} = \frac{|\kappa \sin WL|^2}{|W \cos WL + \alpha \sin WL|^2} \tag{3}$$

where

$$W = (|\kappa|^2 - \alpha^2)^{1/2} \tag{4}$$

and the absorption coefficient α and coupling coefficient κ are given by

$$\alpha = \alpha_0 \left\langle \left\{ \frac{1 + [P(2+P) - \frac{t}{\tau} P^2(1+P)] \exp(-\frac{t}{\tau}(1+P))}{(1+P)^2} \right\} \right\rangle \tag{5}$$

and

$$\kappa^* = i\alpha_0 \left\langle \frac{P\{1 - [1 - \frac{t}{\tau} P(1+P)] \exp(-\frac{t}{\tau}(1+P))\}}{(1+P)^2} \right\rangle \tag{6}$$

Here $\alpha_0 = \frac{1}{2}\sigma n_0$, $P = 4(I/I_s)^2 \cos^2\theta$ where $I_s = \hbar\omega/\sigma\tau$ is the saturation intensity and the angular brackets < > denote averaging over the phase term $\theta = \underset{\sim}{k}_i \cdot \underset{\sim}{r}_i$. In obtaining the above results it is assumed that (a) the transverse relaxation time of the medium is much less than the pulse duration, (b) it is an optically thin saturable absorber and (c) the transition is on resonance [1,3].

The above model has been found to be in good agreement with experiment [1]. It predicts an enhanced transient conjugate reflectivity which has been observed and found to be in good agreement with experiment [1,4

We will now show that this model also predicts the possibility of transient resonant spiking in the time evolution of the conjugate reflectivity after a time equal to the molecular relaxation time of the absorber.

Suppose that the intensity of the pump beams is chosen so that $P = 1$. From equations (5) and (6) we see that after a time $t = \tau$, $|\chi| = \alpha$ i.e. $w = 0$ instantaneously at $t = \tau$. Hence $R = \infty$ at $t = \tau$ (equation (3)). This transient resonant spiking is seen clearly in figure (1) where R is shown as a function of time for $P = 1$, $\alpha_o L = 0{\cdot}5$ and $\tau_\rho/\tau = 1000$. Note that the resonance is truely transient since it is only after a time τ that $|\chi|$ can equal α and from equations (5) and (6) this can only occur when $P = 1$.

Great care would have to be taken in order to observe this spiking experimentally. Apart from having to ensure that $P = 1$, thermal and off-resonant effects would tend to broaden and flatten the resonant spike. Furthermore, any experimental apparatus measures the average value of the conjugate field over some time interval and as a result the spike could be lost in the transient enhancement on which it sits. Any experiment designed to see transient resonant spiking in the conjugate reflectivity would have to ensure that the measuring apparatus averages over time intervals much shorter than the width of the spike.

In conclusion we predict the possibility of transient resonant spiking in the time evolution of the conjugate reflectivity for DFWM in saturable absorbers. This spike occurs after a time equal to the molecular relaxation time of the absorber.

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Figure Captions.

Figure 1. Time evolution of the conjugate reflectivity when P = 1, $\alpha_0 L = 0{\cdot}5$ and $\tau_\rho/\tau = 1000$.

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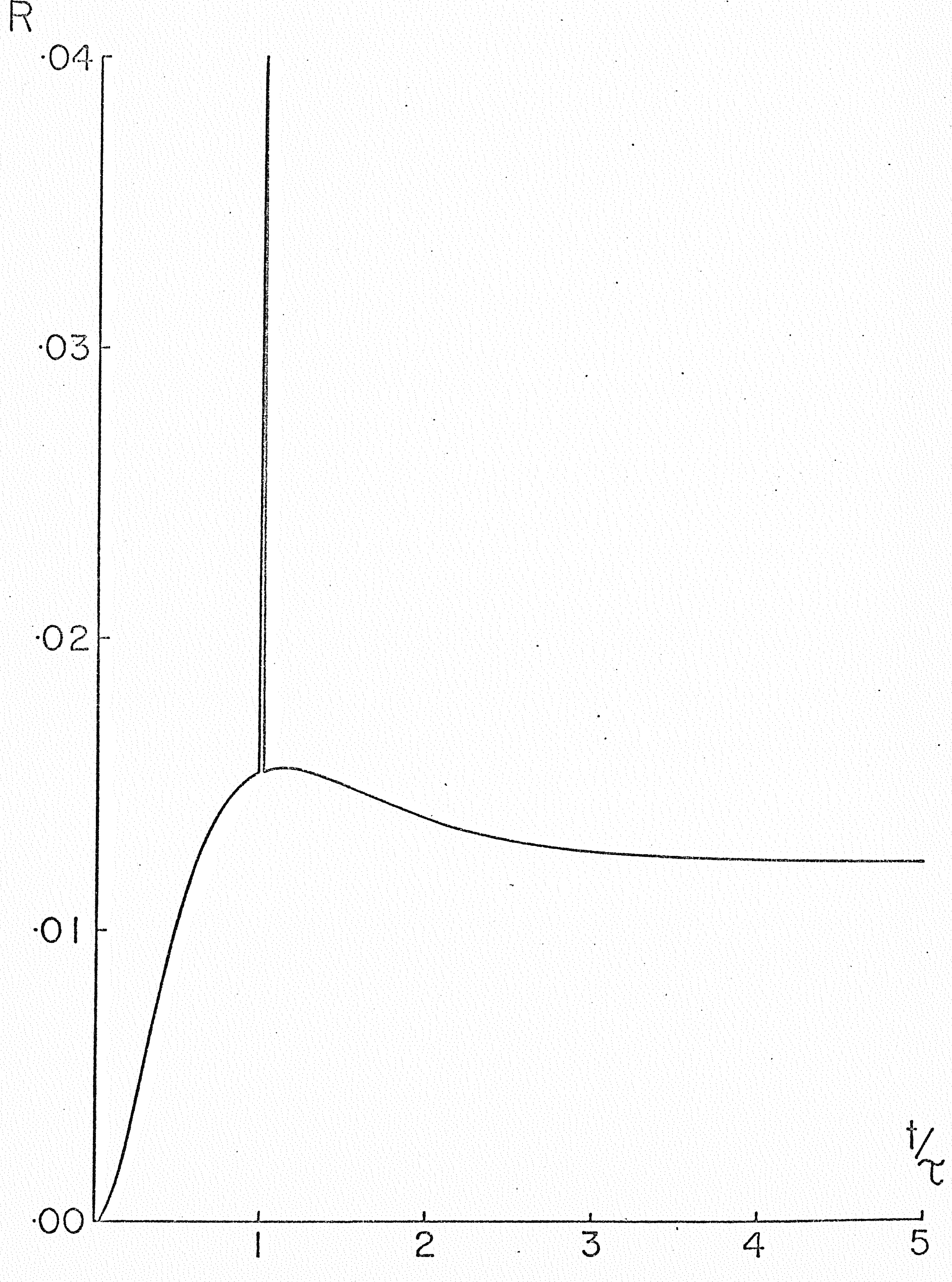

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