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THEORY OF SUB-COOPERATION-LIMIT OPTICAL PULSES IN RESONANT ABSORBERS*

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ABSTRACT

Every resonant dielectric absorber, near enough to one of its resonances to be considered composed of two-level atoms, is characterized by a certain time, which we denote τ_a and call the "cooperation time". The effects arising from this cooperation time have been studied very little and only qualitatively for the most part. Results of a theoretical study with R. A. Marth of resonant absorbers which are interacting with optical pulses shorter than τ_a will be described.

It has been known for some time that the interaction of spins or two-level atoms with a classical radiation field can depend in an interesting way on the existence of a certain coherence time τ_a, defined by[1,2]

$$\frac{1}{\tau_a^2} = \frac{\pi}{2}\mathcal{N}\hbar\omega\,\kappa^2 \qquad (1)$$

Here $\mathcal{N}$ and ω are the density and transition frequency of the atoms or spins, and $\hbar\kappa/2$ is the dipole matrix element of the transition. Following Arecchi and Courtens,[1] I will call τ_a the "cooperation time" for the system of atoms.

For my purposes here, the significance of τ_a is that it establishes a three-part time scale for the interaction of atoms and pulses of radiation. Let's see how that happens. Clearly a pulse might be either longer or shorter than τ_a itself. But the mere coexistence of a lot of atomic dipoles, necessary for τ_a to be defined, necessarily also implies dipole-dipole forces which will act to interrupt dipole-pulse coherence. The associated incoherent interruption time T is easily shown to be related to τ_a:

$$T = \omega\tau_a^2 \; . \qquad (2)$$

Thus there are three pulse length regimes to consider: Pulses longer than both τ_a and T, pulses shorter than both, and pulses with intermediate lengths.

It is easy to show[1] that pulses of the sort encountered in self-induced transparency[3] must be of the intermediate variety. That is,

$$\omega\tau_a^2 > \tau_{sit} > \tau_a \qquad (3)$$

must be satisfied by s.i.t. pulse lengths τ_{sit}.

Now we all understand that unless some Maxwell demon can be recruited to organize the chaotic dipole-dipole interactions, all coherent pulses, not just those of the s.i.t. type, will have to be shorter than $\omega\tau_a^2$. It is the right-hand inequality in (3) that interests me here. Let me begin by writing a more extended chain of times:

$$\omega\tau_a^2 > \tau_{sit} > \tau_a > \tau_{scl} > \omega^{-1} \quad , \tag{4}$$

where τ_{scl} is the length of a hypothetical "subcooperation-limit" pulse. The last two inequalities make it clear that I imagine such subcooperation-limit pulses to be shorter than the cooperation time τ_a, but also long enough to contain a number of cycles of the carrier.

In fact, in order to sustain my two-level atom assumption, I must require the s.c.l. pulses to satisfy

$$\tau_a > \tau_{scl} >> \omega^{-1} \quad . \tag{5}$$

In other words, I imagine subcooperation-limit pulses which are nevertheless slowly-varying pulses.

The first point to settle is the size of the domain left open for s.c.l. pulses. In order for an s.c.l. pulse to be slowly varying, containing at least hundreds of cycles of the carrier, it is necessary that $\tau_{scl} \gtrsim 0.1$ psec. The remaining question is the size of τ_a. As it turns out,[2,4] for many common resonant optical absorbers, (Ruby and alkali metal vapors, for example), $\tau_a \sim 0.1$ nsec.

Thus one finds a three-order-of-magnitude range of pulse lengths available for slowly-varying subcooperation-limit pulses:

$$0.1 \text{ nsec} > \tau_{scl} > 0.1 \text{ psec} \quad , \tag{6}$$

a range comparable to that available for optical self-induced transparency pulses:

$$0.1\ \mu\text{sec} > \tau_{sit} > 0.1 \text{ nsec} \quad .$$

Bob Marth and I have taken the point of view that it should be interesting, and perhaps important, to understand these subcooperation-limit pulses. They are obviously within the ballpark of current experimental capability. We have approached a study of s.c.l. pulses with the idea of seeing in what ways they may be the same as, or different from, the familiar longer s.i.t. pulses which have been studied in great detail lately by Gibbs and Slusher.[5]

Let me borrow some results from Bob's thesis research[4] to show some of the answers he's found to the questions we've raised. Let me mention first two temporary but important assumptions made. Since $\tau_{scl} < \tau_a$ by definition, and since $T_2^* \sim \tau_a$ in many systems of interest, he assumes all interesting pulse lengths τ are enough shorter than T_2^* to make inhomogeneous broadening superfluous for steady-state propagation. Furthermore, for simplicity here, we assume that the pulse carrier frequency and the common atomic resonance frequency are identical. (This latter assumption by itself is removed in Ref. 4. As I'll mention below, the first assumption can also be removed.)

The important experimental quantities associated with steady-state pulses can be identified as follows. The electric field strength is written:

$$\vec{E}(t, z) = \mathscr{E}(t-z/V)\, \mathrm{Re}\left\{(\hat{x}+i\hat{y})\, e^{i\Phi(t, z)}\right\}, \tag{8}$$

where $\mathscr{E}$ is the real steady-state amplitude, and the phase Φ is made up of carrier and pulse contributions:

$$\Phi(t, z) = \omega t - Kz + \phi(t-z/V) \quad . \tag{9}$$

Because of the strong field-atom interaction, the pulse velocity V may be quite different from c, and the wave vector K may differ from ω/c. Note that the instantaneous frequency $d\Phi/dt$ is allowed to differ from ω; that is, $\dot{\phi}$ may be a function of time, leading to the possibility of chirping.

In Figs. 1 and 2 (taken from Ref. 4) are shown V and $\dot{\phi}$ as a function of pulse length τ for four different optically resonant absorbers. The velocity curve is relatively unexciting, since when $\tau < \tau_a \sim 10^{-10}$ sec one sees that $V \sim c$. This is an important result, however, in two respects. First, it is obviously quite different from the very slow velocities associated with s.i.t. Second, it ensures that our neglect of back-scattering is an excellent approximation.[6] It also means that the pulse is not sharing large fractions of its energy with the atoms.

The chirping curve is obviously something new. We see that all pulses, not just s.c.l. pulses, are chirped. In fact, one can easily find from Marth's results[4] a very simple formula for the chirp at pulse center:

$$\dot{\phi}_0 \sim \begin{cases} 3/2\omega\tau^2 , & \tau < \tau_a \\ -3/2\omega\tau_a^2 , & \tau_a < \tau < \omega\tau_a^2 \\ -3/4\tau , & \tau > \omega\tau_a^2 \end{cases} \tag{10}$$

There are no previous quantitative results on chirping in absorbers. As Slusher and Gibbs point out,[5] even estimates of the importance of chirping (especially for s.c.l. pulses) have been difficult to make.

Of course the results for $\tau > \tau_a$ (i.e., for non-s.c.l. pulses) given in (10) and Figs. 1, 2 are to be ignored because inhomogeneous broadening is quite important for such pulses. However, the s.c.l. regime's expression can be taken seriously, and it imparts new information. In the first place, it says that $\dot{\phi}_0\tau \ll 1$, since $\omega\tau \gg 1$. That is, the chirp definitely can never be large enough to affect the bandwidth of the pulse. On the other hand, the chirp is certainly measureable interferometrically, at least in principle. The accumulated phase shift over a one-foot propagation length is about $\pi/2$ for a 1 psec pulse.

Of course, when chirping is present there is no area theorem available to guide the area of the steady-state pulses. In Fig. 3, also taken from Ref. 4, one can see how little the relation between $1/\tau$ and the maximum of the envelope is changed by chirping.

Now, having shown some of the results, I must sketch the theory. Because the s.c.l. pulses are very slowly varying the two-level model is a good one for our atoms. Thus the optical Bloch equations suffice:

$$du/dt = \dot{\phi} v \tag{11}$$

$$dv/dt = -\dot{\phi} u + \kappa \mathscr{E} w \tag{12}$$

$$dw/dt = -\kappa \mathscr{E} v \tag{13}$$

Notice that chirping has been allowed for explicitly, but not detuning, and that incoherent relaxation processes have been ignored as unimportant (for pulses sufficiently short compared to $\omega \tau_a^2 = T$). In the familiar way, Bloch's equations must be made compatible with Maxwell's equations, but we'll ignore that temporarily.

The theory begins by letting the Bloch equations say what the field must be, as follows: First, we recognize that $\kappa \mathscr{E} / \omega$ must be very small for slowly varying pulses. Let us denote this ratio of Rabi frequency to carrier frequency by ρ. Then we assert that

$$\dot{\phi} = \sum_{m=0}^{\infty} \chi_m \rho^{2m} \tag{14}$$

$$w = \sum_{m=0}^{\infty} w_m \rho^{2m} \quad . \tag{15}$$

There is some support for each assertion, but we won't belabor that point here.

The point is that Eqs. (11) - (15) can be combined to yield (to lowest interesting order):

$$\rho(t, z) = \rho_0 \operatorname{sech}\left(\frac{t - t_0}{\tau}\right) \tag{16}$$

as well as

$$\begin{aligned} w_0 &= -1 \\ w_1 &= \frac{1}{2} (\omega\tau)^2 \\ w_2 &= \frac{1}{6} (\omega\tau)^2 \left[\rho_0^{-2} - (\omega\tau/2)^2\right] \quad . \end{aligned} \tag{17}$$

It is now the task of the Maxwell equations to pin down the various pulse parameters, such as maximum amplitude ρ_0, velocity V, chirp coefficients χ_0, χ_1, ..., etc. As I implied at the beginning, Maxwell's equations are up to the task, and among the results are those shown graphically in Figs. 1 - 3.

At this point let me recall my earlier remark that it is possible to accommodate inhomogeneous broadening into the power series scheme of Eqs. (14) and (15). It's amusing that this is the case, because I believe it's the first time that inhomogeneous broadening and chirping have coexisted self-consistently within a steady-state pulse theory for either amplifiers or absorbers.

Although the inhomogeneous broadening results will form the core of another paper,[7] let me at least give here the results corresponding to Eqs. (17) to show how naturally a detuning frequency γ enters into the theory:

$$\begin{aligned} w_0(\gamma) &= -1 \\ w_1(\gamma) &= \frac{1}{2}(\omega\tau)^2 \left[1 + (\gamma\tau)^2\right]^{-1} \\ w_2(\gamma) &= \frac{3}{2} (\omega\tau)^2 \frac{\rho_0^{-2} - (\omega\tau/2)^2 + \frac{2}{3}(\gamma\tau)(\chi_1\tau)}{\left[1 + (\gamma\tau)^2\right]\left[9 + (\gamma\tau)^2\right]} \end{aligned} \tag{18}$$

The $w_0(\gamma)$ and $w_1(\gamma)$ expressions are identical to those derived in the theory of self-induced transparency.[3] Furthermore a 2π unchirped pulse has $\rho_0=2/\omega\tau$ and $\chi_1=0$, leading to $w_2(\gamma)=0$, again in agreement with well known s.i.t. results.[3]

A few concluding remarks are in order. I've sketched a theory of pulses shorter than τ_a. Several results can be emphasized: (1) Slowly-varying steady-state pulses shorter than the cooperation limit are predicted to exist; (2) s.c.l. pulses travel at velocities on the order of c; (3) all s.c.l. pulses are chirped; (4) the chirp can never be large enough to contribute to the pulse bandwidth, but may be detectable interferometrically; and (5) inhomogeneous broadening can be added to the theory relatively easily.

Finally, a remark about the theory itself: Because the presence of chirping demolishes the s.i.t. area theorem,[3] the usual identity of pulse area with dipole turning angle is invalid. The usual theories of steady-state pulses,[3] based upon exploitations of this identity, are not very helpful guides to a chirped-pulse theory. We have used a new approach, embodied in the expansions (14) and (15), which is closer to perturbation theory in spirit.

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LIST OF FIGURES