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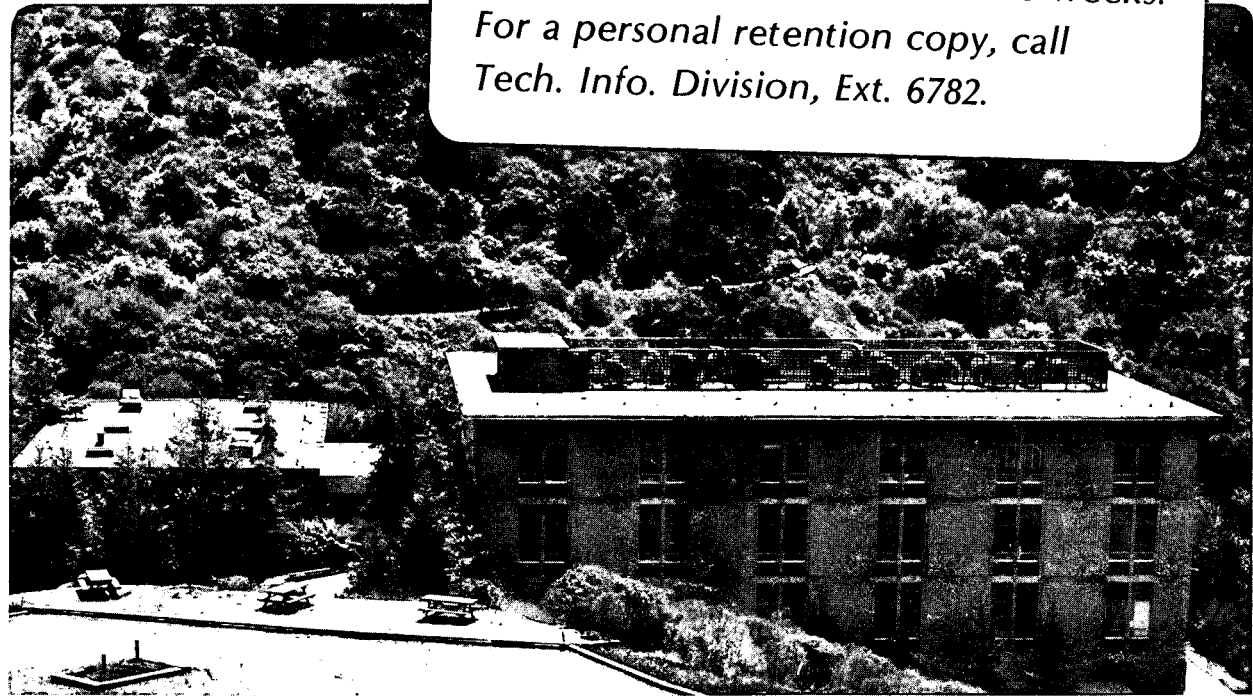
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MODULATED PULSES

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BROADBAND POPULATION INVERSION BY PHASE  
MODULATED PULSES

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Abstract

We present a class of continuously phase modulated radiation pulses that result in coherent population inversion over a large range of transition frequencies. The continuously modulated pulses can be approximated by sequences of discrete phase shifted pulses. Simulations of the inversion properties of the continuously modulated pulses and of the discrete pulse sequences are given.

For many experiments in NMR and coherent optics it is necessary to invert populations over a broad band of frequencies.<sup>1-3</sup> Recently, phase shifted pulses sequences for this purpose have been derived by several methods.<sup>4-5</sup> In this communication, we present an alternative analytical approach to broadband population inversion.

An exact broadband " $2\pi$ " pulse, employing amplitude modulation, exists in the context of self-induced transparency.<sup>6</sup> No analagous exact solution to broadband inversion is known. Allen and Eberly have presented a class of amplitude and phase modulated pulses that invert exactly on resonance.<sup>7</sup> We investigate the off resonance inversion behavior of similar phase modulated pulses. To derive such pulses, consider the Hamiltonian for nuclear spins under radiofrequency (rf) radiation in a rotating frame related to the laboratory frame by the transformation  $T = \exp[-iI_z(\omega t + \phi(t))]$  We call this the frequency modulated (FM) frame. The Hamiltonian is:

$$\mathcal{H} = (\Delta\omega + \dot{\phi}(t))I_z + \omega_1(t)I_x \quad (1)$$

where  $\omega_1(t)$ ,  $\phi(t)$  and  $\omega$  are the laboratory frame rf amplitude, phase and frequency respectively. In the FM frame,  $\Delta\omega$  is the resonance offset and the phase derivative  $\dot{\phi}(t)$  can be viewed as a frequency modulation. The Hamiltonian for a two-level optical system has the same form. The problem is to find  $\dot{\phi}(t)$  and  $\omega_1(t)$  that produce good inversion over a large range of values of  $\Delta\omega$ .

The inversion of on-resonance ( $\Delta\omega = 0$ ) spins can be described by a trajectory from  $+z$  to  $-z$  on a unit sphere in the FM frame.<sup>8</sup> A particularly simple class of inverting trajectories are those which follow a great circle from the  $+z$  to the  $-z$  axis with an azimuthal angle  $\gamma$ . Assuming a constant rf amplitude equal to  $\omega_1^0$ , these trajectories dictate a  $\dot{\phi}(t)$  characterized by a single parameter  $\delta$ . The derived  $\dot{\phi}$  functions are:

$$\dot{\phi}(t) = \frac{\omega_1^0 \delta}{\sqrt{1+\delta^2}} \tan \frac{\omega_1^0 t}{\sqrt{1+\delta^2}} \quad \frac{\pi}{2} \frac{\sqrt{1+\delta^2}}{\omega_1^0} \leq t \leq \frac{\pi}{2} \frac{\sqrt{1+\delta^2}}{\omega_1^0} \quad (2)$$

where  $\delta = \cot \gamma$ . The overall pulse area is  $\pi\sqrt{1+\delta^2}$ . The pulses suggested by Allen and Eberly produce the same on-resonance inverting trajectory but have variable  $\omega_1$  and extend from  $-\infty$  to  $+\infty$  in time.

In Figure 1a, we show calculated plots of the inversion accomplished by the pulse of Equation (2) as a function of the offset  $\Delta\omega$  for various values of  $\delta$ . For all values of  $\delta$ , the on-resonance inversion is perfect. When  $\delta = 0$ , the pulse of Equation (2) reduces to the usual  $\pi$  pulse. As  $\delta$  increases, the range of frequencies over which good inversion is achieved increases.

The inversion of the off-resonance spins can be understood by considering  $\dot{\phi}(t)$  in the FM frame as an adiabatic frequency sweep.<sup>9</sup> Typically, in NMR and optics, an adiabatic sweep is linear, that is  $\dot{\phi}(t) = kt$  where  $k$  is the constant sweep rate.<sup>10-11</sup> A nonlinear sweep can produce adiabatic inversion more efficiently, that is, in a significantly shorter time. For large values of  $\delta$ ,  $\dot{\phi}(t)$  is an example of an efficient adiabatic

sweep. In addition, because  $\dot{\phi}(t)$  derives from the on-resonance inverting trajectory, the inversion for  $\Delta\omega = 0$  is exact for all values of  $\delta$ .

Experimentally, it is usually more convenient to use a sequence of phase shifted pulses rather than a single pulse with a continuously varying phase. To generate such a pulse sequence, we approximate the continuous phase function  $\phi(t) = \int_0^t \dot{\phi}(t') dt'$  by a piecewise constant function. Some representative pulse sequences are shown in Figure 1b. The three pulse sequence is similar to one previously derived by a different method.<sup>4</sup> The details of the approximation method will be described in a forthcoming paper.

The continuously phase modulated pulses presented here invert populations over any desired bandwidth and always invert on resonance. Deriving pulse sequences from the continuously modulated pulses has the advantage, over previous methods, that the discrete sequences can also be constructed for inversion over any bandwidth with minimal computer optimization.

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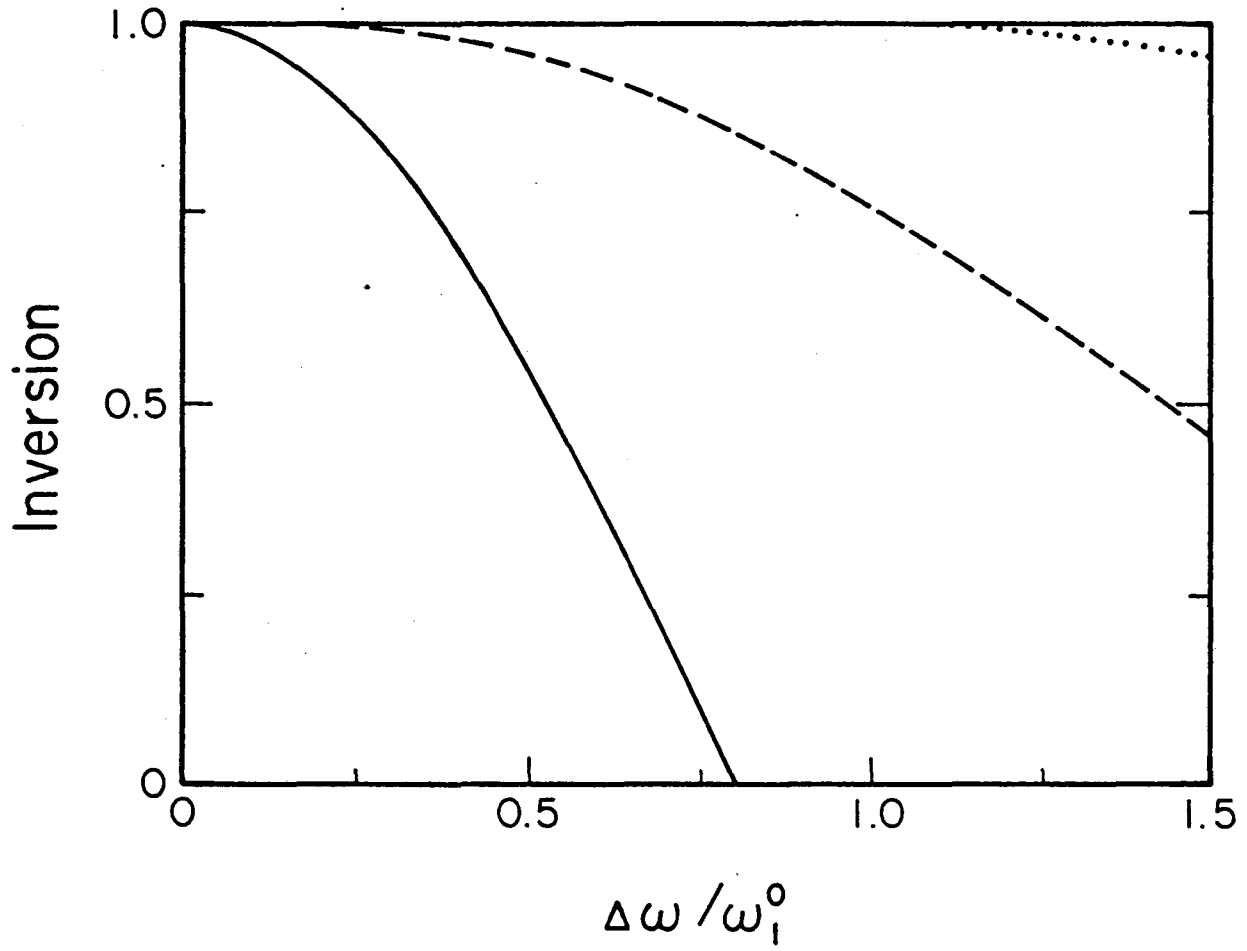


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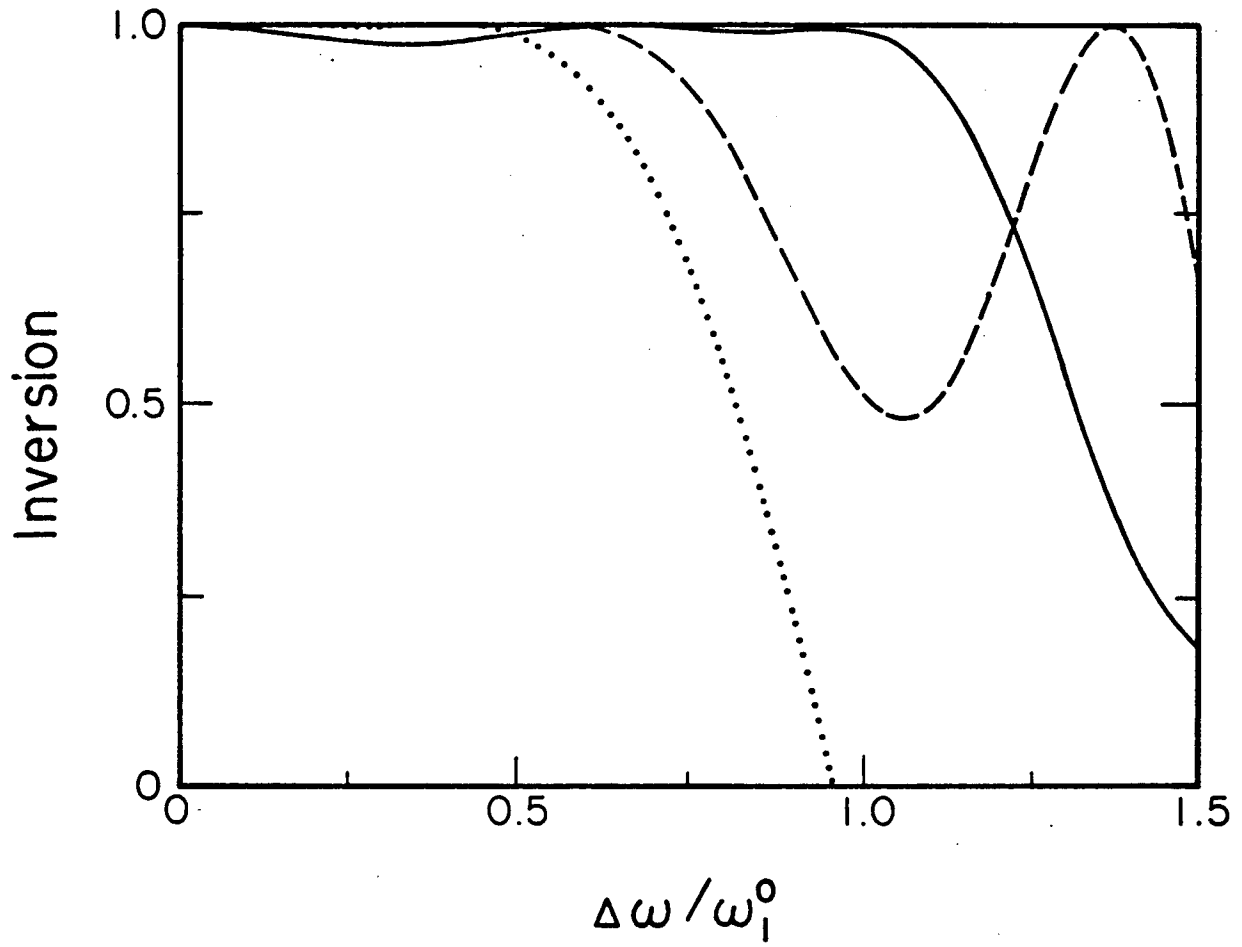
Figure Captions

Figure 1: Simulations of population inversion versus relative resonance offset. a) Continuously phase modulated pulse of Equation (2) for  $\delta = 0$  (solid line),  $\delta = 4$  (dashed line),  $\delta = 9$  (dotted line).  $\delta = 0$  corresponds to a  $\pi$  pulse. b) Discrete pulse sequences derived from continuously phase modulated pulse:  
 $(84)_{94} (251)_0 (84)_{94}$  (dotted line);  $(64)_{322} (122)_{96}$   
 $(310)_0 (122)_{96} (64)_{322}$  (dashed line);  $39_{419} (54)_{209}$   
 $(66)_{139} (84)_{70} (267)_0 (84)_{70} (66)_{139} (54)_{209} (39)_{419}$   
(solid line). The notation here is  $(\theta)_\phi$  where  $\theta$  and  $\phi$  are the flip angles and phases of individual pulses in degrees.



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FIGURE 1a



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FIGURE 1b

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