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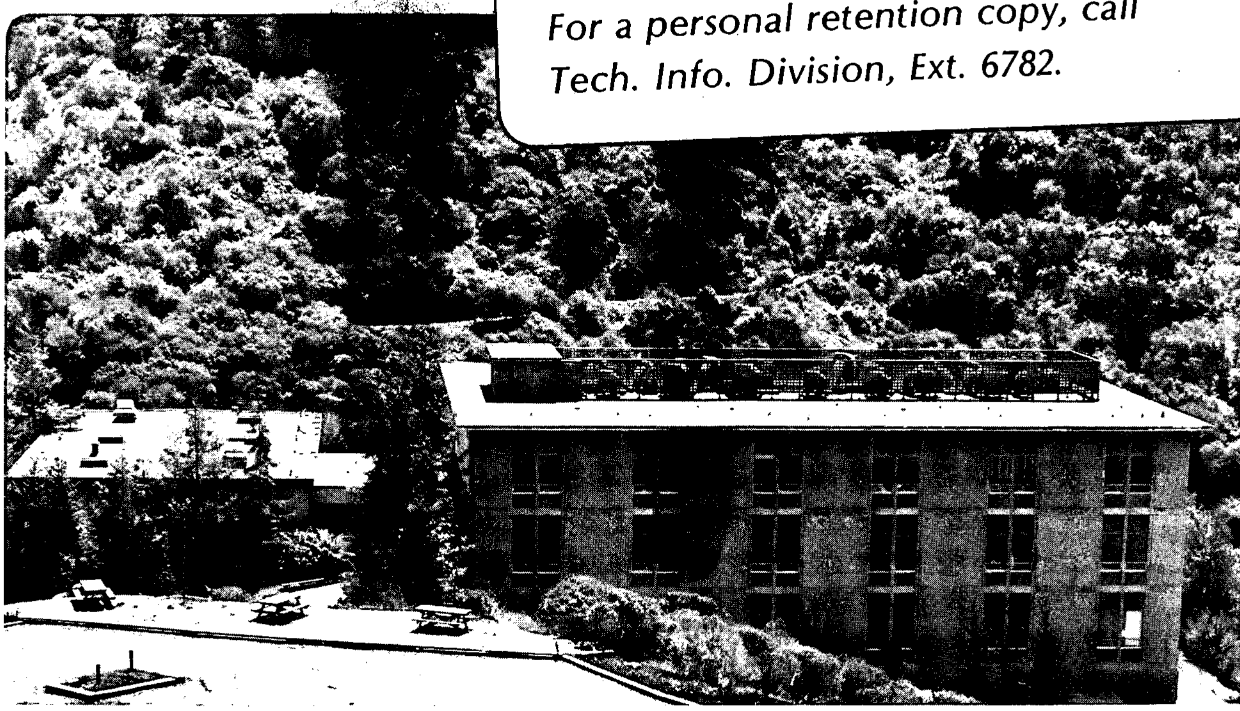

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BROADBAND POPULATION INVERSION

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Abstract

We present a theory for constructing sequences of phase-shifted radiation pulses for coherently inverting populations over a broad band of transition frequencies. Such sequences have applications in nuclear magnetic resonance and coherent optics. Examples of sequences are derived, together with computer simulations of their inversion properties.

The ability to achieve population inversion with coherent radiation is essential to many techniques in pulsed nuclear magnetic resonance (NMR)[1-4] and coherent optics[5-7]. In an ideal system with a single sharp transition frequency, a π pulse exactly at that frequency with a constant phase would produce the desired inversion. However, any real system has transition frequencies over a certain bandwidth resulting, for example, from chemical shifts or spin couplings in NMR or from Doppler broadening or crystal strains in optics. The presence of a range of transition frequencies makes it impossible to achieve a complete resonant inversion, leading us to the important problem of maximizing the inversion over the existing bandwidth. One approach to that problem is simply to use a π pulse with the shortest possible length and the largest possible peak power. Of course, this approach is subject to practical limitations beyond which there is no way to systematically improve its performance. A second approach is to design sequences of coherent, phase-shifted pulses that can effectively invert populations over a larger bandwidth without any increase in peak power[7,8]. In this Letter, we present for the first time a rigorous and systematic theoretical approach to the construction of such sequences for broadband population inversion.

We begin by considering the response of a system to a general sequence of coherent radiation pulses. Using NMR nomenclature, the Hamiltonian governing the response in the rotating frame is:

$$\mathcal{H}(t) = \mathcal{H}_0 + V \qquad (1)$$

$$\mathcal{H}_0 = \omega_1^0(I_x\cos\phi(t) + I_y\sin\phi(t)) \qquad (2)$$

$\mathcal{H}_0$ represents the interaction of the system with the radiation, where ω_1^0 and $\phi(t)$ are respectively the amplitude and phase of the radiation. ω_1^0 is $\kappa\mathcal{E}$ in coherent optics or γH_1 in NMR. I_x, I_y and I_z are components of the spin angular momentum operator in NMR or fictitious spin operators in coherent optics. In general, V may include any other interactions or imperfections in the radiation. For the problem of broadband inversion, we take $V = \Delta\omega I_z$, where $\Delta\omega$ is the difference between the radiation frequency and the transition frequency, commonly called the resonance offset. The evolution of the system during the pulse sequence is then dictated by the propagator U(t), given by:

$$U(t) = T\exp(-i\int_0^t dt'\mathcal{H}(t')) \qquad (3)$$

Here T is the Dyson time ordering operator[9].

The usual initial equilibrium condition of the system is described by a density operator proportional to I_z. If the pulse sequence has a total duration τ, the final condition is $\rho_f = U(\tau)I_zU(\tau)^{-1}$. Flipping a nucleus in NMR or completely exchanging the ground and excited state populations in an optical two-level system corresponds to obtaining a final condition of $\rho_f = -I_z$. Both $\mathcal{H}_0$ and V act simultaneously to bring about the evolution from I_z to ρ_f, as shown in Equation (3). However, we can separate the resonance offset from the pure radiation interaction by rewriting the propagator as follows:

$$U(t) = T \exp(-i\int_0^t dt' \mathcal{H}_0)\ T \exp(-i\int_0^t dt' \tilde{V}(t')) \tag{4}$$

i.e.
$$U(t) = U_0(t)U_V(t) \tag{5}$$

$$\tilde{V}(t) = U_0^{-1}(t)\ V\ U_0(t) \tag{6}$$

In light of Equation (5), the evolution of the system from I_z to ρ_f appears in two steps. First, the presence of V causes a transformation from I_z to an intermediate condition $\rho_i = U_v(\tau)I_zU_v(\tau)^{-1}$. Then $U_0(\tau)$, which would be the propagator for the pulse sequence in the absence of V, takes ρ_i to ρ_f. The division of the overall evolution into two steps forms the basis for our approach to the inversion problem.

By definition, all inverting sequences satisfy the following equation:

$$U_0(\tau)I_zU_0(\tau)^{-1} = -I_z \tag{7}$$

Different sequences have different $U_v(\tau)$ operators, however. For an inverting sequence with $U_v(\tau) = 1$, we would have identically $\rho_i = I_z$; the sequence would then give perfect inversion even in the presence of an arbitrarily large resonance offset, or over an infinite bandwidth. In general, though, we expect to achieve good inversion only over a finite bandwidth. We therefore would like to express the effect of the presence of the resonance offset as a power series in $\Delta\omega$. This can be accomplished by making a Magnus expansion[10-12] of $U_v(\tau)$:

$$U_v(\tau) = \exp\{-i(V^{(0)} + V^{(1)} + \ldots)\tau\} \tag{8}$$

$$V^{(0)} = \frac{1}{\tau} \int_0^{\tau} dt \tilde{V}(t) \quad (9)$$

$$V^{(1)} = \frac{-i}{2\tau} \int_0^{\tau} dt_1 \int_0^{t_1} dt_2 [\tilde{V}(t_1), \tilde{V}(t_2)] \quad (10)$$

The term $V^{(n)}$ in the Magnus expansion is proportional to $\Delta\omega^{n+1}$. Then pulse sequences with $V^{(n)} = 0$ for $0 \leq n \leq N$ should have $U_V(\tau) \approx 1$ for increasingly large resonance offsets as N becomes larger. Consequently, we should see good inversion over an increasingly large bandwidth.

So far, our treatment of broadband inversion has been largely formal. However, it leads to a definite prescription for the derivation of pulse sequences. A sequence composed of m pulses may be described by the notation $(\theta_1)_{\phi_1} \cdots (\theta_i)_{\phi_i} \cdots (\theta_m)_{\phi_m}$, where ϕ_i is the phase of the radiation during the i^{th} pulse and $\theta_i = \tau_i \omega_1^0$ with τ_i being the duration of the i^{th} pulse. The most general sequence of m pulses therefore has 2m variable quantities. According to Equation (6), $\tilde{V}(t)$ in turn depends on those 2m variables, as do all terms in the expansion of Equation (8). To find an inverting sequence with $V^{(n)} = 0$ for $0 \leq n \leq N$, we define a general sequence with enough variables to simultaneously solve those N+1 equations in addition to Equation (7). Thus, the derivation of a pulse sequence for broadband inversion to some order N reduces to the problem of solving a specific set of equations.

In Figure 1, we show computer simulations of the population inversion W as a function of $\Delta\omega/\omega_1^0$ for two sequences derived in the above manner, as well as for a simple π pulse. W is defined by:

$$W = -\frac{Tr(I_z \rho_f)}{Tr(I_z^2)} \quad (11)$$

The three-pulse sequence in Figure 1 has $V^{(0)} = 0$ and has been derived earlier by a different approach[8]. The seven-pulse sequence in Figure 1 has both $V^{(0)} = 0$ and $V^{(1)} = 0$. In accordance with our theory, the inversion bandwidth increases as successive terms in Equation (8) are made to vanish.

The approach outlined above can be extended to the design of pulse sequences that overcome a second obstacle to coherent population inversion, namely the existence of inhomogeneity in the radiation amplitude. Radiation inhomogeneity arises from nonuniform laser beam profiles in coherent optics or from the finite size of the excitation coil in NMR. To treat radiation inhomogeneity we need only take $V = \delta\omega_1(I_x\cos\phi(t) + I_y\sin\phi(t))$ in Equation (1), where $\delta\omega_1$ is the deviation of the radiation amplitude from its nominal value ω_1^0. Then the Magnus expansion of $U_v(\tau)$ becomes a power series in $\delta\omega_1$. The theory is the same in all other respects. In Figure 2, we show W as a function of $\delta\omega_1/\omega_1^0$ for a three-pulse sequence with $V^{(0)} = 0$ as well as for a π pulse.

The theory may be further applied to population inversion in the presence of any other interfering interactions, such as dipolar or quadrupolar couplings in NMR. In addition, pulse sequences that bring the system to a final condition other than a population inversion may be constructed by the same method. For example, under the sequence $(\pi)_{\pi/3}(\pi/2)_{2\pi/3}(\pi)_{\pi/3}$, the system evolves from equilibrium to a final condition of $\rho_f = I_y$ independent of resonance offset to 0^{th} order. This is the broadband counterpart of a coherent $\pi/2$ pulse. We will present such applications in detail in a full paper.

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Figure Captions

Figure 1: Population inversion vs. relative resonance offset for a magnetic nucleus in NMR or two-level optical system. The results of computer simulations are shown for the inversion produced by a π pulse (dashed line), a $(\pi/2)_0(3\pi/2)_{\pi/2}(\pi/2)_0$ sequence (dotted line) and a $(1.867\pi)_0(1.367\pi)_\pi(0.056\pi)_{\pi/2}(0.411\pi)_{3\pi/2}$ - $(0.056\pi)_{\pi/2}(1.367\pi)_\pi(1.867\pi)_0$ sequence (solid line). The effect of a resonance offset on the inversion vanishes to 0^{th} order in our theory for the three-pulse sequence and to 1^{st} order for the seven-pulse sequence. Due to the symmetry of the sequences, the inversion is independent of the sign of the offset.

Figure 2: Population inversion vs. relative deviation in radiation amplitude. The results of computer simulations are shown for the inversion produced by a π pulse (dashed line) and a $(\pi)_0(\pi)_{2\pi/3}(\pi)_0$ sequence (solid line). The effect of deviations in the radiation amplitude on the inversion vanishes to 0^{th} order for the three-pulse sequence.

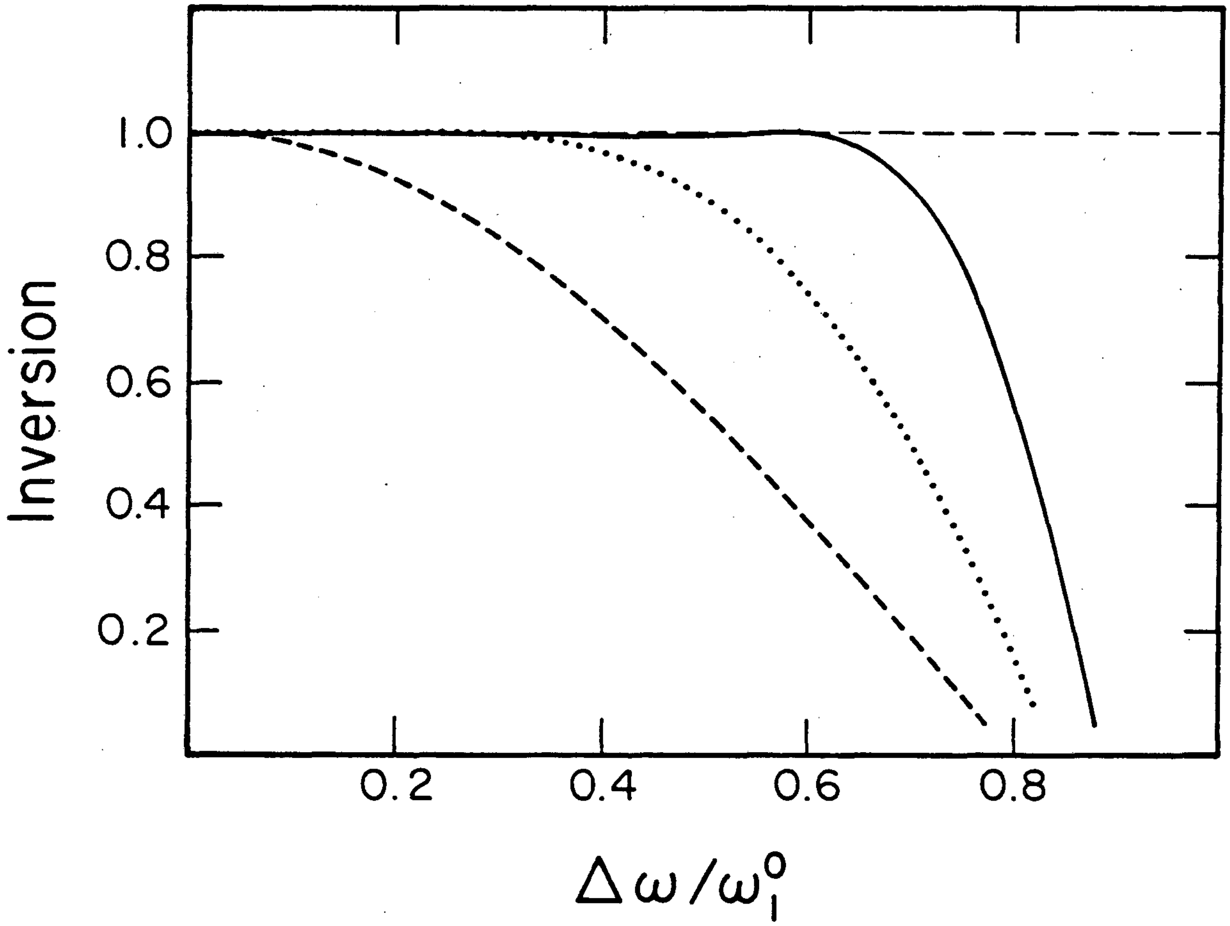

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Figure 1

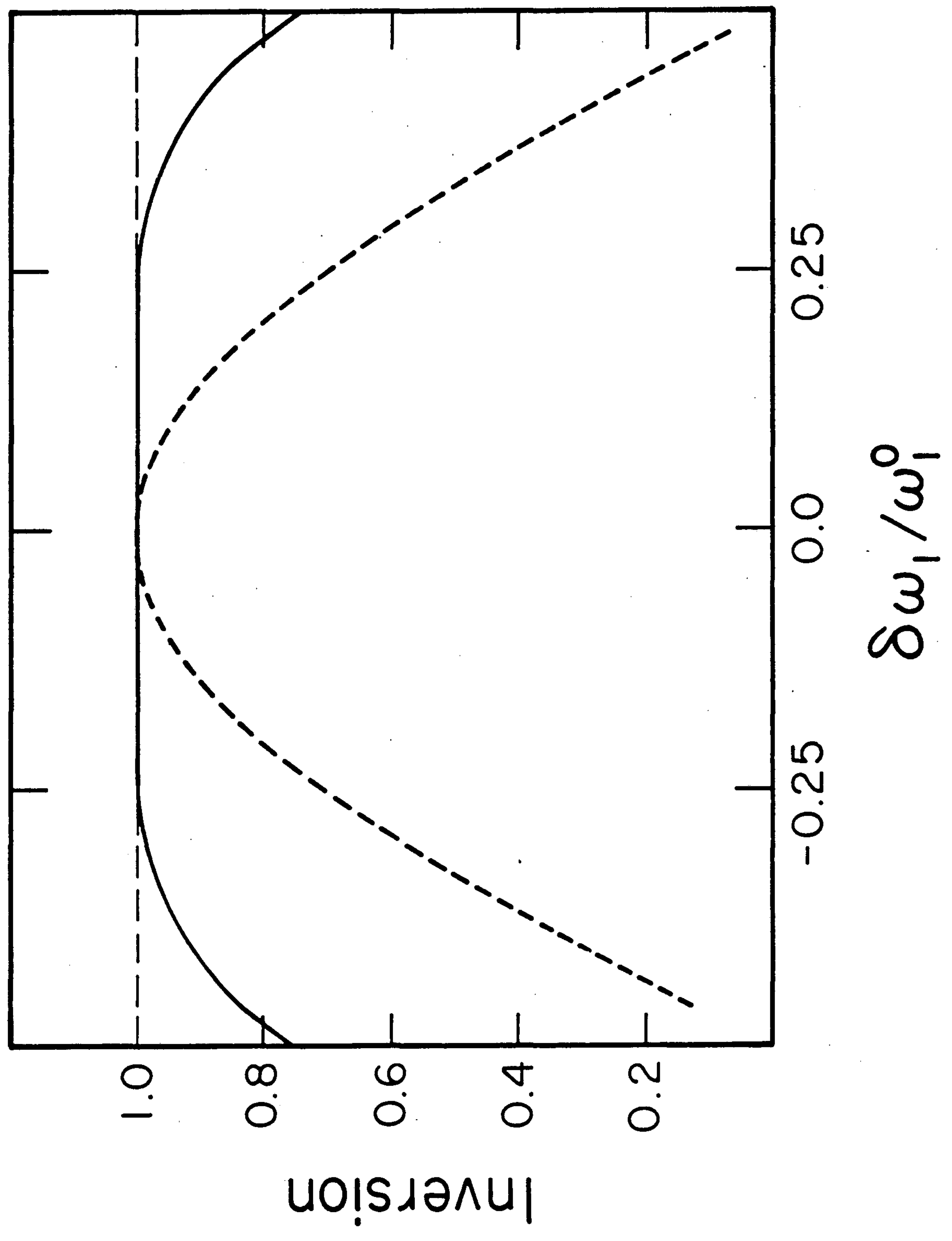

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Figure 2

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