Broadband Population Inversion

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A theory is presented for construction of 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.

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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 more powerful 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, I present a systematic theoretical method for 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. With use of NMR nomenclature, the Hamiltonian governing the response in the rotating frame is

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

$$\mathcal{H}_0 = \omega_1{}^0[I_x \cos\varphi(t) + I_y \sin\varphi(t)]. \tag{2}$$

$\mathcal{H}_0$ represents the interaction of the system with the radiation, where $\omega_1{}^0$ and $\varphi(t)$ are respectively the amplitude and phase of the radiation. $\omega_1{}^0$ 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

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

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 of τ, the final condition is $\rho_f = U(\tau) I_z U(\tau)^{-1}$. 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 Eq. (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.,

$$U(t) = U_0(t) U_V(t), \tag{5}$$

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

In light of Eq. (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 $\rho_i = U_V(\tau) I_z U_V(\tau)^{-1}$. 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 the present approach to the inversion problem.

By definition, all inverting sequences satisfy the following equation:

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

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 $U_V(\tau)$:

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

$$V^{(0)} = \tau^{-1} \int_0^\tau dt\, \tilde{V}(t), \tag{9}$$

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

The term $V^{(n)}$ in the Magnus expansion is proportional to $\Delta\omega^{n+1}$. I call an inverting sequence broadband to Nth order if $V^{(n)} = 0$ for $0 \le n \le N$. As N becomes larger, $U_V(\tau) \approx 1$ for increasingly large resonance offsets. Consequently we should see good inversion over an increasingly large bandwidth.

So far, the treatment of broadband inversion has been largely formal. However, it leads to a definite procedure for the derivation of pulse sequences that are broadband to any desired order N. The problem is to find the specific phase function $\varphi(t)$ in Eq. (2). During a sequence of m pulses, $\varphi(t)$ equals a constant φ_i during the ith time interval, where $1 \le i \le m$. The ith time interval has a duration τ_i. $U_0(t)$ is then a product of rotation operators about axes given by the φ_i. For example, during the second pulse, $U_0(t)$ is

$$U_0(t) = \exp\{-i\omega_1^0 (I_x \cos\varphi_2 + I_y \sin\varphi_2)(t - \tau_1)\} \exp\{-i\omega_1^0 (I_x \cos\varphi_1 + I_y \sin\varphi_1)\tau_1\}. \tag{11}$$

$\tilde{V}(t)$ may then be calculated in terms of trigonometric functions of the $2m$ variables φ_i and τ_i, according to Eq. (6). In turn, the terms in the Magnus expansion are functions of these $2m$ variables, as in Eqs. (9) and (10). The task of finding a pulse sequence then reduces to the problem of solving the $N+1$ equations $V^{(n)} = 0$ for $0 \le n \le N$ in addition to Eq. (7). All of the equations, each of which has I_x, I_y, and I_z components, involve the variables φ_i and τ_i. If we make m large enough, there will be a simultaneous solution, i.e., the desired pulse sequence. In principle, the method may be applied to any order, although in practice the equations are sufficiently complicated for $N > 0$ that they are solved by computer.

In Fig. 1, I 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. Pulse sequences are described by the notation

$$(\theta_1)_{\varphi_1} \ldots (\theta_i)_{\varphi_i} \ldots (\theta_m)_{\varphi_m},$$

where $\theta_i = \tau_i \omega_1^0$. W is defined by

$$W = -\mathrm{Tr}(I_z \rho_f)/\mathrm{Tr}(I_z^2). \tag{12}$$

The three-pulse sequence in Fig. 1 has $V^{(0)} = 0$ and has been derived earlier by a geometric approach which is not easily extended to the derivation of higher-order sequences.[8] The seven-pulse sequence in Fig. 1 has both $V^{(0)} = 0$ and $V^{(1)} = 0$. In accordance with the present theory, the inversion bandwidth increases as successive terms in Eq. (8) are made to vanish.

It should be stressed that the increased inversion bandwidth of the pulse sequences in Fig. 1 does not result from an increase in the width of the Fourier spectra of the sequences themselves. In a highly nonlinear process such as coherent population inversion, there is no simple connection between the Fourier spectrum of the excitation and the spectrum of the response of the system.

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. This problem has also been treated by a geometric approach.[13] To treat radiation inhomogeneity with the present method we need only take $V = \delta\omega_1[I_x \cos\varphi(t) + I_y \sin\varphi(t)]$ in Eq. (1), where $\delta\omega_1$ is the deviation of the radiation amplitude from its nominal value ω_1^0. Then the Mag-

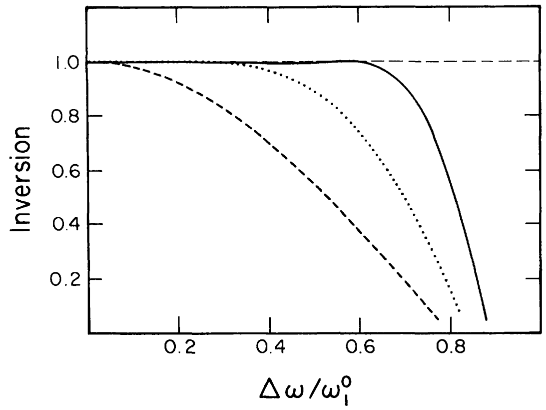

FIG. 1. Population inversion vs relative resonance offset for a magnetic nucleus in NMR or two-level optical system. The results of computer simulations are shown for the inversion produced by a π pulse (dashed line), a $(\pi/2)_0(3\pi/2)_{\pi/2}(\pi/2)_0$ sequence (dotted line), and a $(1.867\pi)_0(1.367\pi)_\pi(0.056\pi)_{\pi/2}(0.411\pi)_{3\pi/2}$-$(0.056\pi)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

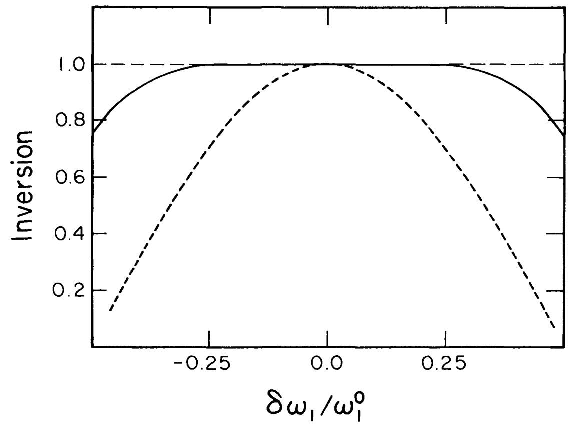

FIG. 2. Population inversion vs relative deviation in radiation amplitude. The results of computer simulations are shown for the inversion produced by a π pulse (dashed line) and a $(\pi)_0(\pi)_{2\pi/3}(\pi)_0$ sequence (solid line). The effect of deviations in the radiation amplitude on the inversion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

nus expansion of $U_V(\tau)$ becomes a power series in $\delta\omega_1$. The theory is the same in all other respects. In Fig. 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.

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 zeroth order. This is the broadband counterpart of a coherent $\pi/2$ pulse.

A further application of the theory that is of particular importance in NMR is the construction of pulse sequences for population inversion in the presence of dipolar or quadrupolar couplings. If V is a dipolar or quadrupolar coupling Hamiltonian, the sequence $(\pi/4)_0(\pi)_{\pi/2}(\pi/2)_\pi(\pi)_{\pi/2}(\pi/4)_0$ is an inverting sequence with $V^{(0)}=0$. The sequence $(\pi/4)_0(3\pi/4)_\pi(3\pi/4)_{\pi/2}(\pi/4)_{3\pi/2}$ creates a final condition of $\rho_f=I_y$ and has $V^{(0)}=0$. The details of such applications will be presented in a full paper.

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