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Selective spin inversion in nuclear magnetic resonance and coherent optics through an exact solution of the Bloch-Riccati equation

M. S. Silver* and R. I. Joseph
The Department of Electrical Engineering and Computer Science, The Johns Hopkins University, Baltimore, Maryland 21218

D. I. Hoult
Biomedical Engineering and Instrumentation Branch, National Institutes of Health, Bethesda, Maryland 20205

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An analytical solution of the Bloch equations is presented using as a driving function a complex hyperbolic secant pulse. The solution indicates that, under the appropriate conditions, the use of such a pulse creates a highly selective population inversion which, above a critical threshold, is independent of pulse (irradiating B_1 field) amplitude and hence B_1 field homogeneity. This result is shown to be in excellent agreement with experiment, and as an aid to comprehension, analogies to adiabatic rapid passage and self-induced transparency are drawn.

It is well known that the disciplines of nuclear magnetic resonance (NMR) and coherent optics have much in common in their descriptions; similar phenomena such as spin and photon echoes, free induction decay, and adiabatic fast passage are observed. Recently, Baum, Tycko, and Pines[1] have published a method for broadband spin inversion using the concept of simultaneous amplitude and phase-modulated B_1 fields based upon analogies to coherent optics. Consequently, we have suggested that the complex form of a hyperbolic secant (sech) B_1 field is a useful alternative analysis for narrow-band spin inversion and is the NMR analog of self-induced transparency.[2,3] McCall and Hahn[4] showed that a pulse of light could propagate through a medium and evolve into a 2π sech pulse, with negligible change in the pulse shape, provided the incident pulse power was above a critical threshold. Similarly, we have shown that the application of a complex, sech radio-frequency B_1 field pulse to an inhomogeneously broadened NMR spin system creates a spin inversion over a sharply defined region within the spectrum, provided the incident pulse power is, once again, above a critical threshold. Outside that region, the spins are returned to their equilibrium positions, and magnetization remains unaffected. In this Rapid Communication we therefore give the theoretical basis for this unique phenomenon and show that our theory is in excellent agreement with experiment.

To begin our analysis, we write the Bloch equations in the rotating frame. Ignoring relaxation effects, we then have

$$\dot{M}+i\Delta\omega M+M_z\Omega(t)=0\ , \tag{1a}$$

$$\dot{M}_z-\tfrac{1}{2}i(M\Omega^*(t)-M^*\Omega(t))=0\ . \tag{1b}$$

Here $i=\sqrt{-1}$, M is the complex magnetization in the x-y plane, M_z the longitudinal magnetization, and $\Omega(t)=-\gamma(B_{1x}+iB_{1y})$ the complex, time-dependent driving function. Let

$$f=\frac{M}{M_0+M_z}\ , \tag{2}$$

where M_0 is the equilibrium magnetization. Equations (1) can then be directly transformed into the Bloch-Riccati (BR) equation:

$$\dot{f}+i\Delta\omega f-\tfrac{1}{2}i\Omega^*(t)f^2+\tfrac{1}{2}i\Omega(t)=0\ . \tag{3}$$

In general, this equation cannot be solved analytically. However, there is one well-known driving function—a hyperbolic secant[5-8]—for which a solution may be found. Consider therefore

$$\Omega(t)=\Omega_0(\operatorname{sech}\beta(t-t_0))^{1+i\mu}\ , \tag{4}$$

where μ is a real constant and Ω_0 is the pulse amplitude. Figure 1 shows that Eq. (4) is a complex B_1 field, with both x and y components applied to the NMR spin system simul-

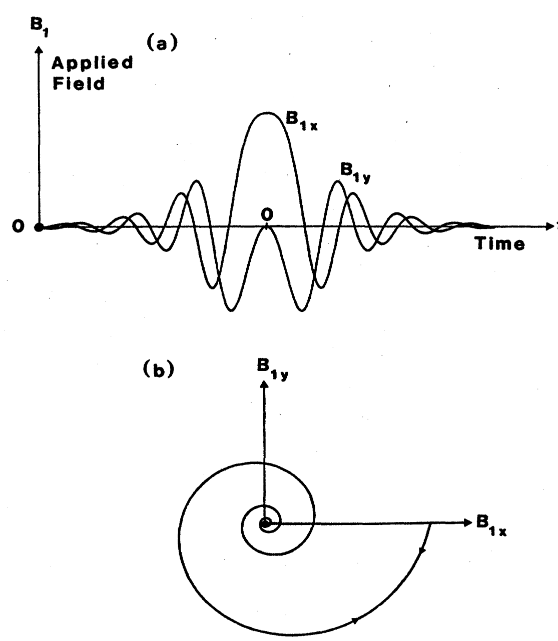

FIG. 1. (a) The complex, hyperbolic secant B_1 field pulse with $\mu=5.0$. (b) A plot of B_{1x} vs B_{1y} in the rotating frame.

taneously. Inserting Eq. (4) into the BR equation, and using the transformations

$$2x=1+\tanh[\beta(t-t_0)]\ ;$$
$$W=\exp\left(-\tfrac{1}{2}i\int_0^t \Omega^*(t')f\,dt'\right) . \quad (5)$$

We obtain

$$x(1-x)W''+W'\left\{-x(1+i\mu)+\left[\tfrac{1}{2}+i\left(\frac{\Delta\omega}{2\beta}+\frac{\mu}{2}\right)\right]\right\}+\left|\frac{\Omega_0}{2\beta}\right|^2 W=0\ , \quad (6)$$

which is a hypergeometric equation whose solutions are of the form

$$W(x)=AF(a,b;c;x)+Bx^{1-c}F(a-c+1,b-c+1;2-c;x)\ . \quad (7)$$

A and B are arbitrary constants, and

$$a=i\frac{\mu}{2}+\left[\left(\frac{\Omega_0}{2\beta}\right)^2-\left(\frac{\mu}{2}\right)^2\right]^{1/2}=i\frac{\mu}{2}+p\ , \quad (8)$$

$$b=i\frac{\mu}{2}-\left[\left(\frac{\Omega_0}{2\beta}\right)^2-\left(\frac{\mu}{2}\right)^2\right]^{1/2}=i\frac{\mu}{2}-p\ , \quad (9)$$

$$c=\tfrac{1}{2}+i\left(\frac{\Delta\omega}{2\beta}+\frac{\mu}{2}\right)=\tfrac{1}{2}+i\nu\ , \quad (10)$$

where we consider solutions for p real and $2p\neq 1,2,3,\ldots$. From the boundary condition $f\to 0$, for $t\to -\infty$ and $x\to 0$, it may be shown that if the magnetization is initially at equilibrium, $B=0$. From Eq. (5), we then write the solution to the BR equation as

$$|f|=\frac{\beta}{\Omega_0}\operatorname{sech}[\beta(t-t_0)]\left|\frac{ab}{c}\right|\left|\frac{F(a+1,b+1;c+1;x)}{F(a,b;c;x)}\right| . \quad (11)$$

We next investigate the behavior of the solution when $t\to\infty$ and $x\to 1$. To this end, we may further reduce Eq. (11) through well-known identities.[9] Writing sech $[\beta(t-t_0)]$ as $2[x(1-x)]^{1/2}$, we have that

$$|f|_{x\to 1}=\frac{2\beta}{\Omega_0}\left|\frac{\Gamma(a+b+1-c)\Gamma(c-a)\Gamma(c-b)}{\Gamma(a)\Gamma(b)\Gamma(c-a-b)}\right| . \quad (12)$$

Through the definitions

$$c-a=\tfrac{1}{2}-p+i(\nu+\tfrac{1}{2}\mu)=(1-\xi)^*\ , \quad (13a)$$

$$c-b=\tfrac{1}{2}+p+i(\nu+\tfrac{1}{2}\mu)=\xi\ , \quad (13b)$$

and $a=z=-b^*$, we then obtain

$$|f|=\frac{2\beta}{\Omega_0}\left|\frac{\Gamma(\xi)\Gamma(1-\xi)}{\Gamma(z)\Gamma(-z)}\right| . \quad (14)$$

Taking the square of the modulus, and using well-known trigonometric and gamma-function identities, we obtain

$$|f|^2=\frac{\cosh^2\pi(\tfrac{1}{2}\mu)-\cos^2\pi p}{\cosh^2\pi(\Delta\omega/2\beta)-\sin^2\pi p} . \quad (15)$$

Expressing M_z, the longitudinal magnetization, in terms of f we see that

$$\frac{M_z}{M_0}=\frac{1-|f|^2}{1+|f|^2} . \quad (16)$$

Inserting Eq. (15) into Eq. (16) we have that

$$\frac{M_z(\Delta\omega)}{M_0}=\tanh\pi\left(\frac{\Delta\omega}{2\beta}+\frac{\mu}{2}\right)\tanh\pi\left(\frac{\Delta\omega}{2\beta}-\frac{\mu}{2}\right)+\cos\left\{\pi\left[\left(\frac{\Omega_0}{\beta}\right)^2-\mu^2\right]^{1/2}\right\}\operatorname{sech}\pi\left(\frac{\Delta\omega}{2\beta}+\frac{\mu}{2}\right)\operatorname{sech}\pi\left(\frac{\Delta\omega}{2\beta}-\frac{\mu}{2}\right) . \quad (17)$$

We now can use this equation to predict the behavior of the inversion for different parameter values.

Equation (17) has several interesting features. First, consider the case where the driving function has no phase modulation (i.e., $\mu=0$). Whenever $\Omega_0\approx 2n\beta(n=1, 2, \ldots)$, $M_z=M_0$, independent of $\Delta\omega$. In other words, a sech pulse of the correct amplitude causes an excursion of magnetization from its starting position, but then returns it to its starting point. However, unlike the phenomenon of self-induced transparency,[4] the effect is not independent of pulse amplitude. Second, consider the limit as $\mu\to\infty$, $\beta\to 0$, $\mu\beta\to C$, a constant, and $\Omega\geq C$. Under these conditions, as $t\to\infty$ (we neglect relaxation), $M_z\to -M_0$. Thus, magnetization is inverted over all frequencies. The driving function then becomes in the region about t_0,

$$\Omega\approx\Omega_0\left[\cos\left(\mu\beta^2\frac{(t-t_0)^2}{2}\right)-i\sin\left(\mu\beta^2\frac{(t-t_0)^2}{2}\right)\right] . \quad (18)$$

Thus, the B_1 field is rotating at a frequency given by

$$\omega_s=\mu\beta^2(t-t_0)\ , \quad (19)$$

and we are effectively performing an adiabatic rapid-passage experiment, for ω_s is swept with time. We therefore see that there is a close relationship between the use of complex sech pulse and a passage experiment.

Third, we note that the second term in Eq. (17) has a maximum value of unity, and that once $\mu\geq 2$, the term has negligible (< 1%) influence on the value of M_z. Thus, provided $\Omega_0\geq\mu\beta$, M_z is essentially independent of the amplitude of the B_1 field, and therefore is independent of the B_1 field homogeneity—an important practical point.

Fourth, as α $[\alpha=\pi(\Delta\omega/2\beta+\tfrac{1}{2}\mu)]$ increases from negative to positive values, so tanhα switches from an asymptote of -1 to $+1$. When $\alpha=\pm 1.47$, tanh$\alpha=\pm 0.9$ and thus the rise from the 5% to the 95% level in the function occurs over the small range of $\Delta\alpha=2.94$. Now for $\Omega_0\geq\mu\beta$, $\mu\geq 2$,

$$\frac{M_z}{M_0}\approx\tanh\pi\left(\frac{\Delta\omega}{2\beta}+\frac{\mu}{2}\right)\tanh\pi\left(\frac{\Delta\omega}{2\beta}-\frac{\mu}{2}\right) , \quad (20)$$

is a localized inversion of magnetization (Fig. 2), of width

$$\Delta\omega=\pm\mu\beta\ . \quad (21)$$

It is clear that as μ increases, the selectivity of the inversion

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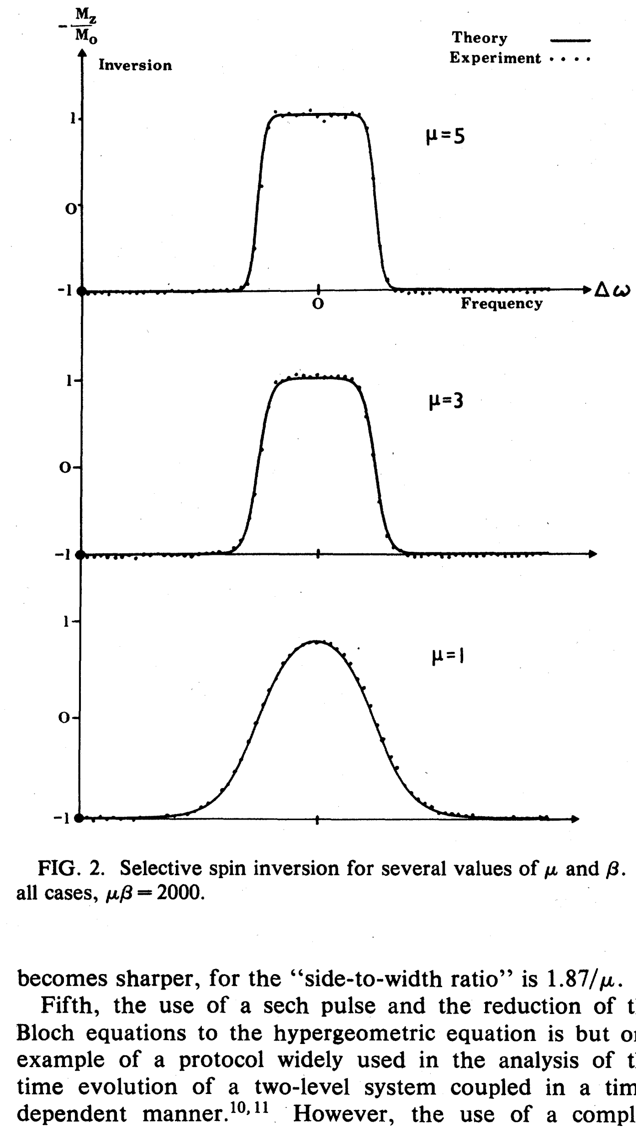

FIG. 2. Selective spin inversion for several values of μ and β. In all cases, $\mu\beta = 2000$.

becomes sharper, for the "side-to-width ratio" is $1.87/\mu$.

Fifth, the use of a sech pulse and the reduction of the Bloch equations to the hypergeometric equation is but one example of a protocol widely used in the analysis of the time evolution of a two-level system coupled in a time-dependent manner.[10,11] However, the use of a complex sech pulse in NMR is novel and the consequent selectivity of the final result is surprising.

Figure 2 compares Eq. (17) with experimental data obtained on an NMR imaging spectrometer using a sample of water at 5 MHz. A single-sideband modulator was a standard part of the equipment and $\mu\beta$ was kept constant at a value of 2000. The pulse was truncated at approximately the 1% level. Thus, for $\mu = 5$, $\beta = 400\ \text{sec}^{-1}$, a 25-ms, 1024 complex point sech function was passed from our array processor via D/A converters, to the spectrometer. Recently, several reports on broadband spin inversion created by phase-modulated pulse sequences have been made[1,12] and attention has focused on the use of such techniques because not many spectrometers possess single-sideband transmitting capability. However, as is well known, the use of a pulse of complex amplitude is equivalent to the employment of a real amplitude pulse with frequency modulation. Hence, our experiment may also be performed with an ordinary ring modulator and a field sweep. Thus Eq. (3) may also be solved if

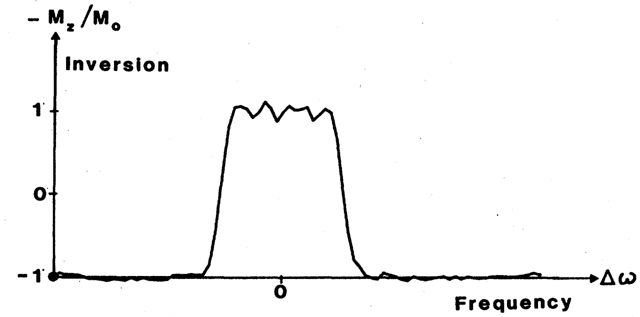

FIG. 3. Selective spin inversion generated by application of a hyperbolic secant, amplitude modulated B_1 pulse, and a hyperbolic tangent B_0 field sweep [Eq. (22)].

$$\Omega = \Omega_0\,\text{sech}\beta t, \quad \Delta B_0 = -\frac{\mu\beta}{\gamma}\tanh\beta t\ , \tag{22}$$

where ΔB_0 is the change in the main field. Figure 3 shows the results obtained.

In conclusion, we have shown that the Bloch-Riccati equation yields an analytical solution for a highly selective, power-independent spin version when the driving function is of a complex sech form. These results are of importance to those performing selective-spin-inversion, spin-decoupling, and adiabatic rapid-passage experiments. We consider the phenomenon to be the NMR analog of self-induced transparency of coherent optics.

Note added. During the reviewing process, a similar theoretical analysis was published independently by F. T. Hioe [Phys. Rev. A **30**, 2100 (1984)] and we are grateful that our attention was drawn to his work.

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*Present address: Siemens Medical Systems, Inc., 186 Wood Avenue South, Iselin, NJ 08830.

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