Two-Level Systems with Relaxation

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A method is presented for the coherent control of two-level systems when T_2 relaxation is significant. The Bloch equations are rewritten as an equation of motion of the stereographic projection, Γ, of the spin vector. This allows a Schur-type iteration used for the design of shaped pulses in magnetic resonance and coherent optics to be extended to include the effect of T_2. In general, the effect of T_2 on Γ cannot be completely compensated for, although in practice it can be to a high degree. An example is presented of a driving field that produces a coherent superposition (no on-diagonal elements of the density matrix) over a chosen band of frequencies, in the presence of relaxation.

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A long-standing problem in the coherent control of two-level systems has been the effect of relaxation while a driving field is being applied. For example in magnetic resonance imaging (MRI), a driving field is typically used to selectively "excite" spins into a coherent superposition, with spins with only a particular range of Larmor frequencies being excited. This enables images to be obtained of an object one slice at a time [1]. Relaxation (i.e., the return of excited spins to equilibrium on a time scale T_1 and the decoherence of spins on a time scale T_2 due to spin-spin interactions) makes this selection less sharp, with less uniform excitation within the slice [2]. Relaxation is also of critical importance in the control of qubits [3,4], imposing a limit on the number of gates that can be implemented in a quantum computer, and in optical coherence techniques [5,6], including the design of ultrafast optoelectronic devices [7].

A great deal is known about two-level systems when relaxation is neglected. In particular, it is known [8] how to design the driving field (or "pulse shape") that will cause the system to end up in any reasonable final state, from a given reasonable initial state (what is "reasonable" is described later). All methods to do this are essentially inverse scattering algorithms for the Zakharov-Shabat eigenvalue problem [9].

When the relaxation times are comparable to, or shorter than, the total duration of the driving field, relaxation can no longer be neglected. In practice, T_1 is often much longer than T_2 and can still be ignored. However, even with this simplification, remarkably little is known about the analytic properties of such systems. No general analytic methods of designing driving fields in the presence of T_2 relaxation are known. It is not even known what constitutes a reasonable final state to specify when calculating a driving field.

It is shown below that the stereographic projection is a particularly convenient way to describe the state when considering a two-level system with T_2 relaxation. A method of compensating for the effect of T_2 relaxation is derived. Crucially, however, it is shown that it is not possible to entirely undo this effect, although it can be undone to a large degree in many cases.

The state of a two-level system without relaxation can be described by a spinor $\psi = \binom{\psi_1}{\psi_2}$ that evolves as [10]

$$\frac{d\psi}{dt} = -\frac{i}{2}\begin{pmatrix} \omega_3 & \omega^\star(t) \\ \omega(t) & -\omega_3 \end{pmatrix}\psi = \frac{1}{i\hbar}\hat{H}\psi, \tag{1}$$

where $\omega_3 = \Delta E/\hbar$ is proportional to the energy separation of the two levels, ΔE, and $\omega(t) = \omega_1(t) + i\omega_2(t)$ is a (complex valued) driving field causing transitions between the levels ($\star$ is the complex conjugate). It is common to define [11] $m = m_x + im_y = 2\psi_1^\star\psi_2$ and $m_z = \psi_1\psi_1^\star - \psi_2\psi_2^\star$, giving the Bloch equations

$$\frac{d\boldsymbol{m}}{dt} = \boldsymbol{\omega} \times \boldsymbol{m}, \tag{2}$$

where $\boldsymbol{m} = (m_x, m_y, m_z)$ and $\boldsymbol{\omega} = (\omega_1(t), \omega_2(t), \omega_3)$. In NMR, $\boldsymbol{m}$ would be interpreted as the magnetization of the sample, and in coherent optics as the fictitious vector polarization. In this Letter, it will be referred to as the spin vector. Equation (1) can also be written as an equation of motion of the (traceless) density matrix ρ,

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[\hat{H}, \rho], \tag{3a}$$

identifying

$$\rho = \begin{pmatrix} m_z & m \\ m^\star & -m_z \end{pmatrix}. \tag{3b}$$

However, for the purposes of designing pulse shapes, it is particularly convenient to recast Eq. (1) as a Riccati equation in the stereographic projection $\Gamma = \psi_2/\psi_1$ [10],

$$\frac{d\Gamma}{dt} = -\frac{i}{2}\omega(t) + i\omega_3\Gamma + \frac{i}{2}\omega^\star(t)\Gamma^2. \quad (4)$$

Knowledge of Γ and $\boldsymbol{m}$ are equivalent, since [12]

$$\Gamma = \frac{m}{|\boldsymbol{m}| + m_z}, \quad (5)$$

and conversely,

$$m = 2|\boldsymbol{m}|\frac{\Gamma}{1+\Gamma\Gamma^\star}, \qquad m_z = |\boldsymbol{m}|\frac{1-\Gamma\Gamma^\star}{1+\Gamma\Gamma^\star}. \quad (6)$$

Here, $|\boldsymbol{m}| = \psi_1\psi_1^\star + \psi_2\psi_2^\star$ is constant in time, assumed known, and often taken as equal to 1.

In the coherent control of systems, it is often required to find a driving field that will cause the system to evolve from an initial state $\Gamma_i = 0$ (for all ω_3) to a final state Γ_f, specified as a function of ω_3. The states are specified as functions of ω_3, in order that the driving field works in the presence of inhomogeneous broadening or, as in MRI, in the presence of a magnetic field gradient. A particularly simple method to calculate such a driving field is to use a Schur-type iteration [13–15]. Without loss of generality, let $\omega(t)$ be a train of n impulses, θ_j, with separation T,

$$\omega(t) = \sum_{j=1}^{n}\theta_j\delta(t - jT). \quad (7)$$

Γ then evolves under the discrete Riccati equation,

$$\Gamma_j = \frac{t_j + z\Gamma_{j-1}}{1 - zt_j^\star\Gamma_{j-1}}, \quad (8)$$

with $z = \exp(i\omega_3 T)$ and $t_j = -i\tan(|\theta_j|/2)\exp[i\arg\theta_j]$, noting that θ_j are, in general, complex valued. Here, $\Gamma_j = \Gamma(t = jT_+)$, i.e., the state immediately following the jth impulse. If Γ_{j-1} can be analytically continued in z from the unit circle ($|z| = 1$, i.e., real ω_3) to $z = 0$, then, from Eq. (8), $\Gamma_j(z = 0) = t_j$.

The algorithm therefore works by setting the final state, Γ_n, equal to the desired state (specified as a function of z), $\Gamma_n(z) = \Gamma_f(z)$. Therefore t_n is simply $t_n = \Gamma_n(z = 0)$. The evolution is then peeled back by one time step by solving Eq. (8) for Γ_{n-1},

$$\Gamma_{n-1} = \frac{1}{z}\frac{\Gamma_n - t_n}{1 + t_n^\star\Gamma_n}. \quad (9)$$

Then $t_{n-1} = \Gamma_{n-1}(z = 0)$, and so on, going backwards in time until all further values of t_j are zero, or close to zero. The values of t_j always tend to zero (as j decreases) for a final state given as a rational polynomial. In practice, then, a reasonable state is one that can be written as a rational polynomial in z (even with poles on the unit circle—which then allows spin inversion, i.e., $m_z = -|\boldsymbol{m}|$, for one or more values of real ω_3). Once the set $\{t_1, t_2, \ldots, t_n\}$ is known, the corresponding impulses θ_j can be calculated (modulo 2π).

When relaxation is not neglected, the Bloch equations are modified to [16]

$$\frac{d\boldsymbol{m}}{dt} = \boldsymbol{\omega}\times\boldsymbol{m} - \begin{pmatrix} m_x/T_2 \\ m_y/T_2 \\ (m_z - m_0)/T_1 \end{pmatrix}, \quad (10)$$

where m_0 is the equilibrium length of $\boldsymbol{m}$. There are no longer equivalent spinor or Riccati equations of motion, and therefore inverse scattering techniques are not immediately usable for calculating driving fields.

The Bloch equations provide a good physical picture of the effect of a driving field and relaxation on a system. However, in order to calculate a driving field, the most convenient representation is again the stereographic projection [Eq. (5)]. In particular, when T_1 relaxation is neglected, system (10) can be written

$$\frac{d\Gamma}{dt} = -\frac{i}{2}\omega(t) + i\left[\omega_3 + \frac{i}{T_2}\frac{1-|\Gamma|^2}{1+|\Gamma|^2}\right]\Gamma + \frac{i}{2}\omega^\star(t)\Gamma^2, \quad (11)$$

i.e., as the Riccati Eq. (4), but with a Γ-dependent imaginary shift in ω_3.

$\Gamma(t)$ is now no longer sufficient to fully determine $\boldsymbol{m}(t)$, since $|\boldsymbol{m}|$ is not constant in time,

$$|\boldsymbol{m}(t)| = |\boldsymbol{m}(0)|\exp\left[-\frac{4}{T_2}\int_0^t\frac{|\Gamma|^2}{(1+|\Gamma|^2)^2}dt'\right], \quad (12)$$

and therefore the complete history of Γ up to time t is needed to determine the state at time t.

The similarity of Eqs. (4) and (11) suggest that the Schur-type algorithm described before can be adapted to obtain a unique driving field giving a final state with stereographic projection Γ_f. It is not possible to additionally specify a desired final $|\boldsymbol{m}|$.

Given a pulse sequence (7), the discrete Riccati Eq. (8) is modified by T_2 relaxation as

$$\Gamma_j = \frac{t_j + z\Gamma_{j-1}f_+(\Gamma_{j-1})}{1 - zt_j^\star\Gamma_{j-1}f_+(\Gamma_{j-1})}, \quad (13)$$

where

$$f_+(\Gamma) = e^\epsilon\frac{|\Gamma|^2 - 1 + \sqrt{4e^{-2\epsilon}|\Gamma|^2 + (|\Gamma|^2-1)^2}}{2|\Gamma|^2}, \quad (14)$$

and $\epsilon = T/T_2$. Then f_+ can be written as a series in ϵ,

$$f_+(\Gamma) = 1 + \frac{|\Gamma|^2 - 1}{|\Gamma|^2 + 1}\epsilon + \frac{1}{2}\frac{(|\Gamma|^2-1)(|\Gamma|^4 + 4|\Gamma|^2 - 1)}{(1+|\Gamma|^2)^3}\epsilon^2 + \cdots, \quad (15)$$

and this will converge, irrespective of $|\Gamma|$, if $\epsilon < \pi/2$.

Let both t_j and Γ_j also be written as series in ϵ,

$$t_j = t_j^{(0)} + t_j^{(1)}\epsilon + t_j^{(2)}\epsilon^2 + \cdots, \quad (16)$$

and

$$\Gamma_j = \Gamma_j^{(0)} + \Gamma_j^{(1)}\epsilon + \Gamma_j^{(2)}\epsilon^2 + \cdots, \quad (17)$$

with the zeroth order $t_j^{(0)}$ as would be calculated ignoring relaxation to give Γ_f. They are therefore known. $\Gamma_n^{(0)}$ will then correspond to the desired final stereographic projection, and $\Gamma_n^{(r>0)}$ will determine the rth order error due to relaxation and the higher-order terms $t_j^{(r>0)}$. The aim is to minimize the total error by choice of $t_j^{(r>0)}$.

It can be shown that $\Gamma_j^{(0)}$ is a (known) rational polynomial of z; i.e., it can be written $\Gamma_j^{(0)} = p_j(z)/q_j(z)$, where both p_j and q_j are polynomials in z of order $j-1$. Without loss of generality, q_j is chosen so that $q_j(0) = 1$. Similarly, let $\bar{\Gamma}_j^{(0)}(z) = \Gamma_j^{(0)\star}(1/z^\star)$. Then $\bar{\Gamma}_j^{(0)}$ equals $\Gamma_j^{(0)\star}$ for z on the unit circle and can be written $\bar{\Gamma}_j^{(0)} = \bar{p}_j(z)/\bar{q}_j(z)$, where $\bar{p}_j$ and $\bar{q}_j$ are polynomials in z of order $j-1$, and $\bar{q}_j$ is chosen to have coefficient of z^{j-1} equal to 1.

The first order term $\Gamma_n^{(1)}$ can be shown to equal

$$\Gamma_n^{(1)} = \sum_{j=1}^{n} \beta_j \frac{z^{n-j}}{q_n^2}[q_{j-1}q_j t_j^{(1)} + z p_{j-1} p_j t_j^{(1)\star}] + \frac{p_n^{(1)}}{z^{n-3} q_n^2}, \tag{18a}$$

where

$$\beta_j = \prod_{k=j+1}^{n} 1 + |t_k^{(0)}|^2,$$

$$p_n^{(1)} = -\sum_{j=1}^{n-1} \frac{\beta_{n-j}^2}{\beta_0} z^{2(j-1)} p_{n-j} q_{n-j}[q_{n-j}\bar{q}_{n-j} - p_{n-j}\bar{p}_{n-j}]. \tag{18b}$$

A key property of $\Gamma_n^{(1)}$ is that it has a series expansion in z, about $z=0$ of the form

$$\Gamma_n^{(1)} = \frac{a_{3-n}}{z^{n-3}} + \cdots + \frac{a_{-1}}{z} + a_0 + a_1 z + \cdots + a_{n-1} z^{n-1} + \cdots. \tag{19}$$

Coefficients $a_{k<0}$ do not depend on the $t_j^{(1)}$. Each coefficient $a_{k\geq 0}$ depends only on $t_{n-k}^{(1)}, \ldots, t_n^{(1)}$ (i.e., not on $t_1^{(1)}, \ldots, t_{n-k-1}^{(1)}$).

If there are no zeros of $q_n(z)$ inside (or on) the unit circle, then the series expansion of $\Gamma_n^{(1)}$ is valid at least as far as the unit circle. Therefore the L_2 measure of first order error in the response, $E^{(1)} = \int_{-\pi}^{\pi} |\Gamma_n^{(1)}(z = \exp[i\theta])|^2 \, d\theta$, is minimized by minimizing the sum

$$|a_{3-n}|^2 + \cdots + |a_{-1}|^2 + |a_0|^2 + |a_1|^2 + \cdots + |a_{n-1}|^2 + \cdots. \tag{20}$$

If n is sufficiently large, it can be assumed that $a_k = 0$ for $k \geq n$. Additionally, the $|a_{k<0}|^2$ terms cannot be modified by changing the $t_j^{(1)}$. Then sum (20) is minimized by requiring $a_0 = a_1 = \cdots = a_{n-1} = 0$. Since a_0 depends (linearly) on $t_n^{(1)}$, but has no dependence on $t_{j<n}^{(1)}$, solving $a_0 = 0$ determines a value for $t_n^{(1)}$. Then solving $a_1 = 0$ fixes a value for $t_{n-1}^{(1)}$, and so on. The minimum value of $E^{(1)}$ is then

$$E_{\min}^{(1)} = 2\pi(|a_{3-n}|^2 + |a_{4-n}|^2 + \cdots + |a_{-1}|^2). \tag{21}$$

This sequential method of obtaining the $t_j^{(1)}$ and the proof that a residual minimum error exists are the main results of this Letter.

This method is easily generalized to when q_n has zeros inside the unit circle. For any polynomial $f(z)$ with zeros z_j inside the unit circle, let

$$B\{f\} = f \times \prod_j \left[\frac{1 - z z_j^\star}{z - z_j}\right]. \tag{22}$$

The Blaschke factors $(1 - z z_j^\star)/(z - z_j)$ move all the zeros to outside the unit circle, but preserve $|f(z)|$ for $|z| = 1$ [17]. Replacing the q_n terms in the denominators in Eq. (18a) by $B\{q_n\}$ allows the series expansion of $\Gamma_n^{(1)}$ to be valid on the unit circle, but does not effect $E^{(1)}$.

In the pathological, but important, case, when q_n has zeros (i.e., $\Gamma_n^{(0)}$ has poles) on the unit circle, giving inversion of the spin vector for real ω_3, it is more meaningful to minimize $|\Gamma_n^{(1)}/\Gamma_n^{(0)2}|$ over the unit circle. Then the previous method can be used, except q_n in the denominators of Eq. (18a) should be replaced by p_n.

This method can be applied to the higher-order errors. For all $r \geq 1$, $\Gamma_n^{(r)}$ has the form

$$\Gamma_n^{(r)} = \sum_{j=1}^{n} \beta_j \frac{z^{n-j}}{q_n^2}[q_{j-1} q_j t_j^{(r)} + z p_{j-1} p_j t_j^{(r)\star}] + \frac{p_n^{(r)}}{z^{n(2r-1)-(4r-1)} q_n^{r+1}}, \tag{23}$$

where $p_n^{(r)}$ is a polynomial in z that does not depend on the $t_j^{(r)}$, but does depend on the lower order pulse values $t_j^{(s<r)}$, and hence is (in principle) known, assuming all the $t_j^{(s<r)}$ have already been determined.

The same arguments as before can then be used when $r > 1$ to choose the $t_j^{(r)}$ to minimize the L_2 error $E^{(r)} = \int_{-\pi}^{\pi} |\Gamma_n^{(r)}(\exp[i\theta])|^2 \, d\theta$. Minimizing successive errors $E^{(r)}$ does not guarantee that the total error $E = \int_{-\pi}^{\pi} |\Gamma_n - \Gamma_n^{(0)}|^2 \, d\theta$ is minimum. However, in practice it works well, as illustrated below.

For example, consider the target final stereographic projection

$$\Gamma_f = \frac{-i(z+1)^8}{B\{(z+1)^8 + [2(z-1)]^8\}}. \tag{24}$$

As this has no poles inside the unit circle, this corresponds to a minimum-phase [18], hence minimum-energy [12], target (however, the results here can be applied to any phase characteristic, e.g., linear or self-refocused [12]). Since, for $z = \exp(i\omega_3 T)$,

$$|\Gamma_f| = \frac{1}{1 + (2\tan\omega_3 T/2)^8}, \tag{25}$$

Eq. (24) corresponds to a Butterworth-type selective function for real ω_3.

The solid line in Fig. 1 shows $|\Gamma_f|$ versus ω_3 for $T = 1$. In the "stop-band," $|\omega_3| \gtrsim 1$, $\Gamma_f \approx 0$ and hence [Eq. (6)] $\boldsymbol{m}$ would be along z after the pulse sequence.

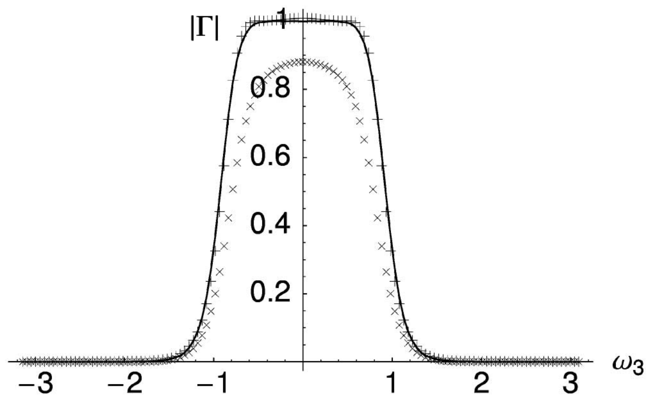

FIG. 1. Solid line: $|\Gamma_f|$ with Γ_f defined in Eq. (24). Crosses: response with $\epsilon = 0.1$ to impulses θ_j of Fig. 2. Pluses: response to $\tilde{\theta}_j$.

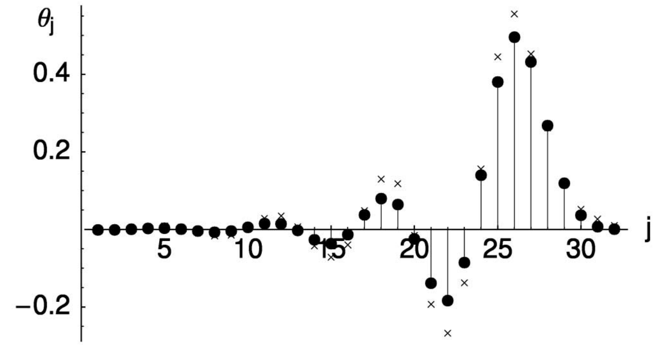

FIG. 2. Solid circles: impulses θ_j, $j = 1, \ldots, 32$, corresponding to the target, Eq. (24), neglecting relaxation. Crosses: impulses $\tilde{\theta}_j$ with relaxation compensated to first order, with $\epsilon = 0.1$.

Equivalently, the density matrix ρ would have no off-diagonal elements. In the "pass-band," $|\omega_3| \lesssim 1$, $|\Gamma_f| \approx 1$, and hence $\mathbf{m}$ would be in the x-y plane after the pulse sequence (equivalently, ρ would have no on-diagonal elements). The pulse sequence would therefore selectively create a coherent superposition depending on ω_3.

Thirty-two–element impulse sequences were calculated from this target response, first neglecting relaxation (giving sequence θ_j), then compensating relaxation to first order in ϵ (and taking $\epsilon = 0.1$), giving sequence $\tilde{\theta}_j$. These are shown as circles and crosses, respectively, in Fig. 2. The symmetry of Γ_f $[\Gamma_f(z) = -\Gamma_f^\star(z^\star)]$ can be shown to result in the impulses all being real valued.

Figure 1 shows the calculated responses (with $\epsilon = 0.1$) to these two pulse sequences compared to $|\Gamma_f|$. The impulses θ_j have a response, shown as crosses in Fig. 1, significantly different from Γ_f. This is in contrast to the impulses $\tilde{\theta}_j$ (the response shown as pluses in Fig. 1), which have an associated total error $E \approx 0.0017$. Although only $|\Gamma|$ is shown, the phase of Γ is also very close to that of Γ_f after the sequence $\tilde{\theta}_j$. The error E can be improved by using higher-order corrections. For example, $E \approx 0.00021$ after applying a fourth-order correction.

In conclusion, the Bloch equations describing the evolution of a spin vector $\mathbf{m}$ under a driving field and with T_2 relaxation decouple to a single Riccati-type equation in Γ, the stereographic projection of $\mathbf{m}$, and an expression for $|\boldsymbol{m}(t)|$ in terms of Γ. An analytic method exists to compensate for the effect of relaxation on Γ. This works well even for relaxation times short in comparison to the total pulse time, e.g., in the previous example, $\epsilon = T/T_2 = 0.1$. Since the total pulse duration was $\tau = 32T$, then $T_2 \approx 0.3\tau$. It is possible to uniformly excite (in the sense described in the example) a two-level system over a given bandwidth in the presence of relaxation. Two important caveats are, first, that it is not possible, in general, to completely undo the effect of T_2 on Γ, although the residual effect can often be made very small. Second, since the algorithm calculates a unique pulse shape, there is no subsequent freedom to influence the final magnitude of $\mathbf{m}$.

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[1] A. N. Garroway, P. K. Grannell, and P. Mansfield, J. Phys. C **7**, L457 (1974).

[2] D. G. Norris, H. Lüdemann, and D. Leibfritz, J. Magn. Reson. **92**, 94 (1991).

[3] J. A. Jones, Fortschr. Phys. **48**, 909 (2000).

[4] K. M. Fonseca-Romero, S. Kohler, and P. Hänggi, Chem. Phys. **296**, 307 (2004).

[5] W. S. Warren and M. Haner, in *Proceedings of NATO Advanced Research Workshop on Atomic and Molecular Processes with Short Intense Laser Pulses, Lennoxville, Quebec*, edited by A. D. Bandrauk (Plenum Press, New York, 1987), p. 1.

[6] D. Goswami, Phys. Rep. **374**, 385 (2003).

[7] A. Neogi, H. Yoshida, T. Mozume, and O. Wada, Opt. Commun. **159**, 225 (1999).

[8] D. E. Rourke and P. G. Morris, Phys. Rev. A **46**, 3631 (1992).

[9] V. E. Zakharov and A. B. Shabat, Zh. Eksp. Teor. Fiz. **61**, 118 (1971) [Sov. Phys. JETP **34**, 62 (1972)].

[10] F. Bloch and A. Siegert, Phys. Rev. **57**, 522 (1940).

[11] E. T. Jaynes, Phys. Rev. **98**, 1099 (1955).

[12] D. E. Rourke and J. K. Saunders, J. Magn. Reson. **115**, 189 (1995).

[13] J. Schur, J. Reine Angew. Math. **147**, 205 (1917).

[14] J. Schur, J. Reine Angew. Math. **148**, 122 (1918).

[15] P. Le Roux, in *Abstracts of the 8th Annual Meeting and Exhibition of the Society of Magnetic Resonance in Medicine, Amsterdam, 1989* (Society of Magnetic Resonance in Medicine, Berkeley, 1989), p. 1168.

[16] F. Bloch, Phys. Rev. **70**, 460 (1946).

[17] S. G. Krantz, *Handbook of Complex Variables* (Birkhäuser, Boston, 1999), Chap. 9.

[18] J. Pauly, P. Le Roux, D. Nishimura, and A. Macovski, IEEE Trans. Med. Imag. **10**, 53 (1991).