PHYSICAL REVIEW A VOLUME 22, NUMBER 2 AUGUST 1980

Theorem on coherent transients

Axel Schenzle,* N. C. Wong, and Richard G. Brewer
IBM Research Laboratory, San Jose, California 95193
and Department of Applied Physics, Stanford University, Stanford, California 94305
(Received 14 January 1980)

A fundamental theorem in nuclear magnetic resonance and quantum optics is derived for an atomic system subject to inhomogeneous broadening and coherent preparation by a pulsed electromagnetic field of arbitrary shape. We show that the coherent emission, free induction decay or echo, which follows is confined to an interval which precisely equals the duration of the applied pulse sequence. The theorem applies to two-level or multilevel transitions for which the inhomogeneous linewidth is infinitely broad and the sample is optically thin.

It is well known in nuclear magnetic resonance (NMR)[1,2] and quantum optics[3,4] that a two-level quantum system can emit coherent radiation after it is prepared by a pulsed resonant electromagnetic field. This effect, known as free induction decay (FID), has been analyzed over the years through solutions[5,6] of the Maxwell and Schrödinger wave equations and exhibits different properties for transitions which are dominated either by homogeneous or inhomogeneous line broadening. For the case of strong inhomogeneous broadening, as in the optical regime, particular analytic solutions which are nonlinear in the field amplitude have been found recently[7] for the thin-sample regime when the inhomogeneous linewidth is infinitely broad, and reveal a striking and fundamental characteristic which appears to have gone unnoticed in the previous NMR and optics literature. Earlier studies[6-9] indicate that *when a pulse of finite duration T (interval $0\leq t\leq T$) prepares a sample, the coherent emission which follows lasts only for an additional period T (interval $T\leq t\leq 2T$).* An example of a calculation[7] which illustrates this behavior is shown in Fig. 1 where the emission for $t>2T$ is zero. The oscillatory behavior, which appears for large pulse areas, is also unusual and is discussed elsewhere.[7] These calculations as well as supporting NMR[8] and infrared[6] experiments suggest that the above statement is actually a theorem. We now prove on very general grounds that this is the case without recourse to particular solutions. The theorem holds for arbitrary population (T_1) and dipole (T_2) decay times, and arbitrary pulse shapes of finite duration T, and can be generalized to multilevel transitions as well. Since the pulse shape is arbitrary, the theorem is equally valid for two- or multiple-pulse echo sequences.

We assume that an electromagnetic pulse of arbitrary shape

$$E(z,t)=\begin{cases}E_0(t)\cos(\Omega t-kz)\,, & 0<t<T\\ 0\,, \quad t<0\,,\ t>T\end{cases} \tag{1}$$

and of duration T coherently prepares an atomic sample. For a two-level quantum system, the atomic density-matrix equations of motion take the familiar form

$$\left(\frac{d}{dt}-i\Delta+\frac{1}{T_2}\right)\tilde{\rho}_{12}=\tfrac{1}{2}i\chi w\,, \tag{2a}$$

$$\frac{dw}{dt}=-i\chi(\tilde{\rho}_{21}-\tilde{\rho}_{12})-\frac{(w-w^0)}{T_1}\,, \tag{2b}$$

where the upper and lower states are $|2\rangle$ and $|1\rangle$, the population difference $w\equiv\rho_{22}-\rho_{11}$, the rapidly oscillating terms in the Schrödinger equation are removed with the substitution $\rho_{12}=\tilde{\rho}_{12}e^{i(\Omega t-kz)}$, and the nonresonant high-frequency terms are neglected. Furthermore, the Rabi frequency $\chi(t)\equiv\mu_{12}E_0(t)/\hbar$ and the tuning parameter $\Delta\equiv-\Omega+\omega_{21}+kv_z$, where μ_{12} is the transition matrix element, ω_{21} is the $1\leftrightarrow 2$ level splitting, and kv_z is a Doppler shift. For an optically thin sample, Maxwell's wave equation yields an emission signal field $E(t)$ which is essentially given by

$$E(t)\sim\langle\tilde{p}(t)\rangle=N\,\mathrm{Tr}\,\langle\mu\tilde{\rho}(t)\rangle\,, \tag{3}$$

where $p(t)$ is the atomic polarization. The bracket denotes an average over the inhomogeneous lineshape, which for an atomic gaseous system is

$$g(\Delta)=\exp[-(\Delta/\sigma)^2]/\sqrt{\pi}\,\sigma, \tag{4}$$

the Doppler width being σ.

By formally integrating (2a) and performing the Doppler average, we obtain the general expression

$$\langle\tilde{\rho}_{12}(t)\rangle=\int_{-\infty}^{\infty}g(\Delta)\tilde{\rho}_{12}(0)e^{(i\Delta-1/T_2)t}d\Delta+\frac{i}{2}\int_{-\infty}^{\infty}d\Delta\int_0^t g(\Delta)w(\Delta,t')\chi(t')e^{(i\Delta-1/T_2)(t-t')}dt'\,. \tag{5}$$

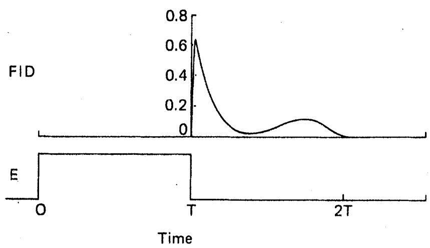

FIG. 1. A computer plot is shown of the free induction decay (upper curve) following a preparative field pulse (lower curve) of amplitude E, duration T, and pulse area $\chi T = 2\pi$. This calculation, which is described in detail in Ref. 7, follows from a solution of the Bloch equations and a numerical integration over an inhomogeneous line shape of finite width σ.

The first term of (5) can be eliminated with the assumption that coherent preparation does not precede the pulse; i.e., $\tilde{\rho}_{12}(0) = 0$.

We now restrict the discussion to physical systems with very large inhomogeneous broadening and will therefore evaluate Eq. (5) in the limit $\sigma \to \infty$, keeping Ng(0) constant. Under the assumption that the order of integration can be interchanged, we first perform the integration over the detuning parameter Δ by closing the path of integration in the upper half-plane and find

$$\oint w(\Delta, t')e^{i\Delta(t-t')}d\Delta = 2\pi i \sum_{(\text{uhp})} R\,, \qquad (6)$$

where the sum is taken over the residues R in the upper half-plane. The contribution to the integral from the half-circle contour can be evaluated easily in the limit of infinite radius $|\Delta| \to \infty$ when we recall that the solution of the Bloch equations has the formal structure[10]

$$w(\Delta, t) = \sum_j A_j \exp(z_j t) + \text{const}\,, \qquad (7)$$

where the quantities z_j are the roots of the characteristic equation

$$(z + 1/T_1)(z + 1/T_2)^2 + \Delta^2(z + 1/T_1) + \chi^2(z + 1/T_2) = 0\,.$$

In the asymptotic limit $|\Delta| \to \infty$ the roots assume the values

$$\lim_{|\Delta| \to \infty} z_j = \begin{cases} -1/T_1\,, \\ -1/T_2 \pm i\Delta\,, \end{cases} \qquad (8)$$

causing the integrand on the circular contour to vanish exponentially when the following relation is satisfied:

$$t > 2t'\,. \qquad (9)$$

Notice that (9) appears to be incompatible with the domain $t > t' > 0$ defined by the integration limits in (5). This difficulty vanishes, however, when allowance is made for the finite pulse, Eq. (1), which changes the upper limit of the time integral from $t \to T$ so that (5) becomes

$$\langle \tilde{\rho}_{12}(t) \rangle = \frac{i}{2\sqrt{\pi}\,\sigma} \int_{-\infty}^{\infty} d\Delta \int_0^T w(\Delta, t')\chi(t')e^{(i\Delta - 1/T_2)(t-t')}dt'\,. \qquad (10)$$

The restricted domain $T > t' > 0$ of (10) is now consistent with (9) when we assume that

$$t > 2T \geq 2t'\,.$$

Equation (10) is a very compact general expression as it includes the preparation or transient nutation effect for the period $0 < t < T$ and FID for the interval $t > T$.

We now evaluate (6). Since the density-matrix equations (2) are a set of first-order differential equations which depend linearly on the parameter Δ, the solutions according to a theorem of Poincaré[11] are entire functions and therefore will be analytic in Δ and will contain *no poles* in the finite complex plane. Consequently, the integral (6) vanishes and we conclude that the emission signal (10) is identically zero for $t > 2T$,

$$\langle \tilde{\rho}_{12}(t) \rangle \equiv 0\,, \quad t > 2T\,. \qquad (11)$$

Since the pulse shape according to (1) is arbitrary, this theorem applies equally well to an entire pulse train. For example, in a two-pulse echo experiment where T is the sum of the two-pulse widths and the delay time, the echo signal terminates precisely at $t = 2T$.

The above discussion assumes an infinite inhomogeneous linewidth $\sigma \to \infty$. The effect of finite σ can be seen analytically if we replace the Gaussian lineshape (4) by the Lorentzian $g(\Delta) = (\sigma/\pi)/(\Delta^2 + \sigma^2)$ and assume a pulse where in (1), $|E_0(t)| < E_0$. We find an upper bound for the polarization (5) by contour integration in the uhp about the pole $\Delta = i\sigma$, and upon integration over time, we obtain

$$|\langle \tilde{\rho}(t) \rangle| \leq (N/4\sigma)f(\sigma)\chi e^{-\sigma(t-2T)}\,, \quad t > 2T\,, \qquad (12)$$

where we have used the formal properties[7] of the Bloch equations giving the upper limit $|w(i\sigma, t')| \leq f(\sigma)e^{\sigma t'}$, where $f(\sigma)$ is analytic. Hence, a large finite inhomogeneous linewidth σ produces a rapid decay for $t > 2T$, in a time $T_2^* \equiv 2/\sigma$, and thus the theorem stands as an excellent approximation for this case as well.

All of these arguments can be extended to echoes or FID within a multilevel quantum system where the energy spacing, the number of applied fields, and their frequencies are arbitrary. The density-matrix Eqs. (2) assume an obvious form given elsewhere.[10] For any one-photon transition $i \to j$, the FID signal is

$$\langle\tilde{\rho}_{ij}(t)\rangle = \int_{-\infty}^{\infty} g(\Delta)\tilde{\rho}_{ij} e^{(i\Delta_{ij} - 1/T_{ij})t} d\Delta + \frac{i}{2}\int_{-\infty}^{\infty} d\Delta \sum_k \int_0^t g(\Delta)\tilde{\rho}_{ik}(\Delta, t')\chi_{kj}(t') \times e^{(i\Delta_{ij} - 1/T_{ij})(t-t')} dt' + \text{c.c.}, \quad (13)$$

where

$$\Delta_{ij} = \omega_{ij} - \Omega_{ij} \pm kv_z .$$

Since the structure of (13) conforms to (5), the previous arguments apply and the same conclusion follows.

The simplicity of this theorem suggests that a simple physical description of the origin of this effect might exist. Thus far, this approach has proved elusive because the FID or echo depends nonlinearly on the applied field and the inhomogeneous broadening introduces a nonobvious interference among the coherently prepared packets.

It is interesting that the fundamental nature of this phenomenon has escaped attention until now. The theorem should prove useful, therefore, in interpreting future coherent transient experiments, especially at optical frequencies.

This work was supported in part by the U. S. Office of Naval Research. N. C. Wong would like to thank the Hertz Foundation for financial support.

---

*On leave from the Department of Physics, Universität Essen, Gesamthochschule, Essen, West Germany.

[1]A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).
[2]C. P. Slichter, *Principles of Magnetic Resonance* (Springer, Berlin, 1978).
[3]M. Sargent, III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, Mass., 1977).
[4]L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).
[5]E. L. Hahn, Phys. Rev. 77, 297 (1950).
[6]R. G. Brewer and R. L. Shoemaker, Phys. Rev. A 6, 2001 (1972).
[7]A. Schenzle, N. C. Wong, and R. G. Brewer, Phys. Rev. A 21, 887 (1980).
[8]A. L. Bloom, Phys. Rev. 98, 1105 (1955).
[9]F. Hopf and M. O. Scully, Phys. Rev. 179, 399 (1969).
[10]A. Schenzle and R. G. Brewer, Phys. Rep. 43, 455 (1978); A. Schenzle and R. G. Brewer, Phys. Rev. A 14, 1756 (1976).
[11]J. H. Poincaré, *Les Méthodes Nouvelles* de la *Mécanique Céleste* (Dover, New York, 1957), Vol. 1, p. 48. See also, F. R. Moulton, *Differential Equations* (Dover, New York, 1958), p. 33.