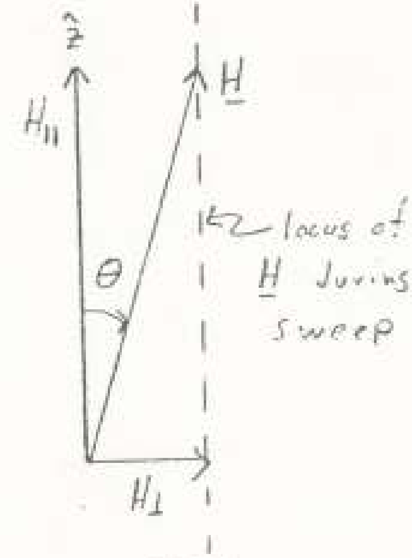
Dynamic Response of Spins to Time Varying Magnetic Fields.

S - 10 - Special Notes

"Resonance" at zero field; adiabatic and sudden passage
R.H. Silsbee, January 1972



A large optical signal is observed, in absence of a radio frequency field, when the total (earth's plus applied) field is swept through zero. The situation is as illustrated, where $\hat{z}$ is taken parallel to the axis of the coils #1 and the earth's field, but where we recognize that the alignment is unlikely to be perfect and that there is probably a residual component, $H_{\perp}$, transverse to the coil axis. We wish to know what happens to the z component of the magnetization, M_z, which determines the optical transparency of the cell, as $H_{\parallel}$ is reversed in sign. We suppose that the passage through zero field occurs rapidly compared with the time in which the optical pumping process can change populations significantly so that the changes in $\underline{M}$ are solely due to the variation in $\underline{H}$.

Classically the spins obey the equation of motion

$$\frac{d\underline{M}}{dt} = \gamma(\underline{M} \times \underline{H}) \; ;$$

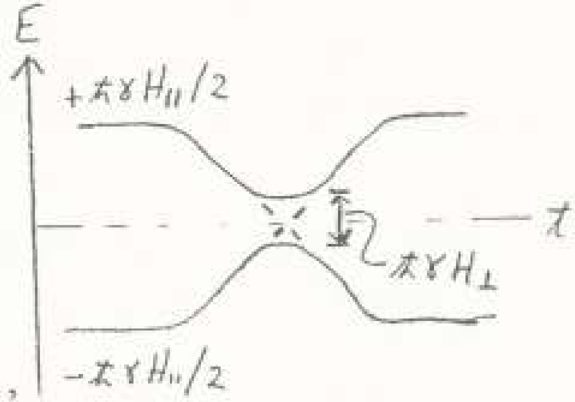
The problem looks (and is) very complicated because of variation of $\underline{H}$ with t, but two important limiting cases may be examined. Suppose the variation of $H_{\parallel}$ is "slow", such that $\underline{M}$ precesses many times about $\underline{H}$ during the time θ changes by a small amount. Then $\underline{M}$ will remain very nearly parallel to $\underline{H}$ and after $\underline{H}$ is reversed, $\underline{M}$ will also have reversed. This is referred to as adiabatic passage and the condition that the variation be "slow" is that

$$(1/H_{\perp})\; dH_{\parallel}/dt \ll \gamma H_{\perp} \qquad (1)$$

or that the rate of change of the angle θ be small compared with the minimum precession rate of the magnetization. This reversal of M_z should result in a large optical signal. The other limit is when $H_{\parallel}$ changes "rapidly", such that the magnetization has no opportunity to follow the field. In this case the magnetization is left in its original direction and the change in optical signal is small. The condition for "rapid" change is of course the inverse of the inequality (1). The simplest case to see is that for $H_{\perp} = 0$. In this case $\underline{H}$ is always parallel or antiparallel to $\hat{z}$, $\underline{M} \times \underline{H} = 0$ always and there can be no torque to invert the spins; $\underline{M}$ is left in its original orientation.

The same qualitative conclusions follow for a quantum mechanical picture for a single spin of one half with the time dependent hamiltonian

$$H(t) = -i\gamma \underline{S}\cdot\underline{H}(t)$$



with the time variation of $\underline{H}$ being that discussed above. Reference to a quantum mechanics text (Schiff. 209 & 211) gives two approaches to a problem such as this when H(t) becomes time independent as $t \to \pm\infty$. If the system is prepared in an eigenstate of $H(-\infty)$ and H varies slowly (adiabatically) in time, the system remains in an eigenstate of the "instantaneous" hamiltonian, and follows this state continuously from $t = -\infty$ to $t = +\infty$. A system initially in the lower energy state ($\underline{M} \parallel \underline{H}$) remains in that state as $\underline{H}$ is reversed and ends up still in the lower energy state, again $\underline{M} \parallel \underline{H}$ giving a reversed magnetization as $\underline{H}$ is adiabatically reversed. On the other hand, if H changes "rapidly", the sudden approximation is valid, and the state occupancies after the transition are obtained by projecting the initial state at $t = -\infty$ onto the eigenstates of $H(+\infty)$, the squares of these projections giving the final state occupations. Since the lower state for $H(-\infty)$ is identical to the upper energy state for $H(+\infty)$, both corresponding to the spin parallel to the initial $\underline{H}$ field, in this sudden passage the spin is left without change of orientation. The inequality validating the quantum adiabatic limit is, of course, just equation 1 and the sudden approximation is valid for the reverse inequality. Again, if $H_{\perp}$ is zero, the levels cross and the sudden approximation is valid whatever the sweep rate.

I know of no exact solution to this problem but see Weger; Bell Syst. Tech. Jour. 39, 1013, '60 (see pages 1028-1030); which gives expressions for the changes in magnetization in the nearly sudden and nearly adiabatic limits. Let $x \equiv \pi\gamma H_{\perp}^2/(dH_{\parallel}/dt)$;

M_o = magnetization at $t = -\infty$, M_f = magnetization after passage;

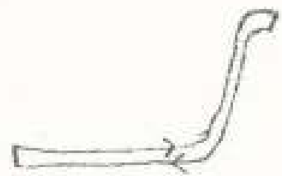
then

$$x \ll 1;\ \text{Sudden limit};\ (\Delta M/M_o) \simeq x^2 \qquad \Delta M \equiv M_o - M_f$$

$$x \gg 1;\ \text{Adiabatic limit};\ (\Delta M/M_o) \simeq e^{-x} \qquad \Delta M \equiv |-M_o - M_f|$$

These expressions are plotted on the graph on the next page and connected by a smooth curve which should give a fair representation to the change in M_z for arbitrary x. As discussed later, this curve is also appropriate for discussing the changes in M_z in the resonance experiment if $H_{\perp}$ is replaced by the magnitude of the radio frequency magnetic field.

Experimentally it is easy to demonstrate the presence of the adiabatic passage; the sudden passage is less easy to show because it is difficult to obtain a sufficiently small $H_\perp$ over the full volume of the optical pumping cell without extensive magnetic shielding. To demonstrate the adiabatic passage, the field is swept back and forth through zero field at a rate somewhat faster than the optical pumping time. If the sweep is centered on zero field one sees the illustrated pattern of successive sweeps giving weak signals always of the same sign, corresponding to the recovery of the optical signal during the half sweep period following the last passage. As the sweep frequency is increased, this signal amplitude decreases because of the shorter recovery time between successive passages. If, however, the mean field is displaced so that the passage occurs near one end of the sweep, then a much larger signal is seen, and is of opposite sign on successive passages. Here the field is of one polarity during almost the whole modulation cycle and nearly the full optical polarization is allowed to develop. Near the end of the sweep, an adiabatic passage reverses the magnetization giving a large signal; before this reversed population is depleted by the optical pumping process, a second adiabatic passage quickly restores the system nearly to the state obtaining before the first passage. Thus two successive adiabatic passages can serve to sample the existence of a large magnetization, without destroying it, allowing the observation of large signals at modulation frequencies high compared with the inverse of the optical pumping time. (This should be demonstrated using a triangular sweep. With a trapezoidal sweep, the dwell time either side of resonance is nearly the same even though the sweep is not centered on the resonance.) If the transverse field is now tuned to zero, the signal should disappear, corresponding to no change in M_z during a sudden passage. Because of inhomogeneities in the transverse field, it is difficult to achieve the sudden condition with a strong inequality, but the change in pattern is striking as one tunes for the minimum mean square transverse field. The signal amplitude is substantially reduced and now has the same sign on successive passages, indicating loss but not reversal of the magnetization.



The distinction between adiabatic and sudden passage is also relevant to a discussion of the resonance at non-zero $H_{\parallel}$ in the presence of radio frequency (r.f.) magnetic fields. By transforming to a coordinate frame, (see Rabi, Ramsey and Schwinger) rotating at the frequency of the radio frequency field, the above considerations can be shown to be equally relevant to the resonance problem.

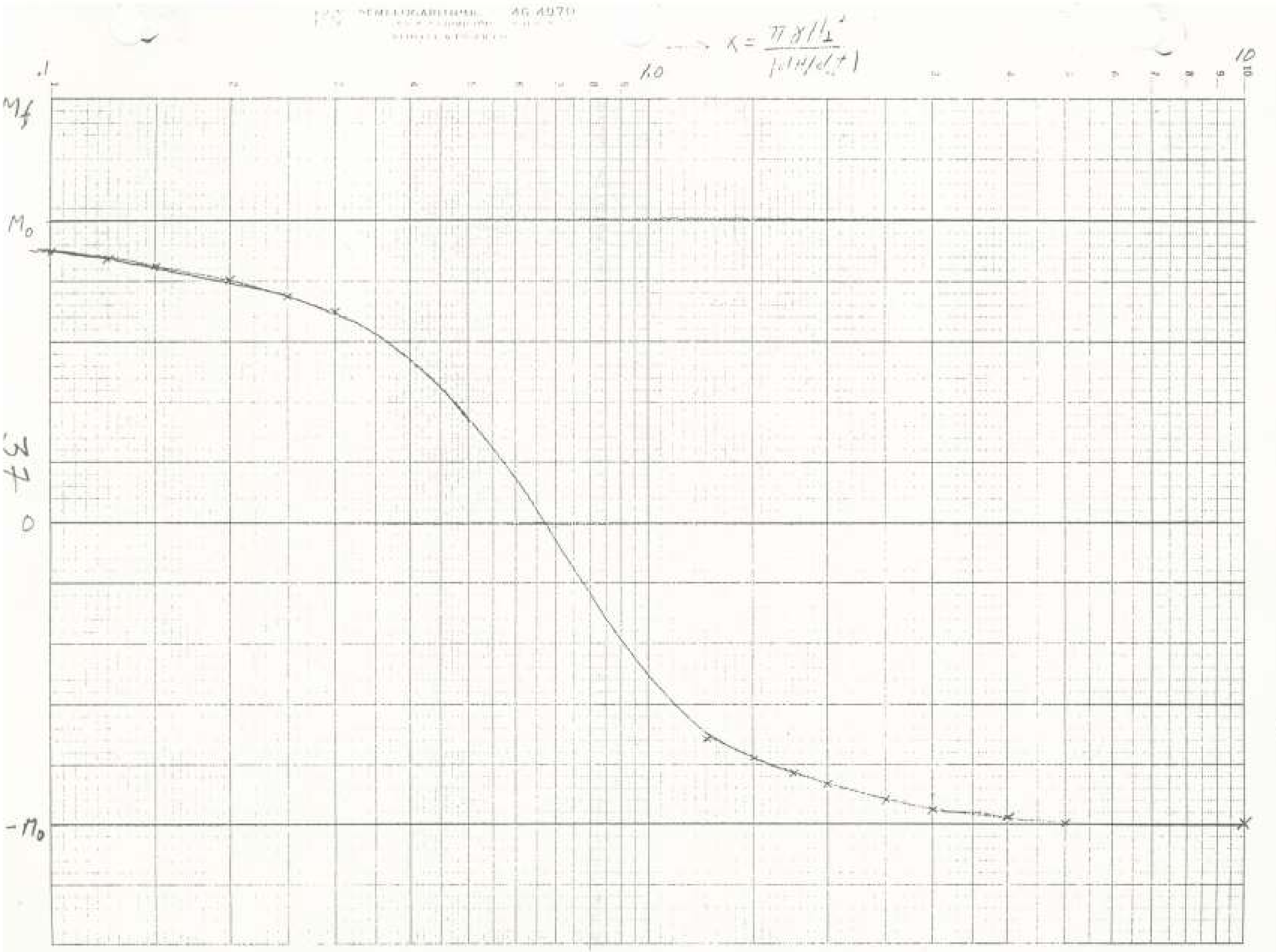
The crucial translations in the arguments are that the "resonance" condition $H_{||} = 0$ is now replaced by

$H_{||} - W_{r.f.}/\gamma = 0$ and $H_{\perp}$ is replaced by $H_{r.f.}$

The adiabatic condition, $\gamma H_{r.f.}{}^2 >> dH_{||}/dt$,

can be achieved at moderate r.f. power levels if the r.f. coil driving the pumping cell is resonated with a capacitor. The presence of the adiabatic passage is demonstrated, just as in the zero field case, by using a triangular sweep with asymmetric dwell times above and below the resonance. The transition to the sudden limit is achieved simply by reducing the amplitude of the r.f. field. The extreme sudden limit is the trivial observation that in absence of an r.f. field, no transitions are induced.

Because the static transverse field and its inhomogeneities play no role in the r.f. experiment (their effects are averaged out by the rapid precession about $H_{||}$), the transition between the adiabatic and sudden approximations may be usefully pursued quantitatively in this experiment, a task made rather difficult in the zero field experiment. On the other hand it is, of course, conceptually much easier to understand the dynamic behavior of the spins in the zero field case. The transformation to a rotating coordinate frame demonstrates the essential equivalence of these two situations.



37