Optical Bloch Equations for Low-Temperature Solids

In an elegant series of experiments, DeVoe and Brewer have recently shown[1] that the conventional optical Bloch equations (OBE) are not correct in the saturation regime. They point out that their observations are consistent with a phenomenological theory given by Redfield[2] in 1955 which provides weak-field and strong-saturation limiting expressions. At intermediate power levels the behavior has not been described analytically.

Here we present the results of a theoretical analysis of a simplified version of the DeVoe-Brewer observations. This analysis differs from Redfield's in that it is based on the theory of stochastic processes rather than on principles of thermostatics. It leads to a relatively simple analytic expression for the free induction decay (FID) linewidth that agrees well with the data of DeVoe and Brewer[1] at low and intermediate power levels as well as in the saturation limit (see Fig. 1).

The damping matrix in the conventional OBE is based on the assumption that a damping process is not affected by an incident field. We avoid this assumption.[3] Following the DeVoe-Brewer observations, such generalizations are of more than academic interest.

In the DeVoe-Brewer experiment on Pr^{3+}:LaF_3 the flip-flop motion of the F nuclear spins gives a time-dependent magnetic field at each Pr^{3+}. We assume that this random magnetic field creates a fluctuation $\delta\omega(t)$ in the resonance frequency of the Pr^{3+} ions. Such a fluctuation is well known to cause damping in the weak-field limit. At optical frequencies, the spontaneous emission rate 2γ $(=1/T_1)$ must also be considered.

Under these conditions we have obtained[3] an effective damping matrix Γ_{ij}. Here, for simplicity, we retain only the most important features of the matrix elements contributing to FID, and we assume that the autocorrelation of $\delta\omega(t)$ can be modeled by $f(\tau)=(\delta\omega)^2\exp(-\tau/\tau_c)$. The result is a simple damping theory in which the damping matrix has only diagonal elements: $\gamma_2(\Omega)$, $\gamma_2(\Omega)$, and 2γ, where $\gamma_2(\Omega)=\gamma+\int_0^\infty d\tau\, f(\tau)\cos\Omega\tau$,

It is now straightforward to compute the steady-state value of the Bloch vector during saturation, and then use this steady-state value as the initial condition for the FID phase of the experiment. If the inhomogeneous linewidth is great enough, then the FID signal is easily shown to decay at the rate

$$R=\frac{1}{T_2}+\left\{\gamma_2^2(\Omega)+\Omega^2\frac{\gamma_2(\Omega)}{2\gamma}\right\}^{1/2}, \qquad (1)$$

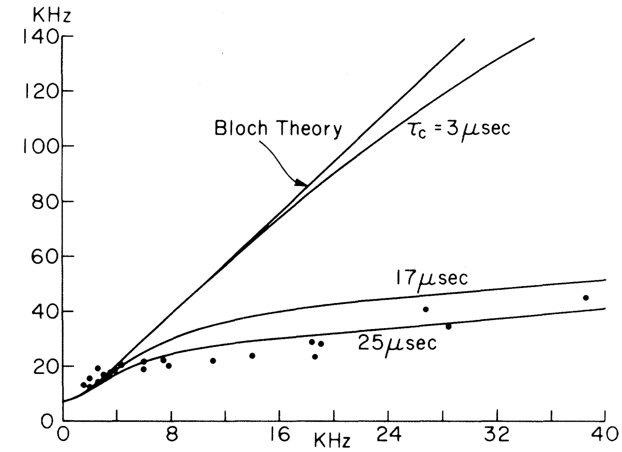

FIG. 1. Three curves showing R as a function of the Rabi frequency Ω on top of the data of Ref. 1. We have used $(\delta\omega)^2\tau_c = 45\times10^3$ sec^{-1} from known values of T_1 and T_2.

where $1/T_2=\gamma_2(0)$. Figure 1 shows R for three values of τ_c. The largest value gives the best fit. It is too close in value to T_2 for complete comfort. Nevertheless, the shape of the curve is clearly about right, for both low and high Rabi frequencies, and expression (1) provides a possibly useful analytic interpolation formula between the two limits. We will describe our theory in detail elsewhere, without the drastic simplifications adopted here.

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[1]R. G. DeVoe and R. G. Brewer, Phys. Rev. Lett. **50**, 1269 (1983).

[2]A. G. Redfield, Phys. Rev. **98**, 1787 (1955).

[3]See M. Yamanoi, in *Coherence and Quantum Optics V*, edited by L. Mandel and E. Wolf (Plenum, New York, 1984), and references therein.