Experimental Test of the Optical Bloch Equations for Solids

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This paper provides the first experimental test of the Bloch equations for an optical transition in a low-temperature impurity-ion solid, $Pr^{3+}:LaF_3$, where the condition $T_1 \gg T_2$ applies. Intensity-dependent optical free-induction-decay measurements are performed by use of an ultrastable cw dye laser (linewidth, 300 Hz) and reveal an $\exp(-2t/T_2)$ decay law at low Rabi frequency (χ), as predicted by the Bloch equations, and an $\exp(-\chi t)$ decay at high χ, which violates the Bloch prediction but agrees with Redfield's characterization of NMR saturation in solids.

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It is commonly assumed in quantum optics and laser spectroscopy that the optical Bloch equations[1,2] provide a fundamental starting point for the quantum description of nonlinear optical phenomena such as saturation and coherent optical transients. The Bloch equations incorporate relaxation processes in the two-level density-matrix equations by two phenomenological parameters, the (diagonal) population decay time T_1 and the (off-diagonal) dipole dephasing time T_2. Implicit in their use is the assumption that T_1 and T_2 are independent of laser intensity. In gas-phase atomic or molecular systems, where $T_1 \sim T_2$, this premise appears to be justified.[3] However, in low-temperature solids, where $T_1 \gg T_2$, the assumption that T_2 is independent of laser intensity may break down in a manner resembling Redfield's description[4,5] of nuclear-magnetic-resonance saturation in solids. In fact, it is surprising in view of the extensive number of earlier laser studies of solids that the validity of the optical Bloch equations in the saturation regime has gone unchallenged.

In this Letter,[6] we report the first detailed study of saturation behavior in a coherently excited, low-temperature, impurity-ion solid, $Pr^{3+}:LaF_3$, and show using measured values of T_1 and T_2 that the Bloch prediction becomes invalid at elevated laser intensities. Instead, we find agreement with Redfield's saturation theory.[4]

Our technique examines the intensity dependence of optical free-induction decay (FID). The Pr^{3+} ions are coherently prepared in a narrow packet of the inhomogeneously broadened line by an ultrastable cw dye laser, and FID follows when the excitation is suddenly removed, the decay time being dependent on the Pr^{3+} saturation behavior and hence the laser intensity in the preparative period.

Consider now the optical FID solution[7] of the Bloch equations

$$\dot{u} + \Delta v + u/T_2 = 0, \tag{1a}$$

$$\dot{v} - \Delta u - \chi w + v/T_2 = 0, \tag{1b}$$

$$\dot{w} + \chi v + (w - w^0)/T_1 = 0, \tag{1c}$$

and also the prediction which results from Redfield's modification[4] of these equations. Equations (1) follow from the 2×2 density-matrix equations of motion for a two-level quantum system, with frequency splitting ω_{21}, interacting with an electromagnetic field of frequency Ω. The Bloch vector amplitudes u, v, and w are related to the density-matrix elements by $\rho_{12} = \frac{1}{2}(u + iv)e^{i\Omega t}$, the dipole term, and $\rho_{22} - \rho_{11} = w$, the population difference term. We also define the tuning parameter $\Delta = -\Omega + \omega_{21}$ and the Rabi frequency as χ. With the assumption of steady-state preparation of an inhomogeneously broadened transition, the Bloch decay law for the heterodyne FID signal,[7] derived from (1), is of the form

$$F_B(t) \sim \exp\{-t[1/T_2 + (1/T_2^2 + \chi^2 T_1/T_2)^{1/2}]\} \tag{2a}$$

$$\rightarrow \begin{cases} \exp(-2t/T_2), & \chi^2 T_1 T_2 \ll 1, \quad (2b) \\ \exp\{-t[1/T_2 + \chi(T_1/T_2)^{1/2}]\}, & \chi^2 T_1 T_2 \gg 1, \quad (2c) \end{cases}$$

where (2b) and (2c) express the low- and high-power limits, respectively. Note in (2a) that the $1/T_2$ term is the dephasing contribution of the FID period whereas the square-root term is the hole-burning contribution of the preparative stage.

Redfield's modification[4] of the Bloch equations involves substituting for T_2 in (1a) an effective dephas-

ing time T_{2e} which is power dependent with limiting values of T_2 at low power and T_1 at high power. At intermediate powers, the behavior is more complex and has not been described analytically. The FID solution becomes

$$F_R(t) \sim \exp\{-t[1/T_2 + (1/T_2 T_{2e} + \chi^2 T_1/T_{2e})^{1/2}]\} \quad (3a)$$

$$\rightarrow \exp\{-t(1/T_2 + \chi)\}, \quad \chi^2 T_1 T_2 \gg 1, \quad (3b)$$

where (3b) expresses the high-power limit, and the low-power limit coincides with (2b). By comparing (2c) and (3b), it will be apparent that in a solid where $T_1 \gg T_2$ the Bloch model saturates as $\chi(T_1/T_2)^{1/2}$, considerably faster than the Redfield result, χ. We now demonstrate that the FID measurements support the Redfield prediction (3b) and not the Bloch result (2c).

The Pr^{3+} transition examined, the lowest crystal-field–split components of the $^1D_2 \leftarrow {}^3H_4$ states at 5925 Å, was chosen because the quantities T_1 and T_2 are well known. In addition, a Monte Carlo calculation[8] and a magic-angle line-narrowing experiment[9] have precisely defined the line-broadening mechanism: The Pr^{3+} ion dephases because of ^{19}F-^{19}F nuclear-spin flip-flops which modulate the Pr^{3+} optical transition frequency via the Pr-F dipolar interaction.

To monitor saturation and FID of a single Pr^{3+} packet, we have actively stabilized a cw ring dye laser to a linewidth narrower than any presently known optical transition in a solid. An rms stability of <300 Hz, corresponding to a fractional linewidth of 6×10^{-13}, has been achieved by an FM sideband phase-locking technique.[10] This technique is far superior to an earlier method of locking to the side of a transmission fringe of a reference cavity for two reasons. First, it utilizes a frequency-locking signal that is shot-noise limited because of the noise-cancellation property of FM detection.[11] Secondly, it permits a faster servo response time with consequently higher servo gain because the laser error signal is instantaneously reflected off the reference etalon rather than undergoing a 100-nsec delay in transmission. A pair of identical 50-cm confocal Invar etalons, acoustically and thermally isolated, serve as the frequency reference and as an independent stability-monitoring system.

Optical FID and nutation are observed by laser frequency switching as described previously.[12] The frequency-stabilized beam first enters an acousto-optic Bragg modulator which acts as an optical gate and a laser frequency switch. The beam then passes through a pair of lenses of 5 and 10 cm focal length that produce beam diameters in the range 0.08 to 1 mm and, with neutral density filters, intensities in the sample from 50 mW/cm^2 to 50 W/cm^2. The generally elliptical beam cross sections are measured with a Reticon diode array to an accuracy of 0.015 mm. The linearly polarized laser field propagates along the c axis of a $5\times 6\times 7$-mm^3 crystal of 0.1-at.% Pr^{3+}:LaF_3 at 1.6 K before striking a p-i-n photodiode An external dc magnetic field of 500 G is applied perpendicular to the c axis. The Pr^{3+} ions are coherently prepared when the modulator is driven by a 400-μsec rf pulse at 95 MHz. At the end of the pulse, the laser frequency is abruptly switched from 95 to 97 MHz for a 100-μsec period, causing FID to occur in the initially prepared packet and nutation in the newly excited packet. Since the sample is illuminated for only 500 μsec every 2 sec, optical pumping is minimized.

The FID signals, which are not averaged, exhibit excellent reproducibility and signal-to-noise ratio as indicated in Fig. 1, a computer plot of 400 experimental points overlaid on an exponentially damped cosine function. The two curves appear to be indistinguishable, and the residuals

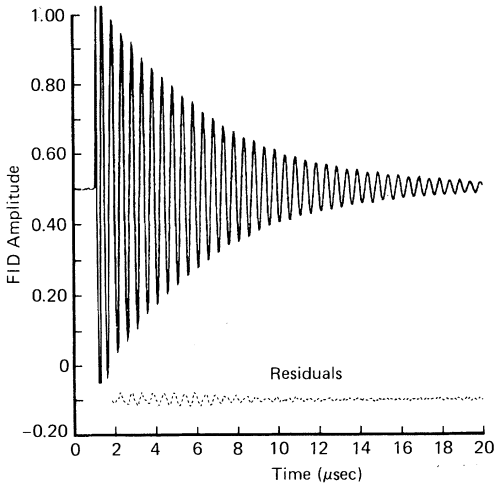

FIG. 1. A computer plot of 400 points of optical FID of 0.1-at.% Pr^{3+}:LaF_3 at 1.6 K. The experimental data are overlaid on a damped cosine, and the residuals indicate that the dephasing time of 5.10 μsec has an uncertainty of less than 1%. The signal is power broadened.

show that the dephasing time can be determined with an uncertainty of less than 1%.

In Fig. 2 is plotted the linewidth of the prepared hole $\Delta\nu$ versus the Rabi frequency $\chi/2\pi$. For the Bloch theory, $\Delta\nu = (1/2\pi)(1/T_2^2 + \chi^2 T_1/T_2)^{1/2}$ half width at half maximum (HWHM), and for the FID experiments, $\Delta\nu = (1/2\pi)(1/\tau - 1/T_2)$, where τ is the observed dephasing time. Here, $T_1 = 0.5$ msec,[13] and χ is obtained directly from the fit of nutation signals by Bloch theory below saturation, which yields 4.7×10^{-4} D for the transition moment. Above saturation, the nutation signals deviate from Bloch predictions and are not used. A heterdyne photon-echo measurement gives $T_2 = 21.7$ μsec in agreement with an extrapolation of the FID data to zero power, Eq. (2b), and a previous value.[14] Scatter in the data is due to the uncertainty in χ.

We see that the two curves of Fig. 2 agree in the low-power regime up to $\chi/2\pi = 3$ kHz and thereafter diverge rapidly. At elevated power, the Bloch prediction approaches $(\chi/2\pi)(T_1/T_2)^{1/2}$, while the experimental linewidths approach $\chi/2\pi$, the ratio of the two being 4.8. The observations, therefore, are in striking agreement with the Redfield prediction (3b).

To establish that the deviation from Bloch saturation behavior, Fig. 2, represents a fundamental effect and not an artifact of our technique, we have examined the effect on FID of (a) finite-duration versus steady-state preparation pulses, (b) spin double-resonance processes arising from fortuitous Zeeman tuning, (c) the intensity dependence of the heterodyne probe pulse, and (d) a theoretical model that incorporates a multilevel instead of a two-level quantum system.

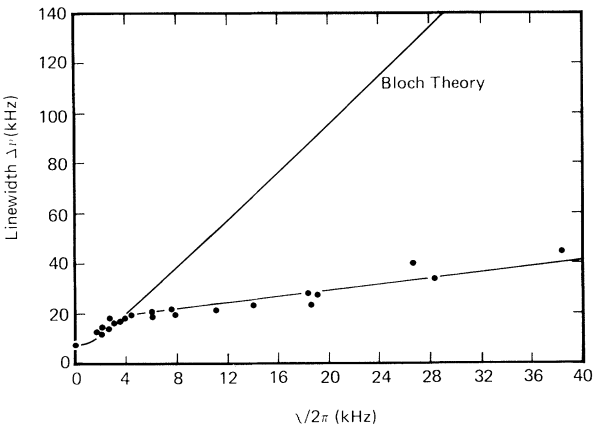

FIG. 2. Optical linewidth $\Delta\nu$ (HWHM) of the prepared hole vs the Rabi frequency $\chi/2\pi$ according to the Bloch theory (upper curve) and FID experiments of Pr^{3+}:LaF_3 (filled circles) where the solid curve is a best fit to the data. The point $\Delta\nu = 1/2\pi T_2$ at $\chi = 0$ results from a photon echo experiment.

(a) Numerical FID solutions of Eq. (1) show that a 400-μsec pulse preparation time (the experimental value) yields a dephasing time that falls less than 20% below the steady-state value (2a). Furthermore, experiments with pulses in the range 400 μsec to 1 msec also reveal a 20% variation in τ in agreement with the numerical solutions. Hence, our transient excitation technique permits a valid comparison with steady-state Bloch theory. (b) By increasing the external magnetic field from 100 to 5000 G in six steps at two fixed Rabi frequencies, $\chi/2\pi = 10$ and 20 kHz, we noted no change in the dephasing time to within 5%. This test rules out the possibility of chance coincidences of the Zeeman-split F and Pr^{3+} hyperfine states that could produce a double resonance effect. (c) By varying the heterodyne probe pulse in the range 4 μW to 4 mW while keeping the preparation pulse intensity fixed ($\chi/2\pi = 20$ kHz), we find that the FID dephasing time remains unchanged to within 5%. Hence, the anomalous dephasing effect seen in heterodyne photon echos is excluded.[14] (d) As is well known, the Bloch equations (1) apply only to a two-level quantum system, and yet population relaxation in Pr^{3+}:LaF_3 involves three or more levels as evidenced by a strong optical pumping cycle. We have numerically calculated the influence of optical pumping and hence a multilevel system on FID by assuming a two-level system ($1 \rightharpoonup 2$) that is now in contact with an n-level reservoir.[15] With this model, the branching ratio of the optically excited state was varied by 5 orders of magnitude, and yet the dephasing time changed by less than 20%. It should also be noted that a preparation pulse of 400 μsec duration prevents more than 30% of the population from leaking out of the optically excited two-level system. Hence, optical pumping of a multilevel configuration does not play an important role.

We have shown that optical saturation in Pr^{3+}:LaF_3 is consistent with Redfield's predictions.[4] It is possible, however, that while our result resembles Redfield's effect phenomenologically, it could have a different physical origin. We briefly sketch which concepts used to understand this phenomenon in NMR are likely to be valid in the optical region. First, Redfield's explanation for the replacement of T_2 by T_1 in Eq. (1a) arises because the decay of u is partially forbidden by energy and entropy considerations above saturation,

an effect that can create local spin order in neighboring nuclei. In the optical case, this mechanism would explain the lengthening of T_2 for Pr^{3+} at high χ since the fluorine spin fluctuations would be reduced because of local F spin order. We plan to test this hypothesis by Raman heterodyne detection of NMR.[16] A second possibility is that the fluorine nuclei are not ordered by the laser, but rather that their coupling to the Pr^{3+} optical transitions via the Pr^{3+}-F dipolar interaction is averaged out by the high optical Pr^{3+} Rabi frequency. We note that the related effect, Pr^{3+} optical line narrowing by applied rf fields which are resonant with the F nuclei, has already been observed in this system.[9] On the other hand, the concept of spin temperature in the rotating frame is not applicable to $Pr^{3+}:LaF_3$ since at 0.1% concentration, the Pr^{3+} ions do not exchange optical energy, and hence cannot come to equilibrium as in a pure solid.

These studies thus emphasize that the optical dephasing parameter T_2 is not always appropriate for coherently excited solids when saturation conditions prevail. Secondly, it seems plausible that other impurity-ion solids should behave similarly to $Pr^{3+}:LaF_3$ in obeying Redfield behavior. Finally, we note that a detailed microscopic theory of optical saturation in solids awaits development.

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