# Experimental test of the modified optical Bloch equations for solids using rotary echoes

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Measurements of optical rotary-echo decay of the $R_1(-\frac{1}{2}),\sigma$ transition of $Cr^{3+}$ in dilute ruby at 2 K are described and compared with predictions of two recent modifications (Gauss-Markov and random-telegraph models) of the optical Bloch equations (OBE's). While the modified OBE approximately describes the free-induction decay behavior both in $Pr^{3+}$:$LaF_3$ and ruby, we show that rotary-echo decay in ruby is instead described by the standard OBE. Various aspects of the spin flip-flop model used in the theories are discussed in regard to ruby, as well as other possible dephasing contributions.

Recent experiments[1] on the free-induction decay (FID) behavior of the $^1D_2$ transition in the solid, $Pr^{3+}$:$LaF_3$, have shown a major deviation from predictions by the well-known optical Bloch equations[2] (OBE). DeVoe and Brewer[1] have suggested that this behavior may occur generally in impurity-ion solids in which dephasing is due to frequency fluctuations induced by host spin flip-flops. In response to this remarkable experiment, several theoretical studies have appeared[3–10] using various statistical models for the frequency fluctuations. The two most widely studied models are the Gaussian-Markov[8] (GM) and random telegraph[9] (RT) each of which can be made to fit the data of Ref. 1 reasonably with an appropriate value for the correlation time $\tau_c$ of the frequency fluctuations. However, as emphasized by Berman,[10] there are inconsistencies in the GM fit and further experiments have been suggested to test different predictions of the theory, e.g., hole-burning shape measurements,[11] rotary-echo decays,[7] and extension to gases.[10] Another requirement of the theory is to give an exponential decay for the photon echo, as observed experimentally. This is fulfilled for both the GM and RT theories (the latter only for $\tau_c \ll T_2$, where $T_2$ is the dephasing time), but it is violated for the model studied by Javanainen.[5] A stronger test of the theories is afforded by rotary echoes[12,13] in which the optical-field–sample interaction is maintained during the entire preparation and observation process. Since the new terms which appear in the modified OBE are intensity dependent, we show that a marked nonexponential decay occurs for both the GM and RT models in contrast to the exponential decay predicted by the standard OBE. In this paper we present experimental and theoretical studies of the modified OBE using rotary echoes in ruby. These studies, together with FID observations,[14] provide, we believe, a comprehensive test of the Gauss-Markov and random-telegraph modified optical Bloch equations.

A dilute ruby sample ($1.6\times4\times10$ $mm^3$, 0.0034 wt. % $Cr_2O_3$), at a temperature of 2 K, was used in a dc magnetic field of 3.5 kG directed along the $c$ axis. The $^4A_2(-\frac{1}{2})\rightarrow\bar{E}(-\frac{1}{2})$ $Cr^{3+}$ transition (at 14 417.57 $cm^{-1}$ measured with a Burleigh wave meter) was resonantly excited using a focused, circularly polarized laser beam propagating along the $c$ axes. 65 mW of single-frequency power was available from a Coherent 699-21 dye laser that was narrowed to root-mean-square linewidth less than 700 Hz, using a FM locking technique.[15] This width is much smaller than the ruby homogeneous linewidth $(\pi T_2)^{-1}=22.7$ kHz. Transparent conducting films were applied to the two $4\times10$ $mm^2$ sample surfaces to allow generation of an electric field in the sample. Rotary echoes were produced by Stark-shifting[16] the energy levels by 25 MHz for 20 nsec at time $T$ (this required about a 38-V pulse) which effectively shifted the laser phase by $\pi$ rad, resulting in rephasing[12] of the nutation at the time region around $2T$. An acousto-optic modulator gated the beam on for 100 $\mu$sec at 10–100 Hz. Optical pumping effects on the decay time were not significant since single-shot data agreed, within 20%, with those averaged at faster rates. The light transmitted through the sample and a variable portion of the incident light was detected by photodiodes, subtracted to minimize amplitude noise and averaged by a Data Precision 6000 digital oscilloscope after preamplification. Finally, the data were transferred to a computer for storage and processing.

The echo amplitude signal $S_e$ was homodyne-detected by beating the incident field $E_0$ against the field $E_a$ emitted by the $Cr^{3+}$ ions so that $S_e\sim E_0E_a$. If the beam is assumed to have a Gaussian cross section, the echo signal at time $t$ is[12,17]

$$S_e(t)\sim\int_0^\infty\int_0^\infty v[T_1,T_2,\Delta,T,\chi(r),t]\exp-(\Delta/\Delta_0)^2 \times\exp-(r/r_0)^2 r dr\, d\Delta ,$$

where $v$ is a transverse component of the Bloch vector $(u,v,w)$, $\Delta_0$ is an inhomogeneous linewidth (1 GHz), $\chi(r)=\chi_0\exp-(r/r_0)^2$, where $\chi_0/2\pi$ is the Rabi frequency at beam center, $T$ is the phase-reversal time, and $r,r_0$ are beam radii. For ruby, $T_1=4200$ and $T_2=14$ $\mu$sec. Numerical methods were used to calculate $S_e$ in the time region $t=2T$ using a matrix analytic solution for the GM, RT, and normal OBE equations derived earlier.[18] In the calculations, The FID during the short phase-reversal time is neglected and the $\pi$ phase shift is produced by reversing the signs of $u$ and $v$ at time $T$.

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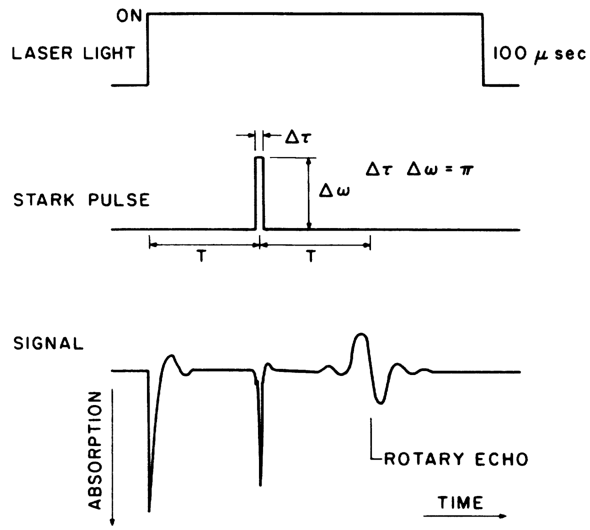


FIG. 1. Pulse sequences used to produce rotary echoes by Stark-shifting and a representative rotary-echo signal (bottom trace) obtained by averaging 256 traces on a Data Precision 6000 digital oscilloscope.

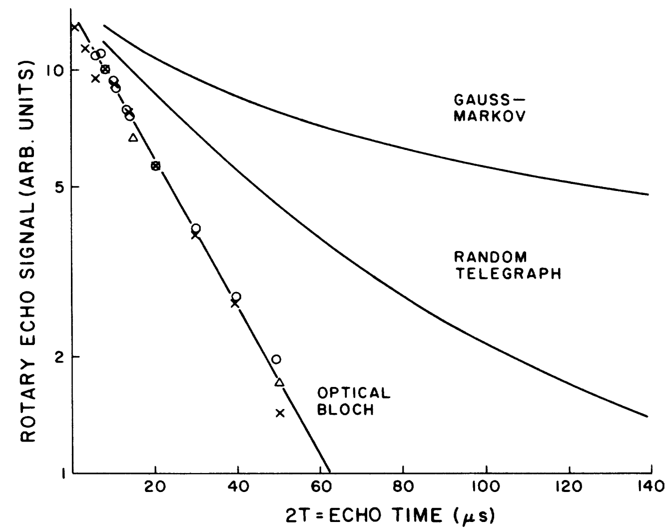


FIG. 2. Theoretical (——) and experimental rotary-echo decay in dilute ruby. The experimental points are for Rabi frequencies of 215 (△), 600 (○), and 1800 (×) kHz. Some Δ points are omitted for clarity since they fall on the × and ○ points. A correlation time $\tau_c = T_2 = 14$ μsec and a Rabi frequency of 1800 kHz are assumed for the theory (see text).

The optical and electrical pulse sequence used is shown in Fig. 1 along with a representative rotary-echo trace. The echo amplitude was taken as the peak-to-peak nutation signal centered at $2T$. Plots of the experimental echo amplitude versus echo time $2T$ are shown in Fig. 2 for various Rabi frequencies. $\chi_0$ was obtained from the observed period of the rotary-echo signal multiplied by a correction factor ($\sim$20%) calculated using the OBE. Also shown are three theoretical curves for $\chi_0/2\pi = 1.8$ MHz using the standard OBE, GM, and RT equations. The OBE curve is scaled vertically to fit the data with the GM and RT curves scaled by the same factor. For the range of $\chi_0/2\pi$ studied, 215–1800 kHz, the theoretical curves changed in shape by less than 5%. A limiting (long) value of $\tau_c = T_2 = 14$ μsec was chosen for the theoretical curves based on experimental FID data[14] which required $\tau_c \approx T_2$ for fitting to theory, similar to the $Pr^{3+}$:$LaF_3$ observations.[7]

It is evident from Fig. 2 that the experimental rotary-echo decay is closely described by the normal OBE (i.e., $\tau_c = 0$), a conclusion that is inconsistent with the FID results[14] which require $\tau_c \approx T_2$. We now consider various possibilities that could account for this discrepancy. (i) The dephasing mechanism in ruby is not determined by the host Al spin flip-flops and (ii) the theoretical model is incorrect.

(i) A principal difference between dopant-host spin interactions in $Cr^{3+}$:$Al_2O_3$ and $Pr^{3+}$:$LaF_3$ is that $Cr^{3+}$ has an electronic spin whereas the $Pr^{3+}$ spin is nuclear. Thus the frozen core of Al spins in ruby is much larger, extending to $\sim$400 Al's atoms (Ref. 19) according to the line-shift criteria of Ref. 20. However, beyond the core, the flipping Al should produce frequency fluctuations, as envisioned by the stochastic models. An important point to address is the possibility of direct Cr-Cr spin flip-flops[21,22] which act like a $T_1$ dephasing mechanism and could explain the observed exponential rotary-echo decay. However, concentration effects are small for our dilute sample, which has (for 3.5 kG) $T_2 = 14$ μsec (0.0034 wt. % $Cr_2O_3$) compared to $T_2 = 13$ μsec (0.005%),[23] 9 μsec (0.01%),[14] falling to 7 μsec at 0.013%.[23] Moreover, spin-spin relaxation measurements[24] with a time resolution of 1 μsec suggest that absence of fast spin flip-flops for 0.0034% material. The possibility of clumping effects (e.g., around defects) remains, however. Another is resonant, ground-state direct spin-flip dephasing by impurities (e.g., Fe). However, this is discounted by the magnetic field independence of the data at high fields.

(ii) Recently, Endo *et al.*[25] have proposed a nonstochastic model[26] in which dephasing occurs because of quantum interference in the multiline "molecule" created by the Cr-Al and Al-Al interactions. Such a model has also been proposed[27] earlier for electron magnetic resonance dephasing. Unfortunately no calculations are available which could indicate differences between the above model and the second-order time-dependent perturbation approach used for the modified OBE. Finally, more complex versions[10] of the stochastic models remain to be numerically investigated.

In conclusion, rotary-echo decay studies show that the Gaussian-Markov and random-telegraph dephasing models are not applicable to ruby. It would be of interest to extend these studies to confirm a similar conclusion advanced by Berman[10] for $Pr^{3+}$:$LaF_3$ from free-induction decay measurements.

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