NRC Publications Archive
Archives des publications du CNRC

Experimental tests of the optical Bloch equations for solids: spectral diffusion

Szabo, Alex

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. / La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version acceptée du manuscrit ou la version de l'éditeur.

Publisher's version / Version de l'éditeur:

Nonlinear Optics Conference Proceedings, 1990

NRC Publications Record / Notice d'Archives des publications de CNRC:
https://nrc-publications.canada.ca/eng/view/object/?id=8dff68df-9491-4a9d-83fc-66d63cfe9a83
https://publications-cnrc.canada.ca/fra/voir/objet/?id=8dff68df-9491-4a9d-83fc-66d63cfe9a83

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at
https://nrc-publications.canada.ca/eng/copyright
READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site
https://publications-cnrc.canada.ca/fra/droits
LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

National Research Council Canada
Conseil national de recherches Canada

To be published:
Vavilov '90
Nonlinear Optics Conf.
Proceedings.

EXPERIMENTAL TESTS OF THE OPTICAL BLOCH EQUATIONS FOR SOLIDS: SPECTRAL DIFFUSION

Alex Szabo

National Research Council of Canada
Ottawa, Ontario, K1A 0R6, Canada

INTRODUCTION AND REVIEW

Recent free induction decay (FID) studies by DeVoe and Brewer [1] have shown that the conventional optical Bloch equations (OBE) fail to describe the saturation behaviour of the $^1D_2$ line (592.5 nm) in Pr^{3+}:LaF_3. This is a surprising and important result both for technological and scientific reasons. For hole-burning memories [2], the effects of such power broadening will play a decisive role in the design of such systems. Scientifically, the study of dephasing (homogeneous linewidth) mechanisms in nuclear and electron magnetic resonance has been actively pursued for some 40 years. In particular, various models of host spin flip induced dephasing have been studied and recently extended to optical transitions [3]. Also several theoretical studies using various models of spin flip induced frequency fluctuations have appeared [4]-[12]. As discussed by Berman [10} however, none of the theories consistently describe the data which includes FID as well as photon echo observations.

Recently we have extended these high resolution FID studies to ruby [13], using an ultra-stable (~ 1 kHz linewidth) dye laser, to subject the theories to further tests with experiment. It might be noted that one contribution to the dephasing spin interaction in ruby occurs between electronic (Cr^{3+}) and nuclear (Al^{3+}) spins whereas in Pr^{3+}:LaF_3 only nuclear spins are involved.

In addition studies of the power dependence of rotary echoes [14] and hole-burning [15] in the frequency domain have been reported. These results of some of these studies are summarized in Fig. 1-2. The FID ruby data in Fig. 1 are similar to that obtained for Pr^{3+}:LaF_3 in that approximate fits of the Gauss-Markov (GM) [8] and random telegraph (RT) [9] modified OBE could be obtained using a correlation time τ_c=T_2, the dephasing time. On the other hand the rotary echo decay was found to disagree with the GM and RT models and was described by the standard OBE. Finally the hole-burning results in Fig. 2, obtained in the frequency domain, are inconsistent with the FID time domain derived hole-widths. However recent theoretical work [16]. that assumes a slow, strong stochastic modulation of the ion frequency appears to consistently describe the FID and rotary echo results. Regarding the frequency domain hole studies, preliminary results are presented in

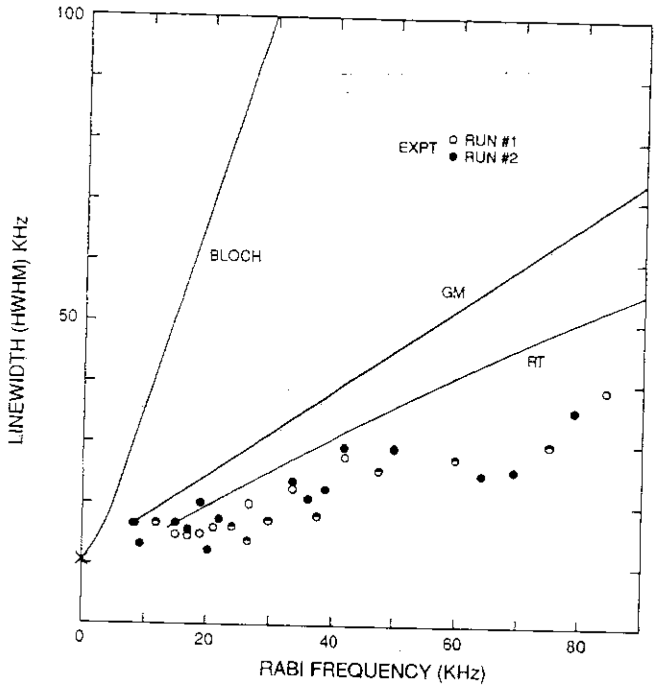

Fig. 1. Optical linewidth (HWHM) of the prepared hole vs Rabi frequency [13]. Theoretical curves for the normal OBE, GM and RT models are shown assuming $\tau_c = T_2 = 15 \mu sec$. The point marked X at zero Rabi frequency was obtained from a photon echo measurement.

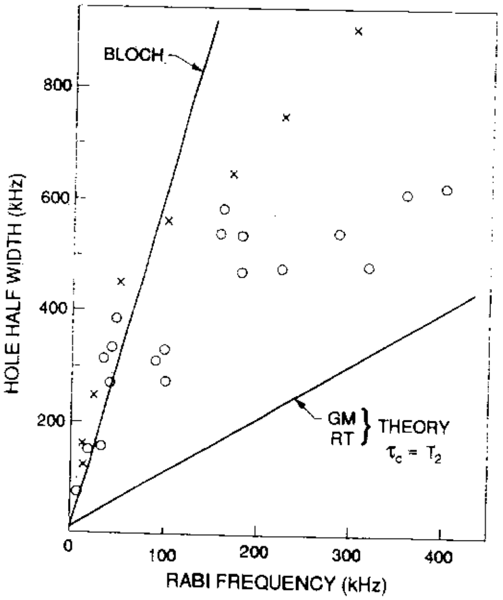

Fig. 2. Optical linewidth of the prepared hole vs Rabi frequency for ruby in the frequency domain [15]. Theoretical curves for the normal OBE, GM and RT models assume $\tau_c = T_2 = 15$ µsec. 0=Stark shift studies, X=AO frequency shifting.

this paper which show that the earlier [15] hole-widths were broadened by spectral diffusion.

SPECTRAL DIFFUSION

The apparatus used is shown in Fig. 3. The $^4A_2(-1/2) \rightarrow \overline{E}(-1/2)$ transition [selected by a circular polarizer (CP)] in 0.0034 Wt% Cr_2O_3 dilute ruby was studied, as in earlier work [15], with a field of 3.6 kG applied along the c axis and a sample temperature of 2K. A Coherent model 699-21 dye laser was modified [13] to give a peak to peak linewidth < 2 kHz. A pump-probe sequence of laser pulses was used at 25 Hz, with a probe intensity ~ 0.1% of the pump and precise probe frequency shifting obtained by a computer-controlled frequency synthesizer driving an acousto-optic (AO) modulator. Typically the pump and probe pulse widths were equal and the boxcar gate width was one-half the pulse width and set on the last half of the probe pulse. The beams before and after the sample were detected and subtracted to reduce amplitude noise and prevent overloading of the amplifier by the pump pulse. Not shown in Fig. 3 are AO switches in front of the diodes to further suppress the pump pulses. Each scan consisted of 101 points taken at 3 second intervals.

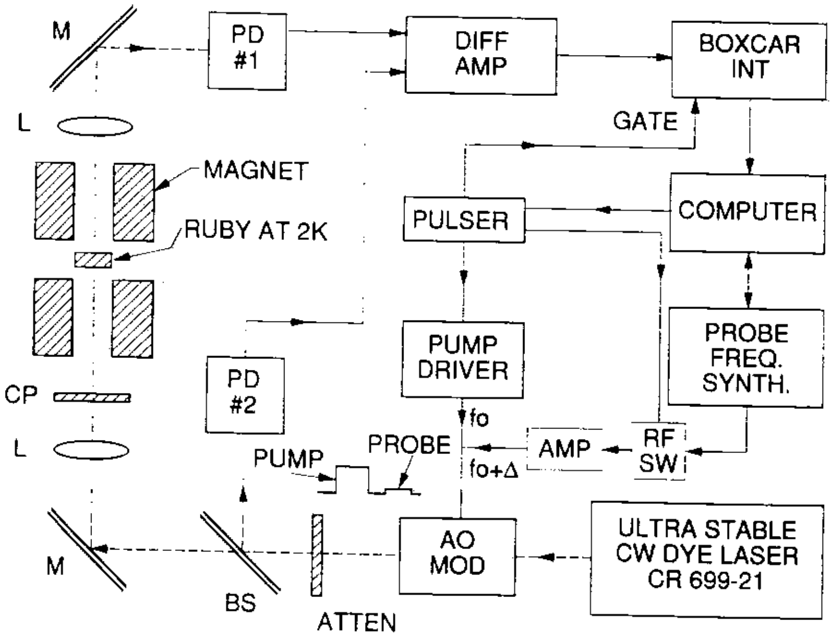

Fig. 3. Apparatus for pump-probe hole-burning studies in the frequency domain (see text).

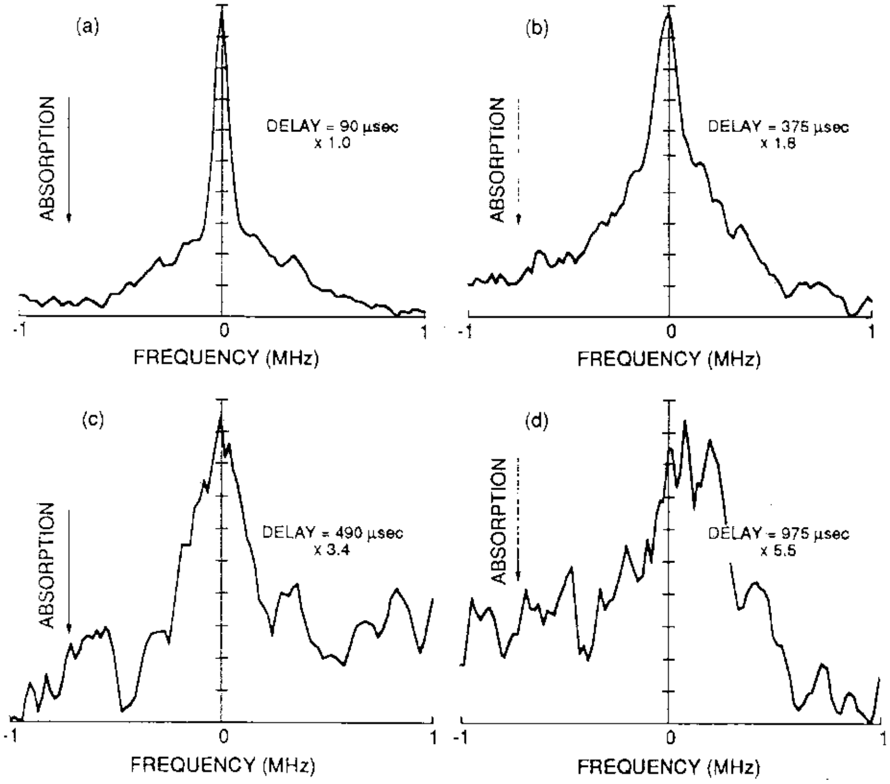

Fig. 4. Hole line shapes observed at various pump-probe delays as defined in the text. The Rabi frequency was 36 kHz and the vertical gain is indicated.

Lineshapes obtained for a series of pump-probe delays (defined as the time from the leading edge of the pump-pulse to the centre of the gate pulse) are shown in Fig. 4. These traces show clear evidence of spectral diffusion over a time scale of ~ 300 µsec, contrary to the earlier studies of Endo et al [17]. (It is possible that the stated [17] power broadening along with frequency jitter may have obscured the diffusion). These and other data were fit to a Lorenztian line shape by regression analysis. For short times (Fig. 4(a)), only the sharp central peak was used for analysis.

The relation between the observed half-width (HW) $\Delta\nu_0$ and $\Delta\nu_h$, the actual hole HW is

$$\Delta\nu_0 = \Delta\nu_h + \Delta\nu_2 + 2\Delta\nu_p \tag{1}$$

where $\Delta\nu_2$ is the dephasing HW (12 kHz) and $\Delta\nu_p$ is the spectral HW due to the finite pulse length. Fig. 5 summarizes the dependence of $\Delta\nu_h$ on pump-probe delay. Fig. 6 shows, at higher signal to noise ratio, that the lineshape is Lorenztian. A final point on the data is that the diffusion appears to occur similar to that observed [18] for phonon-assisted diffusion where a central sharp spike decays into a broad line (fig. 4(a), (b)). For short delays, it is clear from Fig. 5 that the frequency domain holewidth approaches that inferred from FID [13]. Further studies are planned for various Rabi frequencies to confirm this conclusion. Also studies at high field (to 5T) are planned to clarify the field dependent dephasing observed earlier [19].

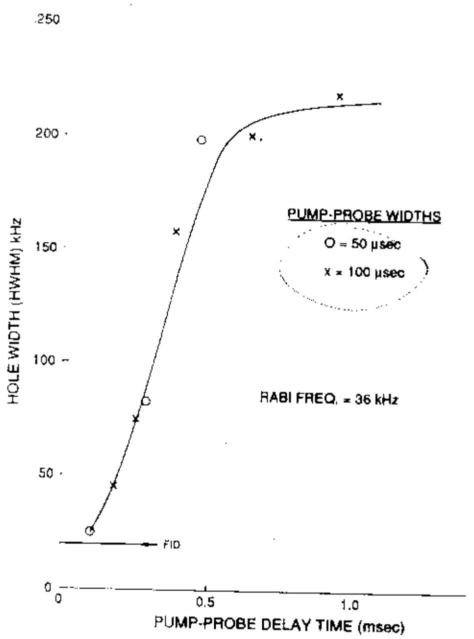

Fig. 5. Plot of deconvoluted hole-width $\Delta\nu_h$ vs pump-probe delay. The horizontal line indicates the value obtained by FID.

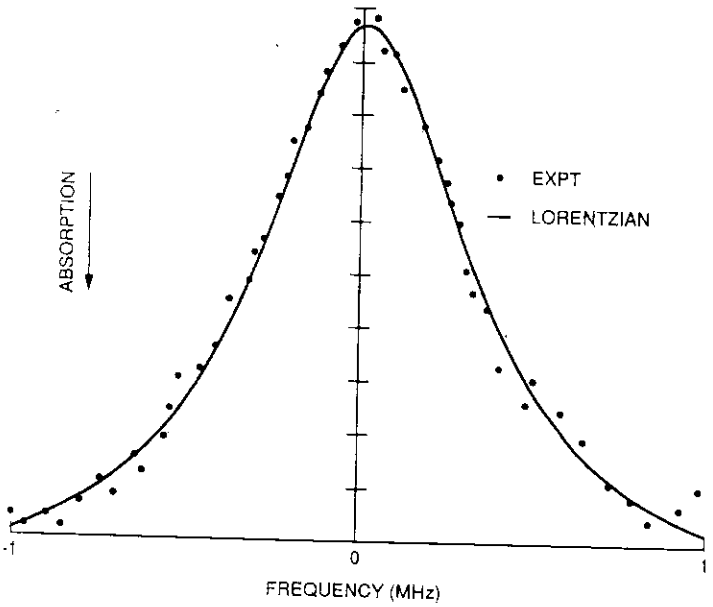

Fig. 6. Regression analysis Lorentzian fit to hole lineshape for a pump-probe delay = 1575 μsec. Rabi frequency = 53 kHz. For clarity not all experimental points are plotted.

Concerning the diffusion mechansism, I conclude that Al^{27} nuclear spin flip-flops outside the frozen core [20] are responsible. Cr^{3+}-Cr^{3+} spin flips cannot contribute since the flip time for 0.0034% ruby has been measured to be ~ 5 msec [21].

ACKNOWLEDGEMENT

The contributions to parts of this work by R. Kaarli and T. Muramoto is gratefully acknowledged as is technical assistance by J. Froemel, discussions with L.E. Erickson, P. Berman and J. Eberly and correspondence with S.Ya. Kilin, A.P. Nizovtsev and R.N. Shakhmuratov.

REFERENCES

1. R.G. DeVoe and R.G. Brewer, Phys. Rev. Lett. 50 (1983) 1269.
2. A. Szabo, U.S. Patent 3, 896,420 (1975). G. Castro, D. Haarer, R.M. Macfarlane and H.P. Trommsdorf, U.S. Patent 4,101,976 (1978).
3. R.G. DeVoe, A. Wokaun, S.C. Rand and R.G. Brewer, Phys. Rev. B23, (1981) 3125.
4. E. Hanamura, J. Phys. Soc. Jpn. 52 (1983) 2258.
5. J. Javanainen, Opt. Commun. 50, (1984) 26.
6. M. Yamanoi and J.H. Eberly, Phys. Rev. Lett. 52 (1984) 1353.
7. A. Schenzle, M. Mitsunaga, R.G. DeVoe and R.G. Brewer, Phys. Rev. A30 (1984) 325.
8. P.R. Berman and R.G. Brewer, Phys. Rev. A32 (1985) 2784.
9. K. Wódkiewicz and J.H. Eberly, Phys. Rev. A32 (1985) 992.
10. P.R. Berman, J. Opt. Soc. Am. B3 (1986) 564; 3 (1986) 572.
11. P.A. Apanasevich, S.Ya. Kilin, A.P. Nizovtsev and N.S. Onishchenko, J. Opt. Soc. Am. B3 (1986) 587.
12. A.R. Kessel, R.P. Shakhmuratov and L.D. Eksin, Sov. Phys. JETP 67 (1988) 2071.
13. A. Szabo and T. Muramoto, Phys. Rev. A39 (1989) 3992.
14. T. Muramoto and A. Szabo, Phys. Rev. A38 (1988) 5928.
15. A. Szabo, T. Muramoto and R. Kaarli, in Coherence and Quantum Optics 6 (Eds J.H. Eberly, L. Mandel and E. Wolf), Plenum Publishing (in press).
16. S. Ya. Kilin and A.P. Nizovtsev (submitted for publication).
17. T. Endo, T. Muramoto and T. Hashi, Phys. Lett. 99A (1983) 128.
18. R. Flach, D.S. Hamilton, P.M. Selzer and W.M. Yen, Phys. Rev. Lett. 35 (1975) 1034.
19. R.F. Liao and S.R. Hartmann, Opt. Comm. 8 (1973) 310.
20. A. Szabo, Opt. Lett. 8 (1983) 486.
21. A. Szabo, T. Muramoto and R. Kaarli (submitted for publication).