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Coherence Effects on Ultra-Narrow Hole Burning in Solids

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Optical hole-burning (HB) of inhomogeneously broadened absorption lines in solids has been of recent interest both for technological and scientific reasons. The use of HB for digital (frequency[1] and time domain[2]) and analogue[3] information storage and processing is presently under development. In part, this activity has prompted a closer look at the fundamental origins of the optical dephasing time T_2 of impurity atoms and molecules in solids. T_2 is important for information storage since it is the ratio of inhomogeneous to homogeneous linewidths that determines the storage capacity.

Magnetic fluctuations due to host and/or impurity spin flipping have been shown to be a limiting mechanism in determining optical dephasing in crystals at low temperatures. Recent photon echo studies[4] of the $^4A_2(-3/2) \rightarrow \bar{E}(-1/2)$ optical transition (hereafter $R_1(-3/2)$) in dilute ruby at high fields (~ 3T) have shown that Cr-Cr electron spin flipping is suppressed leaving only Al superhyperfine nuclear spin flip induced dephasing. In this paper we extend these studies to the $R_1(-1/2)$ transition at high fields using both frequency and time-domain [free induction decay (FID)] techniques. Longer T_2's are expected for the $R_1(-1/2)$ transition since the magnetic splitting rate for the $R_1(-1/2)$ line is 7.6 times smaller than for $R_1(-3/2)$. Other major differences that may affect dephasing are (1) spin mixing is much less for $R_1(-1/2)$ since ground and excited spin states are the same and (2) the ground state frozen core radius of $R_1(-1/2)$ is smaller because of the smaller magnetic dipole.

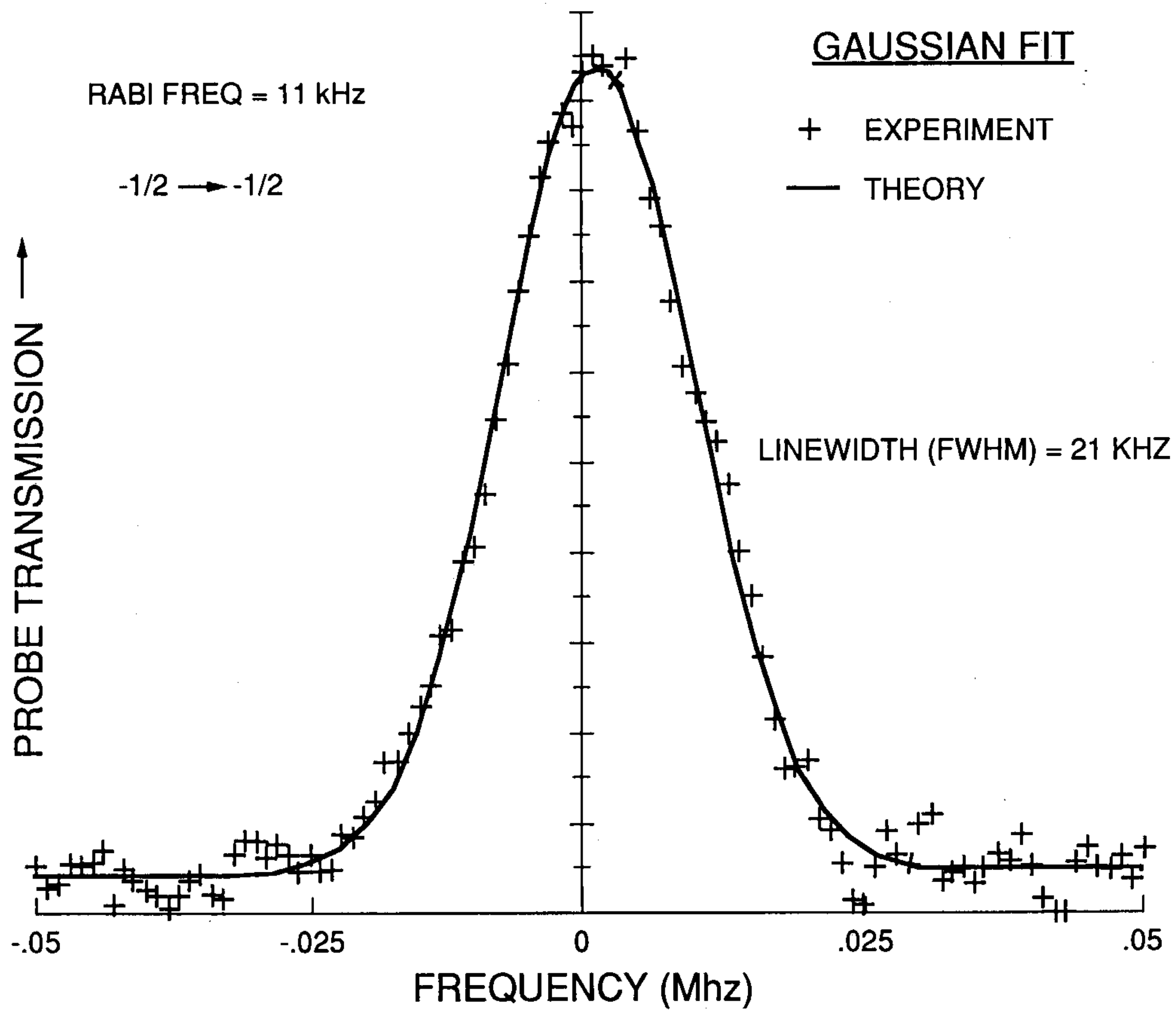
HB studies were done using a pump-probe technique[5]. Pump and probe pulsewidths were in the 50 - 100 μsec range with the probe intensity 1-5% of the pump. Precise tuning of the probe was achieved using an acousto-optic modulator driven by a computer scanned, frequency synthesized, radio frequency oscillator. A ring dye laser, modified to give a peak to peak linewidth of < 2 kHz for times of ~ 1 msec, was the excitation source.

Fig. 1 shows the hole obtained at a field of ~ 4T. This hole is, to the author's knowledge, the narrowest (by factor 5) hole ever observed in a solid. In contrast to the strictly Lorenztian hole shape seen[5] at low fields (~ 0.4T), the lineshape in Fig. 1 is Gaussian. Our original interpretation of this result was that this was an indication of superhyperfine dephasing and consistent with the near Gaussian decay observed[4] for R_1(-3/2) photon echoes. However further experimental studies and numerical analysis of Bloch and modified[5] Bloch equations indicate that the shapes and linewidths observed, for the range of Rabi frequencies and pulsewidths used, are still being determined by coherence rather than dissipative (dephasing and spectral diffusion) processes. This is confirmed by FID studies that imply a homogeneous linewidth < 7.8 kHz. Details of these calculations and measurements will be discussed as well as conditions necessary for observation of dissipation limited linewidths.

1. A. Szabo, U.S. Patent No. 3, 896, 420 (1975).
2. T.W. Mossberg, U.S. Patent No. 4, 459, 682 (1984).
3. A. Szabo, U.S. Patent No. 4, 432, 071 (1984).
4. J. Ganem, Y.P. Wang, D. Boye, R.S. Meltzer, W.M. Yen, R. Wannemacher and R.M. MacFarlane, Phys. Rev. Lett. 66, 695 (1991).
5. A. Szabo and R. Kaarli, Phys. Rev. B, Nov. 1991 (in press).

Figure Caption

Fig. 1 Hole in the ${}^4A_2(-1/2) \rightarrow \overline{E}(-1/2)$ absorption in dilute ruby (0.0023 at %) with a 3.8T magnetic field applied along the c axis. Pump and probe pulsewidths are 50 μsec. Sample temperature is 2 K. Regression analysis for a Gaussian fit gives a 3x smaller residual than for a Lorentzian.


GAUSSIAN FIT
RABI FREQ = 11 kHz
-1/2 → -1/2
+ EXPERIMENT
— THEORY
LINEWIDTH (FWHM) = 21 KHZ
PROBE TRANSMISSION
-.05
-.025
0
.025
.05
FREQUENCY (Mhz)