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Magnetic-field dependence of photon-echo decays in ruby

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Measurements of photon-echo decays in dilute ruby (Cr_2O_3 concentration of 0.018 at. %) in magnetic fields up to 29.5 kG are reported. Echoes from the $^4A_2(-\frac{3}{2})\leftrightarrow{}^2E,\bar{E}(-\frac{1}{2})$ and $^4A_2(-\frac{1}{2})\leftrightarrow{}^2E,\bar{E}(-\frac{1}{2})$ transitions have dephasing times (T_2) that are directly proportional to the ratio of the magnetic field (B) to the temperature (T), for the range $2.5 < B/T < 20$ kG/K. A transition in the decay from exponential to nonexponential behavior for $B/T > 10$ kG/K is observed. Decay times for the $^4A_2(-\frac{1}{2})\leftrightarrow{}^2E,E(-\frac{1}{2})$ echo are roughly a factor of 3 longer than for the $^4A_2(-\frac{3}{2})\leftrightarrow{}^2E,\bar{E}(-\frac{1}{2})$ echo. We attribute the lengthening of T_2 with increasing B/T to the suppression of Cr-Cr spin flip-flops as the population of the upper $^4A_2$ levels decreases.

INTRODUCTION

The photon echo has proven to be a powerful tool in understanding dynamic processes in materials. Ruby was the first material in which photon echoes were produced.[1] The early experiments on ruby established the necessity of a magnetic-field component along the c axis in order to observe photon echoes,[2] but there remains a confusion on the magnetic-field dependence of the decay of the photon echo. In early work[3] using a pulsed ruby laser that produced 15-nsec pulses with about 200-kW peak power, no magnetic-field dependence from 1.5 to 6 kG was reported, except for an anomaly in the decay time at the ground-state level crossing at 2.06 kG. Subsequently, Liao and Hartmann[4] reported the echo decays in ruby to be dependent on magnetic fields up to 5 kG using 1-W, 20-, and 80-nsec-wide pulses obtained by gating a single-frequency cw ruby laser. However, the field dependence of their decay rates began to level off at the higher fields.

In this paper we investigate the decay of photon echoes in ruby for $B > 6$ kG. Our purpose is to clarify an understanding of the photon-echo decay field dependence by extending measurements to magnetic fields much higher than those previously studied. We used a single-frequency-gated cw laser to study the photon-echo decay as a function of an on-axis magnetic field up to nearly 30 kG. Photon-echo decays were measured for the $^4A_2(-\frac{3}{2})\leftrightarrow{}^2E,\bar{E}(-\frac{1}{2})$ and the $^4A_2(-\frac{1}{2})\leftrightarrow{}^2E,\bar{E}(-\frac{1}{2})$ transitions referred to as the $-\frac{3}{2}$ echo and $-\frac{1}{2}$ echo, respectively. As we shall show, decay times increase dramatically as the field is raised. We believe that the increase in the decay times is related to depopulation of the $^4A_2(-\frac{1}{2})$ level. The long decay times provide increased sensitivity to effects not seen before. Whereas, previously[3,5] photon-echo decays in ruby were thought to be independent of temperature below 4 K, we have observed the first temperature-dependent decays at these temperatures. The temperature dependence arises in the same manner as the field dependence because it also controls the population of the $^4A_2(-\frac{1}{2})$ level.

EXPERIMENT AND RESULTS

A 0.018 at. % ruby crystal was placed in an electromagnet with its c axis oriented along the magnetic field. An argon-ion laser pumped a tunable, single-frequency (1 MHz), cw dye laser where the output power at the ruby wavelength was about 10–15 mW. The σ polarized light was focused on the sample with a 4-in lens. Two acousto-optic modulators gated the cw beam. The first was gated on twice providing two pulses about 0.5-μsec wide with a variable delay. The second was gated off at the end of the two-pulse sequence to provide additional rejection of laser leakage during detection of the echo. On the output side of the crystal was a Pockels cell, gated on during the echo, that was used to protect the photomultiplier tube during the preparation pulses. Echo intensities were measured as a function of the delay time between the two pulses.

Photon-echo decays were measured at 2.0 K as a function of magnetic field for both the $-\frac{3}{2}$ and $-\frac{1}{2}$ echoes. At the highest magnetic field (29.5 kG), the decays were also measured at 1.5 K. We only studied transitions from the $^4A_2(-\frac{3}{2})$ level (lowest in energy) and the $^4A_2(-\frac{1}{2})$ (first excited Zeeman sublevel) since, at the high magnetic

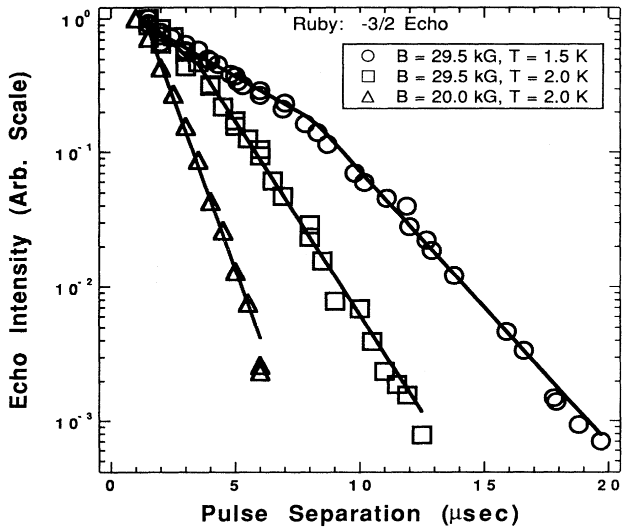

FIG. 1. Photon-echo decays for the $-\frac{3}{2}$ echo. Echo intensity (I) in arbitrary units is shown vs pulse separation (τ) in μsec.

fields and low temperatures at which we worked, these were the only levels significantly populated. Figure 1 shows the measured echo intensities as a function of pulse separation for the $-\frac{3}{2}$ echo. At the highest field (29.5 kG) and lowest temperature (1.5 K), the decay is nonexponential. As the field is lowered and the temperature is raised the decays become shorter and exhibit more single exponential behavior. Figure 2 shows corresponding data for the $-\frac{1}{2}$ echo. Decay times at the same fields and temperatures are about a factor of 3 longer than those in Fig. 1. The decay curves for the $-\frac{1}{2}$ echo, like those for the $-\frac{3}{2}$ echo, are nonexponential at high fields and low temperatures and become shorter and single exponential as the field is lowered.

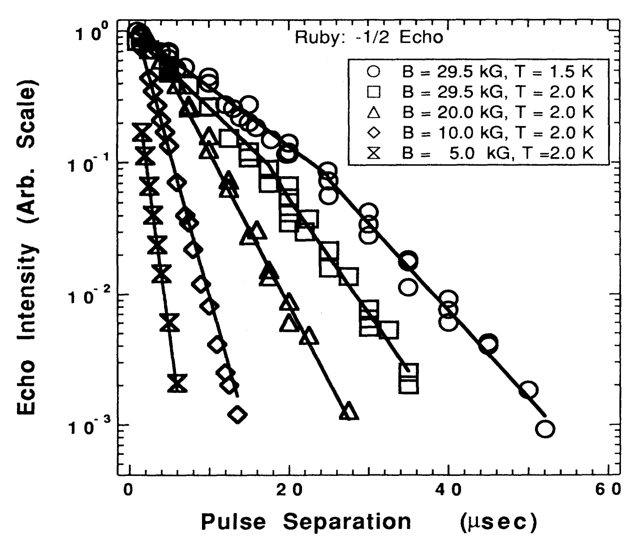

FIG. 2. Photon-echo decays for the $-\frac{1}{2}$ echo. Echo intensity (I) in arbitrary units is shown vs pulse separation (τ) in μsec.

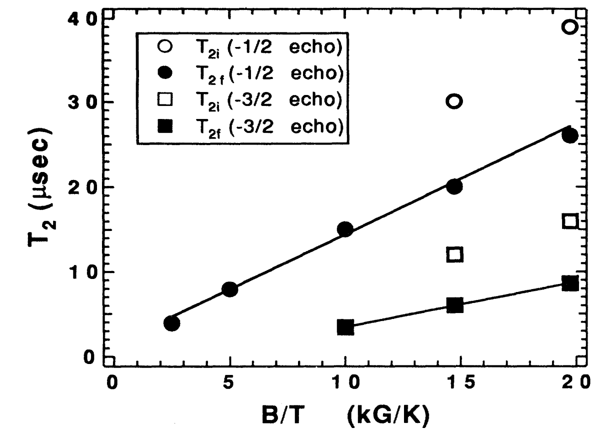

FIG. 3. Photon-echo decay times T_{2f} (the final decay time) and T_{2i} (the initial decay time) for both the $-\frac{3}{2}$ and $-\frac{1}{2}$ echoes in 0.018 at. % ruby vs B/T.

For a single exponential decay, T_2 can be determined by fitting the data to a function of the form,

$$I = I_0 e^{-(4\tau/T_2)}, \tag{1}$$

where I is the measured intensity and τ the separation between the pulses. For nonexponential decays we have fit the intensities to a sum of two terms, each of the form of Eq. (1), with decay times designated T_{2i} (the initial decay time) and T_{2f} (the final decay time). Table I lists T_{2i} and T_{2f} for the nonexponential decays at 29.5 kG. In parentheses next to T_{2i} is a quantity L which is the approximate length of time T_{2i} dominates the decay. Since L shortens as the temperature is raised it seems appropriate to list the decay times for the single exponential decays under the T_{2f} heading. The effect on the T_2's of lowering the temperature is similar to the effect of raising the field. Figure 3 is a plot of the T_2's in Table I as a function of the ratio of the magnetic field to the temperature (B/T). There is a remarkable linear variation of T_{2f}

TABLE I. Optical dephasing times for the $-\frac{3}{2}$ and $-\frac{1}{2}$ echoes at each field and temperature studied. Nonexponential decays are described by times T_{2i} (initial decay time) and T_{2f} (final decay time). L is the time that the initial decay persists. Purely exponential decays are described under the T_{2f} heading.

		$-\frac{3}{2}$ echo		$-\frac{1}{2}$ echo	
B (kG)	T (K)	T_{2f} (μsec)	T_{2i} (L) (μsec)	T_{2f} (μsec)	T_{2i} (L) (μsec)
29.5	1.5	8.6	16 (7)	26	39 (25)
29.5	2.0	6.0	12 (4)	20	30 (20)
20.0	2.0	3.4		15	
10.0	2.0			7.9	
5.0	2.0			3.9	

with B/T. The best-fit lines to the T_{2f} results are shown in Fig. 3. The slopes are 1.3 μsec$\times$K/kG for the $-\frac{1}{2}$ echo and 0.05 μsec$\times$K/kG for the $-\frac{3}{2}$ echo. The linear dependence of T_{2f} on B/T is especially clear for the $-\frac{1}{2}$ echo where all five points fall on a line.

The values of T_2 in 0.018 at. % ruby at 29.5 kG are much larger than those previously reported for 0.0034 at. % ruby at 3.8 kG and $T=2.2$ K, where $T_2(-\frac{3}{2})=6.9$ μsec and $T_2(-\frac{1}{2})=10.3$ μsec.[6] The present results in more concentrated ruby indicate that, by raising the B/T ratio for the 0.0034 at. % sample, a further increase in T_2 would have occurred, suggesting that Cr-Cr interactions are still important in the 0.0034 at. % sample at 3.8 kG, as recently noted by Szabo *et al.*[7]

DISCUSSION

We consider as possible dephasing mechanisms (1) Cr^{3+} interactions with the lattice phonons (T_1 processes), (2) interactions within the Cr^{3+} spin system (Cr-Cr spin diffusion), and (3) Cr^{3+} interactions with the neighboring Al nuclear spins (Cr-Al superhyperfine interactions).

Phonon-induced processes can occur in both the excited and ground states. Two types of processes can contribute in the excited state: single phonon-assisted transitions within the Zeeman levels of the doublet and Orbach processes. However, in the present case, single phonon-assisted transitions with the doublet are forbidden since they require a component of the external magnetic field perpendicular to the c axis.[8] Orbach processes are not effective below 2 K since the small number of resonant 29-cm^{-1} phonons present results in relaxation times that are of order sec.[9] For the ground state at fields of 20–30 kG, spin-lattice-relaxation times, which scale inversely with the cube of the difference between the energy levels, are expected to be about 1 sec.[10,11] Thus, for $B<30$ kG, ground-state single phonon relaxation does not significantly contribute to the direct part of the dephasing. Indirect contributions to the dephasing resulting from single phonon-induced spin flips of neighboring unexcited Cr^{3+} ions may occur but estimates of their magnitude suggest that they are not of major importance.

The dependence of the T_2's on B/T suggests that in 0.018 at. % ruby, Cr^{3+} spin dynamics controls the dephasing. For the $-\frac{3}{2}$ transition, both direct and indirect contributions to optical dephasing arising from spin diffusion should depend on B/T since this parameter controls the relative population of the ground-state spin sublevels. Mutual spin flips between Cr^{3+} ions are governed by the selection rules: $\Delta M_s=0$ for a pair, and $\Delta m_s=\pm 1$ for each spin. Therefore, a spin flip on a Cr^{3+} ion with $-\frac{3}{2}$ spin requires a neighboring ion with $m_s=-\frac{1}{2}$. Since the $-\frac{1}{2}$ spin population falls as B/T increases, the overall spin-flip rate is reduced resulting in an increase in T_2. An increase in T_2 has also been reported by Szabo and Heber[12] for 0.05 at. % ruby when the $-\frac{1}{2}$ spin population was reduced by optical pumping. However, for the $-\frac{1}{2}$ transition, the direct contribution to optical dephasing should increase with B/T since it becomes easier for a $-\frac{1}{2}$ spin to find a $-\frac{3}{2}$ spin neighbor. Both our data and the data of Szabo and Heber[12] also show an increase of T_2 for the $-\frac{1}{2}$ echo as the population of the ${}^4A_2(-\frac{1}{2})$ sublevel is transferred to the $-\frac{3}{2}$ spin state. This would suggest that for the $-\frac{1}{2}$ spin sublevel, indirect contributions play the major role in dephasing which is in agreement with the recent results of Szabo *et al.*[7]

The conclusion that indirect spin-flip processes between the Cr^{3+} ions entirely controls the dephasing for both the $-\frac{1}{2}$ and $-\frac{3}{2}$ echoes is not completely satisfactory. The indirect contribution to the dephasing arising from Cr^{3+} mutual spin flips for the two transitions should produce a dephasing rate which scales as the optical magnetic splitting factor.[12] Therefore, the ratio $T_2(-\frac{1}{2})/T_2(-\frac{3}{2})$ should be 7.6. However, in this work $T_{2f}(-\frac{1}{2})/T_{2f}(-\frac{3}{2})$ is approximately 3 from $B/T=10$ to 20 kG/K. All previous reports of this ratio have been no greater than 3.[6,12] We conclude that, although Cr^{3+} spin diffusion must play an important role in optical dephasing because of the dependence of T_2 on the Cr^{3+} spin-state populations, it does not seem possible to explain the dephasing rates entirely in terms of an indirect contribution from Cr^{3+} mutual spin flips.

Finally, the effects of the Al nuclear spin dynamics on the Cr^{3+} ions must be considered. In ruby, interactions between Cr^{3+} ions and neighboring Al spins are very strong but dephasing of the Cr^{3+} ions is inhibited by a "frozen-core" effect.[6,13] The Cr^{3+} ions with their strong magnetic moments detune the resonances of the neighboring Al spins from one another reducing their rate of mutual spin flips. The Cr^{3+} ions effectively surround themselves with a large core of Al spins that have a spin-flip rate that is generally reduced in comparison to the rate for the bulk Al spins. In the presence of only Cr-Al interactions and excluding frozen-core effects, the ratio $T_2(-\frac{1}{2})/T_2(-\frac{3}{2})$ would still be 7.6.[6] The expected effect of the frozen core is to reduce $T_2(-\frac{1}{2})/T_2(-\frac{3}{2})$ to $7.6/\sqrt{3}=4.4$,[6] a ratio closer to, although still larger than, the observed ratios.

The fact that $T_2(-\frac{1}{2})/T_2(-\frac{3}{2})$ is less than 7.6 for 0.0034, 0.018, and 0.05 at. % ruby suggests that the frozen core affects the dephasing in each of these cases. At the same time, the dependence of the dephasing rates on the Cr^{3+} spin-state populations suggests that Cr-Cr spin diffusion also influences the dephasing. The two dephasing mechanisms, Cr^{3+} spin dynamics and Cr-Al superphyperfine interactions may not be independent of each other since Cr-Cr indirect spin flips probably influence the dynamics of the core.

However, there should be a high-field low-concentration limit where Cr-Cr interactions are completely suppressed and dephasing results only from superhyperfine interactions between the Cr^{3+} electron and Al nuclear spins. The frozen core is not entirely without dynamics of its own. On a sufficiently long-time scale, superhyperfine interactions between the Cr^{3+} electron spins and Al nuclear spins will result in Al spin-flips inside the core. In this limit decays are expected to be highly nonexponential since a correlation exists between the Al spin-flip rate and the Cr-Al interaction strength.

The flipping of nearby Al spins, which occurs occasionally, would result in large frequency shifts for the Cr^{3+} ions. The broadening in time of the Cr^{3+} spectral distribution would result in accelerating nonexponential decay rates. The onset of nonexponential decays at the higher ratios of B/T may indicate the observation of Cr-Al superhyperfine interactions which become more evident as the Cr-Cr interactions are suppressed.

To summarize, for 0.018 at. % ruby, T_2 continues to increase with B/T up to fields of 30 kG at 1.5 K. This indicates the continued dominate role of Cr-Cr mutual spin flips in optical dephasing. However, nonexponential echo decay, which appears at the highest value of B/T, suggests that Al spin dynamics, with its associated frozen core, is becoming competitive with Cr spin dynamics as the latter becomes less effective because of depopulation of the upper ground-state spin sublevels. Experiments in more dilute ruby or at larger values of B/T should eventually lead to the limit of superhyperfine dominated optical dephasing.

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