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Optically induced homogeneous line narrowing

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The R_1 dephasing time in ruby (0.05 wt.% Cr_2O_3) is observed to increase by an order of magnitude when selected transitions of the R_2 line are pumped. Spin flip-flop dephasing in the ground state is suppressed by optically induced depopulation of the $^4A_2(\pm\frac{1}{2})$ levels.

Recently there has been growing interest in optical dephasing (homogeneous linewidth) mechanisms in solids, both for scientific as well as technological[1] reasons. The role of nuclear and electronic spin flip flops (SFF) in producing dephasing has long been recognized[2] in nuclear and electron magnetic resonance and such studies are now being extended to optical transitions.[3-6] In particular, direct proof of host *nuclear* SFF optical dephasing of *unlike* dopant nuclear spins has been achieved by observation of magic-angle homogeneous line narrowing.[6] This Rapid Communication reports on a new technique for homogeneous line narrowing which specifically demonstrates the existence of *electronic* SFF optical dephasing by *like* spins in ruby. The technique relies on selective depopulation of Cr^{3+} ground-state levels by $^4A_2 \rightarrow 2\bar{A}(R_2)$ optical pumping as well as on novel spin thermodynamics produced by the strong dipolar coupling between the Cr^{3+} electronic spins.

Optical dephasing in ruby is measured using photon echoes produced by Stark switching.[7,8] Two stabilized ($\approx$1-MHz linewidth) single-frequency cw dye lasers are used, one to produce the R_1 (693.4 nm) echoes and one to optically pump selected R_2 (692.0 nm) transitions. The R_2 beam is counter propagated to R_1 and is focused in the crystal (0.76 mm thick) to a spot 2× the R_1 diameter of 25 μm. Precise spatial overlap of the two beams is obtained using a beamsplitter with a high-resolution motorized tilt control. To minimize optical pumping,[9] the R_1 beam is gated on at 100 Hz for 70 μsec during the Stark pulse sequence using an acousto-optic modulator. The beams propagate parallel to the C_3 axis, along which a 3-kG field is applied, allowing the use of a circular polarizer for selection of overlapping transitions. Provision was made to measure the R fluorescence emitted perpendicular to the C_3 axis. All studies were done at 2 K with a Czochralski-grown crystal of concentration 0.05 wt.% Cr_2O_3 as specified by the manufacturer.[10]

Table I lists the effect of R_2 pumping on R_1 dephasing times. The largest effect occurs for simultaneous $++$, $--$ pumping (Fig. 1) for which $T_2(-\frac{3}{2})$ lengthens remarkably from 0.45 to 5.4 μsec (Fig. 2), almost equal to the value of 6.9 μsec for a very dilute (0.0034%) sample.[8] This occurs because of almost complete suppression of electronic SFF arising from depopulation of the $^4A_2(\pm\frac{1}{2})$ levels. We now examine various factors involved in this conclusion.

The average spin-flipping rate of an ion in spin state m_i may be written[11]

$$\alpha_i = 2\pi/\hbar \sum_{s,j} |H_{ij}|^2 I_{ij}(E)\rho_j(m_s) \quad , \qquad (1)$$

where $\rho_j(m_s) = n(m_s)/N$ is the probability that an ion is in

TABLE I. R_2 pumping effect on R_1 dephasing times in 0.05% ruby.

Dephasing time (μsec) \ R_2 pump[a] transition	None	$++$, $--$[b]	$++$	$+-$	$--$	$-+$
$T_2(-\frac{3}{2})$[c]	0.45	5.4	3.8	2.4	1.0	0.9
$T_2(-\frac{1}{2})$	1.23	7.4	3.9	3.4	2.1	2.3

[a]R_2 power = 5 mW.
[b]Linear polarization, all other cases used circular polarization.
[c]Quantity in brackets represents ground spin quantum No.

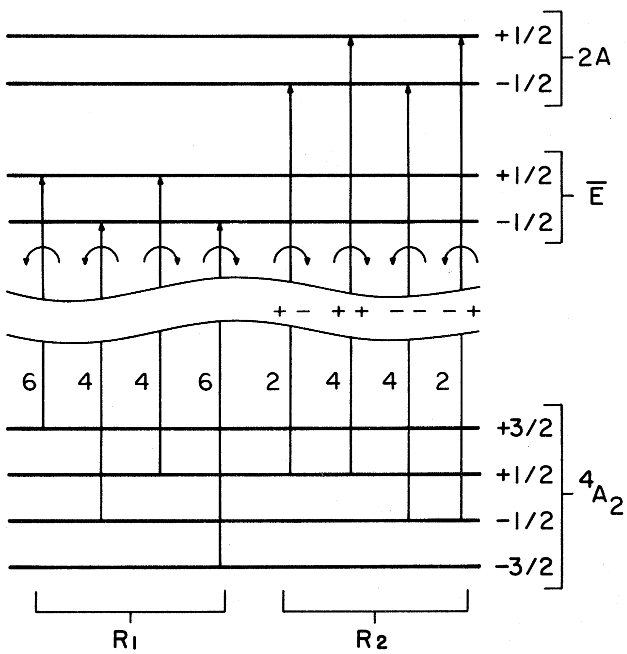

FIG. 1. Cr^{3+} R line energy levels, relative transition probabilities ($E \perp C_3$) and R_2 pump designations in ruby for a large magnetic field along the C_3 axes.

lattice site j with spin m_s; $n(m_s)$ is the density of ions with spin m_s, N the density of sites, H_{ij} the interaction Hamiltonian, and $I_{ij}(E)$ a line overlap function. For $B \parallel C_3$, only spin flips $m_s \leftrightarrow m_s+1$ $(m_s=-\frac{3}{2},-\frac{1}{2},+\frac{1}{2})$ can occur as determined by dipole SFF operators of the form S^+S^-. The observation that maximum lengthening of T_2 is obtained when both $^4A_2(\pm\frac{1}{2})$ levels are pumped is consistent with the above SFF selection rule.

Next we examine how R_2 pumping depopulates the $^4A_2(\pm\frac{1}{2})$ levels. This occurs in two ways. The first is by simple optical pumping whereby the fast R_1 relaxation (≈ 4 msec) results in a transfer of population to the $^4A_2(\pm\frac{3}{2})$ levels ($\approx$1-sec spin-lattice relaxation time). However, since the R_2 homogeneous linewidth[12] is only 300 MHz compared to the inhomogeneous width of several GHz, this alone would not be expected to transfer a significant amount of population. The principal mechanism which transfers the majority of $\pm\frac{1}{2}$ spins to the $\pm\frac{3}{2}$ levels is *ground-state* SFF. As shown in earlier work,[13] this tends to equalize the spin temperature of pumped and unpumped spins. Thus pumping only a small fraction of the ions results in pumping of the entire inhomogeneous line. This is demonstrated in Fig. 3 which shows the frequency dependence of R_2 pumping on the $^4A_2(-\frac{1}{2}) \leftrightarrow \bar{E}(-\frac{1}{2})$ echo. A large decrease in the echo (i.e., ground-state population) occurs by pumping anywhere within the inhomogeneous line. Also, similar to earlier absorption studies,[13] sharp (~ 300 MHz wide) holes appear when the R_2 laser pumps the $\sim$10-MHz bandwidth of R_1 ions involved in the echo. One of these is seen at $++$. Interestingly, under strong R_2 pumping, the echo holes for $--$ and $-+$ pumping become inverted, indicating a population inversion for the $^4A_2(-\frac{1}{2}) \rightarrow \bar{E}(-\frac{1}{2})$ transition.

As discussed by Compaan,[14] SFF dephasing by like spins occurs both by direct and indirect flips. His statistical averaging calculation, however, predicts a nonexponential echo decay which neither we[7,8] nor others[15] observe. [For this reason we have chosen the averaging procedure of Ref. 11 in writing Eq. (1)]. Moreover, the predicted decay rate is considerably faster than observed for our time periods. Because of these discrepancies between theory and experiment, the nature of dephasing in concentrated ruby has remained uncertain. However, the data just presented clearly establish ground-state SFF to be the dominant dephasing mechanism for 0.05% ruby (as opposed to say optical energy

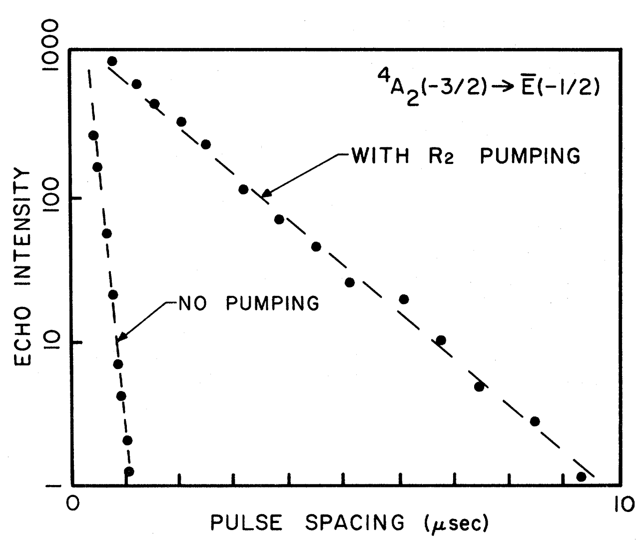

FIG. 2. Photon-echo decay of the $^4A_2(-\frac{3}{2}) \rightarrow \bar{E}(-\frac{1}{2})$ transition in 0.05% ruby showing the effect of simultaneous $(++,--)$ R_2 pumping.

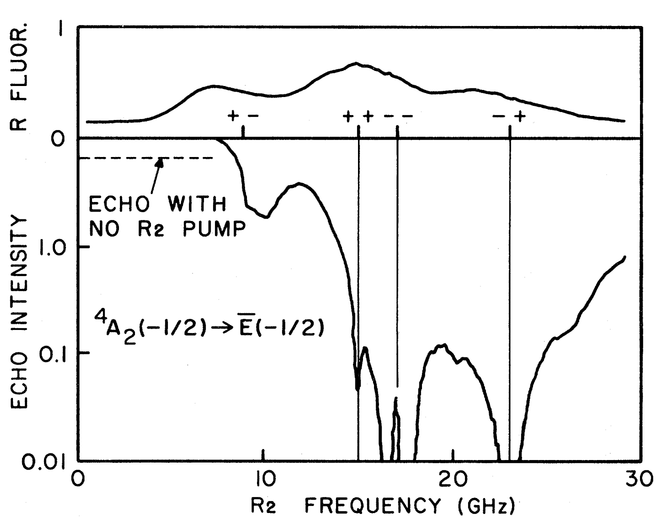

FIG. 3. Dependence of $^4A_2(-\frac{1}{2}) \rightarrow \bar{E}(-\frac{1}{2})$ echo and R fluorescence on the frequency of R_2 pumping. For the short pulse-separation time of 0.4 μsec, the echo is proportional to (population)^2 in $^4A_2(-\frac{1}{2})$ except for "holes" which appear for common R_1 and R_2 ions. A normal hole is seen at $++$ and an inverted one at $--$. The R_2 light is linearly polarized.

transfer). We now estimate the direct and indirect contributions to dephasing.

Since all decays are exponential and are primarily determined by Cr-Cr interactions, we write

$$1/T_2(-\tfrac{3}{2}) = 1/T_I(-\tfrac{3}{2}) + 1/T_D(-\tfrac{3}{2}) ,$$

where I and D represent indirect and direct with a similar equation for $T_2(-\tfrac{1}{2})$. For like spins, T_I scales as the magnetic splitting factor[14] giving $T_I(-\tfrac{1}{2}) = 7.6T_I(-\tfrac{3}{2})$. From Eq. (1),

$$T_D(-\tfrac{3}{2})/T_D(-\tfrac{1}{2}) = [9n(-\tfrac{3}{2}) + 16n(+\tfrac{1}{2})]/9n(-\tfrac{1}{2}) ,$$

where it is assumed that $I(E)$ is independent of spin. For $B = 3$ kG and $T = 1.8$ K, this gives $T_D(-\tfrac{3}{2}) = 2.0T_D(-\tfrac{1}{2})$. The first column of Table I and these equations give $T_I(-\tfrac{1}{2}) = 3.8$, $T_D(-\tfrac{1}{2}) = 1.8$, $T_I(-\tfrac{3}{2}) = 0.5$, and $T_D(-\tfrac{3}{2}) = 3.6$ μsec. Thus for 0.05% ruby we conclude that the $(-\tfrac{1}{2})$ echo decays mainly by direct SFF and $(-\tfrac{3}{2})$ by indirect SFF. The observation (Table I) that $-+$ or $--$ pumping has a small effect on $T_2(-\tfrac{3}{2})$ is consistent with the latter conclusion.

Finally we show in Fig. 4 an echo "excitation" spectrum which together with Table I emphasizes a surprisingly large asymmetry in $++(+-)$ vs $--(-+)$ pumping. The fact that $++(+-)$ lengthens $T_2(-\tfrac{3}{2})$ more ($\approx 3\times$) than $--(-+)$ indicates that more $+\tfrac{3}{2} \leftrightarrow +\tfrac{1}{2}$ flips are seen by a $-\tfrac{3}{2}$ spin than $-\tfrac{3}{2} \leftrightarrow -\tfrac{1}{2}$ flips. This suggests the presence of spin spatial correlation.

In conclusion, this study of like electron spin flipping in ruby directly demonstrates its role in optical dephasing of the R_1 line. While our results resemble that recently reported[16] for a nuclear spin system, i.e., T_2 increases with optical pump power, at first glance the two effects would appear to be unrelated since the optical transitions for the nuclear case are unresolved. However, dynamic nuclear polarization by optical excitation[17] seems feasible, at least for ions slightly out of resonance with the laser (to provide the required asymmetric pumping). Such a mechanism together with strain-broadened domains could produce a host nuclear population change in the vicinity of the impurity ion, accounting for the result of Ref. 16 in a manner similar to the present work.

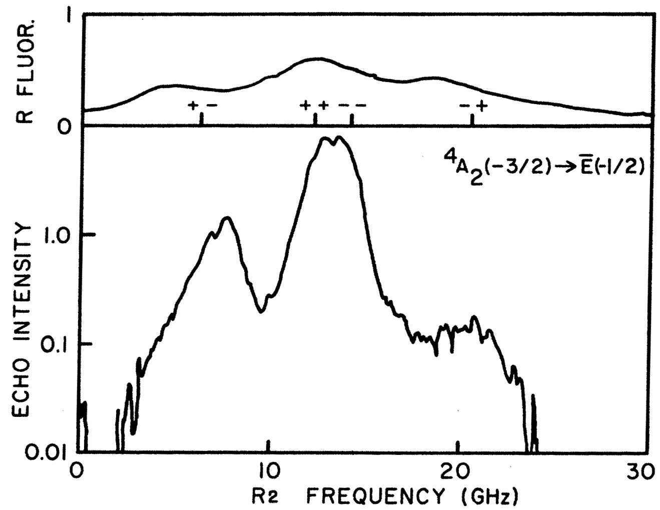

FIG. 4. $^4A_2(-\tfrac{3}{2}) \to \bar{E}(-\tfrac{1}{2})$ echo "excitation," and R_2 fluorescence spectra for R_2 pumping and pulse separation of 7 μsec. No circular polarizer is used so all four R_2 transitions are active.

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