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Zeeman-switched optical-free-induction decay and dephasing in $YLiF_4:Er^{3+}$

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A new technique for performing optical-free-induction-decay experiments is described whereby the ion transition frequencies are rapidly shifted with a pulsed magnetic field. The technique is demonstrated in $YLiF_4:Er^{3+}$ and is used to measure the temperature dependence of the homogeneous linewidths, Γ_h, of the four transitions connecting the Zeeman sublevels of the ground and lowest $^4F_{9/2}$ excited state. Below 4 K, Γ_h is dominated by Er^{3+}-Er^{3+} interactions, and evidence for the presence of both direct and indirect contributions is presented.

I. INTRODUCTION

Coherent transient spectroscopies have proven to be powerful techniques for examining dynamical processes in solids.[1] In one of these, OFID (optical-free-induction decay), a coherent superposition of the ground and excited states is prepared with a single-frequency cw laser. After establishing the coherence on a small frequency-selected subset of ions, a difference in frequency between the laser and the coherently prepared system is rapidly established, such that an optical heterodyne signal is produced due to the interference of the radiation from the laser and the coherently prepared system.[2] The frequency shift has been obtained by changing the laser frequency with either an electro-optic modulator (EOM) placed in the cw laser cavity[3] or with an acousto-optic modulator (AOM) placed external to the cavity,[4] or in a different approach, by shifting the ionic optical transition frequency using a pulsed electric field via the Stark effect.[5]

In this paper we describe a nonlinear OFID experiment in which the ionic transition frequency is switched, for the first time, with a pulsed magnetic field. This technique has potential application in any system which shows a Zeeman effect. This can be a Kramers system, a non-Kramers ion, where one or both states is a doublet, or a singlet in a magnetic field which establishes a magnetic moment in the ground or excited state. The frequency shifts can easily be made quite large, in excess of 1 GHz in some systems.

The technique is demonstrated using transitions among the Zeeman sublevels of the lowest-crystal-field components of the $^4I_{15/2}$ (ground) and $^4F_{9/2}$ (excited) states of Er^{3+} in $YLiF_4$. The observed heterodyne frequency shifts (obtained in the time domain) are equal to those measured with high-resolution spectroscopy (frequency domain). We then use the Zeeman-switched OFID results to measure Γ_h, the homogeneous linewidth, as function of temperature between 1.4 and 3.5 K for all four transitions coupling the Zeeman components of the ground and excited states, and identify the dominant low-temperature dephasing mechanisms operating in these levels.

II. EXPERIMENTAL

This Zeeman-switched OFID experiment is performed in a sample of $YLiF_4:Er^{3+}$, 0.1%, using transitions among each of the two Zeeman components of the ground (g_- and g_+) and excited (e_- and e_+) states, shown in Fig. 1. Also shown in Fig. 1 are the energy levels of the two higher-lying nearest crystal-field levels in the ground- and excited-state manifolds.[6] OFID is detected on all four of the possible Zeeman transitions shown in Fig. 1. The experiments are performed on a sample placed in a pair of Helmholtz coils (total dc impedence 1.0 Ω), wound with 15 turns each, whose diameter is $d = 3.75$ mm and whose separation is $d/2$, as shown in Fig. 2. The c axis of the sample is aligned parallel to the axis of the Helmholtz coil and a large dc magnetic field. The cw laser is focused onto the 150-μm thick sample in a direction perpendicular to the magnetic field, such that it can be polarized π or σ.

A current pulse is applied to the coil with a pulse generator (10 ns rise time, 50 Ω output impedence). The temporal behavior of the current pulse is obtained by monitoring the voltage across a 1.1-Ω resistive wire in series with the coil. The current pulse has a rise time of 50 ns and is limited to a value of 29 mA by the output driver of the pulse generator, for a nominal "1 V" setting on the pulse generator. Current amplitudes up to 350 mA were applied.

An AOM is used to gate the laser onto the sample in

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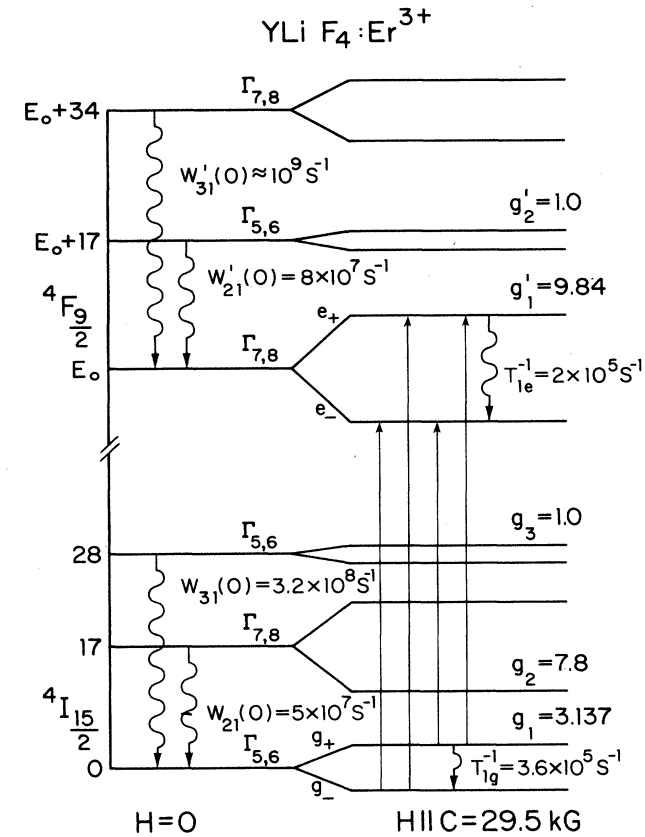

FIG. 1. Portion of the energy-level diagram of $YLiF_4:Er^{3+}$. The symmetries (S_4 site) of the crystal-field levels and the g values of the levels are indicated. The four transitions utilized in the OFID experiments are shown by vertical arrows. The rates $w_{kj}(0)$ next to the wavy arrows indicate the low-temperature spontaneous population relaxation rates.

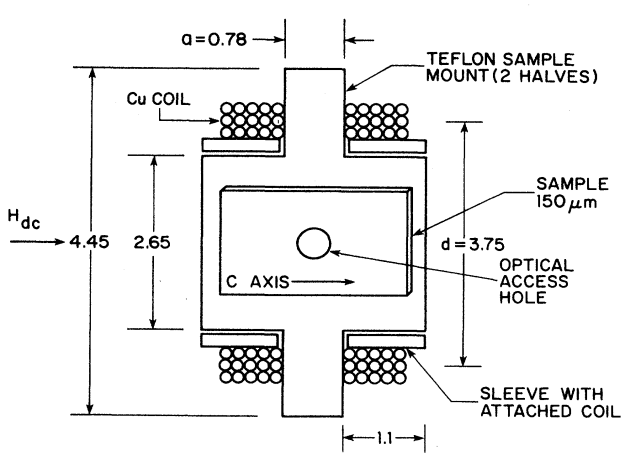

FIG. 2. Cross section of the magnetic field coil and sample holder assembly (dimensions in mm). The Teflon sample mount is constructed in two halves with a groove for the sample. The coil, whose axis is parallel to the c axis, is attached to a sleeve which slips over the sample holder.

order to prevent holeburning. After a 5-μs optical preparation time, the pulsed magnetic field is applied for about 5 μs. The laser is focused with a 21-in. (8 in. for the transitions with short T_2) focal length lens and the light is detected with a 100-MHz avalanche photodiode. The temperature is measured with a calibrated germanium thermistor located near the sample, which is immersed in the liquid helium.

III. ZEEMAN-SWITCHED OFID RESULTS

We first measured directly the field-induced frequency shifts near 29.5 kG from an excitation spectrum taken with a scanning frequency tunable dye laser. High-resolution measurements could be made because the inhomogeneous absorption linewidths of this sample were extremely sharp, consisting, for each Zeeman transition, of a doublet (origin of splitting unknown) separated by 1.3 GHz with a half width for each component of only 0.6 GHz. Excitation spectra were obtained at several fields between 27 and 29.5 kG. The spectroscopically determined frequency shift slopes with respect to field, $(d\nu_i/dH)_{\rm spec}$, are shown in the first column of Table I.

To test the Zeeman-switching technique we utilized the $g_-\ e_-$ transition (see Fig. 1). We first obtained an OFID signal in a 29.5-kG magnetic field by switching the laser frequency in a conventional manner with an EOM inside the laser cavity using a frequency shift of about 5 MHz. The resulting signal is shown in the top trace of Fig. 3. We then obtained the Zeeman-switched OFID signal, shown in the second trace of Fig. 3, for a nominal "1-V" setting on the pulse generator. We obtained a heterodyne frequency of 5.95 MHz. The amplitude and decay of the two OFID experiments are essentially identical. The effect of increasing the current in the coil is also shown in Fig. 3 (lower two traces). The heterodyne frequency is linear with the current in the coil as expected. OFID signals were also obtained on the other three Zeeman transitions labeled in Fig. 1, and they are compared with the results of the $g_-\ e_-$ transition in Fig. 4. A comparison of the measured values of the field-induced heterodyne frequencies, $\Delta\nu_{\rm fid}$, with the spectroscopic measurements, $(d\nu_i/dH)_{\rm spec}$, allows one to calibrate the current-induced pulsed magnetic field in the coil. We find a value of 64.7 G/A compared to a value calculated from the geometry of the coil of 72 G/A.

The calculated field-induced frequency shifts in the absence of mixing of the crystal-field levels by the magnetic field (linear Zeeman effect), is given by

$$d\nu_{\rm calc}(1)/dH = \pm\tfrac{1}{2}(g_e \pm g_g)\beta\ . \qquad (1)$$

TABLE I. Comparison of observed and calculated magnetic field-induced frequency shifts (MHz/G) for $H_{\rm dc}=29.5$ kG$\|c$.

Transition	$\left(\frac{d\nu}{dH}\right)_{\rm spec}$	$\frac{\lvert\Delta\nu_{\rm fid}\rvert}{\Delta H}$	$\frac{d\nu_{\rm calc}(1)}{dH}$	$\frac{\Delta\nu_{\rm calc}(2)}{dH}$
g_-e_-	−2.98±0.05	3.07±0.03	−4.69	−2.74
g_-e_+	11.28±0.05	11.13±0.15	9.09	11.04
g_+e_-	−7.33±0.05	7.11±0.25	−9.09	−7.14
g_+e_+	6.81±0.05	6.91±0.15	4.69	6.63

The g values have been determined by absorption and fluorescence as $g_g=3.24$ and $g_e=9.84$, for the ground and excited states, respectively, in reasonable agreement with electron spin resonance (ESR) results ($g_g=3.137$) (Ref. 7) and previous optical work ($g_e=9.84$).[8] The resulting calculated values of $d\nu_{\rm calc}(1)/dH$ are shown in the third column of Table I, where it is seen that they are inconsistent with the measured quantities.

The additional contribution to the frequency shifts can be explained as resulting from coupling between the crystal-field levels by the magnetic field which produces an additional negative frequency shift, $-A_gH^2$ and $-A_eH^2$, quadratic in the field, for the ground and excited states, respectively. Since the additional observed transition frequency shifts are positive, it follows that the larger shift must occur in the ground state. Assuming that the quadratic shift is identical for both Zeeman components of a crystal-field level, Eq. (2) becomes modified to

$$d\nu_{\rm calc}(2)/dH=\pm\tfrac{1}{2}(g_e\pm g_g)\beta+2(A_g-A_e)H\ , \qquad (2)$$

where a best fit to the observed frequency shifts yields

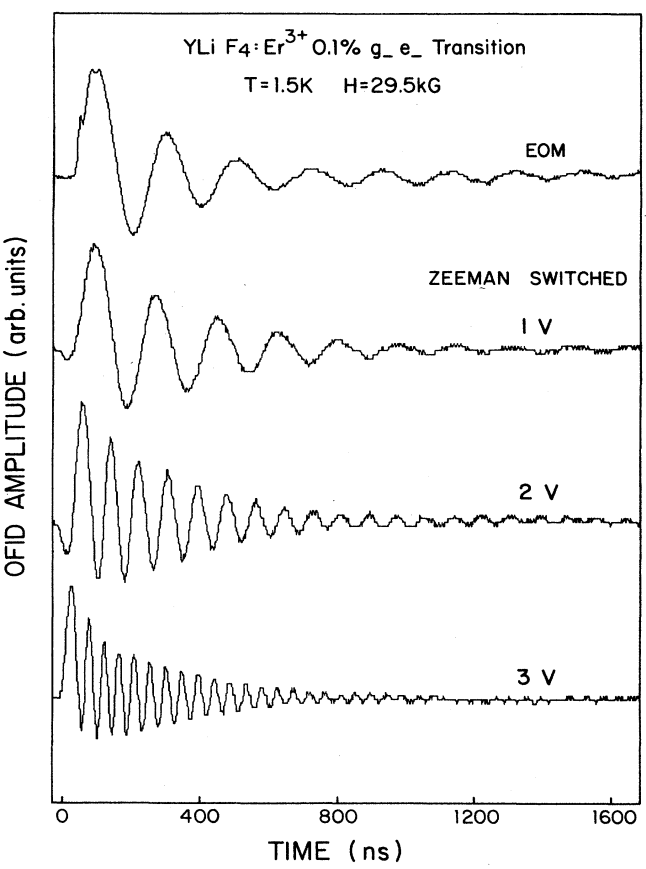

FIG. 3. OFID signals on the g_-e_- transition. Top trace: laser frequency switched with an electro-optic modulator in the cavity. Bottom three traces: ion transition frequency shifted with a pulsed magnetic field using different voltage settings on the pulse generator. Here "1 V"=29 mA to the coil, generating a pulsed field of 1.95 G.

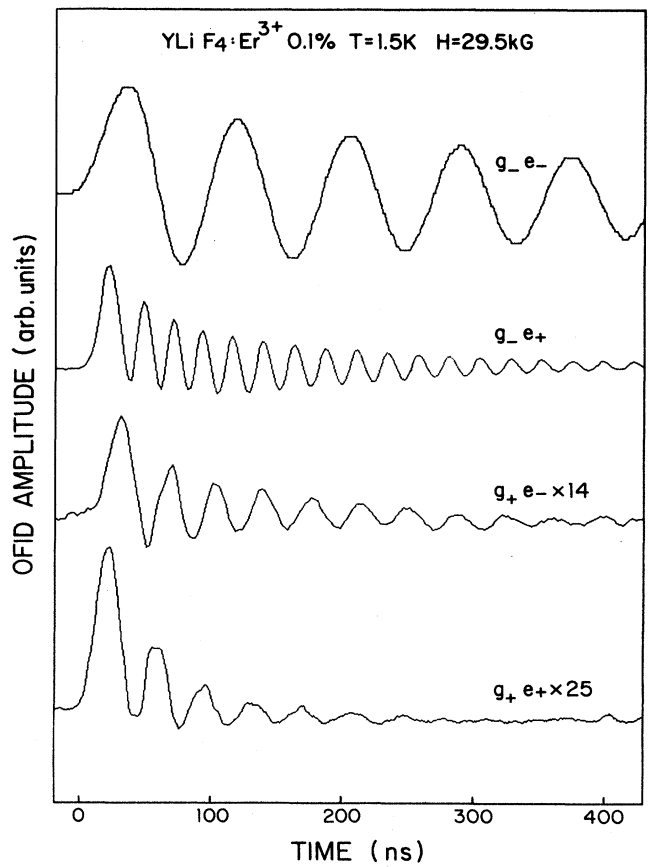

FIG. 4. OFID signals on each of the four transitions at 1.5 K, except for the g_+e_+ transition where the temperature was 2.2 K. The g_-e_- and g_-e_+ signals were obtained in a single shot, while the g_+e_- and g_+e_+ signals were averaged for 100 shots.

$A_g-A_e=33$ MHz/kG^2. These are shown in the fourth column of Table I where it can be seen that there is now good agreement with the observed values of the first and second columns. Since a magnetic field along the c axis does not lower the S_4 site symmetry, only Kramers doublets of the same symmetry can be mixed with the optically coupled states by the field, namely the levels at 28 and 34 cm^{-1} in the ground- and excited-state manifolds, respectively (see Fig. 1).[8]

IV. TEMPERATURE DEPENDENCE OF THE HOMOGENEOUS LINEWIDTH

We used the Zeeman-switched OFID to measure the temperature dependence of Γ_h, the homogeneous linewidth, plotted in Fig. 5 for the four transitions. For the longer values of T_2 (>500 ns), the OFID data were obtained in a single shot, while for shorter values of T_2, data were averaged over 100 shots.

The decay of the nonlinear OFID is determined by the width of the saturation hole burned in the absorption during the optical excitation pulse.[1] In the absence of power broadening the OFID signal decays at a rate

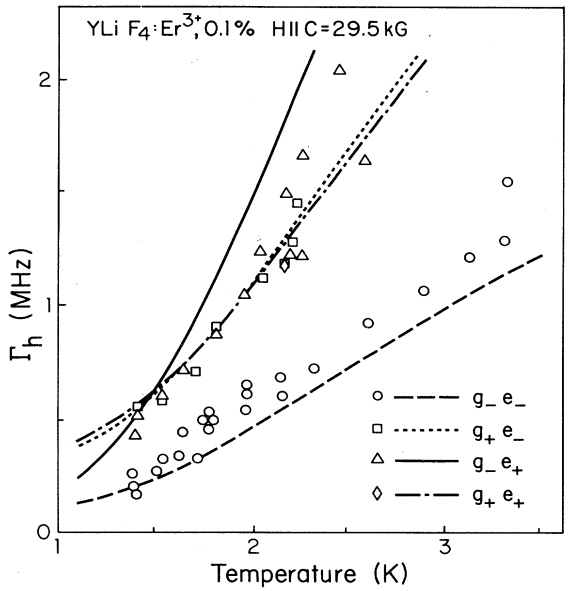

FIG. 5. Temperature dependence of the homogeneous linewidths. The discrete points are the experimental results and the curves are the calculated best fits as described in the text.

$2T_2^{-1}$, where T_2 is the optical dephasing time, related to Γ_h by $\Gamma_h=(\pi T_2)^{-1}$. Both population relaxation and pure dephasing processes contribute to Γ_h. The power dependence of the OFID was checked for power broadening, which was not present. The optical preparation time was varied to maximize the observed value of T_2. For times greater than 5 μs, T_2 decreased, probably due to spectral diffusion, which broadens the hole. Therefore all T_2 measurements were performed with a 5-μs preparation time.

A comparison with previous work in LaF_3:Er^{3+} (Ref. 9), a similar system, suggests that the most important contributions to Γ_h in $YLiF_4$ are likely to include the direct process involving phonon coupling among the two Kramers components in the ground or excited states, resonant phonon coupling to the Zeeman sublevels of the higher-lying crystal-field levels (see Fig. 1), superhyperfine interactions with the neighboring fluorines and Er^{3+}-Er^{3+} dipolar interactions. However, even in small magnetic fields, the fluorines near the magnetic impurity form a frozen core, which is decoupled from their neighbors due to their much stronger interaction with the Er^{3+} magnetic moment, and as a result, F^- spin diffusion is greatly reduced.[10]

Fortunately, it is possible to independently determine some of the contributions to Γ_h using other available information on the phonon-induced relaxation processes. A detailed study of the direct process relaxation in the ground state has been carried out using pulsed ESR.[11,12] Scaling the low-temperature spin-lattice relaxation rate at 36.2 GHz ($H=8.16$ kG) by H^5, an H dependence which is well known in ESR for spin-lattice relaxation within a Kramers pair, we obtain the $T=0$ spontaneous relaxation rate for $g_+ \rightarrow g_-$ at 29.5 kG, $T_{1g}^{-1}=3.6\times10^5$ s^{-1} (Ref. 13). Spontaneous relaxation rates from crystal-field levels 2 and 3, at low fields, are obtained from the temperature dependence of the spin-lattice relaxation rates,[11,12] and are $w_{21}(0)=5\times10^7$ s^{-1} and $w_{31}(0)=3.2\times10^8$ s^{-1}, respectively. These are indicated on Fig. 1. To describe the OFID measurements of this paper, obtained in a large magnetic field, we scale the low-field rates by $[\Delta_g(H)/\Delta_g(H=0)]^3$, the cube of the ratio of the energy-level separation at H to that at low (essentially zero) fields, in order to account for the frequency dependence of the relaxation rates, assuming field-independent ion-phonon matrix elements.

For information on the excited-state relaxation we performed time-resolved fluorescence studies in which we monitored the luminescence of the relevant excited levels after each was selectively excited with a pulsed tunable dye laser. At 29.5 kG, when we excited e_+ we observed a relaxation rate of $T_{1e}^{-1}=2\times10^5$ s^{-1}, which scaled with field as H^5 as expected for a Kramers pair. Excitation of level 2′ ($^4F_{9/2}-\Gamma_{5,6}$), at zero field yielded a relaxation rate $w'_{21}(0)=8\times10^7$ s^{-1}, which could then be scaled by $[\Delta_e(H)/\Delta_e(H=0)]^3$, as already discussed for the ground state. We estimated $w'_{31}(0)\cong10^9$ s^{-1}. Each of these resonant phonon-induced dephasing processes contributes a term to Γ_h of $(2\pi)^{-1}w_{jk}$. Here w_{jk} is the resonant phonon-induced rate coupling the lower and upper states, j and k, respectively, separated by energy Δ_{jk}, and is given by

$$w_{jk}=w_{kj}(0)p(\Delta_{jk});\quad w_{kj}=w_{kj}(0)[p(\Delta_{jk})+1]\ , \qquad (3)$$

where $p(\Delta_{jk})$ is the resonant phonon occupation number. For phonon coupling among the Zeeman sublevels of the ground (g_+ and g_-) or excited (e_+ and e_-) states, j and k are replaced by $-$ and $+$, respectively.

We then add to these phonon-induced contributions, terms arising from the Er^{3+}-Er^{3+} interactions. These take two forms, as described by Mims[10] for electron spins. The "direct" contribution involves mutual spin flips of the coherently prepared ion in state g_- (g_+) with a nearby Er^{3+} ion in the other possible spin state g_+ (g_-), producing an additional source of population relaxation. This rate is proportional to n_+ (n_-), the occupation number for state g_+ (g_-), given by

$$n_\pm=(1+e^{\pm\Delta_g/kT})^{-1}\ . \qquad (4)$$

Its contribution to Γ_h is $A_D n_+$ ($A_D n_-$), for transitions involving states g_- (g_+), where we treat A_D as an empirical constant obtained from a best fit to the data, as described in the following.

The "indirect" contribution results from spin flips of Er^{3+} ions near the coherently prepared ion, producing fluctuating magnetic fields which modulate the optical transition frequencies of the ions. These frequency fluctuations occur during the saturating optical pulse and contribute to its linewdith. Their contributions are proportional to $d\nu_i/dH$, the derivative of the ith optical transition frequency with respect to field (see Sec. III).[9] We make the assumption that the "indirect" contributions to Γ_h are simply proportional to the spin-flip rate of the neighboring Er^{3+} spins. This can be justified in the

early time limit for the sudden jump model described by Klauder and Anderson,[14] where the spectral width broadens at a rate linear in the neighbor spin-flip rate. In our experiments only a small fraction of the neighboring spins undergo a spin flip during the preparation and observation time of the OFID signal ($\cong 5$ μs), so this corresponds to the early time limit.[15,16]

The "indirect" effects can arise from spin-lattice relaxation of the neighboring ion (the T_1 process) or by spin-spin diffusion among two or more neighboring ions (the T_2 process). The temperature dependences of the two processes are slightly different. For the T_1 process we consider that the neighbors can be either in the lower- or upper-ground-state Zeeman sublevels with probability n_- or n_+, respectively, in either case undergoing spin flips at rates given by Eq. (3) with $w_{kj}(0)=T_{1g}^{-1}$. Then the contribution to Γ_h for the ith transition can be written

$$\Gamma_{h,i}(B_I)=B_I\frac{d\nu_i}{dH}R_I(T) , \tag{5}$$

where $R_I(T)$ is the temperature-dependent T_1 spin-flip rate, given by

$$R_I(T)=T_{1g}^{-1}(e^{\Delta_g/kT}-e^{-\Delta_g/kT})^{-1} . \tag{6}$$

B_I describes an effective contribution of all the neighbors, and its value is identical for all four optical transitions. We estimate B_I by equating the linewidth (FWHM) due to spectral diffusion, $\Delta\nu_{\text{spec}}$, with $\Gamma_{h,i}(B_I)$. We have[17]

$$\Delta\nu_{\text{spec}}=\frac{2}{\pi}R_I(T)\Delta\omega_{1/2}\Delta t , \tag{7}$$

where $\Delta\omega_{1/2}$ is the dipolar halfwidth[10] and Δt is the optical preparation time, 5 μs in this case. For an Er^{3+} concentration of 10^{19} cm^{-3}, $\Delta\omega_{1/2}\cong 22$ MHz, yielding $B_I\cong 2.3\times 10^{-5}$ G s.

The "indirect" contribution due to spin-spin diffusion on the neighbors is approximately proportional to n_+n_-, the product of the probabilities of finding a nearby pair of Er^{3+} ions with each ion in a different spin states, n_+n_-. We introduce a proportionality parameter for the indirect contribution, A_I, which is also the same for all four Zeeman transitions. The resulting contribution to Γ_h is

$$\Gamma_{h,i}(A_I)=A_I\frac{d\nu_i}{dH}(e^{\Delta_g/kT}+2+e^{-\Delta_g/kT})^{-1} . \tag{8}$$

We added the contributions of the above-mentioned Er^{3+}-Er^{3+} interactions to the resonant phonon-induced contributions to Γ_h for each of the four Zeeman transitions and calculated the temperature dependence of Γ_h. In the comparison of these calculations with the data (Fig. 5), we have added to the calculated Γ_h a contribution of 70 kHz to all four transitions to account for the effects of laser jitter and the finite (5-μs) preparation time on the observed value of Γ_h. The only adjustable parameters, A_D, A_I, and B_I, have been varied to give a best fit to all of the data. We find that the data are well described with the values $A_D=1$ MHz and $A_I+\frac{9}{7}B_IT_{1g}^{-1}=9$ G. The "direct" term contributes mainly to the difference between the low temperature values of Γ_h for the transitions involving g_- and those utilizing g_+. Most of the temperature dependence below 3.5 K arises from the "indirect" terms containing the parameters A_I and B_I. The temperature dependences of the two "indirect" terms [Eqs. (5) and (8)] are sufficiently similar that it is not possible at low temperatures to distinguish their individual contributions from a fit to the data. If we turn off the T_2 contribution to the "indirect" terms by setting $A_I=0$, we find from a fit to the experimental results, $B_I=2\times 10^{-5}$ G s, a result which is very close to our estimate above. Contributions to Γ_h from phonon-induced coupling to the higher-lying crystal-field levels in the ground and excited manifolds are unimportant below 3.5 K. As a result, the measurement of the temperature dependences of Γ_h on all four transitions allows us to identify the presence of both "direct" and "indirect" contributions of the Er^{3+}-Er^{3+} interactions to the homogeneous linewidth.

V. SUMMARY

We have demonstrated that pulsed magnetic fields can be used to perform Zeeman-switched OFID experiments. The technique is of broad generality; it is applicable to any system where the ground and/or excited state has a magnetic moment. In addition, large frequency shifts can easily be obtained. In our system, $YLiF_4$:Er^{3+}, we have observed optical frequency shifts which are nonlinear in field and these and the ground- and excited-state g values are easily measurable with the Zeeman-switched OFID technique.

Measurements of the homogeneous linewidth on all four transitions have enabled us to identify the presence of both "direct" and "indirect" Er^{3+}-Er^{3+} interactions as contributing to its temperature dependence. The measured temperature dependence at high magnetic fields is adequately described with these interactions alone; the phonon-induced coupling to other crystal-field levels is unimportant in this system below 3.5 K.

ACKNOWLEDGMENTS

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