Optical Decoherence in Er^{3+}-Doped Silicate Fiber: Evidence for Coupled Spin-Elastic Tunneling Systems

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(Received 26 October 2005; published 27 January 2006)

We measured the optical decoherence times T_2, or, equivalently, the homogeneous line width, in an Er-doped optical fiber at low temperature as a function of external magnetic field and temperature using two-pulse photon echoes. The decoherence times were up to 230 ns at fields above 3 T. The magnitude of the line narrowing induced by a magnetic field of 3 T is 2.5 MHz, which is anomalously large compared to that typical for oxide crystals with similar Er^{3+} concentration. This is interpreted as evidence for dephasing by coupled spin-elastic tunneling modes where the normal glass tunneling modes acquire a magnetic character by coupling to the Er^{3+} spin.

DOI: 10.1103/PhysRevLett.96.033602 PACS numbers: 42.50.Md, 71.55.Jv

The optical decoherence time T_2 is a quantity of central importance in characterizing the dynamic interaction of impurities, such as rare-earth-metal (RE) ions in solids, with their surrounding environment. Decoherence is also a critical parameter in applications of frequency selective optical processing, [1,2] spatial-spectral holography [3,4], and quantum information [5]. Disordered materials, exemplified by glasses, provide an environment that results in higher bandwidths through the inhomogeneous linewidths Γ_{inh}. Disorder, however, typically leads to shorter decoherence times (broader homogeneous linewidths) from coupling between the impurity ions and low frequency tunneling modes (two-level systems or TLS) that are characteristic of such materials [6,7] and that contribute a linear term in the heat capacity as a function of temperature in the range of milliKelvin to several Kelvin [8,9]. We report the first measurements of optical decoherence in a glass—here an Er^{3+}-doped fiber—in the presence of an external magnetic field and find an unexpected, large field-dependent contribution to the homogeneous linewidth. The field-dependent contribution is interpreted as evidence for the existence of new coupled spin-elastic TLS, which are the greatest source of optical decoherence. The contribution of these magnetic TLS can be frozen out at moderate fields. A fit to the field dependence of the freezing-out process yields a value for the "effective" or average magnetic moment of the spin-elastic modes of 1.9 Bohr magnetons.

Several groups have measured the dynamic interactions of impurity ions in glasses by spectral hole burning in RE-doped bulk glasses [10–12] and fibers [13,14] and by accumulated photon echoes in fibers [15–17]. However, the inherently long time scale of those measurements (milliseconds to seconds) often results in large contributions to the measured linewidths Γ_h^{eff} from spectral diffusion [18,19], which prevent the measurement of what might be called the intrinsic homogeneous width $\Gamma_h = 1/(\pi T_2)$. Although two-pulse photon echoes provide the best method to measure T_2 on the appropriate time scale, such measurements are difficult in RE-doped glasses because of the short T_2 and low oscillator strength of optical transitions within the $4f$ shell of the RE ions. These problems can largely be overcome in the fiber geometry, given the high light intensity inside the fiber core and the long interaction length that is phase matched for the photon echo. The long sample length also allows substantial absorption at low impurity doping levels, where RE ion-ion interactions are reduced. The only other case where decoherence times have been measured for optical centers in a glass appears to be the pioneering work of Hegarty *et al.* [20], who measured echo decays in a 10 ppm Nd^{3+}-doped silica fiber using a pair of pulsed dye lasers. Their experiments were done between 50 mK and 1 K in a ^{3}He–^{4}He dilution refrigerator. Our expectation was that Er ions couple less strongly to the elastic TLS modes because of their less extended $4f$ orbitals and that photon echoes might, thus, be observable in the more conventional and accessible temperature range of liquid ^{4}He and even up to $T > 4$ K. This turned out to be the case.

The experimental setup for observing photon echoes was as follows: Measurements were made on a sample of 3M Er^{3+}-doped fiber (FS-ER-7A28) of length 96 cm, whose core composition in mole % was 93% SiO_2, 6% Al_2O_3, 1% La_2O_3, and 150 ppm Er. The La_2O_3 is included to prevent aggregation of the Er^{3+} ions, since La^{3+} and Er^{3+} occupy similar sites. The doped fiber was spooled to a diameter of 23.4 mm and immersed in liquid helium in a cryostat containing a superconducting magnet. It was spliced near the spool to a length of undoped Corning SMF-28 fiber which was fed out the top of the cryostat and spliced to undoped connectorized pigtails. The mode field diameter of the Er-doped fiber at 1550 nm was 5.9 μm and that of the undoped fiber was 10.4 μm. The Er doping profile in the 3 μm core was approximately Gaussian. It was important to consider the fiber bending loss $\alpha_B L$, since the diameter of the spool was limited by

the 1 inch bore of the magnet. The loss coefficient α_B is a strong, exponential function of bend radius R and numerical aperture [21,22]. An initially selected erbium-doped fiber with a numerical aperture (NA) of 0.16 exhibited extremely high bending losses at 1534 nm which extinguished the echo signal, so it was replaced by a fiber with a larger NA of 0.25. For this second fiber, the loss coefficient at 1534 nm calculated from the expressions given in Refs. [21,22] was 3.0×10^{-6} cm^{-1}, giving a negligible total bend transmission loss of -0.0013 dB.

An external-cavity cw diode laser followed by an erbium-doped fiber amplifier [23,24] was gated by a 165 MHz acousto-optic (AO) modulator to produce 30 ns pulses that had a peak power of 0.5 mW in the Er fiber. The magnetic field was varied from 9 mT to 5 T, and the temperature was varied in the range of 1.6–4.1 K by adjusting the helium vapor pressure in the sample chamber. After passage through the fiber, the light was coupled out to a second AO modulator that gated the echo signal from the strong exciting pulses onto a 125 MHz (In,Ga)As photodiode. Echo decays were recorded by averaging 64 shots at each time delay as the exciting pulse separation t_{12} was scanned from 60 to 350 ns, depending on the temperature.

The low temperature absorption spectrum shown in Fig. 1 was measured by passing light from a tungsten-halogen lamp through the fiber, followed by a 1 M spectrometer at a resolution of 2 cm^{-1}. The low energy peak centered at 1528 nm corresponds, at least on its low energy side, to the transition from the ground state $^4I_{15/2}(1)$ to the lowest $^4I_{13/2}(1)$ level. We observed a substantial falloff in echo intensity and of T_2 at wavelengths shorter than 1532 nm. This suggests that the strong peak, which has a total width of 79 cm^{-1}, also includes transitions to the $^4I_{13/2}(2)$ level; we estimate the inhomogeneous linewidth of the $(1) \rightarrow (1)$ transition to be $\sim$40 cm^{-1}. The broader peak at shorter wavelengths includes the remaining transitions to higher lying crystal field levels of $^4I_{13/2}$. The absorption coefficient at the peak of 1528 nm was 0.022 cm^{-1} or 9.8 dB/m. Using the manufacturer-provided Er concentration of 150 ppm ($\sim 4 \times 10^{18}$/cm^3), the inhomogeneous width of 40 cm^{-1}, and considering the imperfect overlap between the mode field and the Er doping profile, such an absorption corresponds to an oscillator strength of $\sim 2 \times 10^{-7}$. The light intensity required for a π pulse of 30 ns width is then 40 kW/cm^2. Given the mode diameter in the fiber of 5.9 μm, this corresponds to a peak power of 8 mW. Thus, we estimate that our excitation pulses of 0.5 mW peak power had an area of $\sim \pi/4$. To ensure that the dominant excitation was of the $(1) \rightarrow (1)$ transition, most of the photon-echo measurements were made at 1534 nm on the low energy side of the lowest peak, where the echo signal was a maximum. Measurements were also made between 1537 and 1530 nm. The echo signals under the best conditions were $\sim 10\times$ stronger than the leakage of the exciting pulses through the second AO, even for the delay of 80 ns as shown in Fig. 2(a). Representative photon-echo decay curves are shown in Fig. 2(b) for the residual field of the superconducting magnet (9 mT) and for the highest field used of 5 T. The decays are approximately exponential with a decay constant of $T_2/4$ and are 2 orders of magnitude shorter than typically found in crystalline materials because of dephasing caused by coupling to the low frequency TLS.

One of the most important results that we obtained was the dependence of the homogeneous linewidth, $\Gamma_h = 1/(\pi T_2)$, on magnetic field. Figure 3 shows a reduction of 2.5 MHz in Γ_h from 3.9 MHz at $H_0 = 0$ to 1.4 MHz above $H_0 = 3$ T, after which it is independent of field. This is a novel and unexpected result. In oxide crystalline materials, a similar trend is seen [23,24], but the magnitude of the linewidth reduction for an Er concentration comparable to that in our fiber is approximately 100 kHz, an order

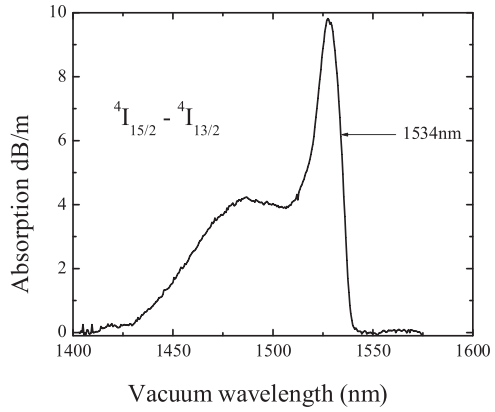

FIG. 1. Absorption spectrum of the $^4I_{15/2}(1)$ to $^4I_{13/2}$ transitions of Er^{3+} in the 3M fiber FS-ER-7A28 at 1.6 K. Most of the photon-echo measurements were made on the low energy side of the absorption at 1534 nm.

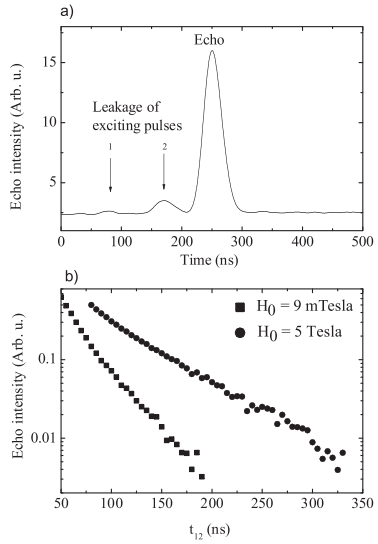

FIG. 2. (a) Photon-echo signal observed after inserting an AO modulator after the Er-doped fiber to reject the exciting pulses. (b) Photon-echo decay curves at the residual magnet field of 9 mT and at the highest field used of 5 T.

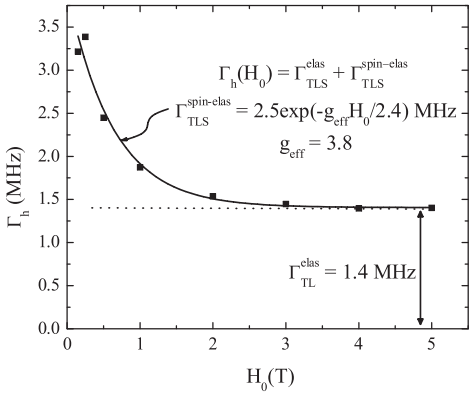

FIG. 3. Magnetic field dependence of the homogeneous linewidth $\Gamma_h = 1/(\pi T_2)$ at 1.6 K. The field-dependent part is attributed to coupling to spin-elastic TLS excitations, and it decreases with field as these freeze out. The fit to the expression shown gives an effective g value of 3.8 for the spin-elastic modes. The purely elastic TLS modes contribute a constant 1.4 MHz as determined from the high field limit where the spin-elastic modes are frozen out.

of magnitude smaller than we see in the glass. In crystals, the dephasing is caused by the magnetic dipole interaction between erbium spins [25], which undergo mutual electron spin flips. This, in turn, creates a fluctuating local field at the Er^{3+} ion whose phase coherence is being considered. These mutual flips are frozen out when $g\beta H_0 > \mathrm{kT}$, where g is the electron g value and β the Bohr magneton. In glasses, it is expected that this magnetic dipolar contribution is smaller than in crystals, because Er^{3+} ions have a very anisotropic g value and the variation in local geometry from site to site in glasses results in a Zeeman splitting that also varies greatly from site to site. This removes the resonance between neighboring Er spins and, hence, slows the mutual spin-flip rate. The spectral density of spin fluctuations moves to lower frequency, substantially reducing their effectiveness in dephasing. Instead of seeing the reduced magnetic field induced effect that this argument implies, we see more than an order of magnitude increase.

This leads us to propose the following picture. The magnetic field-dependent dephasing is not caused by magnetic dipole coupling between fluctuating spins but rather by field-dependent elastic tunneling modes that acquire a magnetic character by coupling to the spins. Such a coupling is not unexpected, since the g values of Er^{3+} are highly anisotropic and contain large orbital contributions. Thus, the g values and magnetic response are sensitive to the configuration of neighboring ions such as those giving rise to the potential wells of the elastic TLS. We propose that two kinds of tunneling modes are involved here: those that are associated with an impurity spin and strongly coupled to the spin and those that are separated from impurity spins, and purely elastic in nature. The involvement of RE ions in TLS modes has been noted earlier in heat capacity measurements of Pr ions in silicate glass [9], and there is other evidence from rf echo experiments of two classes of tunneling modes in Dy-doped glass [26]. In the fiber, we propose that both classes of TLS, to use the most general term, couple predominantly to the optical transition through an elastic dipole interaction. The 1.4 MHz contribution at high field originates from the pure elastic modes. The additional contribution that depends on magnetic field and reaches 2.5 MHz at zero field originates from the coupled spin-elastic tunneling modes. In other words, the TLS that acquire spin character can be frozen into one well in the presence of a magnetic field, while those that are pure elastic modes cannot. This is a new picture of the origins of optical dephasing for Kramers ions in glasses. A fit of the field-dependent linewidth was made to the expression below

$$\Gamma(H_0) = \Gamma(0)\exp(-g_{\mathrm{eff}}\beta H_0/kT), \tag{1}$$

where $\Gamma(0)$ is the linewidth in zero field, β is the Bohr magneton, H_0 is the applied external magnetic field, and g_{eff} is the effective or average g value for the randomly oriented local Er^{3+} sites in the glass. The fit, shown as the solid line in Fig. 3, yields a value of $g_{\mathrm{eff}} = 3.8$. In crystalline systems, the g tensor is generally quite anisotropic, and we have measured g values for bare Er^{3+} spins with magnitudes from less than 1 to greater than 10 for different orientations of the magnetic field with respect to local Er^{3+} site axes.

The results of Huber *et al.* [18] that exponential echo decay implies that the coupling between the rare-earth-metal ion and the TLS mode is dipolar in nature still apply, since the coupling is through elastic dipoles. The temperature dependence of this TLS induced dephasing has been found to be almost linear with temperature below 4 K in a number of glasses [10–12,14,16,17] and disordered crystals [27,28]. This behavior can be explained by a theory proposed by Huber *et al.* [18] in which the linewidth has the form

$$\Gamma_h(T) = AT^{1+\mu}, \tag{2}$$

where μ expresses the energy dependence of the TLS density of states through $\rho(\omega)_{\mathrm{TLS}} = C\omega^{\mu}$, where A and C are constants. We show the temperature dependence of the homogeneous linewidth in Fig. 4 at $H_0 = 2$ T, where essentially all of the spin-elastic contribution is frozen out. The temperature range over which we were able to measure echo decays was less than a factor of 4, so we cannot very precisely establish the functional form of the temperature dependence; however, as shown in Fig. 4, a linear fit provides a good description of the data, indicating that μ is close to 0 in Eq. (2), i.e., that the accessible TLS density of states is almost independent of frequency in this frequency range. The y intercept is essentially zero within experimental error.

We have measured decoherence times using two-pulse photon echoes in an Er-doped optical fiber at 1.5 microns at temperatures up to 4 K in an external magnetic field. The optical decoherence times of up to 230 ns are the longest

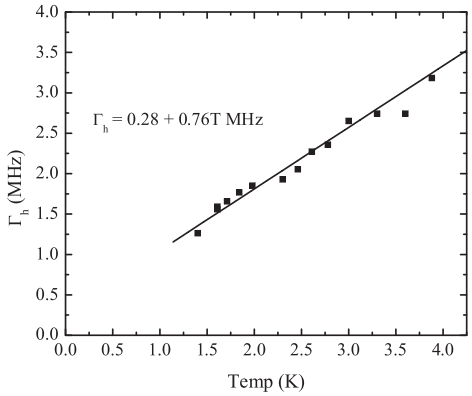

FIG. 4. Temperature dependence of the homogeneous linewidth at 2 T.

measured for an optical center in a glass at liquid ^{4}He temperatures. The good signal to noise ratio observed suggests a potential for application of doped communication fibers in frequency selective optical processing, buffer memories, and spatial-spectral holographic devices.

We propose a qualitatively new picture for optical dephasing in Kramers-ion-doped glasses, i.e., that two distinct classes of tunneling modes are responsible. One class consists of spin-elastic coupled modes in which the electron spin couples to the elastic TLS, and the resulting magnetic character allows this contribution to be frozen out at high magnetic field. From the form of the magnetic field dependence of the linewidth, we obtained an effective g value for the spin-elastic TLS modes of 3.8. The second class consists of TLS that are more distant from the rare-earth-metal ions and that, consequently, do not interact strongly with spins; they produce a magnetic field-independent contribution. It will be interesting to compare the dephasing rates obtained here with heat capacity or far IR absorption data that can determine the number density of TLS modes. In this way, a separation of coupling coefficient effects and TLS number density could be made which should lead to a more systematic understanding of the coherence properties of doped fibers. Together with measurements of acoustic or microwave TLS echoes, such experiments may provide independent evidence for the spin-elastic coupled modes.

We thank Casey Dodge, for his expert assistance in designing and building mechanical components and in fiber splicing, and Jim Onstott, for discussions on fiber composition and properties. This manuscript is based on research sponsored by the Air Force Research Laboratory under Agreement No. F49620-01-1-0313 and by the Montana Board of Research and Commercialization Technology. The views and conclusions contained herein are those of the authors and should not be interpreted as necessarily representing the official policies or endorsements, either expressed or implied, of the Air Force Research Laboratory or the U.S. Government. Sun's work was supported in part by the National Science Foundation/EPSCoR Grant No. 0091948 and by the State of South Dakota.

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