Photon Echoes below 1 K in a Nd^{3+}-Doped Glass Fiber

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Optical dephasing rates T_2^{-1} of a dilute concentration of Nd^{3+} ions in a pure silica fiber have been studied from 0.05 to 1.0 K with use of two-pulse photon echoes. Echoes obtained by resonant excitation of Nd^{3+} into the metastable $^4F_{3/2}$ state from the $^4I_{9/2}$ ground state have yielded $T_2^{-1} = 1.3 \times 10^{-6}$ s^{-1} at 0.1 K which follows a $T^{4/3}$ temperature dependence to 1.0 K. A crossover of T_2^{-1} to a T^2 temperature dependence above 1 K is inferred by comparison with earlier high-temperature linewidth measurements on this transition.

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Relaxation rates of active centers in amorphous soilds are not clearly understood. Disorder plays a poorly defined role in enhancing decay rates in a wide variety of systems studied with spectroscopies ranging from radio[1,2] and microwave[3] frequencies to the optical spectral region. At low temperatures optical dephasing rates of paramagnetic ions and molecules in inorganic and organic glasses are enhanced in comparison to rates in crystalline hosts and show a variety of temperature dependences.[4-10] By use of the techniques of fluorescence line narrowing (FLN),[4-6] spectral hole burning,[7,8,10] and coherent transients,[9] the dephasing rate has been found to increase with temperature as T^m, where m ranges from 1 to 2.2 depending on the system. Various theories[11-13] have been proposed based on an interaction of the optical center with phonons, atomic tunneling systems, and other host excitations, but with modest success. This is due partly to our limited understanding of the nature of the amorphous excitations and partly to the absence of a detectable universal trend in the data.

In this paper we report on optical dephasing below 1 K for a rare-earth–doped amorphous material using photon echoes in a unique geometry, a single-mode silica fiber. We have measured the temperature dependence of the dephasing rate T_2^{-1} of a subset of Nd^{3+} ions in pure silica by selective excitation in the inhomogeneously broadened $^4I_{9/2}(1)$-$^4F_{3/2}(1)$ transition. The dephasing rate varies as $T^{4/3}$ between 0.1 and 1.0 K. Since the homogeneous linewidth of this transition has been well studied by FLN[14] in a variety of Nd^{3+}-doped glasses and shows a nearly T^2 dependence between 30 and 300 K, we infer that a crossover must exist at intermediate temperatures.

The active fiber, prepared by modified chemical vapor deposition (MCVD), is 34 m long with a 6-μm core of pure silica doped with Nd^{3+} to a concentration less than 10^{-4} mole%. Echo studies are particularly advantageous in a fiber geometry since long interaction lengths are available, phase matching is intrinsically satisfied, and the small core size results in modest power requirements. The use of a long fiber to obtain a few absorption lengths results in extremely low Nd^{3+} concentrations minimizing ion-ion interactions and allows for efficient heat sinking.

It is to be noted that in this case Nd^{3+} is in a single-component pure silica matrix in contrast to previous studies in multicomponent systems. We expect that studies in the model glass system SiO_2 will allow closer contact between these experiments and other known physical properties. The $^4F_{3/2}$ level of Nd^{3+} in pure silica shows an unusually large Stark splitting (≈ 430 cm^{-1}) while the inhomogeneously broadened $^4I_{9/2}(1)$-$^4F_{3/2}(1)$ transition has a linewidth of ≈ 100 cm^{-1}. Because of the spectral isolation of this lowest $^4F_{3/2}(1)$ Stark component and its very long lifetime in this host, interband relaxation and multiphonon-emission–limited dephasing at low temperatures[9] are avoided.

The experimental setup is shown in Fig. 1. The active fiber was greased to a copper coil-foil cylinder in good thermal contact with the mixing chamber of a ^{3}He/^{4}He dilution refrigerator. Two undoped single-mode fibers, fusion spliced to the active fiber, guided the light pulses into and out of the refrigerator. Two dye lasers generated the excitation pulses (≈ 10 nsec full width at half maximum) at 0.89 μm which were combined and coupled into the fiber. The echo was easily detected with a photodiode, but for enhanced discrimination against the excitation pulses an electro-optic–modulator/photomultiplier combination with 20 dB attenuation was used. The signals were digitized and averaged over 250 shots typically. The timing and triggering of the two lasers and the modulator were controlled by a

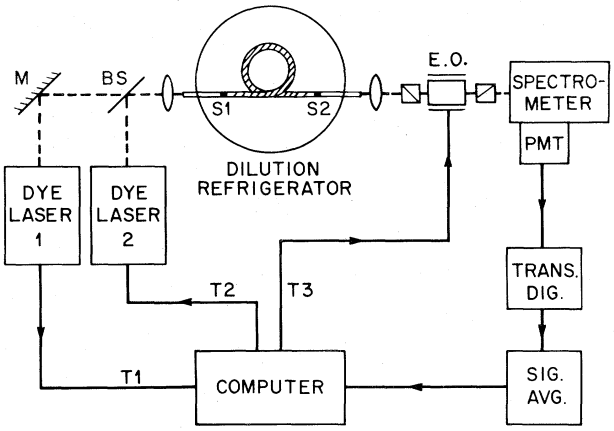

FIG. 1. Experimental setup for generation and detection of photon echoes in a Nd^{3+}-doped fiber. BS, beam splitter; E.O., electro-optic modulator; M, mirror; S1 and S2, fusion splices from the undoped fibers to the doped fiber (shaded area) in the dilution refrigerator; and T1–T3, triggers for the firing of the two lasers and the modulator, controlled by a microcomputer.

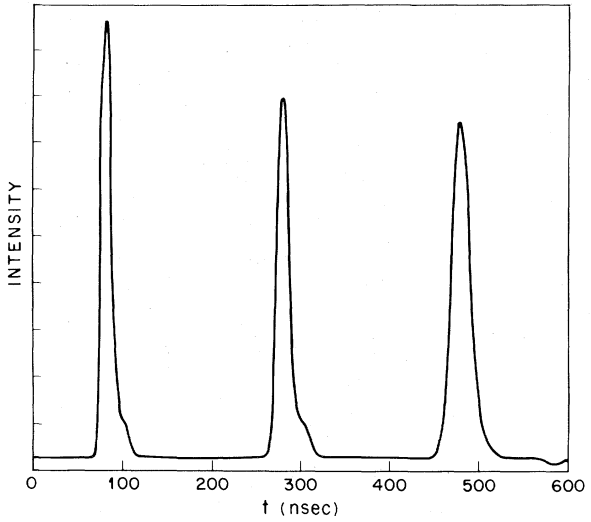

FIG. 2. Two excitation pulses and resulting photon echo in Nd^{3+}-doped SiO_2 single-mode fiber at 50 mK. The excitation pulses have been attenuated by a factor of approximately 100. The enhanced echo width is an artifact due to laser jitter.

microcomputer.

Photon echoes were generated by excitation on the low-energy side of the inhomogeneous line between 0.050 and 1 K. Initially the two excitation pulses were generated from a single dye laser with a fixed optical delay line. No discrimination against the excitation pulses was required, since the echo intensities were comparable to the transmitted excitation pulse intensities. With two dye lasers the echo was weaker and showed a significant shot-to-shot fluctuation. A typical example, in this case, of the transmitted excitation pulses and the observed echo at 50 mK is shown in Fig. 2. On changing the delay τ_{12} between the two excitation pulses, we observed the echo intensity to decay as $\exp(-4\tau_{12}/T_2)$, where T_2^{-1} is the dephasing rate. Figure 3 shows T_2^{-1} as a function of temperature. The solid line represents a $T^{4/3}$ power law which describes the data well[15] between 0.1 and 1.0 K. The reason for the weak temperature dependence below 100 mK is not clear at present. The low-temperature rate is still much larger than the fluorescence decay rate $T_1^{-1} = 2 \times 10^3\ s^{-1}$. We considered heating of the fiber due to nonradiative decay, background absorption, and scattering losses as a possible reason for this behavior. Reduction of the *average* power dissipated in the fiber by decreasing the laser repetition frequency by an order of magnitude has no effect on the dephasing rate at 50 mK. Reducing the *peak* power by about a factor of 3 below the typical excitation power level did not show any change in the dephasing rate at 70 mK either. However, an increase in excitation level by a factor of 5 brought about a $\approx 30\%$ decrease in T_2. Although the power-dependent effects are relatively small we have no independent measurements of the core temperature below 100 mK.

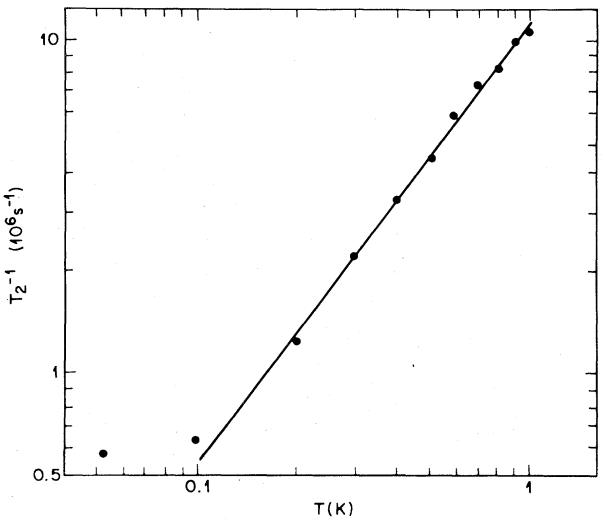

FIG. 3. Temperature dependence of dephasing rate T_2^{-1} of the $^4F_{3/2}(1)$ state of Nd^{3+} in a SiO_2 single-mode fiber. The solid line indicates the temperature dependence $T_2^{-1} \sim T^{4/3}$.

Hence we cannot be confident that T_2^{-1} becomes temperature independent in this region.

The earlier indications of unusual dephasing rates of ions in amorphous solids by FLN[4-6,14] had shown without exception that a near-T^2 temperature dependence persists down to the lowest measured temperature, typically 10 K. Because of resolution problems FLN has not been pursued effectively at lower temperatures and other techniques have been used to study dephasing in the 1.5–25-K region.[7-10] This study represents the first time that very-low-temperature dephasing rates (< 1 K) have been measured on an ion for which there also exist high-temperature FLN data. Our results show that a T^2 dependence cannot persist to arbitrarily low temperatures. We infer that a crossover to a $T^{4/3}$ behavior must occur at a temperature above 1 K. Macfarlane and Shelby[10] have shown that for the $^3H_4$-$^1D_2$ transition of Pr^{3+} in a silicate glass the homogeneous linewidth follows a linear T dependence in the 1.5–25-K region. We expect that in their system a crossover to a high-temperature T^2 behavior may occur.

The most striking feature of the present results is the $T^{4/3}$ temperature dependence of the dephasing rate which is observed for the first time in any inorganic system. Photochemical hole-burning studies on free-base porphin[8] in many organic glasses have shown, however, a $T^{1.3}$ dependence for $0.4 < T < 20$ K. Since the organic system and the measurement technique are quite different from the present case it is difficult to make any quantitative comparison. The remarkable similarity of the T dependence, however, may indicate a fundamental property common to both disordered systems.

Many theoretical models have been developed in an attempt to explain the large amount of data that currently exist on dephasing in glasses. Critical tests of the theories, however, have been hampered by the lack of an extensive set of data on one system. Consequently, the theories have attempted to explain isolated pieces of data. Because of the more extensive knowledge of the dephasing behavior over five decades of temperature for the $^4I_{9/2}$-$^4F_{3/2}$ transition of Nd^{3+} it is instructive to compare our results with some of the theoretical predictions. Theories of Lyo[11] based on an ion-tunneling-system–phonon interaction do not explicitly predict the two distinct power-law regimes occurring in this system. The theory of Huber[13] based on a reexamination of the Raman mechanism predicts a T^2 behavior down to about 20 K but with a faster falloff below this temperature in direct contrast to what we observe. Recently Lyo[16] has examined the role of fractals[17,18] in his theory and has obtained a $T^{1.3}$ behavior for certain interactions. It is unclear at present what role, if any, "fractons" may play in the dephasing process in silica glasses. There is as yet no independent evidence that interactions between tunneling systems and rare-earth ions contribute significantly to dephasing. We expect that a key test of any future theory will be the crossover behavior and how it may represent the blending together of more than one dephasing mechanism.

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