Low-temperature optical dephasing of rare-earth ions in inorganic glasses

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A detailed description is presented of photon echo experiments in Nd^{3+}-doped silica optical fibers at temperatures below 1 K. Dephasing rates T_2^{-1} have been obtained from two-pulse echo sequences at 0.89 μm and are found to follow a $T^{1.3}$ temperature dependence. An interpretation of this result is presented which invokes the interaction of tunneling systems with the optical ions via a dipole-dipole elastic interaction. The dynamics of the interaction originates in a spectral diffusion process and three pulse stimulated echo experiments confirm this interpretation.

I. INTRODUCTION

Relaxation of electronic and magnetic excitations of impurities in amorphous solids shows a characteristically different behavior than in crystalline systems. In various spectroscopies, e.g., electron, nuclear spin resonance[1-3] and optical resonance,[4,5] the relaxation rates in glasses are enhanced compared to their crystalline counterparts and show different temperature dependences at low temperatures (typically $\lesssim 100$ K). This enhancement has indicated the existence of additional relaxation channels, possibly due to extra low-energy excitations, which may be inherent to the amorphous structure.[6] By studying relaxation phenomena in glasses, a probe of such low-lying modes is available and yields information on the excitations and possibly the structure of amorphous systems.

An example is the behavior of the optical homogeneous linewidth of paramagnetic centers in organic and inorganic glasses.[4,5,7] In crystalline compounds the magnitude and the temperature dependence of the homogeneous linewidths are explained by[8] well understood impurity-phonon interactions, such as the Raman process. For a large variety of both organic and inorganic glasses, it has been found that the temperature dependence of the homogeneous linewidth $\Delta\nu_h$ is given by $\Delta\nu_h \sim T^m$, with $1 < m < 2.2$, between approximately 10 and 300 K.[4,5,7,9,10] These temperature dependences and the corresponding magnitudes are consistent with the crystalline values at temperatures $\gtrsim 100$ K, in which regime they are the high-temperature limits of the Raman process. In glasses these temperature dependences surprisingly persist to low temperatures (≈ 10 K) with no obvious "break" in the temperature dependence, as is the case with the crystalline hosts. This unusual behavior has been often ascribed to an interaction of the active center with atomic tunneling systems, the additional excitations which dominate thermal and acoustic properties of glasses at low temperatures.[11-14] However interactions between the impurity and the tunneling system (also called two-level systems or TLS) have not explicitly been demonstrated. Most measurements of $\Delta\nu_h(t)$ in glasses and crystals between 10 and 300 K have used frequency-domain techniques, such as photochemical and nonphotochemical hole burning[5,10,15] and fluorescence line narrowing[4,7,9] (FLN). Below this temperature range, the linewidths become too narrow in the case of inorganic glasses to be conveniently measured with these techniques. In this situation, the linewidth is more readily determined from a direct time-domain measurement of the homogeneous dephasing time T_2. In a simple model, this dephasing time is related to $\Delta\nu_h$ by the relation

$$\Delta\nu_h = \frac{1}{\pi T_2} + \frac{1}{2\pi T_1} , \tag{1}$$

with T_1 the radiative decay time.

In this paper we discuss optical dephasing of Nd^{3+} ions in the pure, amorphous SiO_2 core of an optical fiber below 1 K, in order to investigate deviations at very low temperatures from the previously mentioned temperature dependence.[16] Two- and three-pulse photon echoes have been observed and used to measure the dephasing time T_2 and the lifetime T_1. These coherent transients are the optical analog of spin echoes and have previously been used in crystalline systems to measure homogeneous linewidths or hyperfine splittings.[17-19] In amorphous solids, their use has been rather limited[10,20] due to the much larger decay rates at low temperatures ($\gtrsim 10$ K). We reported previously on a remarkably different temperature dependence of T_2^{-1} below 1 K.[16,21] It was noted that the measured dephasing rates are similar to dephasing rates of the atomic tunneling systems. A model incorporating a spectral diffusion process in which the ion interacts with the tunneling systems through an elastic coupling was shown to account for the echo decay.[21] We have extended these two-pulse echo measurements to three-pulse or stimulated echo sequences and verified the presence of spectral diffusion in the temperature regime below 1 K.

II. EXPERIMENTAL DETAILS

A. Nd^{3+} in amorphous SiO_2

The active Nd^{3+} ions, embedded in the silica core of an optical fiber, were selectively excited to the inhomogene-

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ously broadened $^4F_{3/2}(1)$ state (see Fig. 1). The $^4F_{3/2}(1)$ state of Nd^{3+} was chosen because higher-temperature homogeneous linewidth date ($\gtrsim 10$ K) are available for this level in a variety of inorganic glasses.[9] This allows for a comparison of the results over a large temperature range. A second advantage of measuring the dephasing rate of this state is the fact that the $^4F_{3/2}(1)$ state is spectrally well isolated with a high quantum efficiency. This results in dephasing rates which, at very low temperatures, are not limited by multiphonon-emission or interband relaxation.[20]

The Nd^{3+} ions are in a pure SiO_2 matrix, in contrast to previous studies on homogeneous linewidths in multicomponent inorganic glasses.[4,7,9,10,20] Since the properties of low-energy excitations in pure silica at very low temperatures are well understood[6] it is advantageous to use SiO_2 as the host for an investigation of the role of TLS-Nd^{3+} interaction in the dephasing process. Incorporation of an impurity such as a Nd^{3+} ion into the pure SiO_2 structure requires the breaking of bridging oxygen bonds.[22] This is the function of the network modifier in the multicomponent glasses, but in SiO_2 the rare-earth ion acts as its own modifier. This results in a distorted ligand environment of the Nd^{3+} ion and this is reflected in its spectral properties. From absorption and 1.06-μm luminescence excitation spectra, the splitting of the two Stark components of the $^4F_{3/2}$ manifold in the SiO_2 host was found to be approximately 430 cm^{-1}, which is unusually large compared to the splitting in other Nd^{3+}-doped multicomponent glasses.[9] The inhomogeneously broadened $^4F_{3/2}(1)$ state has a FWHM (full width at half maximum) of ≈ 100 cm^{-1} and is centered at 0.89 μm which is red shifted compared to the same transition in the other glasses. The fluorescence lifetime of the $^4F_{3/2}(1)$ state was found to be 490 μsec at 300 K. This is consistent with values for this transition in the other glasses[9,23] and indicates that fluorescence quenching due to Nd^{3+}-Nd^{3+} cross-relaxation is negligible.

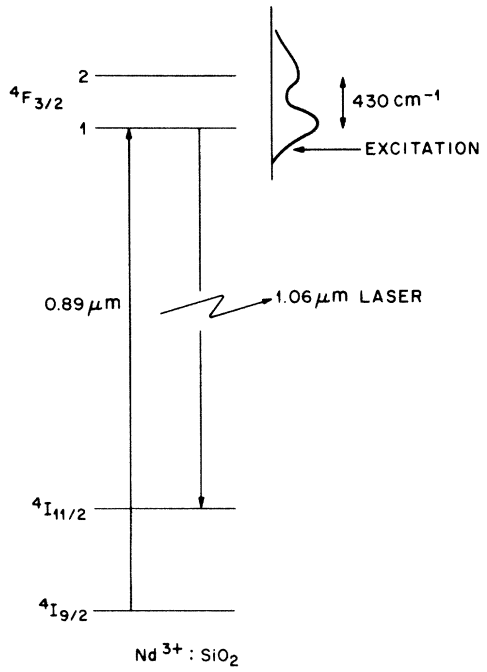

FIG. 1. Schematic energy level diagram of Nd^{3+}:SiO_2 and corresponding absorption spectrum of the $^4I_{9/2} \rightarrow {}^4F_{3/2}$ transition.

B. Optical fiber

As mentioned above, the Nd^{3+} ions are embedded in the core of a single mode optical fiber. This unusual technique of using fibers in a photon echo experiment has several advantages. Excitation pulses travel collinearly because of the waveguide structure of the sample and hence phase matching is automatically satisfied. Also the laser power required for echo generation is moderate because of the high intensities propagating in the small area of the fiver core. A very attractive feature is that any optical density, for a given excitation wavelength, is available by adjusting the fiber length, while keeping the Nd^{3+} concentration low. The latter is important for avoiding ion-ion interactions which may enhance the single-ion dephasing rate. The relatively large length of the 20—30-m sample and the consequent large surface area allows for effective cooling below 1 K in the environment of a ^{3}He-^{4}He dilution refrigerator (as discussed in Sec. II C).

The 75-μm diameter fiber was fabricated from a preform made by the modified chemical vapor deposition process[24] (MCVD) and was designed to propagate the single LP_{01} mode at 0.9 μm. The fiber consisted of a 6-μm diameter Nd^{3+}-doped silica core surrounded by a depressed index, fluorine-doped silica cladding, which is surrounded by silica from the support tube. The Nd^{3+} concentration of approximately 10 ppm contributes a room-temperature attenuation of ≈ 400 dB/km at 0.9 μm. The water content of 0.15 ppm (weight) in the optically guiding region was revealed by an absorption peak of 8 dB/km at 1.38 μm, the first overtone of the fundamental OH stretching vibration.[25]

C. Cryogenics

The optical fiber was cooled to temperatures as low as 50 mK in a ^{3}He-^{4}He dilution refrigerator. The fiber was greased to a 10-cm diameter coil-foil cylinder. This was made by forming a flexible sheet of 0.25 mm copper wires embedded in General Electric 7901 varnish with subsequent formation into a cylinder. Bending losses in the fiber due to differential thermal contraction were reduced by this mounting procedure. Undoped single mode fibers brought the light in and out of the low-temperature environment of the refrigerator and were connected to the active fiber with fusion splices. Transmission losses in the undoped fibers were negligible and splice loss varied between 1 and 3 dB.

Thermometry was accomplished with a doped germanium resistor calibrated against a diluted cerium manganese nitrate susceptibility device. This in turn was calibrated against the National Bureau of Standards superconducting standard. The thermal gradient between thermometer and fiber is probably negligible above 100 mK but may become significant below this. Thus the temperature

dependence of T_2^{-1} below 100 mK may contain contributions from thermometry errors.[16] Heating of the fiber core due to nonradiative decay may also contribute to a thermal gradient.

D. Generation and detection of photon echoes

The experimental configuration to generate two- or three-pulse sequences and detect the resulting photon echoes is shown in Fig. 2. Two-pulse sequences were generated either with dye laser 1 in conjunction with a variable optical delay line (0—50 nsec) or with one pulse each from dye lasers 1 and 2, triggered in a master-slave configuration. Dye laser 1 used LDS 867 dye[26] pumped by the second harmonic of a Nd:YAG laser (YAG denotes yttrium aluminum garnet) and dye laser 2 used a mixture of IR144-IR125 dye[26] pumped by a N_2 laser to generate pulses at 0.89 μm with $\lesssim 10$ nsec FWHM pulsewidths. For stimulated echo measurements dye laser 1 and the delay line generated pulses 1 and 2 and dye laser 2 generated the third pulse. The excitation pulses were launched into the (polarization nonpreserving) fiber with parallel polarization to ensure that each point in the fiber experiences the same state of polarization for all excitation pulses.

Optical nonlinearities such as stimulated Raman (SRS) and Brillouin scattering (SBS) are enhanced in waveguide structures and could possibly interfere (e.g., pump depletion) with echo generation. In the long-length limit (fiber length $\gg 1/\alpha$, with α the nonresonant loss at the corresponding Stokes wavelength) the threshold powers for SRS and SBS are approximately 2 and 0.16 W respectively for the present fiber parameters.[27] The power level for a $\pi/2$ pulse ≈ 10 mW under these conditions[27] and hence no complications arise from SRS and SBS.

Very strong multiple two-pulse echoes were observed in the single-laser configuration. The echo intensity was comparable to the transmitted excitation pulses as shown in Fig. 3. These strong echoes allowed detection with a photodiode and no optical gating of the excitation pulses was required. In order to detect the weak echoes at long

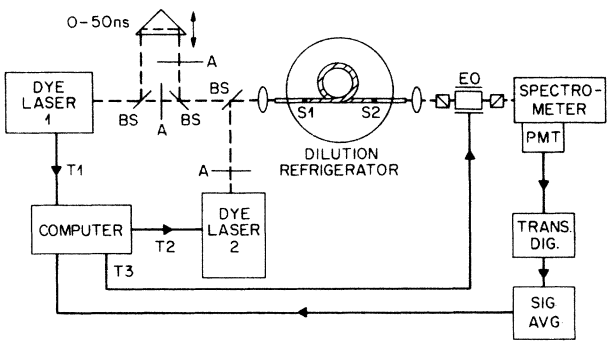

FIG. 2. Experimental setup for generation and detection of two- and three-pulse photon echoes in a Nd^{3+} doped fiber. BS, beam splitter; A, attenuator; EO, electro-optic shutter; S1 and S2, fusion splices between undoped and the doped fiber (shaded) and T1—T3, triggers for synchronizing the firing of the two dye lasers, controlled by a microcomputer.

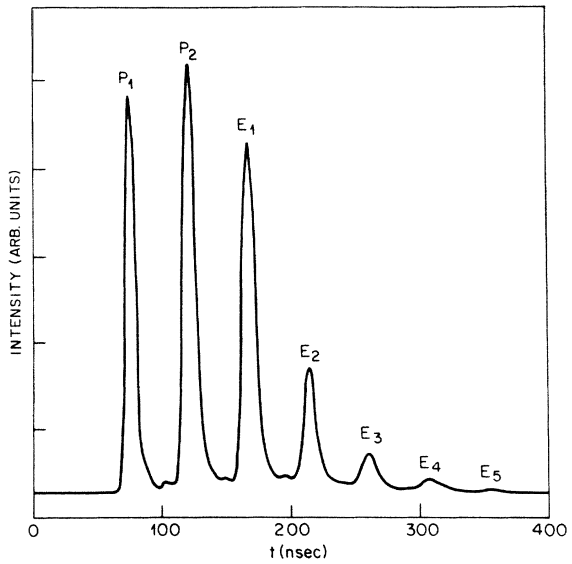

FIG. 3. Two-pulse photon echoes in Nd^{3+}:SiO_2 fiber at $T < 100$ mK. P1 and P2 are (unattenuated) excitation pulses and E1—E5 are generated photon echoes.

delays, an electro-optic shutter—photomultiplier (PMT) combination was used. After detection, the signal from the PMT or the photodiode was analyzed by a transient digitizer and averaged over typically 250 laser shots. The triggering of the two lasers and the synchronization of the electro-optic shutter were controlled by a microcomputer.

III. TWO-PULSE PHOTON ECHO DECAY

The dephasing rate T_2^{-1} was measured between 0.05 and 1 K from the decay of the two-pulse echo intensity as τ_{12} was varied.[16] An example of two (unattenuated) excitation pulses and the multiple resulting echoes (generated with dye laser 1 and the optical delay line) is shown in Fig. 3. The decay of the echo intensity is purely exponential over typically two decades in time. The rate T_2^{-1} follows a simple $T^{1.3}$ power-law dependence between 0.1 and 1 K as shown in Fig. 4. This temperature dependence contrasts with the approximately T^2 dependence above 10 K in similar glasses.[9]

In order to explain the weaker temperature dependence in this regime a model for optical dephasing in glasses has been developed which invokes interactions between the ion and atomic tunneling systems.[21] Not only the temperature dependence but the magnitude of the dephasing rate of the spontaneous photon echo are remarkably similar to the corresponding quantities of the tunneling systems[6] as shown in Fig. 4. Since tunneling system properties have been successfully described with a spectral diffusion model, we take as a starting point a similar model for describing the optical dephasing at these low temperatures. This spectral diffusion model was originally applied by Klauder and Anderson[28] and Mims[29] for interacting two-level spin systems. Applied to the present case, the physical picture is that of modulation of the eigenstates of the ion by strain noise caused by thermal flipping of tunneling systems. A diagonal coupling between the

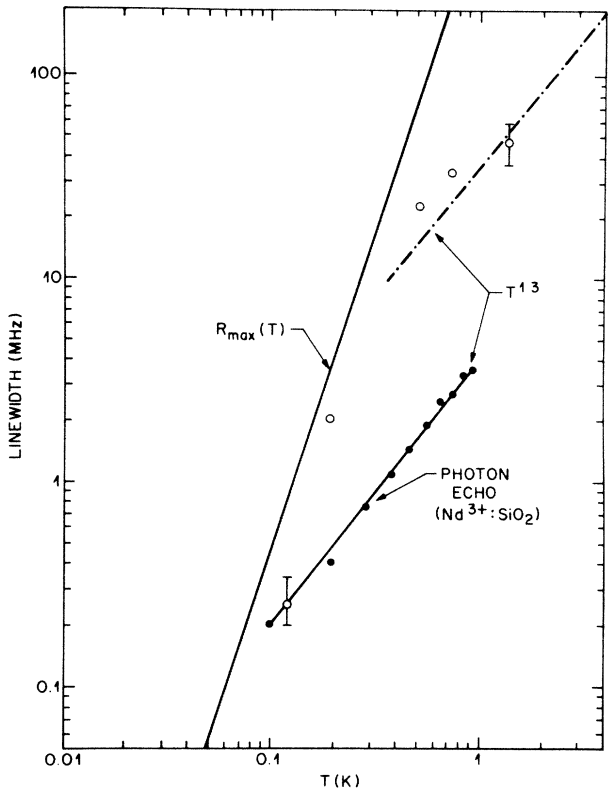

FIG. 4. Homogeneous linewidth of atomic tunneling systems (open circles) in SiO_2 glass and observed dephasing rates of Nd^{3+}:SiO_2. $R_{max}(T)$ is the maximum spin-lattice relaxation rate for tunneling systems with splitting $E=2kT$.

tunneling system and the ion is assumed and the time development of the homogeneous linewidths is described by a diffusion kernel.[21,30]

For a multipolar interaction A/r^n between the ion and a tunneling system the echo amplitude is given by[21]

$$E(2\tau)=\exp[-Cn_0A^{3/n}\langle R^{-3/n}f(R\tau)\rangle] \quad (2)$$

with C a constant, n_0 the spatial density of tunneling systems, and f a function describing the particular frequency modulation model. R is the spin-lattice relaxation rate of the tunneling system:

$$R\sim E^3\exp[-2(\lambda-\lambda_{\min})]\coth(E/2kT) ,$$

with E the tunneling system splitting and λ a tunneling parameter related to the double-well separation.[6] An important step in applying the spectral diffusion model to tunneling systems is averaging over both E and λ as indicated in Eq. (2) with the brackets. This results in

$$\langle R^{-3/n}f(R\tau)\rangle = \int\rho(\epsilon,\lambda)y(E/2kT)R^{-3/n}f(R(E,\lambda)\tau)dEd\lambda , \quad (3)$$

with ρ the normalized density of states of the tunneling systems, $y(x)=\mathrm{sech}^2x$, a thermal cutoff factor limiting the average to tunneling systems with splittings $E\lesssim kT$ and $\lambda_{\min}<\lambda<\lambda_{\max}$. With the following substitutions, $\rho(E,\lambda)\sim E^{\mu}$, $x=E/2kT$, $r=e^{-2(\lambda-\lambda_{\min})}$, $\tilde{R}\sim x^3T^3\coth x$, and $u=\tilde{R}r\tau$, Eq. (3) yields

$$\langle R^{-3/n}f(R\tau)\rangle\sim T^{1+\mu}\int_0^{\infty}x^{\mu}y(x)dx\tau^{3/n}\int_{\tilde{R}r_{\min}\tau}^{\tilde{R}(x,T)\tau}\frac{f(u)du}{u^{1+3/n}} . \quad (4)$$

The lower limit of the last integral in this expression can be set to zero: tunneling systems with large relaxation times much greater than the observation time τ cannot contribute significantly to the dephasing.[31,32] Similarly, as can be seen from Fig. 4, the maximum relaxation rate, $\tilde{R}(x=1)$, is larger than the observed dephasing rates. Hence the upper limit in this integral can be set to infinity. In addition, the asymptotic limit of $f(u)=u^{3/2n}$, and hence the integral converges. This yields

$$E(\tau)=\exp(-KT^{1+\mu}\tau^{3/n}) , \quad (5)$$

with K a constant. Equation (5) is consistent with the observed exponential decay of the two-pulse echo if $n=3$ (dipole-dipole). The observed temperature dependence of T_2^{-1} indicates $\mu=0.3$. This particular energy dependence of the density of states is in agreement with the value obtained from specific-heat measurements on SiO_2.[33] It should be emphasized here that the exponential time dependence follows from averaging over λ in the long-time regime.[21,31,32] Without this averaging, $\ln E(2\tau)$ would vary asymptotically as $\tau^{3/2n}$.

The constant K in Eq. (5) is proportional to $n_0A^{3/n}$. Measurements of the sound velocity in the doped core of the preform from which the fiber was drawn have shown that n_0 is approximately the same in Suprasil W (in which the tunneling system dephasing rates were measured).[34] The interaction strength A can be obtained from a comparison of the photon echo and tunneling system dephasing rate data.[21] This evaluation reveals[21] that the interaction between the ion and the tunneling system is not electrostatic in nature, as had been assumed previously,[11] but rather elastic through the orbit-lattice coupling. A similar analysis[21] of the corresponding optical, homogeneous linewidth shows in general a $T^{(1+\mu)n/3}$ dependence. Although for $n=4$ a roughly quadratic temperature dependence is predicted, the corresponding magnitude is orders of magnitude smaller than the observed linewidths in the regime where the "typical" T^2 dependence is observed ($T\gtrsim 4$ K).[21,34] This suggests that only below ~ 1 K are tunneling systems the dominant contributor to optical dephasing. This conclusion should be contrasted with previous theories[11-13] which attempted to explain the anomalous T^2 dependence of the homogeneous linewidth by assuming the ion-tunneling system interaction to be relevant over the entire temperature range (up to $\simeq$100 K).

IV. THREE-PULSE PHOTON ECHOES

The key concept in our model to explain the dephasing rate is spectral diffusion. The presence of spectral diffusion can be verified directly by using three-pulse (stimulated) photon echoes. In this pulse sequence the first two pulses interfere in time and generate a "frequency comb"

between the ground and the excited state. The third pulse, at time τ_{13} after pulse 1 (with $\tau_{13} \gg \tau_{12}$, normally), scatters off this grating and the amplitude of the resulting (stimulated) echo depends on the extent to which this comb has decayed at the time of the third pulse. This loss of contrast results from population (radiative) decay and additional processes which may wash out the grating modulation depth. Spectral diffusion across the comb is one such process; in the absence of any spectral diffusion the echo amplitude decay is given by

$$E(\tau_{12}+\tau_{13}) \sim \exp(-2\tau_{12}/T_2)[\exp-(\tau_{12}+\tau_{13})/T_1], \quad (6)$$

showing a single exponential decay as a function of τ_{13} governed by T_1. If spectral diffusion is present, however, a fast nonexponential decay occurs for times much less than T_1 with a parametric dependence on τ_{12} in addition to the decay caused by T_1 processes.[6] This τ_{12} dependence results from the grating fringe spacing being proportional to τ_{12}^{-1}. Hence by measuring the decay of the stimulated echo intensity as a function of τ_{13}, the presence of spectral diffusion can be verified.

In the present configuration the first two pulses were generated with dye laser 1 ($0 < \tau_{12} < 50$ nsec) and the third pulse ($0 < \tau_{13} \lesssim 2$ msec) was obtained from laser 2. An example is shown in Fig. 5 which shows the partially gated third pulse and the subsequent stimulated echo. The third pulse and stimulated echo themselves are sufficiently intense to produce additional spontaneous echoes.

Typical decays are shown in Fig. 6 and show an initially nonexponential decay. This decay is much faster than radiative decay, which was independently determined from fluorescence decay measurements to have a (temperature independent) decay time $T_1 = 490\ \mu$sec. The asymptotic decay time of the stimulated echo decay shows good agreement with this T_1 value. The decay function clearly indicates the existence of an additional decay channel of the population grating. Two decay curves are shown for different τ_{12} values and the observed dependence of the initial fast decay on τ_{12} indicates the presence of spectral diffusion.

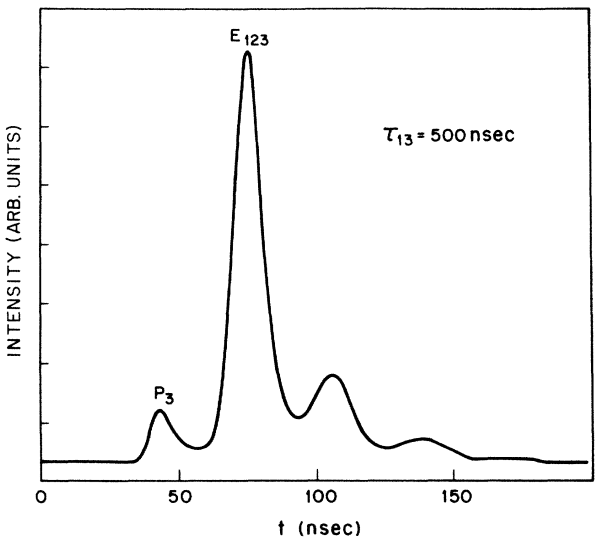

FIG. 5. Stimulated photon echo E_{123} in Nd^{3+}:SiO_2 below 100 mK. The third excitation pulse P_3 is attenuated by ≈ 20 dB. Additional spontaneous echoes are generated by this pulse and the stimulated echo.

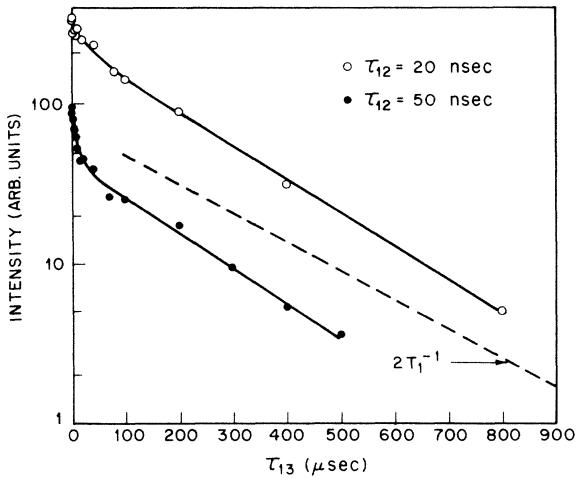

FIG. 6. Stimulated photon echo intensity decay at $T < 50$ mK showing the parametric dependence on τ_{12}. The dashed line indicates the slope of the exponential time dependence the stimulated echo intensity would have if only radiative decay was present.

V. DISCUSSION

The central result of the echo experiment is that the rate T_2^{-1} shows a $T^{1.3}$ temperature dependence below 1 K. This differs significantly from the approximately quadratic temperature dependence of the homogeneous linewidth of the same transition at high temperatures in other organic glasses. The latter behavior is observed in rare-earth-doped inorganic glasses using either FLN or nonphotochemical holeburning techniques. The only exception is the approximately linear temperature dependence between 1.6 and 20 K for a particular transition in a Pr^{3+}-doped glass.[10] Since the density of tunneling systems is roughly constant in various silica glasses, the observation of a linewidth $\sim 40x$ greater than measured here would require a proportionately larger coupling strength for Pr^{3+}, assuming the same interaction mechanism.

A $T^{1.3}$ dependence has been observed in photochemical holeburning experiments in a large variety of organic guest molecules in organic glasses.[5,35] These measurements are typically carried out over one minute, hence they do not necessarily represent a real time measurement of the linewidth. Spectral diffusion has been invoked in some of these compounds[15,36] to explain the time and temperature dependence of the burnt holes. It is not clear, however, whether this same temperature dependence reflects the same underlying physical interaction. In particular quantitative knowledge about tunneling systems in organic systems is very limited or virtually nonexistent. This aspect, the presence of low energy librons, and the possible role of fractal structures of these materials[5,37] in the dephasing process further complicates comparisons between the organic and inorganic systems.

VI. SUMMARY

We have described the observation of two- and three-pulse photon echoes of Nd^{3+} doped in SiO_2 glass fibers below 1 K. The measured two-pulse dephasing rates T_2^{-1} show a different temperature dependence below 1 K than higher temperature homogeneous linewidth data. The similarity of these results with dephasing rates of atomic tunneling systems in SiO_2 glass has led to the quantitative model described herein for optical dephasing mediated by tunneling systems at these temperatures. The model is based on the spectral diffusion concept in which flipping tunneling systems modulate the energy levels of the optical ion through an elastic dipole-dipole coupling and hence dephase the eigenstate of the impurity. Stimulated photon echo measurements clearly indicate the presence of spectral diffusion and independently offer confirmation of the spectral diffusion model.

Finally, this model of the ion-tunneling system interaction is a low-temperature theory. Although higher-order interaction terms yield an approximately T^2 dependence for the dephasing rate, the theoretical values of homogeneous linewidths above 10 K calculated from the inferred strength of the TLS-ion coupling[21] are much smaller than observed. Hence it appears that the contribution of the tunneling systems to optical dephasing in inorganic glasses is significant only for temperatures $\lesssim 1$ K.

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