Absorption Fluctuations and Persistent Spectral Hole Burning in a Nd^{3+}-Doped-Glass Waveguide

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The observation of statistical absorption fluctuations of an inhomogeneously broadened optical transition in a glass is reported. Frequency-modulation spectroscopy in a 10-ppm Nd^3-doped-silica optical fiber has also revealed spectral holes that persist for days at 4.2 K. Homogeneous linewidths Γ_h measured using hole burning and fluctuations are in good mutual agreeement but are significantly larger than those inferred from photon-echo decays in the same system. This difference confirms recent predictions of a time-dependent $\Gamma_h(t)$ arising from tunneling dynamics of the glass.

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Optical transitions of rare-earth ions in glass hosts are inhomogeneously broadened on a scale much greater than the corresponding transitions in crystals as a result of the static disorder in the host. Dynamic changes in the local strain at a site modulate the ion transition energy and result in dephasing of the transition. Using persistent spectral hole burning and by observing the statistical fluctuations in the absorption coefficient as a function of energy, we have measured the dephasing contribution to the homogeneous linewidth of an optical transition of Nd^{3+} in pure silica glass at low temperature. We compare both of these long-time measurements to short-time measurements of T_2^{-1} using two-pulse photon-echo decay in the same system.[1] We conclude that the optical homogeneous linewidth is time dependent, confirming recent predictions for this system.[2] This time dependence arises from coupling of the ion to tunneling systems whose relaxation rates vary over many orders of magnitude. These optical experiments on a time scale of 100–1000 s are thus the analog of time-dependent specific-heat measurements, which have demonstrated the existence of slowly relaxing tunneling systems in a silica glass on similar time scales.[3]

The random nature of the inhomogeneous broadening of optical-absorption lines in glasses (and crystals) implies that there exist fluctuations in the absorption coefficient α as a function of energy that are proportional to $\sqrt{N}$, where N is the number of centers contributing to the absorption. Under some circumstances these fluctuations may be large enough to be observed experimentally.[4] The homogeneous linewidth can be obtained from a measurement of the energy range over which these fluctuations are correlated. We report here the first observation of such fluctuations in a glass host, a single-mode optical-fiber waveguide whose core is pure silica doped with 10 ppm of Nd^{3+} ions. The experimental observation of fluctuations in α is facilitated by centers which produce a large optical absorption. In this case, where the optical dipole moment of the Nd^{3+} transition studied is very small (0.01 D), we have enhanced the sensitivity by adopting a waveguide geometry.

The optical transition studied was the lower Stark component of the $^4I_{9/2}$-$^4F_{3/2}$ manifold of Nd^{3+}, centered at 895 nm. Lifetime broadening contributes only ~ 1 kHz to the homogeneous width, negligible on the scale of the additional pure dephasing mechanisms due to the glass host. Frequency resolution on the scale of the homogeneous linewidth (~ 10 MHz at 1 K, $\sim 10^{-7}$ of the transition energy) is provided by a tunable dye laser with linewidth < 1 MHz. In order to resolve the small differences in absorption due to statistical fluctuations or hole burning, we have used frequency-modulation (FM) spectroscopy[5] with a modulation frequency of ~ 500 MHz allowing detection above the laser amplitude noise bandwidth.

The size of the statistical fluctuations in absorption is dependent on the number of ions in resonance with the laser, i.e., those within one homogeneous linewidth of the laser frequency. If the density of these ions is ρ, then the FM signal is

$$I(\nu_m) \sim \frac{\sigma}{\sqrt{A}} [(\rho L)^{1/2} e^{-2\sigma\rho L}], \quad (1)$$

where σ is the optical cross section of the transition, and A and L are the beam area and length within the sample.[6] The optical cross section of our Nd^{3+} transition is small, and so to maximize the FM signal due to the fluctuations while maintaining low Nd-ion density[7] it is necessary to use small beam areas and long samples, criteria fulfilled ideally by a single-mode optical-fiber waveguide with dimensions $A = \pi \times (3\ \mu\text{m})^2$, and $L = 2$ m. The fiber was immersed in liquid helium, and temperatures were measured using a germanium resistance thermometer calibrated against ^{4}He vapor pressure.

Figure 1 shows the FM signal due to sample absorption versus freuqency at 4.2 K for a region near line center. The upper and lower traces were taken at different times, and show reproducible static fluctuations in the absorption versus frequency. The anplitude of these features is $\Delta\alpha/\alpha \sim 10^{-4}$. We estimate the number of ions contributing to the absorption to be 10^9, implying fluctuations of 3×10^{-5}, in agreement with our observa-

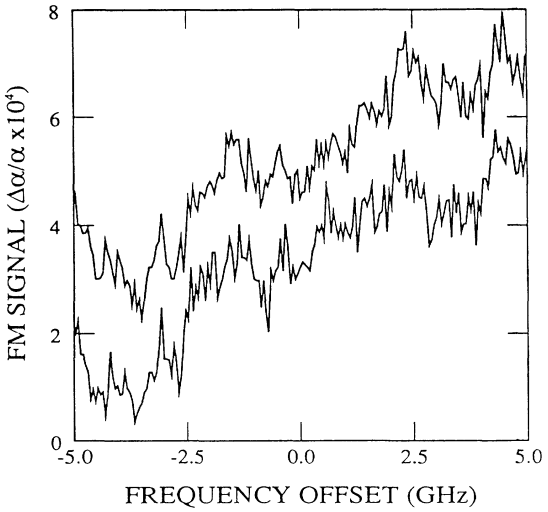

FIG. 1. Two separate measurements of $\alpha(\omega)$ vs frequency, at times separated by 10 min, showing the reproducibility of the low-frequency absorption fluctuations. Nominal temperature was 4.2 K. Vertical scales are slightly offset for clarity. The laser intensity at which these traces were taken was 180 W/cm^2.

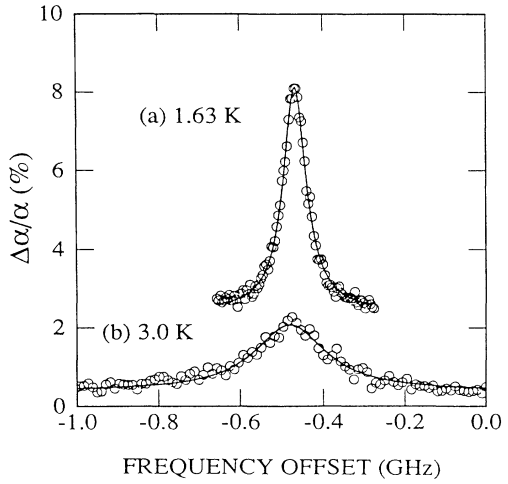

FIG. 2. Persistent spectral holes burnt in the inhomogeneous line at two different temperatures. In (a), the burn time was 200 s and the laser intensity was 0.1 W/cm^2. In (b), 300 s and 0.6 W/cm^2.

tion. Similar absorption fluctuations were seen in adjacent regions of the line, and no such fluctuations were seen when the laser was tuned away from the Nd^{3+} absorption. The value for Γ_h obtained from the autocorrelation of the absorption fluctuations shown in Fig. 1 and those taken in other parts of the line is 400 ± 200 MHz.[8]

Figure 2 shows persistent spectral holes burnt in the inhomogeneous line at different temperatures. The solid lines are fits with a Lorentzian line shape. The maximum depth of hole burnt under any conditions was ~5% of the total absorption, and this depth decayed to 10% of its original value after 24 h. No increase in width was seen at times after 100 s (the typical time scale for burning and reading a hole).

The variation of hole width with temperature is shown in Fig. 3.[9] All these holes were burnt for 300 s with a laser intensity of ~0.1 W/cm^2, with the exception of the point at 4.2 K, which represents the width at the lowest laser intensity for which a hole could be measured. The straight line is a fit of the low-intensity points by $\Gamma = 14T^{1.5\pm0.1}$ MHz. Also plotted are previous measurements of T_2 from two-pulse photon echoes.[1] The straight-line fit to these points is $1/\pi T_2 = 3.6T^{1.3}$ MHz. We note that the width at 1 K extrapolated from the hole-burning data is *4 times larger* than that inferred from the photon-echo decay time. This difference is not an experimental artifact, but represents a real time evolution of the linewidth between the 100-ns time scale of the photon-echo experiment and the 100-s time scale of the hole-burning experiment.

The magnitude of the homogeneous linewidth and its variation with temperature can be explained by dephasing arising from the strain coupling between ions and tunneling systems in the glass.[10] The presence of tunneling systems in glasses explains the thermal properties of glasses at low temperatures, and the standard model[11] visualizes the tunneling systems as atoms or groups of atoms that can tunnel between two almost equal potential wells, with a distribution of energy splittings and barrier heights. The parameters of this distribution have been studied in some detail in silica glass.[12,13]

The time evolution of the optical linewidth due to interactions with the tunneling systems is closely related to similar phenomena seen in phonon-echoes and acoustic-saturation experiments on resonant tunneling systems in

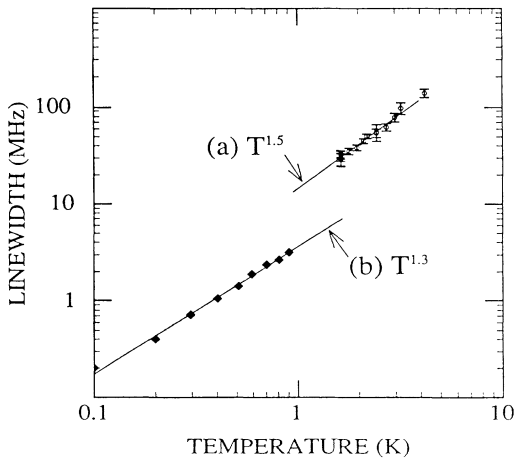

FIG. 3. Homogeneous linewidth vs temperature measured using both hole burning and photon echoes. Circles represent $\Gamma_{hole}/2$, and diamonds represent $1/\pi T_2$ from photon echoes.

glasses.[12,14] The physical basis for the broadening is the shift of the ion resonant frequency by dipolar coupling to the strains produced by thermally induced flips of the surrounding tunneling systems with $\Delta\omega \propto A/r_{ij}^3$. Here A is the coupling constant dependent on the deformation potential of the ion, and r is the distance between ion and tunneling system. The result of the interaction of the ions with many tunneling systems is spectral diffusion of their resonant frequencies, which broadens the line.[15] In order to calculate this broadening for the photon-echo and optical-absorption experiments, it is necessary to average the echo decay, $E(2\tau)$, or line-shape function, $g(\omega-\omega_0)$,

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

$$g(\omega-\omega_0) = \int_0^\infty dt \cos[(\omega-\omega_0)t] \times \exp[-C'n_0A^{3/n}\langle R^{-3/n}f'(R\tau)\rangle] \quad (3)$$

over the tunneling-system energies E and relaxation rates R. Here, n_0 is the density of tunneling systems, and $f, f'(R\tau)$ are functions that depend in detail on the stochastic model considered, but have the asymptotic property $f(R\tau)\sim(R\tau)^{3/2n}$ as $R\tau\rightarrow\infty$. The distribution of tunneling-system energies is assumed to be a slowly varying function of energy, typically $P(E)\propto E^\mu$, where $\mu\approx 0$. The relaxation rate of a tunneling system with energy splitting E is dependent on both E and the tunneling parameter λ, assumed to be uniformly distributed over the interval $\lambda_{\min}$ to $\lambda_{\max}$. The spatial distribution is considered to be random. On averaging over the tunneling-system parameters, one obtains[10] $\langle R^{-3/n}f'(R\tau)\rangle \sim \tau^{3/n}T^{1+\mu}$, predicting exponential decays and corresponding Lorentzian line shapes for dipolar interactions (this is, however, distinct from a lifetime-broadening process) along with a nearly linear temperature dependence of the width.

The absorption line shape and echo decay have recently been considered in more detail[2] using a specific stochastic model in which the tunneling systems are treated in a pseudo-spin-$\frac{1}{2}$ formalism. The temperature dependence of the linewidth and decay time from this calculation are in reasonable agreement with the simpler model outlined in Ref. 11. The magnitude of the optical-absorption linewidth at long times is found to be *larger* than the linewidth inferred from photon-echo decay times, confirmed by the data shown in Fig. 3.

The optical-absorption linewidth as $t\rightarrow\infty$ calculated using the tunneling-model parameters appropriate for silica glass is 7 to 8 times that inferred from the echo experiments. Our experiments indicate an increase of a factor of 4 at 1 K, in reasonable agreement with the theory. Our measured value of the linewidth is not seen to change in experiments with total burn plus read times ranging from 10 to 10^4 s, indicating that the linewidth has already evolved to close to its asymptotic long-time value.

Differences in the homogeneous linewidth as measured by photon echoes and hole burning have been reported for molecules in organic-glass hosts,[16,17] and have been interpreted in terms of the tunneling model as developed for inorganic glasses such as SiO_2. However, no thermal or acoustic measurements have been made on these organic systems at low temperatures, so no information is available about the densities of states or relaxation-rate distributions in these materials.

The optical-absorption linewidth varies[2] as $T^{1.4}$ between 1 and 10 K, compared with a short-time linewidth that varies as $T^{1.3}$. Our observed temperature dependence, $\Gamma_h \propto T^{1.5\pm0.1}$, is in good agreement with this theory. The superlinear temperature dependence of the width arises from assuming a distribution of tunneling-system energies $P(E)$ that varies as $E^{0.3}$, which is appropriate for pure silica[18] but is not a universal feature of glasses. Thus the use of this distribution function in other glassy systems is not necessarily appropriate.

As the measurements of the absorption fluctuations are taken on the same time scale as the hole-burning measurements, the time evolution of the homogeneous linewidth measured is expected to be the same for both experiments. The value for Γ_h obtained from the autocorrelation of the absorption fluctuations is 400 MHz at 4.2 K, which is in agreement with the width obtained from holes burnt by long irradiation using the same laser power. However, both these widths are larger than the narrowest hole width observed at the same temperature by a factor of $\sim$3, indicating that at these high laser intensities there is some heating of the fiber core.

The formation of persistent spectral holes requires an optically induced change in the ion absorption frequency with a lifetime >100 s. We note that in our Nd^{3+} system a deformation potential 10–100 times *larger* than that of the corresponding transition in a crystal must be assumed in order to explain the absolute magnitude of the hole width and echo decay. Our interpretation of this increase in deformation potential is that the Nd^{3+} ion itself forms part of a tunneling system, which allows it to couple more strongly to strain. Reorientation of this extrinsic tunneling system will cause the ion frequency to shift by a large amount, so if optical excitation increases the probability of reorientation then tunneling to those ground-state configurations with long relaxation times will produce long-lived spectral hole burning. If, as seems likely, there is a wide distribution of extrinsic-tunneling-system relaxation rates, then our observation of saturation of the hole depth at $\sim$5% is consistent with only seeing the effects of the slowly relaxing tail of the distribution. A similar model has been put forward for hole burning by proton tunneling.[19]

The low efficiency for hole burning in this system puts a limit on the possibility of observation of holes on shorter time scales, because the laser intensity used to burn the hole must be kept below the threshold for power

broadening. However, the measurement of homogeneous linewidths using statistical fluctuations in absorption coefficient are not subject to time restrictions because of low hole-burning efficiency, and so the possibility of extending the measurements to the intermediate time scales (μs to ms) difficult to reach using hole burning remains open.

We note that according to the formalism of Ref. 17, the width of the hole is related to the Fourier transform of the decay of the stimulated photon-echo signal. From the expressions derived previously[10] for the optical-absorption (OA), photon-echo (PE), and stimulated photon-echo decays at long times one obtains[20] $1/T_2^{\mathrm{HB}}s = \frac{1}{2}[1/T_2^{\mathrm{OA}} + 1/T_2^{\mathrm{PE}}]$. Since the fluctuations in the linear absorption coefficient must measure T_2^{OA}, we might expect a difference between the widths obtained from hole burning and fluctuations. However, a test of this relationship requires more accurate values of the linewidths than are available from our present results.

In summary, we have observed reproducible static fluctuations in the absorption coefficient, as well as persistent spectral hole burning, in a Nd^{3+}-doped silica waveguide. The magnitude of the fluctuations in the absorption coefficient agrees with that calculated assuming a simple statistical model. The reproducibility of the fluctuations indicates that on a time scale of 10^3 s the changes in the ion's immediate environment responsible for the hole filling are infrequent. However, one might expect to see some slow evolution of the fluctuation "fingerprint" in a system with sufficient long-term optical and mechanical stability. Values of the homogeneous linewidth obtained from these fluctuations and persistent spectral holes are in agreement, and are larger than those inferred from previous measurements of the photon-echo decay time. This difference in long- and short-time linewidths confirms recent predictions of spectral diffusion due to the slowly relaxing tunneling systems in the SiO_2 host.

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