ELSEVIER

Journal of Luminescence 60&61 (1994) 753–757

JOURNAL OF LUMINESCENCE

Coherent optical transients in rare earth doped fibers: enhancement of accumulated grating echo signals by the maximum entropy method

A.J. Lucero[1], D.S. Hamilton*

Department of Physics, University of Connecticut, Storrs, CT 06269-3046, USA

Abstract

A train of optical mode-locked pulse pairs is used to generate a frequency grating in the ground state of rare earth ions incorporated into the cores of optical fibers held at low temperatures. Detection of the photon echo stimulated from this grating is complicated by the fact that neither spatial nor temporal discrimination may be exploited to separate the echo signal from the pump and probe pulses in a fiber geometry. This obstacle is overcome by applying the maximum entropy method (MEM) of spectrum reconstruction to the digitized data and extracting the strength of the echo signal from the integration of its Fourier component. For small time delay (maximized echo amplitude) signal to background ratios for the power spectral densities of the echo signals of $\sim 10^6$ were consistently obtained with this method. Temperature dependences (15 to 65 K) of the homogeneous linewidths obtained from the echo decay for the $^3H_4 - {}^1D_2$ transition of Pr^{3+} are reported.

1. Introduction

Time-domain studies of the homogeneous linewidths of impurity centers incorporated as dopants in glasses have augmented and complimented our understanding of the dephasing processes obtained from frequency-domain experiments. Knowledge of the dynamics of non-equilibrium glass structures over a wide range of times and temperatures for a variety of systems is important for testing the fundamental theories developed for glasses, as well as for more applied objectives such as development of new laser materials and investigation of proposed optical memory schemes. The complexity of the amorphous state is underscored by contrasts between different systems in the dependences of the homogeneous linewidth $\Delta\nu_H$. For instance, temperature dependence of the dephasing times of electronic transitions of rare earth ions in a number of glassy hosts has been measured variously to be either linear or quadratic over a broad temperature range, or linear at low temperatures with a cross-over to quadratic behavior [1], as opposed to the exponential or T^7 dependence found for RE ions in crystalline hosts. Additionally, $\Delta\nu_H$ has displayed either an increase or a decrease with change in excitation energy across the spectral profile for different systems [2]. In this set of

* Corresponding author.
[1] Present address: AT&T Bell Laboratories.

0022-2313/94/$07.00 © 1994 – Elsevier Science B.V. All rights reserved
SSDI 0022-2313(93)E0512-V

experiments, we have used the accumulated grating echo to measure the temperature dependence of $\Delta\nu_H$ at the most highly electronically strained sites in the extreme long wavelength regions of the absorption band of $^3H_4 - {}^1D_2$ of Pr^{3+} in silicate glass optical fibers. By exploiting the wave guide characteristics of fibers, reasonable levels of absorption can be obtained in low ion concentration samples, thus avoiding the complications associated with high ion concentration. The apparent problem of separation of input and output signal in this collinear geometry is satisfactorily resolved by transformation of accumulated digitized data into the frequency domain by the *maximum entropy method* [3], a spectral reconstruction algorithm familiar in NMR work.

Following our measurement of dephasing rates of Pr^{3+} in silicate fibers as a function both of temperature and exciting wavelengths in the range of 25 to 70 K, in contrast with previous studies, we see evidence of an evolution from a quadratic temperature dependence to a $T^{3.5}$ behavior as populations farther in the absorption band limits are probed. There have been findings presented recently of increased site coordination in the far wings of the absorption band as manifested for example by *statistical fine structure* [4] or by quasi-molecular complexes formed by the RE ions with the host material [5]. In the latter reference, the authors found that frequency line-narrowed spectra of Eu^{3+} in oxide glasses displayed systematic changes with a sensitive dependence on excitation wavelength. It is thus resonable to conclude that this evolution of dephasing rate temperature dependence parallels the distribution of ions from more glassy to more crystalline or coordinated sites.

2. Experiment and data analysis

The experimental layout is illustrated in Fig. 1. A continuous train of frequency-tunable picosecond pulses is produced by a tunable dye laser synchronously pumped by a mode-locked argon ion laser. The beam of pulses is split by a 50/50 pellicle beam splitter. The separate beams pass through an optical chopper and are modulated at frequencies f and $\frac{5}{6}f$, where f is nominally 2.4 kHz. The delay, τ_{12}, between pulses one and two is continuously adjustable between 0 and 2 ns. The beams of pulses are then rejoined at a second pellicle beam splitter and then coupled into an optical fiber. The autocorrelator and optical spectrum analyzer provide beam diagnostics.

The cryostat contains 20–40 m of optical fiber which has been doped with a low concentration of Pr^{3+}. Two meters of undoped fiber and fusion spliced to the doped fiber to bring the light signal into and out of the low temperature environment.

The repetitive nature of an accumulated grating echo experiment [6] produces an echo signal which is orders of magnitude stronger than that produced with either a two- or three-pulse echo experiment. While a collinear pulse geometry maximizes the amplitude of the signal, this experimental arrangement lacks the advantage of exploiting the directional character (phase-matching condition) of a standard photon echo experiment in which the pump and probe are crossed at a small angle to facilitate detection of the weak signal at the angle between them. In the case of a fiber geometry, the echoes are superimposed on the second pulse of each pulse pair. An additional difficulty in extracting the echo amplitude is that the frequency of pulse arrival is far too high to use any type of electro-optic gating device to discriminate between pulses. However, since for a given pulse-pair phase separation the echo is at a maximum only when both the modulated pump and probe beams (beam 1 and 2) are at their maxima, conversion of the data to the frequency domain by a Fourier transform efficiently extracts the power of the echo signal from both the overlapping pump and probe signals and the background noise. The combined signal from the photodiode detector is routed to a storage oscilloscope and the digitized signal is ported to a Sun workstation for analysis. Data accumulated over many cycles is transformed into frequency space by the maximum entropy method (MEM) with the power of the echo signal inferred from the integration of its Fourier component. When the pump and probe beams are modulated at f and $\frac{5}{6}f$, the Fourier component of the echo signal occurs at the difference frequency $f/6$. The MEM provided several advantages over a standard fast Fourier transform (FFT), while requiring only a modest

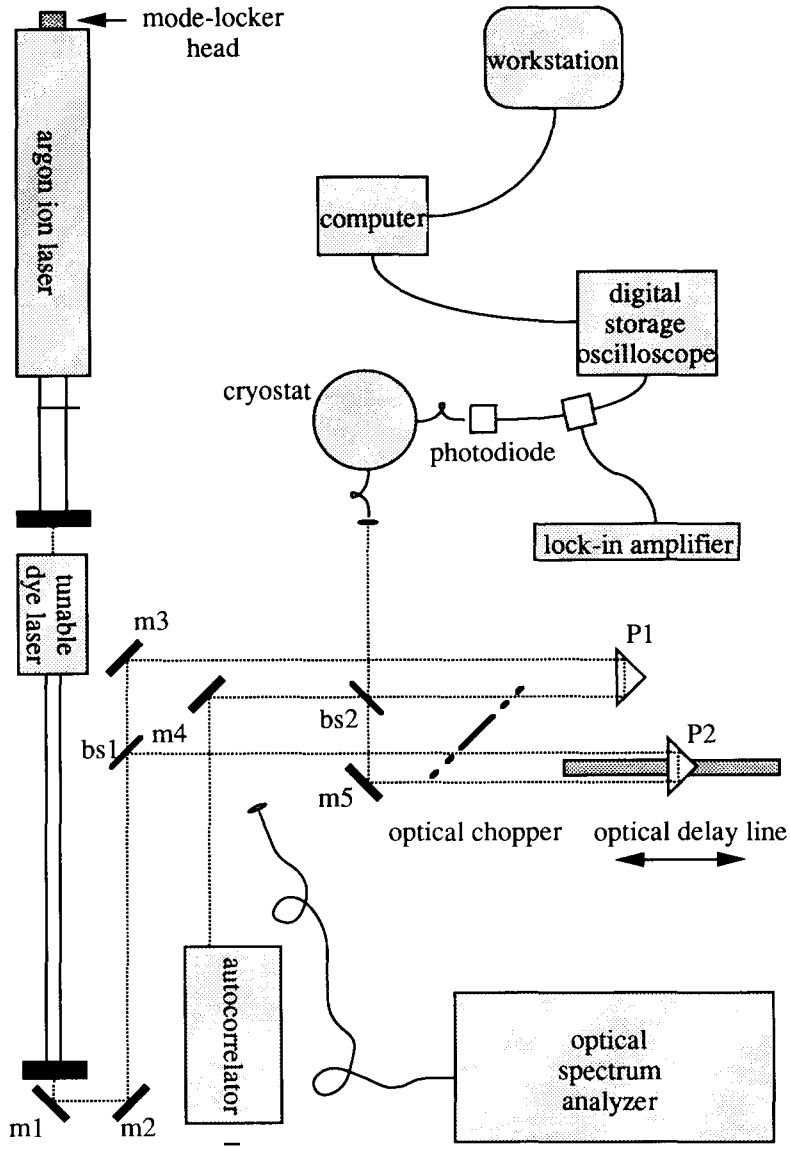

Fig. 1. Experimental arrangement for the accumulated grating echo measurements.

increase in computational time. In addition to giving a much improved signal to noise ratio, the MEM is also free of the constraint of the minimum Nyquist step size, $\delta f = 1/N\Delta t$, where N is the number of data points and Δt is the time step size, thus allowing a spectrum with finer frequency grid to be generated. Finally, by systematically repeating the experiment over an appropriate range of temperatures and exciting wavelengths, the behavior of the dephasing time T_2 over that parameter space may be characterized.

3. Results and discussion

The solid line spectrum in Fig. 2 is an example of the result of raw data processed by the MEM around the 400 Hz center frequency of the echo

signal. To illustrate the contrast between the two transform techniques, the power spectral density as obtained from the standard FFT is also displayed.

The dephasing rates of rare earth and other ions display a markedly different behavior when embedded in a crystal as opposed to a glassy environment. For instance, the temperature dependence of the dephasing rates of Pr^{3+} in LaF_3 is well understood in terms of the two-phonon Raman and the one-phonon direct broadening mechanism [7,8]. The Raman process, which behaves as T^2 above the Debye temperature, falls off as T^7 at lower temperatures, leaving dominant the exponentially activated one-phonon direct process. In glasses the lower power dependence observed at the higher temperatures persists to very low temperatures. This T^m power law ($1.8 < m < 2.2$ depending on the ion), has been well investigated as a function of temperature and to some degree as a function of exciting wavelength. The study of the dephasing times as a function of both of these parameters was the original motivation for an entire set of experiments [9], and has provided an interesting insight into the site coordination of the dopant ion. For example, note in Fig. 3 that the homogeneous linewidth of the Pr^{3+} displays the expected quadratic dependence upon temperature at $\lambda = 618$ nm, but behaves as $T^{3.5}$ slightly further into the longest wavelength region of the absorption band. Taking into consideration the increase in site coordination in the wings of the absorption band referenced in the introduction, we attribute this observation of shifting temperature dependence of the homogeneous linewidths with increasing exciting wavelength to evidence of an evolution from a more glassy environment to a more crystalline one in the farthest low energy wing of the absorption band.

These studies augment the body of data concerning both the dephasing processes in glasses and site-selective spectroscopy, hopefully assisting in elucidating a complex and still not yet fully understood system.

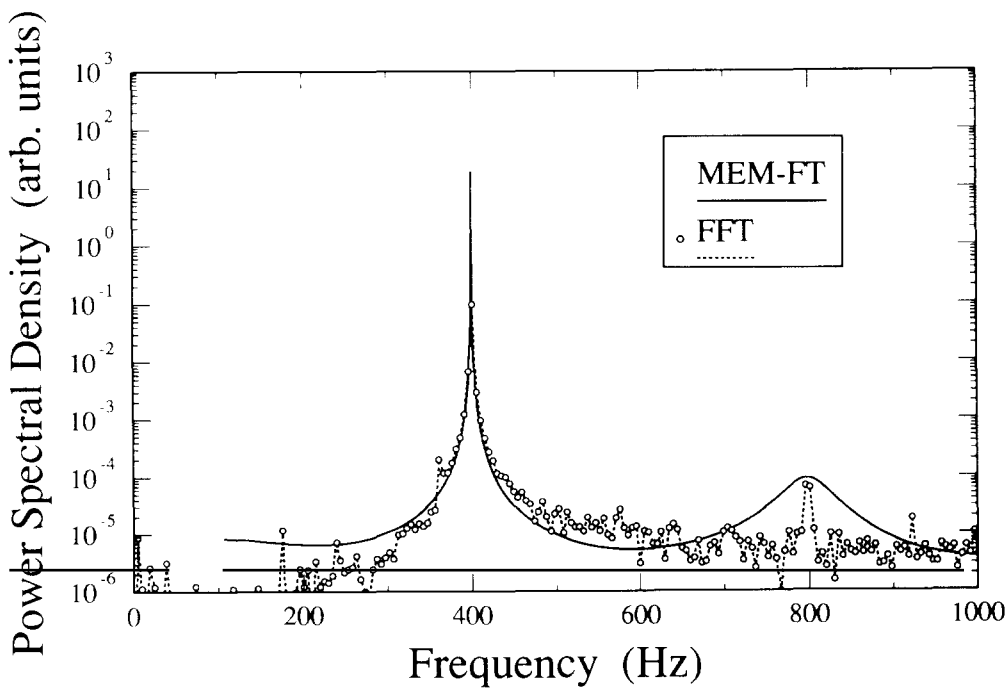

Fig. 2. Detail of a strong echo signal.

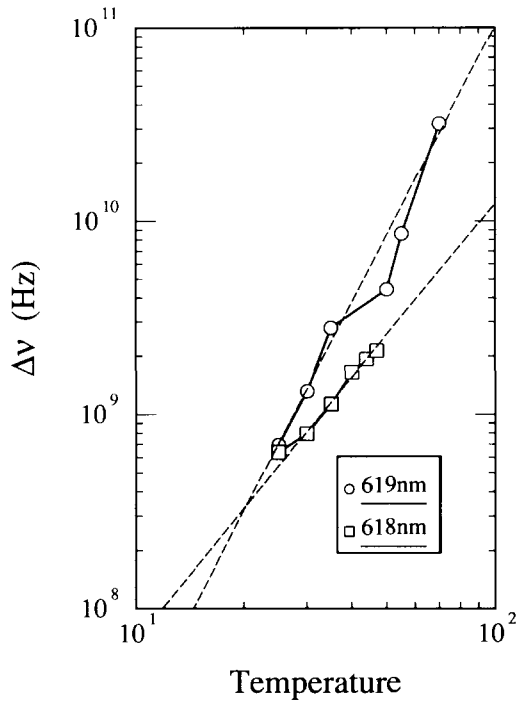

Fig. 3. Homogeneous line widths of Pr^{3+} in a 10^{-6} Pr^{3+} doped silica glass fiber at two different excitation wavelengths.

Acknowledgement

The authors would like to thank Jay Simpson of AT&T Bell Laboratories for providing the RE doped optical fibers used in these experiments.

References

[1] R.M. MacFarlane and R.M. Shelby, J. Lumin. 36 (1987) 179.
[2] H.W.H. Lee, Jour. Lumin. 45 (1990) 99.
[3] W.H. Press, B.P. Flannery, S.A. Teukolsky and W.T. Vetterlin, Numerical Recipes, first ed. (Cambridge, New York, 1986) p. 430.
[4] T.P. Carter, D.E. Horne and W.E. Moerner, Chem. Phys. Lett. 151 (1988) 102.
[5] T.F. Belliveau and D.J. Simkin, J. Non-Cryst. Solids 110 (1989) 127.
[6] W.H. Hesselink and D.A. Wiersma, Phys. Rev. Lett. 43 (1979) 1991.
[7] W.M. Yen, W.C. Scott and A.L. Schawlow, Phys. Rev. 136 (1964) A271.
[8] R. Flach, D.S. Hamilton, P.M. Selzer and W.M. Yen, Phys. Rev. B 15 (1977) 1248.
[9] A.J. Lucero, Coherent Optical Transients in Rare Earth Doped Fibers: Enhancement of the Accumulated Echo Signal by the Maximum Entropy Method, PhD. Thesis, University of Connecticut (1993, unpublished).