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PUMP AND PROBE SPECTROSCOPY OF A Cr^{+3}-DOPED GLASS

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Probe beam modulation in Cr^{+3}-doped glasses induced by a high intensity pump laser is reported. A non-degenerate pump and probe geometry is used. Laser induced clearing and darkening effects and non-linear switching were observed. The effective relaxation time for this system was measured.

1. INTRODUCTION

DURING the past ten years great attention has been given to the study of new optical devices with applications in optical data processing, non-linear switching, phase modulation and bistability [1, 3]. Compared to crystals, glasses are attractive materials for many of these optical technology applications because of their low cost and ease of fabrication. Several authors have suggested the use of Cr^{+3} doped glasses in lasers and other optical devices [4–6]. These glasses have attracted the interest of a number of investigators searching for new hosts to be used as sensitized solid state lasers.

In this paper the results of a study of some non-linear properties of a Cr^{+3}-doped glass are described. A non-degenerate pump and probe technique was used. The glass matrix has a general formula $(SiO_2)_{0.5}$ $(Na_2O)_{0.35}(CaO)_{0.15}$ and is doped with 1% Cr_2O_3. Pump-probe dephasing and switching effects were observed and their dependence on the pump intensity and modulation frequency is reported. A comparison is made with previously reported results in some semiconductor-doped glasses [9].

2. EXPERIMENTAL

The experimental set-up used the usual pump and probe technique (Fig. 1). The pump laser was a CW Argon Ion laser at $\lambda = 5145$ Å and the probe beam was from a CW He–Ne laser at $\lambda = 6328$ Å. Pump and probe beams were focused into the sample using a lens of focal length $f = 200$ mm to a minimum spot diameter of about 30 μm. The pump beam was modulated with a chopper with stability of about 1 Hz and possibility to operate at frequencies up to 4 kHz.

Samples of glasses with a formula $(SiO_2)_{0.5}$ $(Na_2O)_{0.35}(CaO)_{0.15}$, pure or doped with 1% Cr^{+3} were used. These glasses were prepared by melting in air the proper amounts of reagent grade compounds, Na_2CO_3, $CaCO_3$, SiO_2 and Cr_2O_3, in an electric furnace. Melting temperatures in the range 1300–1400° C were required. The melted glass was rapidly cooled to room temperature. From the raw glasses, samples were prepared by careful polishing until obtaining surfaces with good optical quality. These samples were cubes with thickness around 4 mm. All measurements were done at room temperature.

The phase difference between signal and pump was measured with the phase control of a lock-in amplifier. Dephasings of 0° and −180° are associated to instantaneous laser-induced clearing and darkening, respectively. For intermediate values, between 0° and −180° one has delayed darkening.

The time dependence of the probe beam intensity was observed collecting the probe beam, after traversing the sample, on a fast photodiode and feeding the signal to an oscilloscope.

3. RESULTS AND DISCUSSION

Figure 2 shows the absorption spectra of two samples of the glass. The absorption spectrum of Fig. 2a is from an undoped sample, while that of Fig. 2b is from a glass doped with 1% Cr^{+3}. Two

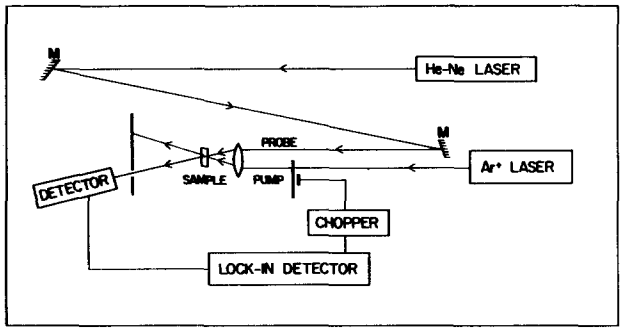

Fig. 1. Diagram of the experimental set-up.

broad bands are seen in the absorption spectrum of Fig. 2b. They are typical of Cr^{+3}-doped oxide glasses [7]. One band near 442 nm is associated to the $^4A_2 \rightarrow {}^4T_1$ transition, and the other band, near 660 nm, is due to the $^4A_2 \rightarrow {}^4T_2$ transition, on which are superimposed Fano resonances of the $^4A_2 \rightarrow {}^2E$ transitions. These bands, extending up to the near infrared, make it possible to study the optical properties of these glasses using lasers in the visible range.

In Fig. 3 it is seen the strong dependence of the pump-probe dephasing with the pump intensity and pump pulse chopping frequency. One can go from laser induced darkening to an instantaneous laser induced clearing (dephasing = 0°). All the intensity dependence results were obtained as the pump intensity was increased from zero to the highest available intensity I_0, while the chopper frequency was kept constant.

The variation of the probe signal intensity with pump frequency was observed and is shown in Fig. 4. Assuming an exponential decay with a relaxation time τ one can reasonably fit the experimental data to a Lorentzian by taking $\tau \cong 18$ ms (Fig. 4). There is good concordance for low and high frequencies. It is clear that the signal behaviour with time is more complex than a simple exponential decay.

These results can be attributed to two non-linear mechanisms:

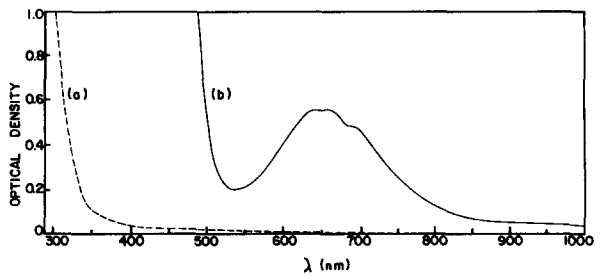

Fig. 2. Absorption spectra of $(SiO_2)_{0.5}(Na_2O)_{0.35}(CaO)_{0.15}$ glasses. (a) undoped sample. (b) 1% Cr^{+3} doped. Thickness = 4 mm.

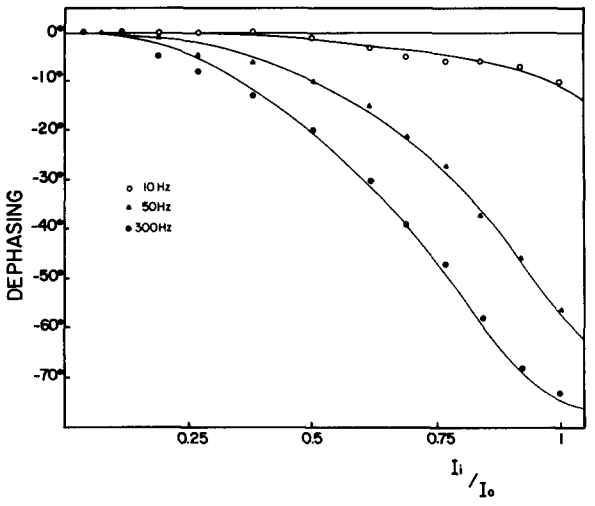

Fig. 3. Dephasing between pump and probe modulation versus pump power. $I_0 = 368\,\mathrm{kW\,cm^{-2}}$, $I_{probe} \sim 0.1\,\mathrm{kW\,cm^{-2}}$.

(a) a clearing or darkening effect connected with the thermal shift of the absorption band due to the absorption of the pump radiation and

(b) heat conduction through the sample according to the temperature transport equation [8]:

$$\frac{\delta T}{\delta t} = \frac{\delta E(\alpha, t)}{\delta t} + \frac{T - T_0}{\tau_1} + D\nabla^2 T \qquad (1)$$

where E is the absorbed energy which depends on the laser intensity, D is the heat diffusion constant, α is the absorption coefficient, T_0 is the surrounding air temperature and τ_0 is the effective relaxation time. The

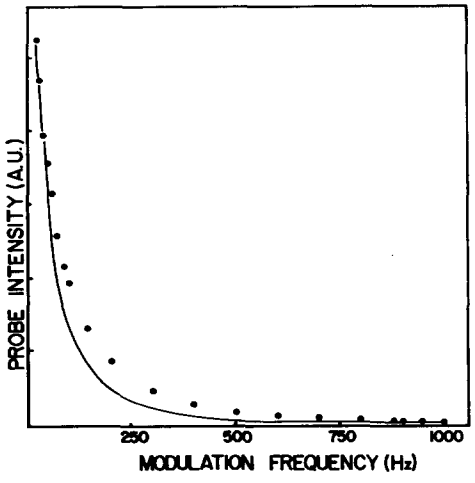

Fig. 4. Probe intensity versus modulation frequency. $I_{pump} = 212\,\mathrm{kW\,cm^{-2}}$, $I(f) = I_0/(\tau^{-2} + f^2)$. Continous line is $I(f)$ with $\tau = 18$ ms and $I_0 = 5000\,\mathrm{Hz^2}$. (●) Experimental data.

magnitude of τ_0 is given by [10]:

$$\frac{1}{\tau_0} = \frac{1}{\tau_l} + \frac{1}{\tau_t} \tag{2}$$

$$\tau_t = r_0^2 (1 + 2 \ln (R/r_0))/(4D) \tag{3}$$

$$\tau_l = \frac{dK}{2Dh}. \tag{4}$$

Here τ_l is the longitudinal diffusion time, τ_t is the transverse diffusion time, d is the sample thickness, K is the heat conductivity, h is the heat convection number, R is the whole sample radius and $2r_0$ is the minimum spot diameter. For the experimental conditions above described, τ_0 is about 2 ms. It can be concluded, from these results, that for high modulation frequencies ($f \cong 300$ Hz, $\tau_{pump} \cong 1.6$ ms $\cong \tau_0$) and high pump intensity the second mechanism is more important in inducing the delayed darkening effect seen in Fig. 3. The effect is smaller for low frequencies ($f \cong 10$ Hz, $\tau_{pump} \cong 50$ ms $\gg \tau_0$) and low pump power. The dephasing behaviour of this system is then different from the one observed in other silicate glasses, doped with semiconductor impurities, where the dephasing is larger for low modulation frequencies [9].

The time behaviour of the probe pulse for different values of the pump power. (Fig. 5) shows a clear process of non-linear switching. This switching behaviour is controlled by the transient heat flow that establishes the steady state temperature profile. At a certain threshold of the pump power the probe transmitted power decreases and there appears a fragmentation of the probe pulse (Fig. 5).

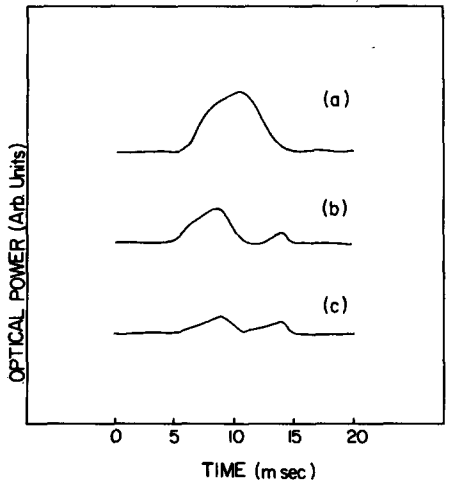

Fig. 5. Induced non-linear probe pulse switching in a Cr^{+3}-doped glass. Vertical scale is arbitrary. Values of I_{pump} are: (a) 212 kW cm^{-2}. (b) 340 kW cm^{-2}. (c) 368 kW cm^{-2}.

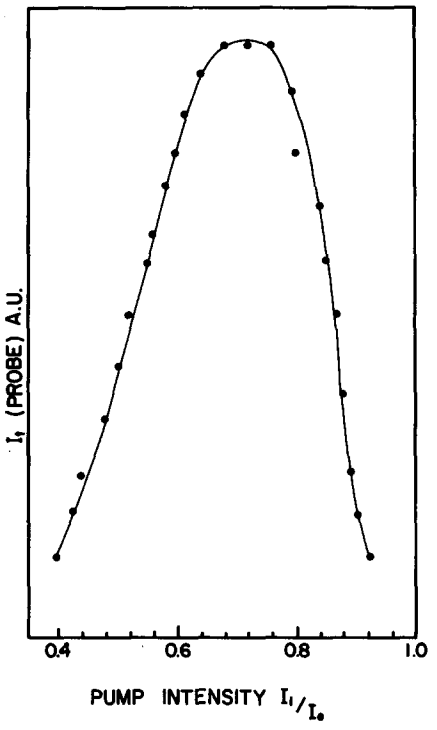

Fig. 6. Transmitted probe intensity versus input pump intensity with $I_0 \sim 400$ kW cm^{-2} [2].

In these experiments the excitation pulses are 5 ms square pulses separated by intervals of 5 ms. The characteristic shape of the induced probe pulse for low intensities of the pump is shown in Fig. 5a. When the pump power is increased the absorption curve edge shifts toward longer wavelengths. This causes a change in the absorption of the probe pulse. The transmitted probe intensity is shown in Fig. 6 as a function of the pump power. There is a maximum transmission for $I \cong 0.7\ I_0$, where I_0 is around 400 kW cm^{-2}.

For pump intensities greater than $0.7\ I_0$ the probe pulse is strongly absorbed. For $I_p \cong 340$ kW cm^{-2} the probe pulse is deformed (Fig. 5b) and for still higher values of the pump intensity the probe pulse is transformed into two pulses resulting from the switching to the high absorption state (Fig. 5c).

A low limit of the switch-off time can be calculated from equation (3) assuming $R = 3$ mm, a spot diameter of 30 μm and $D = 3.8 \times 10^{-3}$. One has then $\tau_0 \cong 1.7$ ms [9]. For these samples, switching times vary from 1 ms to 2 ms, depending on the pump intensity. One has to take into account that transient non-uniformities in the temperature over the sample can affect the switching time behaviour and the effective relaxation time.

In summary, laser induced clearing and darkening

in a Cr^{+3}-doped glass at room temperature were reported. There is a strong dependence of the dephasing between pump and probe with the pump power and with the modulation frequency. Non-linear optical switching was observed in these samples with a effective switching time of about 2 ms. These properties show the potential of Cr^{+3} doped glasses for optical signal processing, switching and transmission at room temperature, using frequencies that go up to a few kHz.

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