Two-step two-color recording in a photorefractive praseodymium-doped $La_3Ga_5SiO_{14}$ crystal

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(Received 12 November 1998; accepted for publication 3 May 1999)

Two-step two-color recording is demonstrated in a photorefractive $La_3Ga_5SiO_{14}$:Pr^{3+} crystal using cw laser radiation. The 488 nm line from an Ar-ion laser is used for gating and gratings are written using a Ti:sapphire laser operating in the range from 788 to 840 nm. The dependence of holographic recording on grating and writing intensity is investigated. A saturation of the sensitivity is found for 2 W/cm^2 of grating intensity. A threshold photon energy of 1.53 eV for the second excitation step is observed. © *1999 American Institute of Physics.* [S0003-6951(99)01726-X]

Several different requirements have to be met before holographic information storage will find use in practical applications. One of the most important demands is that stored information can be read out without erasure. At the same time, however, a reversible process where recorded data can be erased optically is desirable.

An attractive all-optical recording scheme has been suggested in order to meet both the requirements of nonvolatile readout and reversibility.[1] The technique is based on so-called two-step two-color recording. In this process, light with two different wavelengths is used for recording. The first wavelength is used to form an interference pattern (hologram) inside the crystal while the second wavelength is used for gating of the photorefractive process allowing the interference pattern to be transformed into a refractive index modulation. The physics of the gating process is to excite electrons from photorefractive donor centers to the conduction band via a two-step excitation process. In the first step, electrons are excited by a beam at the first wavelength (gating beam) to a localized intermediate state. From this state light of the second wavelength (writing beams) excites the electrons to the conduction band. If the material is insensitive at the wavelength of the writing beams nonvolatile readout using one of the writing beams is obtained simply by shutting off the gating light during readout.

The primary drawback of this technique is that pulsed lasers with intensities of the order of 10^6-10^9 W/cm^2 are usually needed in order to efficiently realize the two-step excitation process.[2–5] In the last few years, however, a variety of materials have been discovered in which two-step excitation can be realized at moderate intensities allowing the use of cw laser sources.[6–8] Common for these materials is the doping with rare-earth Pr ions. Recently, it has been shown that electrons can be excited efficiently, through a two-step excitation process, to the conduction band of $La_3Ga_5SiO_{14}$:Pr^{3+} using the $^3H_4 \rightarrow {}^3P_0$ excitation channel at 488 nm of Pr^{3+}; see Fig. 1.[8] In this letter we investigate the possibility of using this absorption channel for nonvolatile two-step two-color recording.

The crystal $La_3Ga_5SiO_{14}$ is a piezoelectric crystal belonging to the point group 32. For the measurements a sample grown using the Czochralski method with dimensions $5\times4\times3$ mm^3 is used. The crystal is doped with Pr^{3+} with a density of $3{,}5\times10^{20}$ cm^{-3}. In addition to the two-step ionization of Pr^{3+} a secondary center also contributes to the photorefractive effect allowing recording off resonance at 514 nm, see Fig. 1.[9] The main charge transport process involved in the photorefractive process is found to be photovoltaic currents.[9]

Holographic gratings are written using infrared light from a Ti:sapphire laser system operating at wavelengths in the range from 788 to 850 nm. Two TE-polarized writing beams, symmetrically incident at an angle of $\theta=13.5°$, are slightly focused to a diameter of 1 mm onto the sample. The beams propagate along the z axis of the crystal and the grating wave vector is parallel to the x axis. The process is gated using the 488 nm line of an Ar ion laser. The diameter of this beam is 2.5 mm in order to ensure homogeneous illumination of the writing area. The gating beam is polarized at an angle $\beta=53.5°$ with respect to the x axis in order to prevent a strong photovoltaic current to be induced by the gating beam.[9] The diffraction efficiency is measured by switching

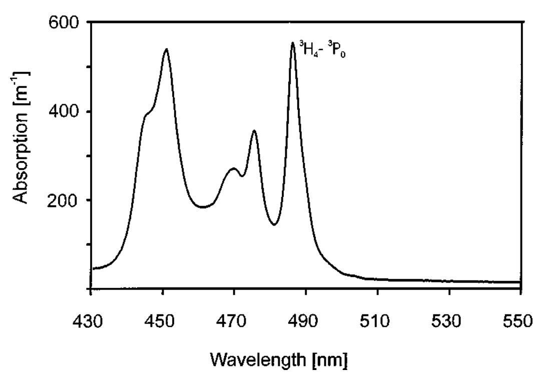

FIG. 1. Absorption spectrum of $La_3Ga_5SiO_{14}$:Pr^{3+} measured at room temperature. The characteristic absorption peak corresponds to interatomic excitations of the $4f^2$ multiplet of Pr^{3+} (Ref. 8).

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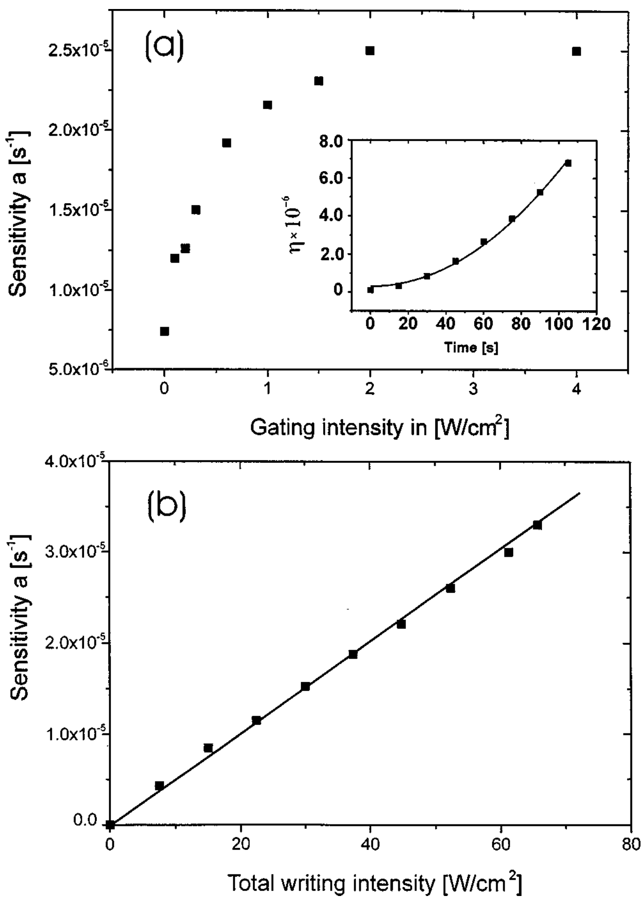

FIG. 2. Dependence of the sensitivity a on light intensity. (a) shows the dependence on grating light intensity with the total intensity of the writing beams ($\lambda = 810$ nm) kept constant at 60 W/cm^2 while (b) shows the dependence of a on total writing light intensity with grating intensity kept constant at 4 W/cm^2.

off one of the writing beams every 15 s for approximately 0.5 s and monitoring the transmitted and diffracted part of the second writing beam.

Using the Kogelnik formula[10] for diffraction and considering the space-charge field to follow a single exponential buildup we obtain in the small diffraction efficiency, short time limit approximation

$$\eta \approx a^2 t^2. \tag{1}$$

In order to obtain saturation values for the diffraction efficiency exposure times longer than 30 min are needed. Thus, buildup of the diffracted signal is monitored in the short time limit, here defined as the first 100 s of recording where a quadratic dependence on time is observed as seen from the inset on Fig. 2(a). The parameter a is used as a measure for the photorefractive sensitivity of the crystal.

In Fig. 2(a) measurements of the sensitivity a is shown as a function of gating light intensity. The wavelength of the writing beams is $\lambda = 810$ nm and the total writing intensity is held constant at 60 W/cm^2. Without the gating light a sensitivity of $a = 7.5 \times 10^{-6}$ s^{-1} is recorded, while at a gating light intensity of 2 W/cm^2 the sensitivity saturates at $a = 2.5 \times 10^{-5}$ s^{-1}. This saturation behavior is similar to that observed before for two-step recording in praseodymium doped $LiNbO_3$.[11] The dependence on the total writing beam intensity, with a constant gating beam intensity of 4 W/cm^2,

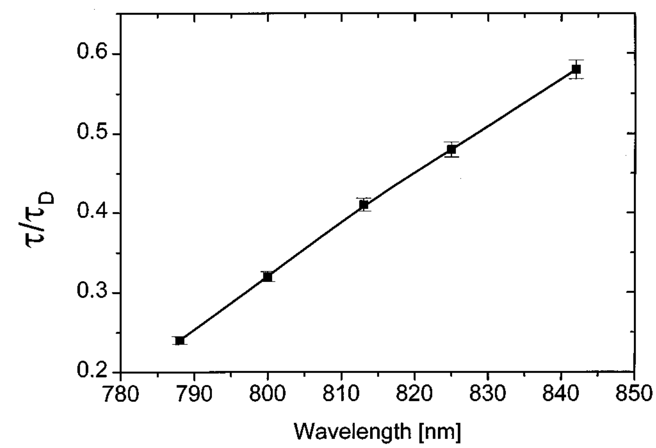

FIG. 3. Wavelength dependence of the ratio of the decay time constant τ to the dark decay time constant τ_D. The intensity of the beam used for measuring is 20 W/cm^2. The error bars indicate the uncertainty of approximately 2% on the fit of the data to a single exponential decay.

i.e., twice the saturation intensity, is shown in Fig. 2(b). The observed linear dependency suggest that the saturation behavior of the sensitivity is related to a saturation of the intermediate level acting in the two-step recording process.

Assume that the two-step excitation process goes through the $^3P_0$ excited state of Pr^{3+}. From the measured absorption coefficients, see Fig. 1 and density of Pr^{3+} ions the excitation cross section is estimated to be as a maximum of the order of 1.5×10^{-20} cm^2. An estimate of the lifetimes of the excited states of Pr^{3+} is obtained measuring the decay of the luminescence emitted from the crystal when illuminated by light with $\lambda = 488$ nm. These measurements reveals a lifetime in the order of 10 μs. Using these values suggests that intensities of the order of 10^5 W/cm^2 are needed in order to obtain saturation which is several orders of magnitude larger than what is observed experimentally. This indicates that the nature of the intermediate levels involved in the process is not excited states of Pr^{3+}. Determining the exact nature of these levels will be the subject of future work.

Saturation values of the diffraction efficiency are measured with and without gating. Using 4 W/cm^2 of gating intensity (488 nm) and a writing intensity of 60 W/cm^2 (788 nm) a diffraction efficiency of approximately 10^{-3} is obtained. In comparison the saturation value obtained without gating is one order of magnitude smaller. The nonzero sensitivity without gating light is assumed to be due to a weak excitation of electrons from the secondary photorefractive center.

The decay of the gratings is investigated under illumination with only one of the writing beams with an intensity of 20 W/cm^2. The decay of the gratings is monitored over a time period of 1 h. In order to compare the decay at different wavelength the data are fitted to an exponential law with a single decay time constant. In Fig. 3 the ratio of the observed decay time constants under readout τ to the dark decay time constant τ_D is plotted as a function of wavelength. Here the dark decay time constant $\tau_D = 8.2 \times 10^4$ s is measured by leaving the crystal in the dark for 1 min and then probing the grating for 1 s using one of the writing beams. The observed increase in decay time constant with increasing wavelength suggests that nonvolatile recording, limited only by the dark

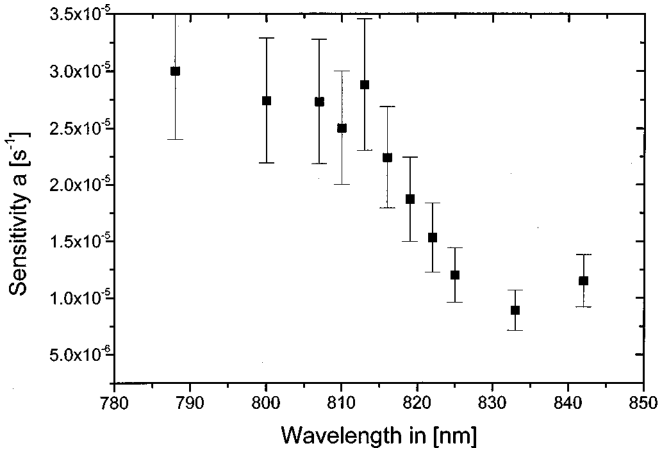

FIG. 4. Dependence of the sensitivity a on the wavelength of the writing beams. The holographic gratings are recorded using a gating light intensity of 4 W/cm^2 and a total writing intensity of 60 W/cm^2. The error bars on the plot indicate an estimated uncertainty of 20% of the measured values which primarily arises from strong sensitivity of the measurements to lineup of the narrow writing beams.

decay time constant, could be achieved by shifting the writing wavelength further into the infrared region. However, as seen in Fig. 4 a drastic decrease in the material sensitivity is observed around 815 nm making the material practically insensitive to gated recording.

The sensitivities reported here are several orders of magnitude lower than the values obtained with gated recording in, e.g., $LiNbO_3$.[11] However, no work has been performed yet to optimize the photorefractive properties of $La_3Ga_5SiO_{14}$. Furthermore, due to the photovoltaic nature of the crystal the sensitivity of the crystal might be increased shifting the wavelength of writing beams towards the visible away from the absorption threshold.

In conclusion, we have realized the first two-step two-color recording in praseodymium doped $La_3Ga_5SiO_{14}$ crystals using cw illumination. Saturation of the sensitivity is observed with increasing gating intensity, whereas a strictly linear dependence is observed for increasing total writing intensity. Diffraction efficiencies of 10^{-3} are obtained. From the dependence of the sensitivity to the wavelength of the writing beams it is found that a threshold photon energy of 1.53 eV (810 nm) is needed to excite electrons from the excited intermediate state to the conduction band. From the saturation behavior of sensitivity on gating intensity it is concluded that excited states of Pr^{3+} cannot provide the intermediate level involved in the recording process.

One of the authors (P.M.J.) acknowledges financial support from the Danish Natural Science Research Council, Grant Nos. 9502764 and 9600852.

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