Fabrication of Photorefractive grating With 800 nm Femtosecond Lasers in Fe: LiNbO₃ Crystal

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Abstract

Nonvolatile and optically reversible photorefractive volume phase hologram can be recorded in Fe: $LiNbO_3$ with 800 nm femtosecond pulse laser based on nonresonant and resonant two-photn absortion via impurity-to-band transition of photoexcited electrons

1. Introduction

Ferroelectric lithium niobate (LiNbO3) crystal is a wide bandgap(~4eV)optical material that is of prime importance for nonlinear optical and holographic applications such as frequency conversion, optical filtering, switching and data storage[1], phase conjugation and optical signal amplification[2].

High intensities and corresponding short pulses are required for efficient nonlinear optical processes and ultrafast holography[1]. LiNbO₃ has been well investigated in the continuous and nanosecond-picosecond ranges and recent years some investigations have been carried-out with femtosecond pulses[1,3,4,5,6,7] in the near infrared regimes.

In ferroelectric crystals, volume phase hologram grating is formed by the photorefractive effect. Illumination of such crystal by the spatially varying light pattern such as from the superposition of two mutually coherent plane waves results in a redistribution of electrons due to diffusion, drift and bulk photovoltaic effect which creates a space-charge field. This field modulates the refractive index of the photorefractive materials via the electro-optic effect and volume phase hologram grating is formed[2,6,8].

Hologram in Fe: LiNbO₃ crystal are generally recorded with blue or green light, as the photorefractive sensitivity is large in this wavelength range[6] by single photon absorption[9]. One of the problems of a volume holographic storage system based on photorefractive effect is that recording and readout require the same wavelength of light. As a consequence if hologram is recorded by the single photon absorption, the recorded information is erased during readout[10].

Thermal fixing and electrical fixing methods have been developed but in these cases optical erasure of the recorded information is not possible. For an all-optical random-accessmemory, optical reversibility has to be maintained[11].

This problem can be solved by recording with two-photon excitation processes namely (1) nonresonant (simultaneous) two-photon absorption and (2) resonant (two-step) two-photon absorption i.e. gated two-photon recording via impurity –to-band transition of the photo-excited electrons[12, 13].

Single photon hologram recording in Fe:LiNbO₃ with 800 nm pulse laser, to the best of our knowledge, has not been performed yet since Fe:LiNbO₃ is not sensitive for direct one-photon recording[8] with 800 nm.

Therefore, photorefractive volume phase grating can be recoded by the two-photon excitation processes with 800nm pulse laser.

2. Non-resonant two-photon absorption process in Fe: LiNbO₃

Fe;LiNbO₃ crystal is not sensitive to 800 nm laser but has good absorption coefficient at 400 nm wavelength. Therefore, simultaneous absorption of two photons of 800 nm which corresponds to the half-wavelength of the light, the crystal absorbs two photons of 800 nm at the 400 nm wavelength[7]. The principle of nonresonant two-photon recording can be explained by the one-center charge transport model.

Fig.1 shows the one-center charge transport model for Fe: LiNbO₃. In this model, only one center is responsible for electrons movement in the crystal. According to this model, the trapped electrons at the Fe²⁺ iron centers are excited into the conduction band by the simultaneous absorption of two photons of interfering beam of 800nm. The photoexcited electrons are retrapped in the empty Fe³⁺ iron centers after movement in the conduction band due to diffusion, drift and bulk photovoltaic effect. Finally they redistribute themselves and create a sinusoidal space-charge electric field which modulates the refractive index of the crystal via electro-optic effect and volume phase hologram is formed.

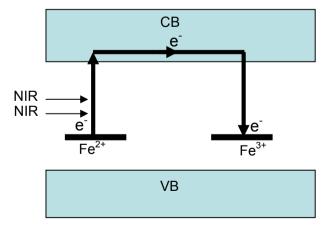


Fig. 1 One-center charge transport model. The arrow indicates excitation and recombination of electrons.

2.1 Experiment

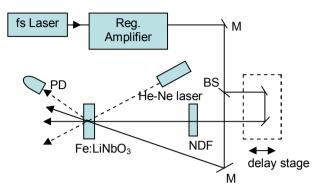


Fig. 2 Experimental setup for nonresonant two-photon hologram recording. BS: Beam Splitter, M: Mirror, NDF: Neutral density filter, Fe: LiNbO₃: Iron doped lithium niobate crystal, PD: Photo-detector.

Fig.2 schematically illustrates the recoding setup for simultaneous two-photon absorption hologram recording. Ordinarily polarized femtosecond laser pulses of 800 nm are generated from Ti: sapphire laser oscillator and then being amplified by the regenerative amplifier. The regenerative amplifier produces pulses of 50 fs duration. These pulses have energies of about 450µJ at a repetition rate of 1kHz with a peak power of 9GW. The beam splitter divides the pulses into two coherent beams. One of the beams passes through the delay line to intersect the other beam in the crystal. These two beams enter the crystal symmetrically in a plane containing the crystal's c-axis so that grating vector becomes parallel to the caxis. By changing the position of the delay stage, the optical path length can be made equal so that they can spatially overlap into the crystal. Readout can be performed by low intensity HeNe laser beam entering the crystal at the Bragg angle with extraordinary polarization. During illumination these two intense laser beams will interfere in the crystal and write the hologram. Readout can be performed by allowing only one of the writing beams of low intensity to enter the crystal which is not able to erase the recorded grating. Erasure can be performed by one of the writing beams having peak power equals to the hologram writing power.

3. Resonant two-photon absorption process in Fe: LiNbO₃

The principle of resonant two-photon absorption process in Fe: LiNbO₃ is explained by means of two-center charge transport model introduced by Jermann and Otten in 1993[14].

Fig.3 shows schematically the two-center charge transport model. According to this model, two photorefractive centers are considered. The iron level is deep and niobium level is called shallow levels[15]. Electrons can be excited from ${\rm Fe}^{2+}$ by the gating light directly either into the conduction band or into the Nb_{Li}^{5+} forming the Nb_{Li}^{4+} . The electrons in the shallow Nb_{Li}^{4+} traps can be excited to the conduction band by both the gating and near-IR light or thermally. The conduction

band electrons can recombine either with $\mathrm{Fe^{3+}}$ or with Nb_{Li}^{5+} . The gating (blue or green light) has sufficient[16] photon energy to excite electrons from $\mathrm{Fe^{2+}}$ centers directly either into the conduction band or into the shallow Nb_{Li}^{5+} or from Nb_{Li}^{4+} centers to the conduction band. Near-infrared (800nm) photons have insufficient energy to excite electrons from $\mathrm{Fe^{2+}}$ to the conduction band but have sufficient energy to excite electrons from Nb_{Li}^{4+} centers to the conduction band.

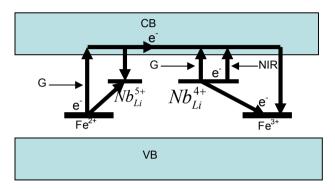


Fig. 3. Two-center charge transport model for Fe: LiNbO₃. The arrow indicates excitation and recombination of electrons.

By uniform illumination of the crystal with 400 nm gating pulse, shallow traps are filled and the crystal will be sensitized for subsequent hologram recording with 800 nm pulses which will excite electrons from Nb_{Ii}^{4+} traps to conduction band.

3.1 Experiment

Fig.4. schematically illustrates the recording setup for resonant two-photon hologram.

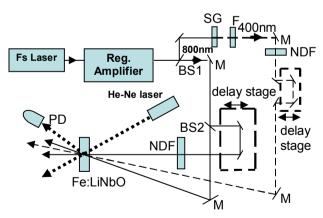


Fig.4. Experimental setup for resonant two-photon hologram recording. BS: Beam splitter, NDF: Neutral density filter, Fe: LiNbO₃: Iron doped lithium niobate crystal, PD: Photodetector, F: Colour filter, SG: Second harmonic generator, M: Mirror

The intense 800 nm fentosecond laser pulses of duration 50 fs having energies of about $450\mu J$ at a repetition rate of 1kHz with peak power of 9 GW are generated from regenerative

amplifier system. The beam splitter BS1 divides the intense 800 nm pulse into two coherent beams. One of these two beams passes through the second harmonic generator(SG) producing 400 nm pulses. The colour filter removes residual component of 800 nm from 400 nm. The second beam splitter BS2 again divides the 800 nm pulses into two coherent beams. These two beams enter crystal symmetrically in a plane containing the caxis of the crystal so that grating vector becomes parallel to the crystal's axis. The beam of 400 nm pulses enters the crystal before or simultaneously with the beams of 800 nm recording pulses.

NDF filters provide different peak intensities of writing and gating pulses. All pulse beams are ordinarily polarized. Hologram is readout by low intensity HeNe laser which enters the crystal at the Bragg angle having extraordinary polarization.

During the writing cycle, recording and gating pulses enter the crystal together and recording 800 nm pulses write the hologram grating.

Nondestructive readout can be performed only allowing one of the writing beams to expose to the crystal. Erasing of the hologram can be performed illuminating the crystal uniformly with the gating pulses.

4. Advantage of two-photon writing over one-photon writing in Fe:LiNbO₃ with 800 nm pulse laser

- (a) LiNbO₃ crystal is insensitive to 800 nm, therefore long recording time will be required for direct one-photon recording with 800 nm laser pulses.
- (b) In one-photon recording, readout results in unwanted erasure of recorded hologram.
- (c) Two-photon excitation processes can be applied to record hologram with 800 nm pulses in a short recording time and at the same time readout without erasure is possible using the pulses of reduced light intensities since the energy of one-photon is not sufficient to excite electrons to the conduction band. On the other hand optical erasure can be performed with the help of two-photon processes [13].

5. Conclusion

Using the advantage of high peak intensities of femtosecond laser pulses, femtosecond hologram can be recorded in $Fe:LiNbO_3$ by using two-photon nonresonant and resonant absorption processes. Both of these recording processes will be an extension of femtosecond holography in the near-infrared spectra with 800 nm pulse laser for nonvolatile and optically reversible multiphoton recording.

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