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Section 3. Femtosecond laser-matter interactions

Femtosecond-laser-encoded distributed-feedback color center laser in lithium fluoride single crystal

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Abstract

Focused infrared femtosecond laser pulses (wavelength \sim 800 nm, emission pulse duration 100 fs) were employed to fabricate optoelectronic devices such as waveguides, micro-gratings and laser active centers in LiF crystals. F₂ color centers of about 2×10^{18} cm⁻³ and refractive index change of about 1% at 633 nm were induced by the fs-laser irradiation. This technique was applied to fabricate a distributed-feedback (DFB) F₂ color center laser structure inside LiF single crystal. The LiF DFB laser exhibited laser oscillation at 707 nm at room temperature. The slope efficiency of \sim 10% and beam divergence of \sim 20 mrad were achieved. © 2006 Elsevier B.V. All rights reserved.

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1. Introduction

Appearance of a Ti:Al₂O₃ femtosecond laser system has stimulated remarkable advances in the laser machining field. Peak power intensity of a focused femtosecond pulse can easily exceed several hundreds TW/cm². Such intense laser pulse allows processing of any transparent dielectric material regardless of their optical bandgaps, and it is possible to machine surface and deep inside of the sample by adjusting the focusing depth. Many groups have reported unique applications of femtosecond laser processing by utilizing these features. For example, digging tiny holes, writing optical waveguides and producing micro cracks and pores inside glass have been reported [1–4]. There is still another important feature of femtosecond laser pulses. We have developed a 'holographic encoding technique by an interfered infrared femtosecond laser pulse' [5,6]. Femtosecond laser pulses generated from a mode-locked laser system have almost perfect temporal coherency due to Fourier-transform-limited pulse nature. This means that high contrast interference patterns may be formed if two coherent pulses are collided in a temporarily and spatially overlapped region. We succeeded in encoding holographic gratings on the surfaces and inside of various non-photosensitive and hard materials such as dopant-free SiO₂ glass, sapphire and diamond. This technique has been applied to fabricating more advanced structures at the nanometer scale such as cross gratings and two dimensional nano-hole array structures using a double exposure technique [7]. Our techniques provide fast methods applicable for producing the integrated optical circuits, which include many optical elementary devices such as lasers, optical waveguides and gratings.

Lithium fluoride (LiF) has best chemical durability among alkali halide crystals. In addition, the laser active color centers formed in LiF have high photothermal stability. For these reason, LiF has been examined for room temperature color center lasers. Especially, F_2 and F_3^+ centers

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in LiF are promising candidates for the visible laser action. Recently, Montecchi reported increase in the refractive index due to the formation of the color centers and fabrication of active channel waveguides [8]. It was expected that grating structures and mirrors were also produced in the same way, and therefore LiF is promising for a host material to integrate optical circuits. However, high-energy deposition techniques such as ion implantation, electron beam and γ -ray irradiation are generally needed for producing the color centers in LiF because of its large optical bandgap energy ~14 eV. It is difficult for these conventional ionization techniques to machine fine patterns inside materials.

In this paper, we demonstrated that an active optical device and a distributed-feedback (DFB) F_2 color-center laser were fabricated by the holographic encoding method using a femtosecond laser pulse.

2. Results and discussion

Regenerative amplified femtosecond pulses from a mode-locked Ti:Al₂O₃ laser system (wavelength: 800 nm, pulse duration: 100 fs, repetition: 10 Hz) were used in this experiment. Polished LiF crystals with thickness of ~1.5 mm containing Ca (7 wt ppm), Fe (1 wt ppm) and Mg (1 wt ppm) impurities were employed as samples. To examine the formation of the color centers, femtosecond pulses (\sim 70 µJ/pulse) focused with a convex lens (f = 60 mm) were irradiated to the surface of the sample. The sample was moved using a computer-controlled stage synchronized with the laser pulse repetition rate. Each point was irradiated by 50 pulses. Fig. 1(a) shows the induced optical absorption spectrum that was obtained by subtracting the spectrum before irradiation from that after irradiation. Two distinct absorption bands are observed at 4.9 and 2.8 eV. The 4.9 eV bands originate from the F centers: i.e., an electron trapped in a fluorine vacancy. The 2.8 eV band is associated with the F_2 and F_3^+ centers. The concentration of the F_2 color center estimated was $2 \times 10^{18} \text{ cm}^{-3}$ using a Smakula's expression [9] and a reported value of the oscillator strength [10]. The luminescence spectrum of the sample irradiated with femtosecond laser pulses is shown in



Fig. 1. (a) Optical absorption spectrum and (b) luminescence spectrum of LiF crystal irradiated with 100 fs laser pulses (\sim 70 µJ/Pulse) at room temperature. The absorption spectrum was obtained by subtracting the spectrum before irradiation from that after irradiation.

Fig. 1(b). Two intense broad emission bands due to F_2 and F_3^+ centers are observed at 650 nm (red luminescence) and 530 nm (green luminescence), respectively. These results show that near infrared femtosecond laser pulses (hv = 1.5 eV) can be used to ionize LiF and produce the color centers instead of the other conventional energetic beams.

Using the color centers induced by femtosecond laser pulses, we produced optical waveguides deep in an LiF crystal. Fig. 2(a) shows a photograph of the embedded optical waveguides and (b) its magnified optical microscope image. The waveguides were written by line-shaped laser pulses with a cylindrical lens (f = 50 mm), and the



Fig. 2. (a) F_2 -center luminescence image of embedded optical waveguides under 450 nm light excitation and (b) an optical microscope image of optical waveguides embedded by femtosecond pulses.

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