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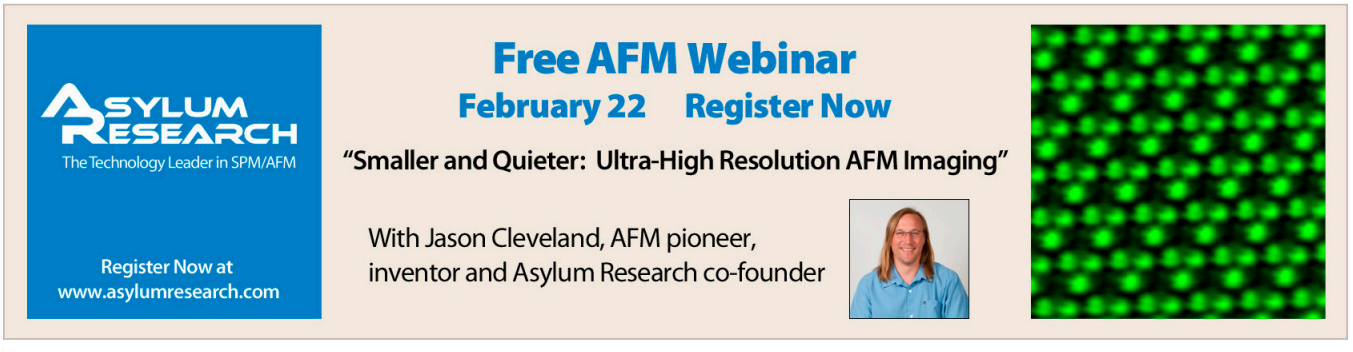

Growth of Nd^{3+}-doped $LiNbO_3$ optical superlattice crystals and its potential applications in self-frequency doubling

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We have grown Nd:MgO:$LiNbO_3$ crystals with periodic ferroelectric domain structures. Absorption and fluorescence spectra measured on these crystals showed little difference from those from Nd:MgO:$LibO_3$ with uniform domain structures. Green fluorescence was generated by self-frequency doubling in a cavity having great losses and pumped by a pulsed dye laser. © *1996 American Institute of Physics.* [S0003-6951(96)02611-4]

Self-frequency doubling (SFD) is one of the most important techniques for constructing compact and reliable short wavelength light sources. The combination of laser oscillation of active ions with nonlinear optical properties of the host material offers an opportunity for producing self-frequency doubling, self-modulated, and self-*Q*-switched lasers as well as miniature waveguide lasers and amplifiers.[1] $Nd_xY_{1-x}Al_3(BO_3)^4$ (NYAB) has been shown to be a promising material for SFD, but difficulties in crystal growth make it unfavorable for practical applications. Nd laser oscillation in $LiNbO_3$ was achieved in 1967, and self-frequency doubling was achieved in 1979.[3,4] $LiNbO_3$ is uniaxially negative. Therefore Nd ion produces radiation polarized perpendicular (σ) and parallel (π) to the *c* axis. The π-polarized (σ-polarized) output occurs at 1084 nm (1092 nm) and is the high (low) gain output.[1] Two problems were encountered in the SFD of Nd:$LiNbO_3$; photorefractive damage and high cavity losses associated with the difficulty in producing low-gain σ-polarized emission required for type I phase matching.[3,5] One can minimize photorefractive damage by maintaining the crystal at elevated temperatures, codoping with MgO, and increasing the wavelength of the pump source from the visible (≈600 nm) to the near IR (≈810 nm). Codoping with MgO reduces the photrefractive damage, but stable cw oscillation in Nd:MgO:$LiNbO_3$ requires operation above 100 °C.[1,5] Cavity losses are increased, because a Brewster window is needed to force the low-gain σ-polarized output to satisfy the noncritical phase-matching conditions. A reduction of operation temperature has been achieved in a Nd:Sc_2O_3:$LiNbO_3$ crystal recently. But the critical requirements for temperature stabilization and type I phase matching condition make the SFD laser difficult to miniaturize.[6] In other aspects, all SFD materials mentioned above can only be used to generate green light. One cannot extend the output wavelength to the blue range. Recent interest in all-solid-state blue and green lasers for optical storage applications has intensified the search for new frequency doubling materials, especially those materials which have high nonlinearity, good laser operation at room temperature, and the ability to generate blue light.

Second-harmonic generation (SHG) in quasiphase matched (QPM)[7] in $LiNbO_3$[8–12] and $LiTaO_3$[13] crystal has recently attracted a great deal of attention. In $LiNbO_3$, by modulating the nonlinear susceptibility with periodic ferroelectric domain structures which have an appropriate period ($LiNbO_3$ optical superlattice), it is possible to quasiphase match at an arbitrary temperature over the crystal's entire transparency range and to use the highest nonlinear coefficient d_{33}. So the combination of laser oscillation and SFD through quasiphase matching technique in rare-earth-doped $LiNbO_3$ crystals with periodic ferroelectric domain structures is possible. It has the advantages of high nonlinearity, easy room temperature quasiphase matching (only π-polarization of the fundamental and harmonic light is required in the process of quasiphase matching), and the ability for generating light from blue (corresponding to SFD of 946 nm line of Nd^{3+} ion) to near infrared (corresponding to SFD of 1.53 μm line of Er^{3+}, for example).

In this letter, we report the growth of Nd_2O_3 and MgO codoped $LiNbO_3$ crystals with periodic ferroelectric domain structures. The absorption spectra and fluorescence spectra were recorded and compared with that of Nd:MgO:$LiNbO_3$ with uniform domain structures. SFD phenomenon were observed in a Nd:MgO:$LiNbO_3$ optical superlattice (OSL) crystal.

The single crystals were grown by the conventional Czochralski method using a Pt crucible in a carefully designed asymmetric temperature system.[9–12] Starting materials were prepared by mixing Nd_2O_3 and MgO with a congruent melt of $LiNbO_3$. Pulling and rotating rates were 3 mm/h and 11 rpm, respectively. The Nd_2O_3 concentration was 0.2 wt % in the crystal and the MgO concentration was 4.5 mol %. The as-grown *a*-axis crystals were transparent and bluish in color. The periodic domain structures were revealed by acid etching the crystal's polished *b* face. The continuous modulation period number reaches over 700, in which over 300 periods are invariable (period fluctuation was kept below 2%). The modulation period measured by an optical microscope is 7.0 μm, as shown in Fig. 1. Sample was cut out parallel to the laminar domains. The sample size is about $3\times6\times2$ ($b\times c\times a$) mm^3. The light-transmitting length (*a*-axis direction) is 2 mm. The sample includes about 570 domain laminas.

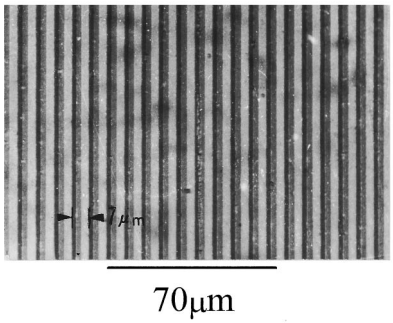

FIG. 1. Photograph of a Nd:MgO:$LiNbO_3$ optical superlattice with a modulation period of 7.0 μm.

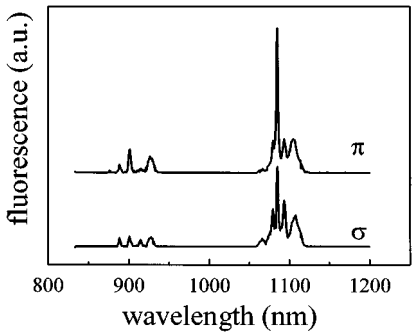

FIG. 3. Fluorescence spectra (σ and π polarized) of a Nd:MgO:$LiNbO_3$ optical superlattice.

Absorption spectra under σ ($E \perp c$) and ($E \| c$) polarized incident light are shown in Fig. 2. In the 750 nm region, four absorption peaks were observed in both the σ and the π polarization cases, though the relative peak intensity is slightly different. The peak position is almost the same. These four peaks are at 743, 752, 757, and 766 nm. From the spectra, only one peak exists in 810 nm region in σ polarization and the maximal absorption is at 814 nm. Whereas in the case of π polarization, there are four peaks in the same region. They are at 805, 809, 814, and 824.6 nm. The maximal absorption is at 809 nm. The absorption spectra of a Nd:MgO:$LiNbO_3$ OSL were similar to those of Nd:MgO:$LiNbO_3$ with a uniform domain structure. It indicates little effect of the periodic domain structures on the Nd absorption characteristics.

Fluorescence spectra were measured by using tunable cw Ti:sapphire laser excitation near the peak absorption. Figure 3 shows the π and σ polarized components of the radiation from the sample obtained with a polarizer. The pumping beam was σ polarized (perpendicular to the crystal's c-axis). The spectral resolution was 0.5 nm in this measurement. The two maximal fluorescence peaks of the π component occurred at 940 and 1084 nm, which is almost the same as that in Nd:MgO:$LiNbO_3$ with a uniform domain structure. In the case of the σ component, the two main absorption peaks also occurred at 940 and 1084 nm. This differs from that in Nd:MgO:$LiNbO_3$ with a uniform domain structure, in which the latter main absorption peak is at 1094 nm as shown in Fig. 4. The 1094 nm peak is clearly seen in Fig. 3, but the peak is not at its highest. This is probably caused by the existence of periodic domain structures, but the reason it still unknown. Both these two lines, 940 and 1084 nm, corresponds to a transition of the Nd ion from state $^4F_{3/2}$ to $^4I_{9/2}$ and from state $^4F_{3/2}$ to $^4I_{11/2}$, respectively.

For characterizing the periodicity of the Nd:MgO:$LiNbO_3$ optical superlattice sample, second-harmonic generation through using third-order QPM technique in the sample has been performed by using a dye laser pumped by a tunable Nd:YAG laser as the fundamental source. The laser has a repetition rate of 10 Hz with a pulsewidth of 7 ns and a linewidth of 0.2 cm^{-1}. The maximal SHG efficiency, which is over 15%, occurred at the fundamental wavelength of about 788 nm. The input energy is about 5 mJ/pulse, and no focusing was used. The full width at half-maximum (FWHM) is less than 0.3 nm. It shows that the periodicity of the sample is very good.

Using our uncoated sample and with no cavity used, no phenomenon could be observed under 809 nm pumping of the above laser. When a cavity was used and with an input pulse energy over 50 mJ/pulse and no focusing, green fluorescence was generated in the sample, but there was no observable green laser output from the output mirror. This green fluorescence may show that some weak laser oscillation of 1084 nm existed in the cavity and then self-frequency doubled by the optical superlattice through QPM. The cavity consisted of an input plate mirror which has high reflectivity at 1084 nm and high transmitance at 809 nm, an output mirror with a radius of 50 mm which is highly reflective for 1084 nm and highly transmitive for 809 nm. The uncoated sample introduced a great loss in the cavity. Thus the difficulties in laser oscillation may be caused by the large loss.

In summary, we have grown a Nd:MgO:$LiNbO_3$ OSL

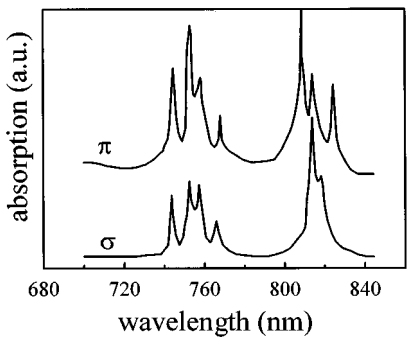

FIG. 2. Absorption spectra (σ and π polarized) of a Nd:MgO:$LiNbO_3$ optical superlattice.

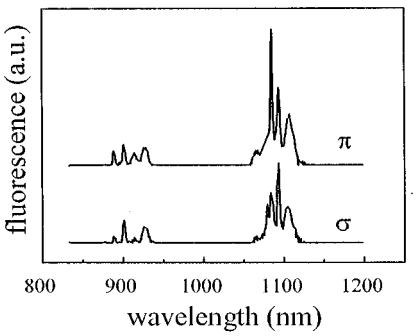

FIG. 4. Fluorescence spectra (σ and π polarized) of a Nd:MgO:$LiNbO_3$ with a uniform domain structure.

with a modulating period of 7.0 μm. Absorption and fluorescence spectra of the crystal were measured, and the results from the crystal show little difference from those from a Nd:MgO:$LiNbO_3$ crystal with a uniform domain structure. SFD green fluorescence has been observed by using a pulsed laser as pumping source in a cavity with great loss.

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