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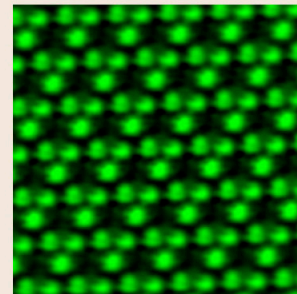
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Growth of optical superlattice LiNbO₃ with different modulating periods and its applications in second-harmonic generation

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Optical superlattice LiNbO₃ crystals with a modulation period number over 200 and a modulation period from 2.0 to over 15.0 μm were grown by the Czochralski method in a carefully designed asymmetric temperature field system. Measurements of the frequency doubling efficiency for generating light from blue to green were performed. The largest efficiency obtained was 24.0% for frequency doubling of a picosecond 980 nm fundamental light. The angle tuning tolerance of a LiNbO₃ optical superlattice was measured to be over 10° of the incidence angle. © 1996 American Institute of Physics. [S0003-6951(96)04320-3]

Quasiphase matching (QPM) was proposed independently by Bloembergen¹ and Franken and Ward² for obtaining efficient frequency conversion in optical second-harmonic generation (SHG) and other nonlinear optical processes. SHG by QPM in a LiNbO₃ optical superlattice (OSL), in both bulk³⁻⁵ and waveguide forms,⁶⁻⁸ has attracted a great deal of attention for its potential applications in constructing compact short-wavelength lasers, and in constructing a QPM optical parametric oscillator (OPO).⁹ A LiNbO₃ OSL in bulk form offers the following advantages: direct frequency doubling of a laser diode without the need for careful light coupling which exists in waveguide devices, and ease of phase matching and angle tuning in QPM OPO. The latter is difficult to achieve in waveguide form. Several methods have been developed for obtaining a bulk LiNbO₃ OSL. Alternating stacks of thin plates of LiNbO₃ were constructed for QPM SHG experiments.¹⁰ Single-crystal fibers with a diameter of 250 μm and having periodically alternating ferroelectric domain structures have been applied to QPM SHG.¹¹ With the development of integrated-optics technology in LiNbO₃,^{7,8,12,13} some field-induced methods present the potential for preparation of a bulk LiNbO₃ OSL.^{9,14,15} Using an electric-field-poling method, a LiNbO₃ OSL with a domain-inversed depth of 0.5 mm has been successfully prepared and has been performed in QPM SHG or OPO.⁹ However, the limitation in the surface area for light-transmitting electric-field-poled samples makes angle tuning difficult in QPM OPO applications. It is also difficult to prepare a LiNbO₃ OSL with a small modulating period, which is desirable for SHG to generate short wavelength light using first-order QPM, by field-induced methods. Compared with field-induced methods, the Czochralski method for bulk LiNbO₃ growth^{3-5,16} grows a LiNbO₃ OSL with a practical dimension, with a modulating period as small as 2.0 μm , and with its period controllable by adjusting growing parameters. However, there are some disadvantages to this method, including the formation of various islandlike domains during growth and period variation along the growing direction. The

islandlike domains occur easily during crystal growth and destroy the periodicity of the LiNbO₃ OSL. The growth of a LiNbO₃ OSL with a period below 4.0 μm by the Czochralski method is also relatively difficult due to the troublesome response of periodic ferroelectric domains to periodic temperature fluctuations during crystal growth.

In this letter, with a carefully designed temperature field system, we report the success in growing a LiNbO₃ OSL with a modulation period ranging from 2.0 to over 15.0 μm . The formation of various islandlike domains was kept at its lowest level. The period fluctuation along the growing direction was limited to 5%. The continuous period number of the LiNbO₃ OSL with a stable modulation period (period fluctuation of less than 5%) can be over 200 when the modulation period is less than about 4.0 μm and easily over 300 when the period is larger than about 4.0 μm . Efficient SHG to generate light from blue to green has been performed with the usage of an OPO as a fundamental source.

Crystals of LiNbO₃ doped with 0.5 mol % yttrium were grown along the crystal's *a* axis by the Czochralski method.³⁻⁵ In designing the asymmetric temperature field system, three parameters—axial temperature gradient above the melt surface, axial temperature gradient below the melt surface, and the radial temperature gradient in the melt surface—were defined as adjusting parameters. In our growing system, the axial temperature gradients above and below the melt surface were kept in the range of 30–40 °C/cm and 6–8 °C/cm, respectively, when the melt depth was about 30 mm in a 40 mm high platinum crucible. The formation of various islandlike domains, which are caused by solute aggregations in solute boundary layers due to solid–liquid interface instabilities, can be effectively suppressed by adjusting these two axial temperature gradients. The measurement of radial temperature gradient is difficult since the temperature system is asymmetric. But the gradient can be adjusted by changing the thickness of the ZrO₂ thermal-insulating layer outside the Pt crucible. Careful adjustment of this thickness can obviously change the shape of the solid–liquid

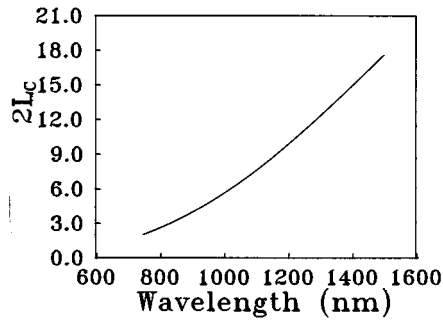


FIG. 1. The relationship between fundamental wavelength and double coherence length of the LiNbO₃ crystal.

interface, and thus affect the response depth of periodic ferroelectric domain structures in the *a*-axis as-grown LiNbO₃ OSL. The response depth is defined as the width of periodic domain structures along the crystal's *z* axis on one side of the main *x*-*z* plane in an *a*-axis grown crystal. One of the most important factors determining this response depth is the shape of the solid-liquid interface. If the shape of the solid-liquid is exactly plane (i.e., exactly in the crystal's *y*-*z* plane), the periodic domain structures cannot occur due to the fact that the polarization is along the crystal's *z* axis. A convex solid-liquid interface is beneficial for generating periodic domain structures. By carefully adjusting these three parameters, we can grow a LiNbO₃ OSL with a modulating period from 2.0 to over 15.0 μm. The difficulty in LiNbO₃ OSL growth will increase as the modulation period decreases. For a modulation period below about 3.0 μm, which is near the limiting response frequency of periodic domain structures to the temperature fluctuations, the periodic response of the ferroelectric domain not only relies on the properties of the material itself and the temperature system, but also depends on the stability of the pulling and rotating systems of the crystal growth unit.

The relationship between the fundamental wavelength in the range of 0.75–1.5 μm and the double coherence length of LiNbO₃, which was calculated from the crystal's Sellmeyer's equation, is shown in Fig. 1, where the fundamental light propagates along the crystal's *a* axis with its polarization along the crystal's optical axis. In this wavelength range of fundamental light, the most practical fundamental sources for constructing an all-solid short-wavelength laser and a QPM OPO are a Nd:YAG or Nd:VO₄ laser at 1.064 μm and a near-infrared laser diode with an output in the range from 800 to 860 nm or from 950 to 980 nm. Thus it requires that the LiNbO₃ OSL has a modulation period in the range from about 2.7 to 6.4 μm. For constructing a QPM OPO with 1.064 μm pumping, it is also desirable that the LiNbO₃ OSL has a modulation period of over 6.4 μm. Figure 2 shows the photographs of a LiNbO₃ OSL with the following modulation periods: (a) 2.7 μm, for frequency doubling of an 809 nm laser diode, (b) 5.2 μm, for frequency doubling of a 980 nm laser diode, and (c) 15.0 μm, which will be used in the QPM OPO. These photographs were taken on the crystal's acid-etched *y* surface. The thickness of the positive domain laminae and that of the negative domain laminae are nearly equal, and the continuity of modulation period and periodicity is very good. SHG experiments were performed by using

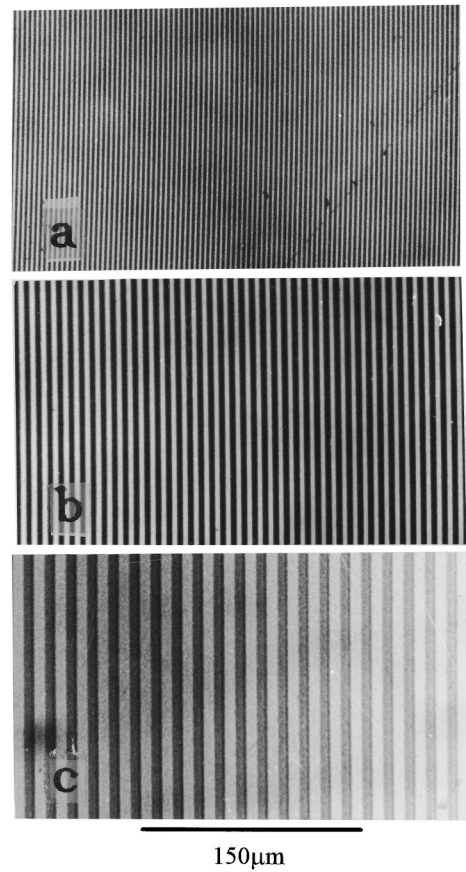


FIG. 2. Photographs of a LiNbO₃ OSL with modulation period: (a) 2.7 μm, (b) 5.2 μm, (c) 15.0 μm.

a picosecond automatic tunable OPO as a fundamental light. The laser has a pulse rate of 1 Hz and a pulse duration of 30 ps. The linewidth of the output light pulse is less than 1 nm at all wavelengths except that at the OPO's degeneracy point of 1064 nm, when the linewidth is about 10 nm. Table I lists the results of SHG efficiency and some parameters of five measured samples with different modulation periods. The SHG efficiencies listed in Table I are difficult to compare, because SHG efficiency is mainly relevant to fundamental power density, fundamental wavelength, modulation period number, and period stability. These factors are different in these five samples. For comparison, we used a commercial 10-mm-long single-domain LiNbO₃ crystal to measure SHG efficiency under similar experimental conditions (the fundamental wavelength is 1064 nm). A SHG efficiency of 18.5% has been obtained under 90° phase-matching conditions. The result shows that sample 4 in Table I (the fundamental wavelength is 1026 nm) has a SHG enhancement of about 18.9.¹⁷

TABLE I. Parameters of LiNbO₃ OSL samples and results of SHG experiments by using an OPO as a fundamental source.

Sample no.	d_{thick} (mm)	Period (μm)	N_{period}	Fluctuation (≅%)	λ_{fun} (nm)	η_{SHG} (%)
1	0.62	2.8	220	5	815	3.0
2	0.78	3.4	230	5	860	4.2
3	1.56	5.2	300	2	980	24.0
4	2.20	6.4	310	5	1026	17.0
5	1.50	8.3	180	2	1130	19.8

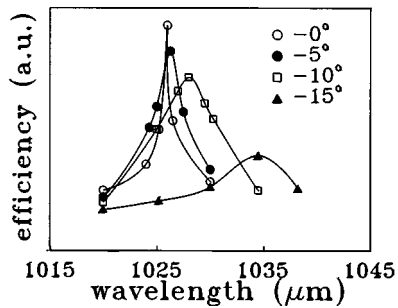


FIG. 3. SHG efficiency vs fundamental wavelength of sample 4 at different incidence angles: (○) 0°, (●) 5°, (□) 10°, (▲) 15°.

It is near the theoretical SHG enhancement of $(d_{33}/d_{31})^2(2/\pi)^2 \approx 23$. The SHG enhancement defined here means that the efficiency comparison is between two crystals (OSL LiNbO₃ and single-domain LiNbO₃) with the same crystal length and the same experimental conditions.

For measuring the LiNbO₃ OSL's angle tunability, we have measured the curve of SHG efficiency with fundamental wavelength at different incidence angles of sample 4 in Table I. The sample was rotated in the crystal $x-y$ plane. The rotation angle is 0° when at normal incidence, then is 5°, 10°, and 15°, respectively. These curves are shown in Fig. 3. The peak SHG wavelength was found to increase as the incidence angle was increased, in agreement with the notion that the effective period will increase when the incidence angle increases. However, the peak SHG efficiency decreases and the full width at half-maximum increases with the incidence angle, due to the increase of reflection of fundamental light at the sample's surface and the relative increase of period fluctuation. The peak SHG efficiency decreases sharply when the incidence angle exceeds 10°. The

results show that nearly 10° of the incidence angle tuning tolerance can be achieved in this uncoated sample, without the evident decrease of SHG efficiency. This result is useful for tuning in LiNbO₃ OSL's applications in SHG or in QPM OPO.

In conclusion, we have grown a LiNbO₃ OSL with a modulation period from 2.0 to over 15.0 μm. Frequency doubling efficiencies for generating blue to green light were measured by using an OPO as a fundamental source. The largest SHG efficiency for doubling 980 nm is 24.0%. The LiNbO₃ OSL's angle tuning tolerance was measured to be over 10° of the incidence angle.

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