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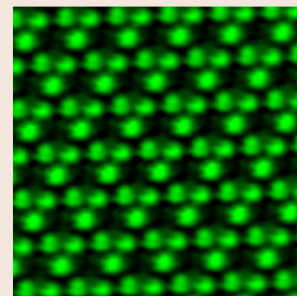
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# Visible dual-wavelength light generation in optical superlattice Er:LiNbO<sub>3</sub> through upconversion and quasi-phase-matched frequency doubling

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Optical superlattice Er:LiNbO<sub>3</sub> was fabricated by inducing a periodic ferroelectric domain structure into the crystal during the growing process. Because of the combination of the nonlinear optical properties of LiNbO<sub>3</sub> and the spectral properties of Er<sup>3+</sup>, the crystal can simultaneously emit the second harmonic light through quasi-phase matching and the green light through upconversion at room temperature. Pumped by infrared diode lasers, violet-and-green and blue-and-green light generation was demonstrated in two samples. The detailed absorption spectrum and emission spectrum of upconversion were measured. The possible physical mechanism was discussed.

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Upconversion phenomena are being exploited for the development of short-wavelength solid state lasers, which have many technical applications including data storage, laser printing, underwater communications, and full color laser display. Erbium doped fibers were used to demonstrate the physical mechanism of two-photon as well as excited-state absorption under near-infrared diode laser excitation.<sup>1-3</sup> Room temperature green upconversion lasers were developed by pumping erbium-doped fibers with infrared lasers.<sup>4-6</sup> As for Er<sup>3+</sup> doped bulk crystals, visible upconversion laser oscillation was also achieved at cryogenic temperatures.<sup>7</sup> On the other hand, the multi-wavelength laser has drawn much attention in recent years because of its applications in precise laser spectrum, laser radar, and nonlinear frequency conversion. Up to date, dual-wavelength laser oscillation has been demonstrated in several crystals including Nd:YAG<sup>8-10</sup> and Nd:YAP.<sup>11-13</sup> However, because of the difficulty of selecting the suitable laser crystal and designing the cavity mirrors so that the two spectral lines have the same oscillation thresholds,<sup>12</sup> it is not easy to obtain the infrared dual-wavelength oscillation at the same time, let alone the visible dual-wavelength laser operation by frequency doubling.

In this letter, we first report on the fabrication of optical superlattice Er:LiNbO<sub>3</sub> (OSL ELN) which is able to simultaneously generate violet-and-green or blue-and-green light pumped by an infrared diode laser at room temperature by means of second-harmonic generation (SHG) and upconversion. The absorption and upconversion spectrums were measured and the phenomena of dual-wavelength light generation were demonstrated.

The optical superlattice (OSL) is a kind of man-made

crystal, with its physical properties being modulated periodically in the span of several microns to several tens of microns, that is comparable with light wavelength. Because of the periodic structure, the OSL will exhibit some novel characteristics which cannot be found in ordinary materials.<sup>14</sup> For example, if the spontaneous polarization of LiNbO<sub>3</sub> (LN) is modulated periodically, the nonlinear optical coefficient will change its sign from the positive domains to the negative domains, so that the quasi-phase matching (QPM) technique can be achieved to compensate the phase velocity dispersion in frequency conversion applications.<sup>15</sup> A significant advantage of QPM is that any interaction within the transparency range can be noncritically phase matched at a specific temperature, even interactions for which birefringence phase matching is impossible. Another benefit is that the interacting waves can be chosen so that coupling occurs through the largest element of the  $\chi^{(2)}$  tensor. In LN, QPM with all waves polarized parallel to the *c* axis yields a gain enhancement over the birefringence phase matched process of  $(2d_{33}/\pi d_{31})^2 \approx 20$ . In our laboratory, since the first demonstration of fabricating OSL LN by the Czochralski method in 1980,<sup>16</sup> we have grown various OSL LN crystals with different dopants and different modulation periods.<sup>17,18</sup> Up to date, CW, picosecond, and femtosecond visible light QPM SHG were obtained.<sup>19-21</sup> However, if we doped some erbium ions into the OSL LN, the crystal will combine the nonlinear optical properties of LN and the spectral properties of Er<sup>3+</sup>. Pumped by adapt infrared light, the crystal is able to emit the upconversion light as well as the second harmonic light at the same time. Since the physical processes of SHG and upconversion are different and SHG has no pumping threshold, the complicated design of the cavity mirrors for obtaining ordinary dual-wavelength output becomes unnecessary. Thus it will be easier to obtain the visible dual-wavelength laser oscillation at the same time. Obviously, such a novel

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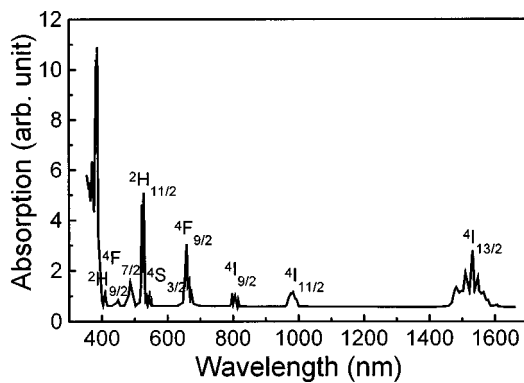


FIG. 1. Absorption spectrum of 0.5%  $\text{Er}^{3+}$  doped optical superlattice  $\text{LiNbO}_3$  measured at 300 K. The corresponding energy levels of major absorption peaks were marked in the figure.

device will have many applications especially in the laser display area.

Our sample for SHG and spectral measurements is a Czochralski grown  $4\text{ mm} \times 4\text{ mm} \times 4\text{ mm}$  cubic OSL LN doped with 0.5 mol %  $\text{Er}_2\text{O}_3$ . The average modulation period of the sample is  $8.2\ \mu\text{m}$  just corresponding to the 3rd-order QPM frequency doubling of 808 nm fundamental light. The two  $a$  faces of the sample were finely polished for light transmission.

Figure 1 shows the absorption spectrum of the OSL ELN sample measured at 300 K in the range of 300–1700 nm. The transmission rates of 808 nm light and 404 nm light are 88% and 90%, respectively. The spectrum appears in a sharp linelike structure, indicating that the spectral character of  $\text{Er}^{3+}$  ions in LN crystal is different from that of  $\text{Er}^{3+}$  doped glass fibers, in which only some peak envelopes can be observed.<sup>21</sup> In comparison with the spectra of  $\text{Er}^{3+}$  in YAG,<sup>22,23</sup> there are similarities in spectrum structure but dissimilarities in peak positions and peak intensities, indicating the effect of a different crystal field.

The diagram of the experimental setup for SHG and upconversion measurements is shown in Fig. 2. The output of an 808 nm GaAlAs diode laser was collimated and focused onto the  $a$  face of the sample. An infrared-cut filter was put behind the sample to block the transmitted fundamental light and pass the second harmonic light into the powermeter. A monochromator at the side of the sample was used to analyze the spectrum and a photomultiplier was used to receive the emitted light from the slit of the monochromator.

In our experiments, when the 808 nm light injected into the sample, remarkable green fluorescence light and violet 404 nm violet second harmonic light were observed even with an incident power lower than 200 mW. With the in-

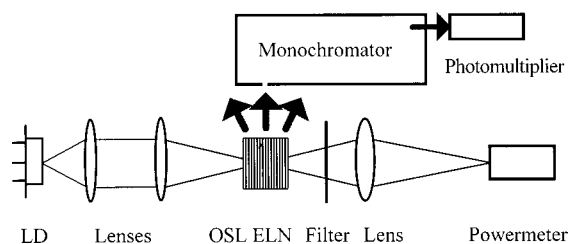


FIG. 2. Experimental setup for measuring the upconversion spectrum and second harmonic generation in optical superlattice  $\text{Er}:\text{LiNbO}_3$ .

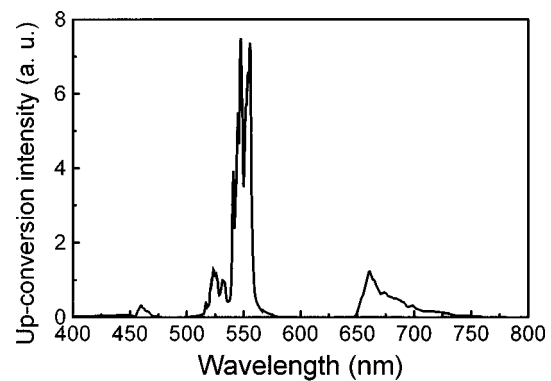


FIG. 3. Emission spectrum of upconversion pumped by an 808 nm GaAlAs diode laser.

crease of the output of the diode laser, the brightness of the two lights increased subsequently. When the input infrared power was 1.0 W, we got 0.8 mW stable violet SHG. The low conversion efficiency (0.08%) maybe due to several factors: for example the period fluctuation, the unpolarized incident infrared light, the 3rd-order QPM, the absorption of the infrared light, and the uncoated transmitted faces. Among them we believe that the period fluctuation is the most severe. In addition, the designed modulation period was calculated according to the refractive indices of the pure LN, not the doped  $\text{Er}:\text{LN}$ . Under the above condition, the fluorescence spectrum of upconversion was recorded as displayed in Fig. 3. It shows that the OSL ELN really possesses strong upconversion emission, at least for the green light of wavelength 500–570 nm. There are two twin-peak structures at this band, one at 547 nm due to the  ${}^4S_{3/2}-{}^4I_{15/2}$  transition, the other at 523 nm due to the  ${}^2H_{11/2}-{}^4I_{15/2}$  transition. The twin-peak structures of these two spectral lines are perhaps contributed by the perturbation of energy levels due to the small difference of crystal fields, since there are two different sites in LN crystal lattice, i.e., the Li site and the Nb site, which can be replaced by  $\text{Er}^{3+}$ . In addition to the green light, the OSL ELN also emitted blue and red fluorescences of upconversion excitation. The central peaks are at 460 and 661 nm which correspond to transitions between the states of  ${}^4G_{9/2}-{}^4I_{13/2}$  and  ${}^4F_{9/2}-{}^4I_{15/2}$ , respectively. Among the four peaks, the 547 nm peak is the most intensive one. It is approximately six times larger than the 661 nm peak and 20 times larger than the 460 nm peak. That is why only green fluorescence can be observed with the naked eye.

For analyzing the physical process of upconversion, it is necessary to realize the relationship between the pumping power and the fluorescence intensity of upconversion. Figure 4 shows the variation of the 547 nm peak intensity as a function of the 808 nm laser power. Just as expected, the fluorescence intensity exhibits a square dependence on the pumping power indicating that the generation of green light is really a two-photon process. The possible detailed transition process may be as follows: Pumped by the 808 nm infrared light, the  $\text{Er}^{3+}$  is excited from the ground state to the  ${}^4F_{9/2}$  state. Since the lifetime is sufficiently long, some of the ions in the  ${}^4F_{9/2}$  level may absorb another 808 nm photon to populate the higher level  ${}^2H_{9/2}$ , which relaxes to the  ${}^4S_{3/2}$  state, causing it to emit the 547 nm green light. Obviously, the above explanation is based on the mechanism of excited-

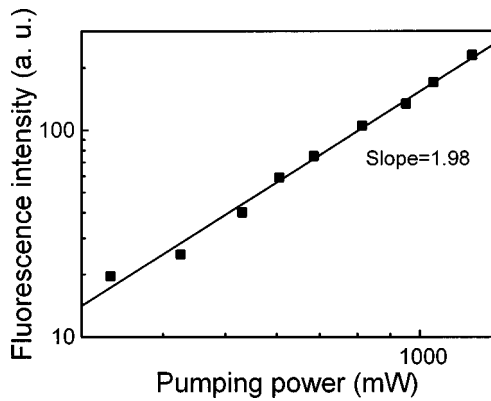


FIG. 4. The intensity of emitted 547 nm green fluorescence of upconversion as a function of the pumped power.

state absorption. However, the two-photon absorption process is also possible if there really exists some “virtual middle state” between the ground state and the high excited state or the  $\text{Er}^{3+}$  is able to absorb two photons simultaneously. Therefore investigating the mechanism of upconversion is needed.

Besides the violet-and-green dual-wavelength light generation, we also demonstrated the blue-and-green-dual-wavelength light emission in another OSL ELN sample with the modulation period of  $5.3 \mu\text{m}$  for the first-order QPM of 980 nm light. By using a similar experimental setup and pumping the crystal with a 980 nm InGaAs diode laser, 490 nm second harmonic light and the green upconversion fluorescence were also achieved simultaneously.

In conclusion, we first demonstrated the visible dual-wavelength light generation in OSL ELN. Remarkable violet-and-green and blue-and-green lights were obtained through upconversion and QPM SHG at the same time. The detailed absorption and upconversion spectrums of upconversion were carefully measured. The possible physical mechanism was discussed. The phenomena demonstrated the possibility of constructing a novel kind of visible dual-wavelength coherent source that might be used in the laser

display and other areas. Our future work is to increase the intensity of upconversion by increasing the concentration of  $\text{Er}^{3+}$  doping or by  $\text{Yb}^{3+}$  co-doping<sup>24</sup> and enhance the SHG by using the electric poling technique<sup>25</sup> to fabricate the OSL ELN with better periodicity.

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