The third-order optical nonlinearity of the stainless steel doped $SrTiO_3$ thin film grown by L-MBE

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Stainless steel-doped $SrTiO_3$ thin films were fabricated by laser molecular beam epitaxy (L-MBE). Nonlinear optical property of the thin film was measured by the single beam Z-scan technique at the wavelength of 532 nm. Two two-phonon absorption coefficient and nonlinear refractive index were determined to be 9.37×10^{-7} m/W and 1.55×10^{-6} esu, respectively. The merit figure T was calculated to be 1.8, satisfying condition $T < 2$ for an optical switch. The thin film has a very promising prospect for the applications in optical device.

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There is currently much interest in the search for novel materials with good nonlinear optical response for possible applications as optical switching and frequency conversion elements in the telecommunication and information industries[1]. Peroverskite oxides are one promising kind of materials for these applications because of their large nonlinear optical coefficients. For example, a series of studies showed that Rh-doped $BaTiO_3$[2] and gold-dispersed $SrTiO_3$[3] demonstrated large third-order nonlinear optical susceptibility. Inspired by these works, we fabricated stainless steel doped $SrTiO_3$ thin film (hereafter: FSTO film) and investigated its third-order optical nonlinearity by Z-scan technique.

The samples were fabricated on the MgO substrates using a laser molecular epitaxy system. A sintered, In-doped $SrTiO_3$ and a piece of stainless steel (1Cr18Ni9Ti) were used as targets. An excimer laser with a wavelength of 308 nm operated at 3 Hz was employed to ablate the targets. The fabrication was began with the deposition of an In:$SrTiO_3$ layer (~ 110 nm) which severed as the epitaxial matrix, then a stainless steel layer (~ 10 nm) was deposited *in situ* on the top of the In:$SrTiO_3$ layer. Finally, another In-doped $SrTiO_3$ layer (~ 270 nm) was deposited on the top of the stainless steel layer. During the FSTO film growth, the substrate temperature and the oxygen pressure were maintained at 810 °C and 3×10^{-3} Pa, respectively. The thickness of the samples was measured to be 390 nm.

The crystallization and orientation of the films were examined by XRD with Cu Kα radiation. The surface condition was investigated by reflective high-energy diffraction (RHEED). The typical XRD patterns of the samples plotted in Fig. 1 have only (100) peaks and Cr-Ni-Ti-Fe (004) peak, which indicate that the thin films are of single phase and c-axis orientation. The pure spotted RHEED pattern shown in Fig. 2, however, implies that the surface of the samples is comprised of polycrystalline grains. From these two figures, it can be inferred that most of metal substances crystallized on the substrate, but some metal atoms penetrate the surface and nucleate as polycrystalline islands.

The optical nonlinearity of the sample was investigated using a single beam Z-scan technique. A Nd:YAG Q-switched laser that delivered pulses of 10 ns at the

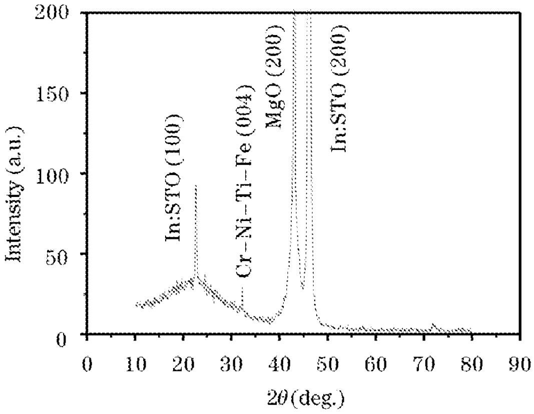

Fig. 1. XRD θ-2θ scan of the stainless steel doped $SrTiO_3$ thin film with MgO substrates.

Fig. 2. Typcial reflective high-energy electron diffraction pattern of the stainless steel doped $SrTiO_3$ thin films grown on MgO substrates.

wavelength of 532 nm was used as the light source. The spatial profile of the laser beam was nearly Gaussian distribution after a spatial filter. Lens with focal length of 120 mm was used for focusing, resulting in a beam waist ω_0 of 30 μm, and a maximum intensity of 6.48×10^7 W/cm^2 at the focal point. The Rayleigh length derived from the relation $Z_R = \pi\omega_0/\lambda$ is 5.3 mm, much longer than either the thickness of the substrate or that of the film. The transmitted beam energy, the reference beam energy, and their ratio were measured by an energy ratiometer simultaneously. A far-field aperture with its linear transmittance, $s = 0.3$, was used in the experiment presented here.

Figure 3 illustrates the transmission spectrum of the sample measured by CARY 2390 spectrophotometer. The transmittance at 532 nm is as high as 90.9%, indicating that no essential linear absorption around 532 nm is experienced in the Z-scan measurement. In addition, the optical band gap is evaluated to be 4 eV.

Figure 4 shows the result of open aperture Z-scan measurement. The shape of the curve with a trough is indicative of the presence of induced nonlinear absorption in the system. Considering the band gap of the sample, the absorption process was dominated by two-photon absorption. The absorption coefficient β can be determined through theoretical fit to the measured trace using Eq. (39) in Ref. [4]. The obtained β value for the FSTO thin film is 9.37×10^{-7} m/W and the corresponding imaginary part of the third-order nonlinear susceptibility $\chi^{(3)}$ is 5.79×10^{-8} esu. The relation between β and $\chi^{(3)}$ is

$$\mathrm{Im}\chi^{(3)}\ (\mathrm{esu}) = \frac{n_0^2 c^2 \beta}{240\pi^2\omega}\ (\mathrm{m/W}) \tag{1}$$

The nonlinear refractive index n_2 can be deduced from closed-aperture Z-scan measurement results. It is known that both the nonlinear refraction and absorption contribute to the transmittance for the closed aperture. To deconvolve them, the transmittance measured with the closed aperture was divided by the transmittance without the aperture. The division result is shown in Fig. 5. The peak-valley configuration of the curve is indicative of a negative nonlinear refractive index n_2 corresponding to self-defocusing material. Based on the best theoretical fit to the empirical data, the difference between the normalized peak and valley $\Delta T_{\mathrm{p-v}}$ is determined to be 0.3. Then the nonlinear refractive index n_2 of the sample is given by

$$n_2 = 2.943 \times 10^6 \times n_0^2 \lambda \tau \omega_0 \frac{\Delta T_{\mathrm{p-v}}}{(1-s)^{0.25} L_{\mathrm{eff}} E}, \tag{2}$$

where λ is the wavelength, τ is the laser pulse width, s is the linear transmittance of the aperture, $E = 18.34$ μJ is the pulse energy. Knowing n_2, the real part of the third-order susceptibility can be obtained by

$$\mathrm{Re}\chi^{(3)}\ (\mathrm{esu}) = \frac{n_0}{3\pi} n_2\ (\mathrm{esu}) \tag{3}$$

The calculated n_2 and $\mathrm{Re}\chi^{(3)}$ are 1.55×10^{-6} and 3.96×10^{-7} esu, respectively. The large third-order nonlinear optical susceptibility of the FSTO is comparable to that of some representative materials, such as high density Cu-doped Al_2O_3 thin films[8], Ce:BaTiO$_3$ quantum dots[9].

To investigate the origin of the large third-order nonlinear optical susceptibility, similar Z-scan measurement was performed on an In-doped SrTiO$_3$ film that has no stainless steel content. The In-doped SrTiO$_3$ film was fabricated under the same condition mentioned above and has the same thickness as the FSTO film. No significant nonlinear response was observed. Therefore the large third-order nonlinear optical susceptibility of

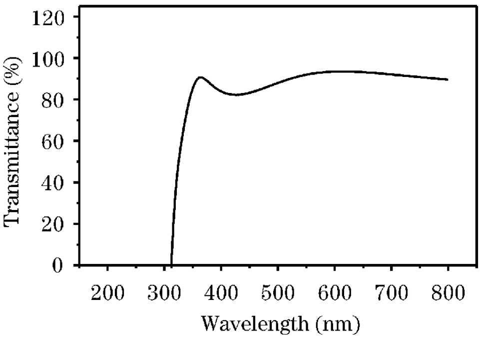

Fig. 3. Transmission spectrum of the thin film.

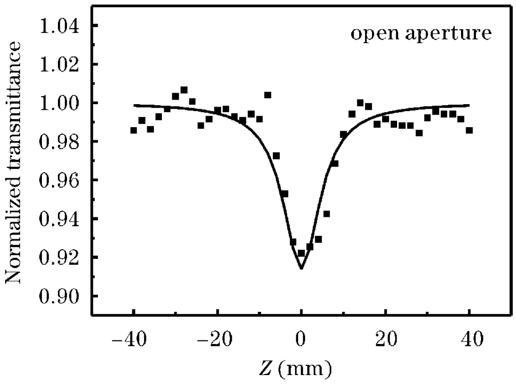

Fig. 4. Normalized Z-scan transmittance curve of the films measured with open aperture. The solid line is the theoretical fit.

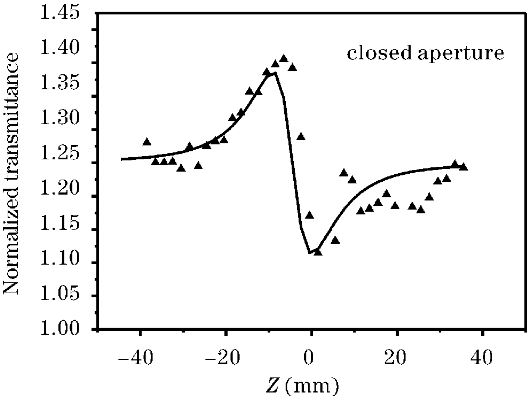

Fig. 5. Normalized Z-scan transmittance curve of the films measured with the closed aperture. The solid line is the theoretical fit.

the FSTO film should result from the nanocomposite structure of the stainless steel dispersed $SrTiO_3$ film.

We calculated the merit figure $T = \beta\lambda/n_2$, which is often used to characterize the suitability of a material for all-optical switching. To carry out the calculation, the value of n_2 has to be converted into m^2/W using the relation, 1 esu$=\frac{40\pi}{n_0 c}$ m^2/W. The obtained T is 1.8 at 532 nm, satisfying the condition for an all-optical switch $T < 2$[5]. Though the wavelength of 532 nm is short for long-haul telecommunications, it may be suitable for short interconnection networks such as local area networks and intra-computer interconnects[6]. Furthermore, since the substrate material MgO can be grown on GaAs, the thin films could be easily integrated into planar optical device structures[7]. Therefore, the stainless steel doped $SrTiO_3$ thin film maybe very promising for optical switching and other optical devices.

In summary, high quality stainless steel doped $SrTiO_3$ thin films have been grown on MgO substrates by L-MBE. Z-scan measurement shows that the thin films possess large third-order susceptibility, the real part and the imaginary part of which are 3.96×10^{-7} and 5.79×10^{-8} esu, respectively. High nonlinear refractive coefficient and a small merit figure make it an attractive material for potential applications in optical devices.

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