

## Magneto-optical and Reflectivity Studies of SrTiO<sub>3</sub>

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Faraday rotation spectra were measured in single crystals of SrTiO<sub>3</sub> between 1.7 and 3.2 eV at room temperature and at 20 K. The rotation showed a monotonous increase as the absorption edge was approached. In a single crystal with thickness of 0.5 mm, it took a value as large as 7.7° at 400 nm for magnetic field of 1 T. The center of dispersion of the magneto-optical spectrum was determined to be 4.19 eV. This value agreed with the peak energy position (=4.2 eV) of the imaginary part of dielectric function,  $\epsilon_2$  deduced from the reflectivity spectrum measured between 0.5 and 20 eV.

**Key words:** strontium titanate, Faraday rotation spectrum, reflectivity spectrum, isolator for short wavelength

### 1. Introduction

Recently short wavelength (blue to violet) laser diodes are appearing into the market. However, optical isolator materials suited for this wavelength region are not easily available. Magnetic garnets, most prevailing optical-isolator materials, are not transparent in the visible wavelengths due to strong absorption band caused by charge-transfer transitions. Although enhanced magneto-optical effect is observed in bismuth-substituted garnets, the figure of merit  $F$  is not sufficient to be applied to optical isolators for wavelengths shorter than 1000 nm, except for the transmission window around 800 nm. For example a Y<sub>2.7</sub>Bi<sub>0.3</sub>Fe<sub>5</sub>O<sub>12</sub> film shows a specific Faraday rotation of  $9 \times 10^4$  deg/cm and an absorption coefficient of  $5 \times 10^4$  cm<sup>-1</sup> at 400 nm, which leads to  $F$  of approximately 0.5 deg/dB. Since the Faraday rotation is known to be proportional to Bi-content, one can get larger rotation by increasing Bi content. It was reported that Faraday rotation as large as  $10^6$  deg/cm was obtained in 100% Bi-substituted garnet film, i.e., Bi<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>(BIG) grown by ion beam sputtering on a Gd<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub> (GGG) substrate.<sup>1)</sup> However even with this film  $F$  is no more than 5 deg/dB. This means that to get 45 deg rotation for optical isolators, the insertion loss amounts to as large as 9 dB. In addition, optical isolator requires thickness of  $4.5 \times 10^{-5}$  cm=450 nm, which exceeds critical thickness for pseudomorphic growth of the BIG film on GGG.

It is for this reason that optical isolators using diluted magnetic semiconductors (DMS) such as Cd<sub>1-x</sub>Mn<sub>x</sub>Te and (Hg<sub>1-y</sub>Cd<sub>y</sub>)<sub>1-x</sub>Mn<sub>x</sub>Te<sub>2</sub> have been investigated for wavelengths in which magnetic garnets are not available, in spite of the demerit that they are in the paramagnetic phase or the spin-glass phase showing no spontaneous magnetization and require stronger magnetic field than garnets to be applied to isolators. The Verdet constant in commercially available CdHgMnTe takes a value of  $\sim 4 \times 10^{-2}$  deg/cmG ( $\sim 2 \times 10^2$  deg/cm for an applied field of

5000 G) at 500 nm. Although the band gap of the DMS can be swept from the IR to UV (350 nm) by varying Mn content, the wavelength region for practical application is limited to  $\lambda > 500$  nm because of the absorption due to the d-d transitions inherent to Mn<sup>2+</sup> ion.

Only a few magnetic materials have been reported to show Faraday effect in short wavelength region at room temperature. FeBO<sub>3</sub> and FeF<sub>3</sub> were intensively studied materials since they are transparent in visible wavelengths and show considerably large Faraday rotation in short wavelengths.<sup>3)</sup> Both FeBO<sub>3</sub> and FeF<sub>3</sub> are canted antiferromagnets with  $T_N=348$  K and 365 K, respectively. Absorption spectrum in a single crystal of FeBO<sub>3</sub> locates the fundamental absorption edges at about 500 nm and two below-gap absorption peaks around 600 nm and 900 nm. Faraday rotation shows a monotonous increase towards shorter wavelengths. At 525 nm where the absorption takes a minimum, the rotation is 2300 deg/cm, corresponding to  $F=15$  deg/dB. On the other hand in FeF<sub>3</sub> an absorption edge at 240 nm and  $F=16$  deg/dB at 400 nm. However both materials suffer an optical birefringence as large as  $10^5$  deg/cm, which makes practical applications of these materials quite difficult. In addition, they are not commercially available as single crystals of good quality.

We investigated magneto-optical effect in nonmagnetic materials. In 1967 Baer reported considerably large Faraday rotations in some of the perovskite transition metal oxides such as SrTiO<sub>3</sub>, BaTiO<sub>3</sub>, KTaO<sub>3</sub> and KTa<sub>0.35</sub>Nb<sub>0.65</sub>O<sub>3</sub> (KTN) as well as in rutile TiO<sub>2</sub>.<sup>4)</sup> Band gap energies are 3.40 eV (365 nm) in SrTiO<sub>3</sub> at 296 K, 3.25 eV (381 nm) in BaTiO<sub>3</sub> at 403 K, 3.77 eV (329 nm) in KTaO<sub>3</sub> at 296 K and 3.54 eV (350 nm) in KTN at 296 K. He plotted spectra of  $n\theta H$  (Verdet constant multiplied by refractive constant) in these materials. The  $n\theta H$  of SrTiO<sub>3</sub> at 3 eV can be read from the plot as  $-2.6 \times 10^{-2}$  deg/cmG for temperatures 296K and 77 K. Assuming  $n=2.6$  Verdet constant can be obtained as  $-1.0 \times 10^{-2}$  deg/cmG. Other perovskites and rutile showed the similar magnitude of Faraday rotation. This study did not attract attention at that time, since single crystals of such oxides were not easily available at that time and there were no needs for short wavelength optical isolators. In 1970, Feldman et al. showed Faraday rotation in Bismuth oxide compounds, such as Bi<sub>12</sub>GeO<sub>20</sub>, Bi<sub>12</sub>SiO<sub>20</sub>, Bi<sub>12</sub>TiO<sub>20</sub>, etc.<sup>5)</sup> According to their report Verdet constants of these compounds are nearly equal to  $-2 \times 10^{-2}$  deg/cmG at 410 nm. However these bismuth compounds show optical activity and cannot be selected as isolator materials.

Recently single crystals of SrTiO<sub>3</sub> with a large size and a good crystalline quality become easily available as substrate materials for high-Tc superconductors. This

prompted us to reinvestigate optical and magneto-optical properties of SrTiO<sub>3</sub>. In the present study we measured Faraday rotation spectra in good quality single crystals at room temperature as well as at 20 K and confirmed that the material shows considerably large temperature-independent Faraday rotation in the short wavelength region around 400 nm. To study the electronic origin of the magneto-optical effect we also measured the reflectivity spectrum for photon energies between 0.5 and 30 eV with the help of synchrotron radiation and deduced dielectric functions using Kramers-Kronig relation.

## 2. Experiments

Specimens for the present study were single crystals of SrTiO<sub>3</sub> obtained from different suppliers as substrates for epitaxial growth of high-T<sub>c</sub> oxide superconductors. All of the crystals were cut parallel to the {100} plane and mechanically polished. Sample thickness was typically 0.5 mm. The samples were set on the cold finger of a helium refrigerator cryostat and were inserted between the pole pieces of an electromagnet, the maximum field of which is 1.2 T. Faraday rotation spectra were measured using polarization modulation technique employing a photoelastic modulator (PEM). A 150 W Xe lamp was used as a light source. The light was passed through a Glan Thomson polarizer whose transmission angle was set to make an angle of 45° with the vertical axis and was focused by a lens through the PEM (Hinds inc. PEM90; modulation frequency was 50 kHz) on the sample surface. The light was then passed through an analyzer (Glan Thomson prism) and focused on the slit of a JASCO CT-25C monochromator with a 1200 lines/mm grating blazed at 750 nm. The light through the output slit was detected by a photomultiplier (Hamamatsu R928). The dc component of the output was kept constant by a feed-back circuit to a high voltage supply for the photomultiplier. The ac component was amplified using a lock-in amplifier tuned to the doubled frequency of the modulator. This method is in principle the same as described for a measurement of Kerr effect proposed by one of the authors (K.S.).<sup>6)</sup> The calibration was carried out point by point by adjusting the angle of the analyzer to get the null signal.

Reflectivity spectra were measured using a Hitachi U-3410 spectrophotometer for energy region between 0.5 and 4 eV and a Seya-Namioka monochromator combined with the synchrotron radiation (SR ring at Institute of Solid State Physics, University of Tokyo) for energies from 2 to 30 eV. Kramers-Kronig analysis was performed with the help of the optical constants determined for 2 to 5 eV by a spectroscopic ellipsometer.

## 3. Results and Discussion

### 3.1 Faraday rotation and optical transmission spectra

SrTiO<sub>3</sub> crystals show a negative Faraday rotation, which is proportional to the applied magnetic field. (Here, a positive sign is defined to be in the same direction as the current in the coils producing the magnetic field.) As shown by dots in Fig. 1 the Faraday rotation of a single crystal of

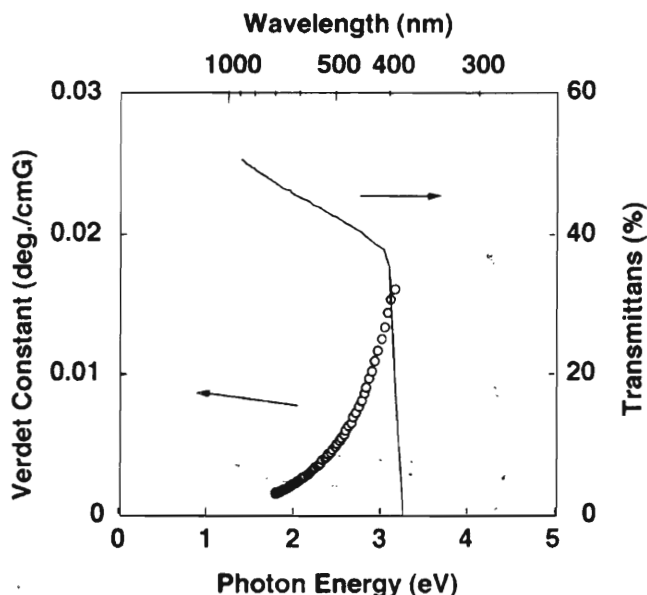


Fig. 1 Spectra of Faraday rotation and transmission of a SrTiO<sub>3</sub> single crystal.

SrTiO<sub>3</sub> increases monotonously with decrease of wavelength. The Verdet constant was determined as  $-1.54 \times 10^{-2}$  deg/cmG at the wavelength of 400 nm. The Faraday rotation should be compared with that of commercially available CdHgMnTe optical isolator materials, which was reported to be  $+3 \sim 4 \times 10^{-2}$  deg/cmG at 980 nm.

In order to investigate whether the magneto-optical effect is intrinsic or due to impurities or defects we measured in Fe-doped crystals and annealed crystals. No change in the value of the Faraday rotation was observed, from which we concluded that the effect is intrinsic.

The optical transmission spectrum of the SrTiO<sub>3</sub> crystal (0.5 mm in thickness) at room temperature is also shown in Fig. 1 by a solid curve. The Verdet constant shows a monotonous increase with photon energy up to 3.2 eV ( $\sim 388$  nm), at which the material loses transparency. Optical transmission was found to be as small as 40% at 400 nm. Such reduction of transmission may be attributed to surface reflection since refractive index of this material is fairly large. If  $n=2.6$  is assumed the normal incidence reflectivity  $R$  amounts to 19.7%, thus leading to  $T=(1-R)^2=0.64$  even if the material is assumed to be completely transparent. Such reflection may be minimized by suitable selection of antireflection coating. However, absorption coefficient after correction of reflection was estimated to be as large as  $\sim 10$  cm<sup>-1</sup> at 400 nm, corresponding to an insertion loss of 40 dB/cm. Such a large value of residual absorption may be attributed to intrinsic defects introduced during growth and surface damages due to polishing process and can be substantially reduced.

Using the Verdet constant of  $1.54 \times 10^{-2}$  deg/cmG and the insertion loss of 40 dB/cm we get figure of merit of  $F=3.85$  deg/dB for applied field of 1T. Reduction of residual absorption is necessary.

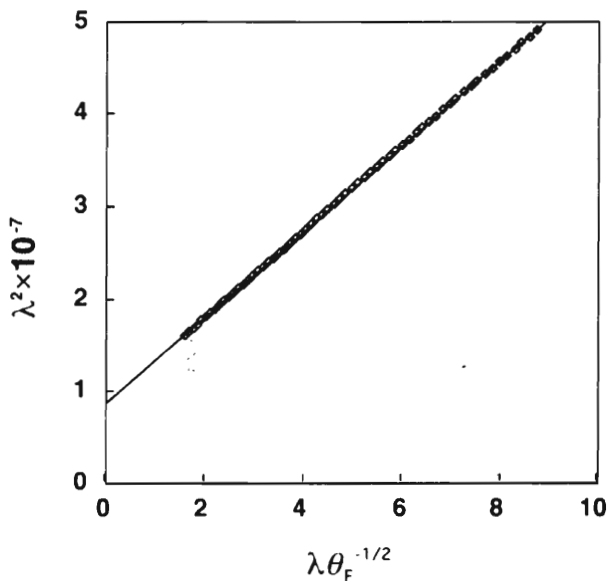


Fig. 2 Plot of  $\lambda(\theta_F)^{-1/2}$  vs.  $\lambda^2$  in SrTiO<sub>3</sub> for determination of parameters in eq (1).

### 3.2 Determination of center of dispersion of the Faraday effect

In order to determine the energy of optical transition from which Faraday rotation originates, we used the following dispersion relation<sup>7)</sup> for the spectral dependence of Faraday rotation:

$$\theta_F = A/\lambda^2(1 - \lambda_0^2/\lambda^2)^2 \quad (1)$$

where  $\lambda$  is wavelength,  $\lambda_0$  center wavelength of dispersion and  $A$  constant. The value of  $\lambda_0$  can be determined by  $\lambda(\theta_F)^{-1/2}$  vs.  $\lambda^2$  plot shown in Fig. 2, from which the photon energy for the center of dispersion was determined as 4.19 eV. This value is larger than that of Baer's analysis in which the center of dispersion was determined as 3.4 eV by using a sophisticated formula for Faraday effect of direct band gap materials.

### 3.3 Temperature dependence

The Faraday rotation spectra measured at 20 K and room temperature are shown in Fig. 3, from which it is found that temperature dependence of the rotation is negligible. This result is also consistent with the Baer's report. The temperature-independent Faraday rotation suggests that this effect does not originate from any paramagnetic centers, for which strong temperature dependence and positive sign are expected. The small temperature dependence observed above is quite favorable for optical isolator application.

### 3.4 Dielectric functions deduced from reflectivity spectrum

The negative rotation in SrTiO<sub>3</sub> was attributed by Baer to the Zeeman splitting of the fundamental absorption edge resulting from the p-d charge transfer transition. He associated the transition energy (3.4 eV) deduced from the theoretical fitting with the shoulder observed at 3.4 eV in the reflectivity spectrum reported by Cardona.<sup>8)</sup>

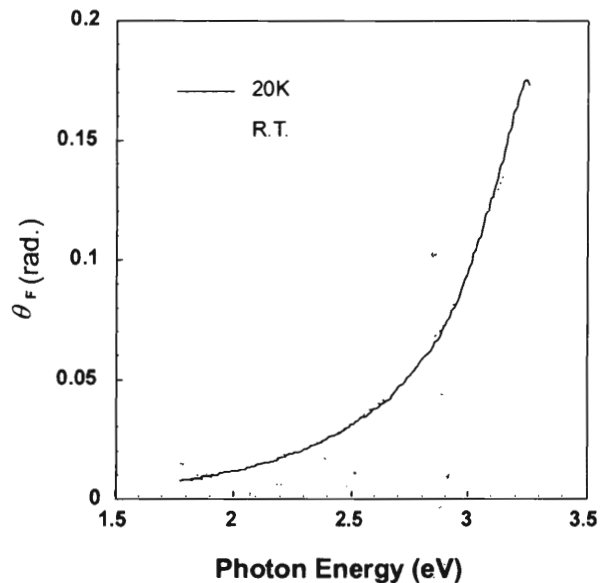


Fig. 3 Faraday rotation spectra of the SrTiO<sub>3</sub> crystal measured at 20 K (solid curve) and room temperature (dotted curve).

We measured a reflectivity spectrum at room temperature between 0.5 and 30 eV in the same sample for which Faraday effect was evaluated. Reflectivity spectrum is shown in Fig. 4. From the reflectivity spectra, real and imaginary parts of the dielectric functions were calculated using Kramers-Kronig relation. To determine appropriate extrapolation parameters for this analysis we used spectra of refractive constant  $n$  and extinction constant  $\kappa$  determined by spectroscopic ellipsometry. The resulting dielectric functions are given in Fig. 5. Here we only show spectra for energies below 10 eV since the Faraday effect of this crystal is essentially related to the fundamental absorption region.

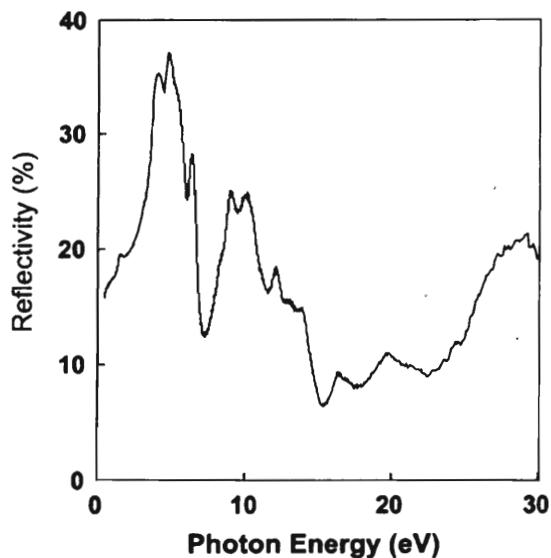


Fig. 4 Reflectivity spectrum of a SrTiO<sub>3</sub> single crystal measured using synchrotron radiation.

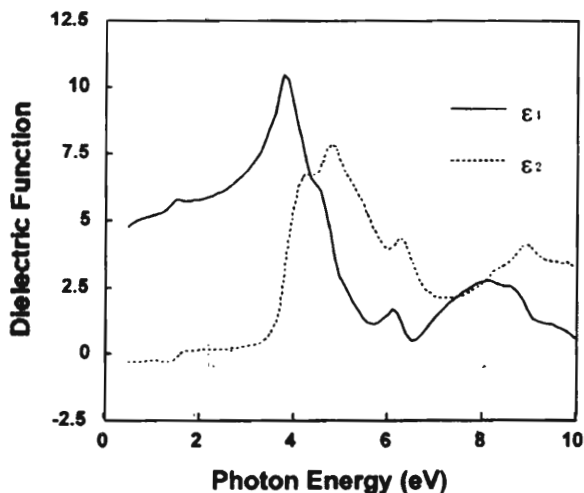


Fig. 5 Real and imaginary part of dielectric functions of SrTiO<sub>3</sub> single crystal.

Contrary to Cardona's data we did not observe any structures around 3.4 eV in the spectrum of the imaginary part of the dielectric function corresponding to the absorption spectrum. The first absorption peak in our spectrum is found at 4.2 eV, which shows a good correspondence with the energy position (4.19 eV) of the center of dispersion determined by the analysis of the Faraday rotation spectrum. We suspect that the 3.4 eV structure observed by Cardona is not an intrinsic one but due to defects or impurities.

#### 4. Conclusion

Magneto-optical spectra were measured in single crystals of SrTiO<sub>3</sub>. Comparatively large temperature-independent Faraday rotation was obtained.

Figure of merit of  $F=3.85$  deg/dB is obtained at 400 nm for applied field of 1T. Further reduction of growth-induced residual absorption and improvement of surface treatment is required to get device quality materials. Fabrication of a short-wavelength isolator using SrTiO<sub>3</sub> is under investigation. Details will be reported in later publications.

The center of dispersion for Faraday rotation was determined as 4.19 eV from the analysis of the spectrum. The energy is consistent with the lowest energy peak position of 4.2 eV in the imaginary part of the dielectric function obtained from the reflectivity spectrum

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