Development of Large Size Sapphire Crystals for Laser Interferometer Gravitational-Wave Observatory

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***Abstract*—Because of its high density and superior quality factor, sapphire crystal, as a candidate material for test masses, has attracted much attention in gravitation wave communities. The use of sapphire crystal, however, is limited by its size, homogeneity, and absorption. An effort has been made to overcome these difficulties at the Shanghai Institute of Optics and Fine Mechanics in collaboration with the Laser Interferometer Gravitational-Wave Observatory Laboratory. By using directional temperature gradient technique (TGT), sapphire crystals of 11 cm in diameter and 8 cm in length were grown at the C plane (0001). The results indicate that a homogeneity of $<5 \times 10^{-7}$ and an absorption of 35–65 ppm/cm have been achieved at wavelength of 1.06 μm. This paper presents the TGT growth of sapphire crystals, their transmittance spectra, optical homogeneity, and absorption. Future applications for gravitational wave experiments are discussed.**

***Index Terms*—Crystal, gravitation wave, interferometer, mirror, sapphire, test mass.**

I. Introduction

ALBERT Einstein predicted the existence of gravitation waves as part of the theory of general relativity [1]. When the space–time curvature varies rapidly it should emanate curvature ripples (gravitation waves), which propagate through universe at the speed of light and carry the imprint of graviton. The Laser Interferometer Gravitational-Wave Observatory (LIGO), at Richland, WA, and Baton Rouge, LA, is designed to study astrophysical gravitational waves as well as their sources.

For each power recycled Michelson interferometer with Fabry–Perot arm cavities, two pairs of mirrors (test masses) at 4 km apart are arranged in an L shape, as shown in Fig. 1. Any difference of the distances between two pairs of mirrors will direct signal light to a photo sensor. Since the gravitational wave changes the difference of the distances between two pairs of mirrors it would cause variations of the signal light on the photo sensor. Two interferometers of this kind are being built by LIGO. One is near Richland, WA, and the other near Baton Rouge, LA. They will operate in unison to reject false signal. The overall detection limit of this kind of interferometer is limited by its noise. Fig. 2 shows the predicted root mean square (rms) noise spectra for LIGO I. While the seismic and photon shot noises dominate the frequency domains of below 70 Hz and above 200 Hz, respectively, the thermal noise,

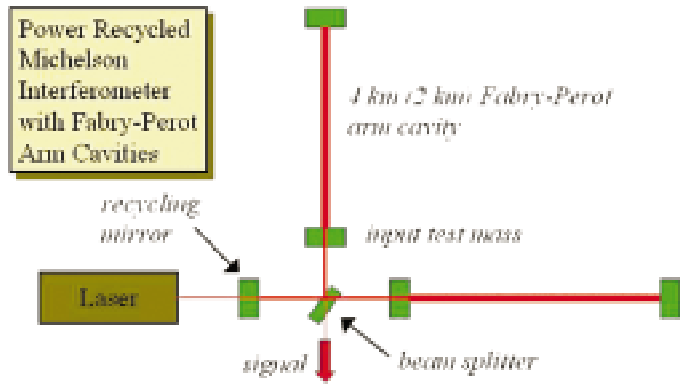

Fig. 1. A schematic showing the configuration of LIGO interferometer.

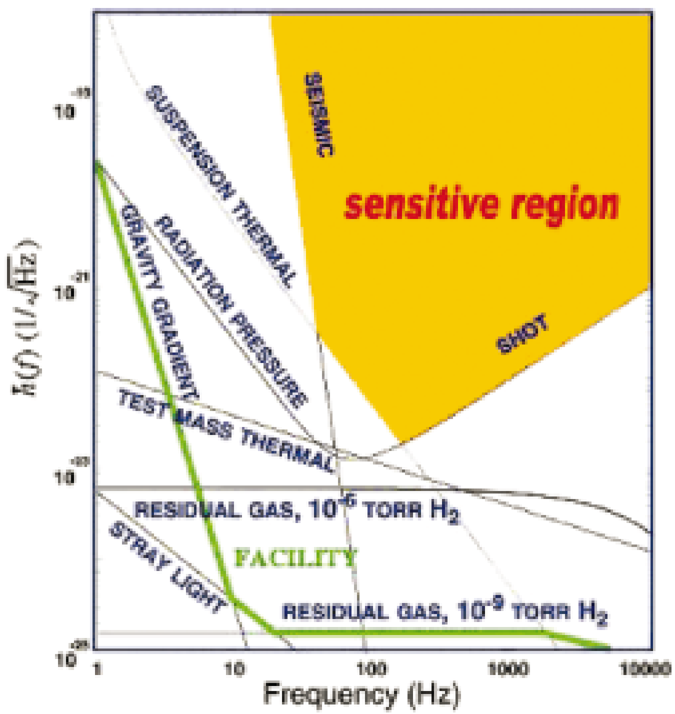

Fig. 2. The rms noise of LIGO I from various sources and corresponding sensitive region are shown as a function of frequency.

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Fig. 3. A photo showing a TGT furnace used to grow sapphire crystals in SIOM.

caused by thermally induced vibrations of the test masses and their suspensions, dominates the frequency domain in the most sensitive region between 70 and 200 Hz.

One of the critical research areas for the enhancement of the LIGO detector is to reduce the test mass thermal noise in the internal modes, labeled as "test mass thermal" in Fig. 2. Because of its excellent optical properties, the maturity of production technology, and good, but not outstanding, thermal noise, fused silica is used for test masses in initial LIGO I detector. Single crystal materials, such as sapphire, are expected to provide a good chance to reduce further the thermal noise. Sapphire crystal is transparent at visible and near-infrared wavelengths. Compared to the fused silica, its higher speed of sound, higher density, and lower mechanical loss factor or higher quality factor, would give a major improvement in the thermal noise due to internal modes. Low-loss sapphire crystals have been successfully developed as substrates in Fabry–Perot reference cavities [2]. Sapphire crystal is commercial available, but not in the size of LIGO requirement, i.e., 35 cm in diameter and 12 cm in length [3]. There are also significant issues that need to be addressed for sapphire application in LIGO, among which the optical homogeneity of $<5 \times 10^{-7}$ and absorption of <10 ppm/cm at wavelength of 1.06 μm are the two most important requirements [3].

Large sapphire crystals of $\phi 28 \times 15$ cm are grown at a plane $(11\bar{2}0)$, m plane $(10\bar{1}0)$ or r plane $(1\bar{1}02)$ by the Heat Exchanger Method (HEM) in Crystal System Inc., Salem, MA [4]. To eliminate the effect of bi-refringence, the orientation (0001), however, is preferred in LIGO application. The existing commercially available crystals also fall short for the required homogeneity and absorption. In the last four years, an extensive research and development (R&D) program has been carried out in the Shanghai Institute of Optics and Fine Mechanics (SIOM), Chinese Academy of Sciences (CAS), Shanghai, China, in a collaboration with the LIGO Laboratory to develop high-quality large size sapphire crystals. In this paper, we present results of this R&D program and discuss its future applications in advanced LIGO.

Fig. 4. A photo showing a sapphire boule grown at SIOM with diameter of 11 cm and length of 8 cm.

II. Development of Sapphire Crystals

Sapphire crystals are grown at SIOM by the directional temperature gradient technique (TGT), which was invented by Prof. Yongzong Zhou *et al.* [5] in 1978 and patented in 1985. This technique uses molybdenum crucible, molybdenum shielding, and graphite heater [5], [6], [7]. Fig. 3 shows a photo of a TGT furnace constructed in SIOM for LIGO. A molybdenum crucible is heated by cylindrical plumbago elements and insulated by multilayer molybdenum shield. The required temperature gradient is obtained by cutting troughs on the heating elements to form an electrical rectangular circuit and by making holes in proper distribution to adjust the resistance of the heating elements. The sapphire growth is conducted in vacuum filled with high purity argon. This allows purification by volatization of impurities with high vapor pressure. Impurities with small segregation coefficient in sapphire are migrated to the outside surface during solidification.

The difference between the TGT and the HEM of Crystal Systems, Inc., is that the TGT does not use helium (or any gas) as heat exchange medium and provides temperature gradient by the resistance of the heater and water cooling for graphite electrodes. Ti : Sapphire crystals of $\phi 11 \times 8$ cm with an orientation of (0001), $(11\bar{2}0)$, and $(10\bar{1}0)$ were routinely grown. Fig. 4 is a photo showing an as-grown sapphire boule grown at c plane (0001) in SIOM for LIGO. It has a diameter of 11 cm and a height of 8 cm.

As-grown sapphire crystal is pinkish because of various contaminations. Fig. 5 shows the longitudinal transmittance of a typical as-grown sample, measured by using a Hitachi U-3210 UV/visible spectrophotometer with double beam and an integrating sphere. The absorption peaks in the longitudinal transmittance spectrum, as shown in Fig. 5, are attributed to Ti^{3+} and Cr^{3+} contaminations.

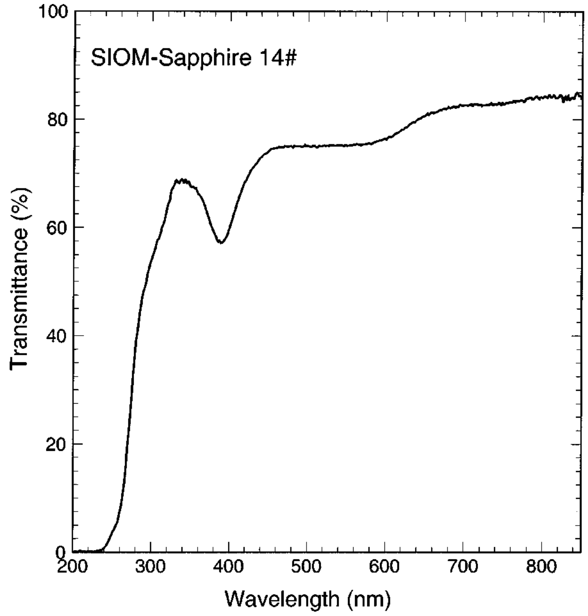

Fig. 5. Longitudinal transmittance spectrum is shown as a function of wavelength for a typical as-grown sapphire sample at SIOM.

TABLE I
AN EXAMPLE OF GDMS ANALYSIS FOR RAW MATERIALS

Element	Japan	Zhejiang	Dalian	Biesterfeld	S.E.I
B	0.4	10	1.0	<0.05	<0.05
F	0.4	10	1.0	<0.05	<0.05
Na	<1	18	16	<0.2	<0.2
Mg	11	2.1	7.6	0.5	0.2
Si	17	15	2.9	0.5	1.0
Ca	20	5.6	1.5	<0.05	<0.05
Ti	0.6	1.1	0.5	0.3	6.0
Cr	<10	18	10	<1	<1
Fe	15	20	<10	<1	<1
Co	3.2	1.1	0.5	<0.005	<0.005
Ni	19	<5	7.5	<0.05	<0.05
Cu	1.4	1.9	0.9	<1	<1
Purity(%)	99.9897	99.9891	99.9948	99.9998	99.9988

To eliminate the pinkish color, glow discharge mass spectroscopy (GDMS) analysis was carried out in Shiva Technologies, Inc. to identify impurities in raw materials as well as sapphire crystals. Table I lists an example of GDMS analysis result on raw materials, in either powder or craquelle forms, from several vendors. The results show that the best raw material is from the Biesterfeld U.S., Inc., which has purity of 5N+ and was produced in the Czech republic. The best Chinese raw material is produced in Dalian, which has purity of 4N+. It also shows that the raw material from Sapphire Engineering, Inc., which was produced in Switzerland, has high Ti contamination. The quality of sapphire crystals is improved by using raw material of higher purity.

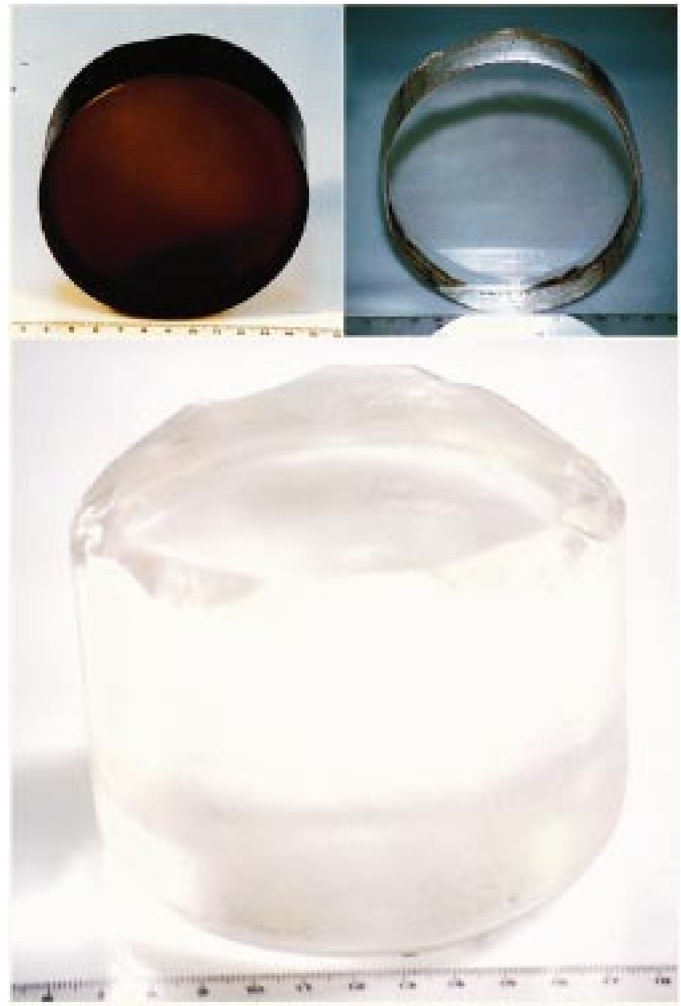

Fig. 6. Photos showing a sapphire sample after annealing in air (top left) and in H_2 (top right) and a full size sapphire sample after two-step annealing (bottom).

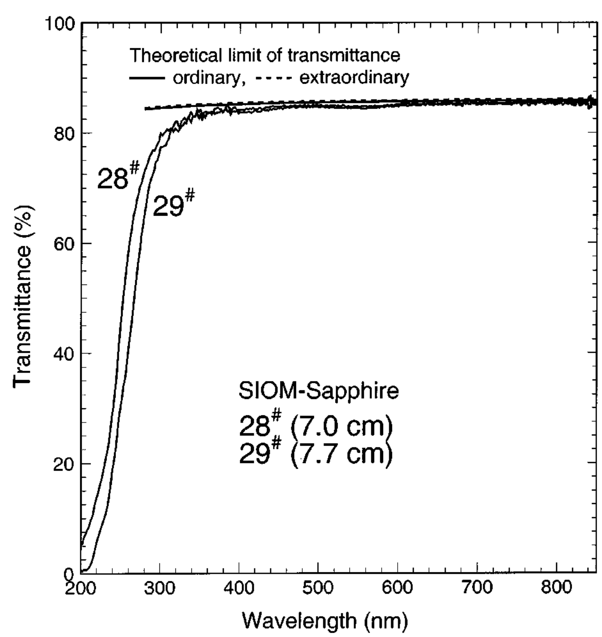

Fig. 7. Transmittance of SIOM samples #28 and #29 is shown as a function of wavelength and compared to the theoretical limit for light of ordinary and extra ordinary polarization.

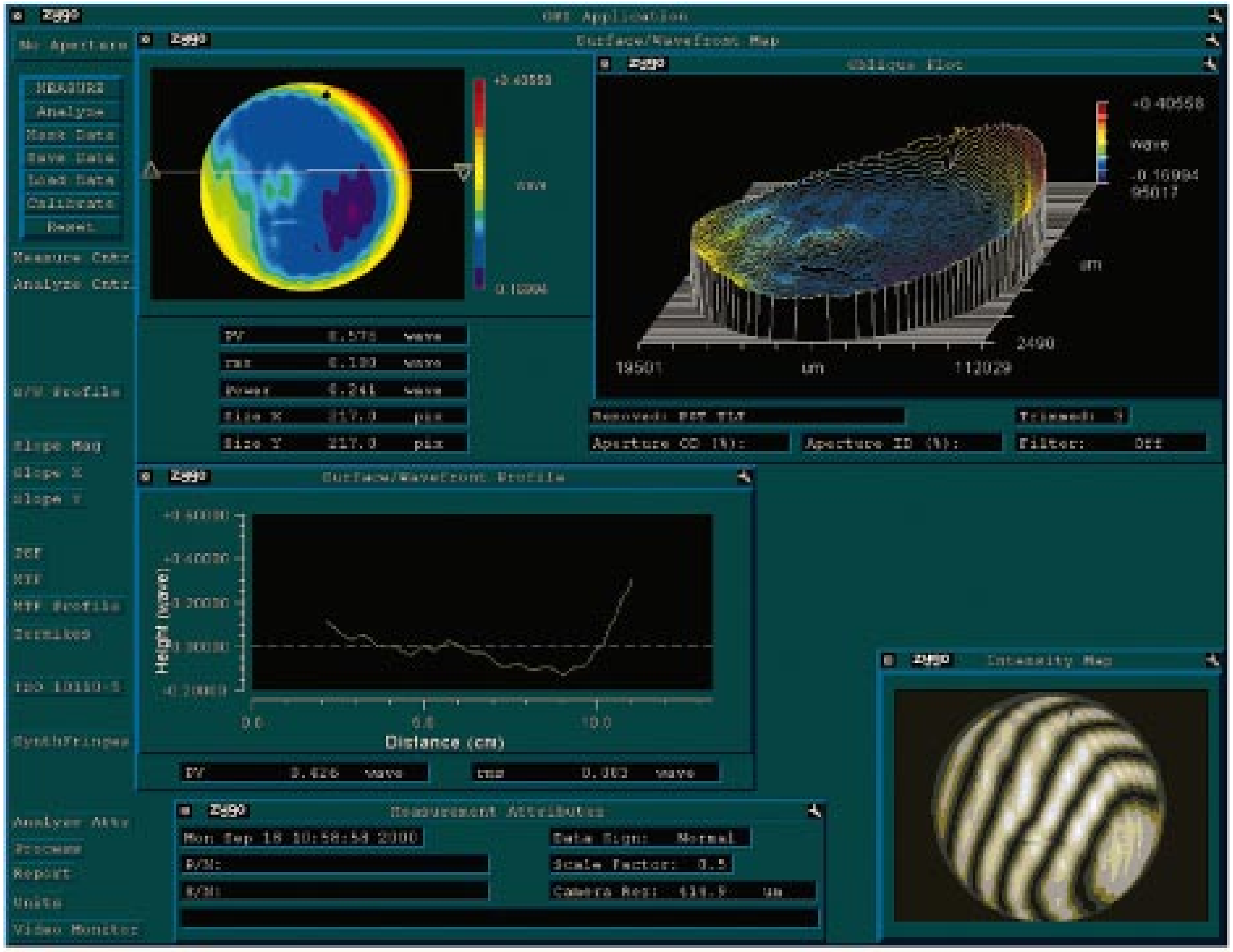

Fig. 8. Display of optical homogeneity measured by a ZYGO Mark-III interferometer at SIOM.

The GDMS analysis also identified trace elements of Fe, Ni, and Mo in sapphire crystals, which are result of contamination during the growth process. Thus, the use of purified raw material only solves a part of the contamination problem. After extensive R&D, a proprietary two step post growth thermal annealing technique, was developed at SIOM. It is understood that the origin of the pinkish color in the as-grown sapphire crystals is caused by contaminations of Ti^{3+} from raw material and carbon from furnace, which causes oxygen vacancies and, thus, form F and F^+ centers. After annealing in O_2 or air, sample turns brownish, as shown in the top left photo of Fig. 6, because of strong oxidation of carbon, but $Ti^{3+} \rightarrow Ti^{4+}$. After annealing in H_2, the carbon contamination is eliminated as hydrocarbon and Ti ions remain as Ti^{4+}, which compensate oxygen vacancies, thus eliminates color centers. The top right photo of Fig. 6 shows the same sample after annealing in H_2. A photo of a full size sample after two step annealing is shown in the bottom of Fig. 6. No visible coloring is observed in samples after two-step annealing. This proprietary two-step post growth thermal annealing technique is effective in eliminating color centers in sapphire crystals.

The longitudinal transmittance spectrum of two ϕ11-cm sapphire samples (#28 and #29) after treated with the two-step annealing process is shown in Fig. 7. The transmittance of these samples with more than 7-cm path length before two-step annealing is similar to that shown in Fig. 5, but approaches the theoretical limits after annealing, indicating very small residual absorption. These theoretical limits were calculated by using sapphire refractive index, taking into account multiple bounces between two end surfaces and assuming no internal absorption [8]. Because of the slight bi-refringence, the theoretical limits for ordinary (solid) and extraordinary (dashes) polarizations are both shown in the figure.

III. Application in LIGO

As discussed in Section I, the advanced LIGO detector may use sapphire crystals of diameter of 35 cm and length of 12 cm. The final specification for the portion of crystals within the central 8 cm is the absorption of <10 ppm/cm and the optical homogeneity of $< 5 \times 10^{-7}$ at $\lambda = 1.06\,\mu$m. Fig. 8 shows a display of optical homogeneity measured by a ZYGO Mark-III interferometer for sample #29 at SIOM. The rms deviation of the surface/wavefront map was measured as 0.05λ at 632 nm, i.e., 32 nm for sample #28, which can be converted to an optical homogeneity of 4.5 $\times$ 10^{-7} at 632 nm. Fig. 9 shows a contour of the optical homogeneity measured at Caltech by a customized Wyko 6000 interferometer for sample #28. The corresponding optical homogeneity at 1064 nm was measured to be 2.4 $\times$ 10^{-7}.

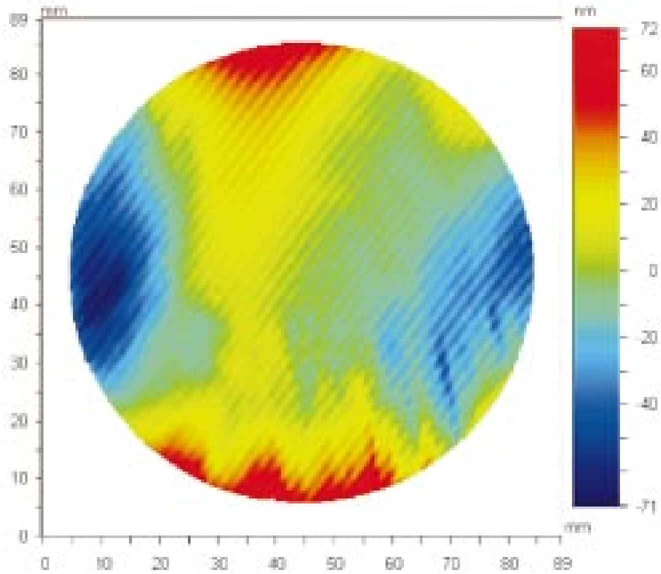

Fig. 9. A contour of optical homogeneity measured by a Wyko 6000 interferometer at Caltech.

TABLE II
LIGO EVALUATION OF SAPPHIRE SAMPLES

Sample	#7	#28	#29	CSI	F. S.
IR Absorption (ppm/cm)	–	35–65	–	80	2–5
Absorption @514 nm (ppm/cm)	–	280–350	–	1,200	< 20
Homogeneity[a]: PV (nm)	165	107	–	177	52
rms (nm)	28	17	–	30	9
SIOM Data[b]: PV (nm)	422	369	362	–	–
rms: (nm)	48	32	52	–	–

a Measured at 1,064 nm.

b Measured at 632 nm.

Table II lists a summary of evaluation for three SIOM sapphire samples and compared to that of typical sapphire samples from Crystal System, Inc. (CSI) and typical fused silica (FS) samples used in the LIGO I. The absorption of sapphire and fused silica samples were measured at Ginzton Laboratory, Stanford University, Stanford CA, by a photothermal deflection technique [9]. The optical homogeneity of SIOM sapphire samples was measured in both SIOM and LIGO at 632 and 1064 nm, respectively. The optical homogeneity of SIOM sapphire samples satisfies LIGO requirement. The key technical difficulty is the absorption. The best SIOM sample #28 is about a factor of five worse than the LIGO final specification (<10 ppm/cm). The sapphire crystals of this quality may find wide applications in optical instruments. For LIGO application, however, larger size and lower absorption are two issues to be addressed in the next phase of the R&D program.

IV. SUMMARY

Beginning July 1997, SIOM has been working on sapphire development for LIGO. Impurities in raw materials and crystals were identified. A two-step after-growth thermal-annealing technique was developed, which is effective in eliminating color centers in sapphire crystals. The LIGO evaluation of SIOM sapphire samples reveals that these samples have attractive quality such as optical homogeneity of $<5 \times 10^{-7}$ and absorption of 35–65 ppm/cm. Although the optical homogeneity of SIOM sapphire samples satisfies the LIGO requirement, their absorption still falls short by a factor of five. This R&D program will be continued to develop large-size high-quality sapphire crystals required by LIGO.

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