



4.9 Developments of Integrated Laser Crystals by a Direct Bonding Method

Akira SUGIYAMA^a, Masaki KATSUMATA^b,
Hiroyasu FUKUYAMA^a, Mitsuhiro TANAKA^b and Yukikatu OKADA^b

^aAdvanced Photon Research Center, Kansai Research Establishment,
Japan Atomic Energy Research Institute,
8-1 Umemidai, Kizu-cho, Souraku-gun, Kyoto, 619-0215, Japan

^bKougaku Giken Co. Ltd.,
135 Nurumizu, Atsugi-shi, kanagawa-ken, 243-0033, Japan

Laser crystal integration using a neodymium-doped yttrium vanadate (or orthovanadate) laser crystal, and non-doped yttrium vanadate crystals that function as cold fingers has been demonstrated. A newly developed dry etching process was adopted in the preparation for contact of mechanically polished surfaces. In the heat treatment process, temperature optimization was essential to get rid of precipitation of vanadic acid caused by the thermo-chemical reaction in a vacuum furnace. The bonded crystal was studied via optical characteristics, magnified inspections, laser output performances pumped by a CW laser diode. From these experiments, it was clear that the integrated Nd:YVO₄ laser crystal, securing the well-improved thermal conductivity, can increase laser output power nearly twice that of the conventional single crystal which was cracked in high power laser pumping of 10 W due to its intrinsic poor thermal conductivity.

Keywords: Direct Bonding, Hydrogen Bonding, Laser Crystals, Nd:YVO₄, ESCA,
Laser Tomography, Transmitted Wavefront, TEM, EDX, LD

1. Introduction

A new technique of material integration for laser crystals has been much in demand to improve heat reduction caused by high power laser pumping. We have investigated obtaining such integrated laser crystals employing a direct bonding technique using commercially available high quality crystals. In our previous studies, we succeeded in bonding Ti:sapphire laser crystals.^{1,2} In this case, the bonding surfaces of a stable oxide crystal were treated with chemical processes to clean up and to create a hydrophilic (-OH) thin layer for hydrogen bonding. However, in applications for different crystal bonding, the chemical processes require the selection of suitable chemical etchants for each crystal to be bonded and optimization in the handling of the etching period and temperature. In most of the cases, hazardous strong acid and alkaline water are used for the etchant in a time consuming procedure. In addition, these etchants cannot be applied for hygroscopic crystals and fluoride crystals such as non-linear borate crystals and YLF laser crystals. From the viewpoint of these problems, we have developed a dry etching process using argon ion beam irradiation instead of the conventional chemical processes. We have succeeded in bonding a surface area of 5mm x 6mm. In this paper, we report our newly developed bonding process of YVO₄ and Nd:YVO₄ laser crystals. Further investigations of the bonded region from the macroscopic to the atomic level are also presented.

2. Experiments

The specimens used in bonding were 1.1-at. % Nd:YVO₄ and non-doped YVO₄ crystals with a bonding surface dimension of 5 x 6 mm², 9 x 10 mm² in a-cut surface, and with a thickness of 0.5mm and 2mm, respectively. Concerning precise procedure for our bonding method, please refer to ref. 3.

3. Results and discussion

3.1. Transmitted wavefront distortion

Transmitted wavefront distortion was measured by a Fizeau interferometer (Zygo: GPI-XP). In order to devise accurate measurements with the system, the specimen was located in the narrow space between transmission and reference flat mirrors to reduce the interference fringe motion by the fluctuation of airflow. The accuracy of the system is 0.01 λ at 633 nm. Table 1 shows the transmitted wavefront of the specimens before and after bonding crystals. From the table, it is clear that each crystal has a small distortion, and the sum of these distortions was close to measured distortion of the bonded crystal. From the result, the bonded distortion was assumed on the order of marginal detection.

Table 1. Transmitted wavefront of crystals.

	Crystals	Distortion (P-V)
Before Bonding	YVO ₄	0.042 λ
	Nd:YVO ₄	0.045 λ
	YVO ₄	0.034 λ
After Bonding	Integrated Crystal	0.116 λ

λ=633nm

3.2. Magnified inspection at the atomic level

The bonded region was also inspected by a field emission TEM (Hitachi: H-9000NAR) equipped with an EDX (Noran: Voyager III M3100) at a magnification power of 4-million. In the measurements, additional fabrication using argon ion milling was required to prepare a specimen extracted from the bonded region. Thickness of the specimen was reduced to around 70 nm in the bonded area. A probe electron beam accelerated by 300 kV was used for TEM measurements. Fig. 1 shows the magnified image of the bonded region. From the measured lattice constant of 0.625 nm in this photo, it is recognized that the bonded interface is parallel to the crystal c-axis.

Although obscure parts of thickness 2 nm slightly appeared along the bonded interface, the atoms in the bonded region were well arranged with the same regularity as inside the crystal. It seemed that the obscure parts were based on the irregularity of the polished surfaces. In the bonding process, most of the irregularity is reduced by elastic deformation and diffusion of the atoms during the long heat treatment while these effects can be smaller for large irregular deformations.

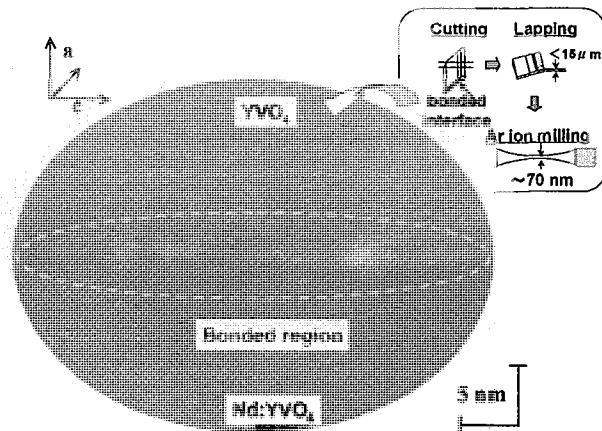


Figure 1 Magnified image taken by TEM.

In the quantitative analysis by EDX, the four elements of neodymium, yttrium, vanadium and oxygen that compose the crystal were selected and measured in spots of 5 nm diameter around the bonded region. As a reference, we also measured the EDX signal at points 50 nm and 1000 nm from the interface. Concerning oxygen, we neglected the measured signal since it was difficult to determine the quantity of the oxygen K_{α} spectra that overlap with the vanadium L_{α} spectra. From the

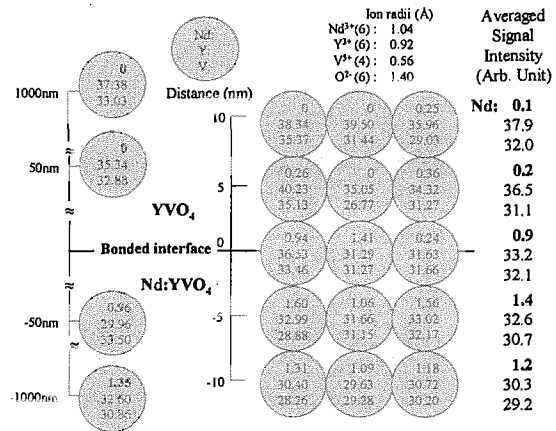


Figure 2 EDX signals of three elements at each point.

averaged signal intensities shown in the right column of Fig. 2, vanadium concentration is almost uniform, that is, it is independent of the distance from the interface. As V^{5+} ion has the smallest ion radius of the four-element ions composing the crystal, V^{5+} ions are transferable by thermal diffusion. However, each V^{5+} ion is surrounded by four O^{2-} ions which have the largest ion radii in the crystal. As a result, each V^{5+} ion would be trapped by O^{2-} ions and could not move in the crystal. Concerning Y^{3+} ions, although at a glance the concentration decreases towards the direction from YVO_4 to $Nd:YVO_4$, there is no clear evidence of the Y^{3+} ion migration by the diffusion because of the intrinsic tendency appearing in the comparison with reference signals at points 50 nm and 1000 nm from the interface. On the other hand, Nd^{3+} ions slightly move towards the YVO_4 side, which is assumed to be driven by the Gorsky effect caused in the thermal diffusion. Here, using the equation $L=(6tD)^{1/2}$ where the time duration of heat treatment (t) is in sec and the diffusion length (L) is in meters,^{4,5} the diffusion coefficient (D) is estimated to be $2.3 \times 10^{-23} \text{ m}^2/\text{sec}$ which is the same order of Al^{3+} ions in Al_2O_3 .⁶ In the low temperature heat treatment at 873 K, Nd^{3+} ions cannot migrate extensively.

3.3. Laser oscillation performance

Figure 3 shows a schematic diagram of the integrated and the normal $Nd:YVO_4$ lasers pumped by a fiber coupled CW laser diode array. The specimen was located in short cavity of length 30 mm. Each specimen coated on both surfaces was held in a copper block cooled by water of which temperature was controlled by a chiller unit at 20°C . In case of normal $Nd:YVO_4$, the crystal was cooled by heat conduction through the copper mount,

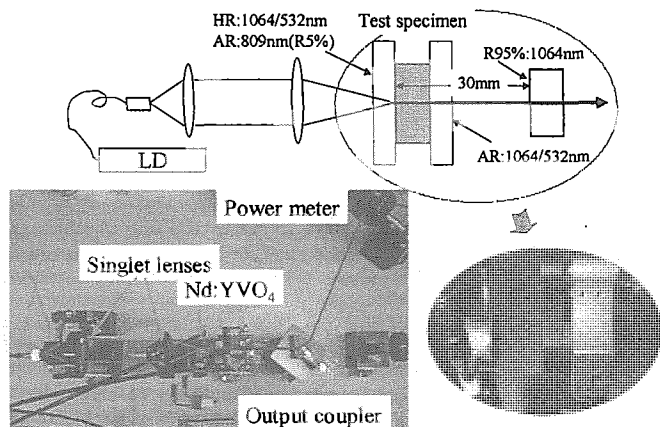


Figure 3 Experimental setup for laser oscillation.

therefore, we used metal adhesives that have a high thermal conductivity of 30~60 W/(m·K) to enhance the fast heat flow. In contrast, the crystal can be directly cooled by water in case of the integrated specimen which has water flow space between the two ends. We measured laser output power in these specimens for comparison. Fig. 4 shows the result of the measurements. From this figure, we cannot find a significant

difference in both output curves till the pump power reaches 10 W. However, in excess of 10 W, the normal crystal was broken by the strong thermal load caused by CW laser pumping. On the other hand, the output power of integrated one still increased, then saturated at the pump power of around 16 W. Laser output power of 3 W is around twice of the output in the normal crystal oscillation.

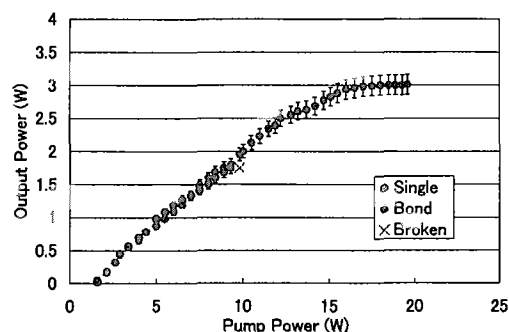


Figure 4 Laser output power of the integrated crystal.

4. Conclusion

Laser crystal integration of Nd:YVO₄ and YVO₄ was achieved by way of our newly developed direct bonding technique. In the preparation of contact, surface improvement was performed by using ion beam irradiation which is an appealing process for the extension of the bonding technique in other hygroscopic and fluoride crystals. The bonded region was evaluated in three different methods such as transmitted wavefront distortion measurements, magnified inspections and laser oscillation. From interferometer experiments, the wavefront distortion caused by the bonded region was assumed to be on the order of marginal detection. Magnified inspection showed that the atoms in the bonded region were well arranged with the same regularity as inside the crystal although obscure parts of thickness 2 nm slightly appeared along the bonded interface. In the EDX quantitative analysis, it was found that Nd³⁺ ion migration was slightly caused by atomic diffusion in the bonded region during heat treatment. The diffusion coefficient of Nd³⁺ ions in the YVO₄ crystal was estimated at 2.3×10^{-23} m²/sec. Experiments of laser oscillation suggested that the heat reduction capacity of the integrated crystal reached twice that of the normal single crystal. As a result, the integrated crystal could increase the laser output power in high power laser pumping.

REFERENCES

1. A. Sugiyama, H. Fukuyama, T. Sasuga, T. Arisawa and H. Takuma, "Direct bonding of Ti:sapphire laser crystals," *Appl. Opt.*, **37**, pp. 2407-2410, 1998.
2. A. Sugiyama, H. Fukuyama, Y. Kataoka, A. Nishimura and Y. Okada, "Feasibility study of a direct bonding technique for laser crystals," *SPIE.*, **4231**, pp. 261-268, 2000.
3. A. Sugiyama, H. Fukuyama, Y. Katsumata and Y. Okada, "Nd:YVO₄ and YVO₄ laser crystals integration by a direct bonding technique," *SPIE.*, **4944-46**, 2002.
4. Y. Fukai, "Physics of diffusion phenomena," Asakura Shoten, Tokyo, 1988 in Japanese
5. P. G. Shewmon, "Diffusion in solids," McGraw-Hill, New York, 1963, translated version in Japanese
6. T. Ando: in "Handbook of crystal growth," Eds., The Japanese Association of Crystal Growth, pp.881, Kyoritsu Shuppan, Tokyo, 1995 in Japanese