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NONLINEARITY AND ELASTIC PROPERTIES OF Cr(3+): Al₂O₃ AND Ti(3+): Al₂O₃ LASER CRYSTALS

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This paper reports the nonlinearity properties such as abbe-number (ν) , nonlinear refractive index (n_2) , nonlinearity refractive index coefficient (γ) and certain elastic properties of bulk, shear modulus, Poisson's ratio for the $Cr(3+):Al_2O_3$ and $Ti(3+):Al_2O_3$ laser crystals. Radiational influences from the Ar^+ laser and the UV sources on these materials, have also been studied.

1. INTRODUCTION

QUITE RECENTLY, a detailed investigation has been carried out on the absorption coefficients and non-radiative quantum efficiencies of ${\rm Ti}^{3+}$: sapphire crystals in different figures of merit [1]. The purpose of the present paper is to examine the non-linearity and elastic properties of ${\rm Cr}^{3+}:{\rm Al}_2{\rm O}_3$ and ${\rm Ti}^{3+}:{\rm Al}_2{\rm O}_3$ laser crystals.

2. EXPERIMENTAL

Crystals of Ti3+: Al2O3 and Cr3+: Al2O3 were grown by the Czochralski pulling technique from a molten mixture of Al₂O₃-Ti₂O₃ and Al₂O₃-Cr₂O₃. The starting materials (Al₂O₃; Ti₂O₃ and Cr₂O₃) used were 99.999% pure. The growth conditions were: pulling rate = $0.3 \,\mathrm{mm}\,\mathrm{h}^{-1}$, rotation rate = $5 \,\mathrm{r.p.m.}$, ambient atmosphere was purified argon, the seed was sapphire 90° (off C axis) orientation. The starting materials were melted by means of r.f. heating in an iridium crucible. Crystals were grown 2.5" in diameter and 10" long. The actual dimensions of the crystals that were used in the work: Cr^{3+} : Al_2O_3 (1.5 cm in diameter and 3 cm long); Ti³⁺: Al₂O₃ (1.5 cm in diameter and 2 cm long). A post growth treatment process was applied, based on earlier practice at Union Carbide. The grown crystals were annealed at 1850°C for 20 h in an atmosphere of 50% H₂ and 50% Ar. Some samples were cut from the boule following this treatment. These samples exhibited low figure of merit (FOM \approx 40). Several samples were then prepared from crystal subjected to further annealing in pure H_2 at 1940°C for 180 h. This type of treatment is known to improve the quality of Czochralski-grown $\text{Ti}^{3+}: \text{Al}_2\text{O}_3$, $\text{Cr}^{3+}: \text{Al}_2\text{O}_3$ [1]. As a result, the overall optical quality of the crystal improved, yielding a much higher figure of merit (FOM \approx 800). Finally, all samples were subjected to "low temperature" annealing at 1100°C for 12 h, following the "rough" fabrication (slicing and grinding). Thin and thick batches of samples were thus prepared.

The ultrasonic measurements were performed by a pulse echo technique with a Matec Pulse Modulator/Receiver Model 6600 with quartz transducers. An elastic pulse was generated by the piezoelectric transducer coupled to one of the sample faces, which was reflected on the opposite face and back detected by the same transducer. The pulse transit time (t) was measured with a Hewlett-Packard Oscilloscope Model 1741 A and the ultrasonic velocity, ν (m s⁻¹), was calculated by the equation,

$$\nu = 2L/t$$

where "L" is the sample length (in m) and "t" is the pulse transit time (in s). According to the transducer used, the longitudinal (ν_l) or transverse (ν_s) velocities were obtained.

Archimede's principle was the basis in measuring the densities of the crystals with the xylene as the immersion liquid. The densities were $4.850 \,\mathrm{g\,cm^{-3}}$ and $4.560 \,\mathrm{g\,cm^{-3}}$ for $\mathrm{Cr^{3+}}:\mathrm{Al_2O_3}$ and $\mathrm{Ti^{3+}}:\mathrm{Al_2O_3}$ laser crystals respectively. The refractive indices $(n_F$ at $\lambda=486.1\,\mathrm{nm})$ n_d at $\lambda=589.3\,\mathrm{nm}$, n_c at $\lambda=656.3\,\mathrm{nm}$) have been measured on a precision refractometer and the results are verified and found comparable with the measured reflectance spectra in the wavelength range of $400-700\,\mathrm{nm}$.

3. RESULTS AND DISCUSSION

From the measured refractive indices (n_d) and densities (d) different other related physical properties of these crystals have been computed using the relevant formulae [2] and tabulated in Table 1.

In addition to the spectroscopic characteristics, non-linearity and elastic properties have also been considered necessary to investigate for the laser materials. Both the laser crystals of Cr³⁺: Al₂O₃ and Ti³⁺: Al₂O₃ are identified as the powerful optical systems among the several transition metal lasers. So, the theoretical parametrization has been carried out in understanding the optical dispersive power and certain acoustical (elastic) properties by measuring the refractive indices at three suitable wavelengths and the ultrasonic velocities. The procedures needed to evaluate the nonlinearity properties have been obtained from the literature [3-7] to assess the optical efficiency of the laser host materials. By employing the relevant formulae, the values of the abbe-number (ν_d) , nonlinearity refractive index (n_2) and its coefficient (ν) have

been evaluated and presented in Table 1, for the laser crystals studied.

In respect of the acoustical properties, the measured longitudinal and shear velocities have been used to determine the elastic characteristics such as longitudinal modulus (L), shear modulus (G), bulk modulus (B), compressibility (C), mean sound velocity (V_m) , Poisson's ratio (σ) and the Young's modulus (y) for the laser crystals by using the well known methods of evaluation given in different references of interest [8-13] and calculated elastic constants and are listed out in Table 2 for making a comparison between the two crystals. An observation of the results given in Table 1 shows that the refractive indices of the crystals are varying significantly depending upon the source of irradiation used. On the irradiation with the UV light (254 nm), the laser materials have shown greater enhancement compared with that of the laser beam (514 and 488 nm). In consequence of this, the nonlinearity properties $(n_2 \text{ and } \gamma)$ are found to be reducing significantly particularly with the UV source. Between the two crystals, the nonlinearity properties of the Ti³⁺: Al₂O₃ have been found to be relatively lower than that of the Cr3+: Al2O3 crystal.

In regard of the elastic properties, as given in Table 2, it is clear that the values of shear modulus, compressibility, mean sound velocity and Young's modulus are found better with the Ti³⁺: Al₂O₃ crystal. In summary, it could be stated that the Ti³⁺: Al₂O₃ is with all lower values in their nonlinearity properties. The notable influences caused due to the irradiation from the UV and the laser sources on these laser crystal materials are

Table 1. Refractive index and nonlinearity properties of Cr^{3+} : Al_2O_3 and Ti^{3+} : Al_2O_3

| • | $Al_2O_3: Cr^{3+}$ | | | | $Al_2O_3:Ti^{3+}$ | | | |
|--|--------------------|--------------------|--------------------|----------------------------|-------------------|--------------------|--------------------|----------------------------|
| | Unirra- diated | 488 nm exposure | 514 nm exposure | UV (254 nm) irradiation | Unirra- diated | 488 nm exposure | 514 nm exposure | UV (254 nm) irradiation |
| Refractive index at | 1.780 | 1.770 | 1.765 | 1.785 | 1.720 | 1.715 | 1.711 | 1.722 |
| $589.3 \text{nm} (n_d)$ | | | | | | | | |
| Refractive index at | 1.786 | 1.776 | 1.770 | 1.790 | 1.727 | 1.722 | 1.718 | 1.727 |
| 486.1 nm (n_F) | | | | | | | | |
| Refractive index at | 1.776 | 1.765 | 1.758 | 1.781 | 1.718 | 1.712 | 1.708 | 1.718 |
| 656.3 nm (n_C) | | | | | | | | |
| Abbe number (ν_d) | 78 | 70 | 64 | 87 | 80 | 72 | 71 | 80 |
| Nonlinear refractive | | | | | | | | |
| index $(n_2) \times 10^{-13}$ es | ı 1.761 | 2.019 | 2.280 | 1.514 | 1.461 | 1.688 | 1.706 | 1.468 |
| Nonlinear refractive | 0.414 | 0.479 | 0.541 | 0.355 | 0.356 | 0.412 | 0.418 | 0.357 |
| index coefficient $(\gamma) \text{ cm}^2 \text{ W}^{-1}$ | | | | | | | | |
| Reflection loss $(R\%)$ | 7.8 | 7.7 | 7.7 | 7.9 | 7.0 | 6.9 | 6.9 | 7.0 |

Table 2. Certain elastic properties of Cr³⁺: Al₂O₃ and Ti³⁺: Al₂O₃ laser crystals

| Properties | $Cr^{3+}:Al_2O_3$ | Ti ³⁺ : Al ₂ O ₃ | |
|---|-----------------------|---|--|
| Longitudinal velocity (ν_1) m s ⁻¹ | 5960 | 6100 | |
| Longitudinal velocity (ν_1) m s ⁻¹ Shear velocity (ν_s) m s ⁻¹ | 2850 | 3045 | |
| Longitudinal modulus $(L) \times 10^{-4} \mathrm{kg}\mathrm{m}^{-2}$ | 17228 | 16968 | |
| Longitudinal modulus $(L) \times 10^{-4} \text{ kg m}^{-2}$ Shear modulus $(G) \times 10^{-4} \text{ kg m}^{-2}$ | 3939 | 4228 | |
| Bulk modulus $(B) \times 10^{-4} \mathrm{kg} \mathrm{m}^{-2}$ | 11976 | 11330 | |
| Bulk modulus $(B) \times 10^{-4} \text{ kg m}^{-2}$ Compressibility $(C) m^2 \text{ kg}^{-1}$ | 8.35×10^{-9} | 8.826×10^{-9} | |
| Mean sound velocity (V_m) m s ⁻¹ | 3205.0 | 3416.2 | |
| Poisson's ratio (σ) | 0.351 | 0.334 | |
| Young's modulus (Y) Nm ⁻² | 1.064×10^{8} | 1.128×10^8 | |

clearly presented in Tables 1 and 2. Because of the fact that the UV-source has got greater power (1000 W), therefore significant variations could be obtained as compared to that of the 1.5 W laser source even after three hours of irradiation. Hence, the UV-source has become a powerful source in enhancing the optical efficiencies of the laser materials reasonably by lowering their nonlinearity properties upon the irradiation.

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