FTD-ID(RS)T-0695-91

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FOREIGN TECHNOLOGY DIVISION

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by

Li Yunkui, Tang Honggao, et. al.

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HUMAN TRANSLATION

FTD-ID(RS)T-0695-91 19 November 1991

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English pages: 7

Source: Zhonggou Jiguang, Vol. 17, Nr. 12, 1990
pp. 750-753

Country of origin: China
Translated by: SCITRAN
F33657-84-D-0165
Requester: FTD/TTTR/Lt. Cason
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SPECTRAL PROPERTIES OF A NOVEL LASER CRYSTAL $Y_3(In,Ga)_2Ga_3O_{12}:Cr^{3+}$

XINXING JIGUANGJINGTI $Y_3(In,Ga)_2Ga_3O_{12}:Cr^{3+}$

By: Li Yunkui, Tang Honggao, Hang Yin, Chen Shuchun

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Abstract: Spectral properties of a novel phonon-terminated laser crystal $Y_3(In, Ga)_2Ga_3O_{12}:Cr^{3+}$ grown by the flux method are reported for the first time. The results show that the spectral properties of this novel crystal are compatible with those of $Gd_3(Se,Ga)_3Ga_3O_{12}:Cr^{3+}$ and is a potential ambient temperature tunable laser crystal.

INTRODUCTION

$Gd_3(Sc,Ga)_2Ga_3O_{12}:Cr^{3+}$ (shortened to GSGG:CR^{3+}) is a type of phonon-terminated laser crystal with excellent capabilities[1]. It has a relatively weak crystal field and relatively strong electron - phonon coupling. At room temperatures a strong terminal phonon emission spectrum with a half width of about 100 nm can be observed. At the same time, experimentally at room temperatures, a wide band continuous tunable laser emission has been observed[2]. Since it has been reported, a great deal of attention has been paid to it. However, since Scandium is rare and expensive its applications are limited. The authors used Indium which has a similar ion radius in place of the Scandium to grow $Gd_3(In,Ga)_2Ga_3O_{12}:Cr^{3+}$ (Shortened to GIGG:Cr^{3+}) monocrystals. It has the same crystal field strength as GSGG:Cr^{3+}, but it has a Stokes Frequency Shift and a fluorescence bandwith less than that of GSGG:Cr^{3+}, with results less than ideal[3].

This operation used Y and In to replace Gd and Sc respectively, and used the flux method to grow $Y_3(In,Ga)_2Ga_3O_{12}:CR^{3+}$ monocrystals (shortened to YIGG:Cr^{3+}. From the room temperature absorption light spectrum, the fluorescent light spectrum and the time discrimination spectrum we determined some of the crystal field parameters and the duration of fluorescence. The results show that the YIGG:Cr^{3+} crystal spectrum

capabilities are clearly superior to those of GIGG:Cr^{3+}, and are comparable with those of GSGG:Cr^{3+}. It is a type of ambient temperature phonon-terminated tunable laser crystal with excellent prospects.

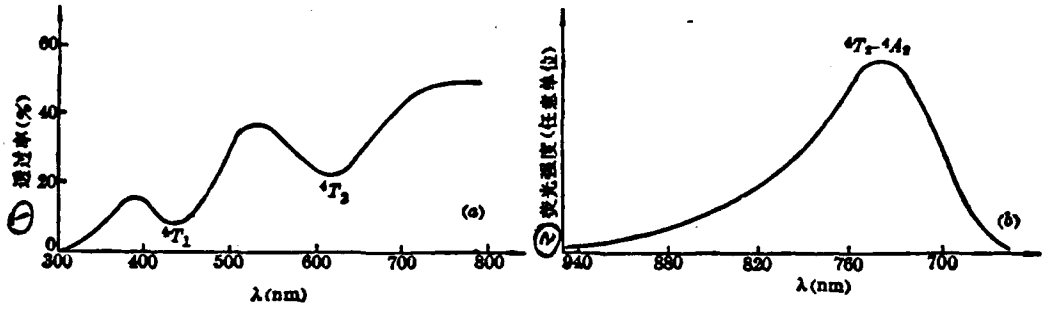
YIGG:Cr^{3+} monocrystals are grown using the flux method. For flux we used PbF and PbO. We also used a small amount of PbO_2 in order to protect the platinum crucible. The materials and the flux were mixed in the following rates: Y_2O_3 6 mol%, In_2O_3 4 mol%, Ga_2O_3 10 mol%, PbF_2 40 mol%, PbO 40 mol%, Cr_2O_3 1 mol%. The purity of the materials were all 4N, and the crystals were grown under gallium rich conditions. The automatic temperature control system was composed of a JWC-802A precision temperature digital programmed timer and a DWT-702. The growth sequence was as follows: The ambient temperature was raised to 800 C at a rate of 100 C/h, then it was raised to 1300 C at a rate of 200 C/h and held there for ten hours. Then it was dropped by 2 C/h until it reached 1280 C. When the crystal temperature was between 1280 and 1100 C, the temperature was dropped 1 C/h. Finally it was dropped by 1.5 C/h to 800 C. Then the furnace was shut down and the crystals were allowed to cool down to room temperature naturally. The crucible was removed from the furnace and 1:1 immersed for several days in a diluted nitric acid solution to separate the crystals. The grown crystals reached sizes up to 5 X 5 X 5 mm^3. They were greenish black in color, transparent, and of excellent quality.

SPECTRUM ANALYSIS

Illustration one (a) and (b) show the ambient temperature absorption spectrum and fluorescent spectrum of YIGG:Cr^{3+} crystals.

The absorption spectrum was measured using the Japanese UV210 spectrophotometer. It is made up of two bands, one of which has a peak wave length of 620nm $[^4A_2 \rightarrow {}^4T_2(t_2^2e)]$ and the other has a wave length of 440 nm $[^4A_2 \rightarrow {}^4T_1(t_2^2e)]$, there were no clear sharp lines within the spectrum.

ILLUSTRATION ONE: THE AMBIENT TEMPERATURE ABSORPTION SPECTRUM (A) AND AMBIENT FLUORESCENCE SPECTRUM (B) OF YIGG:Cr^{3+} CRYSTALS



(1). Penetration (%)

(2). Strength of fluorescence (aroitrary units

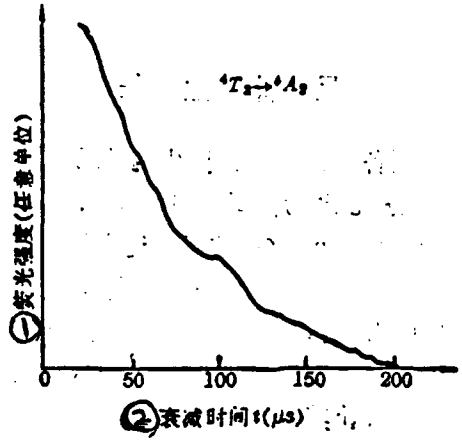
The fluorescence spectrum was stimulated by using a 1.06mm YAK:nd^{3+} pulse laser and a KDP frequency multiplier. The fluorescence produced was passed through a 1 m grate monochromater and then received by a photoelectric multiplier tube. The signal was processed using a Boxcar sampling integrator. Another photoelectric cell was used to receive the laser light as a reference signal to be input into the Boxcar channels. The sampling integrator and the laser trigger were synchronous, and the pulse laser repeat frequency was 7 per second. The fluorescence spectrum (at ambient temperature) observed is shown in illustration one (b). The range of fluorescence was 635 to 950 nm. The peak value wave length was 745 nm. and the half width was 100 nm (between 700 and 800 nm). There were no sharp peaks over the entire width of the fluorescence spectrum. The R line did not appear. This demonstrates that the YIGG:Cr^{3+} crystal as weak field medium, with a fluorescence transition of .

We can find from the absorption spectrum and the fluorescence spectrum that the Stoke's frequency shift is

The duration of fluorescence of the YIGG:Cr^{3+} crystal was calculated at the middle wavelength of fluorescence - 745nm through observing the fluorescence attenuation curve. We can see from illustration two that the attenuation was basically an exponential type. The ambient temperature

calculated was 56ms.

ILLUSTRATION TWO: YIGG:Cr^{3+} CRYSTAL AMBIENT TEMPERATURE ATTENUATION CURVE



(1). Strength of fluorescence (arbitrary units)

(2). Attenuation time t(ms)

CALCULATING THE CRYSTAL FIELD PARAMETERS

With O_h crystal field approximation, by using the Tanabe - Sugano energy matrix[4] it is possible to obtain the crystal field strength parameter D_q and the Racah parameters B and C. D_q is given in the following formula: $10D_q = E_a[^4T_2(t_2^2e)] - E_a[^4A_2(t_2^3)]$

The right side of this formula expresses the energy level position of the peak $^4T_2(t_2^2e)$ in the absorption spectrum as a wave number. With $\lambda[^4T_2(t_2^2e)]$ = 620 nm we obtain the corresponding D_q = 1613cm^{-1}. Racah parameter B can be obtained from the following formula[5]:

$$11D_q + \frac{15}{2}B = E_a[^4T_1(t_2^2e)] - E_a[^4A_2(t_2^3)]$$

Furthermore, it is not necessary to do any additional fitting. Substituting the $^4T_1(t_2^2e)$ peak wave length 44nm (22727cm^{-1}) and D_q = 1613 cm^{-1}, we obtain B = 665 cm^{-1}.

Because the R line field does not appear in the absorption spectrum and the fluorescence spectrum, there is no way to determine the C value. The C parameters and the fit of the crystal field energy levels will have to be determinec later with low temperature parameter measurement.

However, it is possible to use the conclusion proposed in footnote [6] that for gallium garnets, C is generally around 5B. Using rough calculations, we get C is around 3325cm^{-1}.

DISCUSSION

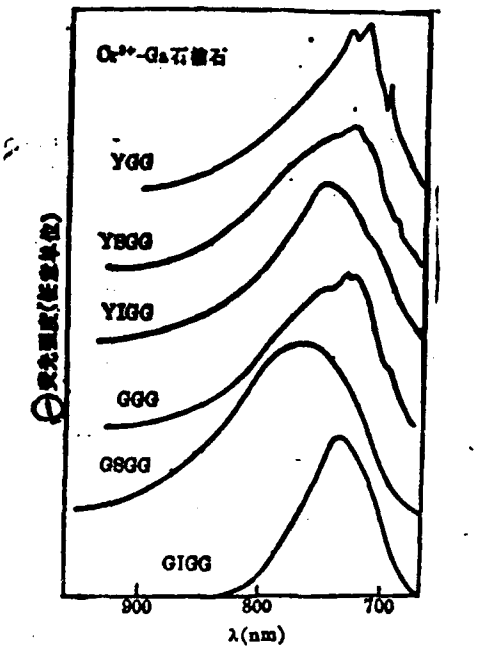
For the sake of clarity, we have listed spectral parameters of gallium garnets in table one[1,3] and the fluorescence spectrum in illustration three[3,7].

TABLE ONE: Cr^{3+} DOPED GARNET SPECTRAL PARAMETERS

	YGG	YSGG	YIGG	GGG	GSGG	GIGG
$E_a(^4T_2)$[cm^{-1}]	16300	16130	16129	15970	15625	15625
$E_a(^4T_1)$[cm^{-1}]	22730	22470	22727	22270	21980	21978
$E_0(^4T_2)$[cm^{-1}]	15100	14790	14775	14740	14480	14628
$E_f(^4T_2)$[nm]	740	760	745	760	760	734
a[Å]	12.277	12.476	12.459	12.376	12.567	12.566
D_q[cm^{-1}]	1630	1613	1613	1597	1563	1563
B[cm^{-1}]	639	630	665	626	638	639
D_q/B	2.55	2.53	2.43	2.55	2.45	2.45
ΔE_s[cm^{-1}]	2400	2680	2706	2460	2290	2000
τ[μs]	240	145	56	160	120	125

From table one we can see that for YIGG:Cr^{3+} crystals, D_q/B = 243, the least of all listed materials. Therefore, the crystal strength is weaker than all the other crystals. According to the criterion proposed in footnote [6], the YIGG:Cr^{3+} zero photon line spacing is about zero, and this is also shown by the relatively short fluorescence duration.

ILLUSTRATION 3: Cr^{3+} DOPED GALLIUM GARNET FLUORESCENCE SPECTRUM (300K)



(1). Intensity of fluorescence (In arbitrary units)

The crystal lattice constant of YIGG:Cr^{3+} is clearly greater than that of YGG:Cr^{3+}, and is close to that of YSFF:Cr^{3+}. This is because the ion diameter of In^{3+} and Sc^{3+} (0.080 nm and 0.075 nm) are somewhat larger than the that of Ga^{3+} (0.062 nm). This is also a direct factor leading to the weakened crystal field strength of YIGG:Cr^{3+}.

YIGG:Cr^{3+} crystal range and half width of fluorescence are similar to those of GSGG"Cr^{3+} and its Stoke's frequency shift is larger. If an average phonon frequency of for YGG:Cr^{3+} is used as an estimate, the Huangkun - Lisi (translator's note: phonetic) factor of YIGG:Cr^{3+} may be as high as 7.3, showing that it has a very strong electron-phonon coupling. The wide band fluorescence of YIGG:Cr^{3+} means it may be possible for this crystal to be a continuous tunable laser crystal in the 700 to 800 nm band.

FOOTNOTES.

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(4). Y. Tanabe *et al.*, *J. Phys. Soc.Japan*, **9**(5), 753(1954)

(5). Tang Honggao, Li Yunkui, et al, "Journal of Optics", 6 (2), 155

(1986).

6. Li Yunkui, "Journal of Optics", 8 (4), 259 (1988).

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