Yb:Lu₂O₃ Hydrothermally-Grown Single-Crystal and Ceramic Absorption Spectra Obtained Between 298 and 80 K

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Abstract

The hydrothermal growth, doping, and low temperature spectral characterization of Yb doped Lu₂O₃ was investigated. The absorption of the lutetia-based sesquioxide laser material Yb:Lu₂O₃ at temperatures of 80, 150, 200, 250, and 298 K, in the wavelength range of 850-1100 nm are reported. Data for both single crystal and ceramic Yb:Lu₂O₃ were obtained. The resulting absorption cross-section data will enable the further evaluation of Yb:Lu₂O₃ as a very promising high power cryogenic laser material.

Keywords: Yb:Lu₂O₃, Sesquioxide, Lutetia, Lasers, Absorption, Spectroscopy, Ytterbium

1. Introduction:

The sesquioxide laser materials Yb:Sc₂O₃, Yb:Y₂O₃, and Yb:Lu₂O₃ have undergone rapid development in the past fifteen years. Good summaries of the early work may be found in the dissertations of E. Mix, L. Fornasiero, and V. Peters [1-3], as well as in two other recent dissertations [4,5] and a number of recently published papers [6-9].

In order to mitigate the difficult high-temperature growth (ca. 2500 °C) of sesquioxide single crystals, sesquioxide ceramic laser materials have also been prepared, investigated, and used in laser demonstrations [10-12]. Recently, measurements of important thermal, mechanical, and thermo-optic parameters of the sesquioxides at room and cryogenic temperatures, including the specific heat, thermal conductivity, thermal expansion coefficient, and thermo-optic coefficient have begun to appear in the literature [13-16].

As described in a recent paper, our group has demonstrated the much lower temperature (≤ 700 °C) hydrothermal growth of Yb:Lu₂O₃ [13,17], in a temperature regime in which defects, impurities, crystal strain, color-centers, and inhomogeneities are minimized. A comprehensive review of the hydrothermal growth process and its application to the development of new and promising laser crystals has also recently been published [18]. In earlier work [17] we presented the first room temperature absorption cross-section data for hydrothermally-grown Yb:Lu₂O₃, as well as the emission cross-sections determined by the method of reciprocity. The sesquioxide laser material Yb:Lu₂O₃ is particularly attractive as a material for high power lasers due to its moderate emission cross-section, ability to be optically-pumped at the low quantum defect ($\approx 5.05\%$) zero-phonon line near 977 nm using powerful, efficient laser diode sources, and the close mass-match between Yb and Lu, leading to reduced impurity phonon scattering and a good thermal conductivity that is nearly independent of Yb doping level. Given the importance of reducing waste heat in extremely high-power lasers, it is essential to obtain a detailed understanding of the absorption event in pumping the laser crystals. In particular it is beneficial to minimize the quantum defect as this represents an inherent loss of efficiency and produces waste heat. In order to fully exploit the advantages of Yb³⁺ as a laser ion, cryogenic operation is most desirable. The recent development of efficient volume Bragg gratings and high power pump diodes operating at or near 976 nm makes zero-phonon line pumping technically feasible. These developments necessitate the detailed understanding of cryogenic absorption behavior of the Yb³⁺ ion doped in a Lu₂O₃ host.

In this paper we present new absorption cross-section experimental data from room temperature to 80 K, obtained from a single crystal of Yb:Lu₂O₃, also grown by the hydrothermal method. A complementary set of emission cross-section measurements will appear in a separate future publication. This paper describes the crystal growth and

preparation, the absorption cross-section data over a wide range of temperatures, calculation of the Yb density, and as well as a comparison to Yb:YAG.

2. Crystal Growth and Polishing:

Crystal growth from hydrothermal solution took place in closed, pure silver reaction tubes which contained the powdered feedstock along with a small amount of aqueous mineralizer to aid in dissolution during the growth cycle. In the case of Yb:Lu₂O₃ discussed here, rare-earth oxide powders (HEFA Rare Earth, 99.997%) of Lu₂O₃ and Yb₂O₃ were measured in the desired doping ratio such that the total powdered material was approximately 1.5 g and added directly to ¼" O.D. x 7" long silver tubes that had been carefully cleaned, weld-sealed, and inspected for integrity. The powdered feedstock, 3 mL of a 20M aqueous KOH solution, was added before the tubes were welded closed.

These loaded tubes were themselves inserted into Inconel autoclaves equipped with two external, individually-controlled ceramic band-heaters with temperature probes and pressure gauge assemblies in order to monitor temperatures and pressure throughout the entire growth process. Water added inside the autoclaves provides the counter pressure necessary to prevent bursting of reaction tubes when heated. The fully loaded and prepared autoclaves were then lowered into insulating pits and covered up to the pressure gauge in vermiculite. A temperature gradient was established with the band heaters of 670°C for the top, growth zone, and 700°C in the lower, dissolution zone, for this prograde solubility system. These temperatures typically resulted in pressures of around 207 Mpa (30,000 psi). The autoclaves were left undisturbed in these insulated pits during the entire growth run of between 12 and 14 days. After this time the heat was discontinued and autoclaves were left to cool naturally over several hours or quick-quenched by an air stream. The cooled reaction tubes were then carefully cut open to reveal various-sized single crystals of up to several mm on a side. While the temperature used in the present study represents a seemingly modest increase over our previous work [13], we observe improved faceting and a greater amount of optically clear regions in the as-grown crystals, compared to those grown at 630-650 °C. We postulate that moving the growth conditions farther away from the Lu₂O₃/LuO(OH) phase transition around 600 °C may be the source of such positive effects.

Elemental analysis by EDX (Energy Dispersive X-Ray Analysis) indicated only the presence of Lu, Yb and O. No spectroscopic abnormalities were observed to suggest the presence of other impurity ions at an appreciable level. Crystals determined to be of sufficient optical quality for spectral analysis were further prepared, if necessary, by polishing. Progressive lapidary discs were used starting with 1200 grit and ending with 100,000 mesh diamond spray.

3. Crystal Spectral Characterization:

3.1. Lu₂O₃ Crystal Properties

Property	Value
Crystal Structure	Bixbyite
Transparency Band Gap (eV)	5.8
Chemical Formula	Lu ₂ O ₃
Dopant	Yb ³⁺
Name	Lutetia
Crystal System	Body-Centered Cubic
Optical Class	Anaxial (Isotropic)
Space Group	Ia $\bar{3}$
Number of Unique Cation Sites	2
Local Point Group Symmetry C ₂	Non-centrosymmetric
Local Point Group Symmetry C _{3i}	Centrosymmetric
Lattice Parameter (Å)	10.391
Unit Cell Volume (Å) ³	1125.84
Number of Formula Units Per Cell, Z	16

Total Number of Cation Sites Per Unit Cell	32
Number of C ₂ Sites Per Unit Cell	24
Number of C _{3i} Sites Per Unit Cell	8
Coordination Number	6
Lu Ion Density (ions/cm ³)	2.85 x 10 ²²

Table 1: Lu₂O₃ Crystal Properties

Lutetia is one of three recently developed isomorphous sesquioxide host crystals: Lu₂O₃, Y₂O₃, and Sc₂O₃. These crystals have been primarily developed to provide possible alternatives, with more favorable physical properties, to the well-developed classic oxide host YAG (Y₃Al₅O₁₂). As shown in Table 1, where for convenience we provide a summary of the crystal properties, Lu₂O₃ is an optically isotropic (anaxial) host that provides a strong, robust, easily doped host for rare-earth ions, which substitute into Lu sites of the body-centered Lu sites. Unlike YAG, however, which has one substitutable rare earth site, Lu₂O₃ has two six-coordinate lutetium sites, with local point group symmetries of C₂ and C_{3i}. The former is non-centrosymmetric and has no inversion center, while the latter is centrosymmetric with a center of inversion. Thus, according to the LaPorte rule, electric dipole transitions are allowed in the C₂ site, while for the C_{3i} site such transitions are forbidden. Indeed, this conclusion is found to be obeyed experimentally, where C₂ radiative sites predominate while the C_{3i} transitions are very weak. The Lu₂O₃ unit cell has been found to contain 32 cations, with 24 being found in the C₂ sites and 8 in the C_{3i} sites. Thus the ratio of C₂ to C_{3i} sites found in the unit cell is 3:1.

3.2. Spectrophotometer and Cryostat Descriptions

Absorption cross-section results for the laser material Yb:Lu₂O₃, were measured at 80, 150, 200, 250 and 298 K. Data was obtained using a 1.42 mm thick hydrothermally-grown Yb:Lu₂O₃ crystal with a nominal Yb doping of 2 at-%, and with polished faces. Absorption was measured using a Shimadzu SolidSpec-3700 DUV high-resolution spectrophotometer that enables optical absorption measurements from 175 to 3300 nm. The spectrometer was modified to incorporate a custom cryogenic optical cell with liquid nitrogen boiloff gas cooling that allows operation from 77 K to room temperature, controlled by suitably adjusting the gas flow. The cell is evacuated to less than 5 x 10⁻⁴ torr to avoid water vapor condensation on the crystal, and incorporates two uncoated SiO₂ windows for transmission measurements. The Yb:Lu₂O₃ crystal was heat sunk using indium foil and the temperature measured close to the crystal using an RTD sensor with a LakeShore Model 218 temperature monitor.

3.3. Calibration of Yb Density

To determine the Yb doping density in our Yb:Lu₂O₃ crystal, we adopted the approach of Kuhn et. al. [20], who showed that by combining the Fuchtbauer-Ladenburg equation for calculating the emission cross-section with the reciprocity method, the ion number density N may be calculated from the equation

$$N_d = 8\pi c \tau \cdot \left(\frac{Z_l}{Z_u} \right) \cdot \int_{\lambda_2}^{\lambda_1} \frac{n(\lambda)^2 \alpha(\lambda) e^{-\frac{\Delta E(\lambda)}{kT}}}{\lambda^4} d\lambda, (1)$$

Here, c is the speed of light, τ the Yb fluorescence lifetime in Lu₂O₃, Z_l and Z_u the lower and upper level partition factors, $\alpha(\lambda)$ the absorption coefficient at the wavelength λ , and $\Delta E(\lambda)$ is the energy difference between a photon at wavelength λ and the zero-phonon line energy value E_{ZL} corresponding to the zero-phonon wavelength λ_{ZL} . λ_1 and λ_2 are the starting and ending wavelength values of the integration. Formally, $\Delta E(\lambda)$ is written

$$\Delta E(\lambda) = E_{ZL} - \frac{hc}{\lambda} = \frac{hc}{\lambda_{ZL}} - \frac{hc}{\lambda}. (2)$$

The partition factors, Z_l and Z_u for the ground state $^2F_{7/2}$ and the excited state $^2F_{5/2}$ manifolds respectively, are calculated with respect to the lowest energy level in each manifold. For the calculation of the number density, we used the energy level assignments of Peters [3], shown in Figure 1. The levels are labeled according to the (i,j) convention, where i represents the manifold number (1 for lower, 2 for upper), and j the energy level number in the

manifold, with the lowest value being at the bottom and the highest at the top. The energy of each level is expressed in wavenumbers.

Figure 1: Yb:Lu₂O₃ Energy Levels

Using the data presented in the following Section III (d), we calculated the Yb number density with the results shown in Table 2. The Yb number density calculated from Eq. (1) yields the number of radiative sites with C₂ symmetry. Here we assume that 75 % of all Yb sites are C₂. By using the calculated Lu number density, we determine a C₂ at-% doping of 1.65 at-%. We also show the calculated density of C_{3i} non-radiative sites, and the total number density, which gives a total Yb doping of 2.2 at-%. Before crystal growth we estimated an at-% Yb doping of 2.0 % based on the mole percentages of the starting materials.

Quantity	Number Density (cm ⁻³) or at-%
Calculated Number Density of Radiative C ₂ Sites	4.69 x 10 ²⁰
Lu Ion Number Density in Lu ₂ O ₃	2.85 x 10 ²²
Calculated at-% Radiative C ₂ Sites	1.65
Calculated Number Density of C _{3i} Sites	1.57 x 10 ²⁰
Total Number Density of C ₂ + C _{3i} Sites	6.26 x 10 ²⁰
Total Calculated at-% Yb	2.20
Estimated at-% Doping From Synthesis	2.00

Table 2: Calculated Number Densities and at-% Values For Yb:Lu₂O₃ Crystal

3.4. Yb:Lu₂O₃ Absorption Cross-Section Data

We begin by showing in Fig. 2 the obtained cross-section data for absolute temperatures of 80, 150, 200, 250, and 298 K. This data set was generated using a hydrothermally-grown Yb:Lu₂O₃ crystal; the optical pathlength was 1.42 mm, the resolution 1.0 nm, and the scan speed 1.67 nm/s. The data have been corrected for Fresnel losses using the Sellmeier equation found in Peters [3]. The spectra show the normal cross-section groupings associated with the (1,1) – (2,3) transition near 903 nm, the (1,1) – (2,2) transition near 950 nm, and the (1,1) – (2,1) zero-phonon transition at about 977 nm. All transitions are associated with the allowed C₂ site, although as we will later see the

zero-phonon band also contains a zero-phonon transition contribution attributed to the C_{3i} Laporte forbidden site. Lattice phonon coupling is evident, particularly in the short wavelength shoulders of the 950 and 977 nm transitions, and to a lesser extent in the longer wavelength shoulder of the 903 nm transition. The phonon transitions become more resolved at lower temperatures; for the 977 nm zero-phonon line the short wavelength shoulder diminishes significantly in amplitude as temperature is lowered while the peaks become better resolved. This feature has also been found in Yb:YAG, where the lattice coupling is less pronounced [21,22]. The value of the zero-phonon peak cross-section at 977.5 nm and room temperature, about $3.8 \times 10^{-20} \text{ cm}^2$, compares favorably with our previous cross-section value of $3.3 \times 10^{-20} \text{ cm}^2$ [17].

Table 3 shows the cross-section amplitudes as a function of absolute temperature for five absorption peaks located at 903, 936.3, 951.7, 977.5, and 1034.5 nm. The 936.3 nm peak corresponds to a phonon line, 1034.5 nm to the emission wavelength, and the other three wavelengths to the (1,1) – (2,1), (2,2), and (2,3) transitions.

Figure 2: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 150, 200, 250, and 298 K

Temperature (K)	903.0 nm	936.3 nm	951.7 nm	977.5 nm	1034.5 nm
298	1.074	0.748	1.516	3.807	0.141
250	1.178	0.790	1.784	4.293	0.128
200	1.324	0.848	2.252	4.287	0.070
150	1.424	0.919	2.585	3.555	0.048
80	1.495	1.020	2.179	1.305	0.041

Table 3: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For Selected Absorption Lines

Figure 3 below shows a plot of the (1,1) – (2,3), (1,1) – (2,2), and (1,1) – (2,1) transitions, corresponding to the wavelengths of 903.10, 951.45, and 977.70 nm transitions of Table 4. For the (1,1) – (2,3) transition at 903 nm, we see that the bandwidth narrows and the amplitude increases as temperature is lowered. This same behavior is seen in Yb:YAG and Yb:LuAG [21-23]. An increase in amplitude is also seen for the (1,1) – (2,2) transition at 951.45, where the line becomes narrower and more intense as temperature decreases, with the exception of the data point at 80 K, which is anomalous. For the (1,1) – (2,1) zero-phonon line at 977.70 nm, however, the behavior becomes even more anomalous, increasing in amplitude and narrowing from 298 to 200 K, and thereafter decreasing in amplitude and continuing to narrow to the lowest temperature of 80 K. These examples of anomalous behavior are instrument related; the zero-phonon line at a temperature below about 150 K becomes narrower than can be resolved using our Shimadzu SolidSpec-3700 DUV spectrophotometer. While line narrowing continues to happen as temperature is lowered, the sharp zero-phonon peak linewidth cannot be resolved and appears as a line of reduced intensity. For Yb:YAG for example, we have shown in a previous publication that the zero-phonon line in Yb:YAG becomes very narrow at 175 K, and then narrows even further to less than 0.2 nm at 75 K [21,22]. A later publication confirmed this behavior and established the linewidth as 0.07 nm at 75 K [24]. Between room temperature and 75 K the intensity of the zero-phonon transition increases by a factor of ≈ 51 .

Figure 3: Yb:Lu₂O₃ Single-Crystal Peak Absorption Cross-Section Values For The Three (1,1) to Upper Manifold ((2,1), (2,2), (2,3)) Transitions, as a Function of Absolute Temperature

Table 4 shows the variation in the (1,1) – (2,1), (1,1) – (2,2), and (1,1) – (2,3) peak transition wavelengths as temperature varies from 298 to 80 K. We also show the wavelength difference between 298 and 80 K. The zero-phonon line (1,1) – (2,1) shifts towards shorter wavelengths by 0.30 nm as temperature is lowered. The smallest shift is -0.05 nm for the (1,1) – (2,2) transition, while the (1,1) – (2,3) transition shifts towards longer wavelengths by about -0.20 nm. These data are conveniently plotted in Figure 4 which reveals no discernable trends. Table 5 lists

the measured FWHM bandwidth values for the (1,1) – (2,1), (1,1) – (2,2), and (1,1) – (2,3) transitions, as a function of absolute temperature. The (1,1) – (2,3) transition at 903 nm shows narrowing as temperature is lowered, by about 2.5 nm. The (1,1) – (2,2) transition at about 952 nm also shows narrowing, by about 7.9 nm, and has a FWHM bandwidth of > 5 nm at 80 K. This width is suitable for efficient pumping using conventional 950 nm diode sources. The quantum defect using this absorption band is about 9.2 %; the same transition in Yb:YAG has a quantum defect of 9.1 %. For the (1,1) – (2,1) zero-phonon transition at 977.5 nm, the FWHM bandwidth also decreases, but by only 2.0 nm to 1.25 nm at 80 K. The reader should be cautioned, however, that these values are instrument limited, and that the real bandwidth is apparently much narrower.

Temperature (K)	(1,1) - (2,1) Transition	(1,1) - (2,2) Transition	(1,1) - (2,3) Transition
298.00	977.70	951.45	903.10
250.00	977.60	951.35	902.80
200.00	977.55	951.55	903.05
150.00	977.45	951.60	903.35
80.00	977.40	951.50	903.30
Difference Between 298 and 80 K (nm):	0.30	-0.05	-0.20

Table 4: Yb:Lu₂O₃ Single-Crystal Transition Center Wavelength Values (nm) as a Function of Temperature For Selected Absorption Lines

Figure 4: Yb:Lu₂O₃ Single-Crystal Transition Center Wavelength Values as a Function of Absolute Temperature, For The Three Transitions (1,1) – (2,1), (1,1) – (2,2), and (1,1) – (2,3)

Temperature (K)	(1,1) - (2,1) Transition	(1,1) - (2,2) Transition	(1,1) - (2,3) Transition
298	3.25	9.8	7.7
250	2.55	7.85	6.7
200	2.1	5.55	5.9
150	1.7	3.45	5.35
80	1.25	1.9	5.2
Difference Between 298 and 80 K (nm):	2	7.9	2.5

Table 5: Yb:Lu₂O₃ Single-Crystal Transition FWHM Bandwidth Values (nm) as a Function of Temperature For Selected Absorption Lines

Figure 5: Yb:Lu₂O₃ Single-Crystal Transition FWHM Bandwidth Values as a Function of Absolute Temperature, For The Three Transitions (1,1) – (2,1), (1,1) – (2,2), and (1,1) – (2,3)

The zero-phonon line anomalous absorption for single crystal Yb:Lu₂O₃ previously shown in Fig. 2 is also observed in ceramic Yb:Lu₂O₃. Figure 6 shows the obtained absorption cross-sections for a low-scattering ceramic Yb:Lu₂O₃ crystal obtained from Konoshima Chemical Company, Ltd. In Japan. This data set was obtained using a 0.36 mm thick ceramic crystal with a calculated total concentration ($C_2 + C_{3i}$ sites) of 10.8 at-% Yb obtained using the same method described in Section III, c), and with 1 nm resolution and a scan speed of 1.67 nm/s.

Figure 6: Yb:Lu₂O₃ Ceramic Absorption Cross-Sections at 86, 100, 150, 200, 250, and 298 K

Temperature (K)	977.5 nm
298	2.88
250	3.43
200	4.01
150	2.95
100	1.53
86	0.99

Table 6: Yb:Lu₂O₃ Ceramic Absorption Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For The (1,1) – (2,1) Zero-Phonon Line

Table 6 shows the obtained cross-section values at 977.5 nm. As observed previously with single-crystal Yb:Lu₂O₃, the cross-section values initially increase as temperature is lowered from 298 to 200 K, and thereafter decrease, in this case to a value of $0.99 \times 10^{-20} \text{ cm}^2$ at 86 K. To further confirm that our spectrophotometer is not resolving the zero-phonon linewidth at lower temperatures, we show in Figure 7 Yb:YAG absorption cross-sections obtained for a 4.87 mm thick Yb:YAG Czochralski grown crystal with 3.7 at-% Yb doping, at 86, 100, 150, 200, 250, and 298 K,

using the same resolution of 1 nm and scan speed of 1.67 nm/s. The doping density here was again obtained using the method of Section III, c). Table 7, which lists the 970.7 nm zero-phonon cross-section values as a function of temperature, shows that the Yb:YAG zero phonon line cross-sections displays the same behavior shown previously in Tables 3 and 6 for single-crystal and ceramic Yb:Lu₂O₃ respectively. Since we know from previous measurements [21, 22, 24] that at 80 K the Yb:YAG zero-phonon line has a width of 0.06 nm, it is clear that the line is not resolved in our present measurements, and as a consequence it is very likely that the Yb:Lu₂O₃ zero-phonon line has a very narrow linewidth at the same temperature.

Figure 7: Yb:YAG Single-Crystal Absorption Cross-Sections at 80, 150, 200, 250, and 298 K

Temperature (K)	970.7 nm
298	0.86
250	0.93
200	0.87
150	0.67
80	0.24

Table 7: Yb:YAG Single-Crystal Absorption Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For The (1,1) – (2,1) Zero-Phonon Line

3.5. Yb:YAG and Yb:Lu₂O₃ Narrower Band Absorption Measurements

In order to further elucidate the zero-phonon linewidth for Yb:Lu₂O₃, we have completed a supplementary set of absorption measurements with a resolution of 0.1 nm for Yb:YAG and resolutions of 0.5, 0.2, and 0.1 nm for single crystal Yb:Lu₂O₃. The measurement results are shown in Figure 8 for Yb:YAG and in Figures 9, 10, and 11 for Yb:Lu₂O₃. Table 8 summarizes the peak cross-section values of Fig's 8, 9, 10, and 11. For Yb:YAG with 0.1 nm resolution, it can be seen that the cross-section value still rolls over starting at about 200 K, and is of course still unresolved. The FWHM bandwidth of the line at 80 K is measured to be 0.68 nm, or about 11.3 times larger than the known value of 0.06 nm. For Yb:Lu₂O₃, the rollover begins at about 150 K for 0.5 nm and 0.2 nm, and at 0.1 nm the rollover begin at about 200 K. We thus see a trend towards a narrower and more intense line as the resolution becomes narrower, as expected. The FWHM linewidth at 80 K is about 0.55 nm. These results imply that the fully resolved minimum FWHM linewidth for Yb:Lu₂O₃ at 80 K will be very narrow, perhaps < 0.1 nm. In the near future we will re-measure the Yb:Lu₂O₃ zero-phonon bandwidth using a much higher resolution method and will report those measurements in a separate publication. Obtaining a higher resolution value for the zero-phonon bandwidth is important to determine whether or not volume-Bragg-grating (VBG) stabilized diode sources operating near 977.4 nm can efficiently optically-pump the transition at cryogenic temperatures, which would result in a quantum defect of only 5.5 %. It should be noted that the laser diode company DILAS recently demonstrated high power VBG stabilized 976 nm diode sources with a bandwidth of < 0.3 nm [25]. This new technology makes zero-phonon line pumping feasible and represents an important advance in high power laser development.

Temperature (K)	Yb:YAG 0.1 nm	Yb:Lu₂O₃ 0.5 nm	Yb:Lu₂O₃ 0.2 nm	Yb:Lu₂O₃ 0.1 nm
298	0.93	4.82	4.82	4.39
250	1.03	5.29	5.44	4.81
200	0.99	5.54	5.75	5.39
150	0.91	5.03	5.12	4.92
100	0.58	3.18	3.44	3.71
80	0.55	2.15	2.88	2.76

Table 8: Comparison Yb:Lu₂O₃ and Yb:YAG Single-Crystal Absorption Peak Cross-Section Values (x 10⁻²⁰ cm²) as a Function of Temperature For Zero-Phonon Line at Selected Resolution

Figure 8: Yb:YAG Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.1 nm Resolution. Sample path length is 4.87 mm and scan speed 0.83 nm/s.

Figure 9: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.5 nm Resolution. Sample path length is 1.42 mm and scan speed 0.83 nm/s.

Figure 10: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.2 nm Resolution. Sample path length is 1.42 mm and scan speed 0.83 nm/s.

Figure 11: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.1 nm Resolution. Sample path length is 1.42 mm and scan speed 0.83 nm/s.

Finally, it should be noted that in Figures 9-11, the observed absorption spectra manifest a short wavelength shoulder that becomes more pronounced as temperature is decreased, located in Figure 11 at 80 K at a wavelength of 976.21 nm. We propose that this secondary absorption peak is associated with the C_{3i} site in Yb:Lu₂O₃ and is a forbidden transition that has been observed at 975.6 nm previously at a temperature of 10 and 4 K [3,26]. This assignment is consistent with the (2,1) energy level value presented by Guyot et. al. [26] of 10264 cm⁻¹, or an absorption wavelength of 974.30 nm. Above 80 K, it has been observed that efficient energy transfer takes place from the C_{3i} to the C₂ site [3].

4. Discussion and Conclusions:

In this paper we present a comprehensive set of absorption cross-section data over a wide temperature range for hydrothermally grown Yb:Lu₂O₃. These data demonstrate it is a promising new sesquioxide laser material that will likely find wide applicability as a suitable host for high power lasers, especially for cryogenic operation. A complementary set of data for ceramic laser material Yb:Lu₂O₃ is also presented. The data, obtained with a resolution of 1.0 nm, span the temperature range from 298 K to 80 K, near the temperature of liquid nitrogen (77 K). We show that the 951.5 nm band associated with the (1,1) – (2,2) transition remains broad at 80 K and is suitable for optical pumping with efficient laser diode sources. The obtained data show anomalous absorption results for the (1,1) – (2,1) zero-phonon line located near 977.5 nm wherein the lowest temperature lines have a cross-section amplitude less than that obtained at room temperature, albeit with much narrower FWHM bandwidths. Higher resolution 0.5, 0.2, and 0.1 nm resolution data were also obtained and presented. Those data show clearly that much larger cross-section amplitudes are obtained as resolution is decreased, and that at 80 K the zero-phonon line has a width of < 0.55 nm, and that it is likely that the fully resolved linewidth is significantly narrower. By use of a higher

resolution monochromator, we are currently working to obtain such fully-resolved zero-phonon line absorption data and will present those results in a separate publication. The development of efficient volume Bragg gratings and new powerful diode pumps that operate efficiently in the 976 nm region make zero-phonon line pumping technically feasible. The enormous gains in energy efficiency and waste heat reduction make this very attractive for high power laser development so the detailed understanding of the pump line width, wavelength, and absorption cross-section are essential. Our cross-section data show clearly that the zero-phonon line contains substantial contributions from both the C_2 and C_{3i} sites, even at room temperature.

Our collaboration is now producing an associated set of hydrothermally-grown emission cross-section data as well, and those results will be presented in a separate publication in the near future.

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Captions

Table 1: Lu₂O₃ Crystal Properties

Figure 1: Yb:Lu₂O₃ Energy Levels

Table 2: Calculated Number Densities and at-% Values For Yb:Lu₂O₃ Crystal

Figure 2: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 150, 200, 250, and 298 K

Table 3: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For Selected Absorption Lines

Figure 3: Yb:Lu₂O₃ Single-Crystal Peak Absorption Cross-Section Values For The Three (1,1) to Upper Manifold ((2,1), (2,2), (2,3)) Transitions, as a Function of Absolute Temperature

Table 4: Yb:Lu₂O₃ Single-Crystal Transition Center Wavelength Values (nm) as a Function of Temperature For Selected Absorption Lines

Figure 4: Yb:Lu₂O₃ Single-Crystal Transition Center Wavelength Values as a Function of Absolute Temperature, For The Three Transitions (1,1) – (2,1), (1,1) – (2,2), and (1,1) – (2,3)

Table 5: Yb:Lu₂O₃ Single-Crystal Transition FWHM Bandwidth Values (nm) as a Function of Temperature For Selected Absorption Lines

Figure 5: Yb:Lu₂O₃ Single-Crystal Transition FWHM Bandwidth Values as a Function of Absolute Temperature, For The Three Transitions (1,1) – (2,1), (1,1) – (2,2), and (1,1) – (2,3)

Figure 6: Yb:Lu₂O₃ Ceramic Absorption Cross-Sections at 86, 100, 150, 200, 250, and 298 K

Table 6: Yb:Lu₂O₃ Ceramic Absorption Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For The (1,1) – (2,1) Zero-Phonon Line

Figure 7: Yb:YAG Single-Crystal Absorption Cross-Sections at 80, 150, 200, 250, and 298 K

Table 7: Yb:YAG Single-Crystal Absorption Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For The (1,1) – (2,1) Zero-Phonon Line

Table 8: Comparison Yb:Lu₂O₃ and Yb:YAG Single-Crystal Absorption Peak Cross-Section Values ($\times 10^{-20} \text{ cm}^2$) as a Function of Temperature For Zero-Phonon Line at Selected Resolution

Figure 8: Yb:YAG Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.1 nm Resolution. Sample path length is 4.87 mm and scan speed 0.83 nm/s.

Figure 9: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.5 nm Resolution. Sample path length is 1.42 mm and scan speed 0.83 nm/s.

Figure 10: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.2 nm Resolution. Sample path length is 1.42 mm and scan speed 0.83 nm/s.

Figure 11: Yb:Lu₂O₃ Single-Crystal Absorption Cross-Sections at 80, 100, 150, 200, 250, and 298 K With 0.1 nm Resolution. Sample path length is 1.42 mm and scan speed 0.83 nm/s.