OPTICAL PROPERTIES OF LITHIUM TERBIUM FLUORIDE AND IMPLICATIONS FOR PERFORMANCE IN HIGH POWER LASERS (POSTPRINT)

David E. Zelmon and Emily C. Erdman

AFRL/RX

Kevin T. Stevens, Greg Foundos, Joo Ro Kim, and Allen Brady

Northrop Grumman

16 October 2015
Interim Report

Distribution Statement A.
Approved for public release: distribution unlimited.

© 2016 OPTICAL SOCIETY OF AMERICA

(STINFO COPY)

AIR FORCE RESEARCH LABORATORY
MATERIALS AND MANUFACTURING DIRECTORATE
WRIGHT-PATTERSON AIR FORCE BASE, OH 45433-7750
AIR FORCE MATERIEL COMMAND
UNITED STATES AIR FORCE
**REPORT DOCUMENTATION PAGE**

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.

<table>
<thead>
<tr>
<th>1. REPORT DATE (DD-MM-YY)</th>
<th>2. REPORT TYPE</th>
<th>3. DATES COVERED (From - To)</th>
</tr>
</thead>
<tbody>
<tr>
<td>16 October 2015</td>
<td>Interim</td>
<td>6 May 2010 – 16 September 2015</td>
</tr>
</tbody>
</table>

**4. TITLE AND SUBTITLE**

OPTICAL PROPERTIES OF LITHIUM TERBIUM FLUORIDE AND IMPLICATIONS FOR PERFORMANCE IN HIGH POWER LASERS (POSTPRINT)

**6. AUTHOR(S)**

1) David E. Zelmon and Emily C. Erdman - AFRL/RX

2) Kevin T. Stevens, Greg Foundos, Joo Ro Kim, and Allen Brady – Northrop Grumman

**7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)**

1) AFRL/RX
Wright-Patterson AFB, OH 45433

2) Northrop Grumman Synoptics,
1201 Continental Blvd, Charlotte, NC 28273

**9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)**

Air Force Research Laboratory
Materials and Manufacturing Directorate
Wright-Patterson Air Force Base, OH 45433-7750

Air Force Materiel Command
United States Air Force

**12. DISTRIBUTION/AVAILABILITY STATEMENT**

Distribution Statement A. Approved for public release: distribution unlimited.

**13. SUPPLEMENTARY NOTES**

PA Case Number: 88ABW-2015-4958; Clearance Date: 16 Oct 2015. This document contains color. Journal article published in Applied Optics, Vol. 55, No. 4, 1 Feb 2016. © 2016 Optical Society of America. The U.S. Government is joint author of the work and has the right to use, modify, reproduce, release, perform, display, or disclose the work.

The final publication is available at http://dx.doi.org/10.1364/AO.55.000834

**14. ABSTRACT (Maximum 200 words)**

LiTbF4 has the potential to replace traditional magneto-optic (MO) garnet materials as a Faraday rotator in high power laser systems due to its high Verdet constant. New measurements are reported of the ordinary and extraordinary refractive indices of LiTbF4 as functions of wavelength and temperature respectively, as well as their corresponding Sellmeier expressions. Consequently, the Verdet coefficient was calculated and plotted as a function of wavelength and temperature. These measurements will aid in further development of LiTbF4 as an optical isolator.

**15. SUBJECT TERMS**

LiTbF4; magneto-optic (MO) garnet materials; Faraday rotator; high power laser; Verdet constant; Sellmeier; optical isolator

**16. SECURITY CLASSIFICATION OF:**

a. REPORT Unclassified
b. ABSTRACT Unclassified
c. THIS PAGE Unclassified

**19a. NAME OF RESPONSIBLE PERSON (Monitor)**

Steven Fairchild (937) 904-4328
Optical properties of lithium terbium fluoride and implications for performance in high power lasers

DAVID E. ZELMON,1,* EILEEN C. ERDMAN,1 KEVIN T. STEVENS,2 GREG FOUNDOS,2 JOO RO KIM,2 AND ALLEN BRADY2

1Materials and Manufacturing Directorate, AFRL/RXAP, 3005 Hobson Way, Wright-Patterson AFB, Ohio 45433-7707, USA
2Northrop Grumman Synoptics, 1201 Continental Blvd., Charlotte, North Carolina 28273, USA

*Corresponding author: david.zelmon@us.af.mil

Received 22 October 2015; revised 15 December 2015; accepted 21 December 2015; posted 22 December 2015 (Doc. ID 252482); published 1 February 2016

LiTbF4 has the potential to replace traditional magneto-optic garnet materials as a Faraday rotator in high power laser systems due to its high Verdet constant. New measurements are reported of the ordinary and extraordinary refractive indices of LiTbF4 as functions of wavelength and temperature, respectively, as well as their corresponding Sellmeier expressions. Consequently, the Verdet coefficient was calculated and plotted as a function of wavelength and temperature. These measurements will aid in further development of LiTbF4 as an optical isolator. © 2016 Optical Society of America

OCIS codes: (160.3820) Magneto-optical materials; (160.4670) Optical materials; (230.3810) Magneto-optic systems.

http://dx.doi.org/10.1364/AO.55.000834

1. INTRODUCTION

Magneto-optical materials have been the subject of intense study for several decades. The independence of the polarization rotation on propagation direction makes magneto-optic devices ideal for a variety of applications including switching, modulation, interferometry, imaging, biomolecular detection, and optical isolation [1–5]. Faraday rotators are the basis for optical isolation and light amplitude modulation. For example, the most popular and principal technique in high power laser systems is linear polarization rotation by Faraday elements (FEs) for laser output extraction of the system. FEs are also used as isolators in laser chains and birefringence compensation in a solid-state laser medium [6]. For FEs being used in high power laser systems, a large Verdet constant, small absorption, small scattering losses, and a small nonlinear refractive index are necessary. These properties are characteristic of low-dispersion fluoride hosts, such as alkali fluorides [7], making them excellent choices for FEs. However, as the power of laser systems grows larger, FEs are exposed to very high field densities which can alter their optical and physical properties due to nonuniform temperatures in the element. In order to compensate for these changes, the temperature dependence of basic optical and mechanical properties such as the refractive index and stress-optic coefficients must be known. In this paper, we describe the measurement of the refractive index and its dependence on temperature of LiTbF4.

2. EXPERIMENTS

Single crystals of LiTbF4 were grown at Northrop Grumman SYNOPTICS by the Czochralski technique, utilizing an inert gas resistance furnace. The system is incongruently melting, with several reported peritectic compositions. A melt containing 63 mol. % LiF, the peritectic composition reported by Weber [3], was chosen as the starting composition. The melt was prepared using 4–99% purity TbF3 and LiF, obtained from suppliers previously qualified by SYNOPTICS. A nitrogen atmosphere was employed for both the melting and growth at temperatures less than 900°C.

Crystals grown from the 63 mol. % LiF melt were just under 4 cm in diameter × 7 cm in length. These crystals had inclusions, as well as scattering centers (precipitates), but did contain areas of high quality material. Starting melt compositions (percent molar compositions) were varied systematically to improve crystal quality. However, more work is required to better understand the melt composition needed for high optical quality crystals.

The method of minimum deviation was used in order to obtain the refractive indices of the LiTbF4 crystal, using the Moller–Wedel divided circle spectrometer [8]. LiTbF4 is a tetragonal crystal with a 4/m point group and is therefore uniaxial. Triangular prisms of LiTbF4 were cut from the boule with the optic axis perpendicular to the triangular faces. In this way, the ordinary and extraordinary indices could be measured.
Temperature was also carried out by means of the method of minimum deviation. A type K thermocouple was mounted inside a small hole drilled into the nontransmitting face of the prism in order to monitor the temperature of the prism. The thermocouple was held in place by Permatex Ultra Copper RTV silicone. The prism sample of LiTbF$_4$ was placed between two copper blocks, which were heated by two cartridge heaters within each block. The temperature was set using an Eurotherm 2416 temperature controller and allowed to stabilize for 45 min before refractive index data was taken. The temperature stability was ±1°C. Refractive indices were measured from 25 to 200 deg in increments of 25 deg.

3. RESULTS

The ordinary and extraordinary refractive indices of LiTbF$_4$ at their corresponding temperatures are shown in Figs. 1 and 2.

The data were fit to a modified version of a temperature-dependent Sellmeier equation discussed by Schlarb and Betzler [10] using the Levenburg–Marquardt algorithm,

$$n^2 = A + \frac{(B + CF)\lambda^2}{\lambda^2 - (\lambda_1^2 + DF)\lambda^2} + E\lambda^2,$$  \hspace{1cm} (1)

where the parameter $F$ is given by

$$F = (T - T_0) \times (T + T_0 + 546.3).$$  \hspace{1cm} (2)

The parameter $T_0$ in the expression for $F$ represents the room temperature (taken as 23°C), and $T$ is the temperature that the LiTbF$_4$ crystal was set at in order to take refractive index measurements. The additive factor of 546.3 represents the conversion of $T$ and $T_0$ to the Kelvin scale. The values for the coefficients are shown in Table 1.

The values for $dn/dT$ can be found by differentiating Eq. (1) in order to obtain

$$2n\frac{dn}{dT} = \frac{(\lambda^2 - (\lambda_1 + DF)^2)C\lambda^2 - (B + CF)\lambda^2(2D(\lambda_1 + DF))}{(\lambda^2 - (\lambda_1 + DF)\lambda^2)^2} \frac{dF}{dT}.$$  \hspace{1cm} (3)

The values of $dn/dT$ at various wavelengths and temperatures are shown in Tables 2 and 3 and Fig. 3.
The data show that LiTbF₄ is positive birefringent with a negative $dn/dT$.

4. DISCUSSION

The effect of temperature on the optical elements used in high power laser systems has been studied extensively [11]. Output beams can be distorted due to nonuniformities in the temperature profile of the various elements in the system, and the performance of each of the elements can be compromised. In the case of magneto-optic devices such as Faraday isolators, the polarization of the output beam is affected by the temperature dependence of the Verdet coefficient and the addition of linear birefringence caused by mechanical stresses due to nonuniform temperature distribution through the elasto-optic effect.

Khanazov [12] has analyzed beam distortion and depolarization of beams in high power laser systems. In his analysis, the temperature dependent refractive index profile is expressed as [13]

$$n(r) = n_0(T_a) + [T(r) - T_a]P,$$

where

$$P = \frac{dn}{dT} - \frac{\alpha n_0^2}{4} \left[ \frac{1}{1 - \nu} (p_{11} + p_{12}) \right],$$

$r$ is the radial distance from the center of the fiber, $dn/dT$ is the change of the refractive index with temperature, $\alpha$ is the thermal expansion coefficient, $\nu$ is the Poisson ratio, and $p_{11}$ and $p_{12}$ are the piezo-optic coefficients. In order to judge the utility of LiTbF₄ versus a more widely used material such as terbium gallium garnet (TGG), knowledge not only of $dn/dT$ but also the thermal expansion coefficient, the Poisson ratio, and the piezo-optic constants is required. Regrettably, these measurements have not been made to sufficient accuracy in TGG [14], and to our knowledge, no such measurements have been performed on LiTbF₄. However, limited measurements of $dn/dT$ have been measured for TGG, and the values are about twice those of LiTbF₄ and have the opposite sign. This may

<table>
<thead>
<tr>
<th>Sellmeier Parameter</th>
<th>$n_o$</th>
<th>$n_e$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1.80878</td>
<td>1.81294</td>
</tr>
<tr>
<td>B</td>
<td>0.34546</td>
<td>0.42548</td>
</tr>
<tr>
<td>C</td>
<td>-2.575 x 10⁻⁸</td>
<td>-3.284 x 10⁻⁸</td>
</tr>
<tr>
<td>D</td>
<td>1.924 x 10⁻⁸</td>
<td>2.087 x 10⁻⁸</td>
</tr>
<tr>
<td>E</td>
<td>-0.00497</td>
<td>-0.00518</td>
</tr>
<tr>
<td>$\lambda_1$</td>
<td>0.14003</td>
<td>0.13492</td>
</tr>
</tbody>
</table>

Table 2. Values of $dn_o/dT \times 10^6$ Calculated from Eq. (3)

<table>
<thead>
<tr>
<th>$\lambda (\mu)$</th>
<th>25°C</th>
<th>50°C</th>
<th>75°C</th>
<th>100°C</th>
<th>125°C</th>
<th>150°C</th>
<th>175°C</th>
<th>200°C</th>
<th>225°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.40</td>
<td>-8.78</td>
<td>-9.53</td>
<td>-10.3</td>
<td>-11.0</td>
<td>-11.8</td>
<td>-12.5</td>
<td>-13.3</td>
<td>-14.1</td>
<td>-14.8</td>
</tr>
<tr>
<td>0.50</td>
<td>-7.30</td>
<td>-7.92</td>
<td>-8.54</td>
<td>-9.16</td>
<td>-9.78</td>
<td>-10.4</td>
<td>-11.0</td>
<td>-11.7</td>
<td>-12.3</td>
</tr>
<tr>
<td>0.60</td>
<td>-6.59</td>
<td>-7.15</td>
<td>-7.71</td>
<td>-8.27</td>
<td>-8.82</td>
<td>-9.39</td>
<td>-9.95</td>
<td>-10.5</td>
<td>-11.1</td>
</tr>
<tr>
<td>0.70</td>
<td>-6.19</td>
<td>-6.72</td>
<td>-7.24</td>
<td>-7.76</td>
<td>-8.29</td>
<td>-8.81</td>
<td>-9.34</td>
<td>-9.86</td>
<td>-10.4</td>
</tr>
<tr>
<td>0.80</td>
<td>-5.95</td>
<td>-6.45</td>
<td>-6.95</td>
<td>-7.45</td>
<td>-7.95</td>
<td>-8.46</td>
<td>-8.96</td>
<td>-9.46</td>
<td>-9.97</td>
</tr>
<tr>
<td>0.90</td>
<td>-5.78</td>
<td>-6.27</td>
<td>-6.76</td>
<td>-7.24</td>
<td>-7.73</td>
<td>-8.22</td>
<td>-8.71</td>
<td>-9.20</td>
<td>-9.69</td>
</tr>
<tr>
<td>1.2</td>
<td>-5.52</td>
<td>-5.98</td>
<td>-6.45</td>
<td>-6.91</td>
<td>-7.38</td>
<td>-7.84</td>
<td>-8.31</td>
<td>-8.78</td>
<td>-9.24</td>
</tr>
<tr>
<td>1.3</td>
<td>-5.47</td>
<td>-5.93</td>
<td>-6.39</td>
<td>-6.85</td>
<td>-7.31</td>
<td>-7.78</td>
<td>-8.24</td>
<td>-8.70</td>
<td>-9.16</td>
</tr>
<tr>
<td>1.4</td>
<td>-5.44</td>
<td>-5.89</td>
<td>-6.35</td>
<td>-6.81</td>
<td>-7.26</td>
<td>-7.72</td>
<td>-8.18</td>
<td>-8.64</td>
<td>-9.10</td>
</tr>
<tr>
<td>1.5</td>
<td>-5.41</td>
<td>-5.86</td>
<td>-6.31</td>
<td>-6.77</td>
<td>-7.22</td>
<td>-7.68</td>
<td>-8.14</td>
<td>-8.59</td>
<td>-9.05</td>
</tr>
<tr>
<td>1.7</td>
<td>-5.36</td>
<td>-5.81</td>
<td>-6.26</td>
<td>-6.72</td>
<td>-7.17</td>
<td>-7.62</td>
<td>-8.07</td>
<td>-8.52</td>
<td>-8.97</td>
</tr>
</tbody>
</table>

Table 3. Values of $dn_e/dT \times 10^6$ Calculated from Eq. (3)

<table>
<thead>
<tr>
<th>$\lambda (\mu)$</th>
<th>25°C</th>
<th>50°C</th>
<th>75°C</th>
<th>100°C</th>
<th>125°C</th>
<th>150°C</th>
<th>175°C</th>
<th>200°C</th>
<th>225°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.40</td>
<td>-11.0</td>
<td>-12.0</td>
<td>-12.9</td>
<td>-13.9</td>
<td>-14.8</td>
<td>-15.8</td>
<td>-16.8</td>
<td>-17.7</td>
<td>-18.7</td>
</tr>
<tr>
<td>0.60</td>
<td>-8.33</td>
<td>-9.03</td>
<td>-9.73</td>
<td>-10.4</td>
<td>-11.1</td>
<td>-11.9</td>
<td>-12.6</td>
<td>-13.3</td>
<td>-14.0</td>
</tr>
<tr>
<td>0.70</td>
<td>-7.82</td>
<td>-8.48</td>
<td>-9.15</td>
<td>-9.81</td>
<td>-10.5</td>
<td>-11.1</td>
<td>-11.8</td>
<td>-12.5</td>
<td>-13.1</td>
</tr>
<tr>
<td>0.80</td>
<td>-7.51</td>
<td>-8.14</td>
<td>-8.78</td>
<td>-9.41</td>
<td>-10.0</td>
<td>-10.7</td>
<td>-11.3</td>
<td>-12.0</td>
<td>-12.6</td>
</tr>
<tr>
<td>0.90</td>
<td>-7.30</td>
<td>-7.92</td>
<td>-8.53</td>
<td>-9.15</td>
<td>-9.77</td>
<td>-10.4</td>
<td>-11.0</td>
<td>-11.6</td>
<td>-12.2</td>
</tr>
<tr>
<td>1.0</td>
<td>-7.16</td>
<td>-7.76</td>
<td>-8.36</td>
<td>-8.97</td>
<td>-9.57</td>
<td>-10.2</td>
<td>-10.8</td>
<td>-11.4</td>
<td>-12.0</td>
</tr>
<tr>
<td>1.1</td>
<td>-7.05</td>
<td>-7.65</td>
<td>-8.24</td>
<td>-8.83</td>
<td>-9.43</td>
<td>-10.0</td>
<td>-10.6</td>
<td>-11.2</td>
<td>-11.8</td>
</tr>
<tr>
<td>1.3</td>
<td>-6.91</td>
<td>-7.49</td>
<td>-8.08</td>
<td>-8.66</td>
<td>-9.24</td>
<td>-9.82</td>
<td>-10.4</td>
<td>-11.0</td>
<td>-11.6</td>
</tr>
<tr>
<td>1.4</td>
<td>-6.87</td>
<td>-7.44</td>
<td>-8.02</td>
<td>-8.60</td>
<td>-9.18</td>
<td>-9.75</td>
<td>-10.3</td>
<td>-10.9</td>
<td>-11.5</td>
</tr>
<tr>
<td>1.6</td>
<td>-6.80</td>
<td>-7.37</td>
<td>-7.94</td>
<td>-8.51</td>
<td>-9.09</td>
<td>-9.66</td>
<td>-10.2</td>
<td>-10.8</td>
<td>-11.4</td>
</tr>
<tr>
<td>1.7</td>
<td>-6.77</td>
<td>-7.34</td>
<td>-7.91</td>
<td>-8.48</td>
<td>-9.05</td>
<td>-9.62</td>
<td>-10.2</td>
<td>-10.8</td>
<td>-11.3</td>
</tr>
</tbody>
</table>
make the problem of compensation for thermal lensing in isolators using LiTbF₄ more tractable.

5. CONCLUSION

We have measured the refractive index of LiTbF₄ as a function of wavelength and temperature. The parameters for a temperature dependent Sellmeier equation have been calculated and can be used for modeling the performance of LiTbF₄ in high power systems. The value obtained for \( dn/dT \) are about half those of TGG and are negative. This implies that the problem of compensating for thermal lensing for a Faraday isolator fabricated with LiTbF₄ should be simpler.

REFERENCES