Nickel element doping impacts on structure features and Faraday effects of magneto-optical transparent holmium oxide ceramics

Mengyao Wang [a,b], Bin Lu [a,c,*], Bo You [a], Ruijie Pei [a], Zhigang Sun [a], Ji-Guang Li [d], Yoshio Sakka [d], Naifeng Zhuang [e,*]

[a] *School of Materials Science and Chemical Engineering, Ningbo University, Ningbo, Zhejiang 315211, China*

[b] *China Railway 20th Bureau Group Co., Ltd., Xi'an, Shaanxi 710016, China*

[c] *Key Laboratory of Photoelectric Detection Materials and Devices of Zhejiang Province, Ningbo, Zhejiang 315211, China*

[d] *Research Center for Electronic and Optical Materials, National Institute for Materials Science, Tsukuba, Ibaraki 305-0044, Japan*

[e] *College of Chemistry, Fuzhou University, Fuzhou, Fujian 350108, China*

[*]Corresponding authors

Bin Lu (Ningbo University); Naifeng Zhuang (Fuzhou University)

Emails: lvbin@nbu.edu.cn (Bin Lu); nfzhuang@fzu.edu.cn (Naifeng Zhuang)

Abstract

The nickel element doped holmium oxide (Ho_2O_3:Ni) transparent magneto-optical ceramics were fabricated by vacuum sintering and the dopant impacts on structure features and Faraday effects were investigated. The starting oxide powders were synthesized by pyrolyzing the resulting layered holmium-based hydroxide nanosheets prepared from a chemical precipitation route using the sodium hydroxide as precipitant at the freezing temperature. Upon high-temperature sintering, the $Ni_i^{\cdot\cdot}$ defect is introduced by Ni^{2+} substitution for Ho^{3+} to form the interstitial solid solution. The 1 at.% Ni^{2+} doped Ho_2O_3 ceramic sample exhibits an in-line transmittance of ~70.04% at 1550 nm with a relative density of ~99.88%, while more Ni^{2+} incorporation (e.g., 2–5 at.%) even leads to a completely opaque state. The magneto-optical transparent Ho_2O_3:1%Ni ceramic developed in this work has Verdet constants of ~-195, -65, and -29 rad/(T·m) at 635, 1064, and 1550 nm, respectively, which are ~1.8-fold higher than the commercial terbium gallium garnet crystal or ~1.4-fold higher than the pure Ho_2O_3 ceramic. This material also possesses relatively large figure of merit of ~14.6 °/T at 1064 nm and relatively high thermal conductivity of ~7.5 W/(m·K) at room temperature.

Keywords: magneto optics, transparent ceramics, rare earths, defects, Faraday effect

1. INTRODUCTION

The Faraday effect refers to a magneto-optical phenomenon caused by alterations in the magnetic field, which induces the change of the transmission characteristics of the incident light beam inside a transparent material.[1-3] Based on it, a magnetically-activated material can be employed as a magneto-optical element (MOE) in systems such as Faraday isolators, Faraday modulators, and magneto-optical switches.[4-6] It can also indirectly monitor the strength of the magnetic field.[6] Common magneto-optical materials can be classified as films, glass, crystals, and ceramics. At present, the most widely used magneto-optical medium is the terbium gallium garnet (TGG) single crystal with a Verdet constant of -134 rad/(T·m) at 633 nm.[7] According to the Faraday effect, the Faraday rotation angle is positively related to the size and magnetic induction. Therefore, a large-size TGG crystal is required to enhance the Faraday effect so as to obtain a large Faraday rotation angle, which may generate more light loss and accumulated heat to possibly induce depolarization effect and thermal lens effect.[8,9] In order to overcome this problem, more and more researchers recently devoted to developing new-type magneto-optical materials with a higher Verdet constant. The most studied magneto-optical ceramics primarily include lanthanide sesquioxides (Ln_2O_3), garnets, pyrochlores, and fluorites,[10-16] among which the sesquioxides generally possess the largest Verdet constant due to the highest Ln^{3+} concentration. In addition, a regular strategy for enhanced Verdet constant in ceramic system is to being doped with another Ln^{3+} cation such as Pr^{3+}, Ce^{3+}, Nd^{3+}, Ho^{3+}, Dy^{3+}, or Tm^{3+} via supper-exchange interaction.[3,17-20] However, these Ln^{3+} cations contain abundant energy levels, frequently causing more optical self-absorption to limit the practical application.

According to the Hund rule and Pauli exclusion principal, electrons are always arranged in pairs with opposite spins. The magnetic moments produced by two

electrons with opposite spin directions would cancel each other out. Thus, unpaired electrons are desired in the outermost layer of the nucleus for achieving magnetic moments. The main elements that meet this condition are Fe, Co, and Ni, among which the Fe element has variable valence while the abundance of the Co element is much less than that of the Ni element. Furthermore, the Ni element has good resistance corrosion and chemical stability. NiO is a typical natural antiferromagnet,[21,22] however, Ho_2O_3 is paramagnetic. Their combined influence on Faraday magneto-optical effect has scarce study so far. In this work, we fabricated transparent Ni^{2+} doped Ho_2O_3 ceramics and studied dopant impacts on structure features and Verdet constants.

In order to obtain a sinterable starting powder, we herein prepared exchangeable layered rare-earth hydroxide (LRH) as the precipitation precursor using a freezing temperature technique, since the obtained LRH nanosheet down to ~7 nm has proved its superiority for the production of high-optical-quality oxide ceramic.[23-26] During the liquid-phase synthesis, the ammonium hydroxide solution is frequently employed as the precipitant. However, such a precipitant is unable to precipitate nickel cation rather than preference for the formation of soluble coordination compound, especially under a high pH level. Considering the sodium hydroxide solution also could provide adequate OH^- anions for LRH synthesis and the soluble Na^+ byproduct could be removed by subsequent washing. Hence, we alternatively utilized the sodium hydroxide solution as the precipitant in the present work, finally leading to transparent Ho_2O_3:Ni ceramics through vacuum sintering. More importantly, unlike the Ln^{3+} cation, the Ni^{2+} dopant never generates additional optical self-absorption in the near-infrared region, which avoids imposing restriction on the application of magneto-optical Ho_2O_3 ceramics. The research findings of this work could provide a reference for the development of high-performance magneto-optical materials.

2. EXPERIMENTAL PROCEDURES

The Ho_2O_3 raw material (99.95% purity, Shanghai Diyang Chemical Co., Ltd., Shanghai, China) was dissolved by excessive amount of nitric acid upon heating. The superfluous nitric acid was fully removed by evaporating the salt solution until dryness. The dried nitrate was dissolved into distilled water to prepare a 0.075 mol/L $Ho(NO_3)_3$ mother liquor. The stoichiometric $NiCl_2{\cdot}6H_2O$ (99.9% purity, Aladdin Biochemical Technology Co., Ltd., Shanghai, China) was dissolved into the mother liquor according to 1–5 at.% cationic concentration. Meanwhile, a 1.0 mol/L NaOH solution was prepared by dissolving its commercial solid powder (98% purity, Shanghai Macklin Biochemical Co., Ltd.) into distilled water as the precipitant. The alkaline precipitant was dropwise dropped into the mother liquor at a rate of 2–3 mL/min under magnetic stirring at a freezing temperature of ~4 ºC till the pH value reached 8.5. The resulting light green suspension, after aging for 1.5 h, was filtered and repeatedly washed with distilled water to remove byproducts. The precipitation was added into $(NH_4)_2SO_4$ solution and the molar ratio of SO_4^{2-} to Ho^{3+} was selected to be 0.03. After ion exchange for 1.5 h, the product was repetitively rinsed with distilled water and absolute ethanol. After drying at 90 °C for 12 h, the precipitation precursor was calcined in a tube furnace under flowing oxygen (~150 mL/min) at 1050 °C for 4 h to yield the oxide particle. The powder was filled into a stainless-steel die for precompression, and then cold isostatically pressed under 300 MPa. The green body was sintered in a tungsten-wire furnace at 1850 °C for 6 h under 10^{-4}–10^{-5} Pa vacuum. The sintered body was finally polished on double faces to improve the surface smooth finish.

Phase identification was performed on X-ray diffractometer (XRD; Model D8 Advance Davinci, Bruker, Karlsruhe, Germany) using nickel filtered Cu $K\alpha$ radiation. The particle size distribution was determined by laser diffraction particle size analyzer

(Model ZS90, Malvern Instruments, Malvern, UK). The microscopic morphologies of the particle and ceramic samples were both observed on a field emission scanning electron microscope (FE-SEM; Model Nova NanoSEM 450, FEI Company, Hillsboro, USA) at 15–30 kV, which is equipped with an Apollo silicon drift detector series for energy-dispersive X-ray spectrometry (EDS). The in-line transmittances of sintered bodies were recorded on a UV/VIS/NIR spectrophotometer (Model Lambda 950, PerkinElmer, Shelton, USA) from 200 to 1700 nm. The Faraday rotation angles were measured using the extinction method on a self-assembled device equipped with three independent laser light sources (635, 1064, and 1550 nm) under adjustable magnetic field from 0 to 0.8 T. Thermal diffusivity and specific heat value were simultaneously obtained from a laser flash diffusivity apparatus (Model LFA467, Netzsch, Selb, Germany) using xenon lamp as the laser heating source from room temperature to 400 °C.

3. RESULTS AND DISCUSSION

Figure 1 shows XRD patterns of the precipitation precursor, calcination product, and sintered bodies with 1−5 at.% Ni^{2+} doping. The precursor exhibits typical LRH structure with characteristic sharp (220) diffraction and a series of (00*l*) (*l* = 2, 4⋯) diffractions.[27] The lattice constant (*c*) of LRH is calculated to be ~1.822 nm using Bragg's equation from the (002) diffraction. The interlayer distance for LRH is half of the *c* constant,[28] and thus its value is ~0.911 nm. After calcining at 1050 °C for 4 h, the diffraction peaks of the product match well with those of the Ho_2O_3 standard card (JCPDS No. 83-0932) and the sharp diffraction peaks indicate the high crystallinity. Owing to the close similarities between (001) direction for LRH and (111) direction for oxide, the phase transformation substantially belongs to quasi-topotactic evolution.[29] The crystallite size (D_{XRD}) can be calculated from the full width at half maximum

(FWHM) of the (222) diffraction band using Scherrer's equation: $D_{XRD} = K\lambda/(\beta Cos\theta)$, where K is the shape factor (K = 0.89), λ is the wavelength of X-ray (λ = 0.15406 nm), β is the FWHM, and θ is the Bragg angle.[30,31] The determined crystallite size of the Ho_2O_3:1%Ni oxide powder is ~39 nm.

After high-temperature sintering, the three Ho_2O_3 bulk specimens with 1−5 at.% Ni^{2+} doping still retain the cubic crystalline phase and no trace of impurities can be observed. The main (222) diffraction peaks gradually shift towards the low angle side with more Ni^{2+} addition. The lattice constants calculated by Bragg's law are ~10.572, 10.586, and 10.592 Å for 1, 2, and 5 at.% Ni^{2+} doped Ho_2O_3 samples, respectively. These phenomena indicate that homogeneous solid solutions have formed. Ho_2O_3 belongs to cubic *C*-type sesquioxide, which has 80 atoms per unit cell containing 32 Ho atoms and 48 O atoms. Its X-ray density (d_{th}) could be determined from Eq. (1):

$$d_{th} = \frac{32\times[(1-m)M_{Ho} + mM_{Ni} + 1.5M_O]}{a^3 N_A} \quad (1)$$

where M_i stands for atomic weight of element i (i = Ho, Ni, and O), a refers to the lattice constant, N_A represents the Avogadro constant, and m denotes the percentage content of Ni atom. Their corresponding theoretical densities are ~8.46, 8.42, and 8.40 g/cm^3. The slightly decreasing theoretical density is because Ni element is lighter relative to Ho element while the increasing lattice volume with more Ni^{2+} doping is another one reason.

The Ni^{2+} doping mechanism is discussed as follows: Since the oxidation states of Ni and Ho elements differ (Ni being 2+ and Ho being 3+), the introduction of low-valent Ni^{2+} may induce one of two possible point defect forms, as described in Eqs. (2) and (3):

$$2NiO \xrightarrow{Ho_2O_3} 2Ni_{Ho}^{'} + V_O^{\cdot\cdot} + 2O_o \quad (2)$$

$$3NiO \xrightarrow{Ho_2O_3} 2Ni'_{Ho} + Ni''_i + 3O_o \quad (3)$$

In the first case, every two divalent Ni acceptors create one $V_O^{\cdot\cdot}$ (Eq. 2), while in the second case, the interstitial $Ni_i^{\cdot\cdot}$ cation is formed (Eq. 3). The Ni^{2+} ionic radius [0.690 Å for coordination number (CN) = 6] is smaller than that of Ho^{3+} (0.901 Å for CN = 6). The formation of oxygen vacancies, as described in Eq. (2), results in the contraction of the unit cell. However, this was inconsistent with the XRD results. Sometimes, an exaggerated mismatch in ionic radii could lead to the formation of an interstitial solid solution according to Eq. (3). Thus, the smaller Ni^{2+} penetrates the Ho_2O_3 lattice, expending the cell dimensions. There are two inequivalent Ho^{3+} sites in Ho_2O_3 unit cell. Either Ho^{3+} is bonded to six equivalent O^{2-} atoms to form a mixture of distorted edge and corner-sharing HoO_6 octahedra or to form a mixture of edge and corner-sharing HoO_6 octahedra. Overall, the Ni^{2+} dopant cannot form substitutional solid solution, and thus Ni^{2+} site is impossible at the octahedral center. The formation of the interstitial $Ni_i^{\cdot\cdot}$ defect in the Ho_2O_3 lattice was confirmed by aforementioned analysis.

Figure 2 exhibits the morphologies of the precipitation precursor synthesized at the freezing temperature and the Ho_2O_3:1%Ni oxide powder calcined at 1050 °C for 4 h. The LRH precursor presents typical two-dimensional nanosheet shape and self-assembles into collective three-dimensional petal-like pattern. After thermal decomposition, the nanosheets collapse into rounded powders with ultrafine size. The average particle size measured by laser diffraction particle size analyzer is ~263 nm [inset in Fig. 2(b)], `while the statistic FE-SEM size is ~83 nm. The former is normally larger than the latter for the non-monodispersed powder due to the self-limitativeness of laser diffraction measurement method.[32] The result of the laser diffraction particle size analyzer could partly reflect the agglomerated extent. Our determined particle size test result is relatively small in fact while the particle size curve in differential volume

distribution exhibits a unimodal particle size distribution without observed hard agglomerates, implying the good particle sinterability for dense ceramic preparation.

Figure 3(a) shows the microstructure of the Ho_2O_3:5%Ni bulk sample, especially focusing on the triple junction region. The grain boundary can be clearly observed after thermal etching treatment and the element mapping results reveal that the elemental distributions of holmium, oxygen, and nickel are relatively uniform without detected secondary phases [Figs. 3(b)-(e)]. That is, the Ni^{2+} dopant has well dissolved into the Ho_2O_3 matrix. The elemental distribution analysis is fully consistent with the XRD result [Fig. (1)]. The qualitative EDS analysis also detects Ho, Ni, and O elements, whereas the additional Pt element ascribes to the surface sputtering for electrical conduction [Fig. 3(f)]. By excluding the Pt-coating effect, the semiquantitative EDS analysis reveals that the elemental contents of Ho, Ni, and O respectively are 74.09, 5.19, and 20.72 at.% in close proximity to our practical stoichiometric proportion, which further verifies that our wet chemical route is feasible for Ni^{2+} and Ho^{3+} precipitation completeness.

Figure 4(a) exhibits appearances of the Ho_2O_3:$x$$Ni^{2+}$ (x = 1−5 at.%) ceramic bodies fabricated by vacuum sintering. The letters covered under the Ho_2O_3:1%Ni bulk can be well read-through. This transparent body presents brown hue similar to the pure Ho_2O_3 ceramic,[13] since the intra-4f^{10} transitions for Ho^{3+} greatly absorb the purple, blue, and green lights in the visible region [Fig. 4(b)]. Additionally, the absorption bands arising from 3d−3d transitions of Ni^{2+} in the visible region almost overlap with the abundant intra-4f^{10} transitions of Ho^{3+} to be difficult to separately distinguish. Based on Tanabe–Sugano diagram, only $^3A_2 \rightarrow {}^3T_2$ transition of Ni^{2+} may generate self-absorption effect in the infrared region, but it has been swallowed in the Ho^{3+} $^5I_8 \rightarrow {}^5I_6$ transition induced absorption band. That is to say, the Ni^{2+} introduction never generates additional

individual optical absorption in the infrared region, indicating its merit for application in this spectrum scope. The overall transparency has not been reduced by 1 at.% Ni^{2+} doping in comparison with our previously reported pure Ho_2O_3 ceramic.[13] Nevertheless, the color of the samples gradually deepens as the increase in Ni^{2+} contents, even leading to completely opaque black close to NiO own color. The theoretical transmittance of the defect-free Ho_2O_3 single crystal has been reported to be ~81.6% at 1550 nm.[25] In this work, the Ho_2O_3:1%Ni sample has a transparency of ~70.0% at the same wavelength, which is ~86% of the theoretical value. Unlike the case in the visible region, the near-infrared range has much less absorption from electronic transitions, and thus this material exhibits good potential to apply to the high-power near-infrared laser system such as Yb^{3+} or Nd^{3+} doped $Y_3Al_5O_{12}$ laser device. The density of the Ho_2O_3:1%Ni ceramic has been measured by the Archimedes method and the result shows that this specimen has an experimental density of ~8.45 g/cm^3. Considering its theoretical density is ~8.46 g/cm^3 (Eq. 1), the relative density is as high as ~99.88% in accordance with the good optical quality.

A self-assembled magneto-optical test equipment is used to measure the Faraday effect of the transparent Ho_2O_3:1%Ni ceramic sample via the extinction method. In this system, three lasers with wavelengths of 635, 1064, and 1550 nm are used as light sources, and the magnetic field intensity is adjusted from 0 to 0.8 T through current-controlled magnet coil. The relationship between Verdet constant (V) and Faraday rotation angle (θ) can be expressed as: $V = \theta / BL$, where B is the applied magnetic field strength and L is the thickness of the transparent material.[33] Thus, a plot of B against θ/L has a linear correlation, where the slope (s) of the fitting is equal to the V parameter. The measured Faraday rotation angle exhibits a good linear relationship with the applied magnetic field [Fig. 5(a)]. The Verdet constants are determined to be ~-195, -

65, and -29 rad/(T·m) at 635, 1064, and 1550 nm, respectively. Table 1 compares the Verdet constants among our developed Ho_2O_3:Ni ceramics and several typical magneto-optical materials.[13,25,34-39] At 1064 nm, the Verdet constant of our sample is ~1.8-fold higher than that of the commercial TGG crystal or ~1.4-fold higher than that of the pure Ho_2O_3 ceramic. Our results demonstrate that the nickel element is a quite effective dopant for enhanced Verdet constant via the supper-exchange interaction. When the temperature remains constant, the V value and the test wavelength (λ) satisfy the following formula:

$$V = \frac{E}{(\lambda^2 - \lambda_0{}^2)} \tag{4}$$

In Eq. (4), λ_0 is relative with the effective migration wavelength of electrons and E is a constant.[34] A plot of V^{-1} versus λ^2 yields a good linear relation with a high degree of fitting (R^2 = 99.94%) as shown in Fig. 5(b). The E and λ_0 parameters are determined to be ~-6.8×10^7 and 232 nm from the slope (E^{-1}) and the intercept ($-\lambda_0{}^2/E$), respectively.

Figure of merit (FoM), defined by the ratio of the Verdet constant to the light absorption coefficient (α), was used to roughly access the quality of a magneto-optical material as shown from Eqs. (5) and (6):

$$\text{FoM} = \frac{|V|}{\alpha} \tag{5}$$

$$\alpha = \frac{1}{L}\lg\left(\frac{1}{T}\right) \tag{6}$$

where T is the transmittance and L is the material thickness.[35,40,41] The calculated FoM values are ~9.8, 14.6, and 7.3 °/T at 635, 1064, and 1550 nm, respectively. The lower FoM values at 635 and 1550 nm ascribe to the strong absorption from *f-f* transitions of Ho^{3+} and longer wavelength based on Eq. (4), respectively. At 1064 nm, the FoM value of our sample is much higher than those of the previously reported undoped Ho_2O_3

ceramic and the commercial TGG single crystal (Table 1).

The measured thermal diffusivity of the Ho_2O_3:1%Ni sample is shown in Fig. 6. The thermal diffusion (α) values are ~2.94, 2.46, 2.07, 1.80, and 1.60 mm^2/s at 25, 100, 200, 300, and 400 °C, respectively. The corresponding specific heats (C_p) are ~0.302, 0.319, 0.347, 0.347, and 0.366 J/(g·K). The thermal conductivity (k) can be deduced from the equation: $k = \alpha \cdot C_p \cdot \rho$, where ρ is the material density (~8.45 g/cm^3 for Ho_2O_3:1%Ni). As a result, the k values are ~7.5, 6.6, 6.1, 5.3, and 5.0 W/(m·K) at 25, 100, 200, 300, and 400 °C, respectively. As the ambient temperature rises, the thermal diffusion together with the thermal conductivity goes a declining trend. This is because the accelerated phonon vibration frequency and the increasing collision probability caused by elevated temperature shorten phonon mean free path. At room temperature, the thermal conductivity of our sample is much higher than those of previously reported Ho_2O_3:5%La ceramic [~4.8 W/(m·K)] and the commercial TGG crystal [~4.9 W/(m·K)],[42,43] further suggesting the good application prospect as Faraday devices.

4. CONCLUSION

Layered holmium-based hydroxide nanosheets were prepared by wet-chemical method upon themolysis into rounded Ho_2O_3:Ni oxide powders with an average particle size of ~83 nm. The optimum Ni^{2+} concentration is 1 at.%, at which the sintered body exhibits an in-line transmittance of ~70.04% at 1550 nm (~86% of the theoretical transmittance) and a thermal conductivity of ~7.5 W/(m·K) at room temperature. The nickel dopant in Ho_2O_3 has been proved to be effective to enhance the magneto-optical effect, by which the Ho_2O_3:1%Ni ceramic has ~1.4-fold higher Verdet constant than the undoped Ho_2O_3 ceramic or ~1.8-fold higher Verdet constant than the commercial TGG crystal. Our developed ceramic materials exhibit good potential to apply to the high-power laser system, especially for use in the near-infrared region.

ACKNOWLEDGMENT

The research was supported by Zhejiang Provincial Natural Science Foundation of China under Grant No. LY23F050007.

CONFLICT OF INTEREST STATEMENENT

The authors declare they have no conflicts of interest.

REFERENCES

1. Vojna D, Slezak O, Lucianetti A, Mocek T. Verdet constant of magneto-active materials developed for high-power Faraday devices. Appl Sci. 2019, 9(15):3160.

2. Feng WX, Hanke J-P, Zhou XD, Guo G-Y, Blugel S, Mokrousov Y, Yao Y. Topological magneto-optical effects and their quantization in noncoplanar antiferromagnets. Nat Commun. 2020, 11(1):118.

3. Wang MY, Lu B, Li HX. Fantastic valence impacts of Pr on microstructures and Faraday magneto-optical effects of transparent $(Ho,Pr)_2O_3$ ceramics. J Eur Ceram Soc. 2021, 41(10):5258–5263.

4. Willamson LA, Chen YH, Longdell JJ. Magneto-optic modulator with unit quantum efficiency. Phys Rev Lett. 2014, 113(20):203601.

5. Sinjab F, Liao ZY, Noting I. Applications of spatial light modulators in Raman spectroscopy. Appl Spectrosc. 2019, 73(7):727–746.

6. Kumari S. A comprehensive study of magneto-optic materials and its applications. Mater Today Proc. 2022, 56(1):100–106.

7. Zhuang NF, Song CG, Guo LW, Wang RF, Hu XL, Zhao B, Lin SK, Chen JZ. Growth of terbium gallium garnet (TGG) magneto-optic crystals by edge-defined film-fed growth method. J Cryst Growth. 2013, 381:27–32.

8. Yin QW, Lu HD, Peng KC. Investigation of the thermal lens effect of the TGG crystal in high-power frequency-doubled laser with single frequency operation. Opt Express. 2015, 23(4):4981–4990.

9. Yakovlev AI, Snetkov IL, Palashov OV, Dai JW, Li XY, Li J. Magneto-optical and thermo-optical properties of Ce, Pr, and Ho doped TAG ceramics. IEEE J Quantum Elect. 2019, 55(5):7000108.

10. Fan W, Bai Y, Liu YF, Li TT, Li BM, Zhang L, Gao CM, Shan SY, Han HC.

Principal element design of pyrochlore-fluorite dual-phase medium- and high-entropy ceramics. J Mater Sci Techno. 2022, 107:149−154.

11. Ikesue A, Aung YL, Makikawa S, Yahagi A. Total performance of magneto-optical ceramics with a bixbyite structure. Materials. 2019, 12(3):421.

12. Slezak O, Yasuhara R, Vojna D, Furuse H, Lucianetti A, Mocek T. Temperature-wavelength dependence of Verdet constant of Dy_2O_3 ceramics. Opt Mater Express. 2019, 9(7):2971–2981.

13. Lu B, Cheng HM, Xu XX, Chen HB. Preparation and characterization of transparent magneto-optical Ho_2O_3 ceramics. J Am Ceram Soc. 2019, 102(1):118–122.

14. Qi HB, Zhou D, Li XH, Jing W, Li WW, Xu JY. Fabrication of 4 at.% La^{3+} ion doped $(Tb_xLu_{0.96-x})_2O_3$ transparent ceramics by using NC-PLSH technology and characterisation of their magneto-optical properties. J Eur Ceram Soc. 2022, 43(3):1156–1160.

15. Zhang KB, Li WW, Zeng JJ, Deng T, Luo BZ, Zhang HB, Huang XG. Preparation of $(La_{0.2}Nd_{0.2}Sm_{0.2}Gd_{0.2}Yb_{0.2})_2Zr_2O_7$ high-entropy transparent ceramic using combustion synthesized nanopowder. J Alloys Compd. 2020, 817:153328.

16. Lai YQ, Lu B, Wang M. Optical properties and Faraday magneto-optical effects of highly transparent novel $Tb_2Zr_2O_7$ fluorite ceramics. Scr Mater. 2023, 227:115282.

17. Furuse H, Yasuhara R, Hiraga K, Zhou SM. High Verdet constant of Ti-doped terbium aluminum garnet (TAG) ceramics. Opt Mater Express. 2016, 6(1):191–196.

18. Li XY, Liu Q, Liu X, Chen XP, Wu LX, Xie TF, Shi Y, Chen HH, Li J. Novel $(Tb_{0.99}Ce_{0.01})_3Ga_5O_{12}$ magneto-optical ceramics for Faraday isolators. Scr Mater. 2020, 177:137–140.

19. Hao DM, Chen J, Ao G, Tian Y, Tang YR, Yi XZ, Zhou SM. Fabrication and performance investigation of thulium-doped TAG transparent ceramics with high

magneto-optical properties. Opt Mater. 2019, 94:311–315.

20. Balabanov SS, Permin DA, Rostokina EY, Palashov OV, Snetkov IL. Characterizations of REE:Tb_2O_3 magneto-optical ceramics. Phys Status Solidi B. 2019, 257(8):1900474.

21. Hou DZ, Qiu ZY, Saitoh E. Spin transport in antiferromagnetic insulators: progress and challenges. NPG Asia Mater. 2019, 11:35.

22. Moriyama T, Oda K, Ohkochi T, Kimata M, Ono T. Spin torque control of antiferromagnetic moments in NiO. Sci Rep. 2018, 8:14167.

23. Lu B, Li J-G, Suzuki TS, Estili M, Liu WG, Sun XD, Sakka Y. Controlled synthesis of layered rare-earth hydroxide nanosheets leading to highly transparent $(Y_{0.95}Eu_{0.05})_2O_3$ ceramics. J Am Ceram Soc. 2015, 98(5):1413–1422.

24. Lu B, Sun ZG, Wang XY, Chen HB, Sakka Y, Li J-G. Photoluminescent and scintillant properties of highly transparent $[(Y_{1-x}Gd_x)_{0.99}Dy_{0.01}]_2O_3$ (x = 0 and 0.4) ceramics. J Am Ceram Soc. 2019, 102(8):4773–4780.

25. Cheng HM, Lu B, Liu YX, Zhao Y, Sakka Y, Li J-G. Transparent magneto-optical Ho_2O_3 ceramics: Role of self-reactive resultant oxyfluoride additive and investigation of vacuum sintering kinetics. Ceram Int. 2019, 45(12):14761–14767.

26. Feng K, Lu B, Hu LB. Down/upconversion luminescence behaviors and temperature-sensing properties of highly transparent $(Er_{1-x}Yb_x)_2O_3$ ceramics. ACS Appl Electron Mater. 2022, 4(2):761–767.

27. Geng FX, Matsushita Y, Ma RZ, Xin H, Tanaka M, Iyi N, Sasaki T. Synthesis and properties of well-crystallized layered rare-earth hydroxide nitrates from homogeneous precipitation. Inorg Chem. 2009, 48(14):6724–6730.

28. Lu B, Wu SF, Cheng HM, Ye RJ, Cai XY, Wang MY, Wang Y. Binary transparent $(Ho_{1-x}Dy_x)_2O_3$ ceramics: compositional influences on particle properties, sintering

kinetics and Faraday magneto-optical effects. J Eur Ceram Soc. 2021, 41(4):2826–2833.

29. Hu LF, Ma RZ, Ozawa TC, Sasaki T. Oriented monolayer film of Gd_2O_3:0.05Eu crystallites: Quasi-topotactic transformation of the hydroxide film and drastic enhancement of photoluminescence properties. Angew Chem Int Ed. 2009, 48(21):3846–3849.

30. Wang XY, Lu B, Xia HP. Novel red-emitting orthorhombic $GdInO_3$:Eu^{3+} perovskite phosphor: Structural resolution, Judd-Ofelt theory, and comparative investigation with hexagonal counterpart. Mater Today Nano. 2023, 21:100291.

31. Ma CC, Wang MY, Xue BW, Lu B. Structural features and photoluminescence behaviors of color-temperature tunable $(Gd,Y)_3Al_5O_{12}$:Ce^{3+}/Cr^{3+} phosphors with varying compositions. J Non-Cryst Solids. 2022, 588:121645.

32. Hu LB, Lu B, Xue BW, Dai SX. Production and characterization of highly transparent novel magneto-optical $Ho_2Zr_2O_7$ ceramics with anion-deficient fluorite structures. J Mater Sci Technol. 2023, 107:217–224.

33. Vasyliev V, Villora EG, Nakamura M, Sugahara Y, Shimamura K. UV-visible Faraday rotators based on rare-earth fluoride single crystals: $LiREF_4$ (RE = Tb, Dy, Ho, Er and Yb), PrF_3 and CeF_3. Opt Express. 2012, 20(13):14460–14470.

34. Furuse H, Yasuhara R. Magneto-optical characteristics of holmium oxide (Ho_2O_3) ceramics. Opt Mater Express. 2017, 7(3):827–833.

35. Wang MY, Lu B. Manufacture and Faraday magneto-optical effect of highly transparent novel holmium aluminum garnet ceramics. Scr Mater. 2021, 195:113729.

36. Lu JY, Guo FY, Chen JZ. Growth and characteristic of $Sr_3Tb(BO_3)_3$ crystal. J Cryst Growth. 2011, 314:157–162.

37. Raja MYA, Allen D, Sisk W. Room-temperature inverse Faraday effect in terbium gallium garnet. Appl Phys Lett. 1995, 67(15):2123–2125.

38. Aung YL, Ikesue A. Development of optical grade $(Tb_xY_{1-x})_3Al_5O_{12}$ ceramics as Faraday rotator material. J Am Ceram Soc. 2017, 100(9):4081–4087.

39. Slezak O, Yasuhara R, Lucianetti A, Mocek T. Wavelength dependence of magneto-optic properties of terbium gallium garnet ceramics. Opt Express. 2015, 23(10):13641–13647.

40. Kruk A, Brylewski T, Mrozek M. Optical and magneto-optical properties of $Nd_{0.1}La_{0.1}Y_{1.8}O_3$ transparent ceramics. J Lumin. 2019, 209:333–339.

41. Xu YT, Guo HT, Xiao XS, Wang PF, Cui XX, Lu M, Lin CG, Dai SX, Peng B. High Verdet constants and diamagnetic responses of GeS_2-In_2S_3-PbI_2 chalcogenide glasses for integrated optics applications. Opt Express. 2017, 25(17):20410–20420.

42. Balabanov S, Filofeev S, Kaygorodov A, Khrustov V, Kuznetsov D, Novikova A, Permin D, Popov P, Ivanov M. Hot pressing of Ho_2O_3 and Dy_2O_3 based magneto-optical ceramics. Opt Mater: X. 2022, 13:100125.

43. Yasuhara R, Snetkov I, Starobor A, Zheleznov D, Palashov O, Khazanov E, Nozawa H, Yanagitani T. Terbium gallium garnet ceramic Faraday rotator for high-power laser application. Opt Lett. 2014, 39(5):1145–1148.

Figure captions

Fig. 1. XRD patterns of the precipitation precursor intended for Ho_2O_3:1%Ni, the calcination product at 1050 °C, and the sintered bodies with 1−5 at.% Ni^{2+} doping for comparison. The inset in the right-hand panel shows the enlarged view of the main (222) diffraction peaks.

Fig. 2. FE-SEM micrographs of the LRH precursor intended for Ho_2O_3:1%Ni (a) and its calcined powder at 1050 °C for 4 h (b).

Fig. 3. FE-SEM micrograph of the Ho_2O_3:5%Ni ceramic sample (a), elemental mappings for Ho (b), O (c), Ni (d) and their comprehensive distributions (e), and EDS elemental analysis (f).

Fig. 4. Appearances (a) and in-line transmittances (b) of the Ho_2O_3:$x$$Ni^{2+}$ (x = 1−5 at.%) ceramics fabricated by vacuum sintering.

Fig. 5. A plot of θ/L versus B for the transparent magneto-optical Ho_2O_3 ceramic doped with 1 at.% Ni^{2+} (a) and a plot of $1/V$ versus λ^2 for the Ho_2O_3:1%Ni bulk sample (b).

Fig. 6. Thermal diffusivity and thermal conductivity of the transparent Ho_2O_3:1%Ni ceramic.