Optical properties of Nd:YAG, Ti:Sapphire and NdF_3 films

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This paper describes the formation and optical characterization of films prepared by conventional electron beam evaporation process. Materials such as Nd:Yttrium Aluminum Garnet (Nd:YAG), Ti: Sapphire and NdF_3 were deposited on BK7, Suprasil, Sapphire and YAG substrates. The films were prepared by using pure single crystals of Nd:YAG and Ti: Sapphire with normal Nd and Ti concentration used in bulk crystals for production of lasers. Samples with higher Nd content were also studied in order to enhance the absorption in the film and to see the effect on optical constants, total integrated scattering and absorption. Energy Dispersive Analysis of X-rays (EDAX) confirmed the presence and quantified the elements Nd, Y, Al and O in Nd:YAG films and Ti, Al, O in Ti:Sapphire films. The X-Ray Diffraction measurements indicated the structure of these films. Refractive index n, in-homogeneities and thickness of these films were determined by taking their transmission spectra. Absorption was measured at 1064 nm by laser calorimetric method as per ISO standard ISO 11551. Total Integrated Scattering was also measured as per ISO standard /DIS 13696 at 633 nm.

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1. Introduction

The ambition to produce micro and sub micro laser systems like Planar Waveguide Laser (PWL) or a Thin Film Laser (TFL) motivated the researchers to study the optical properties of laser active coatings. The bulk laser materials such as Nd: Glass, Nd: Yttrium Aluminum Garnet (Nd:YAG), Nd: Yttrium Aluminum Perovskite (Nd:YAP), Yt:YAG, Er:YAG, Ti: Sapphire etc. are well known and commonly used for the production of solid state lasers. Therefore, as a first step, the films of these materials were prepared and optical characterization was done. It may be noted that the film properties are entirely different from bulk properties. The literature survey [1-9] reveals that the techniques used for the fabrication of such film waveguides were Liquid Phase Epitaxy (LPE), Molecular Beam Epitaxy (MBE) , Magnetron sputtering, Metallo Organic Chemical Vapour Deposition (MOCVD), rf-Sputtering, ion implantation and Pulsed Laser Deposition (PLD). However, we have fabricated these films by normal electron beam evaporation method. Among many laser active materials we selected Nd:YAG and Ti: Sapphire for our study. NdF_3 was also used because of the Nd content.

The films of Nd:YAG, Ti:Sapphire and NdF_3 was grown on amorphous substrates like BK7, Suprasil, Sapphire. Nd:YAG film on crystalline YAG substrate was also studied. Conventional electron beam evaporation and Ion Beam Sputtering (IBS) sources were used to deposit the films. Film thickness was around 1 μm and 1.5 μm. Firstly, a thin film characterisation was made in order to find the index of refraction and extinction coefficients of films from transmission spectra. Absorption and scattering was studied as per relevant ISO standards.

2. Experimental

The bulk material Nd:YAG and Ti:Sapphire used in this study for film preparations was manufactured at Optics Laboratories, Islamabad Pakistan. Thin film samples under study were partly prepared at Optics Laboratories and partly at Laser Zentrum Hannover. Samples Ndyag01-04 with added Nd content were prepared in a BAK 640 vacuum coating plant from M/S Balzers at a base pressure of $\leq$ 1x 10^{-6} mbar. All other samples Ndyag05 -13, Ti: Sapphire and NdF_3 were prepared in two separate BAK 600 (Balzers) vacuum coating plants equipped with electron guns and Ion Beam Sputtering (IBS) source respectively. Films were deposited on normal BK7, Suprasil and Sapphire substrates which were amorphous in nature. Un-doped YAG crystalline substrate was also used for such depositions. The details of evaporation parameter, sources of evaporation, rate, post treatment information have been listed in Table 1.

As the samples were deposited from pure single crystals and in some cases (see Table 1) Nd content was added during evaporation in order to enhance the absorption of pump laser light. The composition of pure Nd:YAG and Ti:Sapphire crystal is well known. In the film form, the composition is not known. Therefore, an Energy Dispersive Analysis of X-rays (EDAX) was performed for the determination of various elements Wt.% present in the films.

An X-ray diffraction (XRD) analysis of the our samples was done to find the structure of the films using a Brag-Brentano Powder Diffractometer and CuK∝ radiation source at 1.54060 Angstrom in the reflection mode. The diffraction angle ranged from 4.850 to 49.850 in 2 θ with a scanning step of 0.02 degrees and exposition time per step was 5 seconds.

For the optical characterisation of the films, transmission T was measured using a Perkin-Elmer Lambda 900 spectrophotometer. From these measurements index of refraction n, extinction coefficient k, film thickness d and other parameters were determined using interference fringes [10]. Absorption and scattering measurements of our samples were done by laser calorimetric and Total Integrated Scattering (TIS) methods. The experimental details have been reported elsewhere [11]. However, these measurements have been standardised during the recent years. This is categorised as ISO 11551 and ISO7DIS 13696 for absorption and scattering respectively.

Table 1. Details of evaporation parameters and post treatment information.

Samples	Substrates	Evaporation rate (nm / sec)	Film thickness (nm)	Substrate temp. (deg.C)	Partial Oxygen Pressure (m bar)	Annealin g temp. (deg.C)	Remarks
Ndyag06	BK7	0.50	1000	380	3x10E-4	----------	**A**
Ndyag04	BK7	0.25	926	380	3x10E-4	400 (4 Hrs.)	**B**
Ndyag01	BK7	0.25	1500	380	3x10E-4	400 (4 Hrs.)	**C**
Ndyag07	Sapphire	0.50	1500	380	3x10E-4	--------	**D**
Ndyag08	YAG	0.50	1500	380	3x10E-4	--------	--------
Ndyag10	Suprasil	0.50	1500	380	3x10E-4	600 (4 Hrs.)	**E**
Ti:Saph03	Suprasil	0.50	1200	--------	--------	--------	--------
NdF_3	Suprasil	0.50	900	360	----------	----------	--------

A. with normal conc. of Nd. Sample Ndyag05 is from same coating run
B. with added Nd_2O_3. Ndyag03 is from same batch
C. with added Nd_2O_3. Ndyag02 is from same batch
D. with normal conc. of Nd. Samples Ndyag08 to 13 are from same batch
E. Nd:YAG powder rubbed on the substrate for seeding effect and later annealed at 600 deg C

3. Results and Discussion

3.1 Thin Film Characterisation

3.1.1 Film Compositional Analysis

Surface analytical techniques such as EDAX was employed to identify and quantify by weight percent the various elements such as Nd , Y, Al ,O ,Ti and F present in Nd:YAG, Ti:Sapphire and NdF_3 films respectively. A Scanning Electron Microscope (Cam Scan2, Pioneer) was used for this purpose. The accelerating voltage was 14.8 KV. Table 2 shows the results of EDAX analysis. Only those elements have been listed which were of our interest.

Table 2. EDAX analytical data of Nd:YAG, Ti:Sapphire and NdF3 films prepared on BK7 substrates.

Sample Description	Elements in the film samples (weight %)					
	Nd	Y	Al	O	Ti	F
Ndyag06	06.98	03.63	42.56	46.83	-------	-------
Ndyag04	08.88	41.93	21.20	28.00	-------	-------
Ndyag01	12.52	41.77	14.88	30.84	-------	-------
Ndyag13	07.35	13.81	36.91	41.93	-------	-------
Ti:Saph03	-------	-------	49.34	48.58	1.06	-------
NdF3	73.71	-------	-------	-------	------	21.15

It is obvious from the EDAX data that depositions made by Nd:YAG normal laser crystal which contains 1.2 atomic percent of Nd produced the Nd:YAG films with all the elements of Nd:YAG. As in bulk material, Nd absorbs the pump energy and then laser action takes place and weight percent of Nd is much more as compared to a thin film. Therefore, samples Ndyag04 and Ndyag01 were prepared with enhanced Nd doping. This is also evident from the EDAX analysis, ndyag04 has 8.88 Wt.% and Ndyag01 shows 12.52 Wt.% of Nd (Table 2). This addition may increase the pump laser absorption efficiency. Similarly, in case of Ti:Saph03 sample Ti_2O_3 was added during evaporation and we could enhance the Ti component in the film which absorbs the pump laser energy as compared to Ti:Sapphire bulk crystal where Nd component is only 0.03 At.% and not even detectable by EDAX method.

3.1.2 Structural analysis

Table 3 lists the results of X-Ray Diffraction Analytical data of Nd:YAG, Ti:Sapphire and NdF_3 films. This analysis provided the information about the structure of the films prepared by different deposition techniques. Although bulk Nd:YAG and Ti:Sapphire materials are crystalline, the film structure of these materials was found to be X-ray amorphous. The recorded absolute intensity versus 2 θ curves indicate no regular peaks but a very low intensity profile over the whole range from 5 to 50 degrees. The glass substrate without any coating also showed similar behaviour. Only NdF_3 coating showed a sharp peak (111) at 28.2 degrees 2 θ angle and some other peaks 110, 112, 300 and 113 at different 2 θ angles were also detected which indicate the crystalline behaviour of the NdF_3 films.

Table 3. X-Ray Diffraction data.

Sample Description	Structure of the Nd:YAG, Ti:Sapphire and NdF3 film samples	
	Amorphous	**Crystalline**
Ndyag06	YES	NO
Ndyag04	YES	NO
Ndyag01	YES	NO
Ndyag10	YES	NO
Ti:Saph03	YES	NO
NdF3	NO	YES

3.1.3 Optical Constants (n, k)

The optical properties of a homogeneous isotropic thin solid film are fully described by its complex index of refraction Nf (λ) = n (λ) – ik (λ) where n (refractive index) is the real part and k (absorption index) is the imaginary part while λ is the wavelength. To determine n and k of the films, transmission spectra of each coating sample were taken in the wavelength region from 400 nm to 2000 nm from a Perkin-Elmer Lambda 900 spectrophotometer. The indices of refraction and extinction coefficients were calculated from spectro-photometric data using well known envelope method [10]. A computer program by H. A. Macleod [12] was used for the evaluation of n and k. All samples were found to be generally homogeneous. However, samples like Ndyag01 - Ndyag04 with added quantities of Nd_2O_3 showed some in-homogeneities in the film because of spitting of Nd:YAG material during evaporation. Figure 1 shows the transmission vs. wavelength of Ndyag05 sample deposited on Suprasil substrate. The periodic behaviour of the curve indicates a high degree of homogeneity in the film.

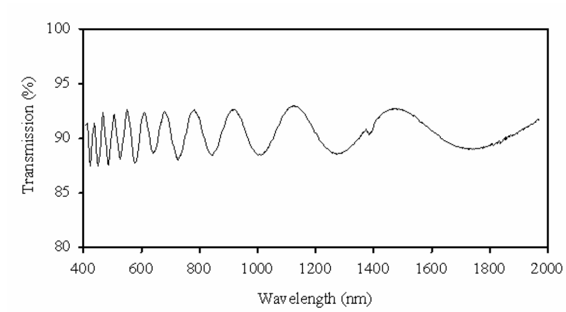

Fig. 1. Transmission spectra of Ndyag05 film.

Fig. 2 shows the transmission curve of Ti: Sapphire film and Figure 3 depicts the transmission vs. wavelength of NdF_3 film in which absorption bands near 800 nm are visible.

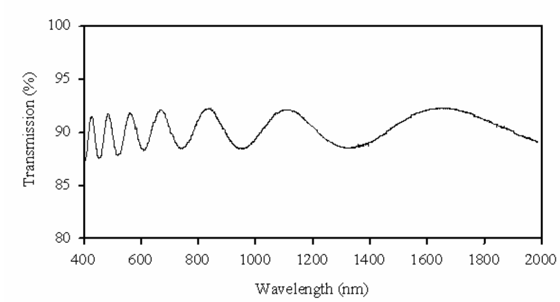

Fig. 2 Transmission spectra of Ti:Sapphire film

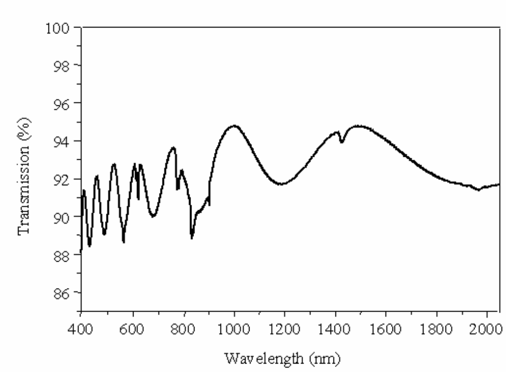

Fig. 3. Transmission spectra of NdF_3 films in which absorption bands near 800 nm are visible

Fig. 4 depicts a typical wavelength dependence of refractive index of Nd:YAG films prepared with different concentrations of Nd by electron beam evaporation method. This dependence was derived from transmission curves of films. At 1064 nm, the index of refraction was found to be 1.608 for Ndyag05 film. However, in the visible region the refractive index is higher. The upper curve next to this sample is Ndyag03 which shows a similar dependence, however it was with added quantities of Nd during deposition and estimated to be 8.88 Wt.% by EDAX analysis. The refractive index is slightly higher because of this addition of Nd concentration in the film as compared to the lower curve where films were deposited by Nd:YAG crystals of normal concentration (Wt.% 6.98). The refractive index comes out to be 1.625 at 1064 nm. The behaviour in the visible region is similar to the curve for Ndyag05 i.e. refractive index is higher in the visible region. The upper most curve in Fig. 4 shows the dispersion data for Ndyag02 film which had even more Nd concentration (12.52 Wt%) and the effect is visible from this curve. The index of refraction was found to be slightly increased in the whole visible and near infrared region. The index of refraction amounts to 1.675, at 1064 nm. It may be noted that in Ndyag02 and 03 samples, films were found to be inhomogeneous and best fits could not be obtained. However, fitted curves were close to the transmitted data.

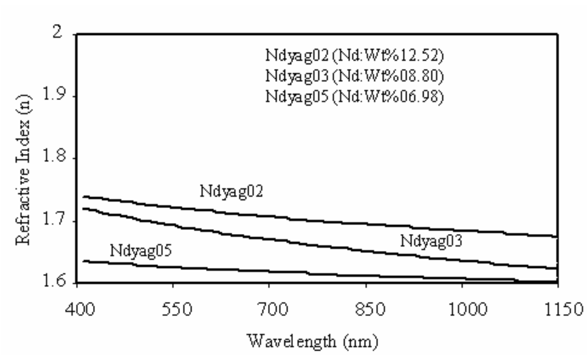

Fig. 4. Wavelength dependence of refractive index of Nd:YAG films.

Fig. 5 depicts the extinction coefficient 'k' vs. wavelength of Nd:YAG films. From the figure it is clear that k is maximum at 806 nm (Pumping wavelength for Nd:YAG lasers) for Ndyag02 which contain more Nd as compare to other two films. At 806 nm value of k for Ndyag02, Ndyag03 and Ndyag05 is 0.00226, 0.00045 and 0.00022 respectively. The absorption index k seems to be increased with the added quantities of Nd_2O_3 at 806 nm as shown in Figure 5.

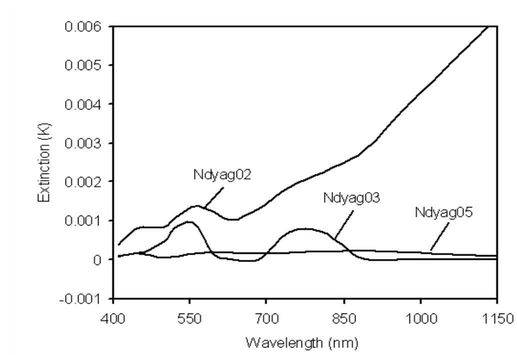

Fig. 5. Wavelength dependence of extinction k of Nd:YAG films

Figs. 6 and 7 depicts the index of refraction and extinction coefficient versus wavelength dependence curve for samples Ti:Saph03 and NdF_3 respectively. It also indicates a similar pattern and refractive index was found to be 1.602 and 1.562 respectively at 1064 nm.

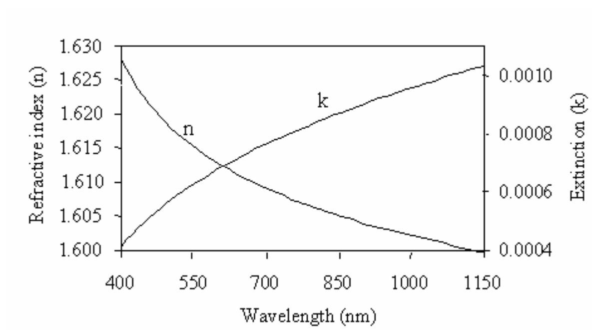

Fig. 6. Wavelength versus refractive index 'n' and extinction 'k' curves of Ti:Sapphire film.

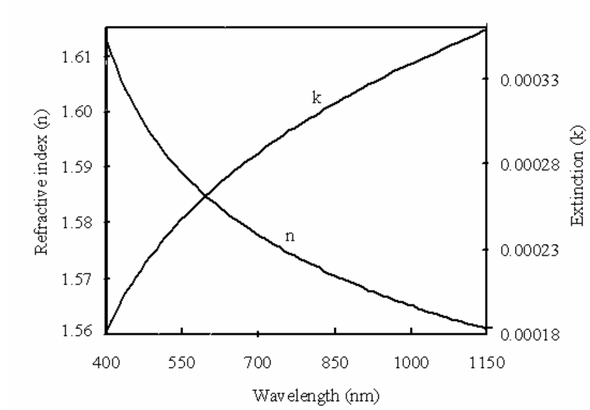

Fig. 7. Wavelength versus refractive index 'n' and extinction 'k' curves of NdF_3 film

3.1.4 Absorption Measurement

Optical coatings are not only characterised by measurements of transmission or reflection but also by measuring absorption and scattering which mainly contribute to the loss mechanisms in the film. As dielectric films should have very low absorption, their measurement technique should also be extremely precise. Absorption in the film cannot be very precisely determined by taking transmission and reflection measurements. Therefore, laser calorimetric methods are being employed for such accurate measurements. Recently, characterisation procedure for absorption measurements in thin films has been standardised [11] and is called ISO11551. The basic principle is rather simple and is reported elsewhere [11]. For the evaluation of the calorimetric data, the recorded temperature curves of heating and cooling cycles are considered. As an example we show the recorded temperature curve of sample Ndyag05 prepared on Suprasil substrate in Fig. 8.

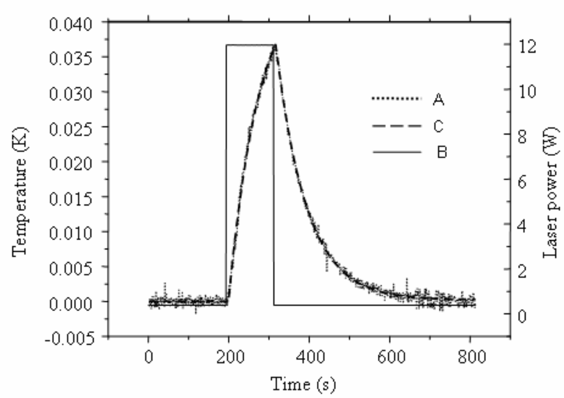

Fig. 8. Typical laser calorimetric measurement curve Ndyag05 sample on Suprasil.

As the absorption is proportional to the rate of heating and cooling cycle, the figure shows the measured curve for heating at known laser power and for a specific time. As the heating cycles is over laser is blocked and cooling occurs at the sample. Absorption is deduced from the formula given in reference [11]. Curve B in Figure 8 represents the laser power during measurement. The curve A shows the heating and cooling cycle and the curve C is the fitted curve. The measured absorption comes out to be 42.8 ppm. Figure 9 displays the results of absorption measurements at working wavelength of 1064 nm of our Nd:YAG, Ti:Sapphire and NdF_3 films.

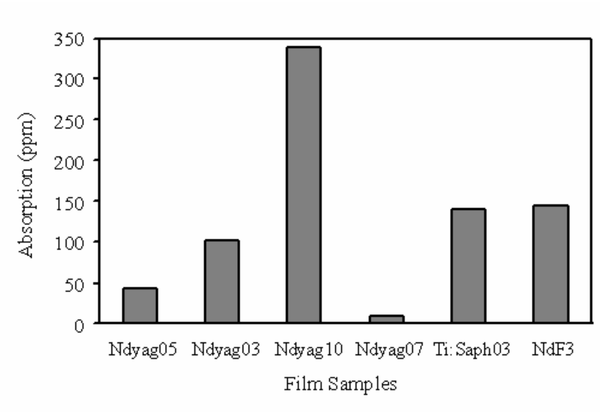

Fig. 9. Absorption data of some of the Nd:YAG, Ti:Sapphire and NdF_3 films measured by Laser Calorimetry.

It is evident from the results that films produced by Nd:YAG material with normal concentration of Nd have less absorption (Ndyag05) as compared to films produced by added Nd content (Ndyag03). Ndyag05 sample shows 42.8 ppm and Ndyag03 gives 102.8 ppm measured absorption. The absorption seems to be increased due to this added Nd content. EDAX data also support this analysis. Ndyag10 depicts the highest absorption i.e. 337.5 ppm. This was also as expected because this sample was treated with fine Nd:YAG powder before deposition so that a seeding effect may take place during evaporation and annealing process. NdF_3 film shows more absorption i.e. 143.8 ppm. This is due to the fact that NdF_3 films are crystalline. XRD analysis also showed a crystalline structure of the film (Table 3). Moreover, EDAX data revealed a very high concentration of Nd component i.e. 73.71 Wt.% which may also be a source of absorption in the film. The lowest absorption was observed in Ndyag07 sample which was prepared on sapphire substrate. Sapphire (bulk) has very low absorption as compared to Suprasil and therefore, Nd:YAG film on this substrate gives the lowest absorption value i.e. 9.057 ppm.

3.1.5 Scattering Measurements

Another parameter which is important for the characterisation of the film is the analysis of surface quality. The evaluation of surface roughness by scattering measurement has also been standardised now and the ISO / DIS 13696 is the standard for this measurement. We also characterised our Nd:YAG, Ti:Sapphire and NdF_3 films as per aforesaid standard. The experimental procedure has been reported elsewhere [11]. The results of the TIS measurements are shown in the Figure 10.

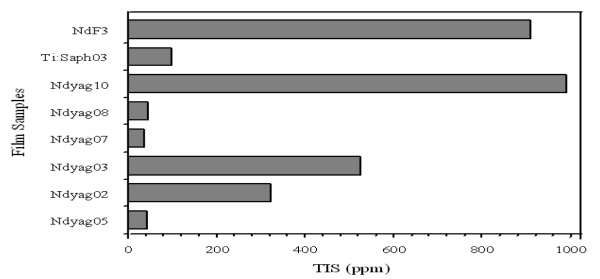

Fig. 10. Shows the Total Integrated Scattering data of Nd:YAG, Ti:Sapphire and NdF_3 films.

The histogram depicts the surface scattering value for various film samples. Scattering was measured as function of position on the surface of the film. The maximum scattering was measured in Ndyag10 (990.88 ppm). This was the sample with fine powder on the substrate before deposition. Therefore, it was expected that a high scattering will appear. However, comparing other samples it was observed that scattering is reasonably good for samples Ndyag05, 06, 07, 08, 09 and 12 ranging from 22 to 82 ppm. All these films were prepared in one batch on different substrates. Film samples Ndyag01, 02, 03 and 04 show relatively higher values of scattering. This is due to the fact that during evaporation Nd:YAG material spit much and cluster were formed on the substrate surface. Moreover, in these samples Nd_2O_3 was added to get better absorption at pump wavelength. The values range from 378 to 536 ppm.

4. Concluding remarks

Thin optical films are generally characterized by their physical properties so that they are well utilised in various applications. In laser applications optical characterization become more important. The experimental results revealed that the laser active material like Nd: YAG, Ti: Sapphire in film form have good physical and optical properties which can be a good candidate to realize laser action in thin films. The absorption in the films can also be enhanced by adding the Nd and Ti content during deposition. Due to addition of these materials, absorption of pump laser radiation around 800 nm in these films can be increased. This result was also verified by the measurement of absorption in these films. NdF_3 was not an laser active material, however characterization was done because it also contained Nd- content. The transmission measurements Nd:YAG and NdF_3 optical films showed absorption peaks near 800 nm.Transmission curve of Ti: Sapphire films showed no absorption peaks.

Optical constants n and k were determined from transmission curves. The Nd: YAG film with higher Nd content showed higher k values. n values with higher Nd content showed high n values at 1064 nm, approximately one order of magnitude less than the bulk material value of n. Similarly, Ti: Sapphire showed a decrease in n and k value with increasing wave length. A similar behavior was observed for NdF_3.

Surface qualitiy of Nd:YAG, Ti:Sapphire and NdF_3 coatings was done by measuring TIS of these films. However, Ti: Sapphire deposited on suprasil substrate showed very low scattering values. Similarly, Nd:YAG film deposited on sapphire substrate showed much less scattering values. Therefore, a dependency of substrate surface can be established on TIS values, because films generally replicate the substrate surfaces. The results indicate that films with low TIS values will have low losses of pump laser light which inturn will give better coupling efficiencies if a planar waveguide laser or a thin film laser is developed.

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