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# Characterization of light propagation in $Nd_xY_{1-x}Al(BO_3)_4$ laser crystal powders

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**Abstract**

In this work, we calculate in two different ways the transport mean-free-paths in $Nd_xY_{1-x}Al(BO_3)_4$ laser crystal powders by using the diffuse spectral reflectance and transmittance of the powders and the absorption coefficient of the crystal materials. Similar results have been obtained from both methods.


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*Keywords:* Random lasers; Laser materials; Wave propagation in random media

## 1. Introduction

The investigation of laser-like behaviour generated in laser crystal powders (LCP) has attracted considerable attention in the last few years [1]. The laser radiation generated in LCP is described as random lasing with non-resonant feedback. The central emission wavelength of such a laser is determined by the resonant wavelength of a gain medium rather by than eigenmodes of the cavity. This type of laser has no spatial coherence, it is not stable in phase, and its photon statistics is strongly different from that of a conventional laser. Their potential applications as compact and mirrorless lasers where the coherence is not necessary or the absence of coherence is desirable, motivate the study of their optical properties and the search for new crystal powder materials. The knowledge of the mean-free-path lengths involved in the scattering and absorption processes that take place in these materials is very important to analyze the behavior of random lasers. Particularly, it has been shown that the threshold of random lasers is strongly dependent on the transport mean-free-path [2]. Therefore, the calculation of this parameter becomes essential in the study of random laser materials. In the present work, we have determined the transport mean-free-path in $Nd_xY_{1-x}Al(BO_3)_4$ laser crystal powders by using the diffuse reflectance and transmittance of the powders and the absorption coefficient of the crystal materials.

## 2. Experimental

In our experiments, we have used polycrystalline powders of $Nd_xY_{1-x}Al(BO_3)_4$ $(x = 0, 0.5, 0.6)$ laser materials which have been prepared from citrate precursors obtained by soft chemistry procedures. Required stoichiometric amounts of analytical grade $Y(NO_3)_3 6H_2O$, $Nd(NO_3)_3 6H_2O$, $H_3BO_3$ and a large excess of $Al(NO_3)_3 9H_2O$ were dissolved in citric acid, and the corresponding solutions were slowly evapo-

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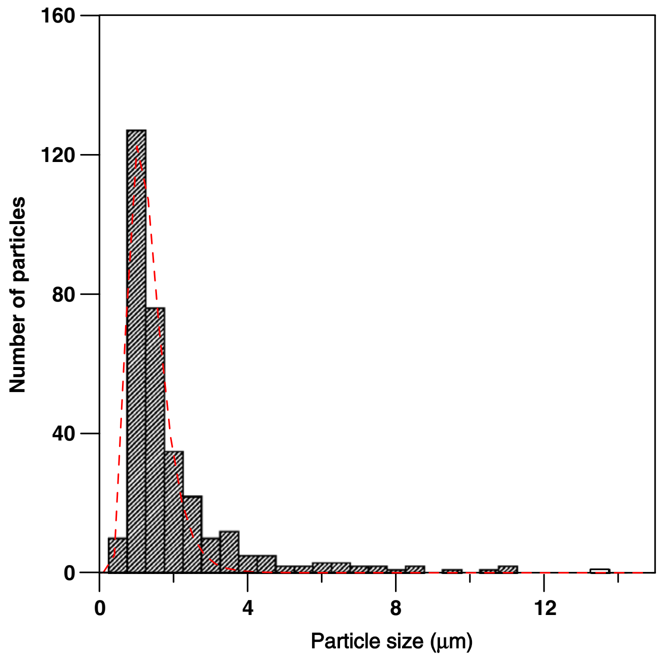


Fig. 1. Histogram of the $Nd_{0.5}Y_{0.5}Al(BO_3)_4$ particle size. The dash line is the log-normal fit from the average particle size is calculated.

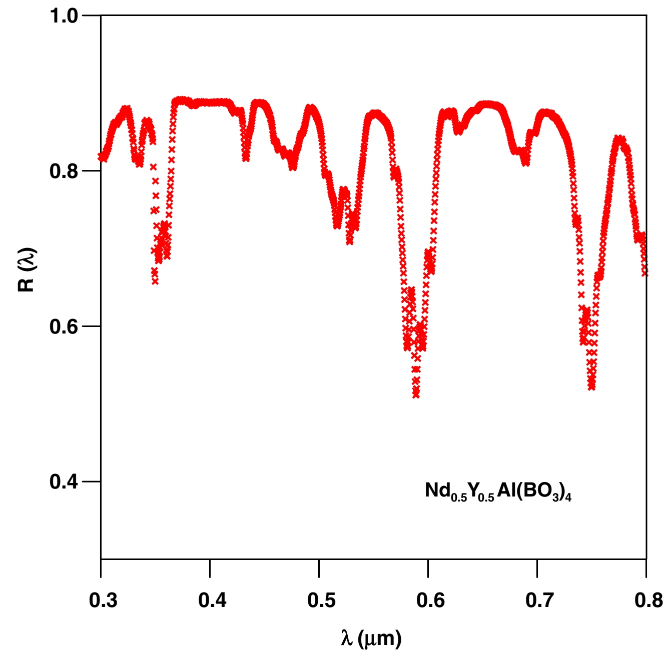


Fig. 2. Spectral reflectance $R(\lambda)$ of $Nd_{0.5}Y_{0.5}Al(BO_3)_4$ powder. The volume filling factor of the powder material is $f = 0.3 \pm 0.05$.

rated, leading to organic resins containing a random distribution of the cations at an atomic level. These resins were first dried at ~120 °C, and then the products were annealed in air up to 900 °C. Rhomboedral R32 (#155) crystal structure and purity of the samples were tested by X-ray diffraction analysis at room temperature using a Bruker D-8 diffractometer with $CuK_\alpha$ radiation. The polydispersity of the measured powders was evaluated from Scanning Electron Microscope photographs. Fig. 1 shows the histogram of the particle size corresponding to the laser crystal powder $Nd_{0.5}Y_{0.5}Al(BO_3)_4$ Fitting the histogram to a log-normal function we obtained the average particle size: $1.3 \pm 0.5$ µm, that is similar for all samples. The volume filling factor of the powder materials has been calculated by measuring volume and weight of the samples. The diffuse spectra of the powders were recorded with an integrated sphere setup. The transmission measurements were made by exciting the samples with an Ar laser and detecting the transmitted signal with a Hamamatsu R928 photomultiplier. The signal was amplified by a standard lock-in technique.

## 3. Results and discussion

Fig. 2 shows the spectral diffuse reflectance $R(\lambda)$ for $Nd_{0.5}Y_{0.5}Al(BO_3)_4$ crystal powder in the 300–800 nm wavelength range. These results correspond to absolute measurements. In Fig. 3 we represent $\mathrm{Ln}(-\mathrm{Ln}(R(\lambda)))$ as a function of $\mathrm{Ln}(1/l_i)$ for the same sample in the 410–630 nm wavelength range. $l_i$ is the inelastic mean-free-path and it is defined as the traveled length over which the intensity is reduced by a factor $1/e$ due to absorption. The inelastic mean-free-paths $l_i$ of the compound have been estimated from the absorption coefficient of the crystal sample [3]. In Fig. 3 it can be seen that the dependence is approximately linear and therefore we can use the expression

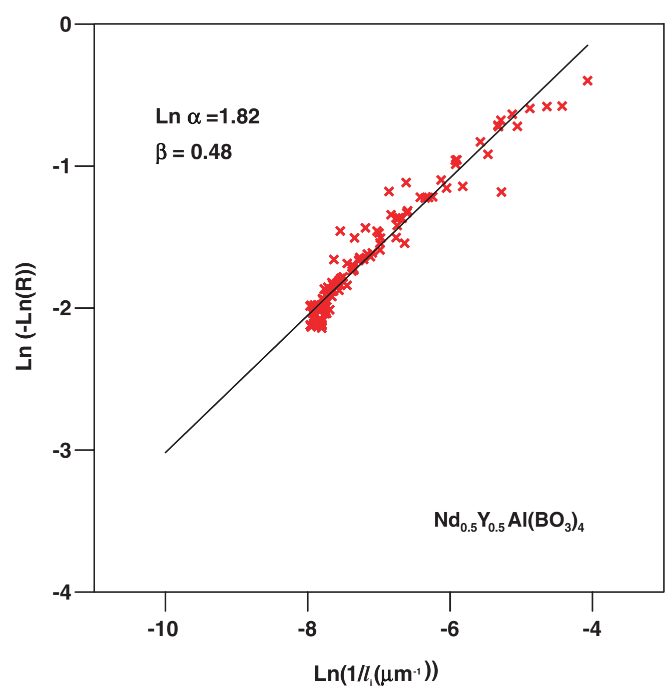


Fig. 3. $\mathrm{Ln}(-\mathrm{Ln}(R(\lambda))$ of $Nd_{0.5}Y_{0.5}Al(BO_3)_4$ powder as a function of $\mathrm{Ln}(1/l_i)$. The crosses represent the experimental points and the solid line is the linear fitting.

$$\mathrm{Ln}(-\mathrm{LnR}) = \mathrm{Ln}\alpha + \beta \mathrm{Ln}\left(\frac{1}{l_i}\right) \quad (1)$$

with $\beta$ parameter constant in that wavelength range [3]. A fitting to Eq. (1) of the experimental points plotted in Fig. 3 gives the values $\mathrm{Ln}\alpha = 1.82 \pm 0.05$ and $\beta = 0.484 \pm 0.007$. The average value of the transport mean-free-path of the crystal powder in that wavelength range has been calculated by solving numerically the equations $\alpha(l_s, \bar{\mu}, r) = 6.17$ and $\beta(l_s, \bar{\mu}, r, l_i) = 0.48$ [3]. $l_s$ is the scattering mean-free-path and $\bar{\mu}$ the anisotropy parameter. The theoretical calculations have been made by assuming a diffusive propagation of light in these materials. The average internal reflectivity of the sample ($r$) has been estimated from the Fresnel reflection coefficients [4] by taking the effective refractive index of the random system, the one given by the Maxwell–Garnet theory ($n \cong 1.21$ for $f = 0.3$). The obtained value of $l_t$ for the analyzed sample is $3.6 \pm 0.9$ μm.

Fig. 4 shows the diffuse transmittance as a function of $(1/l_i)^{0.5}$ at fixed sample thickness ($L = 500$ μm) and at several wavelengths. At large values of sample thickness $L$, the transmission decays exponentially with $L$ as $\exp(-L/l_{abs}) = \exp(-L/(l_t l_i/3)^{0.5})$ [5]. Because of the low absorption of the samples at these wavelengths, the value of $l_t$ is nearly independent on $Nd^{3+}$ concentration, so the fitting of the experimental points of Fig. 4 to the $\exp(-L/(l_t l_i/3)^{0.5})$ function, gives the value of $l_t$ of these powders at each wavelength. The obtained values are shown in Table 1.

It can be observed that the agreement between the $l_t$ values obtained from the two methods is very good. However,

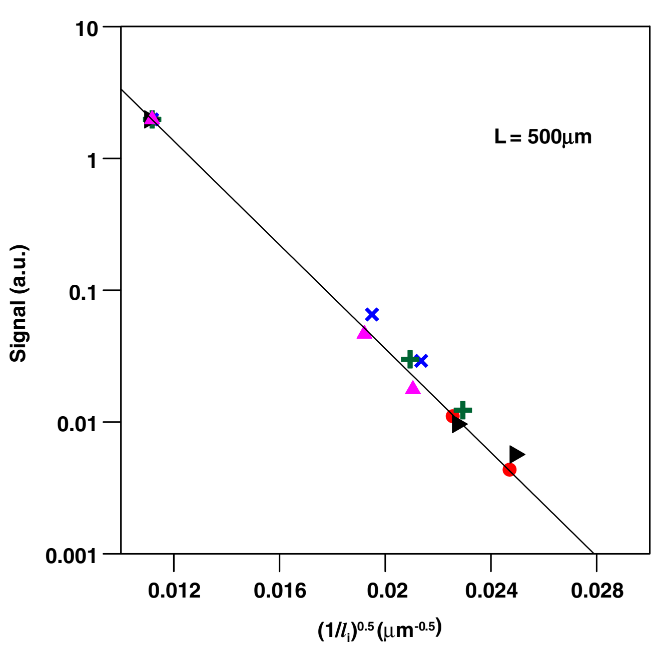


Fig. 4. Transmittance of $Nd_xY_{1-x}Al(BO_3)_4$ ($x = 0$, 0.5, 0.6) powders at $\lambda = 454$ nm (▲), $\lambda = 457$ nm (►), $\lambda = 488$ nm (+), $\lambda = 496$ nm (×) $\lambda = 502$ nm (●). The volume filling factor of the powder materials in these measurements is $f = 0.50 \pm 0.05$. The solid line is the linear fitting to the experimental points corresponding to $\lambda = 502$ nm.

Table 1
Transport mean-free-paths of $Nd_xY_{1-x}Al(BO_3)_4$ powders determined by different methods

| Method | Range (nm) | $l_t$ (μm) |
|---|---|---|
| $R(\lambda)$ vs. $l_i(\lambda)$ | 410–630 | $3.6 \pm 0.9$ |
| $T(l_i^{-0.5})$ | 454 | $3.3 \pm 0.8$ |
| $T(l_i^{-0.5})$ | 457 | $3.9 \pm 0.8$ |
| $T(l_i^{-0.5})$ | 488 | $3.8 \pm 0.8$ |
| $T(l_i^{-0.5})$ | 496 | $3.9 \pm 0.8$ |
| $T(l_i^{-0.5})$ | 502 | $3.6 \pm 0.8$ |

in an independent scattering approximation, the $l_t$ value obtained by the $R(\lambda)$ vs. $l_i(\lambda)$ method should be higher than the other values because the volume filling factor $f$ of the samples used in the reflectance measurements ($f = 0.3$) is lower than the factor used in the transmittance method ($f = 0.5$). Samples with a volume filling factor of 0.5 are closely-packed powders and spatial correlations among scatterers could smooth the decrease of transport mean-free-paths as $f$ is increased [6]. A dependence of $l_t$ with light wavelength has not been detected.

The obtained values of $l_t$ ($\lambda < l_t < L$) ensure a moderate light scattering. As a result, the light spends a long time inside the medium and can be amplified if there is sufficient gain. Taking the values given for threshold gain in similar Nd activated random laser materials [1], we estimate an amplifying length for our samples of 50 μm.

## 4. Summary

The transport mean-free-paths in $Nd_xY_{1-x}Al(BO_3)_4$ ($x = 0, 0.5, 0.6$) laser crystals powders have been experimentally determined by using two different techniques. A good agreement has been achieved between the results obtained from both methods.

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