Rare-Earth-Doped Sesquioxides for Lasers in the Mid-Infrared Spectral Range

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Dissertation

In partial fulfillment of the requirements for obtaining the degree of
Doctor rerum naturalium (Dr. rer. nat.)
at the faculty of Mathematics, Informatics and Natural Sciences
of the Universität Hamburg

Submitted by
Alexander Marc Heuer
Hamburg, 2018

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Date of the disputation	07.05.2018
Examining board leader	Prof. Dr. Dieter Horns
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Berichte aus der Physik

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Shaker Verlag
Aachen 2018

Bibliographic information published by the Deutsche Nationalbibliothek
The Deutsche Nationalbibliothek lists this publication in the Deutsche Nationalbibliografie; detailed bibliographic data are available in the Internet at http://dnb.d-nb.de.

Zugl.: Hamburg, Univ., Diss., 2018

Printed in Germany.

ISBN 978-3-8440-5985-4
ISSN 0945-0963

Shaker Verlag GmbH • P.O. BOX 101818 • D-52018 Aachen
Phone: 0049/2407/9596-0 • Telefax: 0049/2407/9596-9
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Abstract

A. M. Heuer: Rare-Earth-Doped Sesquioxides for Lasers in the Mid-Infrared Spectral Range

The topic of this work is the investigation of rare-earth-doped sesquioxide crystals for lasers operating in the mid-infrared spectral range. Er^{3+}-, Dy^{3+}-, and Pr^{3+}-doped sesquioxides were grown via the cooling down and heat exchanger method. Optimization of those methods led to the first reported growth from the melt of Dy^{3+}:Lu_2O_3. High-quality boules with combinations of the previously mentioned rare-earth dopants and the sesquioxides Lu_2O_3, Y_2O_3, and Sc_2O_3 were grown. Segregation coefficients of various materials were determined.

Spectroscopic characterization of all grown materials was performed. This includes determination of absorption and emission cross-sections, fluorescence lifetimes, positions of energy levels, and combined excited state absorption cross-sections. The combination of narrow energetic gaps between the energy multiplets and a low segregation coefficient led to Pr^{3+} not being investigated further for mid-IR laser operation after determining absorption cross-sections. Dy^{3+}:Lu_2O_3 showed cross-sections up to a factor of six higher compared to its direct competitor Dy^{3+}:ZBLAN. With excellent thermo-mechanical properties, this material is a candidate for the first oxide-based mid-infrared Dy^{3+} laser. Investigations with Er^{3+}-doped sesquioxides showed an overlap regarding their emission with the absorption of Dy^{3+} in the mid-infrared spectral range, opening up the possibility of in-band pumping. The energetic positions of the Stark levels of Dy^{3+}:Lu_2O_3 were determined for the first time.

As a continuation of the author's previous work, preliminary modelocking experiments with Yb^{3+}:Lu_2O_3 were conducted in the near-infrared and led to pulse durations as short as 313 fs while retaining more than 64 % of optical-to-optical efficiency. This constitutes the most efficient mode-locked laser oscillator with any gain material. Laser action with Dy^{3+}:Lu_2O_3 was not obtained, due to increased non-radiative decays out of the upper laser level. Experiments with Er^{3+}:Lu_2O_3 managed to improve on previous results that were obtained with comparable doping concentrations. Er^{3+}:Y_2O_3, while seeing an improved crystal quality, still experiences too much scattering for laser action to start. The first mid-infrared continuous wave laser based on Er^{3+}:Sc_2O_3 was realized and straight away a slope efficiency of 32 % with output powers of up to 611 mW was obtained, exemplarily showing the high promise of the investigated subjects.

Zusammenfassung

A. M. Heuer: Seltenerddotierte Sesquioxide für Laser im Mittleren Infraroten Spektralbereich.

Ziel der vorliegenden Arbeit ist die Untersuchung seltenerddotierter Sesquioxidkristalle bezüglich ihrer Eignung für Laseranwendungen im mittleren infraroten Spektralbereich. Es wurden Er^{3+}-, Dy^{3+}- und Pr^{3+}-dotierte Sesquioxide via der Cooling Down und der Heat Exchanger Methode gezüchtet. Eine Optimierung dieser Methoden führte zu der ersten Zucht aus der Schmelze von Dy^{3+}:Lu_2O_3. Kristalle aus Kombinationen der vorgenannten Dotierionen und den Sesquioxiden Lu_2O_3, Y_2O_3 und Sc_2O_3 wurden gezüchtet. Einbaukoeffizienten von einigen dieser Materialien wurden bestimmt.

Alle gezüchteten Materialien wurden spektroskopisch charakterisiert. Dies umfasst unter anderem die Bestimmung der Absorptions- und Emissionswirkungsquerschnitte, Fluoreszenzlebensdauern, Lage der Energieniveaus und kombinierten Wirkungsquerschnitte der Absorption aus angeregten Zuständen. Dy^{3+}:Lu_2O_3 zeigte bis um einen Faktor sechs höhere Wirkungsquerschnitte als sein direkter Konkurrent Dy^{3+}:ZBLAN. In Verbindung mit seinen exzellenten thermomechanischen Eigenschaften ist es ein Kandidat für den ersten oxidbasierten Dy^{3+} Laser im mittleren infraroten Spektralbereich. Untersuchungen mit Er^{3+}-dotierten Sesquioxiden zeigten einen großen Überlapp zwischen ihrer Emission und der Absorption von Dy^{3+} im mittleren infraroten Spektralbereich. Dies eröffnet die Möglichkeit des resonanten Pumpens. Die Position der Starkniveaus von Dy^{3+}:Lu_2O_3 wurden zum ersten Mal bestimmt.

Aufbauend auf der bisherigen Arbeit des Autors wurden erste Modenkoppelexperimente mit Yb^{3+}:Lu_2O_3 im nahen infraroten Spektralbereich durchgeführt. Pulsdauern bis hinunter zu 313 fs bei über 64 % optisch-optischer Effizienz wurden erreicht. Diese Ergebnisse stellen den bisher effizientesten modengekoppelten Laseroszillator unter Einbezug aller Verstärkungsmaterialien dar. Mit Dy^{3+}:Lu_2O_3 konnte aufgrund von erhöhten nichtstrahlenden Zerfällen keine Lasertätigkeit erreicht werden. Laserexperimente mit Er^{3+}:Lu_2O_3 konnten bisherige Resultate mit vergleichbaren Dotierkonzentrationen übertreffen. Trotz einer gesteigerten Kristallqualität wurde mit Er^{3+}:Y_2O_3, aufgrund von Streuung, keine Lasertätigkeit erreicht. Der erste Dauerstrichlaser mit Er^{3+}:Sc_2O_3 konnte realisiert werden. Hierbei wurden bereits differentielle Wirkungsgrade von bis zu 32 % und Ausgangsleistungen von bis zu 611 mW erreicht, was exemplarisch für die vielversprechenden Perspektiven der untersuchten Materialien steht.

The most exciting phrase to hear in science, the one that heralds new discoveries, is not "Eureka!" (I found it!) but "That's funny ..."

– Isaac Asimov

Contents

Glossary

α	absorption coefficient
A	transition rate
A_{ij}	Einstein coefficient
A_L	cross-section of the laser beam
A_P	cross-section of pumped crystal volume
β	inversion factor
$\beta_{i\to f}$	branching ratio for transition $i \to f$
c	speed of light ($299\,792\,458\,\mathrm{ms^{-1}}$)
C_0	starting doping concentration in the melt
C_S	doping concentration in the solid
E	energy
e	electron charge ($-1.602\,176\,620\,8 \times 10^{-19}\,\mathrm{C}$)
$E_{1,2}$	energy level $1, 2$
E_i	energy of i'th level
E_{ij}	energy difference between i'th and j'th Stark level
$E_\mathrm{p,eff}$	effective pump power density
ϵ_0	vacuum permittivity ($8.854\,187\,817\,\mathrm{Fm^{-1}}$)
$\varepsilon_{\alpha,\beta}$	eigenvalues of the parity operator
$E_\mathrm{r,eff}$	effective internal resonator power density
E_zpl	zero-phonon line energy
F	photon flux
f	frequency
$f_\mathrm{D,A}$	lineshape function (Donator, Acceptor)

γ	logarithmic resonator losses
γ_{i}	logarithmic internal losses
γ_k	logarithmic losses of element k
γ_{oc}	logarithmic output coupling mirror losses
g_i	degeneracy
$\hat{H}$	Hamilton operator
h	Planck constant ($6.626\,069\,572\,9 \times 10^{-34}\,\mathrm{J\,s}$)
η_{abs}	pump absorption efficiency
η_{oc}	output coupling efficiency
η_{opt}	optical-to-optical efficiency
η_{p}	pump overlap efficiency
η_{sl}	slope efficiency
η_{st}	Stokes efficiency
ΔI	transmission spectra intensity difference
I	intensity
I_0	incident intensity
I_{e}	pumped transmission spectrum intensity
I_{fl}	fluorescence intensity
I_{meas}	measured intensity
I_{norm}	normalized intensity
I_{u}	unpumped transmission spectrum intensity
J	total angular momentum
$\mathbf{j}_i$	individual total angular momentum
k	wavenumber
k_{B}	Boltzmann constant ($1.380\,648\,52 \times 10^{-23}\,\mathrm{JK^{-1}}$)
k_{res}	spectral response
k_{s}	segregation coefficient
L	total orbital angular momentum
l	length of the gain medium

L'	effective resonator length
λ	wavelength
λ_{ex}	excitation wavelength
λ_L	laser wavelength
λ_P	pump wavelength
L_{i}	internal losses
$\mathbf{l}_i$	individual orbital angular momentum
l_i	angular momentum quantum number
$\vec{l}_i$	angular momentum operator
l_{R}	resonator length
l'_{R}	effective resonator length
m_e	electron mass ($9.109\,383\,56 \times 10^{-31}\,\text{kg}$)
m_j	total angular momentum quantum number
m_l	magnetic quantum number
m_s	spin quantum number
n	refractive index
$N_{1,2}$	population density of energy level $E_{1,2}$
N_{c}	critical population inversion
N_{dop}	dopant ion density
$N_{\text{dop,melt}}$	dopant ion density in the melt
N_{e}	density of excited ions
n_i	principle quantum number
N_i	population of i'th level
ν	frequency
ν_{l}	laser frequency
ν_{p}	pump frequency
$\hat{P}$	parity operator
p	doping concentration
$\vec{p}_i$	momentum operator
P	power
P_{abs}	absorbed pump power
P_{in}	input pump power

P_{inc}	incident pump power
P_{out}	output power
$P_{\mathrm{out,avg}}$	average output power
P_{thr}	absorbed threshold pump power
Ψ	wave function
Ψ_n	eigenstate n
q	photon count
Q_{A}	integrated absorption cross-section
$\vec{r}$	position operator
R_{dist}	distance between ions
ρ_{ion}	cation density
r_i	nucleus-electron distance
r_{ij}	electron-electron distance
R_k	reflectivity of element k
$\vec{R}_m$	ligand position with respect to nucleus
S	total spin angular momentum
$\mathbf{s}_i$	individual spin angular momentum
$\vec{s}_i$	spin operator
σ	cross-section
σ_{abs}	absorption cross-section
$\sigma_{\mathrm{abs,l}}$	effective absorption cross-section at laser wavelength
$\sigma_{\mathrm{abs,p}}$	effective absorption cross-section at pump wavelength
σ_{em}	emission cross-section
$\sigma_{\mathrm{em,l}}$	effective emission cross-section at laser wavelength
$\sigma_{\mathrm{em,p}}$	effective emission cross-section at pump wavelength
σ_{esa}	excited-state absorption cross-section
σ_{gain}	gain cross-section
σ_{ij}	cross-section for transition $i \rightarrow j$
T	temperature
t	time

τ	lifetime
τ_{fl}	fluorescence lifetime
τ_{nr}	non-radiative lifetime
τ_{rad}	radiative lifetime
τ_{sp}	spontaneous emission lifetime
T_k	transmission of element k
T_{m}	modulation period
T_{oc}	output coupling transmission
V_{out}	lock-in amplifier output signal
$\vec{V}(r_i)$	effective centrosymmetric potential
W_{DA}	transition probability
$W_{i,j}$	transition rate from level i to j
W_{l}	emission rate per unit volume
W_{p}	pump absorption rate per unit volume
Z	atomic number
Z_j	partition function
Z_m	ionic charge of m'th ligand

Acronyms

AR	anti reflective
CW	continuous wave
DFG	difference frequency generation
EDX	energy dispersive x-ray analysis
EM	electromagnetic
ESA	excited-state absorption
ESM	electron scanning microscope
fl	Füchtbauer-Ladenburg
FWHM	full width at half maximum
GDD	group delay dispersion
GSA	ground state absorption
GTI	Gires-Tournois interferometer
HEM	heat exchanger method
HR	highly reflective
M^2	beam quality factor
mcc	McCumber relation
ML	modelocking
OPO	optical parametric oscillator
OPSL	optically pumped semiconductor laser

PID	proportional-integral-derivative
PSD	phase sensitive detector
SESAM	semiconductor saturable absorber mirror
SPM	self-phase modulation
TDL	thin disk laser
Ti:sapphire	titanium-sapphire laser
UC	upconversion
UV	ultraviolet
VECSEL	vertical external cavity surface emitting laser

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