

PLD-Grown Thin Film Saturable Absorbers

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Abstract

FRIEDJOF TELLKAMP: *PLD-Grown Thin Film Saturable Absorbers*

The subject of this thesis is the preparation and characterization of thin films made of oxidic dielectrics which may find their application as saturable absorber in passively Q-switched lasers. The solely process applied for fabrication of the thin films was the pulsed laser deposition (PLD) which stands out against other processes by its flexibility considering the composition of the systems to be investigated. Within the scope of this thesis the applied saturable absorbers can be divided into two fundamentally different kinds of functional principles:

On the one hand, saturable absorption can be achieved by ions embedded in a host medium. Most commonly applied bulk crystals are certain garnets like YAG ($\text{Y}_3\text{Al}_5\text{O}_{12}$) or the spinel forsterite (Mg_2SiO_4), in each case with chromium as dopant. Either of these media was investigated in terms of their behavior as PLD-grown saturable absorber. Moreover, experiments with Mg_2GeO_4 , Ca_2GeO_4 , Sc_2O_3 , and further garnets like YSAG or GSGG took place. The absorption coefficients of the grown films of Cr^{4+} :YAG were determined by spectroscopic investigations to be one to two orders of magnitude higher compared to commercially available saturable absorbers. For the first time, passive Q-switching of a Nd:YAG laser at 1064 nm with Cr^{4+} :YAG thin films could be realized as well as with Cr:Sc $_2$ O $_3$ thin films.

On the other hand, the desirable effect of saturable absorption can also be generated by quantum well structures. For this purpose, several layer system like YAG/LuAG, Cu $_2$ O/MgO, and ZnO/corundum were investigated. It turned out that layer systems with indium oxide (In $_2$ O $_3$) did not only grew in an excellent way but also showed up a behavior regarding their photo luminescence which cannot be explained by classical considerations. The observed luminescence at roughly 3 eV (410 nm) was assumed to be of excitonic nature and its increasing oscillator strength against thinner films was assumed to be due to confining.

The grown thin films were structurally investigated with Reflection High-Energy Electron Diffraction (RHEED). It could be observed that the applied cubic systems like Sc $_2$ O $_3$, In $_2$ O $_3$, or YAG kept the orientation of the substrate, thus they are said to grow in an epitaxial way. Considering thin films of Cr:Sc $_2$ O $_3$, one could observe epitaxial Frank-van der Merwe growth (layer-by-layer) even after the growth of films with thicknesses in a macroscopic scale.

The appearance of particulates is a major drawback when preparing thin films by pulsed laser deposition. Within this thesis, the deposition process could considerably be improved by the implementation of a velocity filter. Despite the filter's installation one could still make use of the measurement equipment like pyrometer, reflectometer, and the RHEED system. Furthermore, the existing facility was improved in such a way that the growth of multilayered systems was possible with only little effort which significantly simplified the growth of the mentioned quantum well structures.

Kurzfassung

FRIEDJOF TELLKAMP: *Mittels PLD hergestellte dünne Schichten als sättigbare Absorber*

Gegenstand dieser Arbeit ist die Herstellung und Charakterisierung von dünnen Schichten aus oxidischen Dielektrika, welche als sättigbare Absorber ihre Anwendung unter anderem in passiv gütegeschalteten Lasern finden. Der für die Herstellung der dünnen Schichten ausschließlich verwendete Prozess ist das Laserstrahlverdampfen (Pulsed Laser Deposition, PLD), welcher sich von anderen Prozessen insbesondere durch seine Flexibilität bezüglich der Zusammensetzung der abzuscheidenden Schichtsysteme unterscheidet. Die im Rahmen dieser Arbeit zur sättigbaren Absorption verwendeten Materialien lassen sich in zwei grundlegende Funktionsprinzipien unterteilen:

Einerseits kann sättigbare Absorption durch in Wirtsmaterialien eingebettete Ionen erzielt werden. Hierfür übliche Volumenkristalle sind YAG ($\text{Y}_3\text{Al}_5\text{O}_{12}$) oder das Spinell Forsterit (Mg_2SiO_4), jeweils mit Chrom als Dotierion. Diese Materialien wurden auf ihre Eigenschaften als mittels PLD hergestellte sättigbare Absorber hin untersucht. Des Weiteren fanden Versuche mit Wirtsmaterialien wie Mg_2GeO_4 , Ca_2GeO_4 , Sc_2O_3 und weiteren Vertretern der Granatgruppe (YSAG, GSGG) statt. Die spektroskopischen Analysen der Schichten aus Cr^{4+} :YAG ergaben, dass deren Absorptionskoeffizienten um ein bis zwei Größenordnungen über denen von handelsüblichen Volumenkristallen liegen können. Erstmals konnte passive Güteschaltung eines Nd:YAG Lasers bei 1064 nm mit Schichten aus Cr^{4+} :YAG realisiert werden, ebenso wie mit den Schichten aus Cr:Sc $_2$ O $_3$.

Andererseits läßt sich der gewünschte Effekt der sättigbaren Absorption auch durch Quantentopfstrukturen (quantum wells) herbeiführen. Hierfür wurden verschiedenste Schichtsysteme untersucht, unter anderem YAG/LuAG, Cu $_2$ O/MgO und ZnO/Korund. Es stellte sich heraus, dass Systeme mit dünnen Schichten aus Indiumoxid (In $_2$ O $_3$) sich nicht nur exzellent auf Sc $_2$ O $_3$, Korund oder Silicium aufwachsen lassen, sondern auch bezüglich ihrer Photolumineszenz Verhalten aufzeigen, die mit klassischen Betrachtungsweisen nicht erklärbar sind. Es wird angenommen, dass eine beobachtete Lumineszenz bei ca. 3 eV (410 nm) von exzitonischer Natur ist und deren sich erhöhende Oszillatorstärke gegen dünner werdende Schichten durch Confinement hervorgerufen wird.

Mittels Reflection High-Energy Electron Diffraction (RHEED) wurden die Schichten *in situ* auf ihre strukturellen Eigenschaften hin untersucht. Bei den verwendeten kubischen Systemen wie Sc $_2$ O $_3$, In $_2$ O $_3$ oder YAG konnte eine Beibehaltung der Orientierung des Substrates beobachtet werden, demnach wuchsen diese Systeme epitaktisch auf. Bei Schichten aus Cr:Sc $_2$ O $_3$ konnte epitaktisches Frank-van der Merwe-Wachstum (layer-by-layer) auch bei makroskopischen Dicken beobachtet werden.

Ein Nachteil der Schichterstellung mittels PLD ist die Ablagerung von Partikeln. Hier konnte der Prozess durch den Einbau eines Geschwindigkeitsfilters deutlich verbessert werden. Trotz des Einbaus des Filters waren Messungen mit Pyrometer, Reflektometer und dem RHEED-System weiterhin möglich. Außerdem wurde die bestehende PLD Anlage derart ausgebaut, dass Multilagenwachstum von dünnen Schichten mit geringem Aufwand möglich ist, welches insbesondere das Aufwachsen der genannten Quantentopfstrukturen erheblich vereinfacht hat.

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