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Title of Doctoral Dissertation

Study on Uniaxially Oriented Films of Lanthanum Hexaboride, Cerium Hexaboride, and Nickel Aluminum Superalloy for Thermophotonics Applications
(光熱変換素子応用のための六ホウ化ランタン、六ホウ化セリウムおよびニッケルアルミニウム超合金による一軸配向膜の研究)

This dissertation explores new categories of durable plasmonic materials, with a primary emphasis on lanthanum hexaboride (LaB_6), cerium hexaboride (CeB_6), and nickel aluminum (NiAl). These materials are investigated for their potential use in high-temperature thermophotonics applications, including thermal emitters, photodetectors, and optical nanoantennas. The motivation behind this research stems from the constraints of conventional refractory materials (RMs) such as molybdenum, tungsten, and nitride group materials, as well as traditional coinage materials like gold, silver, and copper. Although RMs are highly valued for their ability to withstand high temperatures and their mechanical strength, they are hindered by intricate manufacturing procedures, optical inefficiencies, and vulnerability to oxidation. Similarly, the materials used for making coins, although they have strong plasmonic resonances, are restricted by their lower melting points and susceptibility to oxidation.

The first part of the study details the development of a mid-infrared device using LaB_6 . The process entailed creating LaB_6 films using laser direct writing and reactive ion etching to form trilayers consisting of $\text{LaB}_6 - \text{Al}_2\text{O}_3 - \text{LaB}_6$. The films' orientation and crystallinity were carefully examined, emphasizing their influence on the functional parameters of the device. The study conducted a comparison between epitaxial and sputtered LaB_6 films, highlighting the enhanced spectral selectivity of the epitaxial films as a result of their superior crystalline quality.

The second section investigates the potential of CeB_6 as a plasmonic ceramic, providing an alternative to noble and refractory plasmonic metals. The study focused on growing CeB_6 thin films on Si substrates through the process of epitaxial development, utilizing electron beam physical vapor deposition and DC magnetron sputtering. An extensive analysis was conducted on the growth patterns, crystalline properties, and dielectric functions of the films. The results revealed a significant plasmonic response in the infrared region, establishing CeB_6 as a suitable material for plasmonic applications at high temperatures.

The third section offers a comprehensive examination of NiAl films, which are generated through DC magnetron sputtering with in situ heating. The study emphasizes the mechanical and electronic characteristics of the films, such as their smooth surface, excellent crystal quality, and enhanced electrical properties. The films exhibited plasmonic performance that was close to that of a NiAl single crystal and superior to conventional refractory materials,

rendering them appropriate for photothermal energy applications.

The concluding part of the study explores the enhancement of NiAl films through post-annealing and the creation of mid-infrared devices using NiAl – Al₂O₃ – NiAl trilayers. The research showcases the capacity to optimize the performance of these devices by manipulating parameters such as periodicity, width, and thickness of the NiAl strips. The films formed by combining NiAl(110) through in situ heating and subsequent post-annealing demonstrated exceptional spectral selectivity. For the short wavelength regions such as in the visible region surface smoothness plays a major role in yielding higher performance and the effect of crystallinity is moderate. For the near infrared region, crystallinity plays a more important role together with the surface morphology. And in the midinfrared region, crystallinity becomes the most important factor for exhibiting high optical figure of merit.

To summarize, this dissertation makes a substantial contribution to the field of photonic and plasmonic devices by creating new refractory plasmonic materials that are suitable for use in high-temperature environments. The study establishes fundamental principles for the future advancement of photothermal emitters and infrared devices, which could potentially transform the utilization of these materials in the field of thermophotonics.