# Broadband emitting materials doped with thulium and holmium ions for solid-state lasers at 2 µm and beyond

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**Abstract**
Novel laser-active inorganic materials doped with thulium and holmium ions have been investigated for their potential in broadly tunable and ultrafast lasers operating in the short-wave infrared spectral range of 2 to 3 µm. Lasers emitting within this spectral range find numerous applications, including light detection and ranging systems, gas sensing, medicine. They are used as amplifying systems for high harmonic generation and terahertz generation, and further frequency conversion into the mid-infrared region. The present PhD thesis establishes relationships between the structural properties of oxide and fluoride crystalline materials, in the form of single crystals and transparent ceramics, and the spectroscopic properties of thulium and holmium dopant ions, along with their laser characteristics, which are essential for laser development. Moreover, the physical processes underlying unexpected laser behaviour are revealed. This systematic study of laser materials allows for the identification of the most promising compositions for the development of broadly tunable and ultrashort pulse lasers. This study mainly focuses on materials that exhibit significant inhomogeneous spectral line broadening, resulting in broad and structureless absorption and emission spectra of the dopant ions. In order to promote this effect, several strategies have been considered, including rare-earth ion clustering, local structure disorder, compositional disorder in solid-solution compounds, and phonon-sidebands arising from electron-phonon interactions. The formation of thulium and holmium clusters in calcium fluoride crystals enables efficient and broadly tunable laser operation around 2.1 µm. A polarized spectroscopic study of the emission properties of holmium ions in disordered calcium rare-earth aluminate crystals sheds light on its broadband spectral properties and on the role of multiphonon assisted emission, which occurs beyond electronic transitions, in unexpected laser emissions observed in mode-locked lasers. In solid-solution sesquioxide ceramics of the yttria-lutetia-scandia ternary system doped with thulium ions, there is a strong and nearly linear variation of the crystal-field strength with the mean size of the host-forming cation. This relationship allows for the customization of both the position and width of the emission band above 2 µm by appropriately adjusting the host composition. Highly efficient laser operations of thulium- and holmium-doped sesquioxides around 2.1 µm and 2.3 µm have been demonstrated.