

Boron Difluoride Complexes with *N,S*-Schiff-base Ligands: Synthesis, Structure, and Photophysical Properties

Masahiro Ikeshta,* Konan Tanaka, and Takashi Tsuno*

Boron difluoride complexes incorporating *N,S*-Schiff-base ligands are synthesized, and their structural and photophysical properties are investigated. The complexes are obtained via a one-pot synthetic route from 2-mercaptobenzaldehyde, yielding *N,S*-donor frameworks that coordinate with boron atoms to form six-membered chelate rings. Single-crystal X-ray diffraction analysis revealed that these complexes adopt distorted coordination

structures, attributed to the larger atomic radius of the sulfur atom. This geometric distortion results in redshifted absorption and emission spectra, in comparison to analogous *N,O*-Schiff-base boron difluoride complexes. These findings, supported by theoretical calculations, highlight the potential of *N,S*-Schiff-base boron difluoride complexes as tunable optical materials through both electronic and geometric modulation.

1. Introduction

Photoactive organic and organometallic materials play a pivotal role in materials science owing to their versatile optical properties and tunable structures, enabling widespread use in applications such as organic light-emitting diodes,^[1,2] solar cells,^[3] fluorescence imaging,^[4] nonlinear optics,^[5,6] and circularly polarized luminescence materials.^[7–10] Among them, tetra-coordinated boron complexes have attracted considerable attention for their distinctive luminescent behavior.^[11–13] In particular, boron difluoride (BF₂) complexes with Schiff-base ligands, especially those containing *N,O*-chelating atoms, have been extensively investigated.^[14–20] These complexes typically form planar six-membered chelate rings, which contribute to rigid π -conjugated frameworks and result in intense fluorescence. Their structural modularity and strong emission properties render them promising candidates for applications in optoelectronic devices^[21] and fluorescent sensors.^[22]

N,S-Schiff-base ligands, typically derived from the condensation of 2-mercaptobenzaldehyde with amines, offer different coordination geometries compared to *N,O*-chelates, owing to the larger atomic radius and less electronegativity of sulfur. These ligands have been widely applied in coordination complexes with various metal ions, such as V(IV),^[23] Ni(II),^[24–27] Cu(II),^[28] Rh(II),^[29] Pd(II),^[28,29,31] Ir(III),^[32] and Pt(II).^[29,33] In particular, Al(III) complexes have demonstrated significantly enhanced catalytic activity in the ring-opening polymerization (ROP) of ϵ -caprolactone compared to their *N,O*-chelated counterparts,

likely due to the more flexible coordination geometry.^[33] While *N,S*-Schiff-base ligands have been widely explored in metal complexes, their application in organoboron complexes remains limited. Nevertheless, several non-Schiff-base *N,S*-chelating ligands have been successfully employed in the design of organoboron complexes.^[34–36] Notably, boron complexes bearing quinoline-8-thiolate ligands have been reported to exhibit redshifted absorption and emission compared to analogous *N,O*-chelated systems.^[37] These findings suggest that *N,S*-type chelation offers a promising strategy for modulating the electronic and optical properties of boron complexes, yet its potential remains largely underexplored in the context of Schiff-base systems.

In this study, we present the synthesis and characterization of BF₂ complexes, **1a** and **2a**, incorporating *N,S*-Schiff-base ligands derived from 2-mercaptobenzaldehyde (Figure 1). The complexes were synthesized via a one-pot reaction and successfully characterized by single-crystal X-ray diffraction analysis, which revealed their distorted six-membered chelate structures. Compared to structurally analogous *N,O*-Schiff-base BF₂ complexes, **1b** and **2b**, the *N,S*-chelated derivatives exhibited redshifted absorption and emission spectra. Notably, donor-acceptor (D–A) type complexes bearing *N,N*-diethylamino (NEt₂) substituents, **2a** and **2b**, displayed pronounced solvatochromism, accompanied by enhanced intramolecular charge-transfer (ICT). Density functional theory (DFT) and time-dependent (TD)-DFT calculations were performed to rationalize the relationship between geometric distortion and electronic transitions.

2. Results and Discussion

2.1. Synthesis and Structures

The target *N,S*-Schiff-base boron difluoride complexes were synthesized via a one-pot procedure shown in Scheme S1.

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