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Magnetic relaxation in mononuclear Tb and Dy complexes involving chelate nitronyl nitroxide ligand

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# **ACCEPTED MANUSCRIPT**

### Magnetic relaxation in mononuclear Tb and Dy complexes

#### involving chelate nitronyl nitroxide ligand

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#### ABSTRACT

Three new lanthanide-nitronyl nitroxide radical complexes  $[Ln(tfac)_3(NIT-2thz)]$ (Ln(III) = Gd 1, Tb 2, Dy 3; tfac = trifluoroacetylacetonate; NIT-2thz = 2-(2' -thiazolyl)-4,4,5,5-tetramethylimidazoline-1-oxyl-3-oxide) have been synthesized. Single crystal X-ray diffraction analyses reveal that complexes **1-3** are isomorphous. They all crystallized in the *C2/c* space groups. Each central Ln(III) ion is eight-coordinated by three bischelate tfac ligands and one radical ligand. More specifically, NIT-2thz acts as a chelate ligand to link one Ln(III) ion through its nitrogen atom of the thiazole ring and oxygen atom of the N–O group to form a two-spin system. Magnetic studies show that Gd complex exhibits ferromagnetic Gd(III)–radical coupling, Tb and Dy complexes exhibit frequency-dependent out-of-phase signals.

#### **1. Introduction**

Researches on single-molecule magnets (SMMs) which could exhibit slow magnetic relaxation at molecule level have garnered intense attention owing to their possible applications in high density magnetic storage, quantum computation as well as magnetic refrigeration [1–6]. Recently, researches are focused in particular on design and synthesis of lanthanide (Ln) based SMMs due to their huge magnetic anisotropy arising from the large, unquenched orbital angular momentum [7–10]. Up to now, numbers of Ln(III) ions based SMMs have been reported [10–18]. However, the intrinsic drawbacks of Ln(III) ions such as the dipolar spin–spin interactions and quantum tunneling always reduce the effective relaxation energy barrier and sometimes lead to the loss of remnant magnetization [19]. Fortunately, magnetic exchange coupling which generally exist in molecular paramagnetic species has been found to be an effective method to reduce quantum-tunneling relaxation processes and hence might increase the effective relaxation energy barrier [19–23]. In this view,

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