Rich magnetic chemistry of cobalt(II) complexes with*N*-phosphorylthioureas. Crystal structure and solution ^{1}H NMR spectral properties.

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Rok wydania

2007

Czasopismo

Zeitschrift für Anorganische und Allgemeine Chemie

Numer woluminu

633

Strony

2089-2096

DOI

10.1002/zaac.200700185

Kolekcja

Naukowa

Język

Angielski

Streszczenie

Structure and magnetic properties of Co^{II} complexes with *N*-phosphorylthiourea derivatives {RC(S)NHP(O)(O*i*Pr)$_2$} (**HL**) [Co{Et$_2$NC(S)NP(O)(O*i*Pr)$_2$}$_2$] (**1**), [Co{*i*PrNHC(S)NP(O)(O*i*Pr)$_2$}$_2$] (**2**), [Co{[*t*BuNHC(S)NP(O)(O*i*Pr)$_2$}$_2$] (**3**), [Co{p-MeOC$_6$H$_4$NHC(S)NP(O)(O*i*Pr)$_2$}$_2$] (**4**), [Co{p-BrC$_6$H$_4$NHC(S)NP(O)(O*i*Pr)$_2$}$_2$] (**5**) were investigated. Paramagnetic shifts in the ^{1}H NMR spectrum were observed for high-spin Co^{II} complexes with **HL**, incorporating amidophosphate moiety C(S)NP(O). The thermal dependence of the magnetic susceptibility has shown that the extended materials **2**, **3** and **5** show ferromagnetic exchange between distorted tetrahedral metal atoms, while in complex **4** antiferromagnetic behavior takes place. Compound **1** behaves as a spin-canted ferromagnet, an antiferromagnetic ordering taking place below a critical temperature, T_c = 175 K. Complex **2** was investigated by single crystal X-ray diffraction. The cobalt(II) atom in complex **2** is in distorted tetrahedral CoO_2S_2 environment formed by the C=S sulfur atoms and the P=O oxygen atoms of two deprotonated ligands.

Adres publiczny

https://doi.org/10.1002/zaac.200700185

Strona internetowa wydawcy

onlinelibrary.wiley.com

Typ publikacji

Artykuł