Structure, Magnetic Properties and ^{151}Eu, ^{119}Sn Mössbauer Spectroscopy of $Eu_5Sn_3S_{12}$ and $Eu_4LuSn_3S_{12}$

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Z. Naturforsch. **2007**, *62b*, 5 – 14; received August 21, 2006

$Eu_5Sn_3S_{12}$ and $Eu_4LuSn_3S_{12}$ were synthesized and their structures refined from single crystal data (*Pmc*2_1, $Eu_5Sn_3S_{12}$: a = 3.908(1), b = 20.115(4), c = 11.451(2) Å; $wR2$ = 0.0519 for 3048 F^2 and 122 parameters; $Eu_4LuSn_3S_{12}$: a = 3.920(1), b = 20.132(4), c = 11.459(2) Å; $wR2$ = 0.0737 for 3298 F^2 and 122 parameters). The structures contain one-dimensional chains of edge-sharing $SnS_2S_{4/2}$ octahedra and corner-sharing $SnS_3S_{2/2}$ trigonal bipyramids, running parallel to [100]. Five europium sites are seven- or eightfold coordinated by sulfur atoms. Lutetium atoms in $Eu_4LuSn_3S_{12}$ show a strong site preference for one of the two Eu^{3+} positions of $Eu_5Sn_3S_{12}$ and no structural disorder was observed. Both compounds show static mixed valence according to $Eu_3^{2+}Eu_2^{3+}Sn_3^{4+}S_{12}^{2-}$ and $Eu_3^{2+}Eu^{3+}Lu^{3+}Sn_3^{4+}S_{12}^{2-}$, which was confirmed by temperature dependent magnetic susceptibility measurements. The experimental magnetic moments of 14.6(1) ($Eu_5Sn_3S_{12}$) and 14.1(1) ($Eu_4LuSn_3S_{12}$) μ_B/f.u. indicate that each of the two sulfides contains three divalent europium atoms per formula unit. Magnetic ordering for $Eu_5Sn_3S_{12}$ and $Eu_4LuSn_3S_{12}$ sets in below 5 and 3 K, respectively. Both sulfides show metamagnetic or spin-flip transitions in the magnetization curves at 3 K (2 K) with full saturation of the europium magnetic moments at 3 K (2 K) and 80 kOe. ^{151}Eu Mössbauer spectra fully confirm the Eu^{2+} and Eu^{3+} site occupancies. At 4.2 K an increase in line width indicates small hyperfine fields at the europium nuclei.

Key words: Europium Compounds, Mixed Valence, Magnetism, Mössbauer Spectroscopy