Ferromagnetic order up to 300 K in CsCI-type EuX (X = S, Se, Te)

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The Eu(II)-chalcogenides are considered as model systems for Heisenberg magnetism because of the spin-only J = S = 7/2 4f-moment of the Eu²⁺-ions and their simple NaCl structure. The variation of the magnetic ordering temperatures within the chemical series is described by the ferromagnetic J_1 exchange between neighbouring Eu-ions and the mostly antiferromagnetic J_2 exchange via the chalcogen ligands. High pressure studies of the EuX series with conventional ¹⁵¹Eu Mössbauer spectroscopy as well as neutron studies [1, 2, 3] have contributed important information on the pressure dependence of the magnetic interactions of EuX in the NaCl phase.

Here we use the ¹⁵¹Eu nuclear forward scattering (NFS) technique at pressures up to 120 GPa (1.2 Mbar) to perform systematic measurements of the magnetic and electronic properties of EuX in the NaCl phases and, for the first time, in the CsCl-type high pressure phases. We observe a dramatic increase of the ferromagnetic ordering temperatures up to 295 K for EuS at 120 GPa and up to 300 K for EuSe at 77 GPa. The corresponding hyperfine fields in the CsCl structure exhibit a variation with pressure which is markedly different from the behaviour in the NaCl structure, attributed to the different coordination and magnetic exchange paths in the CsCl-type structure. A possible valence change of the Eu-ions towards Eu³⁺ will be discussed in conjunction with the observed isomer shifts, hyperfine fields and magnetic ordering temperatures as well as information from optical and Eu-L_{III} edge spectroscopy.

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