CRYSTAL STRUCTURE AND MAGNETIC PROPERTIES OF THE SPINEL COMPOUND $Fe_{0.76}In_{2.17}S_4$

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In the search for new materials exhibiting both ferromagnetic and semiconducting properties, approximately 10% Fe has been incorporated into the large gap semiconductor In_2S_3, resulting in a new spinel compound $Fe_{0.76}In_{2.17}S_4$. Magnetic measurements show properties of a canonical spin glass state below a freezing temperature of T_F = 5 K (Fig.1). At elevated temperatures (T > 20 K), the magnetic susceptibility of $Fe_{0.76}In_{2.17}S_4$ follows a Curie–Weiss law with a Curie–Weiss temperature of Θ_{CW} = -50 K, thus indicating predominantly antiferromagnetic exchange interactions, and an effective paramagnetic moment of μ_{eff} = 4.6 μ_B/Fe. This is about 7% below the value expected for the high spin configuration (S = 2) of Fe^{2+}.

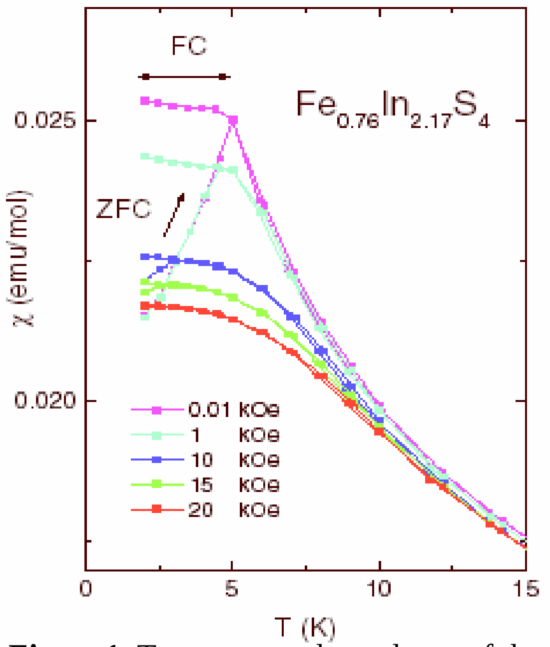

***Figure* 1.** Temperature dependence of the dc-magnetic susceptibility of $Fe_{0.76}In_{2.17}S_4$ for various fields with a clear splitting of the field-cooled (FC) and zero-field-cooled (ZFC) branch, respectively.

Standard powder x-ray diffraction at room temperature revealed the cubic spinel structure. Further structural investigations by high-resolution neutron powder diffraction in the temperature range 1.6 - 280 K (Fig.2) confirmed the spinel structure with an almost complete cation inversion corresponding to an inversion parameter of 0.94. A mutually consistent description of the neutron diffraction data, EDX analysis and magnetic measurements show that the present compound may be written as $(Fe_{0.04}In_{0.89}\square_{0.07})^A(Fe_{0.72}In_{1.28})^BS_4$, with □ denoting vacancies. No magnetic intensities could be observed, in agreement with a spin glass state. ^{57}Fe Mössbauer spectra were recorded between 4.3 K and 295 K in transmission geometry with a constant acceleration spectrometer using a ^{57}CoRh source. An occupation ratio of 0.05:1 was determined for Fe in the tetrahedral A and octahedral B positions [1].

The crystal structure provides the basis for a natural explanation of the spin glass ground state. Frustration characterizes the inability of a system to satisfy all pair-wise interactions and to establish a unique long-range ordered ground state, despite strong interactions. In the present case $Fe_{0.76}In_{2.17}S_4$, the Curie–Weiss temperature of Θ_{CW} = -50 K reflects the energy scale of the effective antiferromagnetic exchange interactions. However, the system remains paramagnetic down to T_F = 5 K, which is an order of magnitude lower than Θ_{CW}. Such a behaviour is a typical signature of dominant frustration effects inhibiting the formation of long-range magnetic order. Instead, a highly degenerate ground state is formed. Frustration combined with disorder provides the key concept to understand the spin-glass state in disordered magnets [2]. Already in 1956 Anderson [3] pointed out that the octahedral B-sites of the spinel structure form a frustrated lattice in which it is possible to achieve perfect short-range order while maintaining a finite entropy. In fact, the B-site sublattice can be directly mapped onto the pyrochlore lattice, and it forms a network of corner-sharing tetrahedra. Due to the constituent triangles, each tetrahedron is geometrically highly frustrated for nearest neighbour antiferromagnetic exchange interactions. The structural refinements of $Fe_{0.76}In_{2.17}S_4$ show that the B-sites are statistically occupied to 37% by magnetic Fe^{2+} ions, which in turn provides a high degree of atomic disorder. The presence of strong geometric frustration combined with a high degree of disorder then naturally leads to the formation of a spin glass ground state.

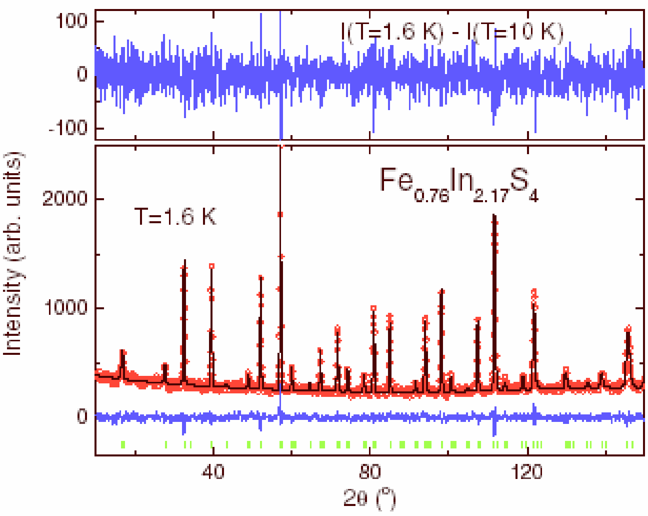

***Figure* 2.** Lower main part: neutron powder diffraction pattern of $Fe_{0.76}In_{2.17}S_4$ at T = 1.6 K. Shown are the experimentally observed (open circles) and the refined calculated (full line) intensities. The difference is shown by the bottom line, and peak positions are indicated by the vertical bars. Upper part: difference of the diffraction intensities at T = 10 K (well above T_F) from T = 1.6 K (well below T_F) in order to evidence the absence of any significant magnetic scattering.

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