Structural and magnetic transitions in α-MnS and MnSe

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This thesis describes the investigations of the low-temperature behaviour of α-MnS and MnSe. From a magnetic point of view, these compounds were often thought to be simple textbook examples of a collinear f.c.c. type-II antiferromagnet in early literature. By the development of vapour-transport crystal-growth techniques and highly sensitive measuring techniques, a renewed interest has arisen for these compounds.

In 1977 Heikens proposed the possible existence of a new phase transition in α-MnS at $T_{tr} = 131K$, $\approx 17K$ below the Néel temperature $T_N=148K$. Examination of the known literature on α-MnSe revealed that also for this compound the magnetic properties are not related with a simple paramagnetic to f.c.c. type-II antiferromagnetic phase transition.

In Chapter 3 we discuss the structural and magnetic properties of MnSe. Low-temperature X-ray diffraction, magnetic susceptibility, neutron diffraction, Mössbauer spectroscopy (collaboration with Dr. J.P. Sanchez, Strasbourg, France) and electron microscopy (collaboration with Dr. J. van Landuyt and Dr. S. Amelinckx) showed that α-MnSe undergoes a partial transformation to a hexagonal (NiAs)-type structure at low temperatures. This NiAs-type phase of MnSe is found to be always magnetically ordered (hexagonal type-I spin arrangement). Microscopic mechanisms for the nature of this (martensitic) transformation are proposed. The magnetic ordering temperature $T_N$ of the cubic phase of MnSe was determined; magnetic susceptibility measurements on the solid solution series $\text{Mn}_1-x \text{Mg}_x \text{Se}$ ($0 < x \leq 0.25$) confirmed the results obtained by neutron diffraction and Mössbauer spectroscopy.

In Chapter 4 the transport properties (resistivity, Hall effect and thermoelectric power) of iodine-grown crystals of MnSe are described. In the observed features, the partial transformation from cubic to hexagonal MnSe (and vice versa) plays an important role. Only a limited temperature region is suitable to draw some conclusions about the transport properties of NaCl-type MnSe. Here, the presence of partly compensated (by substitutional iodine as donor) acceptors (Mn vacancies) is responsible for the observed phenomena.

In Chapter 5 we look in detail at the proposed second phase transition of α-MnS at $T=131K$. The anomalous behaviour of the rhombohedral distortion parameter and the concomitant discontinuities in the magnetic susceptibility of thick platelet-shaped crystals of α-MnS appear to be caused by a
spontaneous redistribution of antiferromagnetic T domains. The presence of S domains in thin transparent platelet-shaped crystals is established by linear optical anisotropy measurements (collaboration with Dr. W. Kleemann, Paderborn, West Germany). Magnetic susceptibility and magnetization measurements on crystal platelets indicate the presence of spin flop phenomena related with the presence of easy and hard in-plane axes of the S domains. The assumption of a second phase transition is not justified.

In the Appendix it is shown by UV photoemission spectroscopy that the top of the valence band of α-MnS has mainly Mn 3d character.

Chapter 2 describes the experimental techniques used in these investigations.