Magnetic and Thermodynamic properties of RNi5 compounds
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Summary

In this thesis, the results are reported of an experimental study of the intrinsic magnetic properties of the intermetallic compounds $\text{RNi}_5$ ($R = \text{Pr, Nd, Gd, Tb, Dy, Ho, Er and Tm}$). The existence of a wide range of $\text{RNi}_5$ compounds makes them specially suited to study the properties of the different rare-earth atoms in this system. The nickel contribution to the system is relatively small and almost constant throughout the $\text{RNi}_5$ series. The small nickel influence enables us to study the properties of the rare-earth atoms in an identical environment.

Magnetization measurements were performed to study the magnetic behaviour. Heat capacity measurements were used to determine the thermodynamic properties of these materials.

In order to make the compound we melted together the pure metals, in a ratio of one atom of the rare-earth metal ($R$) and five atoms of the transition metal Ni. After cooling the material solidifies in a hexagonal (CaCu$_2$) structure. All the materials melt congruently and it was easy to grow large monocrystalline samples. For almost all the experiments we needed large monocrystals of the $\text{RNi}_5$. For the magnetization measurements along the different crystallographic directions we need spheres of 3 mm diameter. Also, the µSR experiments require large monocrystals. These crystals were produced with the Czochralski method in a tri-arc crystal growth equipment of the FOM-ALMOS facility at the Van der Waals-Zeeman Institute.

Most of the properties of the materials under study could be described successfully with our model. This model uses a quantum mechanical description of the rare-earth atoms and a more phenomenological description of the interaction of the magnetic moments. The latter part in this model describes the exchange interaction between the nickel atoms and between the nickel atoms and the rare-earth atoms in the molecular-field approximation. Here, we split the system into two sublattices; one with the nickel only and one with the rare-earth only. The exchange interaction within the rare-earth sublattice and the exchange interaction between the rare-earth sublattice and the nickel sublattice are parameters to be determined in experiment. If we apply a magnetic field, the opposed moments in the nickel sublattice will decrease. In high-field experiments this nickel susceptibility can be determined.

The contributions of the rare-earth atoms itself are decomposed into different contributions to the total hamiltonian. The most important contribution to the total hamiltonian, for us, is the interaction of the atom with the surrounding atoms, the crystal field hamiltonian. We derive the one-ion rare-earth hamiltonian explicitly. Combining the crystal field hamiltonian and the molecular-field approximation in a correct way, we obtain a mathematical description for the whole system.

In the hexagonal symmetry only four crystal field parameters, $B_2^0$, $B_4^0$, $B_6^0$ and $B_8^0$, are necessary to describe the crystal field contribution. Together with the three parameters used in the molecular field approximation, $n_{\text{RR}}$, $n_{\text{RNi}}$, and $Z_{\text{Ni}}$, we obtain seven adjustable parameters.

The interesting phenomena in our material all occur at low temperature. The $\text{RNi}_5$ compounds order magnetically at low temperature (except PrNi$_5$), having Curie temperatures between 4 K and $\sim33$ K. Below these temperatures the magnetic moments in the material align along distinctive directions. In order to study these phenomena we measured the magnetization at 1.5 K along three different crystallographic directions. We performed magnetization measurements in the High-field Installation of the University of Amsterdam in magnetic fields with field strength up to 38 T.
In specific-heat measurement the transition temperatures can be distinguished well. We measured the specific heat of all $R\text{Ni}_5$ from 1.5 K to 200 K. We determined the specific heat of an aligned ErNi$_5$ monocrystal in a magnetic field of 5 tesla along the [001] direction. $\mu$SR measurements on different samples were performed. This is a microscopic measuring method. With these measurements decaying particles, muons (abbreviation $\mu$), are placed in the sample. These particles decay in a direction that depends on the local magnetic field. In this way we could study the magnetic behaviour of our material at different temperatures.

We used all the measurements to search for the best set of parameters describing the different experiments. The theoretical behaviour of the magnetization, specific heat and susceptibility could be calculated and compared with the experimental data. Although, we could find solutions that described individual experiments very well, fitting all data with one set of parameters often resulted in deviations from experiment.

The heat capacity measurements provide us with the accurate temperature of the magnetic transition for almost all compounds (PrNi$_5$ does not order). We could describe the measured heat capacity very well with our theoretical model. The magnetization experiments provide us with insight in the sign and magnitude of the different crystal field parameters. E.g. in ErNi$_5$ the magnetization above 20 T along the [120] direction showed a meta-magnetic transition which forced us to modify the crystal field parameters. From comparison between our set of crystal field parameters and sets reported in literature, it has been concluded that we could fit better most of the results, although incomplete information in literature often obscured the comparison (in many cases the exchange interaction parameters were not published).

Our theoretical model enabled us to describe the various experiments in detail. The main contribution to the magnetic behaviour is caused by the 4$f$-electrons of the rare-earth atom. A minor, but not negligible, contribution to the magnetic behaviour is caused by the 3$d$ electrons of nickel. The nickel contribution is mainly induced by the applied field and the exchange interaction between the nickel and the rare-earth spins. Crystal field effects play an important role by the influence the 4$f$-electrons of the rare-earth on the magnetic properties in these compounds. The properties depend strongly on the decomposition of the lowest multiplet and differ between different rare-earth compounds. We found that all the crystal field coefficients, $A_n^m$, through the $R\text{Ni}_5$ series has a definite sign and are of the same order of magnitude. The nickel susceptibility throughout the series is almost constant.

Muon Knight shift observations in ErNi$_5$ could well be explained with direct dipolar-field calculations. We could establish the 3$f$ site as the stopping site of the muon in ErNi$_5$. The muon measurements, specially in zero field, still lack a thorough theoretical description.

$R\text{Ni}_5$ compounds