Magnetic and Thermodynamic properties of RNi5 compounds
Kayzel, F.E.

Citation for published version (APA):
Kayzel, F. E. (1997). Magnetic and Thermodynamic properties of RNi5 compounds UvA

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Download date: 22 Dec 2018
1 Introduction

In this thesis the intrinsic magnetic properties of $R\text{Ni}_5$ ($R = \text{Pr, Nd, Gd, Tb, Dy, Ho, Er and Tm}$) are studied, with the emphasis on $\text{ErNi}_5$. Among the possible rare-earth and transition metal Ni compounds, hexagonal $R\text{Ni}_5$ has a relatively simple structure. The material is isostructural with the $R\text{Co}_5$ compounds, which alloys show strong magnetic properties that eventually can lead to permanent magnet applications. Compared to the value of the Co moment in these materials, the nickel moment is small and shows a small, or no anisotropy. The existence of a wide range of $R\text{Ni}_5$ compounds (the existence of $\text{PmNi}_5$ and $\text{EuNi}_5$ is unknown) makes them specially suited to study the magnetic behaviour of the different rare-earth atoms in this system.

A study of the intrinsic properties of anisotropic magnetic materials strongly depends on the possibility to produce monocrystalline samples of good quality. The $R\text{Ni}_5$ compounds all melt congruently and most of them can be grown to large monocrystalline batches.

Although these materials are not directly suitable for application (except $\text{PrNi}_5$ which is used in low temperature adiabatic demagnetization cooling stages), the relative simplicity of its crystallographic structure in combination with the small nickel moment opens the possibility of describing the behaviour of the rare-earth in this material. The existence of $R\text{Co}_5$, which has more complicated magnetic contributions from the transition metal, enables comparative studies between these two isostructural materials.

1.1 History

The crystallographic structure of the $R\text{Ni}_5$ compounds was first studied by Wernick and Geller [1959]. The first reports of magnetic measurements appeared in an article by Nesbitt et al. [1962]. They determined the magnetic moment of nickel as the difference of the spontaneous magnetization measured and the theoretical value of the magnetic moment of the free rare-earth ion. Bleany [1963] showed that the strong anisotropy in the magnetization of these compounds can be ascribed to crystal field effects of the rare-earth atoms. The magnetic structure of $\text{ErNi}_5$ and $\text{TbNi}_5$ was studied with neutron diffraction on polycrystalline samples by Corliss and Hastings [1963] and Lemaire and Paccard [1970]. They assigned, within the accuracy of their measurement, a zero value for the moment of nickel.

In 1965, Nowick and Wernick studied the effective field at the Dy atoms by Mössbauer experiments. Comparison of the magnetic moments, found in these measurements and the spontaneous magnetization extrapolated to 0 K yields a small nickel moment of 0.14 $\mu_B$ per Ni atom, antiparallel to the dysprosium moment. From paramagnetic resonance data Shaltiel et al. [1964] and Burzo and Ursu [1971] derived a negative exchange interaction between the gadolinium atoms and the conduction electrons. The resulting polarization leads to a small reduction of the magnetic moment of gadolinium in GdNi$_5$. In 1976, Gignoux et al. studied monocrystalline GdNi$_5$ and YNi$_5$. YNi$_5$ was found to be a Pauli paramagnet enhanced by exchange. In GdNi$_5$, where the anisotropy is very small, a polarization of 0.16 $\mu_B$/Ni opposes the gadolinium moment and is attributed to the conduction electrons.

Magnetization measurements in fields up to 15 T on monocrystalline material were primarily studied at the Institute Louis Neél in Grenoble by Escudier et al. [1977] (ErNi$_5$),
Specific heat measurements were performed by Wallace and collaborators. The results were published in several articles, Nasu et al. [1971] (LaNi$_5$, CeNi$_5$, PrNi$_5$, NdNi$_5$ and GdNi$_5$), Craig et al. [1972] (PrNi$_5$), Marzouk et al. [1973] (LaNi$_5$, CeNi$_5$, NdNi$_5$ and GdNi$_5$) and Sankar et al. [1974] (DyNi$_5$, HoNi$_5$ and ErNi$_5$). Later, specific-heat experiments were extended for LaNi$_5$ by Ohlendorf and Flotow [1980] and for PrNi$_5$ by Ott et al. [1976] and Sahling et al. [1982]. In the thesis of Naït Saada [1980] the specific heat measurements of PrNi$_5$, NdNi$_5$, TbNi$_5$, DyNi$_5$, HoNi$_5$, ErNi$_5$ and TmNi$_5$ are presented.


1.2 Aim and outline of the thesis

The aim of this thesis is to characterize experimentally the intrinsic magnetic and thermodynamic properties of the $\text{RNi}_5$ series of intermetallic compounds, and to interpret the experimental results in terms of a physical model. In this model crystal electric field interactions and exchange interactions play a dominant role.

Within this model we searched for a single parameter set for each compound explaining simultaneously the magnetization, specific heat and susceptibility experiments.

In this thesis we extended previous magnetization measurement. Magnetization measurements in magnetic fields from 15 T to 38 T have been performed. The heat capacity of the samples was determined with high accuracy. Of monocrystalline ErNi$_5$ we measured the heat capacity in magnetic fields up to 5 T. For the first time muon spin relaxation measurements and muon Knight shift measurements were performed on several monocrystalline samples.

The possibility to grow monocrystalline samples of high quality at the University of Amsterdam in the Czochralski tri-arc equipment enabled us to produce sufficient material for all these experiments. After this introducing chapter the outline of this thesis is as follows.

In chapter 2 we introduce the essential theoretical concepts to understand the experimental data properly. The behaviour of the transition metal nickel in these compounds is described. After treating the interactions of the rare earth, the molecular-field model is introduced and the formulas necessary for the calculation are derived. The theoretical approach of the description of different experimental data is discussed.

Single-crystal growth, sample preparation and characterization are discussed in chapter 3. The binary phase diagram of ErNi$_5$ and the crystallographic structure are introduced. The new stainless-steel tri-arc, where many of our crystals were grown, is described. A small problem around the nomenclature of axes is resolved.

Chapter 4 introduces the high-field installation of the Van der Waals-Zeeman Institute of the University of Amsterdam and describes the magnetization measurement in this system. The specific-heat measuring method is treated briefly. The muon experimental setup is described and the different installations where we performed our experiments are introduced.
In chapter 5, ErNi$_5$ is presented as prototype system. For this compound the specific heat, the magnetization and the susceptibility have been measured. We present specific-heat measurements in different magnetic fields. Inelastic-neutron scattering experiments are performed to verify the energy level scheme. Additional muon experiments were performed which represent new possibilities to analyse the magnetic behaviour. For ErNi$_5$ we deduced a set of parameters that can describe all measurements very well.

For YNi$_5$, LaNi$_5$, PrNi$_5$, NdNi$_5$, GdNi$_5$, TbNi$_5$, DyNi$_5$, HoNi$_5$ and TmNi$_5$, magnetization and specific heat measurements are presented and analysed in chapter 6. For all these compounds we give the crystal-field parameters and additional parameters describing our measurements.

In chapter 7, finally, the deduced values for the magnetic and crystal electric field parameters are discussed in more detail and compared with the other parameters in the series.

1.3 Acknowledgement

The discovery of Nd$_2$Fe$_{14}$B in November 1983 has renewed the interest in 3d-4f intermetallic compounds in order to understand the outstanding magnetic properties of these materials. In 1985 this interest initiated a special programme called Concerted European Action on Magnets (CEAM) by the European Commission. More than 80 European institutes participated in this programme and created a systematic study on magnetic materials that resulted in several projects (CEAM-I, CEAM-II and CEAM-III). The present work has been part of the research programme on Basic Interactions in Rare-Earth Magnetism (BIREM contract BREU-0068). The BIREM programme started within the scope of the European Commission research and technology development programme (BRITE/EURAM). The objective of the BIREM project was to study the basic interactions that govern the electronic and magnetic properties of the 3d-4f intermetallic compounds.
$\textit{RNi}_3$ Compounds