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Neutron powder diffraction determination of the magnetic structure of Nd_2Al

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Abstract. We have determined the magnetic structure of Nd_2Al by neutron powder diffraction. This orthorhombic intermetallic compound orders ferromagnetically below 36 K with the Nd moments aligned along the b-axis. Even at 1.7 K, the larger of the two Nd moments is only 2.3(2) μ_B, about 70% of the 'free-ion' value of 3.27 μ_B. This reduction is a consequence of the substantial crystal-field effects at the Nd^{3+} sites.

1. Introduction

The R_2Al intermetallic compounds (R = rare earth) crystallize in the orthorhombic Co_2Si-type structure (space group *Pnma*, #62) in which the R atoms occupy two crystallographic sites (both 4c) and the Al occupies a third 4c site. In 1978, Sill and Biggers [1] showed that Nd_2Al is a ferromagnet with a Curie temperature of 36 K and they quoted a "fully-stretched" Nd magnetic moment of 3.27 μ_B. More recent work [2, 3, 4] suggested that the Nd magnetic moment at low temperatures is actually quite strongly reduced from the free-ion value of 3.27 μ_B and it was proposed that this reduction in moment may be due to either strong crystal-field quenching or antiferromagnetic components in the magnetic order, either intrinsic or as clusters. In this paper we present neutron powder diffraction measurements we recently made on Nd_2Al. In particular, we confirm that Nd_2Al is ferromagnetic with a strongly quenched Nd moment. No antiferromagnetic components in the magnetic order were observed.

2. Experimental Methods

The Nd_2Al sample was prepared by arc melting stoichiometric amounts of the pure elements (Nd 99.9 wt.%, Al 99.99 wt.%). The sample was turned and remelted several times in order to ensure homogeneity. The alloyed button was then sealed under vacuum in a quartz tube, annealed for 3 weeks at 700 °C and quenched in water. Cu-K_α x-ray powder diffraction and EDAX analysis confirmed the majority phase to be the intended orthorhombic Nd_2Al phase. It proved impossible to prepare a single-phase sample. Refinement of the x-ray diffraction pattern using the GSAS/EXPGUI package [5, 6] showed the presence of impurities of about 5 wt% each

of NdAl (orthorhombic *Pbcm* [7]) and Nd_3Al (cubic $Pm\bar{3}m$ [8]), with a trace of unreacted Al (cubic $Fm\bar{3}m$) also present. Basic magnetic characterization was carried out on a Quantum Design PPMS susceptometer/magnetometer operated down to 1.8 K.

Neutron diffraction experiments were carried out on the *Echidna* high-resolution powder diffractometer at the OPAL reactor in Sydney, Australia [9]. The neutron wavelength was 2.44160(2) Å, calibrated against a standard Al_2O_3 sample (NIST SRM676). All refinements of the neutron diffraction patterns employed the GSAS/EXPGUI package [5, 6]. The neutron diffraction data were corrected for absorption effects.

3. Results and Discussion

In figure 1 we show the refined neutron diffraction pattern of Nd_2Al obtained at 106 K, at which temperature Nd_2Al is paramagnetic and the neutron diffraction pattern exhibits only nuclear scattering.

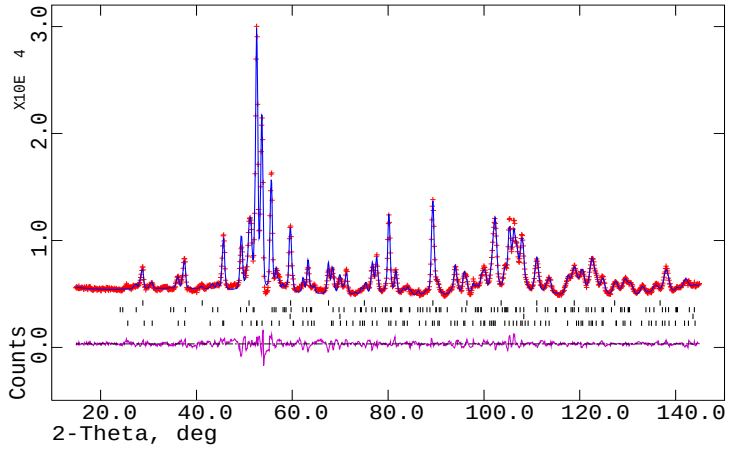

Figure 1. Neutron diffraction pattern of Nd_2Al obtained at 106 K ($\lambda =$ 2.44160(2) Å). The Bragg markers (bottom to top) represent Nd_2Al, Al, NdAl and Nd_3Al.

The refined lattice parameters at 106 K are a = 6.6825(4) Å, b = 5.2342(3) Å and c = 9.7321(7) Å. In table 1 we give the refined atomic position parameters of Nd_2Al, deduced from the refinement of the 106 K neutron powder diffraction pattern. The conventional refinement R-factors (%) are R(p) = 3.4 and $R(F^2) = 3.0$.

Table 1. Crystallographic data for Nd_2Al obtained by refinement of the 106 K neutron powder diffraction pattern.

Atom	Site	x	y	z
Nd	4c	0.0184(5)	$\frac{1}{4}$	0.7040(3)
Nd	4c	0.1929(5)	$\frac{1}{4}$	0.0724(4)
Al	4c	0.2045(12)	$\frac{1}{4}$	0.4026(6)

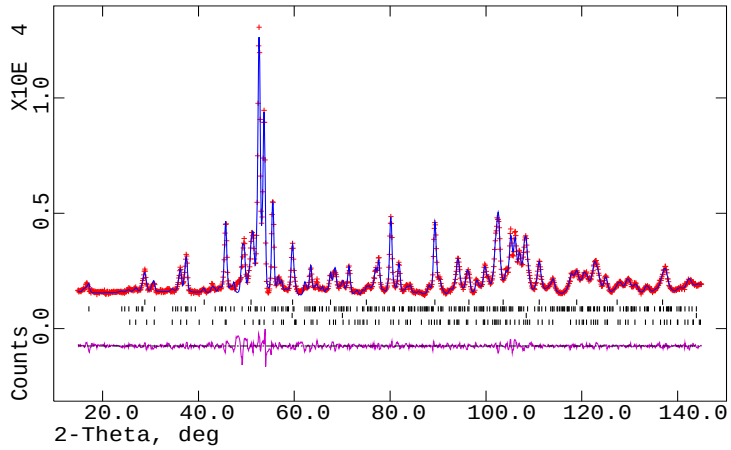

Figure 2. Neutron diffraction pattern of Nd_2Al obtained at 1.7 K ($\lambda =$ 2.44160(2) Å). The Bragg markers (bottom to top) represent Nd_2Al, Al, NdAl and Nd_3Al.

In figure 2 we show the refinement to the neutron diffraction pattern obtained at 1.7 K. The magnetic contributions to the 1.7 K Nd_2Al diffraction pattern occur only at the nuclear peak positions and correspond to the propagation vector $\mathbf{k} = [0\ 0\ 0]$. We find no evidence for additional *magnetic-only* peaks from the Nd_2Al phase. In order to consider all possible magnetic structures allowed for Nd_2Al, we carried out Representational Analysis for the Nd site using the SARA*h* program [10]. The decomposition of the magnetic representation comprises eight one-dimensional representations:

$$\Gamma_{Mag}^{4c} = 1\Gamma_1^{(1)} + 2\Gamma_2^{(1)} + 2\Gamma_3^{(1)} + 1\Gamma_4^{(1)} + 1\Gamma_5^{(1)} + 2\Gamma_6^{(1)} + 2\Gamma_7^{(1)} + 1\Gamma_8^{(1)} \quad (1)$$

and the basis vectors of these irreducible representations are given in table 2.

Table 2. Representational Analysis for the Nd(4c) site in Nd_2Al with a propagation vector [0 0 0]. The respective atomic positions are $(x,\ y,\ z)$, $(\frac{1}{2}+x,\ y,\ \frac{1}{2}-z)$, $(-x,\ -y,\ -z)$ and $(\frac{1}{2}-x,\ -y,\ z+\frac{1}{2})$.

Representation	Ordering Mode	First component	Second component
Γ_1	G_Y	$+-+-$	0
Γ_2	$C_X\ A_Z$	$++--$	$+--+$
Γ_3	$F_X\ G_Z$	$++++$	$+-+-$
Γ_4	A_Y	$+--+$	0
Γ_5	F_Y	$++++$	0
Γ_6	$A_X\ C_Z$	$+--+$	$++--$
Γ_7	$G_X\ F_Z$	$+-+-$	$++++$
Γ_8	C_Y	$++--$	0

We can immediately rule out the purely antiferromagnetic representations Γ_1, Γ_2, Γ_4, Γ_6 and Γ_8 because magnetometry measurements make it clear that Nd_2Al is at least "predominantly

ferromagnetic" [4]. This leaves three possible magnetic structures, namely Γ_3, Γ_5 and Γ_7. The only allowed ordering directions for the Nd(4c) magnetic sublattices with $\mathbf{k} = [0\ 0\ 0]$ are either along the b-axis (Γ_5) or in the ac-plane (Γ_3 and Γ_7). The best refinement to the measured diffraction pattern is with the Nd(4c) sites ordered ferromagnetically in the F_Y mode, along the crystal b-axis, corresponding to the Γ_5 representation. Of particular note are the Nd^{3+} magnetic moments, 1.2(2) μ_B and 2.3(2) μ_B, both of whch are substantially smaller than the 'free-ion' value of 3.27 μ_B for the Nd^{3+} ion.

Our refinements show that there are no antiferromagnetic components associated with the magnetic order of the Nd sublattices, either as an intrinsic canting or as clusters. The reduction in the Nd magnetic moments in Nd_2Al is therefore most likely the result of crystal-field quenching. It is known that the crystal-field acting on the R^{3+} sites in the orthorhombic R_2Al compounds is quite large and this, coupled with the fact that the magnetic exchange interaction is relatively weak (T_C = 36 K for Nd_2Al), leads to the observed quenching. In a ^{169}Tm Mössbauer study of Tm_2Al, for example, one of us showed that the strong effect of the crystal-field leads to unusually slow electronic relaxation of the Tm^{3+} ion [11].

4. Conclusions

We have determined the magnetic structure of Nd_2Al by neutron powder diffraction. The magnetic ordering temperature is 36(2) K. At 1.7 K, the magnetic order of the Nd(4c) sublattices is ferromagnetic along the orthorhombic b-axis. Significant crystal-field quenching of the Nd^{3+} magnetic moments is present.

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