

Magnetic and Chemical Order in Heusler Alloys Containing Neodymium and Molybdenum

N. A. Khalefa *

*(Physics Department, Al-Anbar University,)

ABSTRACT

Saturation magnetization X-ray and neutron diffraction measurements have been made on alloys at the compositions Nd_2MoCd , Nd_2MoZn , Nd_2MoCu , Nd_2MoTi , Nd_2MoAg , Nd_2MoZr and $NdMoZr$. The alloys containing Cu and Ag have fully ordered Heusler, $L2_1$, Chemical structure. Nd_2MoCd is similarly ordered but with some partial Mo-Cd disorder. The alloys Nd_2MoZn and Nd_2MoTi each contain a secondary phase in addition to the primary Heusler phase Nd_2MoZr contains the two phases $Nd_{122}MoZr$ and Nd and $NdMoZr$ is ordered in the Cl_b structure. The alloys containing the group IIIB or IVB elements Cd, Zn, Cu, Ti or Ag are ferromagnetically ordered, with the magnetic moment associated with the ordered Nd sites. The two alloys containing the group VB elements Zr have vacant chemically ordered 'Nd' sites but are paramagnetic.

Keywords - Neutron, Nd_2MoCd , Heusler, IVB elements and VB elements.

I. INTRODUCTION

The HEUSLER alloys are group of ternary. Intermetallic compounds formed at the stoichiometric composition X_2YZ . With the doubly ordered $L2_1$ type structure shown in Fig. 1. Generally in most Heusler alloys X is a transition metal Y is normally Mn and Z is a B sub-group such Cd, Zn, Cu, Ti, Ag, In or Zr. In particular $L2_1$ alloys occur in the series Cu_2MnZ [1,3,4], Ni_2MnZ [5,8,9], Pd_2MnZ [5,8-10], Au_2MnZ [11, 12, 14], and Co_2MnZ [5, 6, 8, 13,18]. Investigations of their magnetic properties have shown that in all but the latter series the alloys have ordered magnetic structure with moments of approximately $4 \mu_B$ on the Mn site most of the alloys are ferromagnetic but some anti-ferromagnetic structure have been observed [7,8,10].

In the series Co_2MnZ it was shown [18] that. In addition to the moment on the Mn sites a substantial moment also associated with the Co sites. This results in an increase in the exchange interactions and correspondingly higher Curie temperature. The present work reports a similar investigation of the chemical and magnetic structure of the Heusler alloys series Co_2TiZ and the Cl_b type alloys $Nd Mo Zr$ in which the former magnetic Mn atoms have been replaced by Molybdenum atoms.

II. ORDER IN $L2_1$ AND Cl_b TYPE ALLOYS

The unit cell illustrated in Fig 1. is comprised of four interpenetrating f.c.c sublattices A,B,C,D. In this structure Bragg reflections occur when the miller indices are unmixed. This gives rise to the following three types of structure amplitude.

h, k, l odd

$$F(111) = 4 [(f_A - f_C)^2 + (f_B - f_D)^2]^{1/2}$$

$$h+k+l = 4n+2$$

$$F(200) = 4 [f_A - f_B + f_C - f_D] \dots\dots(1)$$

$$h+k+l = 4n$$

$$F(220) = 4 [f_A + f_B + f_C + f_D]$$

Where f_A, f_B, f_C and f_D are the average scattering factors for the A,B,C and D sites respectively.

Alloys at the composition Nd_2MoI , ordered in the $L2_1$ Heusler structure have the A and C sites occupied by Nd atoms, the B sites occupied by Mo atoms and the D sites occupied by the Z atoms, for such alloys the structure amplitudes reduce to

$$F(111) = 4 [f_{MO} - f_Z]$$

$$F(200) = 4 [2f_{MO} - f_Z] \dots\dots(2)$$

$$F(220) = 4 [2f_{MO} + (f_{MO} - f_Z)]$$

Reflections of the type (111) with h,k,l all odd and these of type (200) with h,k,l all even and $(h+k+l) = (4n+2)$ are super-lattice reflections with intensities dependent upon the state of order. The remaining reflections of type (220) with h,k,l even and $(h+k+l) = 4n$ are order independent principal reflections. Johnston and Hall [19] and Webster [20] discussed in detail the determination of the state of order in ternary alloys of this type from X-ray and neutron diffraction measurements. If random disorder occurs all the superlattice reflections are reduced in intensity by the same factor S^2 , where S is a long-range ordering parameter defined as in the binary case i.e

$$S = \frac{C - C_D}{C_O - C_D}$$

Where C, Co and CD represent the numbers of atoms located on their ordered sites in the existing states in the fully ordered state, and in the randomly ordered state respectively. However, if preferential disordering occurs between certain sites only, the two sets of super-lattice reflections are affected differently. The preferential disordering may be described in terms of an additional preferential disordering parameter α . Although it is not in general possible to define uniquely the state of long-range order in a ternary compound using only these two ordering parameters in practice provided the preferential disordering is not too large and the scattering factors differ sufficiently-graphs of F^2 vs α often indicate that one solution is possible in this series, Nd_2MoZ , as in the series Co_2MnZ only one type of preferential disordering is observed, namely B_2 type disorder between atoms of the B and D sites only, in the alloys containing Neodymium in this case α (Mo-Z) is defined as the fraction of Mo atoms preferentially occupying D sites. In disordering of this type of the even super-lattice lines are affected only by any random disorder and are reduced in intensity by the factor S^2 , whereas the odd super-lattice lines are reduced by the factor $(1-2\alpha)^2 S^2$. When $\alpha=0.5$ complete order exists between the Mo and Z atoms, the two sites cease to be distinguishable, and the structure reduces to the B_2 , CSCI type [29].

The small difference between the atoms scattering factors of Nd and Mo and absorbing correction, often permit only a qualitative indication of the type of chemical order from X-ray data with neutron absorption corrections are usually small and essentially isotropic, and the scattering lengths of Nd and Mo differ both in magnitude and in sign. This normally permits an unequivocal quantitative description of the chemical order to be made especially when also supported by the X-ray data [15].

In Cl_b type compounds, such as $NdMoZr$, the problem of ordering is essentially that of a quaternary alloy containing three types of atom and one vacancy orders on the four sites. A general solution is again not possible but, providing the degree of disorder is not large, the use of both X-ray and neutron diffraction data usually enables a unique description of the long-range order to be made. The neutron diffraction Bragg peaks from ferromagnetic materials contain both nuclear and magnetic contributions. For unpolarised neutrons there is no coherence between the nuclear and magnetic scattering amplitudes and so the total structure factor F is related to the nuclear and magnetic structure factor F_n and F_m respectively by the equation $F^2 = F_n^2 + q^2 F_m^2$

F_n is obtained by substituting the appropriate nuclear scattering lengths b^{02} in equation (2) and F_m by substituting the appropriate magnetic scattering length P where

$$P = 0.269 \mu f_0$$

μ is the atomic magnetic moment, in Bohr magnetons, and f_0 is the magnetic form factor. The form factor used for Nd was that given by Nathans and Paoletti [21, 27] $q^2 = \sin^2 \psi$. Where ψ is the angle between the magnetic and scattering vectors. In cubic ferromagnetic materials $q^2 = 2/3$ in zero field, but may be reduced to zero by the application of a saturating magnetic field along the scattering vector. The application and removal of such a field can be used to separate the nuclear and magnetic structure factors, and hence to obtain details of the chemical and magnetic structures.

III. EXPERIMENTAL PROCEDURES

a) Alloys Preparation

Alloys were fabricated in the form of 30 g ingots by melting together the appropriate quantities of spectrographically pure elements in an Argon arc furnace specimens suitable for magnetic analysis were cut from different parts of the ingots and the remainders were crushed to provide power samples for X-ray and neutron diffraction analysis. The samples were sealed in evacuated quartz ampoules homogenized at 2620°C for 24 hr then quenched. The weight losses recorded on melting are shown in (table 1).

b) Structure analysis

X-ray powder diffraction photographs were taken using a Philips camera with filtered Iron $K\alpha$ radiation to minimize fluorescence. Lattice parameters were in agreement with those expected from density measurements [30]. Preliminary assessments of the chemical structures were made from visual estimates of the line intensities. Quantitative information about the chemical ordering and magnetic structure was obtained from neutron diffraction power measurements made at A.E.R.E Harwell [28]. The measurements were made using the CURRAN diffractometer of wavelength 1.06 Å from the DID0 reactor [17]. The powder samples were enclosed in vanadium cans and neutron diffraction patterns were obtained at 4.2° 2θ and at room temperature. A superconducting magnet was employed to apply a horizontal magnetic field at 7 KOe across the sample when necessary.

c) Magnetic analysis

With the exception of NdMoZr all alloys were ferromagnetic with Curie temperatures between 100 and 400°k. Specimens for magnetic investigation were shaped into rough ellipsoids approximately 3.5mm long by 1.5mm dia. and chemically polished to remove surface contaminants Bulk magnetization measurements were made using a sucksmith ring-balance and a Foner vibrating sample magnetometer (VSM). Calibrated against pure nickel using the data of Crangle and Good Man [22]. Magnetic isothermals were measured in applied field up to 16 KOe at series of temperatures between 4.2 and 400°k. demagnetization corrections were estimated and subtracted. The spontaneous magnetization σ_0T was obtained from extrapolation of σ^2 Vs H/σ plots [23]. Extrapolation of the resulting spontaneous magnetization curves to 0°k was made using the $T^{3/2}$ law to give the saturation magnetization σ_0 . Paramagnetic susceptibilities were measured using a sensitive sucksmith ring-balance calibrated against pure palladium ($\chi_{pd} = 5.23 \times 10^{-6}$ e.m.u g^{-1} at 293°k). Measurements were made at several applied fields up to 15 KOe.

Nd₂MoCd

Gladyshevskij et al. [23] investigated an alloy at the composition Nd₂MoCd and concluded from X-ray back reflection photographs that it had Heusler. L₂₁ structure with a lattice parameter 5.847 Å.

An alloy was prepared at the composition Nd₂MoCd its structural properties, together with those of other alloys investigated in this series, are summarized in Table 1. The magnetic properties of the alloys are summarized in Table 2 and Fig. 2. Nd₂MoCd is ferromagnetic with a small moment and a Curie temperature below room temperature. The nuclear and magnetic structure factors, calculated from neutron diffraction data collected above and below the Curie temperature respectively at 293 and 4.2°k, are compared with theoretical values in Table 3. The comparison of nuclear structure factors indicates that Nd₂MoCd is essentially ordered in the L₂₁ structure but with a small amount of preferential (Mo-Cd) disorder. Corresponding to $\alpha(\text{Mo-Cd}) = 0.029$, as shown by the reduction in the 'odd' super-lattice structure factors only. The magnetic structure factors obtained by subtraction of the normalized room temperature data from the 4.2°k data agree well with the structure factor calculated of the assumption that the entire measured moment is associated with the Nd sites.

Nd₂MoZn

X-ray diffraction photographs taken of an superlattice lines would have intensities of the order of 10^{-4} that of the intensities of the principal lines. The alloy was ferromagnetic with a curie temperature 130°k, as shown in Table 2 and Fig. 2.

The existence of a fully ordered Heusler structure and the location of the alloy fabricated at the composition Nd₂MoZn indicated a polyphases structure containing about 50 percent of a Heusler phase with a lattice parameter 5.740°Å. the alloys was ferromagnetic with curie temperature 375°k

Nd₂MoCu

Markiv et al. [24] reported, on the basis to X-ray data that a Heusler alloy with a lattice parameter 5.848°Å could be made at the composition Nd₂MoCu An alloy was fabricated at the stoichiometric composition; X-ray data indicated an L₂₁ structure with a lattice parameter 5.848°Å. The 'odd' super-lattice lines were weak but visible but the 'even' superlattice lines could not be resolved. This is consistent with, but not conclusive evidence of an ordered Heusler structure for which the 'even' magnetic moments at the Nd sites are confirmed by the room temperature and 4.2°k neutron diffraction data compared in Table 4.

Nd₂MoTi

Gladyshevskij et al. [23] concluded from X-ray back reflection powder photographs that an alloy at composition Nd₂MoTi had the Heusler structure with a lattice parameter 5.823°Å.

An alloy was prepared at the stoichiometric composition. The ensuing X-ray photograph showed a large proportion (about 80 percent) of Heusler phase. The alloy was ferromagnetic with a Curie temperature of 386°k.

Nd₂MoIn

Attempts were made to form an alloy at the composition Nd₂MoIn but they were unsuccessful because did not enter into the solution.

Nd₂MoAg

Gladyshevskij et al. [23], reported from X-ray observation a Heusler alloy at the composition Nd₂MoAg with a lattice parameter 6.059°Å. Fujita et al [25], confirmed the X-ray structure and investigated the bulk magnetic properties.

A single phase alloy was made at the composition Nd₂MoAg. The X-ray data was consistent with that expected for a single phase Heusler structure with a lattice parameter 6.073°Å. the alloy was strongly ferromagnetic with a Curie temperature of 359°k, as indicated in Table 2 and Fig 2.

Neutron diffraction patterns were recorded at 4.2°k with and without a magnetic field to separate the nuclear and magnetic contribution to the diffraction peaks. The theoretical and measured structure factors are compared in Table 5. The results confirm the existence of a fully ordered Heusler structure with magnetic moments associated with the Nd sites.

Nd₂MoZr

An alloy was prepared at the stoichiometric composition Nd₂MoZr X-ray photographs indicated a 2 phase system. The small amount of secondary phase was indexed at f.c.c with a lattice parameter

3.547 Å consistent with small regions of free Nd. The principal phase had a f.c.c superlattice with a lattice parameter 5.882 Å and line intensities consistent with a structure intermediate between the $L2_1$ and Cl_b structures and composition $NdXMoZr$. The alloys was strongly ferromagnetic with a saturation magnetization $27.2 \text{ e.m.u.g}^{-1}$, over the temperature range 4.2-750 °k the magnetization decreased by only 10 percent, indicating a Curie temperature in excess of 1000°k commensurate with that of free Neodymium. Assuming the moment of the free Neodymium to be $1.716 \mu_B$ the amount of free Neodymium was estimated to be 17 percent. Identification of the primary phase from the neutron diffraction data summarized in Table 6, is difficult because of the relatively small scattering length of Neodymium. The nuclear peaks are in sensitive in the case of this particular compound Nd_xMoZr to ordering intermediate of $L2_1$ and Cl_b (i.e $2 > X > 1$). The best fit was however obtained for $x=12$. A comparison of the field on/off data showed no moment associated with the principal phase.

NdMoZr

Gladyshevskij et al. [23] reported from X-ray investigation that $NdMoZr$ had the Cl_b structure with a lattice parameter 5.884 Å. Terada et al. [26] reported that the alloy was paramagnetic with temperature independent susceptibility with a slight field independence dependent upon previous heat-treatment.

An alloy was prepared at stoichiometric composition $NdMoZr$. The X-ray photographs indicated a single phase structure with Cl_b ordering and a lattice parameter of 5.884 Å.

The magnetic results were consistent with a temperature independent susceptibility of $1.9 \pm 0.3 \times 10^{-6} \text{ e.m.u.g}^{-1}$ together with a very small ferromagnetic precipitate with magnetization $\sigma_{00} = 0.018 \pm 0.003 \text{ e.m.u.g}^{-1}$ giving a slight field dependence.

The neutron diffraction structure factors obtained at 4.2°k compared in Table7 are consistent with an alloy fully ordered in the Cl_b structure. There was no indication of any second phase on the neutron diffraction pattern.

IV. DISCUSSION

The chemical and magnetic ordering of these alloys show marked similarities with those already reported for the alloy series Nd_2MoZ [18], except that unlike the Mn, Mo carries no magnetic moment. The chemical structure is very similar. In both series the alloys containing Cd exhibit some B_2 type disorder, and alloys containing Zr have vacant 'Nd' sites. The series differ in that the alloys Nd_2MnAg and Nd_2MoTi are single phase whereas the alloys Nd_2MoZn and Nd_2MoTi are not. As in the Nd_2MnZ series alloys containing sub-group IVB elements

have smaller lattice parameters than those containing adjacent IIIB elements and within each sub-group the lattice- parameters increase with atomic number. The magnetic properties depend upon the B sub-group from which the element Z comes. As with the Nd_2MnZ series the highest Curie temperature and largest moments are observed when Z is from group IVB, and lowest when from group IIIB.

The Cl_b type alloys $NdMoZr$ differs from the Heusler alloys in that it contains only half the number of Nd atoms. The alloy is paramagnetic with the Nd atoms arranged on an f.c.c sub-lattice with shortest Nd-Nd distances $a/2 \sim 4.1 \text{ Å}$.

Whereas the Heusler alloys have their Nd atoms ferromagnetically ordered on a simple cubic sub-lattice with shortest Nd-Nd distances $a/2 \sim 3 \text{ Å}$.

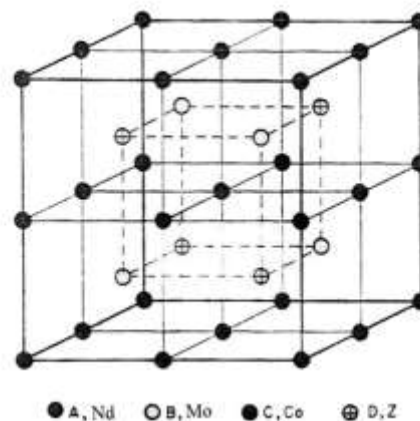


Fig. 1: The Heusler structure

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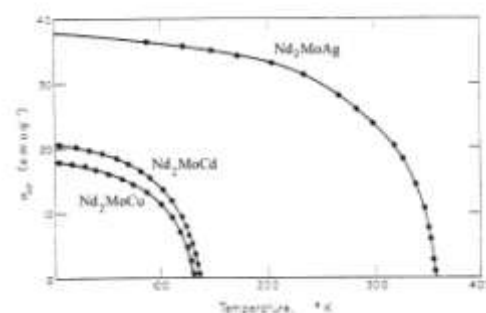


Fig. 2: Spontaneous magnetization $\sigma_0 \tau$ Vs temperature for Nd_2MoCd , Nd_2MoCu and Nd_2MoAg

Table1. Details of alloys structure and preparation

Alloys	Weight loss (%)	Lattice parameter (Å)	Structure
Nd ₂ MoCd	0.2	5.848	L ₂₁ +some B ₂
Nd ₂ MoZn	0.8	5.704	50% L ₂₁ +polyphase
Nd ₂ MoCu	0.7	5.845	L ₂₁
Nd ₂ MoTi	0.3	5.831	80% L ₂₁ +polyphase
Nd ₂ MoAg	0.5	6.073	L ₂₁
Nd ₂ MoZr	1.0	5.882	85% Co ₁₂ MoZr+15% Nd
NdMoZr	1.0	5.884	Cl _b

Table2. A summary of the principal magnetic features of alloys Nd₂MoCd, Nd₂MoZn, Nd₂MoCu, Nd₂MoTi, Nd₂MoAg and NdMoZr.

Alloy	Curic temp. (k)	σ ₀₀ (e.m.u.g ⁻¹)	μ ₀₀ /mol (μ _B)	μ ₀₀ /Nd (μ _B) (from neutron diffraction)
Nd ₂ MoCd	138±3	20.6 ± 0.6	0.71 ± 0.03	0.35 ± 0.1
Nd ₂ MoZn	375 ± 4	31.4 ± 0.9	0.73 ± 0.03	0.37 ± 0.1
Nd ₂ MoCu	130 ± 3	17.8 ± 0.5	0.75 ± 0.03	0.4 ± 0.1
Nd ₂ MoTi	386 ± 4	41.6 ± 1.0	0.77 ± 0.03	0.45 ± 0.1
Nd ₂ MoAg	359 ± 4	37.8 ± 1.0	1.93 ± 0.07	1.03 ± 0.1
NdMoZr	361 ± 4	39.8 ± 1.0	1.88 ± 0.06	1.05 ± 0.1

Table3. A comparison of the calculated and observed neutron diffraction structure factor for Nd₂MoCd

hk1	L ₂₁	F ² Nuclear		F ² magnetic	
		Calculated α(Mo-Cd)=0.029	observed	calculated μ (Nd)=(0.37) μ _B	Observed
111	0.477	0.433	0.433 ± 0.008	0	0.003 ± 0.01
200	0.242	0.245	0.235 ± 0.005	0.019	0.015 ± 0.008
220	0.263	0.260	(0.260)	0.013	(0.014)
311	0.478	0.423	0.414 ± 0.008	0	0.00 ± 0.01
222	0.249	0.245	0.236 ± 0.005	0.008	0.009 ± 0.008

Table4. A comparison of the calculated and observed neutron diffraction structure factor for

Nd ₂ MoZn				
hk1	F ² Nuclear		F ² magnetic	
	calculated L ₂₁	observed	calculated μ (Nd)=(0.40) μ _B	Observed
111	1.124	1.143 ± 0.008	0	0.00 ± 0.01
200	0.014	0.014 ± 0.008	0.024	0.025 ± 0.008
220	0.774	(0.774)	0.018	(0.018)
311	0.124	1.111 ± 0.008	0	0.001 ± 0.01
222	0.014	0.015 ± 0.005	0.011	0.012 ± 0.008

Table5. A comparison of the calculated and observed neutron diffraction structure factors for Nd₂MoAg.

hk1	F ² nuclear		F ² magnetic	
	calculated L ₂₁	observed	μ (Nd) = 1.03 μ _B	observed
111	0.903	0.870	0	0.006 ± 0.01
200	0.053	0.074	0.171	0.173 ± 0.01
220	0.593	(0.593)	0.131	0.147 ± 0.01
311	0.903	0.930 ± 0.01	0	0.00 ± 0.01
222	0.053	0.035 ± 0.01	0.085	0.080 ± 0.01

Table 6. A comparison of calculated and observed neutron diffraction structure factor for Nd₂MoZr

hk1	F ² nuclear calculated		observed	F ² magnetic observed
	L ₂₁ , Nd ₂ MoZr	Nd _{1.2} MoZr		
111	0.774	0.814	0.807 ± 0.01	0 ± 0.01
200	0.090	0.010	0.009 ± 0.05	0 ± 0.005
220	0.490	0.250	(0.250)	0 ± 0.005
311	0.774	0.814	0.817 ± 0.01	0 ± 0.01

Table 7. A comparison of calculated and observed neutron diffraction structure factor for NdMoZr

hk1	F ² Calculated Cl _b	F ² nuclear observed
111	0.837	0.827 ± 0.01
200	0.003	0.009 ± 0.05
220	0.203	(0.203)
311	0.837	0.815 ± 0.01
222	0.003	0.007 ± 0.05

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