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Evidence of weak ferromagnetism in the C14 Laves phase $(Fe_{1-x}Co_x)_2Nb$ system

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ABSTRACT

In this work are studied the structure and magnetic properties of the $(Fe_{1-x}Co_x)_2Nb$ alloys with X-ray diffraction and magnetization measurements. The results confirm the formation of the C14 Laves phase in the Co concentration range $0 \le x \le 0.55$. The unit cell volume decreases with the increase of the cobalt concentration and is larger than in Fe_2Nb. Magnetization measurements indicate that this system is weakly ferromagnetic at low temperatures for $x \le 0.55$. From the behavior of the $M(T)$ curves a cluster-glass-like mechanism is predicted for this system below 50 K.

1. Introduction

Most of hexagonal C14 compounds are weakly magnetic or nonmagnetic. C14 Fe_2Nb was found to be weakly antiferromagnetic (AF) below 10 K. However, this is impaired by a slight deviation of the composition from the stoichiometry; deviation towards both Nb- and Fe-excess sides leads to a ferromagnetic ordering [1–5]. Actually it has been found that both ferro- and antiferromagnetic spin fluctuations coexist in Fe_2Nb [4]. Theoretical predictions from band structure calculations have indicated that the antiferromagnetic state is nearly degenerate in energy with a paramagnetic state, although the former has been confirmed as the most stable [6,7]. The Fermi level in Fe_2Nb lies on the right side slope of a prominent peak. Hence, in the framework of the rigid band approximation, the stability of the magnetic state of this compound is very sensitive to substitutions for Fe, as a consequence of the variation of the d-electron number. Moreover, volume effects are also expected to play an important role in the stability of the magnetic ordering.

The magnetic properties of the C14 hexagonal Laves phase Fe_2Nb compound is thus very sensitive to the concentration ratio of Fe and Nb. Recent experimental results have shown that the Al substitution for Fe enhances the antiferromagnetism of Fe_2Nb, the V, Cr and Mn substitutions leads to the appearance of ferromagnetism, while the system becomes nonmagnetic with the Co and Ni substitutions [5]. In this study we investigate the effect of the substitution of Co for Fe on the structure and magnetic properties of the $(Fe_{1-x}Co_x)_2Nb$ system. The Nb content was kept fixed at 33 at.%. We found that the C14 Laves phase is formed in this system for a Co concentration up to $x = 0.55$, and it is weakly ferromagnetic at low temperatures.

2. Experimental

Three specimens of $(Fe_{1-x}Co_x)_2Nb$ alloys with nominal compositions $x = 0.10, 0.33$ and 0.55 were prepared by arc melting the appropriate quantities of the constituent elements under argon atmosphere. The buttons thus obtained were annealed at 1000 °C for 3 days in evacuated quartz tubes with a further quenching in iced water. The compositions of the studied alloys were determined by energy dispersive X-ray analysis (EPMA). X-ray powder diffraction data were obtained by using the Cu K α radiation collected on a Bragg-Brentano diffractometer using a 2θ step size of 0.05°. $M(H)$ curves were taken at 4.2 K with a vibrating sample magnetometer in fields up to 15 kOe. The temperature dependence of the magnetization

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Table 1
Lattice parameters (a, c) of the hexagonal C14 Laves phase, the c/a ratio, unit cell volume and Curie temperature for the $(Fe_{1-x}Co_x)_2Nb$ alloy system

Alloy	Structure	a (Å)	c (Å)	c/a	V_o (Å^3)	T_C (K)
0.10	Hex	4.903	7.960	1.623	165.71	35
0.33	Hex	4.859	8.012	1.649	163.81	14
0.55	Hex	4.883	7.879	1.613	162.69	28

$M(T)$ was registered from 5 K up to 350 K in a low external magnetic field (10 Oe) using a superconducting quantum interference device magnetometer. The sample was firstly brought in a zero-field-cooled state (ZFC) by cooling it from 390 K down to 5 K under zero applied field, and then, after applying a field of 10 Oe, the measurements were taken on heating. Subsequently the data were collected under the same field while decreasing temperature down to 5 K (FC).

3. Results

The chemical composition of the studied alloys, shown in Table 1, was determined on the homogenized specimens by EPMA (EDS and WDS), which confirmed within the experimental precision the nominal composition. A profile analysis of the X-ray diffraction patterns with the Rietveld method has shown that these alloys indeed are single phased with the structure of the hexagonal C14 Laves phase, as can be seen in the diffractograms shown in Fig. 1. From Table 1 it can be seen that both a and c depend strongly on the composition and present unsystematic behavior with the increase of the Co concentration. The unit cell volume is larger than in Fe_2Nb (159.24 Å^3) [8], and decreases with the increase of the Co concentration up to $x = 0.55$.

In Fig. 2 are shown the $M(T)$ curves of the temperature dependence of the magnetization for these alloys. The ZFC data were taken with rising temperature. Somewhat surprisingly, although weak, a ferromagnetic behavior has been observed at low temperatures in all the studied alloys, on the contrary to earlier investigations, which reported a paramagnetic behavior for this system [5]. The ferromagnetic behavior of these alloys is thus a remarkable new feature. For $x = 0.10$, the collapse of the total magnetization occurs at about 35 K, which was then taken as the Curie temperature T_C at this composition. For $x = 0.33$, $T_C = 14$ K. No other transition was observed. In the ZFC mode, the decrease of the magnetization at low temperatures, below about 35 K, as observed for $x = 0.10$ and $x = 0.55$, is thought to be associated with the existence of frustrated couplings and a spin-glass like behavior. However, the ZFC curve retraces the FC curve (taken on cooling) only above 175 K. Such a irreversibility of the $M(T)$ curves is an indication of a cluster-glass-like mechanism. At $x = 0.55$ the collapse of the magnetization takes place at about 28 K.

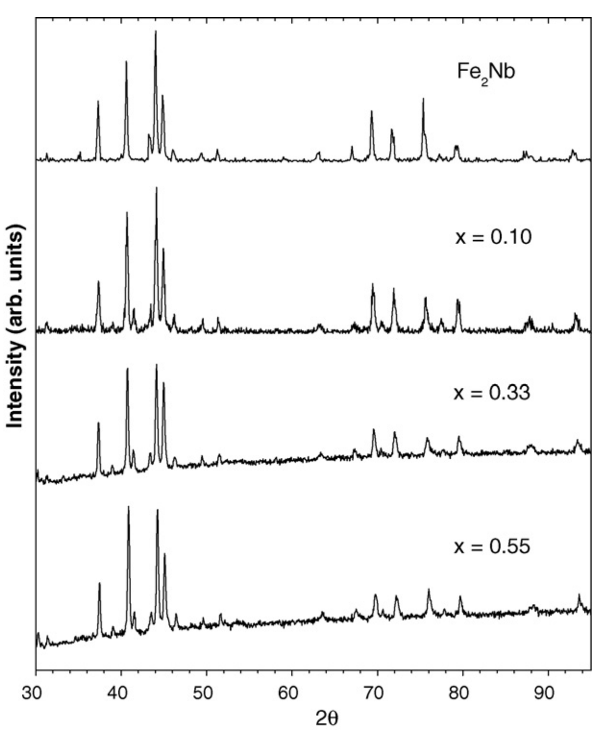

Fig. 1. X-ray diffraction diagrams of $(Fe_{1-x}Co_x)_2Nb$ alloys. A calculated pattern for Fe_2Nb is shown at the top, for the sake of comparison ($a = 4.830$ Å and $c = 7.882$ Å [8]).

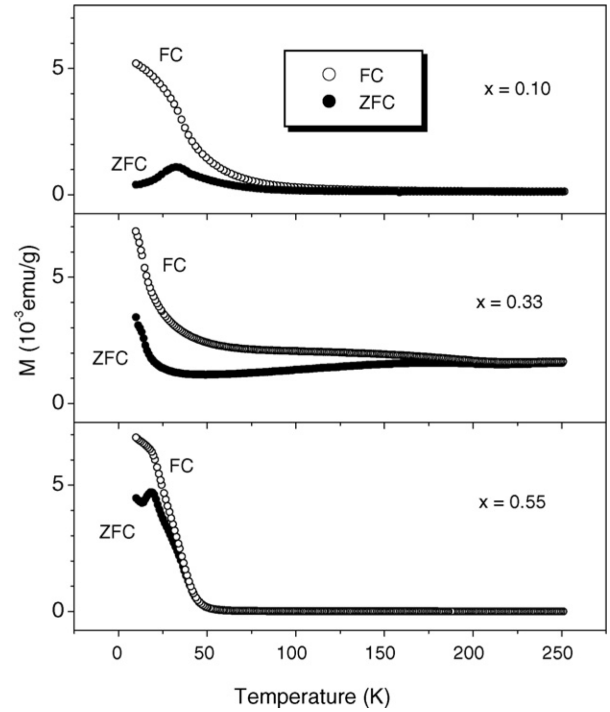

Fig. 2. Field-cooled (FC) and zero-field-cooled (ZFC) curves for $(Fe_{1-x}Co_x)_2Nb$ alloys, taken in a measuring field of 10 Oe.

In Fig. 3 are shown the hysteresis loops for these alloys at 4.2 K in applied fields up to 15 kOe. The registered curves are characteristics of soft ferromagnetic materials. Although saturation was not attained in the applied field, it can be observed from the curves in Fig. 3 that the saturation magnetization M_s increases with the Co concentration. Despite remanence is observed in all the alloys, a small coercivity is only observed for $x = 0.10$.

4. Discussion

It can be seen from Table 1 that the unit cell volume of the Laves phase for $(Fe_{1-x}Co_x)_2Nb$ alloys, for $0 \le x \le 0.55$, is larger than in Fe_2Nb (159.24 Å^3) [8]. According to Yamada et al. [5], ferromagnetism is observed in the C14 Laves phase alloys containing V, Mn and Cr when the cell volume is larger than in Fe_2Nb, at around 161 Å^3. Hence, the statement that the $(Fe_{1-x}Co_x)_2Nb$ alloys are nonmagnetic is based on the assumption that these alloys possess a smaller unit cell volume than in Fe_2Nb. However, the present investigation shows that the cell volume in the $(Fe_{1-x}Co_x)_2Nb$ system indeed is larger than in Fe_2Nb, and the alloys are weakly ferromagnetic at low temperatures. Higher Curie temperatures are observed when the c/a ratio is close to 1.632, as it is in Fe_2Nb. Further-