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NEUTRON DIFFRACTION AND MAGNETOSTRICTION OF CUBIC $La(Fe_xAl_{1-x})_{13}$ INTERMETALLIC COMPOUNDS

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The ferromagnetic and antiferromagnetic states of $La(Fe_xAl_{1-x})_{13}$ were studied by neutron diffraction and magnetostriction measurements. We present a model for the magnetic structure of the antiferromagnetic state, consisting of ferromagnetic clusters, coupled antiferromagnetically. Magnetostriction data exhibit a strong Invar character and contain information about the origin of the magnetic moments.

$La(Fe_xAl_{1-x})_{13}$ intermetallic compounds can be stabilized in the cubic $NaZn_{13}$-type structure with iron concentration x between 0.46 and 0.92 [1]. In this limited concentration range the compound may be found in three magnetic structures (see fig. 1): (i) at low iron concentration a mictomagnetic state was found. (ii) At higher iron concentration a soft ferromagnetic state exists where the Fe-moments follow closely the Slater–Pauling curve. (iii) At the highest iron concentrations an antiferromagnetic state was found, with sharp metamagnetic transitions to the fully saturated ferromagnetic state, accompanied by a large hysteresis of order of 5 T and a strong volume change [2]. The collapse of ferromagnetic order for $x < 0.86$ to antiferromagnetic order for $x > 0.86$ in this Fe-based intermetallic compound may be linked to the collapse of ferromagnetic order in the Fe–Ni Invar alloys and the antiferromagnetism in γ-Fe. Here, comparable mechanisms might be present due to the special crystal structure of $La(Fe, Al)_{13}$, which permits a high Fe–Fe coordination number up to 12 with fcc-like local symmetry.

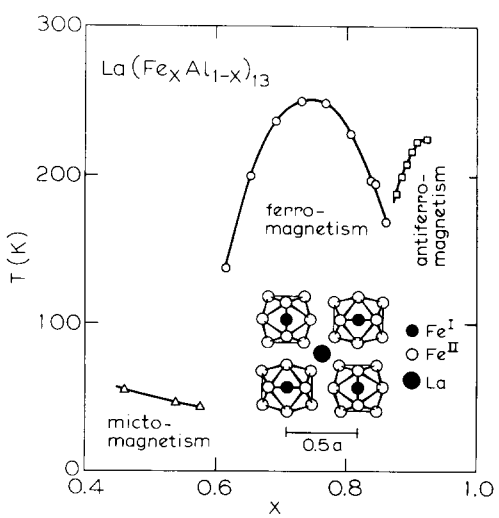

Fig. 1 Magnetic phase diagram of $La(Fe_xAl_{1-x})_{13}$. The inset shows a projection along the c-axis of four icosahedra plus central atoms (clusters).

The cubic $NaZn_{13}$-type structure has two different sites for the Fe atoms in a ratio 1 : 12. The Fe^I atoms and the La atoms form a CsCl structure. Each Fe^I atom is surrounded by an icosahedron of 12 Fe^{II} atoms and thus has an fcc-like local symmetry. The icosahedra are packed in alternate directions so that each unit cell contains 8 icosahedra and therefore 104 spins. Each Fe^{II} atom has 1 Fe^I and 9 Fe^{II} nearest neighbors.

Neutron powder diffraction measurements were performed on a ferromagnetic $x = 0.69$ sample and an antiferromagnetic $x = 0.91$ sample at 4.2 K and at room temperature [3]. The refinement analysis of the diffractograms at room temperature showed that the Fe^I sites are predominantly (> 97%) occupied by Fe. The Al atoms are thus statistically distributed only over the Fe^{II} sites, thereby favoring the fcc-like coordination of the Fe atoms.

The refinement analysis of the diffractogram of the antiferromagnetic $x = 0.91$ sample at 4.2 K is more difficult because each unit cell contains 104 spins. Therefore, we have modelled the system by considering each icosahedron plus central atom as a cluster. This cluster model is the only possible symmetric arrangement, in which each Fe atom is unambiguously assigned to a single cluster. Several antiferromagnetic cluster structures were constructed of which the model illustrated in fig. 2 fitted best with the neutron diffractogram [3]. The best fit was obtained with the central Fe^I spin parallel to the cluster spin, with a different Fe^I moment with respect to the surrounding Fe^{II} moments. The final results of the refinement analysis are $2.14(3)\mu_B$/Fe for the Fe^{II} moment, and $1.10(7)\mu_B$/Fe for the central Fe^I moment. These values have to be compared with $2.13(2)\mu_B$/Fe, obtained by magnetization measurements of the spin flipped state. Our model

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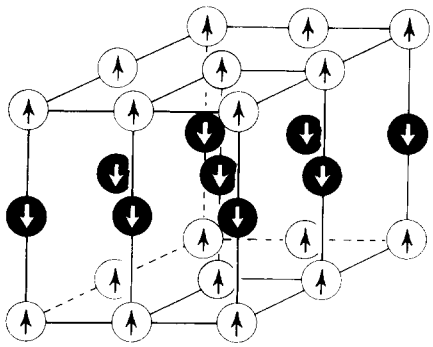

Fig. 2. Model for the antiferromagnetic structure of $La(Fe_xAl_{1-x})_{13}$. Each spin represents a cluster of thirteen atoms.

is consistent with the observation that the spin-flip fields are low when compared to the magnetic ordering temperature. The occurrence of the small magnetic moment ($1.1\mu_B$/Fe) in the antiferromagnetic ground state might reflect the instability of the iron moment in an fcc local environment [4].

The spontaneous volume magnetostriction ($\omega_s = \Delta V/V$) was measured for three ferromagnetic and one antiferromagnetic sample from 6 K up to room temperature [1]. The results are shown in fig. 3 after subtracting the phonon term. At low temperature ω_s can be described by a combined local moment and band model, as proposed by Shiga [5]:

$$\omega_s = \kappa C_{loc} \sum_{ij} \langle \boldsymbol{m}_i \cdot \boldsymbol{m}_j \rangle + \kappa C_{band} \sum_i m_i^2(T)$$

with κ the compressibility, C_{loc} and C_{band} the magnetovolume coupling constants due to the local moments and the band splitting, respectively, and m_i the local moments. For the ferromagnetic state this relation can be reduced to:

$$\omega_s = \kappa(C_{loc} + C_{band}) M^2(T) = \kappa C M^2(T).$$

By comparing the saturation magnetization and the spontaneous volume magnetostriction at 0 K, we find a value for the magneto-elastic coupling constant $\kappa C = 1.75 \times 10^{-8}$ cm^6/emu^2 for both the ferromagnetic and the spin-flipped antiferromagnetic state near the magnetic phase boundary. This value is about twice as large as for bcc-Fe, FeNi Invar and Fe_3Pt.

We now can separate the band and local moment term, by calculating these parameters within the cluster model, obtained from the neutron diffraction results. Going from the antiferromagnetic ground state to the induced ferromagnetic state, the spin–spin correlation function increases from 0.59 to 1.00, and m_i^2 increases

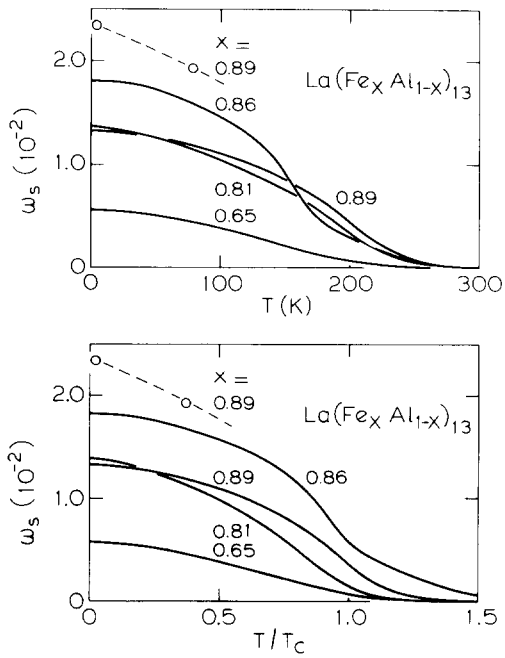

Fig. 3. Spontaneous volume magnetostriction ω_s vs. temperature T and reduced temperature T/T_c.

6%. Experimentally we found that ω_s in the antiferromagnetic state is 57% of the value in the ferromagnetic state. Although the accuracy of these values must not be overestimated, we conclude that the volume-magnetostriction in $La(Fe, Al)_{13}$ mainly arises from the local moment contribution. It can be inferred from fig. 3 that ω_s starts to increase at a distinct temperature (280 K). As we expect the correlation between the spins to scale with T/T_c, this means that the local moments start to increase or even to form from 280 K downwards, independent of the magnetic ordering temperatures.

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