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# Abnormal thermal expansion in $NaZn_{13}$-type $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ compounds


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## Background

- The thermal expansion could give rise to the failure of materials due to undesirable mismatch of coefficient of thermal expansion (CTE). Therefore, the control of thermal expansion has already become the focus studied and front field.
- Over the past 20 years, abnormal thermal expansion (ATE) has been found in some materials. Some expand with decreasing temperature presenting negative thermal expansion (NTE) behaviour, and some almost neither expand nor contract as the temperature changes showing near-zero thermal expansion (NZTE) behaviour. Nevertheless, the challenge still remains for the control of ATE materials. Especially, in most cases, ATE property occurs in only a narrow temperature range.
- The cubic $NaZn_{13}$-type $La(Fe, M)_{13}$-based (M = Si, Al) compounds are recently developed as promising abnormal thermal expansion (ATE) materials. Here, we report that colossal negative thermal expansion (NTE) and near-zero thermal expansion (NZTE) are accessible in $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x = 0, 0.04, 0.06 and 0.08) compounds accompanied with metamagnetic transitions.

## Objectives

- Preparation of $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x =0, 0.04, 0.06 and 0.08) compounds using the arc melting method.
- Study the thermal expansion of $LaFe_{11.4}Al_{1.6}$ and the influence of the partial substitution of Co for Fe on the abnormal thermal expansion behaviours.
- Investigate the correlation between the magnetic ordering and the thermal expansion properties.

## Conclusion

- A group of $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x = 0, 0.04, 0.06 and 0.08) compounds with cubic $NaZn_{13}$-type structure were synthesized and the admixture of Co element into the $La(Fe, Al)_{13}$ compound plays a significant role in controlling thermal expansion.
- The $La(Fe, Al)_{13}$ and $La(Fe_{0.92}Co_{0.08})_{11.4}Al_{1.6}$ compounds show near-zero thermal expansion behaviour at low temperature region (77 K-200 K).
- $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x = 0.04, 0.06 and 0.08) compounds show NTE behaviour accompanied with a metamagnetic transition ferromagnetic-paramagnetic, shifting toward higher temperature region with increasing the amount of Co dopant.
- The ATE behaviour occurs at the entire temperature range (77 K-300 K) for $La(Fe_{0.92}Co_{0.08})_{11.4}Al_{1.6}$ compound, which exhibits ZTE from 77 K to 200 K, and shows NTE from 200 K to 300 K.

## Methods

### Samples preparation

**$La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x = 0, 0.04, 0.06 and 0.08)**

The raw materials of Fe, Co, Al, and La were all at least 99.9% pure. An excess 10 at % of La over the stoichiometric composition was added to compensate for the loss during melting.

The samples were prepared in an arc melting furnace under a high-purity argon atmosphere.

The arc-melted ingots were sealed in a quartz tube filled with high-purity argon gas, subsequently homogenized at 950 °C for 10 days, and finally quenched quickly into ice water.

### Performance test

X-ray diffraction (XRD) was employed to identify the phase purity and crystal structure at room temperature.

The linear thermal expansion data (Δ L/L) were measured using a strain gage over the temperature range of 77K-300K.

Measurements of magnetization at different temperatures were performed on a physical property measurement system (PPMS).

PPMS-14T

## Results

### XRD patterns

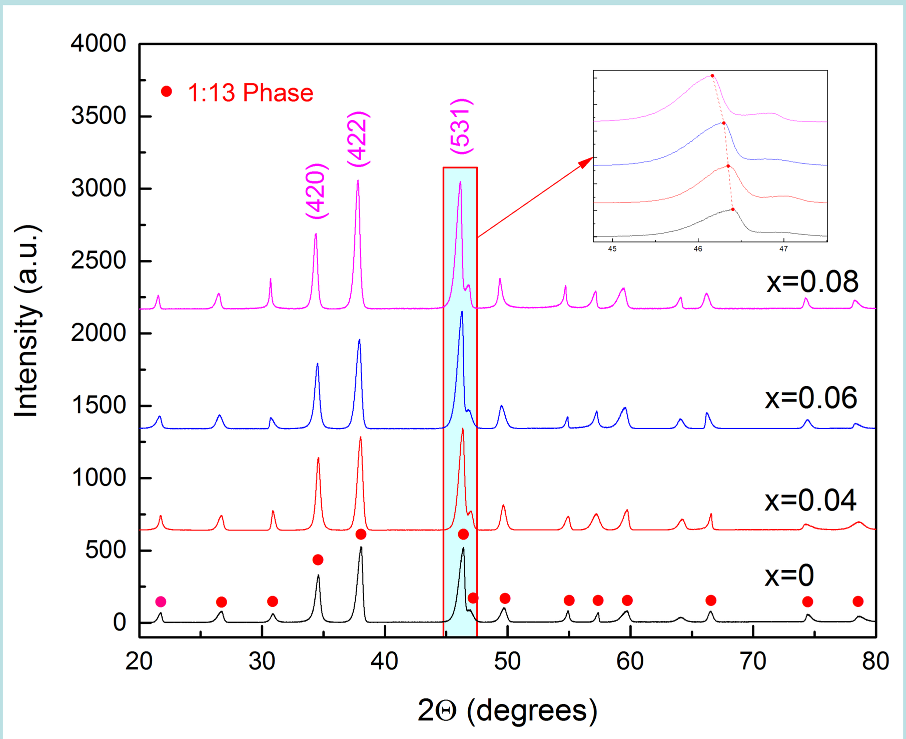


**Figure 1.** XRD patterns for $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x = 0, 0.04, 0.06 and 0.08) compounds. The inset shows an expanded view of the (531) peak for the samples.

- No visible diffraction peaks of pure elements or other detectable secondary phases exist.
- The XRD patterns exhibit a phase with the cubic $NaZn_{13}$-type structure (space group: Fm3c).
- The inset shows that the lattice parameter increases with increasing the Co content, in accordance with the larger size of the atomic radius of Co than that of Fe.

### Thermal expansion

- The coefficients of thermal expansion (CTE) are -9.02 × $10^{-7}$ $K^{-1}$ and -2.89 × $10^{-7}$ $K^{-1}$ in the temperature range 77 K-200 K for $La(Fe, Al)_{13}$ and $La(Fe_{0.92}Co_{0.08})_{11.4}Al_{1.6}$ compounds, respectively, indicating a near-zero thermal expansion at low temperature.
- Giant negative thermal expansion occurs in the samples of $La(Fe_{1-x}Co_x)_{11.4}Al_{1.6}$ (x = 0.04, 0.06 and 0.08). Meanwhile, with increasing the amount of Co from x = 0.04 to x = 0.08, the NTE operation-temperature window moves toward higher temperature region with a slight reduction in CTE.

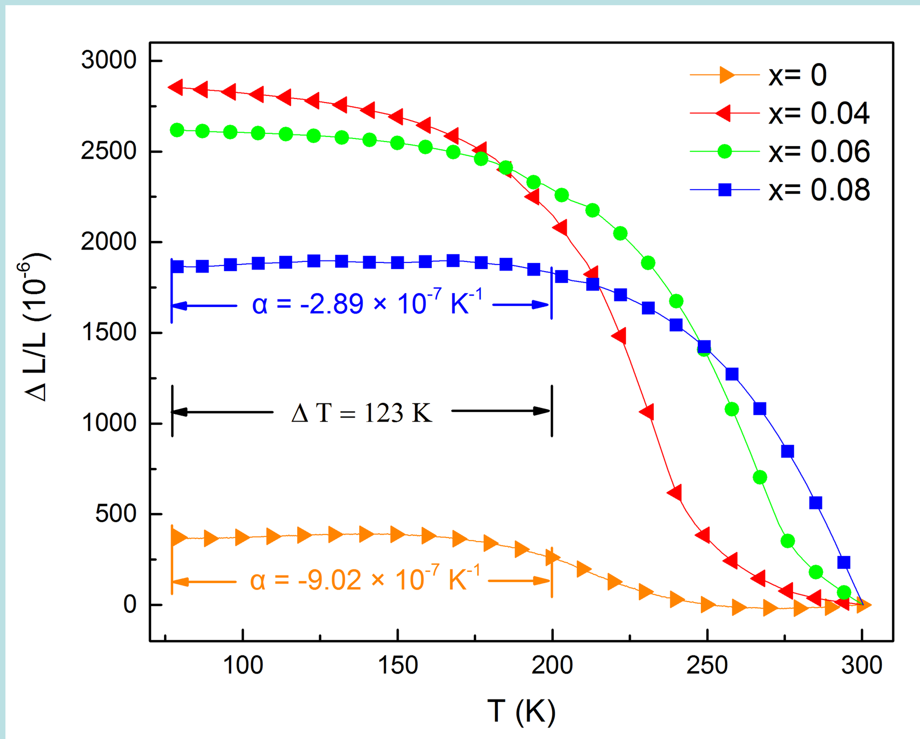


**Figure 2.** Temperature dependence of linear thermal expansion Δ L/L (reference temperature: 300 K) from 77 K to 300 K.

### Magnetic properties

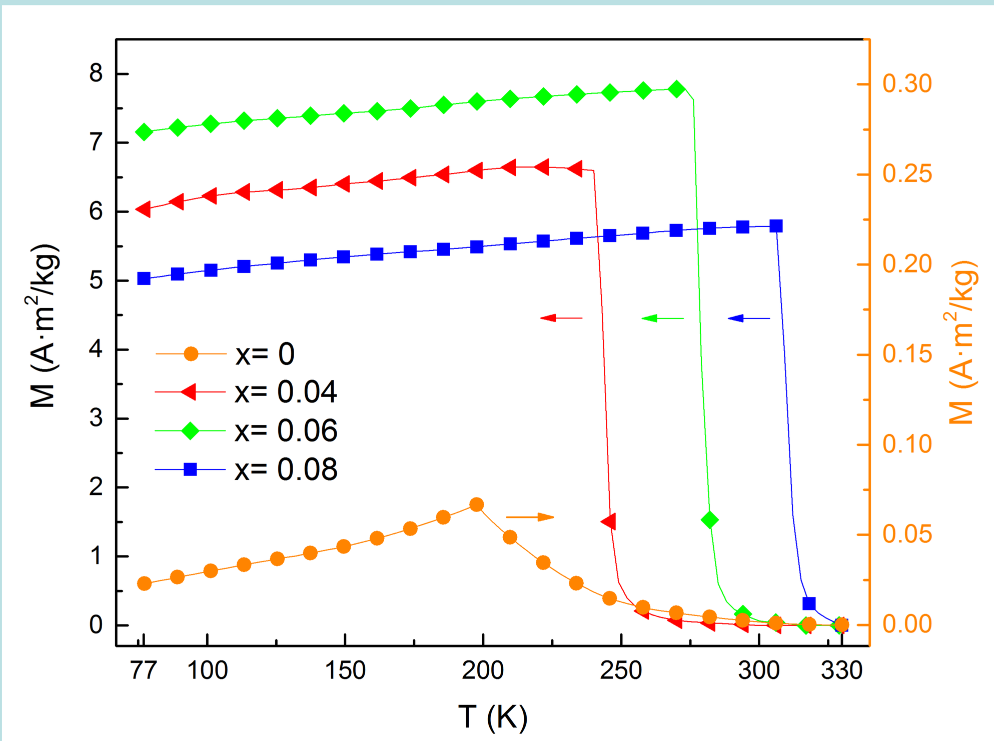


**Figure 3.** Temperature dependence (77 K-330 K) of magnetization $M(T)$ measured under a field of 100 Oe.

- Introducing a small amount of Co can convert the antiferromagnetic (AFM) coupling to the ferromagnetic (FM) one for $LaFe_{11.4}Al_{1.6}$ compound with an AFM ground state.
- The large values of Δ L/L is attributed to the magnetic transition from AFM to FM by doping Co at the ground state.
- The $T_C$ moves to higher temperature with the increasing amount of Co element, which is in good agreement with the temperature dependence of linear thermal expansion.


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