Supplementary information for

Abnormal thermal expansion property in cubic NaZn$_{13}$-type La(Fe, Al)$_{13}$ compounds

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**Figure S1.** X-ray diffraction patterns for samples of LaFe$_{13-x}$Al$_x$ (x=1.8, 1.9, 2.1, 2.3, 2.5 and 2.7) at 300K.

Figure S1 presents XRD patterns for all samples of LaFe$_{13-x}$Al$_x$ (x=1.8, 1.9, 2.1, 2.3, 2.5 and 2.7). From this figure, the indices of crystal face indicate that these samples have a dominating phase with the NaZn$_{13}$-type structure. Also, there exists visible miscellaneous diffraction peaks in the X-ray diffraction patterns, which reveals the existence of $\alpha$-Fe phase according to the analysis of XRD data. As a normal positive thermal expansion material, $\alpha$-Fe does not perform negative thermal expansion (NTE) behavior. Therefore, it is concluded that the NTE property of the LaFe$_{13-x}$Al$_x$ compounds comes from the 1:13 phase due to the unique magnetic transition properties. Moreover, it is easy to understand that there would be more remarkable NTE performance if the purity of 1:13 phase is higher. The impurity of the LaFe$_{13-x}$Al$_x$ compounds is caused by various reasons, such as the oxidation of raw materials, heterogeneous melting and inappropriate annealing time. We believe that the enhanced purity suggests more potential applications of the LaFe$_{13-x}$Al$_x$ compounds as NTE materials.
Figure S2. Temperature dependence of linear thermal expansion $\Delta L/L$ (reference temperature: 300K) from 4.2K to 300K for samples of $\text{LaFe}_{13-x}\text{Al}_x$ ($x=2.5, 2.7$).

Figure S2 displays a larger version of $\Delta L/L$-T curves for samples of $\text{LaFe}_{13-x}\text{Al}_x$ ($x=2.5, 2.7$). In this figure, $\Delta L/L$-T curves decrease above 250K, which act the same behavior as that of normal positive thermal expansion. However, below the temperature of 250K, the two curves undergo slight change: increase first, then decrease and begin to rise again with the decreasing temperature. Especially, for the sample of $x=2.5$, the average CTE is $-1.69\times10^{-6}\text{K}^{-1}$ between 155K and 245K in which the $\Delta L/L$-T curve increases with the decrease of temperature. Similarly, the average CTEs for 80K-155K and 5K-80K are $0.45\times10^{-6}\text{K}^{-1}$ and $-1.23\times10^{-6}\text{K}^{-1}$, respectively.

As discussed in our communication, NTE is induced by magnetovolume effect (MVE) in the $\text{La(Fe, Al)}_{13}$ compounds. When the volume expansion caused by MVE almost equals the contraction due to the atomic anharmonic vibration, the $\text{La(Fe, Al)}_{13}$ compounds perform zero thermal expansion (ZTE) behavior. Although the $\Delta L/L$-T curves changes slightly, the average coefficient of thermal expansion (CTE) are too small to expand or contract for the $\text{La(Fe, Al)}_{13}$ compounds. Such unique thermal property suggests these compounds as promising ZTE materials.