Experimental details

Growth procedure:

The single crystal of NHO was grown using a traveling floating zone furnace technique starting from a polycrystalline NHO sintered rod. Because of the high melting point of NHO, a furnace equipped with high-performance Xenon lamps was necessary. The polycrystalline NHO source powders for the floating zone growth were synthesized by a calcination of HfO_2 (Alfa-Aesar, 99.9 %) and $Nd(OH)_3$. $Nd(OH)_3$ was prepared by soaking Nd_2O_3 (Alfa-Aesar, 99.9 %) powder in distilled water. The resulting $Nd(OH)_3$ was dried at 60 °C overnight. Before using the $Nd(OH)_3$ powders, phase purity was assured by X-ray powder diffraction (XRD). Subsequently, a 1:1 stoichiometric mixture of $Nd(OH)_3$ and HfO_2 powders was ground in an agate mortar, loaded into an alumina crucible and calcinated at 1500 °C for 48 hours. Grinding and calcination were repeated until a phase pure NHO powder was obtained. For the floating zone crystal growth the powder was isostatically pressed to a rod (diameter ∼8 mm) and sintered at 1500 °C. the floating zone growth was carried out in a 8 bar oxygen atmosphere, further details can be found in Ref. 1.

Measurement details:

XRD powder diffraction patterns were collected on a STOE Stadi P and a Bruker D8-Advance diffractometer using Cu$K_{\alpha 1}$ radiation in both of the polycrystalline source powder and the crushed NHO single crystal which shown in fig.1.

The single crystal was sliced to thin plate (thickness ∼0.3 mm) for the thermal expansion measurement and the capacitance measurement. The thermal expansion coefficient measurement was performed by using a Wien-type micro-dilatometer mounted into a physical properties measurement system (PPMS, Quantum Design). The dielectric properties were measured versus temperature and frequency by capacitance spectroscopy measurements using an Agilent E4980A Precision LCR meter.

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[1] Y. Liu, Z. C. Li, W. P. Liu, G. Friemel, D. S. Inosov, R. E. Dinnebier, Z. J. Li, and C. T. Lin, *Supercond. Sci. Technol.* **25**, 075001 (2012).

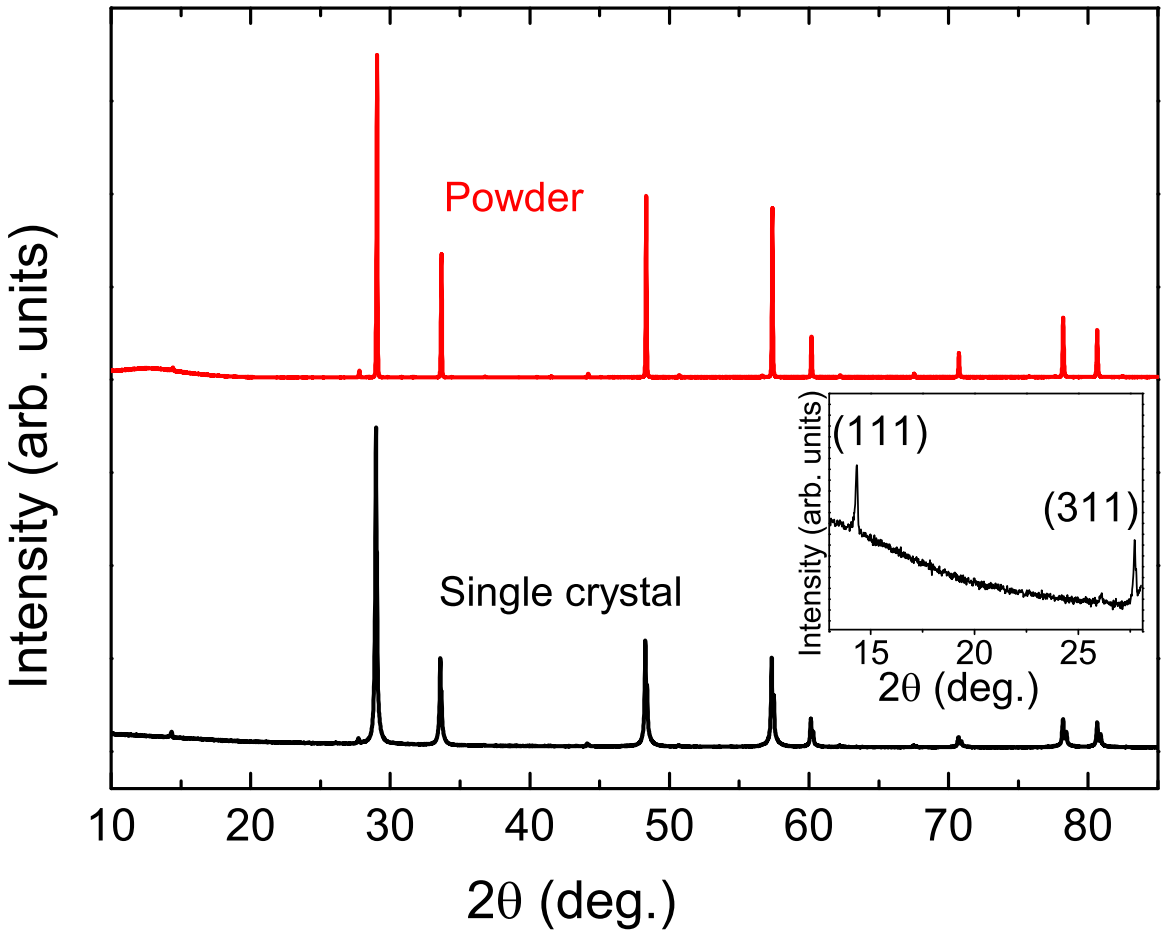

Figure S 1: (Color online) XRD patterns of the polycrystalline source powder (upper) and the powder obtained by crushing a small piece of the single crystal $Nd_2Hf_2O_7$ (lower). All the peak positions and their relative intensities are well matched the previous $Nd_2Hf_2O_7$ powder XRD result reported by Ubic *et al.*. Inset : The Enlarged low angle XRD pattern of the single crystal $Nd_2Hf_2O_7$. The weak Bragg peaks indexed as (111) and (311) are characteristic fingerprints of the pyrochlore structure.

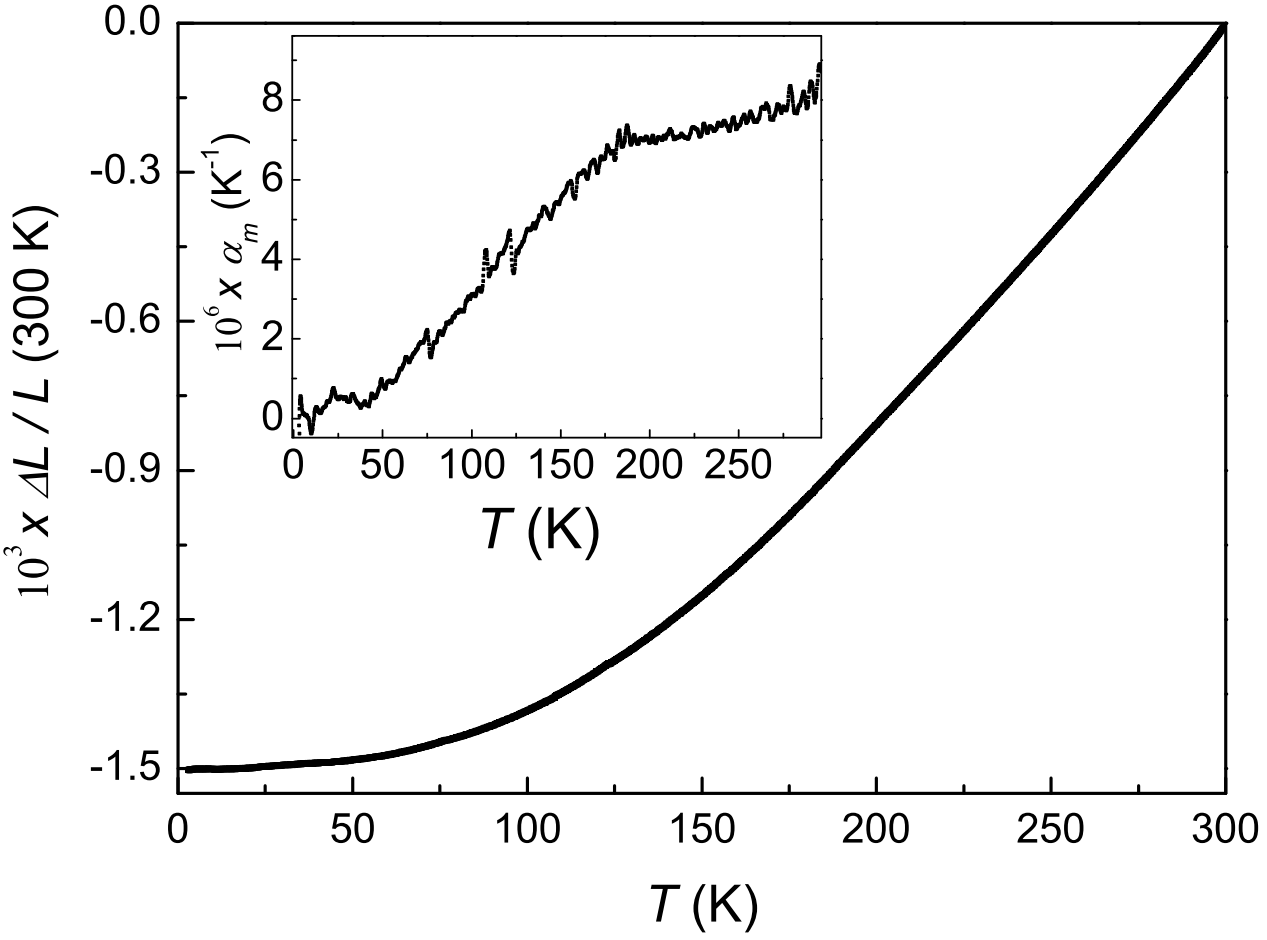

Figure S 2: Length change of the single crystal of NHO from 300 K to 2 K. Inset : The linear thermal expansion coefficient of the single crystal.