Growth of $La_4Ni_3O_{10-\delta}$ Single Crystals Using the High-Pressure Optical-Image Floating Zone Technique

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Abstract: **The quest to discover new high-temperature superconductors has ignited significant scientific interest. $La_4Ni_3O_{10-\delta}$ offers a unique opportunity to explore the fundamental mechanisms of superconductivity in nickelates. In this study, we report the successful growth of $La_4Ni_3O_{10-\delta}$ single crystals using the high-pressure optical-image floating zone technique. Our investigation into the physical properties of these crystals reveals their high quality, underscoring their potential for further research.**

Polycrystalline $La_4Ni_3O_{10-\delta}$ synthesis

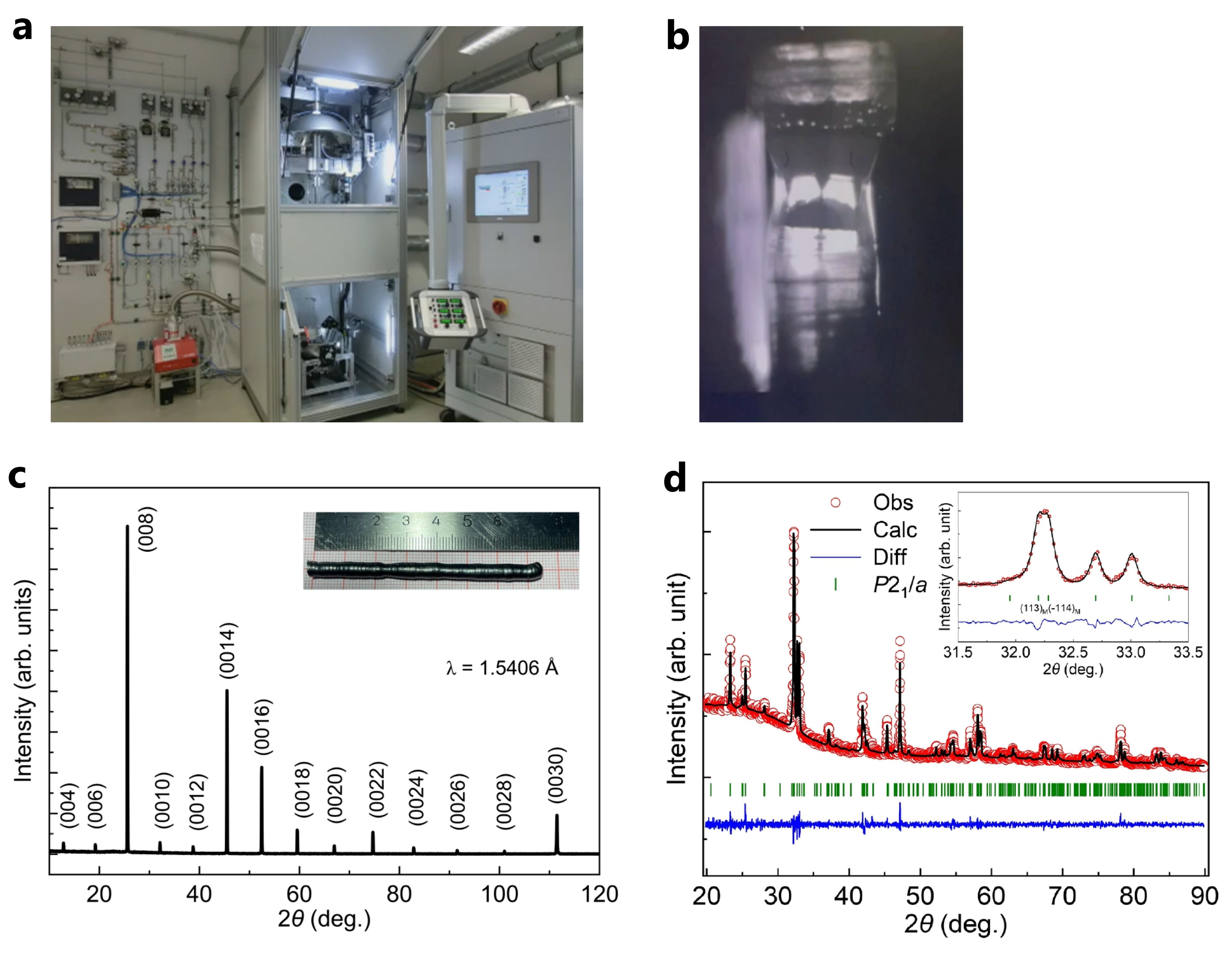
The precursor powder for the $La_4Ni_3O_{10-\delta}$ compound was prepared using the conventional solid-state reaction method. First, La_2O_3 (Aladdin, 99.99%) and NiO (Aladdin, 99.99%) were calcined at 1273K for 12h to remove the absorbed water. Then, they were mixed stoichiometrically and ground while an additional 0.5% of NiO was added to prevent potential NiO volatilization. The powder mixture was calcined in a box furnace at 1373K for 24h; this calcination process was repeated twice to ensure complete and homogeneous reaction.

In-situ lab-based XRD measurements on powder were carried out on a Bruker D8 Venture diffractometer to prove the high quality of $La_4Ni_3O_{10-\delta}$ polycrystalline.

The obtained powder was reground and stored in a rubber tube for isotropically pressed at 300 MPa to form a cylindrical feed rod.

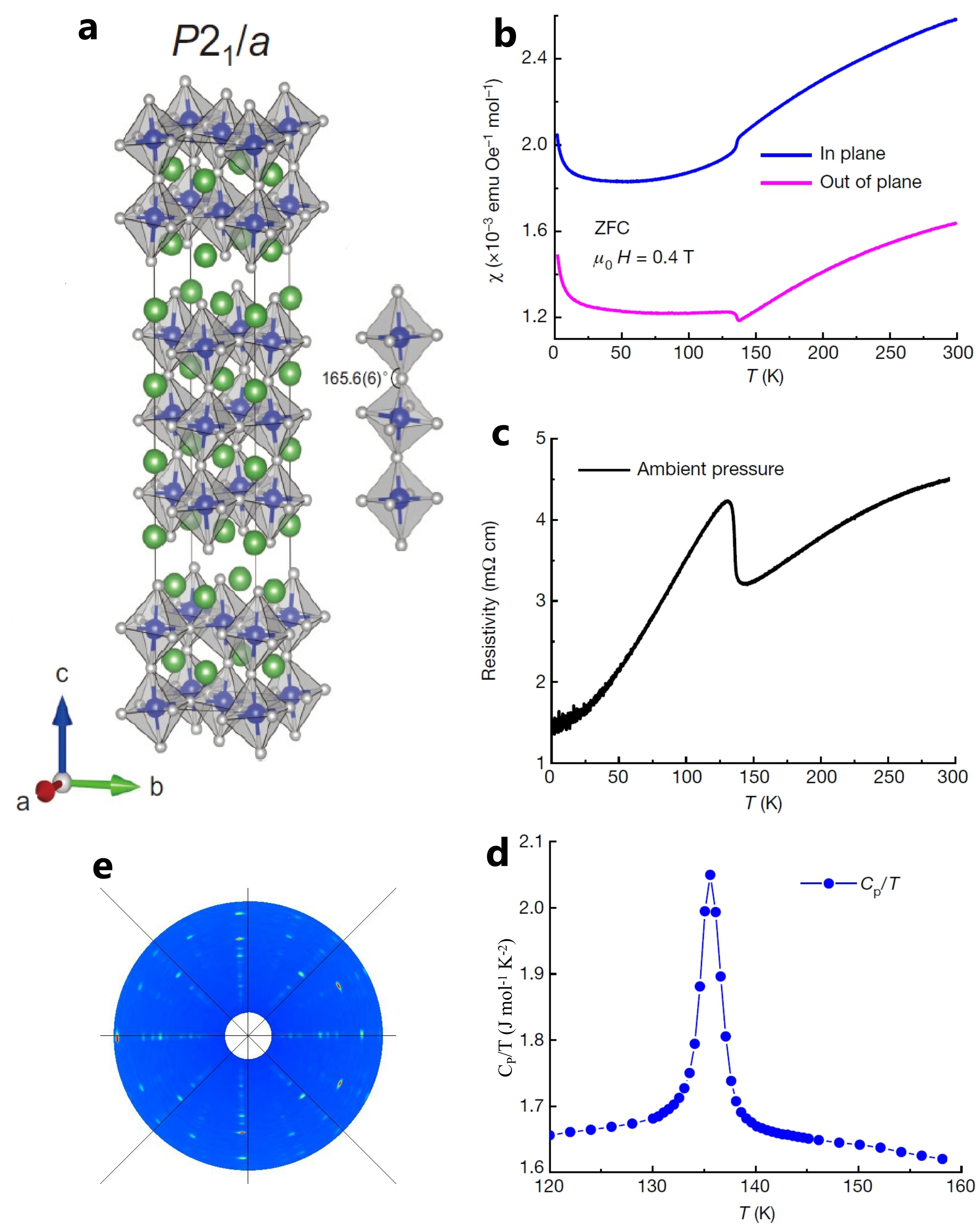
Growth of $La_4Ni_3O_{10-\delta}$ single crystals

The cylindrical rod, approximately 13cm in length and 6mm in diameter, was then underwent once sintering at 1673 K for 12h in air. Single crystals were grown using **a vertical optical-image floating-zone furnace (Model HKZ, SciDre)**. During the crystal growth process,we carefully maintained an oxygen pressure of 18–22 bar, and used a 5-kW Xenon arc lamp as the light source. The rod was rapidly traversed through the growth zone at a speed of 15mm h^{-1} to enhance the density, after which a growth rate of 3mm h^{-1} was maintained to get high quality single crystal.

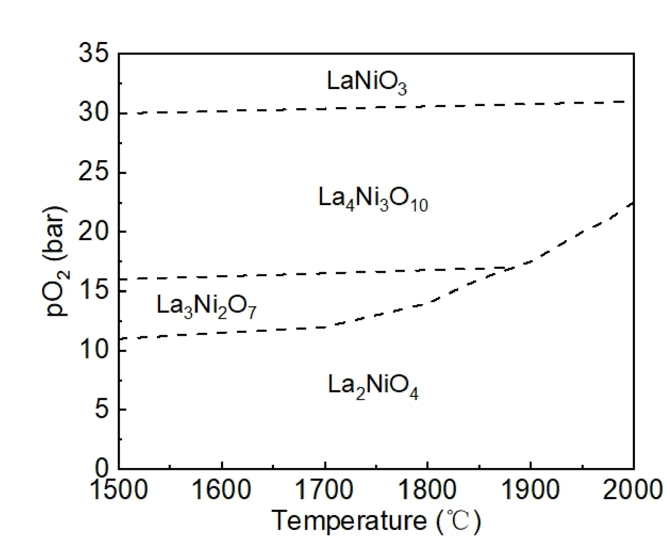


a, Vertical optical-image floating-zone furnace (Model HKZ, SciDre). **b**, Single crystal growing in a floating zone furnace. **c**, XRD measurements of a $La_4Ni_3O_{10-\delta}$ single crystal along the ab plane, revealing no detectable impurity phases. **d**, Rietveld refinement of a lab-based XRD pattern for powdered La4Ni3O10-δ single crystals at ambient pressure and room temperature. This dataset fits well with the $P2_1/a$ space group. The inset shows the details near 32.2° , where the $(1\ 1\ 3)_M$ and $(-1\ 1\ 4)_M$ peaks are captured by the $P2_1/a$ model.

Fundamental physical properties of high quality single crystals



a, Crystal structure of $La_4Ni_3O_{10-\delta}$ at ambient pressure $P2_1/a$ and the Ni–O–Ni angle between adjacent NiO_2 layers is 180° . **b**, Magnetic susceptibility of $La_4Ni_3O_{10-\delta}$ measured from 2K to 300K with an applied field of 0.4T, parallel and perpendicular to the ab plane. The SDW/CDW transition characterized by a kink in the χ(T) curve occurs at $\mathbf{T_N \approx 136K}$. **c**, Resistivity profile of $La_4Ni_3O_{10-\delta}$ in the ab plane at ambient pressure, using a current of 100μA. **d**, Specific heat of $La_4Ni_3O_{10-\delta}$ near T_N. **All of the fundamental physical properties indicate the nearly same $\mathbf{T_N}$.** **e,** X-ray Laue pattern of $La_4Ni_3O_{10-\delta}$ single crystal along ab plane.



Schematic drawing of Ruddlesden-Popper phase predominance as a function of pO_2 and temperature.

Reference

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