Electronic Supplementary Information (ESI):

Epitaxial antiperovskite superconducting $CuNNi_3$ thin films by chemical solution deposition

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Experimental Details

1. Thin film preparation

Epitaxial CNN thin films were grown on $LaAlO_3$ (LAO, 001) substrates by chemical solution deposition. Cupric nitrate and nickel nitrate were dissolved into glacial acetate acid and 2-methoxyethanol (in volume of 1:1) at 60 ºC, and the solution was stirred at this temperature for 20 minutes, and then stirred at room temperature for more than 12 hours. The solution concentration was preciously controlled to 0.4 M. Before the deposition process, all the substrates were ultrasonically cleaned with acetone, ethyl alcohol and deionized water, and then cleaned in a plasma-cleaner for 10 minutes. All the thin films were fabricated by the spin-coating method with a rotation speed of 5000 rpm and a time of 10 s, and then baked in air at 150 ºC and 350 ºC for 2 and 10 minutes, respectively. In order to prepare thicker film, the above processing was repeated for 8 times. Finally, the thin films were annealed at 600 ºC for 2 hours in air and nitrogenised at 600 ºC for 2 hours under flowing ammonia atmosphere with 1 atm pressure.

2. Characterization

The crystal phase and quality were analyzed by a high-resolution X-ray diffraction with a monochromatic Cu-K_a radiation on a Philips X'pert Pro machine (XRD, Nickel filter and Bragg-Brentano geometry, PANalytical B. V., Almelo, Netherlands). The composition, surface morphology and film thickness were determined by energy dispersive spectroscopy (EDS) and a field-emission scanning electronic microscopy (FE-SEM, FEI-designed Sirion 200, FEI, Hillsboro, OR). The crystallite size and the interface were checked by a transmission electron microscopy (TEM) inspection (JEM-2010, JEOL Ltd., Japan). Electrical transport properties were measured on a physical properties measurement system (PPMS, Quantum-designed) using the standard four-point probe technique. The magnetization measurements were performed with a Quantum Design superconducting quantum interface device (SQUID) magnetic property measurement system (MPMS) system.

3. Theoretically calculations

The origin of the superconductivity of $CuNNi_3$ was discussed theoretically based on the density functional theory. Figure 1a shows the electron density of states (DOS) of $CuNNi_3$. Around the Fermi energy (E_F), the 3*d* electrons of Ni contribute majorly to the DOS. Similar to the case of $MgCNi_3$,[1,2] there is a van Hove singularity very close to E_F, leading to a large DOS at E_F ($N(E_F)$) with the value of 6.6 states/eV. Figure 1b show the phonon dispersions and the electron-phonon coupling strengths for different modes of $CuNNi_3$ calculated based on the density-functional perturbation theory.[3] The three high frequency phonon branches are contributed by the lighter N atom, while the twelve low frequency branches are mainly from the heavier Cu and Ni atoms as shown in Figure 1c. A significant softening of the acoustic modes can be found at R point, generating the large electron-phonon coupling strengths at these modes. According to the BCS theory, a large $N(E_F)$ can lead to strong electron-phonon coupling. On the other hand, as the case in $MgCNi_3$, the van Hove singularity adjacent to E_F may cause the phonon modes softening, which can largely enhance the electron-

phonon coupling.[4,5] Both the two sides existing in $CuNNi_3$ make it have a sizeable electron-phonon coupling with the total coupling strength of λ= 0.58. The T_c is estimated using the Allen-Dynes formula[6] with a typical Coulomb pseudopotential μ^*= 0.12. The calculated T_c of 3.552 K is very close to the experimental value.

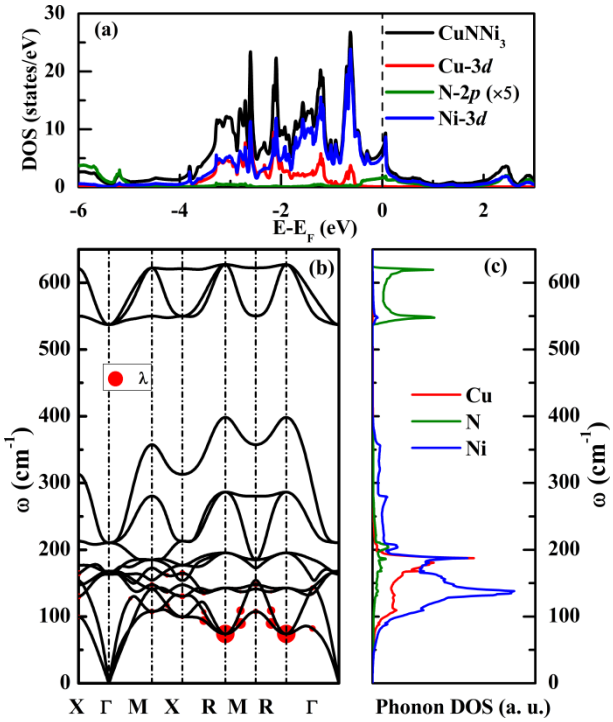

Fig. 1 (a) Electron DOS; (b) phonon dispersions; (c) phonon DOS of $CuNNi_3$. (b) is decorated with symbols, proportional to the partial electron-phonon coupling strength λ_q^v.

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