After being expected to be a promising analog to cuprates for decades, superconductivity has recently been discovered in infinite-layer nickelates, providing new opportunities to explore mechanisms of high-temperature superconductivity. However, in sharp contrast to the single-band and anisotropic superconductivity in cuprates, nickelates exhibit a multi-band electronic structure and an unexpected isotropic superconductivity as reported recently, which challenges the cuprate-like picture in nickelates. Here, it is shown that strong anisotropic magnetotransport behaviors exist in La-based nickelate films with enhanced crystallinity and superconductivity \((T_{\text{onset}} = 18.8\, \text{K}, T_{\text{zero}} = 16.5\, \text{K})\). The upper critical fields are anisotropic and violate the estimated Bardeen–Cooper–Schrieffer (BCS) Pauli limit \((H_{\text{Pauli}} = 1.86 \times T_{c, H=0})\) for in-plane magnetic fields. Moreover, the anisotropic superconductivity is further manifested by the cusp-like peak of the angle-dependent \(T_c\) and the vortex motion anisotropy under external magnetic fields.

1. Introduction

Great amounts of efforts have been made to seek for high-temperature superconductivity (HTSC) beyond cuprates to help understand its elusive pairing mechanisms.\(^1\) Infinite-layer nickelates have long been considered as a promising candidate owing to the same layered square-planar crystal structure and 3\(d^9\) electron counts as cuprates.\(^2\) Indeed, superconductivity has been realized in rare-earth nickelates recently,\(^3–7\) which provides a new platform for understanding the physics of HTSC. As is known, cuprates exhibit anisotropic superconductivity due to their layered structure and quasi-2D electronic structure,\(^8,9\) and recent experiments show that HTSC can even exist in monolayer cuprate membranes.\(^10\) As for nickelates, they host many electronic features that are distinctly different from those of cuprates even though sharing a similar layered structure.\(^11–12\) For example, in contrast to the single-Cu-3\(d_{x^2−y^2}\) band scenario in cuprates,\(^9\) infinite-layer nickelates possess multiband Fermi surfaces consisting of quasi-2D hole pockets derived from Ni-3\(d_{x^2−y^2}\) orbitals, and (one or two) 3D electron pockets mainly from rare-earth-5\(d\) orbitals depending on the doping level (Figure 1a).\(^11–14\) Some experimental observations, like the metallic ground states\(^3–5,15\) and the absence of long-range magnetic orders,\(^16–18\) are thought to be related to such multi-orbital features.\(^11–14,19–24\) Moreover, superconductivity in nickelates displays strikingly unexpected 3D isotropic behavior as reported by recent magnetotransport measurements\(^25,26\) which challenges the cuprate-like picture in nickelates.\(^11,27,28\) As such, whether nickelates are ideally analogous to cuprates is still a question.
under hot debates and the clarification of above puzzles will
greatly contribute to the understanding of superconductivity in
infinite-layer nickelates.

Here, we report the synthesis of La$_{0.8}$Sr$_{0.2}$NiO$_2$ thin films
on SrTiO$_3$ substrates with improved crystalline quality and en-
hanced superconductivity by reactive molecular beam epitaxy
(MBE). Transport measurements show the anisotropic upper crit-
cal field $H_{c2}$, a cusp-like peak of angle-dependent $T_c$ and vortex-
motion anisotropy under external magnetic fields. All these ex-
perimental observations suggest the anisotropic nature of super-
conductivity in lanthanum nickelates, similar to that in cuprates.

2. Results and Discussion

It is well known that the synthesis of superconducting infinite-
layer nickelates are extremely challenging, and the growth
and reduction parameters require careful optimizations.[3,15,29–31]
The lanthanum nickelate films are chosen for this work since
they have smaller lattice mismatch with SrTiO$_3$ substrates
(1.69%) compared with neodymium- and praseodymium-based
counterparts.[15,32,33] Using reactive MBE[29] we first synthe-
size undoped LaNiO$_3$ films, during which the La/Ni flux ra-
tio is carefully optimized to ensure the most “compact” lattice
structure, in other words, the smallest $c$ lattice constants. After
that, the Sr-doped samples are grown using the co-deposition
method,[34] during which the doping level can be precisely con-
trolled by adjusting the Sr shutter time. Reflection high-energy
electron diffraction (RHEED), x-ray diffraction (XRD) and trans-
port characterizations confirm the high crystalline quality of
the as-grown La$_{0.8}$Sr$_{0.2}$NiO$_3$ perovskite films and as-reduced
La$_{0.8}$Sr$_{0.2}$NiO$_2$ infinite-layer films (Figure S1, Supporting Infor-
mation). More importantly, the cross-sectional scanning trans-
mision electron microscopy (STEM) and corresponding Fourier
transforms show no signature of Ruddlesden–Popper (RP)-type
stacking faults (Figure 1b; Figure S2, Supporting Information),
a common defect observed in infinite-layer nickelates.[4,5,25,30]
Meanwhile, sharp interfaces without obvious cation intermix-
ing can be ascertained through the atomic-resolved energy-
dispersive x-ray spectroscopy (EDS) maps (Figure S2, Support-
ing Information). Temperature-dependent resistivity measure-
ments on these samples reveal a greatly enhanced supercon-
ductivity compared to other infinite-layer nickelates grown on
SrTiO$_3$ substrates.[15,32,33] A superconducting transition begins at
$\approx 18.8$ K and hits zero-resistance at $\approx 16.5$ K (Figure 1c,d), man-
ifesting a clean system to explore inherent properties in these
compounds. Interestingly, an enhancement of the normal-state
conductivity (also known as the excess conductivity) can be ob-
served below $\approx 25$ K, after subtracting the linear-extrapolated re-
sistivity data between 30 and 50 K (Figure 1d). This observa-
tion may manifest that the superconducting fluctuations in lan-
thanum nickelates is a relatively prominent effect, as that in
cuprates.[35] In addition, we also observe a resistivity drop in our
undoped LaNiO$_2$ samples at $\approx 13$ K (not shown), which is most likely to be the onset of the superconductivity observed in previous reports.$^{[15, 36]}$

Systematic magnetotransport measurements were performed under external magnetic fields perpendicular ($H \parallel c$) and parallel ($H \perp c$) to the NiO$_2$ planes under low fields up to 12 T and pulsed fields up to 56 T (Figure 2a, b; Figure S3, Supporting Information). Clearly, the nickelate films display highly anisotropic response to different magnetic field orientations, in sharp contrast to the isotropic behavior reported previously.$^{[25, 26]}$ Specifically, in our experiments, superconductivity is robust against the field strength up to 12 T for both field orientations, but shows significantly broader transition width for $H \parallel c$ than $H \perp c$ as field strength increases.

To quantitatively describe the observed anisotropic behavior, the upper critical field $H_{c2}(T)$ under both field orientations are extracted using the criterion of 50% $\rho_{xy}(T)$, where $\rho_{xy}(T)$ is determined through the linear fit to the normal state resistivity between 25 and 30 K (Figure 3). Evidently, the $H_{c2}(T)$ displays a linear dependence for the out-of-plane magnetic fields, while it follows a square-root relation ($T_c - T$)$^{1/2}$ for the in-plane ones (Figure 3c). The linear dependence of $H_{c2}(T)$ can be well described by the Ginzburg–Landau (GL) model$^{[37]}$ $H_{c2}^{\text{GL}}(T) = \Phi_0/[2\pi \xi_{ab}(0)](1 - T/T_c)$, where $\Phi_0$ denotes the magnetic flux quantum, $\xi_{ab}(0)$ is the zero-temperature in-plane GL coherence length. Using this relation, we obtain the $\xi_{ab}(0)$ of $27.5 \pm 0.2$ Å, which is comparable to that of cuprates.$^{[18]}$

As the ($T_c - T$)$^{1/2}$ dependence of $H_{c2}$ for $H \perp c$ can be originated from the paramagnetic de-pairing effects, GL model is inadequate to explain the experimental data since it only takes the orbital de-pairing effects into account.$^{[39]}$ Instead, the single-band Werthamer–Helfand–Hohenberg (WHH) theory,$^{[40]}$ which considers both the orbital and paramagnetic de-pairing effects, is adopted for the analysis of the $H_{c2}$ data. The overall fitting quality is quite well, yielding the Maki parameter $\alpha_M$ and spin-orbital scattering parameter $\lambda_M$ equal to (0.0) and (56.5, 0.8) for $H \parallel c$ and $H \perp c$, respectively. The essential difference between $\alpha_M$ for two field orientations corresponds well with the anisotropic superconductivity as observed in cuprates,$^{[41]}$ that is, orbital-limited for $H \parallel c$ and Pauli-paramagnetic-limited for $H \perp c$. In addition, the relatively large $\lambda_M$ signals the spin-orbit effects cannot be ignored here, which may also explain the surpass of the Bardeen–Cooper–Schrieffer (BCS) Pauli limit ($H_{c2, \text{Pauli}} = 1.86 \times T_c$) for the in-plane magnetic fields. In order to confirm the violation of BCS Pauli limit, we also performed high field measurements at 2 K in pulsed field up to 56 T at Wuhan National High Magnetic Field Center (Figure S3, Supporting Information). It can be seen that $H_{c2}$ determined from the high-field data at 2 K is indeed above the BCS Pauli limit for $H \perp c$, indicating the reliability of our fitting analysis based on the low-field data. Note that other effects such as spin-orbit coupling and disorders may influence the Pauli-limit in nickelate system, further work is necessary to examine these factors. We also note that $H_{c2}$ for $H \parallel c$ deviates from the WHH fitting at 2 K, which resembles the anomalous low-temperature upturn of $H_{c2}$ reported in Nd-based nickelates,$^{[25]}$ and is subsequently confirmed by our systematic high-field measurements.$^{[42]}$ However, the origin of such anomalous upturn is not fully understood yet.$^{[25, 42–44]}$ and needs further explorations. And after we finished the manuscript, we noticed that this Pauli-limit violation has been also confirmed by other high-field transport measurements performed in La-based nickelates.$^{[43, 44]}$ The large $H_{c2}/T_{c, \text{H} = 0}$ ratios approaching $T = 0$ K can be possibly explained as the presence of spin-triplet pairing$^{[43]}$ or strong phase fluctuations$^{[44]}$ in La-based nickelates. In addition, the Pauli-limit violation is also found to be more prominent in the in-plane magnetic fields with $H_{c2}(0)/\rho_0$ equals to $\approx 2$, which qualitatively agrees with what we observed here (Table S1, Supporting Information). Considering the similar $H_{c2}(T)$ behavior regardless of $T_{c, \text{H} = 0}$ in different reports, the underlying mechanism for such greatly enhanced $H_{c2}$ at low temperatures in La-based nickelates may be the same as well.

Also, the anisotropic ratio $\Gamma$, which is defined as $H_{c2}^{\text{GL}}(T)/H_{c2}^{\text{HH}}(T)$, is calculated based on above fitting results and is summarized in Figure 3d. The derived $\Gamma$ values are comparable to those of iron-based superconductors.$^{[45, 46]}$ but
Figure 3. Temperature-dependent upper critical field for magnetic fields a) along c axis and b) in the a-b plane. Here, $H_{c2}$ at low fields is defined as the field strength at which resistivity reaches 50% $\rho_n(T)$. $H_{c2}$ at high fields is determined as shown in Figure S3 (Supporting Information). The black arrows denote the BCS Pauli limit of $H_p = 1.86 \times T_c$, $H = 0$ (where $T_c, H = 0$ is the superconducting transition temperature at $H = 0$ T). The shaded blue and orange area indicate the orbital-limited and Pauli-limited superconductivity, respectively. The grey dashed lines are the corresponding linear fits. d) The temperature-dependent anisotropic ratios $\Gamma(T) = H_{c2}^{H||c}(T)/H_{c2}^{H\perp c}(T)$ for LSNO films with different thicknesses, which are derived from the corresponding fitted $H_{c2}$ data based on the WHH theory. The inset shows the enlarged view near $T_c$, where calculated $H_{c2}$ data are superimposed. The error bars represent the uncertainty of temperature measurements and the linear extrapolation process of $\rho_n(T)$.

Figure 3. Temperature-dependent upper critical field for magnetic fields a) along c axis and b) in the a-b plane. Here, $H_{c2}$ at low fields is defined as the field strength at which resistivity reaches 50% $\rho_n(T)$. $H_{c2}$ at high fields is determined as shown in Figure S3 (Supporting Information). The black arrows denote the BCS Pauli limit of $H_p = 1.86 \times T_c$, $H = 0$ (where $T_c, H = 0$ is the superconducting transition temperature at $H = 0$ T). The shaded blue and orange area indicate the orbital-limited and Pauli-limited superconductivity, respectively. The grey dashed lines are the corresponding linear fits. d) The temperature-dependent anisotropic ratios $\Gamma(T) = H_{c2}^{H||c}(T)/H_{c2}^{H\perp c}(T)$ for LSNO films with different thicknesses, which are derived from the corresponding fitted $H_{c2}$ data based on the WHH theory. The inset shows the enlarged view near $T_c$, where calculated $H_{c2}$ data are superimposed. The error bars represent the uncertainty of temperature measurements and the linear extrapolation process of $\rho_n(T)$.

smaller than those of cuprates. Similar magnetotransport measurements were also performed on thinner films (12 u.c.) for comparison, revealing nearly identical anisotropic behavior as the thicker ones (Figure S4, Supporting Information). Combining the fact that the isotropic $H_{c2}$ found in nickelate films of nearly the same thickness in a previous study and thickness-independent anisotropic ratio (Figure 3d), the anisotropic behavior observed here cannot originate from the 2D geometric confinement scenario caused by thin film geometry. Therefore, the overall anisotropic behavior should be qualitatively reliable, as evidenced by the good reproducibility of the fitting results obtained on multiple samples (Tables S1 and S2, Supporting Information).

Then we performed angle-dependent measurements of $T_c$ under fixed magnetic fields (Figure 2c), as widely used to confirm the dimensionality of superconductivity in 2D materials as well as in cuprates. As expected, the $T_c(\theta)$ exhibits a strong dependence on the magnetic field orientations, manifesting a strikingly sharp cusp-like feature near $\theta = 0^\circ$ ($H_{c2}$). The $T_c(\theta)$ extracted using 1% $\rho_n(T)$ and 90% $\rho_n(T)$ criterion reveal similar behaviors (Figure S5, Supporting Information), demonstrating that this cusp-like feature is criterion-independent and is
intrinsictoLa-basednickelates. It is known that the angular dependence of \( T_c \) can be described by the Tinkham model for 2D superconductors\(^{18,50} \)

\[
T_c(H, \theta) = T_{c0} - \left( T_{c0} - T_{c0}^{H||c}(H) \right) \sin \theta - \left( T_{c0} - T_{c0}^{H\perp c}(H) \right) \cos^2 \theta
\]

(1)

or the anisotropic GL model for 3D superconductors\(^{50} \)

\[
T_c(H, \theta) = T_{c0} + \frac{H}{dT_c^{H||c}(H)/dT} \left( \sin^2 \theta + \gamma^{-2} \cos^2 \theta \right)^{1/2}
\]

(2)

where \( T_{c0} \) is the transition temperature at zero field, \( T_c^{H||c} \) and \( T_c^{H\perp c} \) are the transition temperature for the out-of-plane and in-plane magnetic fields, respectively, while \( \gamma \) represents the effective mass ratio of the in-plane and out-of-plane electron motion. For our data, the cusp-like feature near \( \theta = 0^\circ \) (\( H \perp c \)) can only be well reproduced by the 2D Tinkham model rather than the 3D GL model.\(^{38} \) This result may indicate the possible quasi-2D nature of the superconductivity in lanthanum nickelates, in line with the anisotropic \( H_c^0 \) discussed above. Further detailed angle-dependent \( T_c \) with more data points within the range of 5 degrees from \( \theta = 0^\circ \) also confirms our conclusion (Figure S6, Supporting Information).

Since the superconducting transition exhibits clear broadening effects for \( H \parallel c \), the thermally-activated vortex motion could exist as well in nickelates, as widely studied in cuprates\(^{51,52} \) and iron-based superconductors.\(^{45,46} \) According to the thermally-activated flux flow (TAFF) model,\(^{52} \) the Arrhenius relationship

\[
\rho(T, H) = \rho_0 \exp \left( - \frac{U_0}{k_B T} \right)
\]

should be expected, where \( \rho_0 \) is the resistivity prefactor, \( U_0 \) is the activation energy and \( k_B \) is Boltzmann’s constant. Indeed, the resistivity data follow the Arrhenius dependence over several orders of magnitude above the measurement noise floor (Figure 4). And the activation energy \( U_0 \), which is determined from the slope of the linear parts in corresponding Arrhenius plots, exhibits strong anisotropy for two field orientations (Figure 4c). For \( H \parallel c \), the field-dependent activation energy decreases moderately as \( \theta \) increases from 0° to 90°, indicating the coexistence of single-vortex pinning and collective creep at low fields.\(^{46} \) On the other hand, the activation energy \( U_0 \) shows a rather weak power-law dependence as \( H^{0.11} \) for \( H \perp c \). Here, the larger power-law exponent \( n \) for \( H \parallel c \) is consistent with the broader superconducting transition width, representing stronger response of vortex motion with increased magnetic field compared to \( H \perp c \).\(^{52} \) The fundamental difference of the power-law exponent \( n \) here manifests the strong anisotropy of the pinning strength in the \( \text{La}_0.8\text{Sr}_{0.2}\text{NiO}_2 \) films, as reported in iron-based superconductors\(^{45,51} \) and cuprates.\(^{52} \) Besides, the crossover behavior of the power-law exponent \( n \) at \( \approx 90^\circ \) may be related to the competition between collective creep (at low field) and plastic creep (at high field), and their interaction with different types of pinning defects.\(^{53} \) However, further characterizations such as field-dependent critical current and temperature-dependent magnetization curves are needed to help understand the complex vortex dynamics in nickelates. In addition, the difference in \( U_0 \) values for two field orientations reaches about an order of magnitude at 12 T, implying the strong anisotropy of vortex motion along in-plane and out-of-plane directions. Further
detailed magneto-resistance measurements in fine steps below 1 T also confirm our conclusion about anisotropic vortex motion (Figure S7, Supporting Information).

Similar magnetotransport experiments have been performed on many infinite-layer nickelate films, which reproduce our observations, indicating the superconductivity in lanthanum nickelates is anisotropic in nature. Therefore, the previous observations of isotropic superconductivity in nickelates may be related to the vertical running faults,[25,26] as well as the formation of a-axis oriented domains,[15] which naturally reduce the anisotropy of the c-axis oriented infinite-layer structure. Note that one of these previous observations of isotropic behaviors was performed on the Nd$_{0.8}$Sr$_{0.2}$NiO$_3$ films grown using our MBE system as well.[32] Compared to lanthanum nickelates, the neodymium counterparts have the larger lattice mismatch with SrTiO$_3$ substrates, which could lower the crystalline quality, as evidenced by the unavoidable extra impurity diffractions in the RHEED patterns.[29] Also, the rare-earth 4f moments may play a very important role in the superconducting anisotropy in nickelates, as proposed recently.[44] Other than the enhanced crystallinity, the improved T$_c$ together with 2D character of superconductivity observed here may be related to the increased c lattice constants of La-based nickelates compared to Nd-based ones.[13,34,55] However, further strain-dependent measurements on nickelate films are needed to confirm the impact of c lattice constants on T$_c$ and dimensionality of superconductivity, which is beyond the scope of the present work.

3. Conclusion

In summary, we have synthesized infinite-layer La-based nickelate thin films without detectable RP-type stacking faults using reactive MBE and obtained the highest reported superconducting transition temperature on SrTiO$_3$ substrates at ambient condition up to date. The upper critical field $H_{c2}$ is found to be strongly anisotropic, with the observation of Puali-limit violation. The angle-dependent T$_c$ and vortex motion anisotropy under external magnetic fields further confirm the anisotropic nature of the superconductivity in lanthanum nickelates. Our findings thus suggest that the quasi-2D hole bands derived from Ni-3d$_{x^2-y^2}$ orbitals may play a more important role than the 3D electron bands in pairing formations in nickelates. As such, the cuprate-like picture may be still valid in describing the superconductivity in infinite-layer nickelates.

4. Experimental Section

**Film Growth and Sample Preparation:** The pristine perovskite La$_{0.8}$Sr$_{0.2}$NiO$_3$ films were grown on the TiO$_2$-terminated SrTiO$_3$ (001) substrates by reactive MBE using a DCA R450 MBE system, with the substrate temperature of 600 °C and an oxidant background pressure of 1 × 10$^{-3}$ Torr using the distilled ozone. During the film growth, RHEED was employed in situ to monitor both the growth process and surface quality (Figure S1a, Supporting Information). The growth parameters, such as cation stoichiometry, were optimized as reported previously.[29] To obtain the infinite-layer phase, the La$_{0.8}$Sr$_{0.2}$NiO$_3$ samples together with CaH$_2$ powder (0.1 g) were sealed in a vacuum chamber with background pressure lower than 1 × 10$^{-3}$ Torr, and then heated to 310 °C for 4 h, with warming (cooling) rate of 10–15 °C min$^{-1}$. The obtained superconducting films show single phase and are fully strained to the substrates, as confirmed by XRD (Figure S1b, Supporting Information). And the positions of the main film peaks and Laue fringes in 2θ-ω scan can be well captured by the corresponding Laue function fitting,[24] which also reflects the good crystallinity of the infinite-layer structure (Figure S1c, Supporting Information). The slight discrepancy between the fitted (5.5 ± 0.4 nm) and nominal (6.8 nm) film thickness may reside in the following reasons. First, the formation of La$_3$(Ti,Ni)O$_7$ intermediate layer at the top and bottom La$_0.8$Sr$_0.2$NiO$_3$/SrTiO$_3$ interfaces.[57] Second, neglecting the coherent interference effects between diffracted waves for film and substrate during the fitting process due to the lack of corresponding crystal polarizability factor of La$_0.8$Sr$_0.2$NiO$_3$ structure, which could underestimate the film thickness.[34]

**Structural Characterization:** Specimens for the cross-sectional STEM were fabricated using the focused ion beam (FIB) techniques on a Thermo Scientific Helios C4 X FIB system. The atomic-resolution STEM-HAADF and EDS images were obtained on a double spherical aberration-corrected FEI Titan G2 60–300 system at 300 kV with a field emission gun. To further analyze the lattice constants of the infinite-layer structure, we performed the fast Fourier transform (FFT) (Figure S2b, Supporting Information) and the intensity line profile (Figure S2c, Supporting Information) of the HAADF-STEM image. In the FFT image, the diffraction spots from the substrate and film align well along a axis while show different periodicities along c axis, manifesting the in-plane coherency and difference on the c lattice constants. The average c lattice constant of the La$_0.8$Sr$_0.2$NiO$_3$ film is 3.46 Å as extracted from adjacent 11 peaks, which is consistent with previous reports.[31] XRD was performed using a Bruker D8 Discover diffractometer, with the Cu-K$_α$ radiation of 1.5418 Å wavelength.

**Transport Measurements:** The low magnetic field measurements were performed using the standard six-probe configuration at the Cryogen Free Measurement System (CFMS, Cryogenic) and the physical property measurement system (PPMS, Quantum Design). The high magnetic field measurements were performed at 2 K using the standard four-probe configuration in pulsed field up to 56 T at Wuhan National High Magnetic Field Center (Figure S3, Supporting Information). The ohmic contacts were made through the ultrasonic aluminum-wire bonding. The magnetotransport measurements were performed by aligning the film c axis parallel or perpendicular to the applied magnetic field via a rotator, while the “parallel” is defined as the position with maximum magnetoresistance under applied magnetic fields (H || c).

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request.
