

Thickness-driven structural evolution and vacancy-linked superconductivity in epitaxial NbN thin films

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Epitaxial δ -NbN thin films (9–205 nm) were grown on MgO(001) and MgO(110) to examine how thickness-driven structural evolution controls normal-state transport and superconducting performance, and how these changes are linked to vacancy-mediated lattice evolution. Structural characterization shows that increasing thickness generally improves the microstructural coherence length. The room-temperature resistivity generally decreases with thickness when a single epitaxial registry is preserved. However, the 205-nm NbN(110) film, which exhibits a weak secondary (111) component, shows a higher resistivity despite its larger microstructural coherence lengths, indicating that resistivity is not governed by structural coherence alone. All measured films exhibit a negative temperature coefficient of resistivity, and the broad minimum in $d\rho_{xx}/dT$ appears for all samples around 50–80 K, which correlates more closely with the disorder parameter $k_F\uparrow$ than with microstructural coherence lengths. Hall measurements indicate that the effective carrier density n_{eff} increases with thickness and correlates with the evolution of unit-cell volume. Together with density functional theory calculations showing that percent-level vacancy disorder can produce comparable changes in lattice volume, this supports a thickness-driven vacancy-linked lattice evolution accompanied by strain relaxation, coinciding with an increase of T_c (8.5–12.3 K). The largest estimated upper critical field is 44.9 T for the 205-nm NbN(110) film, corresponding to a superconducting coherence length of 2.7 nm.

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I. INTRODUCTION

Niobium nitride (NbN) remains a workhorse superconductor for thin-film technologies because it combines a relatively high transition temperature with a short coherence length and large upper critical fields, enabling operation in nanoscale geometries and under strong magnetic fields. These attributes underpin applications ranging from superconducting nanowire single-photon detectors to hot-electron bolometer mixers, where film uniformity and reproducible transport are as critical as T_c itself [1–3].

Beyond its device utility, NbN has long served as a model system for exploring how disorder reshapes metallic transport in a conventional s -wave superconductor. As early as the 1970s, NbN thin films were reported to remain superconducting while exhibiting a negative temperature coefficient of resistance, $d\rho/dT < 0$, in the normal state [4].

Importantly, magnetotransport work showed that NbN samples with negative $d\rho/dT$ can display magnetoresistance signatures consistent with three-dimensional weak localization, directly linking the anomalous T dependence of $\rho(T)$ to quantum-interference physics in a disordered conductor [5]. In this broader context, NbN provides a concrete materials platform where the “bad-metal/disorder-crossover” phenomenology (often discussed in relation to Mooij-type correlations in disordered transition-metal systems) becomes experimentally tractable while superconductivity remains robust over much of the parameter space [6].

Epitaxial NbN provides a controlled platform to disentangle the coupled roles of disorder, strain, and stoichiometry. In particular, reactive magnetron sputtering on MgO(001) enables systematic tuning from relatively low-disorder metallic films to disordered diffusive regimes, as indicated by $k_F l$, while measuring transport and Hall response [7]. Hall measurements further indicate that the effective carrier density can vary substantially with growth conditions and has been reported to correlate with T_c ; correspondingly, $\rho(T)$ and $R_H(T)$ can show nontrivial temperature dependences across a wide disorder range [8,9].

At the same time, δ -NbN is intrinsically nonideal and can accommodate vacancy populations that modify lattice metrics and electronic structure; experiments and first-principles studies have linked defect content to lattice-parameter or

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volume changes and to transport trends [10,11]. However, microstructural metrics (texture, coherence lengths, boundary character) and electronic parameters (n_{eff} , $k_F l$, T_c , B_{c2}) are still often discussed separately, and transport-focused datasets for epitaxial NbN on MgO(110) are comparatively scarce relative to MgO(001). MgO(001) provides a high-symmetry reference for NbN epitaxy, whereas MgO(110) enables the influence of lower in-plane symmetry on strain relaxation and defect evolution to be examined. The thickness range of 9–205 nm was selected to follow the progression from interface-dominated growth to a nearly relaxed thick-film state. Here we perform a thickness-controlled comparison of epitaxial NbN grown on MgO(001) and MgO(110), combining microstructural-coherence analysis, longitudinal and Hall transport measurements, upper-critical-field estimates, and vacancy-containing density functional theory (DFT) calculations. This approach allows us to examine how thickness-driven structural evolution, such as vacancy-linked lattice evolution, and microstructural coherence length, coevolves with transport and superconducting properties.

II. METHODOLOGY

NbN thin films were grown in a Mantis UHV dc magnetron sputtering system (base pressure $< 5 \times 10^{-8}$ Pa). Single-crystal MgO(001) and MgO(110) substrates ($10 \times 10 \times 0.5$ mm³; offcuts $< 0.3^\circ$ and $< 0.5^\circ$, respectively) were cleaned as in Ref. [8] and annealed *in situ* under vacuum at 800 °C for 30 min before deposition. Films were reactively sputtered from a 3-in. Nb target (99.7%) in an Ar/N₂ discharge using gas flows of 80 sccm Ar and 12 sccm N₂, at 800 °C, with a working pressure of 4.5 mTorr and substrate rotation of 10 rpm. The target was presputtered for 20 min with shutters closed. The deposition rate was approximately 0.3 Å/s, and the film thickness was controlled by varying the deposition time. After growth, samples were cooled at a rate of 5 °C/min. For each nominal thickness, MgO(001) and MgO(110) substrates were mounted side-by-side and coated in the same deposition run.

θ -2 θ X-ray diffractometry (XRD) in Bragg-Brentano geometry was used to record the overall structure of the films using a PANalytical X'Pert PRO diffractometer (Cu $K\alpha$, 45 kV/40 mA) operated with a Ni filter to suppress Cu $K\beta$. High-resolution XRD (to confirm epitaxy and compare crystalline quality) was then performed on a PANalytical Empyrean (Cu $K\alpha_1$, 45kV/40mA). φ scans used an x-ray lens (incident) and a 0.27° parallel-plate collimator (diffracted); ψ was set to 25.2° to access the 113 reflection for MgO(001) and was set to 31.5° to access 311 reflection for MgO(110). Reciprocal space maps (RSMs) were acquired with a two-bounce Ge(220) hybrid monochromator, an open detector, and a 255-channel area detector in linescan mode; asymmetric grazing-exit geometry was used for nonpreferred directions. X-ray reflectivity (XRR) was measured on the same instrument (mirror optics and 0.27° collimator) and the film thickness (for $t < 100$ nm) was obtained by data fitting using the software genx (Ref. [12]).

Film thickness with $t > 100$ nm was measured by cross-sectional scanning electron microscopy (SEM; Zeiss Sigma 300 VP Gemini, operated with an acceleration voltage of

5 kV). Elemental composition was determined by Time-of-flight Elastic-recoil detection analysis (ToF-ERDA) complemented by Rutherford backscattering, performed at the Tandem Laboratory (Uppsala University). ToF-ERDA used a 36-MeV 127 I⁸⁺ beam with a 45° recoil angle and a 67.5° incidence angle relative to the surface normal. Compositional depth profiles were extracted from time-energy coincidence spectra using potku 2.0 [13].

Selected films were examined by cross-sectional transmission electron microscopy (TEM; FEI Tecnai G2 TF 20 UT, 200 kV). Samples suitable for TEM analysis were prepared using focus ion beam milling and a lift out approach using a ThermoFisher Helios 5 UC instrument. Each sample was coated with a tungsten protective layer while milling out a 2–3- μ m-wide windows to a final thickness of 60–70 nm. High-resolution high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) was performed with an aberration-corrected FEI Titan³ operated at an acceleration voltage of 300 kV. Images were processed by inverse fast Fourier transform (FFT) using the Gatan DigitalMicrograph software.

The single-crystal nature of the deposited film was confirmed on the macroscale by electron backscatter diffraction (EBSD; Zeiss 560 Gemini scanning electron microscope equipped with an Oxford Instruments Symmetry diffractometer) The specimen was analyzed as grown with no additional preparation or treatment.

Electrical resistivity was obtained from the film sheet resistance measured with a four-point probe (Jandel RM3000) using $\rho = R_s t f$, where R_s is the sheet resistance, t is the film thickness determined by SEM or XRR, and f is the geometry-dependent correction factor. Low-temperature transport measurements were performed in a Physical Property Measurement System (PPMS Quantum Design Model 6000) using dc current excitation. PPMS resistance data were converted to resistivity using the independently measured thickness and were cross-calibrated to the room-temperature resistivity obtained from the four-point probe measurements. Electrical contacts were prepared by ultrasonic wire bonding (see Ref. [14] for additional details of the low-temperature measurement setup). Hall-effect measurements were performed by sweeping the magnetic field from -9 to $+9$ T in the normal state, above the onset of the superconducting transition. The Hall coefficient, R_H , was obtained from the slope of the transverse resistivity as a function of magnetic field. The effective carrier density was calculated within a single-band approximation as

$$n_{\text{eff}} = \frac{1}{e|R_H|},$$

where e is the elementary charge. No correction for electron-electron interaction effects was applied.

DFT calculations were performed with quantum espresso [15] using the generalized gradient approximation [16] and projector augmented-wave pseudopotentials [17]. Plane-wave cutoffs were 80 Ry for wave functions and 800 Ry for charge density; convergence with respect to the wave-function cutoff was checked at 60–90 Ry with no meaningful change beyond 80 Ry. Rocksalt NbN ($Fm\bar{3}m$) was modeled using vacancy-containing supercells of 64 sites (atoms + vacancies).

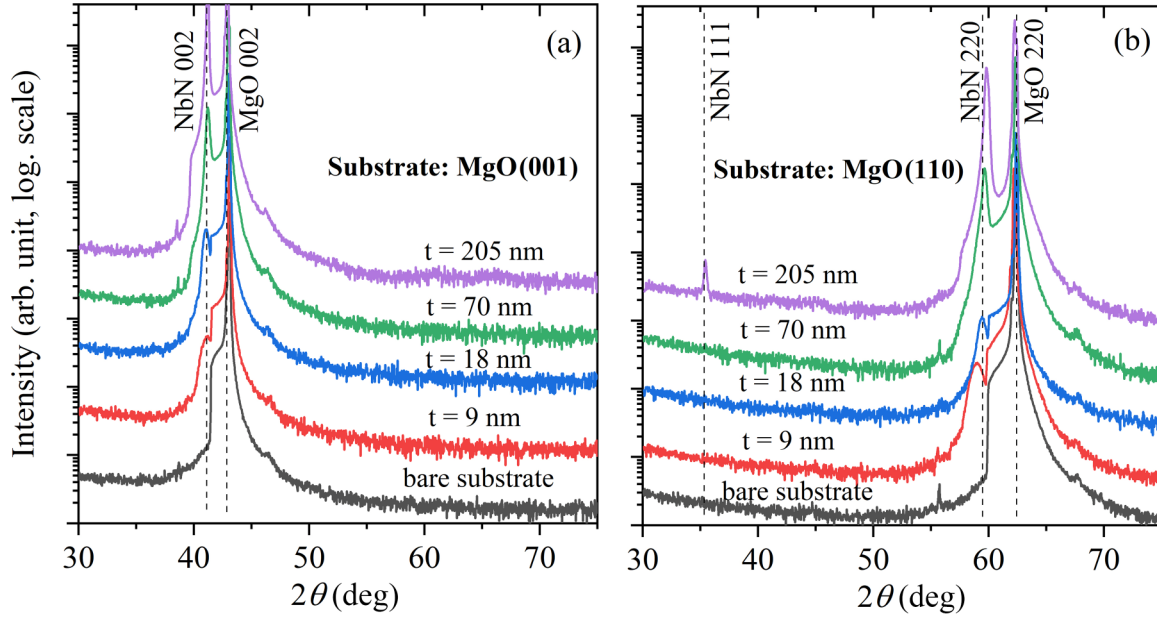


FIG. 1. X-ray diffraction (θ - 2θ) patterns of NbN thin films with thickness $t = 9, 18, 70$, and 205 nm grown on (a) MgO(001) and (b) MgO(110), together with the corresponding bare-substrate scans. The NbN film reflections and MgO substrate peaks are labeled; dashed lines mark the expected peak positions.

Both ordered and disordered vacancy configurations were considered; disordered structures were generated as SQSs (special quasirandom structures) using the mcsqs code [18]. Chemical potentials were referenced to bcc Nb ($Im\bar{3}m$) and the N_2 molecule.

Brillouin-zone sampling used Monkhorst-Pack meshes with reciprocal-space spacing below $0.04 \text{ \AA}^{-1}$. Electronic occupations were treated by cold smearing [19] with width 0.1 eV. Lattice parameters and atomic coordinates were relaxed until stresses were below 10^7 Pa ($\approx 0.06 \text{ meV/\AA}^{-1}$) and forces were below $4.1 \times 10^{-11} \text{ N}$ ($\approx 26 \text{ meV/\AA}$). The formation energy was computed as

$$E_f = \frac{E_{\text{tot}} - \sum_s n_s \mu_s}{\sum_s n_s},$$

where E_{tot} is the supercell total energy, n_s is the number of atoms of species s , and μ_s is the corresponding chemical potential ($s = \text{Nb}, \text{N}$).

III. RESULTS AND DISCUSSION

A. Epitaxial microstructure and thickness evolution

Figure 1 establishes the dominant out-of-plane texture for all films. For NbN on MgO(001) [Fig. 1(a)], only the 002 reflection is observed over the full thickness range (9–205 nm), indicating predominantly (001)-oriented growth. For NbN on MgO(110) [Fig. 1(b)], the diffraction is dominated by the 220 reflection, consistent with (110) texture; a weak secondary (111) contribution is detected for the thickest film. The epitaxial growth is confirmed by in-plane diffraction and reciprocal-space mapping (Fig. 2), and by electron microscopy. In particular, azimuthal φ scans of the off-axis 113 reflections for NbN/MgO(001) and the corresponding 311 reflection for NbN/MgO(110) show fixed

in-plane alignment between film and substrate [Fig. 2(a)], while the RSM confirms well-defined film and substrate peaks [Fig. 2(b)]. Cross-sectional TEM and selected-area diffraction further support coherent epitaxial growth [Fig. 2(c)]. The mottled contrast may arise from local strain fields associated with misfit and threading dislocations, together with possible variations in lamella thickness and bending during specimen preparation. TEM-energy-dispersive x-ray spectroscopy (EDS) showed no detectable compositional variation across the film. EBSD mapping (Supplemental Material Fig. S1 [20]) was used as a complementary large-area orientation check, confirming lateral uniformity of the film orientation over at least a millimeter beyond the local and reciprocal-space information obtained from XRD/RSM and TEM.

Thickness-dependent lattice parameters extracted from x-ray diffractometry are summarized in Table I. The crystallographic convention used to assign the lattice parameters for the two growth orientations is illustrated schematically in Fig. S2. The lattice mismatch between relaxed NbN ($4.37 \text{ \AA}$) and MgO ($4.212 \text{ \AA}$) is as follows:

$$f = \frac{a_{\text{NbN}} - a_{\text{MgO}}}{a_{\text{MgO}}} \times 100 = 3.75\%.$$

For NbN/MgO(001), the in-plane lattice constant increases from $a = b = 4.25 \text{ \AA}$ (9 nm) to $4.37 \text{ \AA}$ (205 nm), while c decreases from $4.399 \text{ \AA}$ to $4.375 \text{ \AA}$; the unit-cell volume correspondingly increases from 79.45 to $83.55 \text{ \AA}^3$. For NbN/MgO(110), a and b decrease from 4.41 and $4.43 \text{ \AA}$, respectively, for 9-nm film to $4.37 \text{ \AA}$ for 205-nm film, while c increases from 4.17 to $4.37 \text{ \AA}$, with the volume rising from 81.5 to $83.3 \text{ \AA}^3$. The measured lattice evolution cannot be explained by elastic strain alone. For the thinnest NbN(001) film, the contraction of the in-plane lattice parameter from

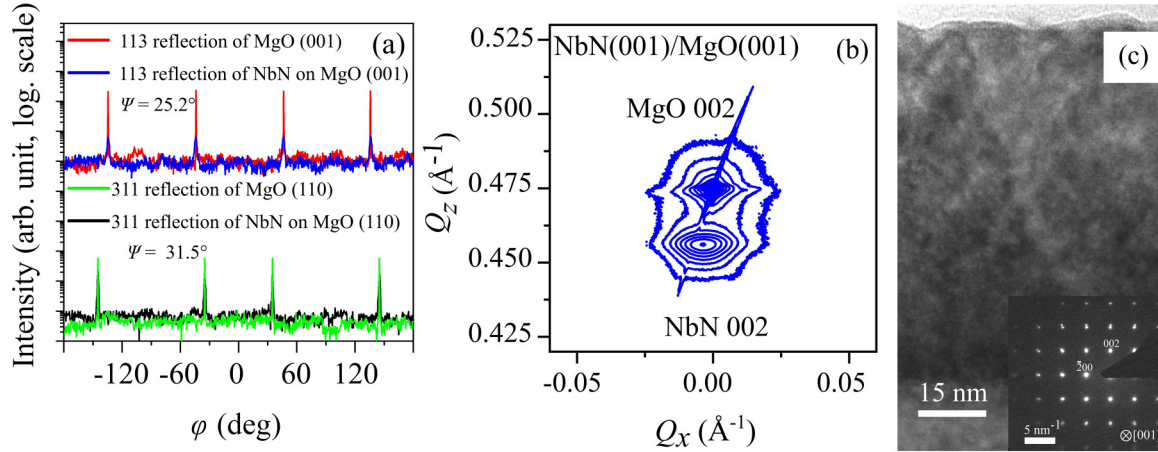


FIG. 2. Structural characterization of 70-nm-thick NbN films grown on MgO substrates. (a) Representative azimuthal φ scans of the off-axis MgO and NbN 113 reflections for the MgO(001) sample at $\psi = 25.2^\circ$, and the corresponding 311 reflections for the MgO(110) sample at $\psi = 31.5^\circ$. (b) Reciprocal-space map around the symmetric 002 reflections of NbN/MgO(001). The feature at higher Q_z corresponds to the MgO 002 substrate reflection, whereas the feature at lower Q_z is assigned to the NbN 002 film reflection. (c) Cross-sectional bright field TEM image of NbN/MgO(001); inset: selected area electron diffraction pattern (zone axis [001]).

approximately 4.37 to 4.25 Å would, for a Poisson ratio of 0.20–0.30, predict an out-of-plane lattice parameter of approximately 4.43–4.47 Å under an isotropic biaxial-elastic model. The measured value, 4.399 Å, is lower than this elastic prediction, indicating an additional lattice-contracting contribution. Together with the DFT results, which show that vacancy disorder can produce comparable reductions in lattice parameter and volume, this supports a vacancy-linked contribution to the thickness-dependent lattice evolution. The chemical compositions were measured for 70 and 205 nm samples and showed similar ratios of Nb/N ratios equal to ~ 1 . The chemical compositions of thinner samples cannot be determined with sufficient certainty using ERDA due to overlapping of N and O signals from the substrate. At low thickness, both NbN(001) and NbN(110) films adopt tetragonal structures with comparable volumes. However, $c/a > 1$ for NbN(001) and $c/a < 1$ for NbN(110), showing that they exhibit different tetragonal distortions when thin, which relax as the film thickness increases.

First-principles calculations provide a consistency check for the structural and energetic effects of vacancies. Figure 3(a) shows that increasing vacancy concentration reduces the lattice constant and unit-cell volume, with stronger contraction for Nb-deficient and coupled Nb-N vacancy configurations than for N-deficient NbN. Figure 3(b) shows that

moderate vacancy concentrations are energetically accessible, whereas highly vacancy-rich configurations become less favorable. The calculations therefore support vacancy-related lattice contraction but do not define a unique vacancy concentration for each film.

Quantitatively, we estimate the microstructural coherence lengths from symmetric reciprocal-space widths [21] via

$$L_{\parallel} = \frac{2\pi}{\Delta Q_x} = \frac{\lambda}{2\beta_{\omega} \sin \theta_B}, \quad L_{\perp} = \frac{2\pi}{\Delta Q_z} = \frac{\lambda}{\beta_{2\theta} \cos \theta_B},$$

where ΔQ_x and ΔQ_z are the full widths at half maximum of the NbN peak along the in-plane and out-of-plane directions in reciprocal space, respectively [Fig. 4(a)]. These coherence lengths should be interpreted as effective values that incorporate finite domain size as well as mosaic and strain broadening.

Figure 4 quantifies the thickness evolution of microstructural coherence by analyzing RSM peak widths and comparing with TEM. Both the lateral coherence length $L_{\parallel}$ and the vertical coherence length $L_{\perp}$ increase with thickness for each orientation [Fig. 4(a)]. The vertical coherence length grows from approximately 7–9 nm in the thinnest films to about 29–34 nm in the thickest films, while the lateral coherence length increases from about 4–5 nm to roughly 7–14 nm depending on orientation. The STEM images in Figs. 4(b)

TABLE I. Out-of-plane and in-plane lattice constants and corresponding unit-cell volume of δ -NbN films grown on MgO(001) and MgO(110) as a function of film thickness. The lattice parameters [a and b for MgO(001) and a, b, c for MgO(110)] are reported to three significant figures due to error propagation (Sec. S4 in the Supplemental Material [20]).

Thickness (nm)	NbN on MgO(001)			NbN on MgO(110)			
	$a = b$ (Å)	c (Å)	V (Å ³)	a (Å)	b (Å)	c (Å)	V (Å ³)
9	4.25	4.399	79.4	4.41	4.43	4.17	81.5
18	4.28	4.408	80.8	4.40	4.41	4.20	81.5
70	4.36	4.387	83.4	4.38	4.38	4.31	82.8
205	4.37	4.375	83.5	4.37	4.37	4.37	83.3

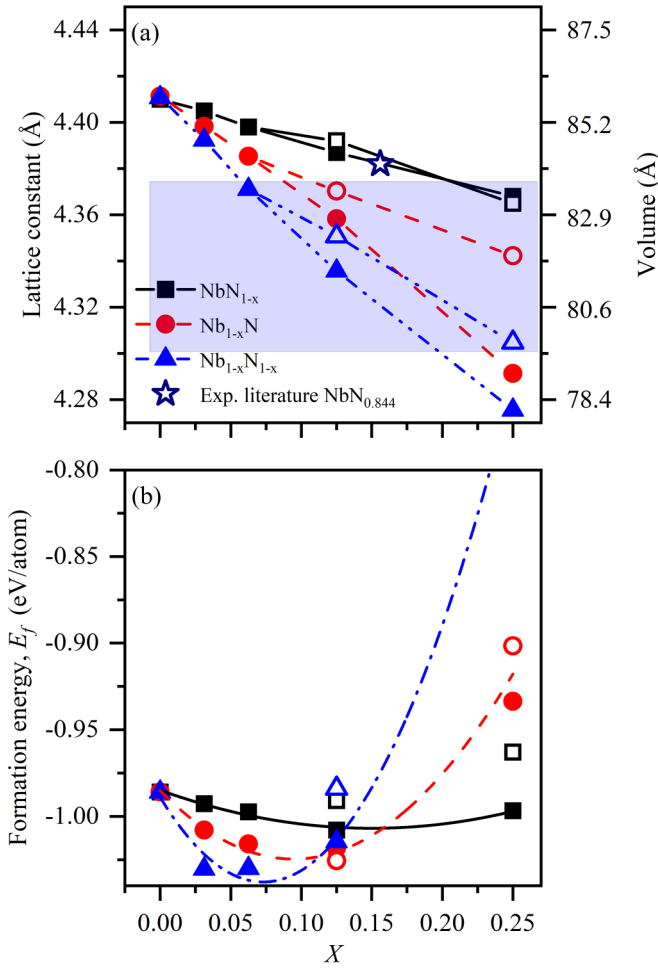


FIG. 3. (a) DFT-relaxed lattice constant a (left axis) and unit-cell volume V (right axis) of cubic (B1) NbN as a function of vacancy concentration x for N-deficient NbN_{1-x} (black squares), Nb-deficient Nb_{1-x}N (red circles), and coupled Nb + N vacancies $\text{Nb}_{1-x}\text{N}_{1-x}$ (blue triangles). Filled symbols correspond to ordered vacancy configurations, while open symbols correspond to SQS (disordered) supercells. The shaded band indicates the range of unit-cell volumes measured for NbN films of different thicknesses grown on MgO(001) and MgO(110). The star marks an experimental literature value for $\text{NbN}_{0.844}$ [25]. For comparison with experiment, the DFT lattice constants in (a) are uniformly shifted downward by ~ 0.02 Å; the unshifted data are provided in the Supplemental Material (Fig. S3) [20]. (b) DFT-calculated formation energy (E_f) vs x for the same vacancy configurations (lines are guides to the eye).

and 4(c) directly visualize dislocation networks and contrast variations consistent with a finite coherence length. We interpret these results as evidence that the thickness dependence is governed mainly by a defect-rich region near the film/substrate interface: dislocations are concentrated near the interface and become less dense as the film thickens and relaxes, reducing mosaicity (and associated broadening) while preserving epitaxial alignment [22,23].

In summary, Figs. 1–4, Table I, and Fig. S1 display a consistent structural picture: (i) the films are predominantly single texture and epitaxial on both MgO(001) and MgO(110), with only the thickest NbN(110) film showing a minor secondary

component, as also reported for other thicker epitaxial films [24]; (ii) lattice metrics evolve systematically with thickness; and (iii) microstructural coherence increases with thickness, reflecting a gradual reduction of microstructural disorder. This controlled structural axis provides a foundation for interpreting the normal-state and superconducting transport trends below.

B. Normal-state transport and superconducting length scales

Figure 5 summarizes the room-temperature resistivity as a function of thickness and microstructural coherence length. The observed thickness dependence is unlikely to originate primarily from classical surface or boundary scattering, because the transport-derived mean free paths, approximately 0.07–0.99 nm, are much smaller than both the film thicknesses and the XRD-derived coherence lengths. Instead, the resistivity evolution is attributed mainly to thickness-dependent atomic-scale disorder, including vacancies, off-stoichiometry, and strain-related defect populations, which affect both the effective carrier density and the carrier mobility. In thinner films, the defect-rich and highly strained region near the film-substrate interface represents a larger fraction of the conducting volume.

The 205-nm NbN(110) film deviates from the otherwise decreasing resistivity trend. This sample contains a detectable secondary (111) component, which may introduce additional structural disorder and interfaces between differently oriented regions. Thus, the resistivity is governed by the combined evolution of electronic disorder, carrier density, and microstructural connectivity rather than by film thickness or structural coherence length alone.

For the low-temperature resistivity measurements, we selected five samples: three from the NbN(110) series (including the thinnest and thickest films) to see the effect of thickness and two from the NbN(001) series for comparison.

Figure 6(a) compares $\rho_{xx}(T)$ of the studied films. The inset highlights that all films become superconducting, but their T_c and the transition width vary across the series. Figure 6(b) shows a pronounced minimum in $d\rho_{xx}/dT$ around 50–80 K for all samples, i.e., the magnitude of the negative $d\rho_{xx}/dT$ is maximal in this intermediate-temperature window. A broadly negative $d\rho_{xx}/dT$ in a metal is a well-known hallmark of transport close to the Mott-Ioffe-Regel/“Mooij” regime, where increasing disorder correlates with a suppressed (and sometimes negative) $d\rho_{xx}/dT$. A weak-localization-based interpretation of the Mooij rule exists and emphasizes that quantum-interference corrections can reduce the effective electron-phonon contribution and drive $d\rho/dT < 0$ when the mean free path approaches the interatomic scale [26–28]. However, such an explanation is not conclusive and typically requires corroboration from magnetoresistance and dimensionality analysis. In strongly disordered epitaxial NbN thin films, $d\rho/dT < 0$ (and even $dR_H/dT < 0$) is reportedly persisting up to ~ 300 K for $k_F\lambda \lesssim 8$, together with an empirical scaling between $R_H(T)$ and $\rho(T)$ that arguably do not reconcile with standard weak localization alone [8]. From this perspective, the 50–80 K dip is best interpreted as a crossover temperature scale where the anomaly is strongest, while at lower T the inelastic channel progressively freezes out and

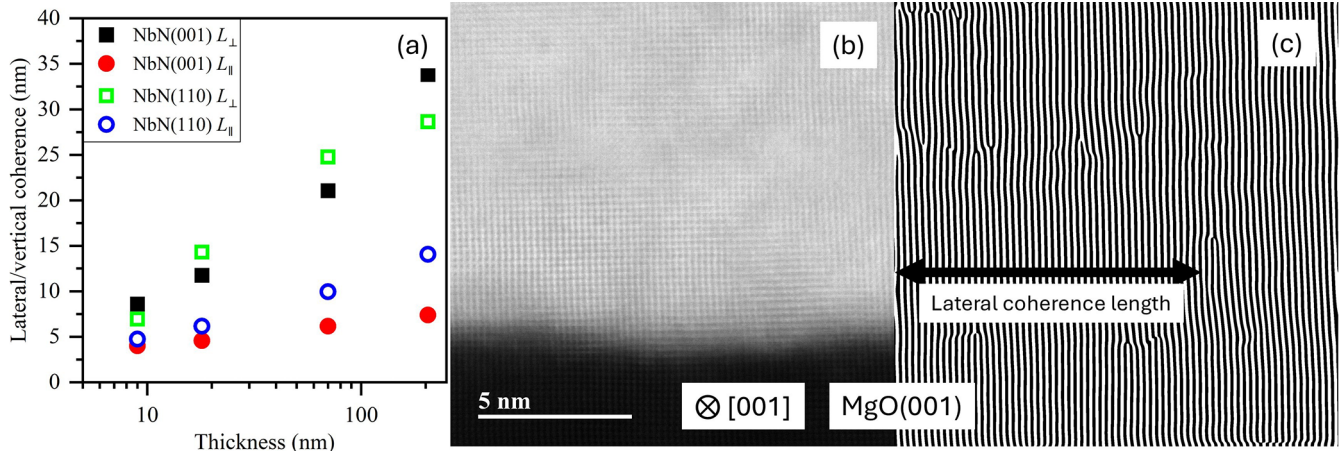


FIG. 4. (a) Thickness dependence of the lateral ($L_{\parallel}$) and vertical ($L_{\perp}$) coherence lengths of NbN films grown on MgO(001) and MgO(110), extracted from RSM via peak broadening along Q_x (in plane) and Q_z (out of plane), respectively. (b) High-resolution HAADF-STEM image of a 70-nm NbN film on MgO(001). (c) Inverse FFT of the STEM image highlighting vertical defect contrast; the arrow indicates the lateral coherence length scale.

$d\rho/dT$ moves back toward zero. The thick NbN(110)–205 nm film displays a larger magnitude of $d\rho_{xx}/dT$ and a deeper minimum than the other samples, whereas the remaining films show comparatively similar, shallow minima. Based on the data presented in Table II, we see that 205 nm NbN(110) film has a high disorder ($k_F l = 1.1$) and a short mean free path ($l = 0.7$ Å). We therefore treat the derivative minimum as an empirical signature of enhanced disorder to transport well above T_c .

Table II summarizes the longitudinal transport metrics, Hall response, and microstructural coherence length extracted from XRD across the thickness/orientation series. The Hall coefficient is negative for all samples: for NbN(110), R_H evolves from -6.1 to -2.0 to -0.5 ($\times 10^{-10} \Omega \frac{m}{T}$) as thickness increases from 9 to 70 to 205 nm, and for NbN(001) it changes from -10.1 to -2.0 ($\times 10^{-10} \Omega \frac{m}{T}$) as thickness increases from 70 to 205 nm. Within a single-band Drude estimate [29] this corresponds to an increase in an effective

carrier density with thickness (as listed in Table II); however, in strongly disordered and/or microstructurally heterogeneous films, R_H can also reflect mobility weighting and current-path connectivity, so we treat n , k_F , l , and $k_F l$ as effective parameters used to compare trends.

When considered together with the structural evolution established from diffraction and the vacancy-related volume trends supported by DFT, this Hall response points to a thickness-dependent defect/strain evolution rather than a purely geometric size effect. Both the in-plane and out-of-plane lattice parameters were determined, showing that the thickness evolution cannot be attributed to strain alone. The systematic increase of the unit-cell volume with thickness indicates an additional contribution beyond purely elastic relaxation (Table I). The DFT results provide a consistency check: varying the vacancy content produces volume changes comparable to the experimentally observed range, and the calculated formation energies vary only weakly over a finite

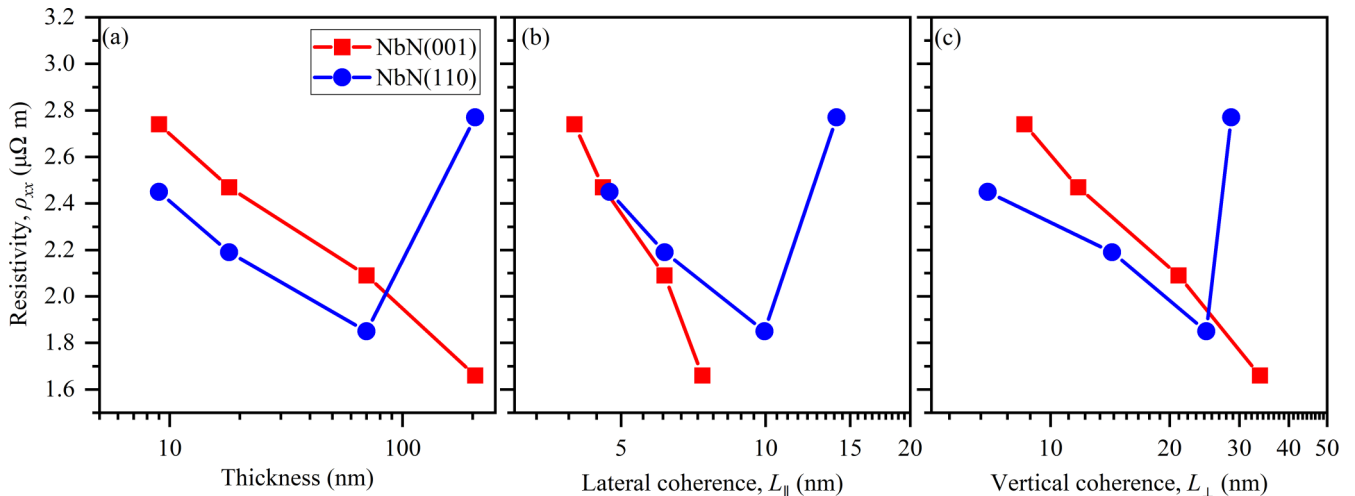


FIG. 5. Room-temperature resistivity of NbN films as a function of (a) film thickness, (b) lateral coherence length, and (c) vertical coherence length. Red squares represent NbN(001) and blue circles represent NbN(110); lines are guides to the eye.

TABLE II. Summary of normal-state and superconducting parameters for NbN films grown on MgO(110) and MgO(001) with different thicknesses. Listed are the longitudinal resistivity at 300 K $\rho_{xx}(300\text{ K})$, 14 K $\rho_{xx}(14\text{ K})$, residual resistivity ratio (RRR), which is $\rho_{xx}(300\text{ K})/\rho_{xx}(14\text{ K})$, Hall constant R_H , effective carrier density n_{eff} , Fermi wave vector k_F , mean free path l , disorder parameter $k_F l$, superconducting transition temperature $T_{c,50\%}$ and $T_{c,95\%}$, $\Delta T_c = T_{c,95\%} - T_{c,1\%}$, zero-temperature upper critical field $B_{c2}(0)$, and zero-temperature superconducting coherence length $\xi_{\text{SC}}(0)$ estimated from $B_{c2}(0)$, and structural lateral ($L_{\parallel}$) and vertical ($L_{\perp}$) coherence length.

	ρ_{xx} (300 K) ($\mu\Omega\text{ m}$)	ρ_{xx} (14 K) ($\mu\Omega\text{ m}$)	RRR	$R_H \times 10^{-10}$ ($\Omega\text{ m/T}$)	$n_{\text{eff}} \times 10^{28}$ m^{-3}	k_F ($\text{\AA}^{-1}$)	l ($\text{\AA}$)	$k_F l$	$T_{c,50\%}$ (K)	$T_{c,95\%}$ (K)	ΔT_c (K)	$B_{c2}(0)$ (Tesla)	$\xi_{\text{SC}}(0)$ (nm)	$L_{\parallel}$ (nm)	$L_{\perp}$ (nm)
NbN(110)-9 nm	2.45	3.88	0.63	-6.1	1.02	0.67	6.9	4.6	8.5	9.3	1.86	17.0	4.4	4.7	6.9
NbN(110)-70 nm	1.85	3.25	0.57	-2.0	3.12	0.97	3.9	3.8	8.5	8.9	1.36	14.9	4.7	9.9	24.8
NbN(110)-205 nm	2.77	7.29	0.38	-0.5	12.5	1.57	0.7	1.1	11.2	11.9	1.37	44.9	2.7	14.0	28.6
NbN(001)-70 nm	2.09	3.80	0.55	-10.1	0.62	0.57	9.9	5.6	10.5	10.8	0.65	18.0	4.3	6.2	21.1
NbN(001)-205 nm	1.66	3.39	0.49	-2.0	3.12	0.97	3.8	3.7	12.3	12.4	0.36	31.8	3.2	7.4	33.9

vacancy fraction (Fig. 3), implying that vacancy concentrations can be altered without a large energetic penalty during growth. Since elastic strain can couple to point-defect thermodynamics and growth-induced defect incorporation, it is plausible that strain relaxation and vacancy-linked structural evolution occur together during growth and subsequent relaxation. In this picture, the thickness-driven approach toward larger unit-cell volumes is consistent with a possible reduction in cation-vacancy content consistent with the DFT mapping, and it aligns with the concurrent reduction in $|R_H|$ (increase in n_{eff}) with thickness. Since the nominal deposition conditions were identical across the series, we associate this trend primarily with thickness- and orientation-driven changes in strain/vacancy concentration, rather than with variations in growth parameters. The link between effective carrier density trends and superconducting properties in epitaxial NbN is also consistent with prior Hall-based analyses in this materials family [7].

The transport-derived mean free paths listed in Table II range from approximately 0.07 to 0.99 nm, whereas the XRD-derived lateral and vertical coherence lengths range from approximately 4.7 to 33.9 nm. The electronic mean free path is therefore substantially shorter than the characteristic microstructural coherence dimensions for every measured sample. This separation of length scales indicates that momentum relaxation is not primarily limited by the finite XRD coherence length, but is more consistent with scattering from atomic-scale disorder, including point defects, off-stoichiometry, and short-range structural disorder.

Turning to the superconducting transition, Table II shows the thicker measured films exhibit higher T_c values for both orientations, together with higher effective carrier densities, in parallel with the increase of the effective carrier density n_{eff} . This trend mirrors the behavior reported for epitaxial NbN by Chockalingam *et al.* [7], where variations in T_c track changes in carrier density. The resistivity values in the present work, 1.66–2.77 $\mu\Omega\text{ m}$, fall within the broader range of 9.4–3.83 $\mu\Omega\text{ m}$ reported by Chockalingam *et al.* [7]. While they induced vacancy-related changes by varying the deposition conditions, our fixed growth conditions indicate that thickness-dependent strain relaxation and vacancy evolution can produce comparable variations in resistivity and T_c . In our case, the coupled structural/DFT picture provides a natural pathway for this correlation: the thickness-driven increase in unit-cell volume (Table I), together with the DFT vacancy-

volume mapping (Fig. 3), is consistent with a trend consistent with a reduction in Nb-vacancy content, which would increase the electronic filling and therefore n_{eff} , concomitantly raising T_c . We emphasize that n_{eff} is an effective single-band estimate and that scattering/inhomogeneity still influences transport (as reflected by $k_F l$ and transition widths), but the observed co-evolution of T_c and n_{eff} indicates that carrier-density changes linked to strain/vacancy evolution are a primary driver of the thickness dependence of T_c in the present series.

We define the transition width as $\Delta T_c = T_{95\%} - T_{1\%}$, where $T_{95\%}$ and $T_{1\%}$ are the temperatures at which $\rho_{xx}(T)$ is 95% and 1% of $\rho_{xx}(14\text{ K})$, respectively. ΔT_c is generally larger for the NbN(110) films than for the NbN(001) films. Within the NbN(110) series, it decreases from the 9-nm film to the thicker films but remains nearly unchanged between 70 and 205 nm. This behavior does not correlate with the disorder parameter $k_F l$, which is similar for the NbN(001) and NbN(110) series [except for the 205-nm NbN(110) sample]. Likewise, ΔT_c shows no clear dependence on the microstructural coherence length. The different tetragonal distortions, $c/a > 1$ for NbN(001) and $c/(a \text{ or } b) < 1$ for NbN(110) at low thickness, also do not track ΔT_c , since the thickest films are essentially cubic ($c/a \approx 1$) and structurally very similar. The remaining distinction between the two sets is the growth orientation; at present we cannot establish a direct connection to ΔT_c , and the microscopic origin of this difference remains an open question.

$B_{c2}(0)$ is estimated using the Werthamer-Helfand-Hohenberg (WHH) orbital-limited expression in the dirty limit,

$$B_{c2}(0) = -0.693 T_{c,95\%}(0) \left. \frac{dB_{c2}}{dT} \right|_{T=T_c},$$

where the numerical prefactor 0.693 is the standard WHH value for dirty superconductors when orbital pair breaking dominates (see Sec. S5 in the Supplemental Material for detailed calculations [20]). To quantify how superconductivity evolves across thickness and orientation, we extract the zero-temperature coherence length from $\xi_{\text{SC}}(0) = \sqrt{\Phi_0/2\pi B_{c2}(0)}$ where Φ_0 is the flux quantum [30]. The thickest films exhibit the largest $B_{c2}(0)$ estimates, reaching 31.8 T for NbN(001) and 44.9 T for NbN(110), together with superconducting coherence lengths of 3.2 and 2.7 nm, respectively. These large B_{c2} values are consistent with dirty-limit scattering and short

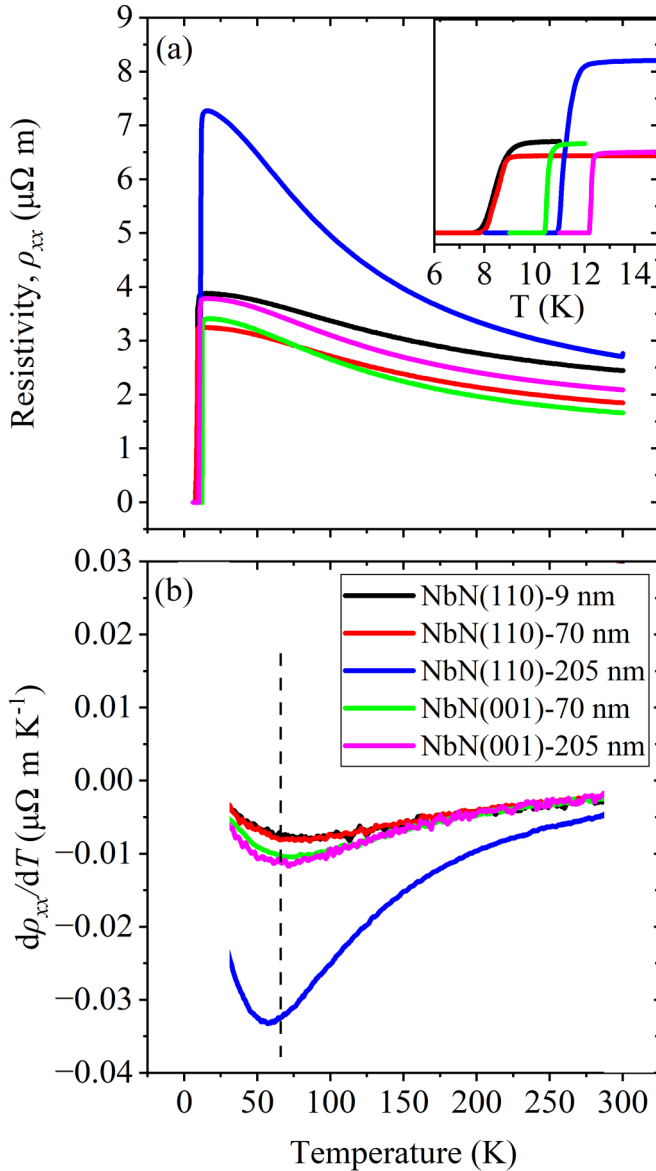


FIG. 6. Temperature dependence of the longitudinal resistivity ρ_{xx} and its derivative for NbN films of different thicknesses on MgO substrates. (a) $\rho_{xx}(T)$ for NbN on MgO(110) with thicknesses of 9, 70, and 205 nm and NbN on MgO(001) with thicknesses of 70 and 205 nm; the inset shows an enlarged view of the superconducting transition. (b) Corresponding $d\rho_{xx}/dT$ curves for the films. The color code for the different films is indicated in panel (b).

superconducting coherence lengths. However, the present data do not allow a unique microscopic mechanism to be assigned to the thickness dependence of B_{c2} . The largest values occur in samples with relatively low $k_F l$, but B_{c2} does not follow the XRD-derived microstructural coherence lengths.

IV. CONCLUSION

In this thickness/orientation series of epitaxial δ -NbN on MgO, the normal-state transport is governed by an inter-

play between structural evolution occurring across the film thickness, microstructural coherence length, and electronic disorder. At room temperature, ρ_{xx} generally decreases with thickness in films that retain a single epitaxial registry. In contrast, the 205-nm NbN(110) film, which exhibits a weak secondary (111) texture, shows an increase in ρ_{xx} despite its larger microstructural coherence lengths, indicating that resistivity is not governed by microstructural coherence alone. All measured films show a negative temperature coefficient of resistivity, and the broad minimum in $d\rho_{xx}/dT$ around 50–80 K correlates more closely with the disorder parameter $k_F \uparrow$ than with the microstructural coherence lengths, indicating that the relevant momentum-relaxation physics is set primarily by atomic-scale electronic disorder rather than grain-scale coherence. Hall data show that the effective carrier density n_{eff} increases with thickness and correlates with the evolution of unit-cell volume in a manner consistent with vacancy-linked structural evolution, likely coupled to strain, which coincides with the observed increase in T_c . The transition width is generally larger for NbN(110) than for NbN(001), but no clear correlation with microstructural coherence length, disorder parameter, mean free path, or superconducting coherence length is established. Finally, the largest B_{c2} estimates occur in the thickest films and correspond to short superconducting coherence lengths, consistent with dirty-limit behavior; however, the present data do not establish a unique microscopic origin for the thickness dependence of B_{c2} .

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During the preparation of this work, the authors used ChatGPT, a language model developed by OpenAI, in order to improve the language and grammar of the manuscript. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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