

Epitaxial YbN thin films grown by nitrogen plasma-assisted molecular beam epitaxy

Y. Chen^{1,2}, A. Meléndez-Sans¹, S. Kimura^{2,3,4}, L. H. Tjeng¹, and S. G. Altendorf¹¹*Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, 01187 Dresden, Germany*²*Department of Physics, Graduate School of Science, The University of Osaka, Toyonaka 560-0043, Japan*³*Graduate School of Frontier Biosciences, The University of Osaka, Suita 565-0871, Japan*⁴*Institute for Molecular Science, Okazaki 444-8585, Japan*

(Received 30 October 2025; accepted 3 December 2025; published 24 December 2025)

We report the successful growth of stoichiometric, epitaxial ytterbium nitride (YbN) thin films via molecular beam epitaxy under ultrahigh vacuum conditions using activated nitrogen supplied by a plasma source. Through systematic optimization of the growth parameters, we achieved high-quality YbN films with excellent crystallinity and a well-defined (100) out-of-plane orientation on MgO(100) and LaAlO₃(100) substrates, as determined by *in situ* reflection high-energy electron diffraction and *ex situ* x-ray diffraction. *In situ* photoelectron spectroscopy results unambiguously demonstrate the semiconducting character of YbN and the fully trivalent valence state of the Yb ions. Using the photon-energy dependence of the valence band spectra we were able to reveal a significant hybridization between the Yb 4*f* and N 2*p* states.

DOI: [10.1103/chf5-cc7n](https://doi.org/10.1103/chf5-cc7n)

I. INTRODUCTION

Rare-earth (RE) mononitrides (RENs) crystallized in the fcc rocksalt structure show interesting magnetic properties due to the strongly correlated and localized 4*f* electrons. Most RENs exhibit semiconducting behavior with a ferromagnetic ground state, making them promising candidates for various electronic and spintronic devices, attracting significant research interest [1–6].

In the ferromagnet-dominated REN family [1,7,8], ytterbium nitride (YbN) stands out due to its antiferromagnetic ground state with a reported bulk Néel temperature of 0.79 K [9]. Like the other RENs, YbN adopts the rocksalt crystal structure and it has the second smallest lattice parameter (4.7852 Å) within the series [10]. Early studies on bulk crystals suggested a metallic or self-compensated semimetallic behavior [11]. However, the transport characteristics as well as the electronic structure have been subject of extensive debate. Meanwhile, spectroscopic characterization of bulk samples is limited, and the presence of divalent ytterbium has also been reported [12].

There is a general consensus that the controversies about the intrinsic properties of YbN can be traced back to material issues. Many samples, particularly the early bulk samples, suffered from poor crystallinity, nonstoichiometry, or partial oxidation. Also theoretical predictions of the electronic structure and properties of YbN and the RENs in general are intricate, given the necessity of treating the extended N 2*p* and RE 5*d* bands simultaneously with the strongly correlated localized 4*f* states. This sparked a debate within the band

structure community regarding the nature of the material, particularly whether it exhibits a (semi-)metallic or semiconducting character [13,14].

During the last two decades, experimental studies on RENs have focused on thin-film samples synthesized in ultrahigh vacuum (UHV) systems, which provide better control of stoichiometry and cleanliness, and are more suitable for the fabrication of potential devices, such as spin-filter Josephson junctions [2,4,5]. Molecular beam epitaxy (MBE) has become the preferred method for REN thin films for its high crystal quality results [15]. Most RE atoms catalyze the breakdown of molecular nitrogen, allowing REN layers to form simply by depositing RE atoms in an N₂ atmosphere [16]. Numerous studies on the growth of polycrystalline GdN [3,17–19], SmN, DyN, ErN, and LuN films [20,21], as well as epitaxial GdN [22–25], SmN [24–31], DyN [25], and HoN [32] films have been reported, although achieving good crystalline quality (i.e., epitaxial and with well-defined crystalline orientation) and stoichiometry remains challenging.

For the growth of YbN thin films, two aspects must be considered: first, ytterbium has two possible valence states (2+ and 3+), and second, it is incapable of catalyzing the dissociation of nitrogen molecules [16]. Therefore, the synthesis of YbN thin films requires an activated nitrogen source. As a result, in contrast to the extensive research on other RENs, the literature on YbN thin films is limited. Warring *et al.* reported the synthesis of polycrystalline YbN films with (111) texture on Al₂O₃(0001) substrates by MBE using a Kaufman-type ion source and suggested a semiconducting nature together with a fully trivalent state of Yb ions [33]. More recently, another report on the synthesis of YbN(111) thin films on MgO(100) substrates by magnetron sputtering also indicated that YbN is a semiconductor, while a mixed-valence state of Yb ions was observed [34].

In this work, we systematically investigate the MBE growth of YbN on MgO(100) and LaAlO₃(100) substrates

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using a RF plasma source to supply activated nitrogen. By employing the nonpolar (100) surface orientation, we circumvent the inherent polar surface instability associated with the (111) growth direction [35–38]. Our objective is to synthesize high-quality YbN thin films with epitaxially ordered crystalline structure and controlled stoichiometry, and to clarify the Yb valence state as well as the semiconducting or (semi-)metallic nature of YbN. The crystalline structure, chemical composition, Yb valence state, and electronic structure of the films are studied by a combination of reflection high-energy electron diffraction (RHEED), x-ray photoelectron spectroscopy (XPS), and ultraviolet photoelectron spectroscopy (UPS) performed *in situ* (i.e., without removal from the UHV environment) as well as *ex situ* x-ray diffraction (XRD).

II. METHODS

YbN thin films were grown by MBE on epi-polished MgO(100) substrates purchased from Crystec GmbH and LaAlO₃(100) substrates purchased from Crystal GmbH. The lattice parameters of MgO and LaAlO₃ are 4.21 and 3.79 Å (5.36 Å when considering 45° in-plane rotation), resulting in a lattice mismatch of −13.7% and +10.7% in comparison with the reported YbN bulk lattice parameter of 4.7852 Å [10]. Before being loaded into the UHV system, the substrates were cleaned in acetone and isopropanol using an ultrasonic bath. The substrates were then annealed in an O₂ atmosphere ($P_{O_2} = 1 \times 10^{-6}$ mbar) at 600 °C for 2 h in the growth chamber to produce a well-ordered surface for epitaxial growth.

The base pressure of the MBE system was approximately 1×10^{-10} mbar. High-purity ytterbium metal with an initial purity level of 99.9%, that was additionally refined at 1000 °C in a Ta ampoule, was evaporated from a standard effusion cell. The Yb flux rate was measured at the deposition position using a quartz crystal thickness monitor, and varied from 1 to 10 Å/min ($T_{Yb} = 335\text{--}410$ °C) for growth optimization. By thorough degassing of the Yb effusion cell, the background pressure was maintained below 5×10^{-9} mbar, even at the highest Yb flux rate of 10 Å/min during deposition. Nitrogen was supplied via an RF plasma source (UNI-Bulb RF Plasma Source from Veeco), operating in the high-brightness mode. The power of the plasma source was varied from 100 to 200 W, and the nitrogen flow was adjusted from 0.1 to 0.4 SCCM (cubic centimeter per minute at STP). During growth, the pressure in the growth chamber was 2.5×10^{-6} mbar at 100 W/0.1 SCCM, and increased to 1×10^{-5} mbar at 200 W/0.4 SCCM. The distance between the plasma source and sample surface was set to 10 cm. Different substrate temperatures (T_{sub}) varying from 30 to 400 °C were chosen for each film.

During the deposition process, an *in situ* RHEED system (STAIB Instrument RH35, acceleration voltage = 20 kV) was employed to characterize the surface atomic structure of the substrates and the films. The electronic structure of the fabricated thin-film samples was analyzed by *in situ* XPS and UPS. Monochromated Al $K\alpha$ radiation ($h\nu = 1486.6$ eV) was used for XPS measurements, and the energy resolution was estimated to be 0.4 eV. UPS measurements were taken using He I radiation ($h\nu = 21.2$ eV) from a helium discharge lamp without monochromation, with an energy resolution of

approximately 0.1 eV. During the synthesis, sample transfer, and all the above-mentioned measurements, the films were maintained in a UHV environment at a background pressure of about 1×10^{-10} mbar.

The detailed crystalline structure of the films was investigated using *ex situ* XRD. Before removal from the UHV system, approximately 10-nm-thick Cr capping layers were deposited on the top of the sample surface to prevent degradation in air. XRD measurements were conducted using a PANalytical X'Pert PRO diffractometer with monochromatic Cu $K\alpha_1$ radiation ($\lambda = 1.54056$ Å).

III. RESULTS AND DISCUSSION

We first investigate the optimal conditions for the MBE synthesis of YbN thin films. Our attempt to grow YbN with molecular nitrogen at a high nitrogen pressure of 1×10^{-5} mbar, Yb flux rate of 3 Å/min, and substrate at room temperature proved unsuccessful. The XPS spectra showed no signals of the ytterbium reacting with the nitrogen, demonstrating that the presence of molecular nitrogen does not affect the Yb deposition, resulting in a still purely Yb metal film. These results are discussed in more detail in Appendix A. This finding reinforces the importance of using activated nitrogen in the synthesis of YbN, in accordance with other reports [16,33]. In this study, a RF nitrogen plasma source is utilized for this purpose.

Figure 1 shows the RHEED patterns of a MgO(100) substrate and a series of thin films grown at different substrate temperatures ranging from 30 °C to 400 °C. The power of the RF plasma source, nitrogen flow, and Yb flux rate were set to 100 W, 0.1 SCCM, and 5 Å/min, respectively. As shown in Fig. 1(b), at room temperature ($T_{sub} = 30$ °C), a RHEED pattern with rings and faint transmission spots was observed, suggesting that the thin-film sample is primarily polycrystalline with some three-dimensional growth components. At a higher substrate temperature of 200 °C [Fig. 1(c)], the rings remain visible in the RHEED pattern. At the same time, the intensity of the transmission spots increases, becoming comparable to the rings, indicating improved crystalline quality with enhanced island growth and a reduced amount of randomly oriented domains. At even higher temperatures, as shown in Figs. 1(d)–1(f), intensity-modulated streaks appear in the RHEED patterns and become crisper with increasing temperature, suggesting epitaxial layer growth with surface steps. We noticed that faint ringlike structures appear in some of the epitaxial films grown at $T_{sub} > 300$ °C. This may be related to tiny regions of polycrystalline character, or more likely to a minor disorder on the surface of the sample, as no significant effect on the XRD and XPS measurement results was observed.

By comparing the spacing of the RHEED streaks with that of MgO(100) [Fig. 1(a)], the lattice parameter of the synthesized films was determined to be 4.73 ± 0.06 Å, which agrees with the reported bulk lattice parameter of YbN (4.7852 Å [10]) within the measurement accuracy. Furthermore, the real-time RHEED measurements indicate that the YbN films become fully relaxed after approximately 0.5 nm of growth, with no sign of the formation of an oxide layer.

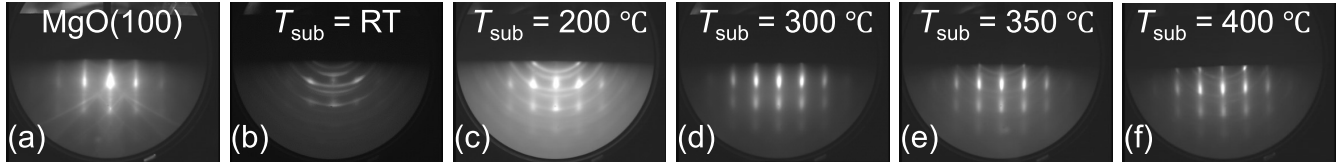


FIG. 1. *In situ* RHEED images of (a) the MgO(100) substrate cleaned by annealing in O_2 atmosphere and of (b)–(f) YbN films grown at a Yb flux rate of $5 \text{ \AA}/\text{min}$, RF plasma source power of 100 W, nitrogen flow of 0.1 SCCM, and indicated substrate temperatures. The incident electron beam was aligned parallel to the [001] crystallographic direction of the MgO(100) substrate.

In addition to the substrate temperature, we studied the effect of the Yb flux rate on the crystalline quality, which has only a minor influence within the investigated range of 1–10 $\text{\AA}/\text{min}$. We also attempted to grow YbN films at higher plasma power and nitrogen flow, which resulted in degraded crystallinity, likely due to nitrogen plasma-induced sputtering. The growth of YbN thin films on $\text{LaAlO}_3(100)$ substrates has also been studied, resulting in epitaxial films of similar quality. Details are provided in Appendix B.

The composition and electronic structure of the fabricated thin films were subsequently investigated using XPS. Figure 2 shows the wide-range XPS spectra of the YbN films fabricated at different substrate temperatures up to 400 °C. Overall, the results are comparable. All of the prominent peaks in the spectra correspond to Yb or N, with mostly similar peak intensities across the sample series. No significant contamination was observed. Only minor peaks assigned to the O 1s core level and O KLL Auger emission were detected, likely originating from oxidation during the XPS measurements, even in an ultrahigh vacuum environment. Evidence of the oxidation process during XPS measurements is presented in Appendix C.

Further detailed XPS results for the core levels and valence band are shown in Fig. 3, along with the reference spectra of Yb metal (predominantly divalent) and Yb_2O_3 (trivalent) [39,40]. Since Yb_2O_3 is a good insulator, an energy shift to higher binding energy due to the charging effect was observed during the XPS measurement. Therefore, the spectra of Yb_2O_3 were shifted 1.7 eV toward lower binding energy to align the 4d peak position with that of the YbN thin film grown at 400 °C.

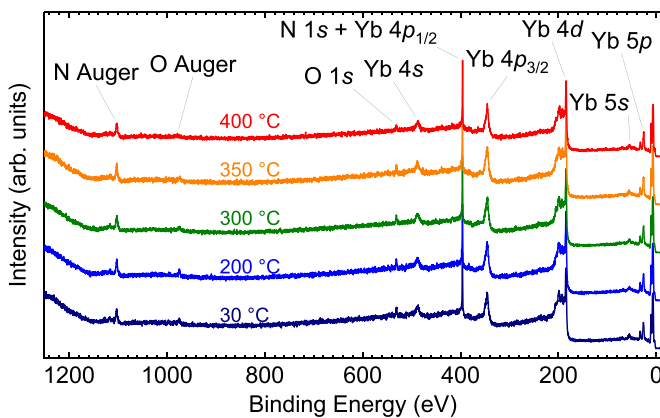


FIG. 2. XPS wide-range spectra of the YbN thin films grown at the indicated substrate temperatures. Each spectrum is shifted upward for clarity.

Figure 3(a) shows the spectra of the N 1s and Yb 4p regions. The sharp peak at 396 eV corresponds to the N 1s core level, which overlaps with the Yb $4p_{1/2}$ peak. To examine the N 1s core level in more detail, the spectra in the N 1s region are expanded and shown in the inset of Fig. 3(a). The spectra are normalized to the integrated area of the corresponding Yb 4d core level to allow for a quantitative comparison of the N 1s intensities of the different films. One can easily find that the N 1s peak retains nearly the same intensity, shape, and binding energy position in samples grown at substrate temperatures up to 350 °C. In contrast, for the sample grown at 400 °C, the N 1s peak shows higher intensity and a sharper shape, suggesting a slightly higher nitrogen concentration and thus fewer nitrogen vacancies. Meanwhile, its position shifts by approximately 0.25 eV toward lower binding energy. A similar shift to lower binding energy is observed for all other XPS peaks, i.e., the entire spectrum, suggesting that the Fermi level of the sample deposited at 400 °C undergoes a downward shift relative to that of films grown at lower substrate temperatures. This behavior was found for all films grown at highest temperatures [also for the films on $\text{LaAlO}_3(100)$ substrates]. Given the established role of nitrogen vacancies as electron donors, one may speculate that this shift can be attributed to the reduced electron doping of films grown at the highest substrate temperature, which would lower the Fermi level.

We then look into the valence state of the Yb ions. The peaks at around 400 and 345 eV in Fig. 3(a) can be assigned to the Yb 4p core levels of Yb^{3+} , as they closely match those in the Yb_2O_3 reference spectrum. On the other hand, in the spectrum of the Yb metal sample, where Yb ions are predominantly in the divalent state [41], a different set of peaks appears at around 390 and 340 eV, which are absent in the YbN samples. The absence of these peaks suggests that Yb ions in the films are in a trivalent state.

A similar conclusion can be drawn from the Yb 4d core level and the valence band spectra displayed in Figs. 3(b) and 3(c). All YbN films exhibit Yb 4d spectra similar in shape to Yb_2O_3 , with no detectable peaks at 182 and 192 eV, which would indicate the presence of Yb^{2+} as shown in the spectra of Yb metal. In the valence band spectra, the YbN thin films show features similar to the Yb $4f^{13}$ multiplets of Yb_2O_3 , but with a sharper spectral shape. Additionally, broad structures centered around 3 and 4 eV are observed, which are attributed to the N 2p and O 2p states in YbN and Yb_2O_3 , respectively. In contrast, sharp $4f^{14}$ doublets appear at around 1.2 and 2.4 eV in the Yb metal spectrum. These Yb^{2+} features are absent in the spectra of the YbN thin films. With an XPS detection limit of down to $\approx 1\%$ concentration for Yb^{2+} , we can exclude even relatively small amounts of divalent Yb ions.

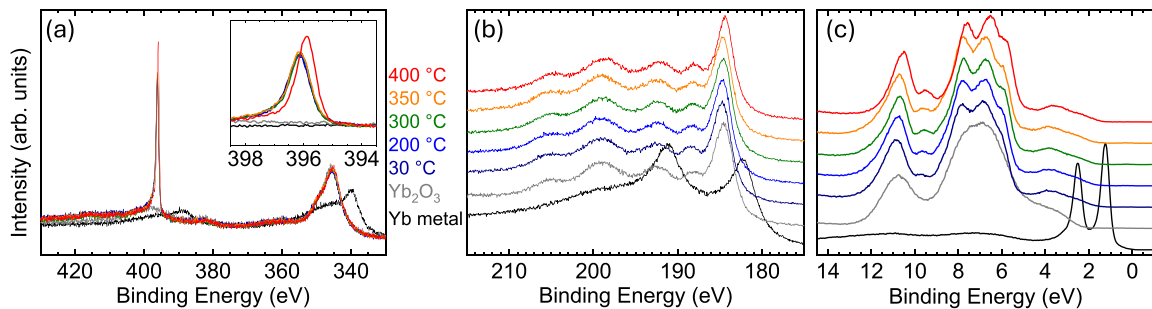


FIG. 3. The XPS spectra of YbN thin films grown at the indicated temperatures: Yb₂O₃ (gray) and Yb metal (black). (a) Yb 4*p* and N 1*s* spectra normalized to the integrated area of the corresponding Yb 4*d* core level. The inset shows the spectra zoomed in on the N 1*s* region. (b) Yb 4*d* spectra normalized to their highest peak. (c) Valence band spectra normalized to their highest peak. Each spectrum is shifted upward for clarity in (b) and (c). The spectra of Yb₂O₃ are shifted by -1.7 eV, aligning the Yb 4*d* core level with that of the YbN film grown at 400 °C, to compensate for the charging effect during XPS measurements.

Therefore, the absence of the Yb²⁺ contribution in both the core-level and valence band spectra confirms that the Yb ions are entirely trivalent in YbN.

It is noteworthy that the growth of YbN, which requires activated nitrogen, differs from the growth of other non-Yb-based RENs. While studies on various REN thin films grown using molecular nitrogen suggest that the higher substrate temperatures favor the formation of nitrogen vacancies [1,28,29,32,42,43], YbN exhibits an even higher and sharper N 1*s* peak for the film grown at the high substrate temperature of 400 °C [Fig. 3(a)], indicating a better nitrogen stoichiometry. Furthermore, the YbN films grown at high substrate temperatures show no signs of degradation in crystalline quality [Fig. 1(f)] or oxidation (Fig. 2) compared to the films grown at lower temperatures. These observations contrast with a study on HoN thin films grown with molecular nitrogen [32], where an increased oxygen content along with a degraded RHEED pattern was observed when grown at comparable high substrate temperatures.

The differences may be explained by the enhanced reactivity of radical nitrogen from the RF plasma source compared to the catalytic reaction of molecular nitrogen with rare-earth atoms. This results in excessive nitrogen supply during YbN growth, preventing oxidation and degradation of the thin films. The presence of nitrogen in excess is confirmed by the fact that the composition and electronic structure remained nearly unchanged (while the crystalline quality improved significantly) when the substrate temperature was increased from room temperature to 350 °C, which is already clearly above the distillation temperature of elemental ytterbium at the deposition position, and also by the fact that the films grown under higher nitrogen flow rates and plasma powers show no significant increase in nitrogen contents.

To characterize the crystalline structure in more detail, an *ex situ* XRD measurement was conducted for one of the epitaxial YbN thin-film samples. Before removal from the UHV system, the thin-film sample was capped with a 10-nm chromium layer to prevent degradation in air. Figure 4(a) presents the XRD ω -2 θ scan of a Cr-capped YbN film fabricated at 300 °C. The XRD data show peaks corresponding to the MgO(2*n*00) and YbN(2*n*00) reflections. A minor peak near 65° can be attributed to the (200) reflection of the Cr capping layer, which was also observed in the XRD of

Cr-capped SmN thin films in our previous study [29]. The XRD-derived lattice parameter is 4.786 ± 0.008 Å, in good agreement with our RHEED measurements and the reported bulk value [10]. The inset of Fig. 4(a) shows the rocking curve of the YbN(200) diffraction peak. The FWHM is estimated to be 2.1°, consistent with other epitaxial REN thin films grown by MBE with molecular nitrogen [29,32].

Figure 4(b) shows the in-plane ϕ scan of the YbN(111) and MgO(222) reflections. The fourfold symmetry of the YbN(111) peaks aligned with MgO(222) confirms the epitaxial growth of YbN(100) on MgO(100) without in-plane rotation. The x-ray reflectivity (XRR) result is shown in Fig. 4(c), from which the film thickness was determined to be approximately 13.5 nm, indicating a YbN growth rate of about 2.8 Å/min.

Finally, the electronic structure of the fabricated YbN thin films was investigated in more detail by comparing the valence band spectra obtained at two different photon energies on the same sample. Figure 5 displays the valence band spectra of YbN measured using both UPS and XPS, along with the XPS spectrum of the Yb₂O₃ reference film. As previously discussed, the XPS spectra of YbN and Yb₂O₃ share similar structures above 5 eV, which can be assigned to the Yb 4*f*¹³ multiplets. On the other hand, the features at around 2–5 eV differ markedly in shape and energy position, which is considered to be due to the difference of O 2*p* orbitals in Yb₂O₃ and N 2*p* orbitals in YbN, respectively. In the UPS spectrum of YbN, the structure at approximately 3.6 eV is significantly enhanced compared to the XPS spectrum, and a new strong peak, which is absent in the XPS spectrum, appears at around 4.7 eV. Additionally, the intensities of the spectral features above 5 eV are significantly suppressed. To identify the contributions to the valence band, we consider the differences in the photoionization cross sections for the two photon energies. The value of the N 2*p* orbitals changes from 0.72×10^{-4} Mb for XPS (1486.6 eV) to 9.688 Mb for UPS (21.2 eV) with an enhancement factor of about 10^5 , while the value of the Yb 4*f* orbitals changes from 0.82×10^{-1} Mb to 1.19 Mb with merely an enhancement factor of about 10 [44]. Therefore, the features above 5 eV, which are suppressed in UPS, can be identified as Yb 4*f*¹³ multiplets; and the significantly enhanced structures around 3 eV and the newly appearing peak at approximately 4.7 eV in the UPS

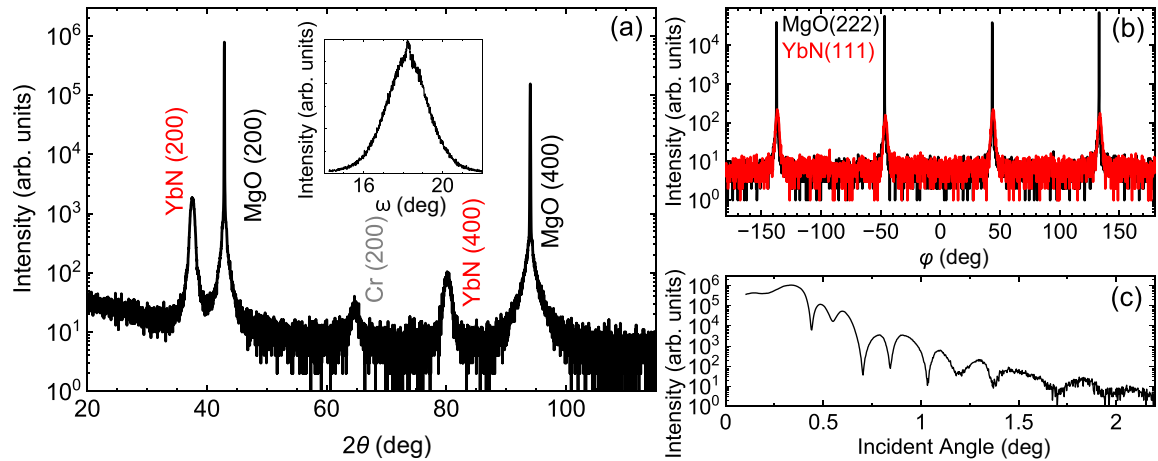


FIG. 4. Crystallinity of the YbN thin film grown at 300 °C, capped *in situ* with about 10 nm of Cr metal, investigated by XRD measurements. (a) XRD ω - 2θ scan. The inset shows the rocking curve of YbN(200) reflection. (b) In-plane ϕ scan of YbN(111) and MgO(222) reflections. (c) X-ray reflectivity (XRR) curve.

spectrum can be considered to be related to the N 2*p* orbitals. Meanwhile, the intensity of the structure assigned to the N 2*p* orbitals is in the same order as the Yb 4*f* multiplets in the XPS spectrum, despite the negligible cross section of N 2*p* compared with Yb 4*f* for the XPS energy. This indicates the presence of significant *p*-*f* hybridization in YbN, consistent with theoretical calculations [13], and similar to experimental results on SmN [29].

The XPS and UPS spectra of YbN also clearly indicate a semiconducting nature of the YbN films. By linear extrapolation of the leading edge of the UPS spectra, the valence band maximum was found to be at 1.64 eV, defining the lower limit of the band gap. This value is in agreement with the previous reported optical band gap of YbN for 1.5–2 eV [33,34]. Some residual intensity near the Fermi level indicates possible defect-induced occupied Yb 5*d* states.

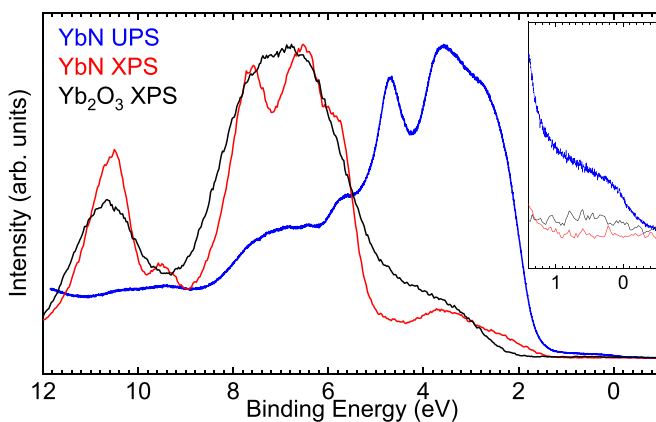


FIG. 5. Valence band spectra measured with UPS (blue) and XPS (red) of a YbN film grown on MgO(100) at the substrate temperature of 400 °C, together with an XPS spectrum (black) of the Yb₂O₃ reference sample. To correct for charging-induced energy shifts, the XPS spectrum of Yb₂O₃ was shifted 1.7 eV toward lower binding energy to align the Yb 4*d* core level and Yb 4*f* multiplets with those of the YbN sample. The inset shows the enlarged view of the Fermi level region.

IV. CONCLUSION

In summary, we successfully grew epitaxial YbN thin films on MgO(100) and LaAlO₃(100) substrates via MBE using an RF plasma nitrogen source, with carefully controlled growth conditions. We investigated their electronic and structural properties using *in situ* XPS, UPS, and *ex situ* XRD measurements. XRD results confirmed that the YbN thin films exhibit a pure (100) orientation and comparable crystalline quality to other REN epitaxial thin films grown by MBE with molecular nitrogen, indicating a significant improvement in crystallinity over previous polycrystalline YbN thin films.

Photoelectron spectroscopy measurements provide direct experimental evidence that the Yb ions in YbN are fully trivalent. The spectra also demonstrate the presence of a band gap, which confirms the semiconducting nature of YbN. Comparing the XPS and UPS valence band spectra with the photon-energy dependence of the elemental photoionization cross sections reveals a significant hybridization of the N 2*p* and Yb 4*f* states.

ACKNOWLEDGMENTS

The authors want to express their gratitude to K. Höfer and T. Mende for their skillful technical assistance, to the department of Claudia Felser for the use of their thin-film XRD instrument, and to P. Höhn for the purification of the Ytterbium. A.M.-S. acknowledges the Material Science Ph.D. program of Universitat Autònoma de Barcelona and M. I. Alonso from ICMAB-CSIC for her supervision. Y.C. is grateful to the Ph.D. support program for Pioneering Quantum Beam Applications (PQBA), and the support from JST, the establishment of university fellowships towards the creation of science technology innovation, Grant No. JPMJSP2138.

DATA AVAILABILITY

The data that support the findings of this article are openly available [45].

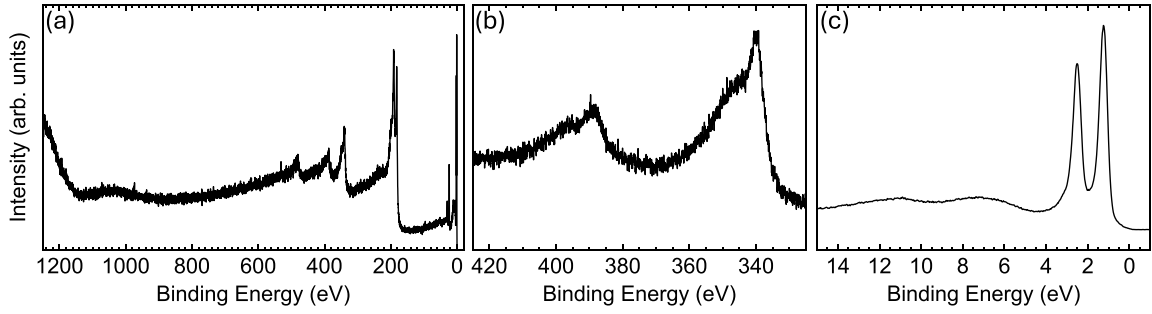


FIG. 6. XPS spectra of a thin-film sample grown on MgO(100) with molecular nitrogen at a nitrogen pressure of 1×10^{-5} mbar, Yb flux rate of $3 \text{ \AA}/\text{min}$, and room temperature. (a) Wide-range spectrum. (b) Yb $4p$ spectrum. (c) Valence band spectrum.

APPENDIX A: ATTEMPT TO GROW YbN THIN FILMS WITH MOLECULAR NITROGEN

Figure 6 shows the XPS results of a sample grown on a MgO(100) substrate with molecular nitrogen at $P_{N_2} = 1 \times 10^{-5}$ mbar and at room temperature. In the wide-range spectrum shown in Fig. 6(a), peaks attributed to Yb and minor peaks attributed to O $1s$ core level and O KLL Auger emission are observed, while no clear signal from nitrogen was detected. The XPS spectrum of the Yb $4p$ core level is shown in Fig. 6(b). Peaks appear at around 390 and 340 eV, which can be attributed to the $4p$ core level of Yb^{2+} , while the features of Yb^{3+} or N $1s$ are absent. In the valence band spectrum exhibited in Fig. 6(c), sharp $4f^{14}$ doublets at 1.2 and 2.4 eV,

together with a very weak feature of $4f^{13}$ multiplets, are observed, similar to the spectrum of Yb metal. The XPS results indicate that the measured thin film contains no nitrogen and that the Yb ions are predominantly divalent. Thus, the film is Yb metal rather than YbN. This finding supports the report on Yb's inability to split nitrogen molecules catalytically, and thus the impossibility of growing YbN thin films with molecular nitrogen [16].

APPENDIX B: GROWTH OF YbN THIN FILMS ON $\text{LaAlO}_3(100)$

Figure 7 shows the results of a YbN film grown at a substrate temperature of 350°C , plasma source power of 100 W,

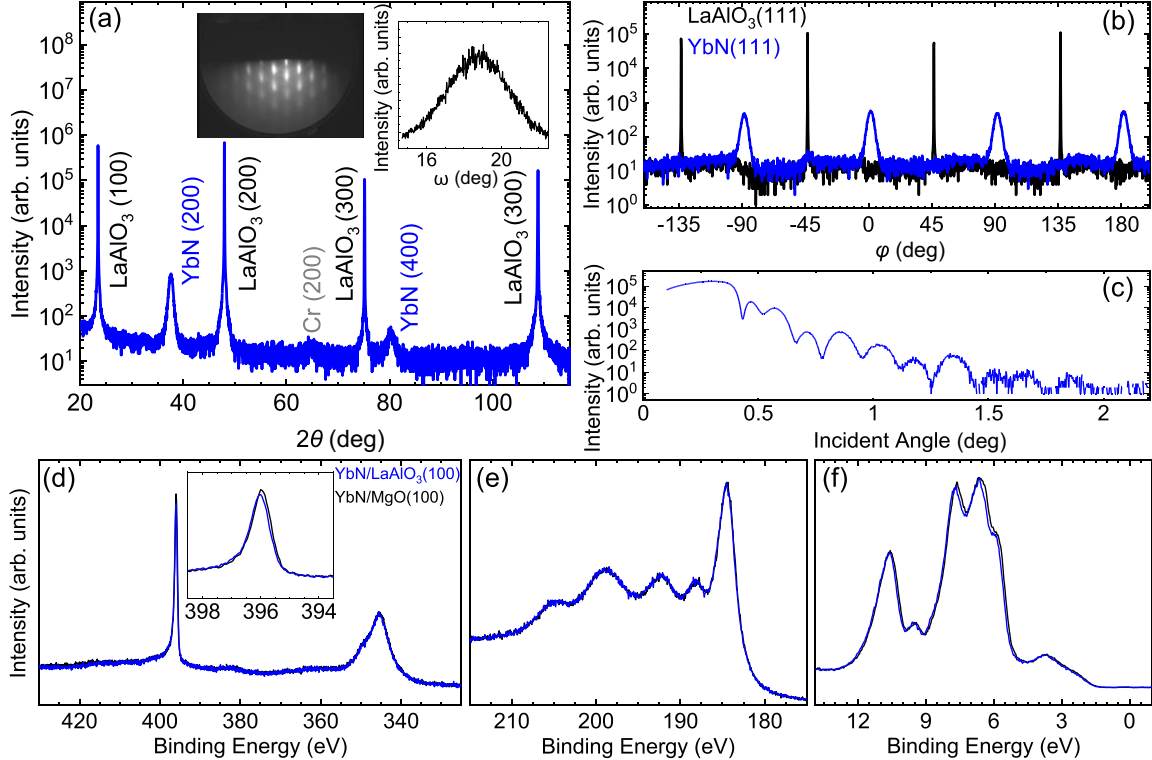


FIG. 7. Characterization of YbN thin films on $\text{LaAlO}_3(100)$ substrates. (a)–(c) XRD of a Cr-capped sample grown at 350°C and Yb flux rate of about $3 \text{ \AA}/\text{min}$ for 80 min. (a) ω - 2θ scan. The RHEED pattern measured *in situ* just after growth, and the rocking curve of the YbN(200) peak is shown in the inset. (b) In-plane ϕ scan of YbN(111) and $\text{LaAlO}_3(111)$. (c) XRR curve. (d)–(f) XPS spectra measured *in situ* of a sample grown at 400°C and on a $\text{LaAlO}_3(100)$ substrate (blue), together with the XPS results of a YbN thin film grown on a MgO(100) substrate under similar conditions (black). (d) Yb $4p$ and N $1s$ core-level spectra. (e) Yb $4d$ core-level spectra. (f) Valence band spectra.

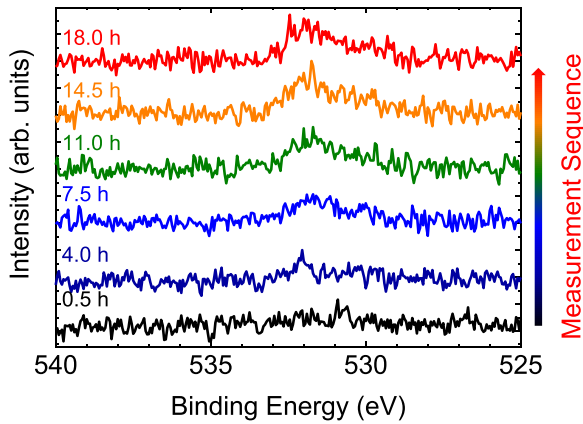


FIG. 8. Time-dependent series of XPS measurements of the O $1s$ core-level region for a YbN thin film grown on a MgO(100) substrate at $T_{\text{sub}} = 400^\circ\text{C}$ and with a Yb flux rate of $5 \text{ \AA}/\text{min}$. For each spectrum, the time after the start of the measurement is indicated. The spectra are shifted upwards in the measurement sequence for clarity.

nitrogen flow of 0.1 SCCM , and Yb flux rate of $3 \text{ \AA}/\text{min}$ on a $\text{LaAlO}_3(100)$ substrate. The XRD ω - 2θ scan is shown in Fig. 7(a), where peaks corresponding to $\text{LaAlO}_3(n00)$ and $\text{YbN}(2n00)$ reflections, and a minor feature of the $\text{Cr}(200)$ reflection at 65° are observed. The lattice parameter is calculated to be $4.783 \pm 0.013 \text{ \AA}$, consistent with YbN grown on MgO(100). The RHEED pattern and the rocking curve of the $\text{YbN}(200)$ reflection are shown in the inset of Fig. 7(a). Similar to the films grown on MgO(100), intensity-modulated streaks and weak additional features appear in the RHEED pattern. The FWHM of the $\text{YbN}(200)$ rocking curve is estimated to be 3.6° , which is broader than that of the YbN on MgO(100), indicating a slightly lower crystalline quality. Figure 7(b) shows the in-plane ϕ scan of the $\text{YbN}(111)$ and $\text{LaAlO}_3(111)$ reflections. A series of fourfold $\text{YbN}(111)$ peaks appears at 45° relative to the $\text{LaAlO}_3(111)$ reflections, revealing a 45° in-plane rotated growth of YbN layers on

LaAlO_3 substrates, in agreement with previous reports of REN thin films on $\text{LaAlO}_3(100)$ [29,32]. The XRR result in Fig. 7(c) indicates a film thickness of approximately 15.5 nm , corresponding to a growth rate of YbN of about $1.9 \text{ \AA}/\text{min}$. Figures 7(d)–7(f) show the XPS spectra of a YbN thin film grown on $\text{LaAlO}_3(100)$ at 400°C in comparison to the one grown on MgO(100) under similar conditions. In general, the Yb $4p$, Yb $4d$, and valence band spectra are similar for both samples grown on $\text{LaAlO}_3(100)$ and MgO(100) substrates. These results suggest that the epitaxial growth of YbN thin films on $\text{LaAlO}_3(100)$ is also possible; however, there is no improvement in crystalline quality compared to the growth on MgO(100). This differs from the growth of SmN or HoN, for which LaAlO_3 proved to be the superior substrate [29,32]. This difference may be due to the smaller lattice parameter of YbN compared to SmN or HoN, resulting in similar lattice mismatches with MgO and LaAlO_3 .

APPENDIX C: TEMPORAL DEVELOPMENT OF THE O $1s$ CORE LEVEL

Although the YbN films were grown and measured by XPS within the UHV system with a base pressure of about $1 \times 10^{-10} \text{ mbar}$, the XPS measurements indicate the presence of a small amount of oxygen for all samples. To investigate the origin of this contamination, we evaluate the time dependence of the XPS measurements. For each film, six rounds of XPS measurements were performed in the same sequence: wide scan, O $1s$, N $1s$ +Yb $4p$, Yb $4d$, and valence band. Each full sequence took approximately 3.5 h to complete. Figure 8 shows the temporal development of the O $1s$ spectra for each iteration of a YbN thin film grown on a MgO(100) substrate under the optimized conditions. The O $1s$ peak was initially negligible but intensified over time. This indicates that the oxygen detected in the YbN films is not related to the growth process. Instead, oxidation occurs gradually during the XPS measurement, despite the ultraclean conditions. A similar oxidation during XPS measurements has been reported previously for HoN epitaxial thin films [32].

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