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RESEARCH ARTICLE

The Evolution of Magnetism in a Thin Film Pyrochlore Ferromagnetic Insulator

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ABSTRACT

The pyrochlore vanadates are compelling candidates for next-generation dissipationless devices. $Lu_2V_2O_7$ and $Y_2V_2O_7$ are ferromagnetic insulators ($T_c \sim 70$ K) that are believed to exhibit the magnon Hall effect and are expected to host topological magnons. Their completely dissipationless magnon edge states could be harnessed to realize low-power information transport in spintronic or magnonic devices. As a crucial step in the realization of devices, we synthesize the first thin films of pyrochlore $Y_2V_2O_7$ on isostructural $Y_2Ti_2O_7$ substrates and explore the evolution of their magnetic properties down to the ultrathin limit. All films are insulating ferromagnets with transition temperatures of up to the bulk value ($T_c \sim 68$ K) that decrease with thickness according to finite-size effects. Our films also exhibit a change in anisotropy from in-plane to out-of-plane easy axis coincident with the development of partial strain relaxation and nonzero magnetic hysteresis in an applied field. This evolution demonstrates the impact of strain on magnetic anisotropy and paves the way to tunable magnon topology.

1 | Introduction

The ferromagnetic insulating pyrochlore vanadates ($A_2V_2O_7$, A = Y, Lu) are promising platforms for low-power spintronic [1–3] or magnonic [3–7] devices. In addition to interest as ferromagnetic insulators ($T_c \sim 70$ K) [1, 3, 8–11], the vanadate pyrochlores are predicted to host topological magnons with fully dissipationless magnon transport in edge states [2, 5–7, 12]. This nontrivial topology of magnons leads $Lu_2V_2O_7$ to exhibit the magnon Hall effect [3, 4, 12–14] and suggests that the pyrochlore vanadates can transport information in the form of edge-confined, topologically-protected spin waves in low-power magnon-based devices.

The vanadates belong to a family of isostructural materials, the pyrochlore oxides, with generic formula $A_2B_2O_6O'$ where both the A and B cations form separate but interpenetrating

Margaret A. Anderson and Megan E. Goh contributed equally to this work.

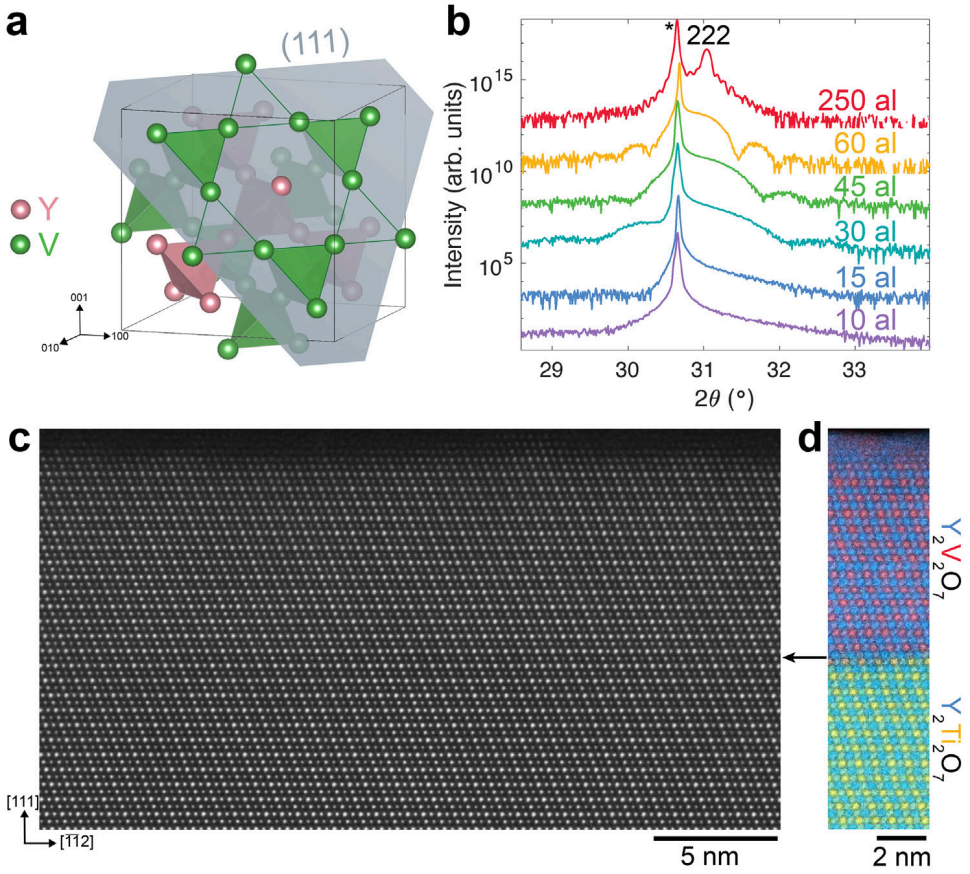

FIGURE 1 | Pyrochlore structure and characterization. (a) The cation structure of $Y_2V_2O_7$ showing the separate interpenetrating pyrochlore sublattices of corner sharing tetrahedra and an isolated 111 vanadium Kagome plane (b) X-ray diffraction about the 222 substrate ($Y_2Ti_2O_7$, denoted by asterisk) and film ($Y_2V_2O_7$) peak for the thickness series (al = atomic layers) showing thickness fringes indicative of smooth, high-quality films (c) HAADF-STEM micrograph of the 30 atomic layer $Y_2V_2O_7$ film along $\langle 1\bar{1}0 \rangle$ showing nearly indistinguishable interface between substrate and film (d) An EELS micrograph of the same film as (c) at approximately the same scale showing a clear interface between film (top) and substrate (bottom).

sublattices of corner sharing tetrahedra (Figure 1a) often called the pyrochlore sublattice [15]. This sublattice, the 3D analog of the Kagome lattice, is geometrically frustrated.

In $A_2V_2O_7$, V^{4+} ($3d^1$, $s = \frac{1}{2}$) sits in a nearly octahedral oxygen coordination environment with a slight trigonal distortion that splits the lower energy T_{2g} states of the octahedral environment into a lowest lying A_{1g} state and higher energy doubly-degenerate E'_g states (Figure S1) [1, 10, 16, 17]. In this configuration, the V^{4+} ions host isotropic Heisenberg exchange and weak but nontrivial Dzyaloshinskii-Moriya (DM) superexchange allowed by Moriya's rules and enabled by spin-orbit coupling [4, 10, 14, 18, 19]. From this relatively simple model on the pyrochlore lattice, myriad unexpected properties emerge. $Y_2V_2O_7$ and isostructural $Lu_2V_2O_7$ are ferromagnetic insulators contrary to the expectation that the unpaired spins leading to a net magnetic moment should produce a metallic state [1, 8–10]. The collinear ferromagnetism is thought to be stabilized by orbital order leading to nontrivial higher-order electron hopping [20] or via thermal order-by-disorder that selects a subset of spin configurations from an O(3) manifold of degenerate states [21]. Although the DM interaction cancels out across nearest-neighbor bonds, preserving the collinear spin arrangement [4, 7, 21], the fictitious magnetic flux caused by ring exchange about inequivalent loops in the pyrochlore structure enables topological magnon edge states and the magnon Hall effect [12–14]. $Lu_2V_2O_7$ powder exhibits a light-induced metal-to-insulator transition [8] while single crystals show colossal magnetoresistance near the ferromagnetic transition temperature. Theoretical studies suggest device applications of pyrochlore vanadates ranging from a field-induced spin transistor [1] to a topological magnon waveguide [6].

Early efforts to synthesize single crystals of $Y_2V_2O_7$ yielded micron-scale crystallites embedded in a matrix of unreacted starting material [11, 22]. Higher-quality floating zone single crystals of $Lu_2V_2O_7$ enabled the observation of orbital ordering [17], ferromagnetic-cluster-mediated colossal magnetoresistance [23], and the first observation of the magnon Hall effect [4]. Recently, macroscopic single crystals of $Y_2V_2O_7$ have been reported as well [3]. While single crystals are ideal for thermal transport and neutron scattering measurements, the thin film geometry is key to realizing practical devices. In addition to enabling device fabrication and integration, thin film synthesis offers a variety of pathways to modify and improve the functionality of materials. Film properties can be tuned with epitaxial strain, modulated doping, and interfacial effects [24, 25]. In thin films of $Y_2V_2O_7$, strain and dimensional confinement could modify the strength of magnetic exchange and tune the ferromagnetic ordering [2] or

magnetic anisotropy, thereby impacting the topological magnon states [6, 7, 12, 26, 27].

Here, we synthesize the first thin films of $Y_2V_2O_7$ and characterize their magnetic properties as a function of thickness, strain, and dimensional confinement. We further take advantage of the Kagome-triangular plane layering in the [111] direction (Figure 1a) of the pyrochlore structure and the atomic precision of molecular beam epitaxy to isolate sub-unit-cell thicknesses and explore the unusual ferromagnetism of $Y_2V_2O_7$ in the ultrathin limit. We find that our high-quality thin films of pyrochlore $Y_2V_2O_7$ exhibit the ferromagnetic insulating state found in bulk crystals down to sub-unit-cell thicknesses. However, the Curie temperature decreases in thinner films following finite-size effects. The thin films exhibit hysteresis under an applied field with a coercivity that increases with thickness. Finally, the $Y_2V_2O_7$ shows a change in anisotropy coincident with the onset of partial relaxation in thicker films and counter to fundamental expectations based on shape and surface anisotropy. Understanding the interplay of dimensionality, strain, and magnetism in these films is critical to the fabrication and realization of magnonic devices and sheds light on the intersection of topology and frustration in thin films writ large.

2 | Results

2.1 | Synthesis and Structural Characterization

Using reactive-oxide molecular beam epitaxy (MBE), we realize the first thin films of pyrochlore $Y_2V_2O_7$ on the isostructural substrate $Y_2Ti_2O_7$(111). YVO_x on yttria-stabilized zirconia (YSZ) substrates forms a phase with pyrochlore-like reflection high energy electron diffraction (RHEED) and x-ray diffraction (XRD) patterns that indicate the expected doubling of the out-of-plane lattice parameter of the film compared to the substrate (Figure S2). However, magnetic susceptibility measurements yield $T_c \approx 140$ K; twice as high as expected from bulk $Y_2V_2O_7$ crystals (Figure S3). Subsequently, high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) imaging reveals a non-pyrochlore phase with alternating 111 planes, but no discernible in-plane order (Figure S4). Based on the film's fast Fourier transform (FFT) and comparison with the diffraction patterns of defect fluorite and pyrochlore crystals, we identify the phase as an anisotropic defect fluorite with pyrochlore-like ordering in the 111 direction. While this phase may be of interest as a highly insulating high-T_c ferromagnetic insulator, it lacks the 111 vanadium kagome planes that lead to nontrivial magnon topology in $Y_2V_2O_7$. Therefore, films on YSZ are not promising platforms for low dissipation magnonic devices.

Moving away from the more common YSZ substrate, we successfully stabilize pyrochlore $Y_2V_2O_7$ films on noncommercial $Y_2Ti_2O_7$ pyrochlore substrates (Figure S5). In situ RHEED indicates smooth, highly crystalline films (Figure S6). Atomic force microscopy confirms that the film surfaces are atomically smooth with RMS roughness on the order of 150 pm (Figure S7). As on YSZ, XRD exhibits the expected pyrochlore peaks with thickness fringes that are a further testament to film quality (Figure 1b; Figure S8). HAADF-STEM imaging shows an exceptionally high-quality film, nearly indistinguishable from the isostructural substrate, with the ideal checkerboard-like cation ordering of a pyrochlore viewed along $\langle 1\bar{1}0 \rangle$ (Figure 1c; Figure S9). The film exhibits minimal cation disorder and no extended defects, such as antiphase boundaries that are common to pyrochlore thin films on YSZ substrates [28]. Atomic resolution electron energy loss spectroscopy (EELS) mapping of elemental concentration further presents a well-ordered pyrochlore film atop a sharp substrate-film interface with at most one monolayer of interdiffusion (Figure 1d; Figure S10). Both EELS and x-ray absorption spectroscopy (XAS) measurements of the V-$L_{2,3}$ edge show the expected peak shape for V^{4+} in a nearly octahedral coordination environment.

Based on the lattice mismatch between $Y_2V_2O_7$ and $Y_2Ti_2O_7$, fully strained films experience approximately 0.9% tensile strain. Reciprocal space mapping (RSM) shows that our $Y_2V_2O_7$ films are fully strained up to at least 45 atomic layers (12.8 nm). Beyond this critical thickness, films show partial relaxation indicated by a diffuse background spread toward the bulk $Y_2V_2O_7$ peak position (Figure S11).

To probe the magnetic properties at the ultrathin limit of $Y_2V_2O_7$, we leverage the atomic layer precision of MBE to synthesize a series of $(Y_2Ti_2O_7)_m/(Y_2V_2O_7)_n$ superlattices where $m, n \in \mathbb{N}$ are the number of atomic planes in each repeat. Superlattices enable the characterization of the magnetic properties of the film in the ultrathin limit by maintaining low dimensionality while increasing the total magnetic signal produced by multiple non-interacting repeats. While these samples allow us to avoid sample volume limits for our magnetic measurements, some prior studies have suggested that interlayer coupling can have an effect on magnetic properties [29, 30]. We thus label each data point in magnetic data that arises from superlattice samples. In each superlattice, we hold $m = 6$ so that each nonmagnetic $Y_2Ti_2O_7$ is a full unit cell thick to ensure negligible interlayer coupling between $Y_2V_2O_7$ layers: the magnetic exchange interaction is known to be vanishingly small beyond the nearest neighbors [18]. We vary $n \in \{2, 6, 10\}$ to compare directly with the thickness series and smoothly probe unit-cell and single tetrahedra thicknesses (Figure S12). We adjust the number of superlattice repeats to maintain 30 total atomic layers of $Y_2V_2O_7$ to ensure a measurable magnetic signal. To confirm our measurements are not dominated by the effects of interdiffusion, we further synthesized a set of fully disordered $Y_2Ti_xV_{2-x}O_7$ samples with the same Ti:V ratios and overall thicknesses as the superlattices.

In situ RHEED shows the superlattices are smooth and highly crystalline, similar to single phase $Y_2V_2O_7$ or $Y_2Ti_2O_7$ films. Due to the similar atomic weight of titanium and vanadium, the superlattice layering is challenging to characterize with diffraction techniques (Figure S13). Only the $(Y_2Ti_2O_7)_6/(Y_2V_2O_7)_6$ superlattice shows a single weak XRD satellite peak about the 222 film peak near the expected superlattice peak position. This peak is absent in the corresponding Y_2TiVO_7 film. The low noise floor of synchrotron XRD more clearly reveals this presumed superlattice peak and confirms the absence of defect phases (Figure S14). STEM imaging shows the superlattice forms the pyrochlore structure with minimal *A-B* site cation disorder (Figure S15), while atomic resolution EELS maps of vanadium and titanium concentration show increasing interdiffusion deeper in the film (Figure S16).

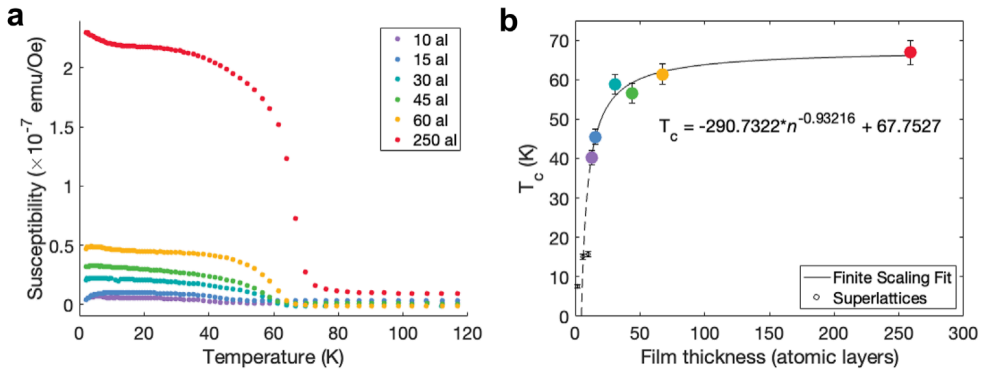

FIGURE 2 | Magnetic susceptibility of $Y_2V_2O_7$ thickness series. (a) Field-cooled magnetic susceptibility measured in a field of 500 Oe perpendicular to ⟨111⟩ for each sample in the thickness series. (b) Ferromagnetic transition temperature versus film thickness (n, atomic layers) with a finite-size scaling fit (solid line, extrapolated along dotted line) that shows excellent agreement with both thickness series (filled circles) and superlattice samples (open circles).

2.2 | Magnetic Properties

All films are ferromagnetic insulators like bulk $Y_2V_2O_7$ and $Lu_2V_2O_7$. $Y_2V_2O_7$ films show a high surface resistance that increases in thinner films. Resistivity versus temperature shows fully insulating behavior (Figure S17). Magnetic susceptibility measurements of the thickest $Y_2V_2O_7$ film in a 500 Oe in-plane applied field reproduces the bulk crystal susceptibility behavior with a sharp ferromagnetic transition at $T_c = 67 \pm 3$ K and a plateau at low temperatures (Figure 2a, red). The field-cooled and zero-field-cooled curves diverge below T_c (Figure S18). Fitting the high temperature inverse susceptibility to the Curie-Weiss law yields $\theta_{CW} \approx 69$ K and $\mu_{eff} \approx 1.83\ \mu_B$ per vanadium atom (Figure S19). For thinner films, the qualitative susceptibility behavior remains the same (Figure 2a; Figure S20) however the ferromagnetic T_c decreases following the finite-size effect:

$$T_c(n) = An^{-B} + C \tag{1}$$

where $B = 1$ for a simple mean-field theory and C is the bulk Curie temperature [31]. The fit of this function to the measured T_c of $Y_2V_2O_7$ films with thicknesses ranging from about 10–250 atomic layers (Figure 2b) yields a scaling factor $B = 0.93$, similar to that expected from mean-field theory, and $C = 67.7$ K, consistent with the bulk Curie temperature $T_{c,bulk} = 68$ K [3, 11]. The superlattice transition temperatures also align well with an extrapolation of the finite-size fit, suggesting that T_c approaches 0 K in the monolayer limit, in contrast to theoretical predictions that the ferromagnetic transition temperature may be enhanced to room temperature or above in few monolayer films of the pyrochlore vanadates (Figure S21) [2].

X-ray magnetic circular dichroism measurements at the V-$L_{2,3}$ edge of a 60 atomic layer $Y_2V_2O_7$ film begin to show a signal near the ferromagnetic transition temperature ($T_c \sim 61$ K) of susceptibility measurements (Figure S22). This confirms that the ferromagnetism arises from the V^{4+} ions, as expected.

As hinted by the splitting of the field-cooled and zero-field-cooled susceptibility below the transition temperature, the magnetization of the thickest (250 atomic layer) film shows hysteresis under an applied field (Figure 3a, red). The narrow coercive field (at most 133 ± 7 Oe at 1.8 K) confirms that thin films of $Y_2V_2O_7$ behave as soft ferromagnets like fully oxidized bulk crystals of both $Lu_2V_2O_7$ and $Y_2V_2O_7$, which lack measurable hysteresis entirely [3, 32]. The hysteresis persists largely unchanged in films of approximately 60 atomic layers (Figure 3a,b). In thinner films, the coercivity shrinks approaching the ultrathin limit (Figure 3c; Figure S26). In magnetic thin films, coercivity tends to increase in the thinnest films as increasing strain induces defects, which act as pinning sites for magnetic domain walls [33]. Here, the growth in coercivity accompanies partial relaxation as film thickness increases past the critical value determined with RSM. This suggests that the 0.9% tensile strain induced by the slightly larger $Y_2Ti_2O_7$ stabilizes the defect-free pyrochlore phase while relaxation leads to the formation of defects. A similar effect occurs in Ni/Cu thin films as the nickel thickness is increased past the critical value, leading to relaxation [34]. In all films, the saturation magnetization is on the order of 1 μ_B per vanadium atom as expected for a collinear d^1 ferromagnet. The small volumes of the films lead to sizable uncertainty in the conversion to μ_B per vanadium atom, which complicates precise interpretation of the saturation value and direct comparison between films. However, the consistent magnitude throughout the thickness series suggests film quality does not degrade significantly down to at least 15 atomic layers. The $n = 10$ superlattice exhibits a larger saturated magnetization than the 10 atomic layer $Y_2V_2O_7$ film, indicating the high quality of the vanadate layers within the superlattice (Figure S27). As n decreases, the magnetic moment decreases until no signal is measured in the $n = 2$ film even at 1.8 K, as expected from the vanishing T_c.

Comparing the magnetization in a field applied within the plane of the film (perpendicular to ⟨111⟩) and out-of-plane (along ⟨111⟩), the magnetically easier axis transitions from out-of-plane in the thickest films to in-plane for films below 45 atomic layers thick (Figure 4a–e). This transition is coincident with both the onset of partial strain relaxation and the development of measurable ferromagnetic hysteresis. Accordingly, in films that exhibit this partial relaxation (60 and 250 atomic layers, Figure 4a,b), the out-of-plane magnetization conforms to the in-plane behavior at low fields. This indicates that the relaxed

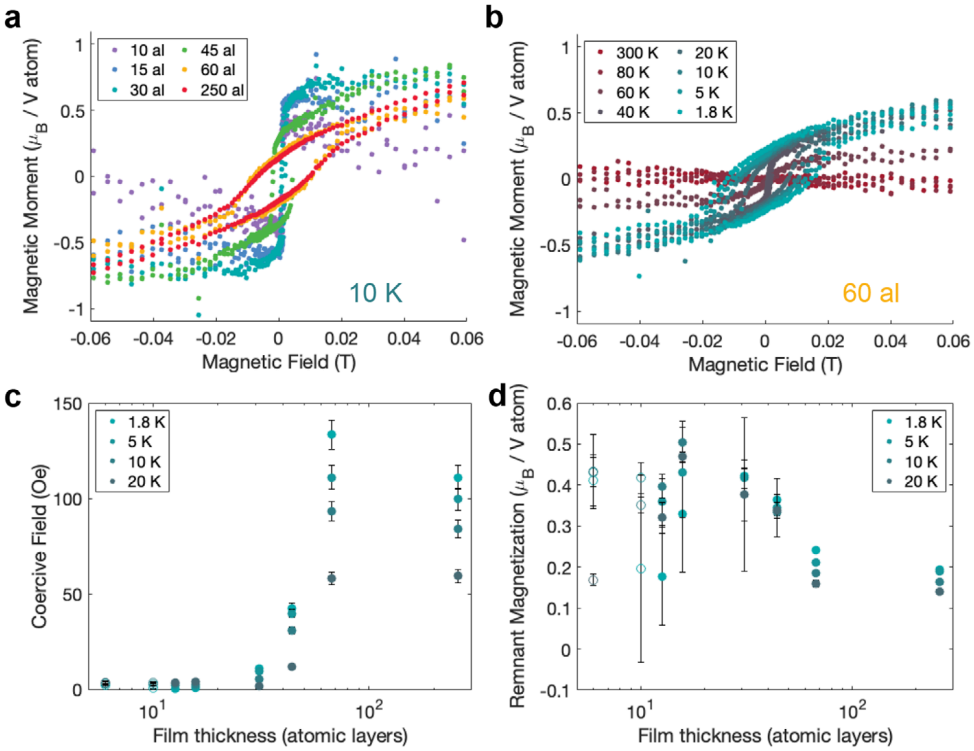

FIGURE 3 | Magnetization versus applied field loops. (a) Thickness dependence of magnetization versus applied field loops measured at 10 K with field perpendicular to ⟨111⟩. (b) Temperature dependence of magnetization versus applied field loops with field perpendicular to ⟨111⟩ for 60 atomic layers of $Y_2V_2O_7$, which has $T_c = 63$ K. (c) Coercive field (width) of magnetization loops versus film thickness and temperature with single-phase films (filled circles) and superlattices (open circles). (d) Remnant magnetization of $Y_2V_2O_7$ thin films versus thickness and temperature.

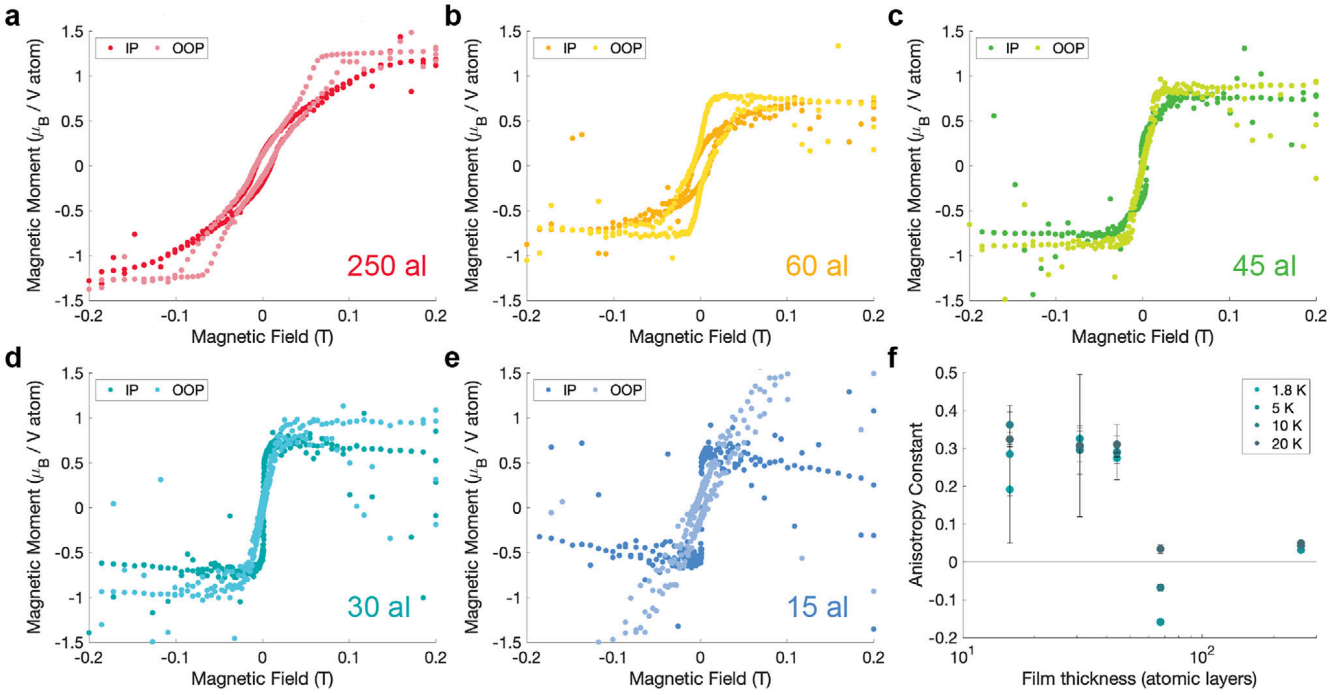

FIGURE 4 | Magnetic anisotropy of $Y_2V_2O_7$ thin films. (a–e) Magnetization versus applied field loops at 10 K for a field applied within the plane of the film (IP, perpendicular to ⟨111⟩) and out-of-plane (OOP, field along ⟨111⟩) for films with thickness (a) 250, (b) 60, (c) 45, (d) 30, and (e) 15 atomic layers (f) The anisotropy constant $\Delta M_r = M_{r,IP} - M_{r,OOP}$, assuming constant 1 μ_B saturation, showing a change in anisotropy between the 45 and 60 atomic layer films.

portion of the film may have distinct anisotropy. This change in anisotropy in partially relaxed films can also be seen in XMCD signal measured at normal and grazing incidences (Figures S23–S25). A change in easy axis represents a change of anisotropy, which can be quantified by an anisotropy constant defined as $\Delta M_r/M_s = (M_{r,IP} - M_{r,OOP})/M_s$. Here, due to the uncertainty in the exact value of the magnetic moment between films, we take M_s to be the bulk value, 1 μ_B/V^{4+} ion. Quantifying the anisotropy in our films, we see a transition from positive (in-plane easy axis) toward negative (out-of-plane) anisotropy between the 45 and 60 atomic layer films (Figure 4f), as expected. Full characterization of magnetic behavior of the $Y_2V_2O_7$ thickness series in an out-of-plane field is presented in Figures S28– S30. The $n = 10$ superlattice shows a strong in-plane easy axis (Figure S31). The anisotropy is less dramatic in the $n = 6$ superlattice, possibly due to the effects of interdiffusion.

3 | Discussion and Conclusion

We explore magnetism in ultrathin films of the pyrochlore ferromagnetic insulator $Y_2V_2O_7$. As an important step toward the development of low-power magnonic devices, we synthesize the first reported vanadate pyrochlore thin films. While films on isostructural $Y_2Ti_2O_7$ substrates readily form the pyrochlore phase, YVO_x deposited on commercial YSZ(111), a popular substrate for thin film pyrochlore synthesis, forms a distinct defect phase, which lacks the kagome vanadium planes that lead to nontrivial magnon topology. $Y_2Ti_2O_7$, in contrast, forms the pyrochlore phase on YSZ(111) and may be used as a buffer layer to seed pyrochlore $Y_2V_2O_7$ films. Based on structural characterization with in situ RHEED, XRD, and HAADF-STEM imaging, our films on $Y_2Ti_2O_7$ substrates are higher quality than most bulk crystal $Y_2V_2O_7$ [11, 22]. The films, especially those that are fully strained, are smooth, highly crystalline, and single phase with very little cation disorder. $(Y_2Ti_2O_7)_6/(Y_2V_2O_7)_n$ superlattices are similarly high quality with respect to strain and pyrochlore structure. However, marked interlayer cation diffusion within these samples may affect their magnetism, and although they display higher ferromagnetic ordering temperatures than their fully disordered counterparts (Figure S21), further superlattice studies are needed to determine whether these samples can fully allow us to probe magnetism down to a two atomic layer ($n = 2$) limit.

The magnetic properties of the thickest film (88 nm, approximately 250 atomic layers) largely reproduce the behavior of bulk crystals. A Curie-Weiss fit of the inverse susceptibility yields a μ_{eff} of 1.83 μ_B per vanadium ion, which is fairly consistent with the theoretical free ion value, 1.73 μ_B, and that found from fitting the inverse susceptibility of $Lu_2V_2O_7$ bulk crystals, 1.75 to 1.89 μ_B per vanadium [8, 11, 23, 35]. The overestimation of this value derives from spin-orbit coupling [35]. Our fit also gives a θ_{CW} of 69 K, consistent with the ferromagnetic transition temperature determined by the maximum rate of change of the susceptibility. Intriguingly, all Curie-Weiss fits to bulk crystal $Lu_2V_2O_7$ data yield a θ_{CW} about 30 K higher than the true Curie temperature and show a deviation from linear Curie-Weiss behavior at 130 – 170 K, which is attributed to the formation of ferromagnetic clusters or magnetic polarons above the true ferromagnetic transition [8, 16, 23, 32, 35]. The $Y_2V_2O_7$ thin film, in contrast, conforms well to linear Curie-Weiss behavior down to the ferromagnetic transition temperature, which suggests the formation of ferromagnetic clusters is absent or suppressed.

As film thickness decreases, the measured Curie temperature falls according to finite-size effects with a scaling factor similar to that expected from mean-field theory. Unfortunately, our results suggest that the T_c of ultrathin pyrochlore vanadates will not approach room temperature as hoped [2]. However, further exploration of the pyrochlore-like defect phase that forms on YSZ (with $T_c \approx 140$ K) may suggest pathways to enhance the ferromagnetic transition temperature in $Y_2V_2O_7$ films. Apart from the decay of T_c with decreasing thickness, our films also exhibit magnetic hysteresis that manifests as the films begin to relax. The partial relaxation of films beyond the critical thickness may be accompanied by the formation of defects, which act as domain wall pinning sites. This behavior, also noted in Ni/Cu thin films [34], is contrary to the typical behavior of thin films that form defects to relieve increasing strain. We also find a consistent saturation magnetization of about 0.75 μ_B per vanadium atom for all films, which suggests that the crystal quality of the films is maintained throughout the thickness series down to at least 15 atomic layers.

Questions about magnetic anisotropy in the pyrochlore vanadates have attracted significant interest because of its potential impact on magnon transport. Disagreement about the strength of Dzyaloshinskii-Moriya (DM) interactions led to the suggestion that V^{4+} in the pyrochlore structure possess significant single-ion anisotropy [10, 16], contrary to the fact that such a term should be trivially proportional to identity in a quantum spin-1/2 system [19]. Neutron inelastic scattering suggests that magnetic exchange is largely isotropic in $Lu_2V_2O_7$ [18], though fits to magnetic susceptibility may indicate a change in anisotropy below 170 K [16] or the ferromagnetic Curie temperature [35]. Higher order anisotropic exchange is alternately determined to be non-negligible [19] or dismissed as perturbatively small [21]. Regardless, anisotropy is critically important to the topological behavior of magnons in the pyrochlore vanadates [7, 12, 20] and may be modified in thin films via strain, doping, or dimensionality [7].

In thin films, we see a marked transition of anisotropy that accompanies the onset of partial relaxation between films of about 45 and 60 atomic layers. In thicker films, the magnetically easier axis lies in the out-of-plane 111 direction. At about 45 atomic layers thick, the easy axis transitions to within the plane of the film, partial relaxation occurs, and hysteresis opens. This change in anisotropy is opposite of that expected based on shape anisotropy in the thin film geometry, where the thinnest films should exhibit an out-of-plane easy axis to reduce demagnetizing effects [36]. Bulk $Lu_2V_2O_7$ hosts a 100 magnetic easy axis [4], while theoretical studies suggest that individual V^{4+} ions have an easy axis along 111 [10, 16, 21]. We see unique behavior that suggests strain may indeed modify magnetic anisotropy and therefore impact magnon topology in thin films of $Y_2V_2O_7$. The tensile strain may distort the nearly octahedral coordination environment about vanadium atoms, leading to a change in inter- and intralayer exchange interactions and thus the overall

magnetic anisotropy [7]. Future explorations of the magnetism of vanadate pyrochlore thin films under strain may open the door to tunable anisotropy and magnon topology and the selection of ideal properties for fully dissipationless magnonic devices.

4 | Methods

The $Y_2Ti_2O_7$ substrate crystal growth was performed at the IKZ Berlin using a conventional RF-heated Czochralski setup equipped with a crucible balance. The crystal was grown by the top-seeded solution growth technique (TSSG) using a flux based on TiO_2 and BaB_2O_4 at an initial melt temperature of about 1600°C. An iridium crucible (about 58 mm inner diameter) embedded in ZrO_2 and Al_2O_3 insulation was used. An actively heated iridium afterheater and a lid with an opening for the seed holder were placed on top of the crucible. The growth of the crystal occurred at a rate of 0.35 mm·h^{-1} using automatic diameter control and a rotation rate of 10 rpm. An Ar/O_2 gas mixture (0.93% O_2) at atmospheric pressure was used as the growth atmosphere. Substrates of various sizes were prepared by CrysTec GmbH (Berlin, Germany) from the grown crystals.

We synthesize pyrochlore $Y_2V_2O_7$ thin films and $(Y_2Ti_2O_7)_m/(Y_2V_2O_7)_n$ superlattices with reactive-oxide molecular beam epitaxy in a Riber C21 dual chamber system. Yttrium, titanium, and vanadium are evaporated from elemental sources with fluxes of approximately 10^{12} $\frac{\text{atoms}}{\text{cm}^2\cdot\text{s}}$ roughly calibrated in situ with a quartz crystal microbalance (Figures S7 and S32). YVO_x deposited on $(111)(ZrO_2)_{0.905}(Y_2O_3)_{0.095}$ (yttria-stabilized zirconia or YSZ) forms a pyrochlore-like defect phase (Figures S2– S4). The true pyrochlore phase forms on noncommercial, isostructural $Y_2Ti_2O_7(111)$ substrates at 800°C in 2×10^6 torr O_2. Film crystallinity and surface morphology were monitored and optimized with in situ reflection high-energy electron diffraction (RHEED).

Based on a monolayer deposition time inferred from RHEED intensity oscillations and ex situ x-ray reflectivity measurements of calibration samples, we synthesize a series of pyrochlore $Y_2V_2O_7$ films simultaneously on YSZ(111) and $Y_2Ti_2O_7(111)$ with thicknesses ranging from 10–260 atomic layers (1.67–43.33 unit cells). We first explore proof-of-concept pyrochlore superlattice growth of high atomic contrast $(Y_2Ti_2O_7)_m/(Tb_2Ti_2O_7)_n$ films on YSZ(111) (Figure S33). We then synthesize a set of superlattices with six atomic layer (1 unit cell) non-magnetic $Y_2Ti_2O_7$ spacer layers separating 2, 6, or 10 atomic layer $Y_2V_2O_7$ with a total of 30 atomic layers of $Y_2V_2O_7$ in each film. As a control, we produce submonolayer-shuttered disordered $Y_2Ti_xV_{2-x}O_7$ films with the same Ti:V ratio and overall thickness as each superlattice.

Using a Malvern Panalytical Empyrean four circle diffractometer (Cu Kα_1 radiation, $\lambda = 1.5406$ Å) equipped with a Ge(220)x2 monochrometer and Pixcel3D detector, we characterized the structural quality and strain state of our films with x-ray reflectivity (XRR), x-ray diffraction (XRD), and reciprocal space mapping (RSM).

With an Asylum MFP-3d Origin+ atomic force microscope (Oxford Instruments) in tapping mode, we measure sample surface roughness and visualize its topography.

We prepare lamellae for scanning transmission electron microscopy with a Helios 660 focused ion beam system. To verify film structure and superlattice layering, we conduct high-angle annular dark field (HAADF) STEM imaging and electron energy loss spectroscopy (EELS) on a ThermoFisher Scientific Themis Z STEM operating at 200 kV with a convergence angle of 18.9 mrad and collection angle range of 64–200 mrad. Low-angle annular dark field imaging (LAADF) with collection angles from 23–128 mrad yield higher substrate-film contrast. For each micrograph, a stack of short exposure scans were cross-correlated and summed to correct for beam instabilities and drift.

We measure the magnetic properties of the films on a Quantum Design Magnetic Property Measurement System 3 at Harvard's Laukien-Purcell Instrumentation Center. For measurements with the magnetic field applied within the plane of the film, samples were mounted on a quartz rod using GE Varnish (Oxford Instruments) within a clear plastic straw. For out of plane fields, the samples were wedged in place in a straw and secured with clear gel capsules. We measured the magnetic background of a $Y_2Ti_2O_7$ substrate annealed in the MBE at growth conditions. Samples were field-cooled in a 2 kOe applied field and susceptibility was measured in a 500 Oe field unless otherwise specified. Susceptibility measurements were corrected for the substrate background and the temperature independent diamagnetism, then normalized by volume to yield units of μ_B/V atom. Magnetization versus applied field curves were corrected with substrate subtraction and volume normalization. Ferromagnetic critical temperatures were defined as the temperature at the highest rate of change in susceptibility.

X-ray absorption spectroscopy (XAS) carried out at beamline 6.3.1 at the Advanced Light Source confirms the expected oxidation state and local symmetry environment of V^{4+} in the $Y_2V_2O_7$ thin films. Further, we conduct x-ray magnetic circular dichroism measurements under ±1.4 T magnetic field to verify the ferromagnetic behavior of the vanadium ions.

Further x-ray magnetic circular dichroism measurements were performed at the I10-1 beamline at the Diamond Light Source. Measurements were done on the V-$L_{2,3}$ edge at 20 K at grazing and normal incidences. For measurements at 1.5 T, we measured with both positive and negative circularly polarized light, as well as at both positive and negative fields. For measurements at 0.02 T, we measured with only a positive field so as to probe the same magnetic state with each measurement. Field dependent data were taken at 514.08 eV with background taken at 510 eV. After background subtraction, field dependent data were normalized to the total of the background and on-peak data.

Synchrotron surface x-ray diffraction measurements were performed at the beamline sector 7-ID-C of the Advanced Photon Source, Argonne National Laboratory, using a six-circle Huber diffractometer in Psi-C geometry. The incident x-ray beam was monochromated to 17.5 keV ($\lambda = 0.70846$ Å) using a Si(111) double-crystal monochromator ($\Delta E/E \approx 1 \times 10^{-4}$), and focused to a 30 μm (vertical) × 50 μm (horizontal) beam spot size using Kirkpatrick–Baez mirrors. The total photon flux at the sample was approximately 3×10^{12} photons/s. Specular (00L) Bragg rod measurements are collected with an Eiger2 X 500K area detector up to $L = 3.9$ reciprocal lattice units (r.l.u.), with incident and

exit angles matched. Raw 2D detector images were corrected for background scattering, geometric factors, and pixel response non-uniformity. Reciprocal lattice vector (L) was converted to total angular deflection (2θ) using $L = (\frac{2c_0}{\lambda})\sin(\frac{2\theta}{2})$ where c_0 is the substrate lattice parameter (10.01 Å for $Y_2Ti_2O_7$) and λ is the x-ray wavelength (0.70846 Å).

Electronic transport measurements were conducted in a Physical Property Measurement System (Quantum Design) with a Dynacool closed-loop helium compression system.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

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

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