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ABSTRACT
Alternate stacking of a highly conducting metallic layer with a magnetic triangular layer found in delafossite PdCrO$_2$ provides an excellent platform for discovering intriguing correlated quantum phenomena. Thin film growth of delafossites may enable not only the tuning of the basic physical properties beyond what bulk materials can exhibit, but also the development of novel hybrid materials by interfacing with dissimilar materials, yet this has proven to be extremely challenging. Here, we report the epitaxial growth of metallic delafossite PdCrO$_2$ films by pulsed laser epitaxy (PLE). The fundamental role of the PLE growth conditions, epitaxial strain, and chemical and structural characteristics of the substrate is investigated by growing under various growth conditions and on various types of substrates. While strain plays a large role in improving the crystallinity, the direct growth of epitaxial PdCrO$_2$ films without impurity phases was not successful. We attribute this difficulty to both the chemical and structural dissimilarities with the substrate and volatile nature of the PdO sublayer, which make nucleation of the right phase difficult. This difficulty was overcome by growing CuCrO$_2$ buffer layers before PdCrO$_2$ films were grown. Unlike PdCrO$_2$, CuCrO$_2$ films were readily grown with a relatively wide growth window. Only a monolayer thick buffer layer was sufficient to grow the correct PdCrO$_2$ phase. This result indicates that the epitaxy of Pd-based delafossites is extremely sensitive to the chemistry and structure of the interface, necessitating near perfect substrate materials. The resulting films are commensurately strained and show an antiferromagnetic transition at 40 K that persists down to as thin as 3.6 nm in thickness. This work provides key insights into advancing the epitaxial growth of the broader class of metallic delafossites for both studying the basic physical properties and developing new spintronic and computing devices.

ABO$_2$ delafossites are a class of materials offering a wide range of fascinating physical properties due to the inherent combination of electronic conduction and magnetism. Uniquely to this class of materials is the layered structure consisting of alternating A and BO$_2$ layers as shown in Fig. 1(a). The natural heterostructuring of the A and BO$_2$ layers makes delafossites highly attractive to study the itinerate behaviors of electrons combined with localized non-collinear magnetism. In addition, the intrinsically long mean free path leading to an extremely high conductivity as well as the two-dimensional (2D) nature of the transport merit this material system for transformative applications. The A layer is composed of closed-packed cations with A = Cu, Pd, Ag, or Pt, whereas the BO$_2$ layer is composed of slightly distorted edge-sharing BO$_6$ octahedra with B = Al, Cr, Fe, Co, or Rh. The delafossites with
A = Cu, Ag are semiconductors and have long been studied as p-type transparent conducting oxides. This characteristic is in contrast to delafossites with A = Pd or Pt, which are highly metallic and exhibit conductivities as high as copper. Furthermore, the triangular in-plane connectivity gives rise to non-collinear antiferromagnetic states for materials, such as CuFeO$_2$, CuCrO$_2$, AgNiO$_2$, and PdCrO$_2$. While several interesting studies were reported from bulk materials, including large magnetoresistance, unconventional anomalous Hall effect, field-induced magnetic transition, and magnetoelectric effect, many interesting
questions, including the strain tuning, proximity effect, and dimensional control of interlayer coupling, are yet to be understood as there have been only a few attempts on the epitaxial growth of metallic delafossites. Among delafossites, PdCrO$_2$ is highly attractive owing to the strong coupling between highly conducting Pd 2D layers and CrO$_2$ non-collinear antiferromagnetic layers and, consequently, exhibits unusual transport properties. However, due to the volatile nature of PdO and deleterious nucleation of impurity phases that are hard to avoid, the successful growth of the Pd-based delafossites remains yet to be achieved.

Here, we have systematically studied the epitaxial synthesis of the delafossite PdCrO$_2$ by pulsed laser epitaxy (PLE) on various substrates. By systematically tuning the growth conditions, mainly including growth temperature $(T)$, oxygen partial pressure $(P_O)$, and laser fluence $(I)$, it was found that the formation of high-quality, phase pure PdCrO$_2$ epitaxial films requires a delicate balance of the growth conditions. Although the growth conditions that favor the growth of PdCrO$_2$ are nominally independent of the substrate, the highest quality PdCrO$_2$ was found to grow on low-lattice-mismatched delafossite buffer layers, which significantly reduced the appearance of impurity phases. The quality of the buffer layer was found to be the limiting factor for the growth of PdCrO$_2$, which indicates that further improvement of the buffer layer or development of a new generation of bulk crystals will ultimately enable synthesis of high-quality PdCrO$_2$-epitaxial thin films.

The pulsed-laser epitaxy of PdCrO$_2$ films used a sintered polycrystalline target. Based on the previous reports on bulk synthesis, the polycrystalline PdCrO$_2$ target was prepared using a combination of Pd, PdCl$_2$, and LiCrO$_2$. These powders were mixed in a stoichiometric ratio and then sintered at 900 °C in a vacuum furnace, which resulted in PdCrO$_2$ mixed with LiCl. The mixture was then ground and washed with distilled water for 30 min to remove the LiCl byproduct. Note that LiCl is highly soluble in water, unlike PdCrO$_2$. The resulting PdCrO$_2$ polycrystalline powder was then dried by heating to 120 °C in air for approximately 12 h. This powder was then pelletized and sintered at 800 °C in atmosphere to form the final target. For the PdCrO$_2$ single crystal, a mixture of the obtained polycrystalline PdCrO$_2$ and NaCl flux were annealed at 900 °C for 24 h and slowly cooled to 800 °C as described elsewhere.

We used single crystals to compare the optical properties with our thin films. For the film growth, the growth conditions were widely varied $(T = 500–800°C, P_O = 10–1000 mTorr$, and $J = 1–2 J/cm^2)$, whereas the repetition rate of the KrF excimer laser $(\lambda = 248 nm)$ was fixed at 10 Hz. After the growth, the samples were cooled to room temperature in $P_O = 100 Torr$. CuCrO$_2$ thin films, used as a buffer layer, were grown directly on Al$_2$O$_3$ at $T = 700°C$ under an oxygen pressure of $P_{O_2} = 10 mTorr$. We note that the epitaxy of CuCrO$_2$ films was quite straightforward and can be readily achieved over a wide growth window. Thus, as the growth of PdCrO$_2$ was difficult, we used CuCrO$_2$ to see if the use of a buffer layer with the same crystal structure would facilitate the nucleation of PdCrO$_2$. The crystal structure was characterized by x-ray diffraction (XRD) using a four-circle high-resolution x-ray diffractometer (X’Pert Pro, PANalytical; Cu K$_{α1}$ radiation), and the thickness of the film $(d)$ was calibrated by x-ray reflectivity (XRR). The surface morphology measurements were taken by atomic force microscopy (Veeco Dimension 3100). High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images were collected using a fifth order aberration-corrected NION UltraSTEM 200 operated at 200 kV using a 30 mrad convergence semi-angle. The transport properties were measured using the van der Pauw configuration with a 14 T Quantum Design PPMS using aluminum wires directly wire-bonded to the PdCrO$_2$ films. Optical properties of PdCrO$_2$ thin films, the PdCrO$_2$ single crystal, and the SrTiO$_3$ substrate were measured using spectroscopic ellipsometry for UV-visible range (M-2000, J.A. Woollam Co.) with photon energies between 0.74 eV and 5.86 eV at an incident angle of 65°. For films and substrates, infrared ellipsometry (IR-VASE, J.A. Woollam) was used for energy below 0.74 eV. The spectra were fitted using a two-layer model (film/substrate) to obtain physically reasonable dielectric functions. For the single crystal, the complex optical conductivity was extracted using the Kramer–Kronig analysis after measuring the near-normal reflectance (Vertex 80v FT-IR spectrometer, Bruker).

PdCrO$_2$ has a rhombohedral structure (space group R-3m) with lattice parameters of $a = b = 2.930 Å$ and $c = 18.087 Å$ ($a = β = 90°, γ = 120°$). The triangular in-plane geometry requires the use of substrates with a triangular (hexagonal) symmetry. This lattice symmetry can be obtained from the same crystallographic structure or (111) oriented cubic perovskites that are available with a wide range of lattice constants. As shown in Fig. 1(b), we chose (111)-SrTiO$_3$, (0001)-Al$_2$O$_3$, (111)-MgO, and (0001)-4H-SiC. We note that there have been reports on successful synthesis of bulk materials, including CuCrO$_2$, CuAlO$_2$, CuFeO$_2$, CuGaO$_2$, PdCrO$_2$, PdRhO$_2$, and PtCoO$_2$. Among them, a few Cu-based delafossites, including CuAlO$_2$ and CuCrO$_2$, were successfully grown as thin films. In Fig. 1(b), we compare lattice parameters of various delafossites and commercially available single crystals as potential buffer layers or substrates. The lattice mismatches $(δ/(a_i − a_s)/a_i × 100$, where $a_i$ is the lattice parameter of the substrate) of PdCrO$_2$ $(a_{PdCrO_2} = 2.930 Å)$ with various substrates are indicated in parentheses. Note that the surface lattice size $(a_s)$ of the (111)-oriented cubic substrates is taken as a triangular unit to interact with the oxygen lattice of the delafossite, as illustrated in Fig. 1(b). Overall, this approach gives a wide range of lattice mismatches, i.e., (111)-SrTiO$_3$ $(δ = −6.1%)$, (0001)-Al$_2$O$_3$ $(δ = −5.9%)$, (111)-MgO $(δ = 1.7%)$, and 4H-SiC $(δ = 4.8%)$. The majority of previous studies of thin-film growth of delafossites have focused on Al$_2$O$_3$ (0001) as a substrate due to its close lattice match and the hexagonal symmetry as well as the chemical similarity of the oxygen terminated surface of Al$_2$O$_3$ to the BO$_2$ terminating surface of the delafossites. Thus, our wide selection of substrates allows us to study the influence of strain in achieving high-quality delafossite films.

To understand the role of the substrate in nucleation of both the delafossite and the impurity phases, we grew PdCrO$_2$ films on the substrates mentioned earlier, as well as on an additional buffer layer, i.e., single-monolayer PLE-grown CuCrO$_2$ $(δ = 1.3%)$ (~0.6 nm in thickness) on (0001) Al$_2$O$_3$. Initial growth parameters were optimized for PdCrO$_2$ films grown on Al$_2$O$_3$, i.e., $P_O = 100 mTorr$, $T = 700°C$, and $J = 1.5 J/cm^2$. XRD 2θ–θ scans for ~30 nm thick PdCrO$_2$ films grown under the same optimum conditions on all of the substrates and buffer layers are shown in Fig. 2(a). As one can find, the strong appearance of the 0006 PdCrO$_2$ peak indicates that the PdCrO$_2$ phase is well-established. The most intense PdCrO$_2$ peaks occur for the substrates and buffer layers with the
closest lattice match, Al$_2$O$_3$ and delafossite CuCrO$_2$ buffer layer. The PdCrO$_2$ peaks are suppressed, in general, as the lattice mismatch increases. Despite a small mismatch, no sign of PdCrO$_2$ film peaks was observed on MgO substrates. The full width at half maximum (FWHM) from the XRD rocking curve $\omega$-scans measured for the 0006 peak is plotted as a function of the lattice mismatch in Fig. 2(b). This result shows similar dependence on the lattice mismatch. For films grown with a low mismatch, the rocking curve width is rather low, $\Delta \omega \lesssim 0.1^\circ$ (Al$_2$O$_3$ and CuCrO$_2$), whereas FWHM values of up to $1^\circ$ are found for 4H-SiC and SrTiO$_3$. We also note that, while the sign is opposite, PdCrO$_2$/SiC ($\delta = 4.8\%$) has a similar size of the lattice mismatch with PdCrO$_2$/Al$_2$O$_3$ ($\delta = 5.9\%$), but the FWHM of PdCrO$_2$ phases show a huge difference. This may be because the tensile strain increases oxygen deficiency due to the reduced formation energy or increased oxygen exchange kinetics.\textsuperscript{6,28}

The role the substrate plays in nucleation can be additionally seen by comparing the intensity of secondary phases. Specifically, in Fig. 2(c), the intensity of the Cr$_2$O$_3$ peak at $39.7^\circ$ is plotted as a function of the lattice mismatch. The films grown on a buffer layer of CuCrO$_2$ and on 4H-SiC showed highly suppressed Cr$_2$O$_3$ impurity phases, whereas films grown on Al$_2$O$_3$, SrTiO$_3$, and MgO showed Cr$_2$O$_3$ peaks with a much larger intensity. This XRD result shows that the nucleation of Cr$_2$O$_3$ does not solely depend on the lattice mismatch, but rather is highly dependent on the crystal structure of the substrate. Specifically, Cr$_2$O$_3$ shares the same crystal structure as that of Al$_2$O$_3$ and, thus, seems very energetically favorable to form epitaxially on Al$_2$O$_3$, explaining the dominant formation of Cr$_2$O$_3$.

This result further contrasts with films grown on both buffer layer and 4H-SiC, where there is only a very weak Cr$_2$O$_3$ peak, likely indicating that the formation of the Cr$_2$O$_3$ phase is unfavorable on these surfaces. In addition to the crystallographic symmetry, we further note that the formation of Cr$_2$O$_3$ impurities might be suppressed by preventing the loss of volatile PdO by using, for instance, Pd-rich targets as demonstrated for PdCoO$_2$\textsuperscript{14}.

In general, for heteroepitaxy, the closer the film and the substrate crystal structures to one another, the higher the film quality thereon. For example, PdCoO$_2$ films grown on Al$_2$O$_3$ have shown epitaxial twins due to multiple ways to crystallographically connect the delafossite structure to the corundum structure.\textsuperscript{16-20} As data in Fig. 2 show, the CuCrO$_2$ delafossite buffer layer gives rise to the highest crystalline quality PdCrO$_2$. In addition to the small lattice mismatch and identical crystal symmetry of R-3m, CuCrO$_2$ is electrically insulating, which readily allows transport measurements of PdCrO$_2$.

Thin film of CuCrO$_2$ was grown using PLE on Al$_2$O$_3$ at $T = 700^\circ$C, $P_{O_2} = 10$ mTorr, and $J = 1.5$ J/cm$^2$. An XRD scan is shown in Fig. 3(a) for CuCrO$_2$ where the delafossite 0003n peaks are the only peaks resolved. Moreover, the rocking curve FWHM of the 0006 peak from a thick CuCrO$_2$ film (10 nm) is $-0.1^\circ$, indicating a higher film quality than those in previous reports.\textsuperscript{24} Using a single monolayer CuCrO$_2$ buffer layer, the growth conditions of PdCrO$_2$ were further mapped systematically. Figure 3(b) shows a full XRD scan and an $\omega$-rocking curve for a PdCrO$_2$ film grown under optimum conditions, which confirms a good epitaxy of PdCrO$_2$.

Figure 3(c) summarizes the results for PdCrO$_2$ films grown at different $T$ and $P_{O_2}$, where the contour plot indicates the rocking curve FWHM values of the 0006 PdCrO$_2$ peak, and the symbols indicate whether the sample is metallic (solid green circles) or insulating (blue stars). The conditions, where there were no PdCrO$_2$ peaks observed, are marked with red crossed circles. From these data, the optimum growth regime is found to reside in $600^\circ$C $\leq T \leq 700^\circ$C and 200 mTorr $\leq P_{O_2} \leq 300$ mTorr. Within this optimal growth window, the rocking curve width is minimal, and the films are metallic, whereas the films grown outside of the optimal window are found to be totally insulating (beyond our instrumental limit) even in the case with good crystallinity. The insulating nature was found to be extrinsic as islands were rather dominantly grown instead of a continuous film. The rocking curve width ($\Delta \omega = 0.1^\circ$) is nominally the same as the underlying CuCrO$_2$, which indicates that the structural quality is largely limited by the underlying buffer layer. Finally, it is important to point out that these growth conditions are nominally identical to those found for PdCrO$_2$ directly grown on Al$_2$O$_3$, which confirms that the formation of the impurities is entirely driven by the character of the surface. In addition, as shown in Fig. 3(d), the XRD reciprocal space mapping confirmed that this film is fully strained. The HAADF-STEM image shown in Fig. 3(e) for PdCrO$_2$ grown on a single-unit-cell thick CuCrO$_2$ buffered Al$_2$O$_3$ substrate further confirms the high crystallinity of the epitaxial thin film. From this image taken along the $\langle 1100 \rangle$ direction, the layered delafossite crystalline structure with alternatively stacked Pd (bright) and Cr (less bright) layers is clearly resolved on the Al$_2$O$_3$ substrate, which is the darker region on the bottom of the image. The atomic structure at the interface is resolvable, for which there are several interesting aspects: First, the nucleation layer on the Al$_2$O$_3$ substrate turns out to be the Cr–O sublayer, followed by stacking of the Pd sublayer,
due to the triangular symmetry of the CrO$_6$ layer. Second, as Cu (atomic number $Z = 29$) is much lighter than Pd ($Z = 46$), the growth of the CuCrO$_2$ buffer layer should be manifested by appearance of the less bright Cu layer than that of Pd on top of Cr–O (note the STEM brightness $\approx Z^2$). However, we found no obvious sign of the Cu layer from our Z-contrast imaging. This result might be due to the fact that the first top Cu layer was replaced or mixed with Pd instead of nucleation of a Cr–O layer on top of the Cu layer. Third, the substrate side of the lattices shows a sign of disorder that might originate from intermixing with the film. Nevertheless, the overall structure seems well-maintained without a hint of severe structural disorder or impurity phases. This can be further understood by more extensive studies utilizing element-specific atomic resolution electron energy loss spectroscopy (EELS), which we leave out as a future study.

Figure 4 shows the thickness dependent transport data for the PdCrO$_2$ films grown on CuCrO$_2$ buffered Al$_2$O$_3$. As shown in Fig. 4(a), our films ($d = 3.6–33$ nm in thickness) show clear metallic behaviors, except for the thinnest ($d = 3.6$ nm) film, of which $\rho(T)$ shows a slight upturn at low temperature, consistent with the onset of a localization transition. Below $3.6$ nm, PdCrO$_2$ thin films were no longer conducting due to the finite thickness, typical of conducting oxides such as SrRuO$_3$. $\rho(T)$ for PdCrO$_2$ films was nearly independent of the thickness above $\sim 4$ nm, but the value was about an order of magnitude higher ($\rho = 100 \mu\Omega$ cm at 300 K) than that of a single crystal ($\rho_{\text{single}} = 8.2 \mu\Omega$ cm). The overall quality of the films is captured by the residual resistivity ratio (RRR, the ratio of the room temperature $\rho$ to the low temperature $\rho$). The maximum RRR value is 2.1 for the 33-nm-thick film. This value is comparable to other PdCoO$_2$ thin films grown by pulsed laser deposition, but

![Figure 3](https://scitation.aip.org/content/aip/journal/apm/8/5/10.1063/1.5144743)

**FIG. 3.** (a) XRD 2θ-θ scans of a CuCrO$_2$ thin film (thickness 10 nm) grown on an Al$_2$O$_3$ (0001) substrate under optimum growth conditions (left). The individual 0003 delafossite peaks are indicated, while the substrate is indicated by an asterisk (*). The corresponding rocking curve for the CuCrO$_2$ 0006 peak is shown on the right. (b) XRD 2θ-θ scans of a PdCrO$_2$ thin film (thickness 13 nm) grown on a CuCrO$_2$ buffer layer (left). The corresponding rocking curve for the PdCrO$_2$ 0006 peak is shown on the right. (c) Contour plot of FWHM of the PdCrO$_2$ 0006 peak as functions of oxygen partial pressures, $P_{O_2}$, and growth temperature, $T$, showing the optimal growth window (top) and oxygen partial pressure dependence of the residual resistivity ratio (RRR) (bottom). A larger RRR value was achieved within the optimal growth window. (d) XRD reciprocal space map of a 23.5-nm-thick film recorded around the Al$_2$O$_3$ 1126 reflection and the PdCrO$_2$ 1018 reflection. (e) A cross-sectional HAADF-STEM image of a PdCrO$_2$ thin film, showing that the Cr–O layer is the first nucleation layer formed on the substrate surface. We note that, while a monolayer-thick CuCrO$_2$ buffer layer was used, we could not find a clear signature of Cu at the interface. We further note that a couple of layers at the interface of the substrate reveal some structural and chemical disorder that might stem from the buffer layer growth. See the text for a more detailed explanation.
This comparison indicates that the film quality may have room for further improvement. $\rho(T)$ also gives insight into the magnetic properties of PdCrO$_2$ films. Bulk PdCrO$_2$ exhibits non-collinear antiferromagnetism with a Néel temperature of $T_N = 37.5$ K,\textsuperscript{22,24} which is reflected in transport measurements as a kink in $\rho(T)$\textsuperscript{15,24} This feature is more clearly seen in $d\rho/dT$ data, which show clear kinks at $T_N = \sim 37$ K. (c) The antiferromagnetic ordering temperature ($T_N$) from PdCrO$_2$ films with different film thicknesses shows a good agreement with the bulk value.

Finally, optical conductivity spectra [$\sigma_i(\omega)$] of PdCrO$_2$ thin films and a single crystal at the room temperature are shown in Figs. 5(a) and 5(b), which were extracted from spectroscopic ellipsometry. Overall, $\sigma_i(\omega)$ of the thin films show a consistent behavior with that of the single crystal. First, all films and single crystal exhibited a low-energy Drude peak below around 1 eV, which reflects the metallic nature. They also showed several interband transitions at 1.3 (referred to as $E_1$), 2.3 ($E_2$), 4.5 ($E_3$), and above 6.0 eV ($E_4$). From electronic structure calculations,\textsuperscript{15} a schematic density of states (DOS) is provided as shown in Fig. 5(c). From the schematic, the $E_1$ peak at 1.3 eV can be attributed to the on-site $d-d$ transition between the hybridized Cr orbital states, and the $E_2$ peak at 2.3 eV to the $d-d$ transition between the hybridized Cr or Pd orbital states. The $E_3$ and $E_4$ peaks at $\sim 4.5$ and above 6 eV are attributed to the charge transfer transition from the O to the Cr or Pd states. By considering the Drude and Lorentz oscillators, we fitted $\sigma_i(\omega)$ as shown in Fig. 5(b). The Drude parameters obtained from the fit were plasma frequency, $\omega_p \approx 28$ 000 cm$^{-1}$, and scattering rate, $\gamma_i \approx 1450 \pm 50$ cm$^{-1}$ (wavenumber). We note that $\omega_p$ of our thin film is in good agreement with that of our PdCrO$_2$ single crystal ($\omega_p \approx 35$ 000 cm$^{-1}$, $\gamma_i \approx 220$ cm$^{-1}$) and a sister compound PdCoO$_2$ single crystal ($\omega_p \approx 33$ 000 cm$^{-1}$, $\gamma_i \approx 97$ cm$^{-1}$),\textsuperscript{24} but the value of $\gamma_i$ is 7–15 times larger than that from the single crystal. The obtained Drude parameters are summarized in Table I. The huge difference of $\gamma_i$ is consistent with the higher resistivity of thin films. The Drude parameters were converted into relaxation time of $\tau \approx 3.65 \times 10^{-15}$ s, carrier density of $n_{10}^{opt} \approx 1.3 \times 10^{22}$ cm$^{-3}$, mobility of $\mu^{opt} = 5.27 \pm 0.5$ cm$^2$/V s, and DC conductivity of $\sigma^{opt} = 9000$ $\Omega^{-1}$ cm$^{-1}$, corresponding to $\rho^{opt} \approx 110$ $\mu\Omega$ cm. The obtained values were consistent with the values from transport measurements. The data were found to be nominally thickness-independent, which indicates that the broad electronic properties are preserved down to the thinnest sample. The Drude parameters, however, show clear thickness dependence as listed in Table I. The scattering rate $\gamma_i$ increases as the film thickness decreases. This finding implies that disorder may play a dominant role in the ultrathin limit. We also note that the optical conductivity, $\sigma_i(0)$, and DC conductivity, $\sigma_{DC}$, show a significant difference for the 3.6 nm-thick sample. We attribute this difference to potential inhomogeneity, e.g., twin boundaries and Cr$_2$O$_3$ impurity phases in the vicinity of the interface.

In summary, we have shown that, albeit on the border of instability, PdCrO$_2$ can be grown epitaxially by pulsed laser epitaxy. Epitaxial strain and the chemical and structural characteristics of the substrate are of key importance to the phase purity of PdCrO$_2$. However, substrates that are at low strain states yet closely match impurity phases strongly enhance the formation of these secondary phases. As such, achieving epitaxial PdCrO$_2$ films required a different approach. We found that the use of a CuCrO$_2$ buffer layer as thin as a single monolayer not only helped reduce the impurity phases but also improved the crystalline quality of the films in comparison to the films grown directly on Al$_2$O$_3$. PdCrO$_2$ epitaxial thin films exhibited a clear magnetic transition down to 3.6 nm. Overall, our results show that overcoming these significant growth challenges for this family of materials is the first step toward a new
generation of complex oxide thin films and creation of novel quantum heterostructures.

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**TABLE I.** The Drude model parameters of PdCoO$_2$ thin films fitted to the optical conductivity and DC conductivity, which are compared to the values from a PdCoO$_2$ single crystal. Literature values for a PdCoO$_2$ single crystal is also shown as a comparison.

| Thickness | $\omega_p$ (cm$^{-1}$) | $\gamma_1$ (cm$^{-1}$) | $\sigma_1(0)$ (Ω$^{-1}$ cm$^{-1}$) | $\sigma_{DC}$ (Ω$^{-1}$ cm$^{-1}$) |
|-----------|----------------|----------------|---------------------------------|----------------|
| 3.6 nm    | 25 000        | 3310           | $4.5 \times 10^3$              | $0.2 \times 10^3$ |
| 5.5 nm    | 28 000        | 2260           | $6 \times 10^3$                | $8.3 \times 10^3$ |
| 9.7 nm    | 28 000        | 1450           | $9 \times 10^3$                | $9.1 \times 10^3$ |
| PdCoO$_2$ single crystal | 220          | $0.8 \times 10^5$ | $1.22 \times 10^5$ |
| PdCoO$_2$ single crystal | 97           | $1.85 \times 10^5$ | $3.84 \times 10^5$ |