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The spin polarization of palladium on magneto-electric Cr_2O_3

Takashi Komesu^{*} , Will Echtenkamp, Christian Binek 
and Peter A Dowben 

Department of Physics and Astronomy and the Nebraska Center for Materials and Nanoscience,
University of Nebraska-Lincoln, Jorgensen Hall 855 North 16th Street, Lincoln, NE 68588-0299, United
States of America

E-mail: tkomesu2@unl.edu

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Abstract

While induced spin polarization of a palladium (Pd) overlayer on antiferromagnetic and magneto-electric $\text{Cr}_2\text{O}_3(0001)$ is possible because of the boundary polarization at the $\text{Cr}_2\text{O}_3(0001)$, in the single domain state, the Pd thin film appears to be ferromagnetic on its own, likely as a result of strain. In the conduction band, we find the experimental evidence of ferromagnetic spin polarized in Pd thin films on a $\text{Cr}_2\text{O}_3(0001)$ single crystal, especially in the thin limit, Pd thickness of around 1–4 nm. Indeed there is significant spin polarization in 10 Å thick Pd films on $\text{Cr}_2\text{O}_3(0001)$ at 310 K, i.e. above the Néel temperature of bulk Cr_2O_3 . While $\text{Cr}_2\text{O}_3(0001)$ has surface moments that tend to align along the surface normal, for Pd on Cr_2O_3 , the spin polarization contains an in-plane component. Strain in the Pd adlayer on $\text{Cr}_2\text{O}_3(0001)$ appears correlated to the spin polarization measured in spin polarized inverse photoemission spectroscopy. Further evidence for magnetization of Pd on Cr_2O_3 is provided by measurement of the exchange bias fields in $\text{Cr}_2\text{O}_3/\text{Pd}(\text{buffer})/[\text{Co}/\text{Pd}]_n$ exchange bias systems. The magnitude of the exchange bias field is, over a wide temperature range, virtually unaffected by the Pd thickness variation between 1 and 2 nm.

Keywords: magneto-electric material, spin polarized electronic structure,
spin polarized inverse photoemission spectroscopy

(Some figures may appear in colour only in the online journal)

1. Introduction

The boundary polarization, especially as the surface/interface, has been regarded as essential to the successful magneto-electric devices fabricated using chromia $\text{Cr}_2\text{O}_3(0001)$ [1–24]. This boundary spin polarization at the surface of $\text{Cr}_2\text{O}_3(0001)$ was predicted independently by Andreev [25] and Belashchenko [26] and has been experimentally verified

by a variety of approaches [27–33] including spin polarized inverse photoemission (SPIPES) [28]. These SPIPES studies [34, 35] have been extended to boron doped chromia that retains a much higher Néel temperature as well as persistent surface polarization, although more canted off the surface normal than undoped chromia. At issue is whether the surface or boundary spin polarization of chromia $\text{Cr}_2\text{O}_3(0001)$ induces spin polarization in the palladium (Pd) adlayer or is the Pd adlayer ferromagnetic.

Induced polarization across a ferromagnetic/paramagnetic interface is known and can be understood in the context of the Landau–Ginzburg equation [36–42], a mean field model without a microscopic Hamiltonian. This approach has been applied to ferromagnetic layers adjacent to Pd [36–39]. While chromia is not ferromagnetic, the boundary spin polarization

* Author to whom any correspondence should be addressed.



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should play a similar role [32]. On the other hand, strained Pd layers have been observed to be ferromagnetic [43–46]. Pd layers on chromia are certainly strained. This dichotomy would have a profound effect on the mechanism for exchange bias in $\text{Cr}_2\text{O}_3/\text{Pd}/[\text{Pd}/\text{Co}]_n$ structure [27, 29, 47, 48].

Pd, as a heavy metal similar to Pt, is often used in Hall-bar structures to read out the magnetic state of an adjacent magnetic layer [27, 49–54]. The read-out mechanism is widely considered to be the spin Hall magnetoresistance effect, which originates from a combination of the spin Hall and inverse spin Hall effects. The spin Hall effect in turn gives rise to a family of spin Hall effect devices [55, 56] and heavy metal Hall bars have been utilized in many spintronic device proposals. The presence of magnetization in the sensor layer, however, affects the Hall signal and has thus important implications for the interpretation of the measured signal. For example, the presence of intrinsic magnetization in the Pd sensing layer will give rise to a contribution to the Hall signal which does not originate from spin Hall magnetoresistance but rather anomalous Hall effect. Therefore, it is important to know and investigate how strain in Pd films can give rise to magnetization.

We have investigated Pd on magneto-electric chromia, $\text{Cr}_2\text{O}_3(0001)$, using spin SPIPES, as well as spin polarized photoemission, but here there is an emphasis on the spin polarization determined by SPIPES, because inverse photoemission is extremely surface sensitive [57, 58]. The experimental evidence points to ferromagnetism in Pd overlayers on $\text{Cr}_2\text{O}_3(0001)$ and that Pd on $\text{Cr}_2\text{O}_3(0001)$ is more than just a paramagnet with an induced polarization arising from the chromia boundary polarization.

2. Experimental

We have investigated Pd adlayers on single crystal of $\text{Cr}_2\text{O}_3(0001)$. The clean $\text{Cr}_2\text{O}_3(0001)$ single crystal was prepared by Ar^+ ions sputtering, with 1.5 keV in 2×10^{-5} Torr of Ar gas pressure, typically for 60 min at a sample emission current of about 6 μA . The $\text{Cr}_2\text{O}_3(0001)$ single crystal was subsequently annealed in oxygen (O_2) gas pressure, 8×10^{-8} Torr for about 20 min, to 1000 K, for several cycles, to obtain an ordered surface with no O_2 vacancies. X-ray photoemission spectroscopy (XPS) was used to confirm a contamination free surface while low energy electron diffraction (LEED) measurements was used to establish the crystallographic order of the single crystal $\text{Cr}_2\text{O}_3(0001)$ and the Pd overlayer. The lattice constant of the Pd adlayer was extracted from the LEED measurements. The reduction of oxygen vacancies contributed to the insulating character of the $\text{Cr}_2\text{O}_3(0001)$ single crystal surface and this in turn leads to an XPS core level shift to higher binding energies, after annealing with O_2 , as expected [59–66]. The Pd adlayers were grown through Pd evaporation, via e-beam bombardment of a crucible, at a deposition rate of about 1 min \AA^{-1} and film thickness determined with a film thickness monitor and confirmed by XPS in the thin film limit.

The SPIPES experiments were carried out with a transversely polarized spin electron gun based on a prior Ciccacci design [67], as described elsewhere [68, 69]. The spin

polarized electron gun used a GaAs single crystal as the spin polarized electron source ‘activated’ (with O_2 and cesium alternative deposition) in a separate chamber, with the initial photocurrent circular generated by circularly polarized laser beam. The spin polarized electron source was designed to be vacuum continuous with the spectrometer chamber equipped with an iodine-based Geiger–Müller isochromat photon detector described some details else where [69]. This SPIPES system is designed so that the incident spin polarization direction is transverse (perpendicular) with respect to the incident electron momentum and parallel (or antiparallel) with the applied field for all sample angles, to maximize our spin-sensitivity. There are 2 different modes for investigation of the boundary spin polarization, with the SPIPES system, whose value depends on the magnetic characteristics of the sample. In determining surface in-plane spin polarization of ‘ferromagnetic materials’ an external pulsed magnetic field, of about 400 Oe, is applied to the sample prior in one direction then the spin polarized inverse spectrum taken for that magnetization direction at remanence. Then the direction of pulsed external magnetic field is reversed and then the spin polarized inverse spectrum taken for the opposite magnetization direction at remanence. In this mode, incident electron spin direction as spin polarized electron is fixed. Because we have used $\text{Cr}_2\text{O}_3(0001)$ single crystal, not a thin film where parasitic magnetization can interact with an applied magnetic field giving rise to a magnetically selected single domain state [70], the 400 Oe pulse field is an insufficient field to reverse the antiferromagnetic spin configuration in the bulk. As a result, the net spin polarization was determined by switching of polarization direction of incident spin polarized electrons with reversal of the circular polarized light helicity used to generate the spin polarized photocurrent. This results in reversal of the outgoing spin polarized electron spin direction. SPIPES is characterized by low count rates, so many sets of data are averaged together for each spectrum shown here in this work, as is typical of SPIPES. The spectra were obtained at remanence with the electron incidence along the surface normal and the energy resolution was approximately 400 meV. Typical of such instruments, the electron gun spin-polarization is 28%. As is typical of SPIPES, all spin-resolved spectra have been renormalized to a 100% hypothetical beam polarization. Instrumental asymmetries in SPIPES have been checked and removed by alternating the magnetic field after each sweep of electron gun energy. The Fermi level was established from tantalum or gold in electrical contact with the sample for all the measurements described herein.

3. Spin polarization at room temperature

Room temperature (294 K) is only slightly below the chromia Néel temperature (307 K), yet the exchange bias field, H_{EB} , in a chromia/Pd/(CoPd) $_n$ multilayer stack persists very close to the chromia Néel temperature, as seen in figure 1. The exchange bias field, H_{EB} , observed after isothermal switching through an applied magnetic field as seen in figure 1, has been seen before [27, 29, 48]. The magnitude of the exchange bias field changes with temperature, but remains nonzero at 294 K,

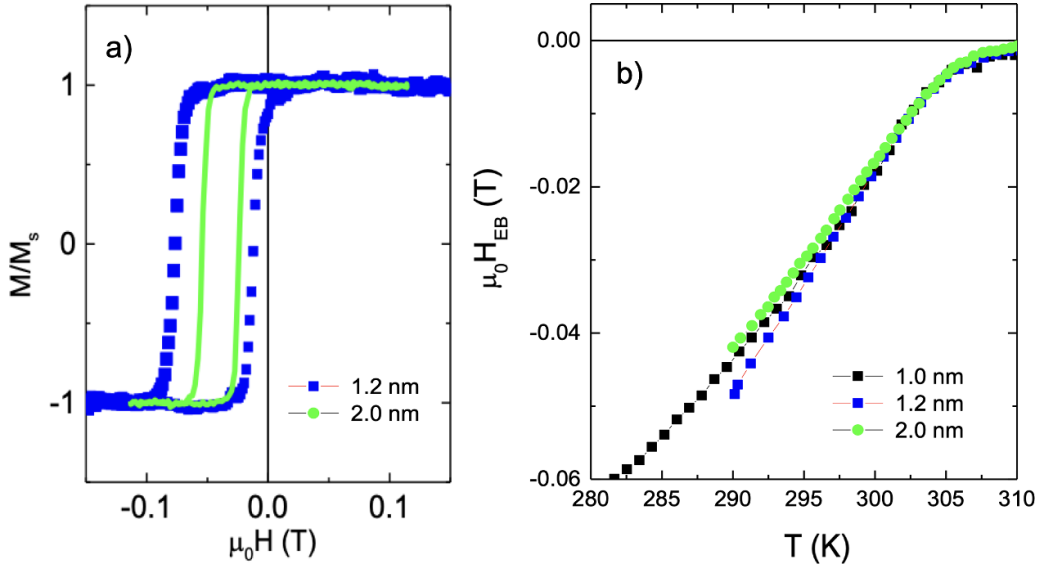


Figure 1. The temperature dependence of exchange bias in various chromia/Pd/(CoPd)_n heterostructures with for Pd buffer layer thicknesses of 1.0, 1.2, and 2.0 nm. Typical exchange bias at 292 K, as a function of applied field for chromia/Pd/(CoPd)_n heterostructures with for Pd buffer layer thicknesses of 1.2 (blue) and 2.0 nm (green) and (a) and the exchange bias plotted as function of temperature for chromia/Pd/(CoPd)_n heterostructures with for Pd buffer layer thicknesses of 1.0 (black), 1.2 (blue), and 2.0 nm (green).

suggestive of a persistent boundary polarization at the interface between Pd and Cr₂O₃(001).

Most remarkably, on increasing the thickness of the Pd buffer layer separating the ferromagnetic Co/Pd multilayer from the antiferromagnetic pinning layer, one would expect an exponential decrease of the exchange bias with increasing buffer layer thickness [39]. Such a dependence or any significant dependence for the three buffer layer thicknesses of 1.0, 1.2 and 2.0 nm is not observed (see figure 1(b)). This indicates that the Pd layer does not behave as a nonmagnetic buffer layer but rather is magnetized and behaves as part of the overall ferromagnetic film.

For the SPIPES studies, the Cr₂O₃ sample was magneto-electrically field cooled in applied *E* and *B* fields through the chromia Néel temperature (307 K), in order to put Cr₂O₃ in a single domain state and enhance the net surface polarization. Figure 2 shows the in-plane SPIPES of the unoccupied density of states for various thicknesses of the Pd overlayers deposited on top of Cr₂O₃. The in-plane SPIPES reveals some Pd spin projection parallel (SPIPES) to the film plane at 294 K temperature. Although the boundary spin polarization of chromia is generally regarded as along the surface normal, the canting this combination of measurements implies is not unusual and has been seen in previous measurement for Co on Cr₂O₃(0001) single crystals [71], while in-plane net polarization has been obtained for Cr₂O₃(0001) single crystals in SPIPES [28], as noted above.

The surface net spin polarization of the unoccupied Pd 4d-bands to 1 eV above the Fermi level retain net spin polarization with increasing Pd film thickness as is evident in the SPIPES spectra of figure 2. Figure 2 shows the SPIPES results as a function of the Pd layer thickness grown on a Cr₂O₃(0001) single crystal. The feature just above the Fermi level that increases with increasing Pd cover is associated with

the unoccupied Pd d-band [72] and has been seen in prior inverse photoemission studies of Pd(111) [72, 73]. Since the spin majority does not align with spin minority, throughout the conduction band region, there is clearly net spin polarization, and the spin asymmetry is band dependent. As the Pd film thickness exceeds the ultra-thin film limit of 5 Å, the unoccupied Pd 4d band has majority spin polarization, as is evident in the SPIPES, as shown in figure 2. Because SPIPES is so surfaces sensitive, we can conclude that the net surface spin polarization of the Pd adlayer generally, and especially the spin asymmetry of the unoccupied Pd 4d-band, appears to decrease with increasing Pd film thickness, greater than 10 Å, as summarized in figure 3.

4. The correlation between strain and in-plane Pd layer surface polarization

The Pd adlayers grown on a Cr₂O₃(0001) single crystal are well ordered. The LEED shows that not only is the clean Cr₂O₃(0001) single crystal well ordered but that there is retention of surface order up to a Pd thickness of 120 Å deposited on a Cr₂O₃(0001) single crystal, as seen in figure 4. The clean Cr₂O₃(0001) single crystal gives the hcp type hexagonal pattern seen before in LEED [74–77], but because of the surface, the symmetry is known to be C_{3v} symmetry, i.e. three fold symmetry, not six fold.

A more subtle aspect of the Pd thin films deposited on a Cr₂O₃(0001) single crystal is that the Pd-Pd distance, as determined from LEED, is larger than the accepted in-plane Pd-Pd 2.751 Å spacing [78–80]. The maximum lattice expansion of Pd on Cr₂O₃(0001), from the analysis of the LEED, is about 14%–15% at a Pd layer thickness of about 1 nm (10 Å) on Cr₂O₃(0001). This expanded lattice constant in the thinnest

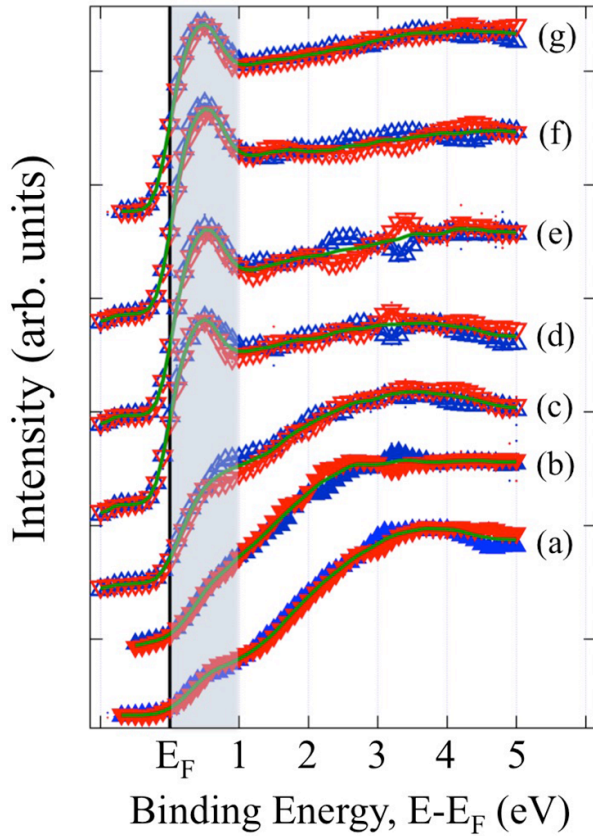


Figure 2. The spin polarized inverse photoemission spectroscopy (SPIPES) results as a function of (a) none, (b) 5 Å, (c) 10 Å, (d) 30 Å, (e) 40 Å, (f) 80 Å, and (g) 120 Å Pd layer thickness grown $\text{Cr}_2\text{O}_3(0001)$ at 294 K. The blue upward triangles are for the spin up majority state and the red downward triangle is for spin down minority state. The green curve indicates the spin average. The unoccupied state binding energies given relative to the Fermi level ($E-E_F$). The spectra shown with filled symbols were taken by reversing the incident electron spin polarization direction, the rest of the spectra taken by reversing the pulse magnetic field.

Pd films is larger than the 2.88 Å Pd–Pd spacing that would put the Pd layer in registry with the Cr_2O_3 surface as has been observed previously for Pd thin films on Cr_2O_3 [80]. At a film thickness of 2 to 4 nm, the measured in-plane Pd–Pd lattice constant does cluster about 2.88 Å, which is consistent with prior work [80], and places the Pd in registry with the Cr_2O_3 surface. As the Pd film thickness increases the LEED indicates that the Pd–Pd spacing decreases with increasing Pd adlayer film thickness on the $\text{Cr}_2\text{O}_3(0001)$ substrate, as summarized in figure 4. Further deposition of Pd on $\text{Cr}_2\text{O}_3(0001)$ leads to a relaxation of the expansive strain to relaxed value of bulk lattice constant of close to 3.89 Å [78, 79], or Pd–Pd distance of 2.75 Å [78–80]. Strain relief is very much expected for metal thin films with increasing film thickness [81].

By plotting the in-plane Pd–Pd lattice constant, determined from LEED, and the net spin polarization determined from SPIPES, as seen in figure 3. It is clear that both the in-plane Pd lattice constant from LEED and the spin polarization extracted from SPIPES follow very similar trends. As the Pd layer thickness on $\text{Cr}_2\text{O}_3(0001)$ increases, the spin polarization that the surface of the Pd film decreases, but so does the lattice

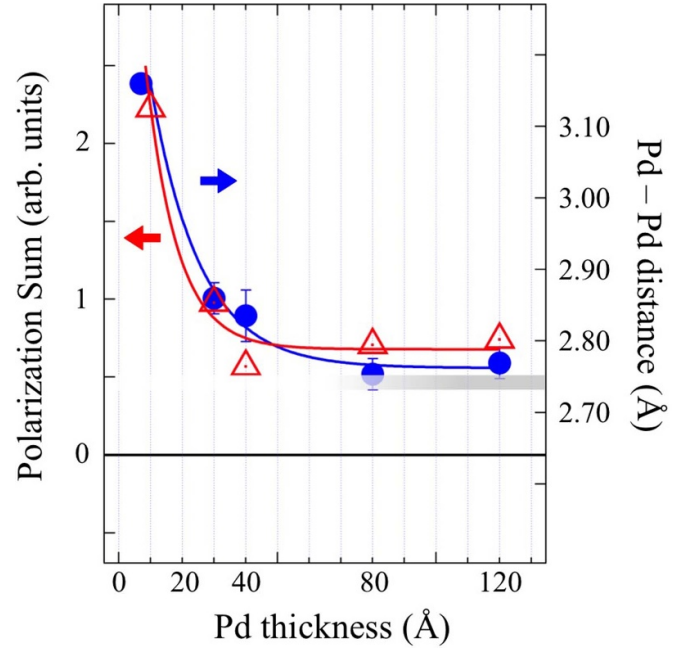


Figure 3. The summed spin polarization (red triangles) between the Fermi level (E_F) and 1 eV above the Fermi level, $E-E_F$, as determined from spin polarized inverse photoemission (figure 4) compared to the Pd–Pd distance (blue circles) as determined from low energy electron diffraction (LEED), shown in figure 6, both for Pd layers $\text{Cr}_2\text{O}_3(0001)$ as a function of the Pd layer thickness. The in-plane Pd–Pd distance of bulk Pd(111) is indicated by the grey stripe.

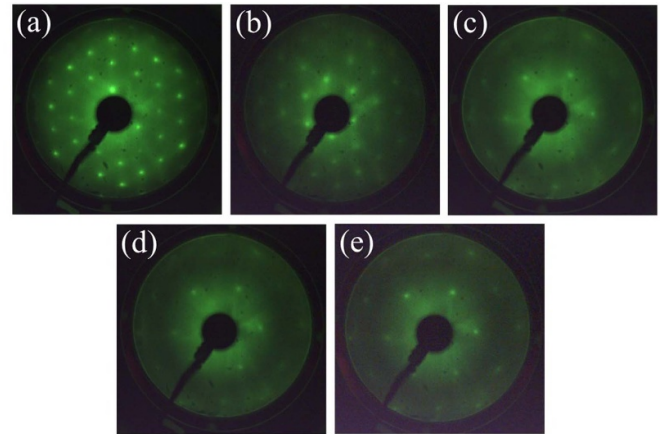


Figure 4. The low energy electron diffraction (LEED) patterns as a function of the Pd layer thickness grown $\text{Cr}_2\text{O}_3(0001)$ at 294 K. The LEED diffraction shown are for Pd layer thicknesses on the clean $\text{Cr}_2\text{O}_3(0001)$ of (a) clean $\text{Cr}_2\text{O}_3(0001)$, (b) for 10 Å, (c) for 40 Å, (d) for 80 Å, (e) for 120 Å respectively. The electron energy was 230 eV for all images.

expansion measure in LEED. This suggests that the measured spin polarization is related to strain. Strain, however, is related to Pd thin film ferromagnetism [43–46, 82–85], as noted in the introduction. The expansive strain that appears to be associated to ferromagnetic Pd in prior work is about 1%–2% [45, 84], while here the expansion of the Pd thin film lattice is 15% for the thinnest Pd films and reducing to 2% at Pd film thicknesses of 80 Å.

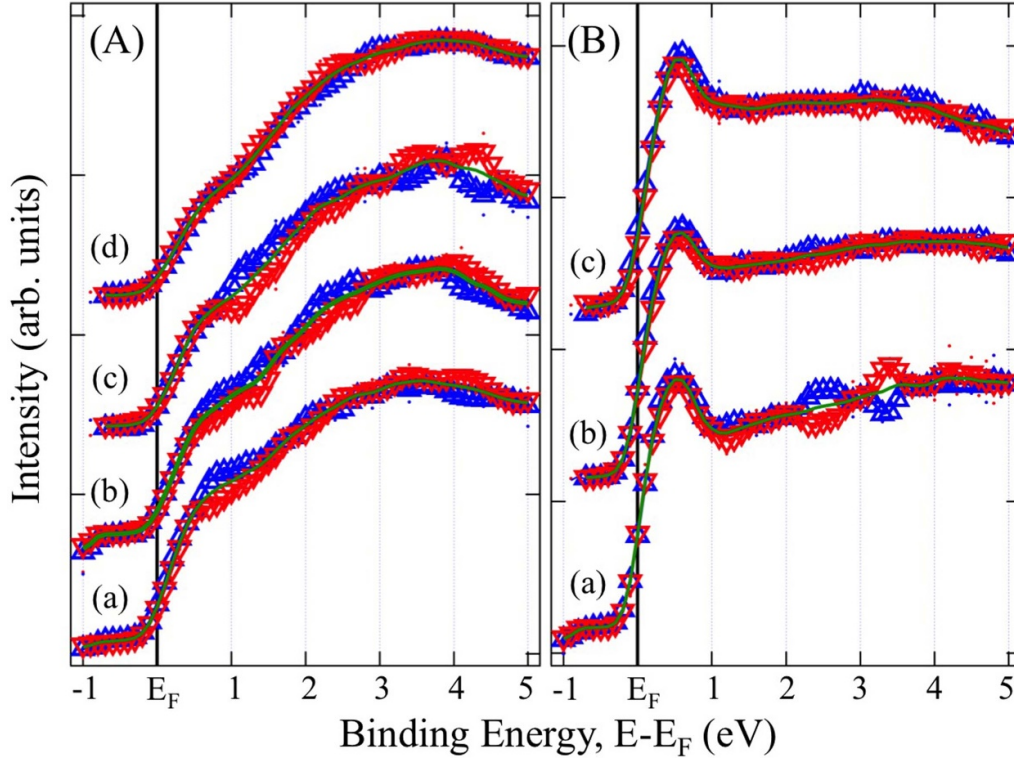


Figure 5. Evidence of net spin polarization of the unoccupied states for 10 Å (A) and 40 Å (B) thick Pd films on Cr₂O₃(0001), from spin polarized inverse photoemission. This temperature dependent SPIPES measurement of Pd thickness of 10 Å (A) and 40 Å (B) on Cr₂O₃(0001) and summarized. For 10 Å thick Pd films on Cr₂O₃(0001) (A) the SPIPES spectra were taken at approximately (a) 294 K, i.e. room temperature, (b) 305 K, (c) 310 K, and (d) 470 K. For 40 Å thick Pd films on Cr₂O₃(0001) (B) the SPIPES spectra were taken at approximately (a) 294 K, i.e. room temperature, (b) 330 K, and (c) 470 K.

5. Ferromagnetic Pd thin films on Cr₂O₃(0001)

The weight of the evidence indicates that Pd layers less than 80 Å to 120 Å thick are ferromagnetic on Cr₂O₃(0001). There are several indicators that all point to a ferromagnetic Pd layer Cr₂O₃(0001).

The Landau–Ginzburg mean field approach in describing the induced polarization in a paramagnet on a ferromagnet would have induced magnetization decay decays as $M(z) = Re^{-kz}$ where z is the distance from the paramagnet/ferromagnet interface and k^{-1} is the paramagnetic correlation length [40–42, 86]. Cr₂O₃ is, of course not a ferromagnet, but an antiferromagnetic, but it is a magneto-electric and the Cr₂O₃(0001) surface exhibits a spin polarized boundary layer in the single domain state [27–33], often referred (incorrectly) as a magnetic surface layer. The boundary between the spin-polarized layer of Cr₂O₃(0001) and a Pd overlayer does exhibit the push-pull of magnetization across the boundary that is reminiscent of the Landau–Ginzburg mean field approach [32], Landau–Ginzburg mean field approach does not apply. While the spin polarization Pd adlayer, measured from SPIPES, does appear to decrease roughly as an exponential with the increasing Pd film thickness on Cr₂O₃(0001), as plotted in figure 3, the spin polarization does not go to zero as would be expected for induced polarization in a paramagnet. But the Pd-Pd in-plane strain, determined from LEED, has not gone to zero at a Pd film thickness of 80 Å on Cr₂O₃(0001).

This persistent spin polarization is consistent with the observation that exchange coupling in chromia/Pd/(CoPd)_n heterostructures, as seen in figure 1, remains little perturbed as the Pd buffer layer thicknesses is increased from 10 Å, to 12 Å and then to 20 Å. Here again, in the simple picture from Landau–Ginzburg, we would expect an exponential decay if the Pd adlayer is paramagnetic, which is not seen.

Because there is a measure net spin polarization at remanence with a rough 400 Oe pulsed magnetic field (figure 2) which is too small an applied field to reverse the domain state of a chromia single crystal [70], this implies that there is a reversal of the remnant polarization of the Pd adlayer.

Perhaps most compelling is that the Pd layer on Cr₂O₃(0001) retains net spin polarization in SPIPES, at temperatures above the Néel temperature of chromia (307 K), as seen in figure 5. Again, since the spin majority does not align with spin minority, throughout the conduction band region, there is clearly net spin polarization, and the spin asymmetry is band dependent. This is clear for the feature just above the Fermi level for the 40 Å thick Pd films on Cr₂O₃(0001) (figure 5(b)) and in the region of 1–3 eV above the Fermi level for 10 Å thick Pd films on Cr₂O₃(0001) (figure 5(a)). For chromia, Cr₂O₃(0001), the spin polarization is lost above the Néel temperature [27, 28]. The general trend is that the net spin polarization decreases with increasing temperature, but significant spin polarization is seen at 310 K for 10 Å thick Pd films on Cr₂O₃(0001), where in fact there should be none.

6. Summary

We have performed spin polarized electronic structure investigations of Pd overlayers grown on Cr₂O₃(0001) single crystal. All-in-all, the data cannot be reconciled with a paramagnetic Pd layer on Cr₂O₃(0001) where the spin polarization is due solely to the induced polarization arising from the uncompensated spins at the Cr₂O₃(0001) surface. We cannot exclude the induced polarization contributes to the net polarization of the ferromagnetic Pd adlayer, but this cannot fully explain the spin polarization of the Pd adlayer above the Neel temperature of chromia nor the exchange coupling effects seen.

The LEED indicates that significant expansive strain occurs in the 10 Å to 40 Å, 1–4 nm, thick Pd(111) layers Cr₂O₃(0001). As in other work [43–46, 70–73], strain is implicated as contributing to the ferromagnetism in the Pd film. We note that here the ferromagnetism is in Pd(111) while in prior work the films are textured along Pd(001).

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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ORCID iDs

Takashi Komesu  <https://orcid.org/0000-0002-2826-4193>

Christian Binek  <https://orcid.org/0000-0002-0026-0772>

Peter A Dowben  <https://orcid.org/0000-0002-2198-4710>

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