Supplementary materials for

unravelling the sign reversal of anomalous Hall effect in ferromagnet/heavy metal ultrathin films

Yao Zhang[1,2], David Cortie[3,4], Thomas LaGrange[5], Waitung Lee[4], Tane Butler[1], Bart Ludbrook[1] and Simon Granville[1,2*]

[1]Robinson Research Institute, Victoria University of Wellington, Wellington 6140, New Zealand.

[2]MacDiarmid Institute for Advanced Materials and Nanotechnology, Wellington 6012, New Zealand.

[3]Institute for Superconducting and Electronic Materials, University of Wollongong, Wollongong, NSW 2500, Australia

[4]The Australian Nuclear Science and Technology Organisation, Lucas Heights, NSW 2232, Australia.

[5]Laboratory of Ultrafast Microscopy and Electron Scattering, École Polytechnique Fédérale de Lausanne EPFL, Lausanne1015, Switzerland.

*Email:simon.granville@vuw.ac.nz

(1) Methods

Polarized neutron reflectometry (PNR): PNR was conducted using the PLATYPUS reflectometer at the Australian Nuclear Science and Technology Organisation (ANSTO). PNR is an ideal tool for studying magnetic thin films, surfaces, and multilayers by measuring the depth profiles of magnetic moment distributions in thin films or multilayers from the surface to the substrate. PLATYPUS is a time-of-flight instrument that uses neutrons with wavelengths ranging from 2.8 to 12.5 Å (when polarized). The spinning mechanical choppers can be adjusted to achieve variable wavelength resolution. In this work, a combination of two choppers (labelled “1” and “3” on the instrument) operating at 33 Hz was chosen, yielding a theoretical wavelength resolution of $d\lambda/\lambda = 3.5\%$. A four-slit system was used to collimate the beam and provide a matching angular resolution. A Fe/Si supermirror with 99.3% polarization was used to polarise the beam. The reflection pattern is plotted as a function of the out-of-plane scattering vector, $Q_z = 4\pi \sin\theta/ \lambda$, where θ is the scattering angle. The reflection of the two neutron spin states (R^{++} and R^{--}) shows a strong dependency on Q. The superscripts “++” and “--” denote whether the neutron spin is parallel or antiparallel to the applied magnetic field for the incident (first superscript) and reflected beams, respectively (second superscript). The PNR data was fitted with the RefNx software, and the thin-film thickness measured from X-ray reflectivity was used to constrain the fitting model. Complementary fitting and Monte Carlo Markov Chain (MCMC) sampling were performed using the RefNX software package to reveal the details of the magnetic interfaces and the associated uncertainties.

(2) Two-contribution model

Two-contribution model is based on the electric current flowing through three separately considered conducting layers (see Fig. S1(a)) in which only Mn_2CoAl and the intermixed CoPd layer can result in an anomalous Hall voltage, as shown in Fig. S1(b).

The measurement is of the anomalous Hall voltage, divided by the total applied current to result in an anomalous Hall resistance as a function of the thickness.

$$R_{AHE}(t) = \frac{V_{AHE}(t)}{I_T},$$

where V_{AHE} is the anomalous Hall voltage, R_{AHE} is the anomalous Hall resistance, and I_T is the total applied electric current.

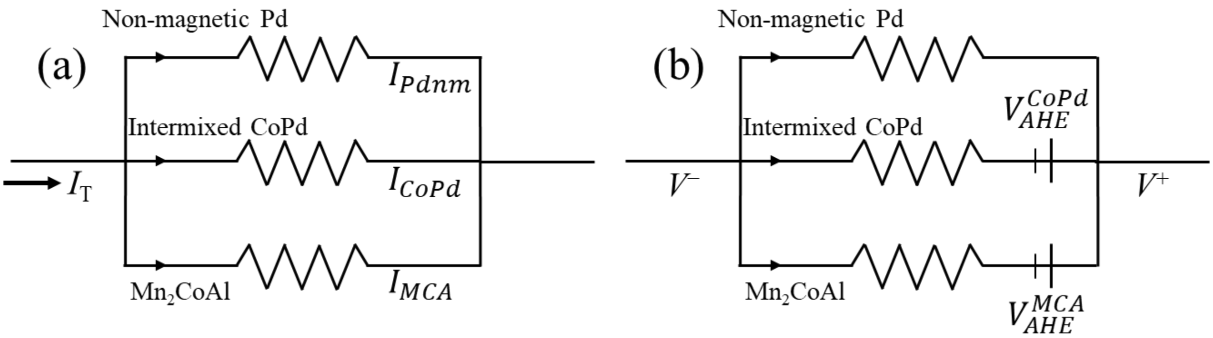

Fig. S1 Schematic circuit diagram (a) electrical current along the Mn_2CoAl, the intermixed CoPd and the non-magnetic Pd layer, (b) anomalous Hall voltage for the Mn_2CoAl and the intermixed CoPd layer.

(i) For region 1, the intermixed CoPd layer on the top:

V_{AHE} is the sum of contributions from the fixed thickness of the Mn_2CoAl layer and the intermixed CoPd layer with variable thickness.

$$V_{AHE}(t) = R_{AHE}^{MCA}\, I_{MCA} + R_{AHE}^{CoPd}(t)\, I_{CoPd},$$

where $R_{AHE}^{MCA}, R_{AHE}^{CoPd}$, I_{MCA} , and I_{CoPd} are the anomalous Hall resistance of Mn_2CoAl, anomalous Hall resistance of CoPd intermixing layer, electric current in the Mn_2CoAl, and electric current in the CoPd intermixing layer, respectively

Divide by I_T to get

$$R_{AHE}(t) = R_{AHE}^{MCA} \frac{I_{MCA}}{I_T} + R_{AHE}^{CoPd}(t) \frac{I_{CoPd}}{I_T} \quad (1)$$

The Mn_2CoAl layer and the intermixed CoPd layer are in parallel, so the longitudinal resistive voltage drop is the same in both of them:

$$V_R^{MCA} = V_R^{CoPd}$$

$$\frac{\rho_{MCA} l}{w t_{MCA}} I_{MCA} = \frac{\rho_{CoPd} l}{w t_{CoPd}} I_{Pd},$$

where $\rho_{MCA}, \rho_{CoPd}, w,$ and l are the resistivity of Mn_2CoAl, resistivity of intermixed CoPd layer, width, and length, respectively

And, using $I_{MCA} + I_{CoPd} = I_T$,

$$\frac{\rho_{MCA} l}{w t_{MCA}} I_{MCA} = \frac{\rho_{CoPd} l}{w t_{CoPd}} (I_T - I_{MCA})$$

so

$$I_{MCA} = I_T \left(\frac{\rho_{CoPd}\, t_{MCA}}{\rho_{MCA}\, t_{CoPd} + \rho_{CoPd}\, t_{MCA}} \right) (2)$$

and taking $I_{CoPd} = I_T - I_{MCA}$, we also have

$$I_{CoPd} = I_T - I_T \frac{\frac{\rho_{CoPd}}{t_{CoPd}}}{\frac{\rho_{MCA}}{t_{MCA}} + \frac{\rho_{CoPd}}{t_{COPd}}} = I_T \left(\frac{\rho_{MCA}\, t_{CoPd}}{\rho_{MCA}\, t_{CoPd} + \rho_{CoPd}\, t_{MCA}} \right) (3)$$

Substitute equation (2) and (3) into equation (1):

$$R_{AHE} = R_{AHE}^{MCA} \left(\frac{\rho_{CoPd}\, t_{MCA}}{\rho_{MCA}\, t_{CoPd} + \rho_{CoPd}\, t_{MCA}} \right) + R_{AHE}^{CoPd} \left(\frac{\rho_{MCA}\, t_{CoPd}}{\rho_{MCA}\, t_{CoPd} + \rho_{CoPd}\, t_{MCA}} \right),$$

(ii) For region 2, with a fraction of the top Pd layer not being mixed with Co:

The current passing through the unmixed Pd layer will contribute nothing to the anomalous Hall voltage. Hence, we have to split the I_{Pd} into I_{Pdnm} and I_{CoPd} parts, where I_{Pdnm} is the current passing through the non-magnetic Pd layer.

Using the same development as above,

$$\frac{\rho_{CoPd}l}{wt_{CoPd}}\ I_{CoPd} = \frac{\rho_{Pd}l}{wt_{Pdnm}}\ I_{Pdnm},$$

where t_{Pdnm} is the non-magnetic Pd layer on the top.

And $I_{Pd} = I_{CoPd} + I_{Pdnm} = I_T\left(\frac{\rho_{MCA}\, t_{Pd}}{\rho_{MCA}\, t_{Pd}+\rho_{Pd}\, t_{MCA}}\right)$ (4), so

$$\frac{\rho_{CoPd}l}{wt_{CoPd}}\ I_{CoPd} = \frac{\rho_{Pd}l}{wt_{Pdnm}}(\,I_{Pd} - I_{CoPd})$$

using $t_{Pd} = t_{CoPd} + t_{Pdnm}$, and replacing I_{Pd} with the earlier expression (4):

$$I_{CoPd} = I_T\left(\frac{\rho_{MCA}\, t_{Pd}}{\rho_{MCA}\, t_{Pd} + \rho_{Pd}\, t_{MCA}}\right)\left(\frac{\rho_{Pd}\, t_{CoPd}}{\rho_{CoPd}[\, t_{Pd} - t_{CoPd}] + \rho_{Pd}\, t_{CoPd}}\right)$$

Thus,

$$R_{AHE} = R_{AHE}^{MCA}\left(\frac{\rho_{CoPd}\, t_{MCA}}{\rho_{MCA}\, t_{CoPd}+\rho_{CoPd}\, t_{MCA}}\right) +$$

$$R_{AHE}^{CoPd}\left(\frac{\rho_{MCA}\, t_{Pd}}{\rho_{MCA}\, t_{Pd}+\rho_{Pd}\, t_{MCA}}\right)\left(\frac{\rho_{Pd}\, t_{CoPd}}{\rho_{CoPd}[\, t_{Pd}- t_{CoPd}]+\rho_{Pd}\, t_{CoPd}}\right) \text{(5).}$$

(3) Fuchs-Sondheimer (FS) model

Here, we employ the FS model to fit the Pd thickness dependence of the resistance of trilayer so that the value of ρ_{MCA} and ρ_{Pd} can be obtained.

The total resistance of the trilayer can be written as

$$R_T = \frac{l}{w}\left(\frac{\rho_{MCA}\, \rho_{Pd}}{\rho_{MCA}\, t_{Pd} + \rho_{Pd} t_{MCA}}\right)$$

By using the well-known FS model $\rho = \rho_0[1 + \frac{3}{8\lambda}(1-p)]$ [1], where ρ_0, λ and p are the bulk resistivity, the ratio of the film thickness to the electron mean free path, and the specularity parameter described the surface scattering [2], respectively, so

$$R_T = \frac{l}{w}\left\{\frac{\rho_{MCA}\, \rho_{Pd}[1+\frac{3}{8\lambda}(1-p)]}{\rho_{MCA} t_{Pd}+\rho_{Pd}[1+\frac{3}{8\lambda}(1-p)] t_{MCA}}\right\}.$$

This equation can give a nice fitting, as shown in Fig. S2. The fitting value p = 0.92, ρ_{MCA} = 219.5 μΩ cm and ρ_0^{Pd} = 11.1 μΩ cm, which are very close to our previous results [3] and other references [4,5].

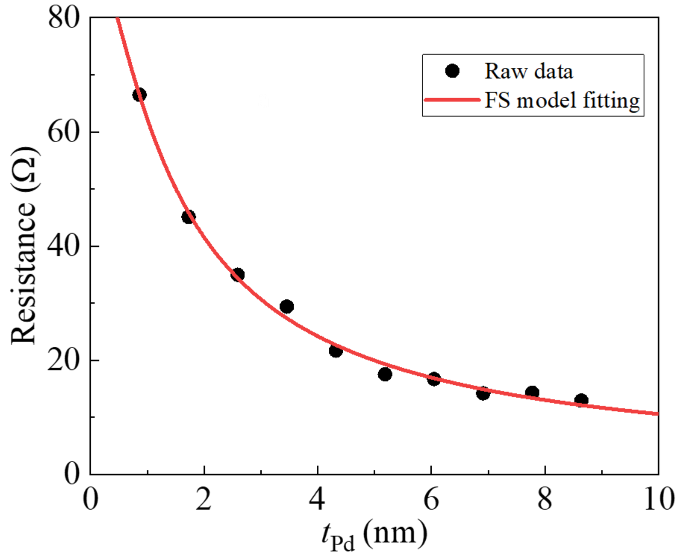

Fig. S2 Resistance as a function of thickness of Pd layer for MgO/Mn_2CoAl/Pd(t_{Pd}) trilayers.

(4) Alternative model based on magnetic Mn_2CoAl and proximity magnetized Pd

We have tried the fitting with contributions from the Mn_2CoAl and a proximity magnetized Pd layer, as shown in Fig. S3. By using the same derivation as shown in the two-contribution model section, R_{AHE} can be written as:

$$R_{AHE} = R_{AHE}^{MCA}\left(\frac{\rho_{Pd}\, t_{MCA}}{\rho_{MCA}\, t_{Pd}+\rho_{Pd}\, t_{MCA}}\right) + R_{AHE}^{Pdm}\left(\frac{\rho_{MCA}\, t_{Pd}}{\rho_{MCA}\, t_{Pd}+\rho_{Pd}\, t_{MCA}}\right)\left(\frac{\rho_{Pd}\, t_{Pdm}}{\rho_{Pdm}[\, t_{Pd}- t_{Pdm}]+\rho_{Pd}\, t_{Pdm}}\right) \quad ,$$

where R_{AHE}^{Pdm} is the anomalous Hall resistance of the magnetized Pd, t_{Pdm} is the thickness of the magnetized Pd. We suppose the resistivity of Pd and magnetized Pd is the same [6]. It is known that the thickness of magnetized Pd is usually less than 1 nm with a very weak magnetization [7,8]. Fig. S3 shows the fitting. One can see that AHE from the magnetic proximity effect is very small and only narrows in 1 nm, which cannot give a good fitting.

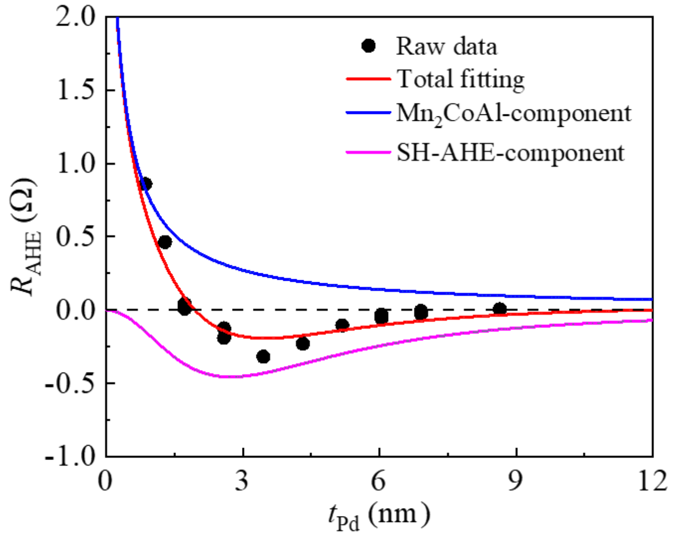

Fig. S3. The experimental R_{AHE} as a function of t_{Pd} with the fitting by a model based on the Mn_2CoAl and the magnetic proximity effect.

(5) Alternative model based on magnetic Mn_2CoAl and spin Hall-AHE

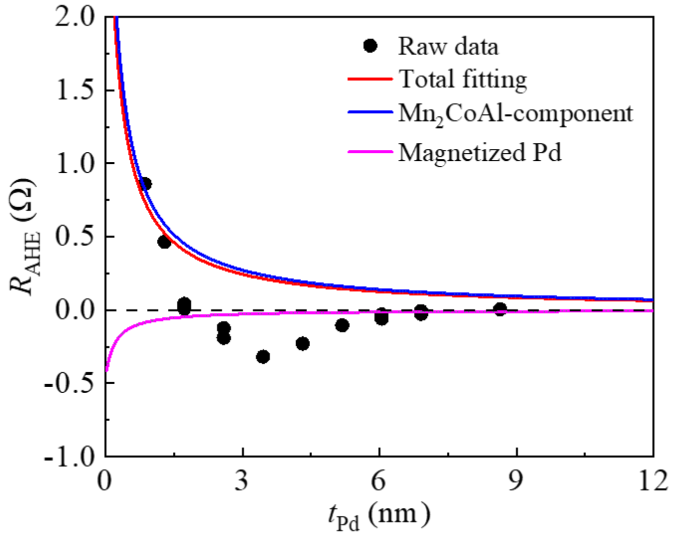

Fig. S4. The experimental R_{AHE} as a function of t_{Pd} with the fitting by a model based on the Mn_2CoAl and SH-AHE.

We also have tried the fitting with contributions from Mn_2CoAl and a spin-Hall AHE term, as shown in Fig. S4. The details have been explained in the Manuscript.

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