

Effect of seed layer thickness on Ta crystalline phase and spin Hall angle

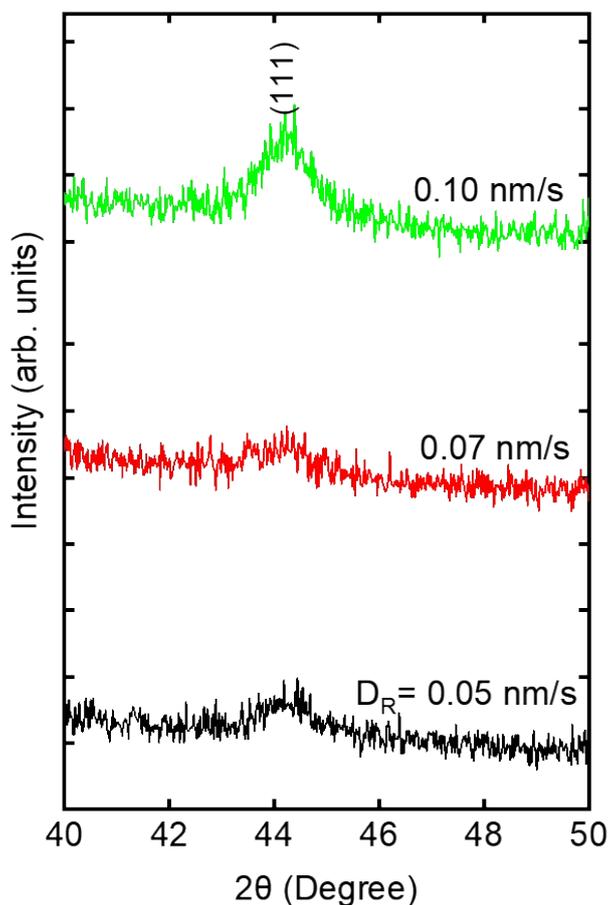
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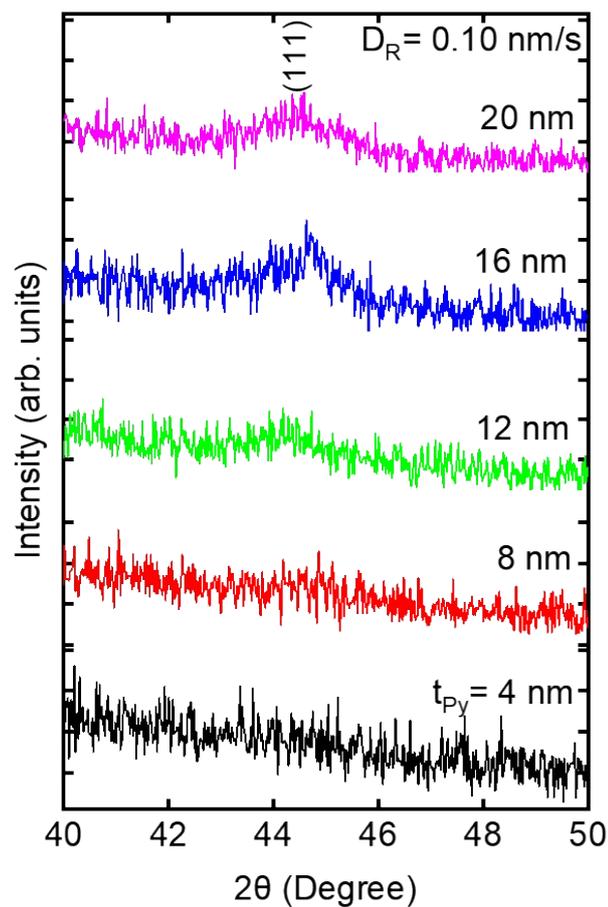
Supplementary information S1



Supplementary figure S1: GIXRD of Py at different deposition rates deposited on Si substrate

The following observations are made from Permalloy (Py) thin films deposited on naturally oxidized Silicon (Si) substrate.

1. We have deposited Permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) by varying deposition power to control the deposition rates by keeping all other sputtering conditions unchanged.
2. The crystalline Permalloy (111) peak is observed from 0.05 nm/s to 0.10 nm/s as shown in figure S1 and there is no prominent XRD peak below 0.05nm/s deposition rates.



Supplementary figure S2. GIXRD of Permalloy (Py) at various thicknesses ($t_{\text{Py}}=4, 8, 12, 16$ and 20 nm) deposited at a deposition rate 0.10 nm/s on Si substrate.

3. We have observed that 0.10 nm/s deposition rate exhibits small inhomogeneous linewidth broadening (ΔH_0) below 10 Oe from ferromagnetic resonance (FMR) measurements. The small inhomogeneous linewidth broadening suggests that the films are homogeneous. The effective magnetization, $4\pi M_{eff}$ is found to be 10000 ± 123 Oe in our Py films deposited at 0.10 nm/s.
4. Based on the optimization, we have deposited Py at 0.10 nm/s deposition rate with various thicknesses $t_{Py=4, 8, 12, 16}$ and 20 nm as shown in figure S2.
5. The above figure S2 shows the GIXRD plots of Permalloy (Py) thickness ($t_{Py=4, 8, 12, 16}$ and 20 nm) deposited at identical sputtering deposition conditions with a deposition rate 0.10 nm/s.
6. Py deposition rate at 0.10 nm/s shows the nucleation of single-phase cubic crystal system with Face-Centered-Cubic (FCC) lattice.
7. The observed 2θ position of FCC Py is 44.2° which is high intensity (111) plane matches with ICDD database.
8. The crystalline peak is visible only from 12 nm Py whereas 4 and 8 nm thick Py have not exhibited any prominent diffraction peaks which may be due to relatively low lateral volume of grains in Py thickness^{1,2}.

Supplementary information S2

In this work, ISHE experiments were strategically performed by selecting samples where the implications of two different phases of Ta can be explored. Therefore, the Py(8)/Ta(18) sample with α -Ta phase and Py(20)/Ta(18) sample with $(\alpha+\beta)$ -Ta phase have been

investigated in detail and the results are presented in the main manuscript. Due to the limited access of the resources, we are not able to estimate the spin Hall angle other than two samples. However, we have now included (Table S1&S2) the phase information, effective damping parameter and spin mixing conductance for all the samples.

Table S1. Effective damping, spin-mixing conductance for α -Ta of Py (4 & 8 nm)/Ta (18nm) bilayer structure.

| Structure | Ta phase | α_{eff} | $g_{\uparrow\downarrow} (\times 10^{18} m^{-2})$ |
|----------------|--------------|----------------|--|
| Py (4)/Ta (18) | α -Ta | 0.0105 | 6.2 |
| Py (8)/Ta (18) | α -Ta | 0.0088 | 7.9 |

Table S2. Effective damping, spin-mixing conductance for $(\alpha+\beta)$ -Ta of Py (12, 16 & 20 nm)/Ta (18nm) bilayer structure.

| Structure | Ta phase | α_{eff} | $g_{\uparrow\downarrow} (\times 10^{18} m^{-2})$ |
|-----------------|----------------------|----------------|--|
| Py (12)/Ta (18) | $(\alpha+\beta)$ -Ta | 0.0078 | 7.9 |
| Py (16)/Ta (18) | $(\alpha+\beta)$ -Ta | 0.0079 | 9.1 |
| Py (20)/Ta (18) | $(\alpha+\beta)$ -Ta | 0.0076 | 10.1 |

In addition, we would like to mention here that, we did investigate a different set of samples in order to confirm our claim on the trend of spin Hall angle being higher in $(\alpha+\beta)$ -Ta phase than α -Ta phase (not shown in the manuscript). In this regard, similar samples Py(8)/Ta(18) sample with α -Ta phase and Py(20)/Ta(18) sample with $(\alpha+\beta)$ -Ta had been obtained by using different deposition rate. Importantly, the results indicate $(\alpha+\beta)$ -Ta displays relatively higher

spin Hall angle compared to α -Ta which is same as the observations reported in the manuscript.

References

- 1 A. Fillon, G. Abadias, A. Michel, C. Jaouen and P. Villechaise, *Phys. Rev. Lett.*, 2010, 104, 2–5.
- 2 A. Fillon, G. Abadias, A. Michel and C. Jaouen, *Thin Solid Films*, 2010, 519, 1655–1661.