Origin of fieldlike spin-orbit torques in heavy metal/ferromagnet/oxide thin film heterostructures

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On the origin of field-like spin-orbit torques in heavy metal/ferromagnet/oxide thin film heterostructures

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We report measurements of the thickness and temperature (*T*) dependencies of current-induced spin-orbit torques, especially the field-like (FL) component, in various heavy metal (HM)/normal metal (NM) spacer/ferromagnet (FM)/Oxide (MgO and HfO_x/MgO) heterostructures. The FL torque in these samples originates from spin current generated by the spin Hall effect (SHE) in the HM. For a FM layer sufficiently thin that a substantial portion of this spin current can reach the FM/Oxide interface, *T*-dependent spin scattering there can yield a strong FL torque that is, in some cases, opposite in sign to that exerted at the NM/FM interface.

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Since the discovery that an in-plane charge current density J_e in certain HM thin films can be utilized to effectively manipulate the magnetization state of an adjacent FM layer [1–6], two classes of spin orbit interactions mechanisms have been considered as candidate sources for these spin-orbit torques (SOT): a strong Rashba-Edelstein (RE) interaction [1,2,7–9] at the ferromagnet interface(s) resulting in the generation of a non-equilibrium conduction election spin polarization at the interface; and the effect of an incident transverse spin current density J_s on the ferromagnet arising from a strong spin Hall effect (SHE) within certain HMs [3–6,10]. Both mechanisms can result in the exertion of damping-like (DL) torque τ_{DL} and a FL torque τ_{FL} on the FM. These can be characterized by the DL(FL) spin torque efficiency ξ_{DL} (ξ_{FL}) or the equivalent effective fields generated per unit J_e, $\Delta H_{DL} / \Delta J_e = \xi_{DL}\hbar / (8\pi e M_s t_{FM})$ and $\Delta H_{FL} / \Delta J_e = \xi_{FL}\hbar / (8\pi e M_s t_{FM})$, where M_s is the saturation magnetization of the FM layer, t_{FM} is its thickness, $\hbar$ is Planck's constant and e is the electron charge [10,11]. The RE effect is generally expected, at least within the context of a Boltzmann equation or drift-diffusion analysis [10], to exert a larger FL than DL torque, while in the SHE case absorption of the transverse polarized component of J_s exerts a larger τ_{DL} and reflection with some spin rotation can result in a smaller τ_{FL}.

Studies of SOT excitation of nanomagnets and domain wall motion in HM/FM heterostructures have generally shown that these processes can be well explained by a DL torque due to the SHE of the HM, with an interfacial Dzyaloshinskii-Moriya interaction also important in the case of domain wall displacement [12–14]. It is therefore quite puzzling that heterostructures made of the same materials can, when the FM is thin and magnetized out of plane, exhibit $\tau_{FL} > \tau_{DL}$ [12,15]. This is true even though experiments in which a NM layer of

variable thickness (with minimal SHE) is inserted between the HM and FM, confirm the nonlocal nature of τ_{FL} in those experiments [16–18]. The origins of τ_{FL} are therefore under active debate -- there are reports showing that the magnitude and even the sign of τ_{FL} can greatly depend on the thickness of the FM [17,19], the type of FM [20], the type of HM [16,21], the direction of the magnetization in the FM [22,23] and temperature [15,23].

Here we report measurements of SOTs in various in-plane magnetized (IPM) and perpendicularly magnetized (PM) HM/NM/FM/Oxide (MgO and HfO_x/MgO) heterostructures, using Ta, Pt, and W for the HM. We have examined τ_{FL} as a function of NM and FM thickness, t_{NM} and t_{FM}, and as a function of temperature *T* from 300 K to 5 K. The spin torque efficiencies, ξ_{DL} and ξ_{FL}, are measured by spin torque ferromagnetic resonance (ST-FMR) for IPM samples [3,24] and ΔH_{DL} and ΔH_{FL} by the harmonic response (HR) method [19,25,26] for PM samples. The t_{NM} dependent measurements reveal that the FL torques observed in all of our samples are due to spin current that originates in the HM via the SHE. By varying t_{FM} for PM cases we find that the FL torque in samples with very thin FM layers can be strong, and differ significantly between FM/MgO and FM/HfO_x/MgO interfaces. Whereas the τ_{DL} is invariably only weakly sensitive to *T,* and similarly for the relatively smaller τ_{FL} present in samples with a thick FM layer, the FL contribution from the FeCoB/Oxide interface is strongly *T* dependent, seemingly going to zero at low *T* in a quasi-linear manner, consistent with previous measurements [15,23].

We will first establish that τ_{FL} in our HM/NM/FM/Oxide heterostructures is caused by J_s that originates in the HM, rather than by a local RE effect at a FM interface. The IPM multilayer samples used in this study (see supplementary material [27] for sample fabrication

and processing details) were W(4)/Hf(t_{Hf})/$Fe_{60}Co_{20}B_{20}$(t_{FeCoB})/MgO(2)/Ta(1), where $t_{Hf} = 0.25 - 2\text{nm}$ and $t_{FeCoB} = 2 - 7$ nm, and the numbers in parentheses represent the nominal thickness in nm. We used ST-FMR to define a FMR spin torque efficiency ξ_{FMR} from the ratio of the symmetric (*S*) and anti-symmetric (*A*) components of anisotropic magnetoresistance response at the ferromagnetic resonance [24,28]. *S* is proportional to τ_{DL} and *A* is due to the sum of the Oersted field torque and any spin-orbit-induced τ_{FL}. The spin-orbit torque efficiencies ξ_{DL} and ξ_{FL} can then be obtained from the dependence of ξ_{FMR} on t_{FM}, assuming that ξ_{DL} and ξ_{FL} do not have a strong dependence on t_{FM} in the range examined [24,28]:

$$\frac{1}{\xi_{FMR}} = \frac{1}{\xi_{DL}}\left(1 + \frac{\hbar}{e}\frac{\xi_{FL}}{4\pi M_s t_{FM} t_{HM}}\right) \qquad (1)$$

Here t_{HM} is the thickness of the HM. Figure 1(a) shows as an example the results for ξ_{FMR} as a function of FeCoB thickness for two different Hf spacer thicknesses. The strong variation with t_{FM} is indicative of a significant τ_{FL}. From fits to Eq. (1) (dashed lines in Fig. 1(a)) we determined ξ_{DL} and ξ_{FL} as a function of t_{Hf}, with the results shown in Fig. 1(b). The signs of both ξ_{DL} and ξ_{FL} are negative, indicating a negative spin Hall ratio for W and a FL effective field that is in opposition to the current-generated Oersted field. As t_{Hf} increases from 0.25 nm, we find that both $|\xi_{DL}|$ and $|\xi_{FL}|$ decrease in concert, and extrapolate to negligible values at large Hf thicknesses[†]. The solid and dashed lines in Fig 1(b) are fits of the same spin current attenuation function to both sets of data [27], which indicate an effective attenuation length

[†] While there are reports that Hf can sometimes generate a quite substantial spin current [29,37], our amorphous Hf layers have a minimal SHE, consistent with a previous experiment [31] .We tentatively attribute this difference to the different phases that Hf thin films can possess, as in the case of W.

$\lambda_s^{Hf} \approx 0.9$ nm. This indicates that both ξ_{DL} and ξ_{FL} are the result of a diffusive spin current from the underlying W that passes through the Hf layer. Similar non-local FL terms have been reported for Py/Cu/Pt and CoFeB/Cu/Pt IPM heterostructures [16,17]. The fit for ξ_{FL} suggests that there is a small residual FL component (-0.010 ± 0.004) when the Hf is in the thick limit, which may be attributable to a comparably weak RE effect at the Hf/FeCoB interface [29]. In contrast to the results that we will discuss for PM samples with much thinner FM layers, for the IPM samples neither ξ_{DL} nor ξ_{FL} has a significant T dependence [27].

The FL torque can be considerably stronger in PM samples, which have by necessity much thinner FM layers. We will show that the degree of this enhancement is strongly dependent on both t_{FM} and the details of the FM/Oxide interface. To establish the first dependency we fabricated two series of PM samples: Ta(4)/FeCoB(t_{FeCoB})/MgO and Ta(6)/FeCoB(t_{FeCoB})/Hf(t_{Hf}) (oxidized)/MgO. In the latter case, $0 < t_{\mathrm{Hf}} \leq 0.4$ nm, and the Hf was oxidized to form a thin HfO_x layer during the subsequent sputter deposition of the MgO rather than the usual case of oxidation of the FM surface [30], as confirmed by the interfacial anisotropy energy, $K_s \approx 2$ erg/cm^2, that was obtained without any post-fabrication annealing [27]. Figure 2(a) shows the effective fields as measured by HR as a function of t_{FeCoB}^{eff} for $t_{\mathrm{Hf}} = 0$ and 0.2 nm. The stated value of t_{FeCoB}^{eff} for the two sample has been corrected to account for a very thin (0.1 ± 0.1 nm) magnetic dead layer as measured by vibrating sample magnetometry (VSM) (see supplementary material [27]). (Note also that all HR results reported in this paper have been corrected for the transverse (planar Hall) magnetoresistance contribution to the HR measurement [26,27], and that small Oersted field contributions have been subtracted

from the $\Delta H_{FL} / \Delta J_e$ results.) For both the FM/MgO and FM/HfO_x/MgO cases we find that $\Delta H_{FL} / \Delta J_e$ decreases rapidly as t_{FM} increases from 0.7 nm to 1.0 nm. For a given value of t_{FM}, the strength of τ_{FL} is very different -- stronger for the FM/HfO_x/MgO samples by approximately a factor of 2 compared to FM/MgO.

Even though $\Delta H_{FL} / \Delta J_e$ depends strongly on both t_{FM} and the composition of the FM/Oxide interface for the PM samples, our previous studies as a function of the thickness of a Hf spacer between the HM and the FM in PM samples show that the origin of this enhanced τ_{FL} is still a spin current emitted from the HM, and the spin-orbit torque decay as a function of increasing Hf spacer thickness and extrapolate to negligible values at large t_{Hf} [31]. Therefore we conclude that the enhanced FL torque in the PM samples must be due to the portion of J_s from the HM that can pass through the FM layer and reach the FM/Oxide interface before dephasing and/or relaxing [32], so that spin scattering at this interface is able to affect the amount of spin accumulation in the very thin FM layer.

As noted above, for a given t_{FM}, $\Delta H_{FL} / \Delta J_e$ for FeCoB/HfO_x is approximately twice that for FeCoB/MgO. The importance of the FM/Oxide interface in the enhancement of τ_{FL} in PM structures is also illustrated by measurements of $\Delta H_{DL} / \Delta J_e$ and $\Delta H_{FL} / \Delta J_e$ as a function of the thickness of an oxidized Hf passivation layer for a series of Ta(6)/FeCoB(0.8)/Hf(t_{Hf})/MgO. As t_{Hf} increases from approximately one atomic layer (0.2 nm before oxidation) to two (0.4 nm) there is only a small change in ΔH_{DL}, while ΔH_{FL} decreases markedly, by nearly a factor of two (Fig. 2(b)). The strong variation of the FL term with t_{Hf} is presumably related to the more complete passivation of the FM surface by a slightly thicker Hf layer. Our finding that the

perpendicular anisotropy field H_a increases as t_{Hf} becomes thicker, as shown in the inset to Fig. 2(c), supports this attribution. In Fig. 2(c) we plot $\Delta H_{FL} / \Delta J_e$ as a function of $1 / H_a$, which indicates that as the strength of the interfacial anisotropy increases, the room-temperature value of τ_{FL} decreases linearly. This behavior is repeated with Ta/FeCoB/MgO samples where we found that the room-temperature values of $\Delta H_{FL} / \Delta J_e$ for that system also vary as $1/H_a$ when different annealing temperatures were employed to modify the PMA [27]. This suggests a tradeoff between the spin relaxation mechanism responsible for τ_{FL} at room temperature and the SOI and crystal field interactions that create the PMA at FCB/Oxide interfaces [33].

While there is only a weak *T* dependence for ξ_{DL} and ξ_{FL} in the IPM samples, for the thin PM samples, as we will show, the FL torque contribution from the FM/Oxide interface is strongly temperature dependent, weakening dramatically at low *T*. We tentatively ascribe this behavior to *T*-dependent spin-flip scattering at the FM/Oxide interface. When the spin scattering is suppressed near $T = 0$, the enhancement of FL torque generated by the FM/Oxide interface is suppressed as well, even though the DL torque is largely unaffected.

We performed *T*-dependent HR measurements of $\Delta H_{DL} / \Delta J_e$ and $\Delta H_{FL} / \Delta J_e$ on a number of different PM samples. Fig. 3(a) and 3(b) show results obtained respectively from an annealed Ta(4)/FeCoB(0.8)/MgO sample and an un-annealed Ta(6)/FeCoB(0.8)/Hf(0.2) (oxidized)/MgO sample, that is without and with the Hf passivation layer. While $\Delta H_{DL} / \Delta J_e$ is nearly invariant with *T,* there is a strong variation in $\Delta H_{FL} / \Delta J_e$ in both cases as *T* goes towards zero. The behavior of $\Delta H_{FL} / \Delta J_e$ for the sample without the Hf passivation is quite similar to that reported previously [15,23]. Below 250 K, ΔH_{FL} decreases quasi-linearly with decreasing *T,*

approaching zero around 70 K, and then departs from linearity to vary more slowly, becoming slightly negative as *T* goes to zero. Here we use the convention that a negative ΔH_{DL} corresponds to a negative spin Hall angle. The degree to which ΔH_{FL} depends on t_{FeCoB} also shows a strong variation with temperature. While ΔH_{FL} at 300 K decreased by over a factor of five when we increased t_{FeCoB} from 0.7 to 1.0 nm in the Ta samples without Hf (Fig. 2 (a)), as *T* approaches 5 K this dependence on t_{FeCoB} is largely absent, with all of the ΔH_{FL} results converging to a very similar small, negative, value (see [27] plot S7). Over this range of temperature the *T* dependence of the magnetization itself is negligible, varying by less than 20% (see section S8 in [27]).

The FL torque in the Hf passivated sample (Fig. 3(b)) also shows a strong *T* dependence that is even more linear for *T* below 200 K to at least 50 K. For additional insight we also studied PM samples with different HM base layers and with a thin Hf spacer layer between the HM and the FM: Ta,Pt,W(4)/Hf(t_{Hf})/$Fe_{60}Co_{20}B_{20}$(t_{FeCoB})/MgO(2)/Ta(1), where $t_{Hf} = 0.5$ or 1 nm and $t_{FeCoB} = 0.7 - 1$ nm. As mentioned previously we initially employed such Hf spacer layers to enhance the PMA in the HM/Hf/FeCoB/MgO heterostructures and to study the role of the spin current from the HM in determining the strength of the SOT [31]. Subsequently, analytical electron microscopy studies [34] have shown that there is diffusion of Hf to the bottom of the MgO overlayer, both before and more so after annealing, resulting in the formation of a thin HfO_x layer at the top of the FM. We conclude that this FM/HfO_x interface is responsible for the linear *T* dependence of the FL torque in all these samples at low *T*.

Figure 3(c) shows $\Delta H_{DL}(T)$ and $\Delta H_{FL}(T)$ as generated in a Ta and a Pt based sample with the Hf spacer and Fig. 3(d) shows results from a W sample ($t_{Hf} = 1$ nm for the Ta and W samples and $t_{Hf} = 0.5$ nm for the Pt sample). ΔH_{DL} in all three cases is nearly constant, increasing just slightly with decreasing *T*. The different signs for ΔH_{DL} correlate with the different signs of the spin Hall ratio θ_{SH} (negative for Ta and W, positive for Pt). The sign of $\Delta H_{FL}(T)$ at *T* = 300 K is in all cases opposite to that of θ_{SH}, being + for Ta and W, and – for Pt. In all cases $\Delta H_{FL}(T)$ also decreases quite linearly with decreasing *T* down to ~ 5 K and in the process exhibits a sign change at low *T* for Ta and W, one that is most strongly seen in the W sample, which has the highest spin Hall ratio. This provides the strongest incident spin current at the Hf/FM interface and results in the largest field-like torque being exerted there, in comparison to the Ta/Hf/FM and Pt/Hf/FM samples. $\Delta H_{FL}(T)$ at the lowest temperature for the W sample corresponds to a FL spin torque efficiency $\xi_{FL} \approx -0.03$, which is consistent with the IPM ST-FMR measurement for the same structure with the same Hf thickness (Fig.1 (b)).

To further confirm that the $\Delta H_{FL}(T)$ behavior in these latter samples cannot be the result of a strong RE generated effective field at the Hf/FeCoB (or FeCoB/HfO_x) interface, we measured $\Delta H_{DL}(T)$ and $\Delta H_{FL}(T)$ in a control sample having only a 4 nm Hf base layer, as also shown in Fig. 3(d). $\Delta H_{DL}(T)/\Delta J_e$ is negligible over the full *T* range, while $\left|\Delta H_{FL}(T)/\Delta J_e\right|$ is quite small, $\leq 1\times10^{-6}$ Oe/(A/cm^2) with little *T* variation.

We conclude that the dominant mechanism in generating ΔH_{FL} is the scattering of the incident SHE-generated J_s at each of the two interfaces of the FM. For the spin torque that is

exerted only at the NM/FM interface (*e.g.*, in the IPM samples) the result is a comparatively small field-like torque, $\Delta H_{FL} < \Delta H_{DL}$, and is at most weakly T dependent. When in thin PM samples a significant J_s reaches the FeCoB/Oxide interface the result is a stronger contribution to τ_{FL} at $T \sim 300$ K with a sign opposite to θ_{SH} and with $\Delta H_{FL} \geq \Delta H_{DL}$, but this contribution decreases apparently close to zero at low T, leaving only the weaker spin current dependent contribution to ΔH_{FL} from the HM/FM interface.

The spin scattering at the HM/FM interface can perhaps be treated via the scattering-matrix spin mixing conductance scenario where spin rotation during the reflection of part of J_s will result in a field-like torque. Initial calculations indicated that this effect should be weak, but until recently such calculations assumed no strong SOI at the interface, which is generally not the case in the HM/FM thin film systems of interest here [35,36]. Typically we find experimentally that $\Delta H_{FL} \sim 0.2 - 0.3\ \Delta H_{DL}$ [24]. At the FM/Oxide interface, the strength of H_a is inversely correlated with the strength of the room-temperature field-like torque (see Fig. 2(c) and [27], Fig. S6), which indicates that there may be competition between the electronic states at the FM/Oxide interface that generate the interfacial anisotropy and those that provide the spin relaxation pathway.

In summary, we have measured the thickness and T dependence of the DL and FL SOTs in a range of HM/NM/FM/Oxide heterostructures. The strength of the FL torque varies in a manner principally dependent upon: (1) a nonlocal J_s generated by the SHE in the HM, and (2) scattering of this J_s at the FM interfaces on which it impinges, rather than dependent on a large local RE effect in which a spin polarization is generated by the J_c flowing at a FM interface. We infer that spin scattering at either the HM/FM or FM/Oxide interface can promote formation of a

net spin accumulation in the FM layer that generates an effective field ΔH_{FL}. The FM/Oxide interface can result in the stronger ΔH_{FL}, provided that a significant portion of the spin current incident on the FM reaches that interface prior to dephasing, which requires a very thin FM layer, and provided that there is a high density of interfacial states that act as strong spin scattering centers. This latter appears to vary inversely with the interfacial anisotropy energy density in the FeCoB/Oxide interfaces studied here.

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Fig 1

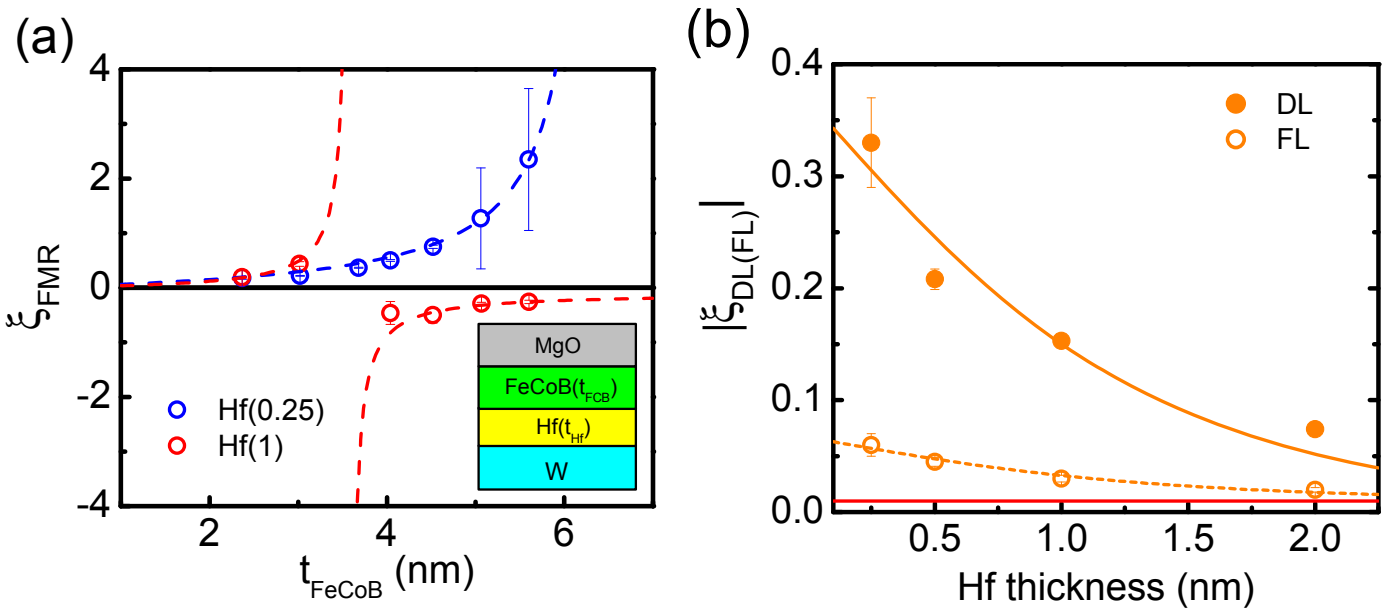

Fig. 1.(a) ST-FMR spin torque efficiency ξ_{FMR} as a function of the FeCoB thicknesses t_{FCB} for two in-plane magnetized samples W(4)/Hf(0.25,1)/FeCoB/MgO. (b) Damping-like and field-like spin torque efficiencies ξ_{DL} and ξ_{FL} as a function of Hf thickness, determined using fits to Eq.(1) for t_{FeCoB} spanning the range 2 -7 nm. The solid (dashed) orange lines are fits to the two sets of data (see [27]). The signs of both ξ_{DL} and ξ_{FL} are negative. The red solid line indicates a small residual FL spin torque efficiency (-0.010 ± 0.004).

Fig 2

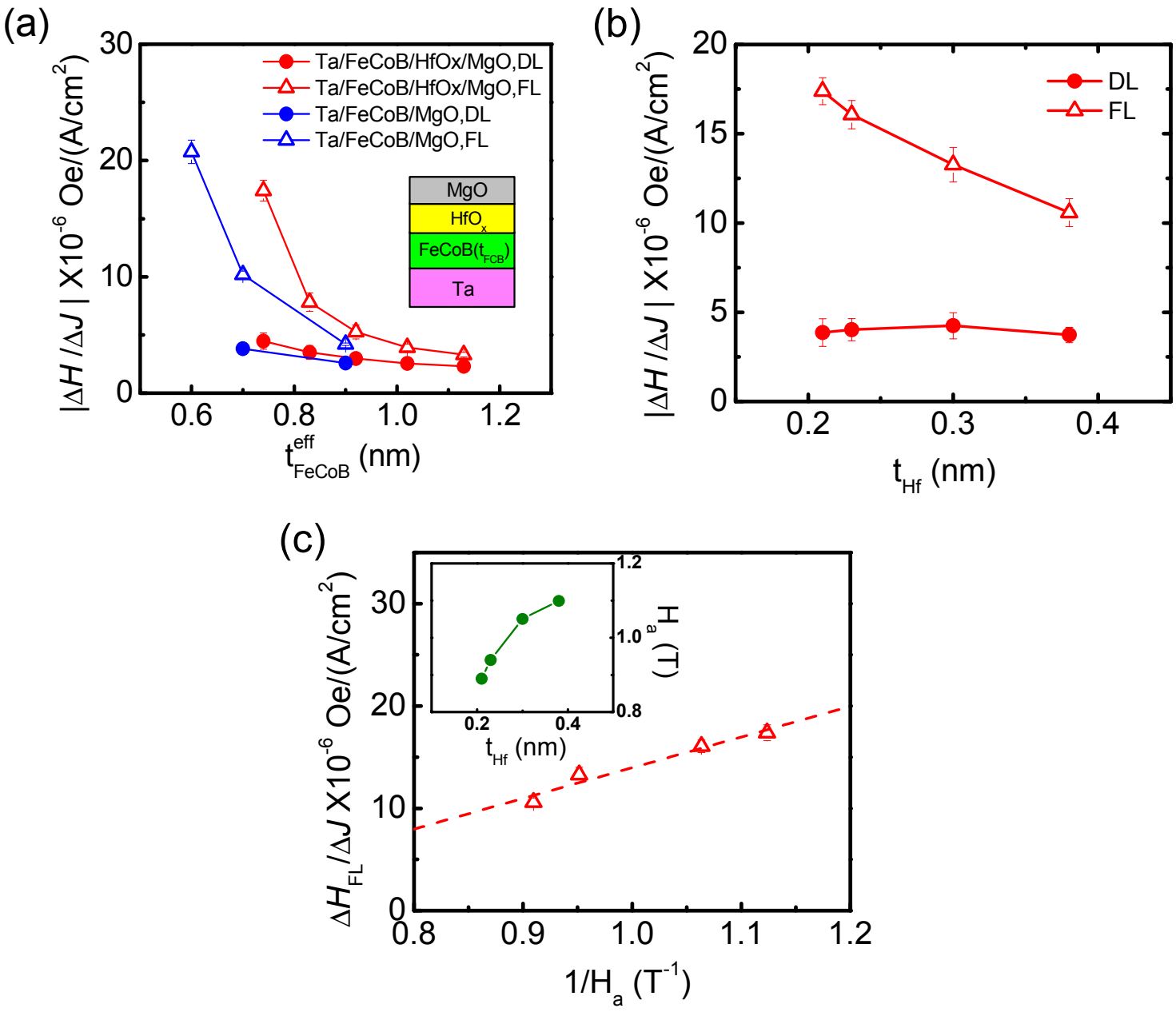

Fig. 2.(a) The current-induced effective fields $\Delta H_{DL}/\Delta J_e$ and $\Delta H_{FL}/\Delta J_e$ as a function of t_{FeCoB} for a Ta/FeCoB/MgO and a Ta/FeCoB/HfO_x(0.2)/MgO sample. (b) $\Delta H_{DL}/\Delta J_e$ and $\Delta H_{FL}/\Delta J_e$ as a function of the Hf passivation layer thickness. (c) $\Delta H_{FL}/\Delta J_e$ as a function of the inverse of the anisotropy field H_a. Inset: H_a for different Hf passivation layer thicknesses.

Fig 3

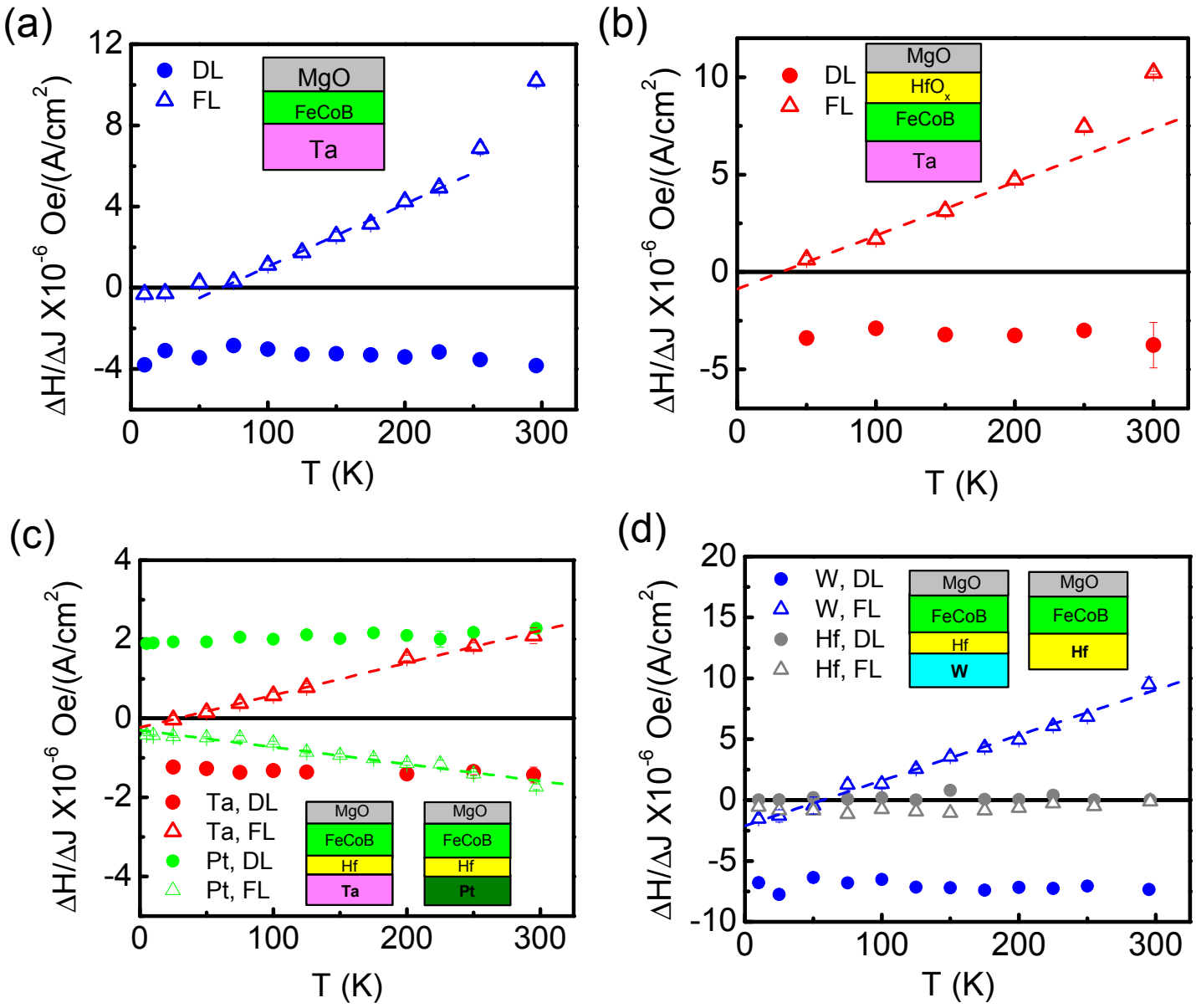

Fig. 3. Spin-orbit torque effective fields as a function of temperature for different samples: (a) Ta(4)/FeCoB(0.8)/MgO; (b) Ta(6)/FeCoB(0.8)/HfO_x(0.2)/MgO; (c) Ta(4)/Hf(1)/FeCoB(1)/MgO and Pt(4)/Hf(0.5)/FeCoB(1)/MgO; and (d) W(4)/Hf(1)/FeCoB(1)/MgO and Hf(4)/FeCoB(1)/MgO. The dashed lines are fits to the linear portion of the $\Delta H_{FL}(T)/\Delta J_e$ variation.