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M. Haidar  ; H. Mazraati ; P. Dürrenfeld ; H. Fulara ; M. Ranjbar; J. Åkerman 

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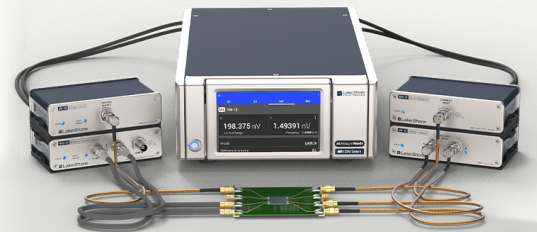
  
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M. Haidar,<sup>1,a)</sup>  H. Mazraati,<sup>2,3</sup>  P. Dürrenfeld,<sup>4</sup>  H. Fulara,<sup>4</sup>  M. Ranjbar,<sup>4</sup> and J. Åkerman<sup>4</sup> 

## AFFILIATIONS

<sup>1</sup>Department of Physics, American University of Beirut, Riad El-Solh, P.O. Box 11-0236, Beirut 1107-2020, Lebanon

<sup>2</sup>NanOsc AB, Kista 164 40, Sweden

<sup>3</sup>Department of Materials and Nanophysics, School of Engineering Sciences, KTH Royal Institute of Technology, 100 44 Stockholm, Sweden

<sup>4</sup>Department of Physics, University of Gothenburg, 412 96 Gothenburg, Sweden

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<sup>a)</sup> Author to whom correspondence should be addressed: [mh280@aub.edu.lb](mailto:mh280@aub.edu.lb)

## ABSTRACT

We demonstrate the compositional effect on the magnetodynamic and auto-oscillation properties of  $\text{Ni}_{100-x}\text{Fe}_x/\text{Pt}$  ( $x=10-40$ ) nanoconstriction-based spin Hall nano-oscillators. Using spin-torque ferromagnetic resonance performed on microstrips, we measure a significant reduction in both damping and spin Hall efficiency with the increasing Fe content, which lowers the spin pumping contribution. The strong compositional effect on spin Hall efficiency is primarily attributed to the increased saturation magnetization in Fe-rich devices. As a direct consequence, higher current densities are required to drive spin-wave auto-oscillations at higher microwave frequencies in Fe-rich nanoconstriction devices. Our results establish the critical role of the compositional effect in engineering the magnetodynamic and auto-oscillation properties of spin Hall devices for microwave and magnonic applications.

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The recent demonstration of pure spin current-induced spin-transfer torque arising from the spin Hall effect (SHE) represents an efficient route to controlling the magnetization dynamics in magnetic nanostructures.<sup>1,2</sup> In a ferromagnet (FM)/heavy metal (HM) bilayer, a pure spin current is generated when a longitudinal charge current passes through the HM and induces a transverse spin current due to the strong spin-orbit coupling in the HM. The conversion of the charge current density ( $J_C$ ) into pure spin current density ( $J_S$ ) is characterized by the spin Hall angle ( $\theta^{\text{SHA}}$ ) in these bilayers. The pure spin current may be sufficient to excite perpetual self-oscillations in the magnetization, which can further induce spin waves in these systems. This is of particular interest for spintronics applications and has led to a new class of microwave devices called spin Hall nano-oscillators (SHNOs).<sup>3-11</sup> Generating higher spin current densities through a higher  $\theta^{\text{SHA}}$  is necessary to improve the performance of SHNOs and to avoid higher charge current densities. So far, studies have focused on different HMs with various spin-orbit coupling strengths to generate higher spin current—for example, the  $\beta$ -phase of W,<sup>12-14</sup> Ta,<sup>15,16</sup> or

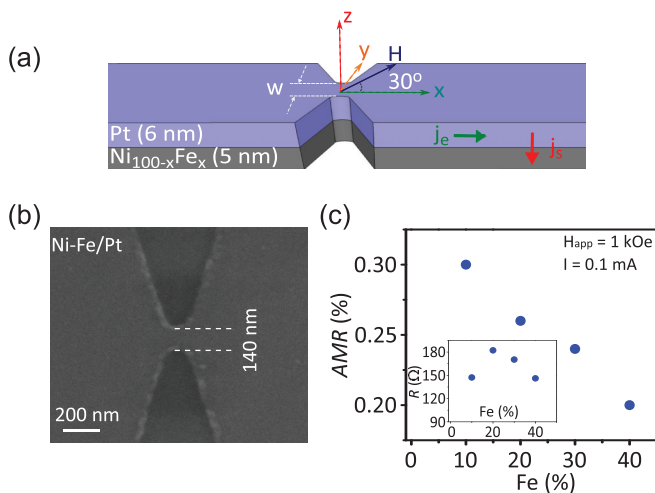
$\text{Ni}_x\text{Cu}_{1-x}$ .<sup>17</sup> It has also been shown that modifying the interface with Hafnium<sup>18,19</sup> or an antiferromagnet<sup>20,21</sup> enhances the transmission of the spin current between the HM and the FM. The amplitude of the spin current generated in the FM/HM bilayer is also controlled by the magnetodynamics of the ferromagnet<sup>22,23</sup> and the transparency of the FM-HM interface.<sup>24</sup>

In this work, we use spin torque-induced ferromagnetic resonance (ST-FMR) and auto-oscillation measurements to study the impact of the alloy composition on the magnetodynamics and spin-torque properties of spin Hall devices made of  $\text{Ni}_{100-x}\text{Fe}_x$  (5 nm)/Pt (6 nm) bilayers, where  $x = (10-40)$  denotes the Fe content in the FM in percent. First, we measure an increase in the magnetization, a decrease in the Gilbert damping, and a reduction in the impact of spin torque on the ST-FMR linewidth as the Fe content increases. Interestingly, we find a reduction in the spin Hall angle in Fe-rich devices, which scales in inverse proportion to the magnetization. Then, we study the microwave auto-oscillation characteristics as a function of Fe content in nanoconstriction SHNOs. In Fe-rich devices, the auto-oscillations have higher frequencies accompanied by low

output power; in addition, higher threshold current densities are required to drive auto-oscillations.

The layout of our devices is sketched in Fig. 1(a). The devices are fabricated from  $\text{Ni}_{100-x}\text{Fe}_x/\text{Pt}$  bilayers deposited in a high vacuum magnetron sputtering chamber with a base pressure below  $2 \times 10^{-8}$  Torr on sapphire C-plane substrates at room temperature. The different Ni-Fe alloys were co-sputtered under the same conditions and from the same pure Ni and Fe targets, where the composition was established by varying the respective plasma powers. Energy-dispersive x-ray spectroscopy (EDX) measurements showed that the actual alloy composition is within  $\pm 3$  at.% error of the nominal composition. Two kinds of spin Hall devices were fabricated from each film: (1)  $8 \times 16 \mu\text{m}^2$  rectangular stripes and (2) nanoconstriction-based SHNOs. The nanoconstriction devices are designed as  $4 \times 12 \mu\text{m}^2$  rectangles with two indentations of 50 nm tip radius forming a nanoconstriction in the center of the strip with a nominal width of 140 nm. For the  $\text{Ni}_{60}\text{Fe}_{40}/\text{Pt}$  device, the nominal width of the nanoconstriction is 120 nm. These patterns were fabricated using electron beam lithography followed by an etching process. The devices are protected from long-term degradation by a 50-nm-thick SiOx layer. A scanning electron micrograph of the nanoconstriction is shown in Fig. 1(b). We established electrical connection to the devices using coplanar waveguides, which we fabricated through a liftoff process from 1  $\mu\text{m}$  copper and 20 nm gold. Figure 1(c) shows the anisotropic magnetoresistance (AMR) and the resistance (inset) of the nanoconstriction devices as a function of the percentage of Fe. We measured a reduction in AMR in Fe-rich devices. The resistance drop for devices with the concentration below 20% in the inset of Fig. 1(c) is both consistent with the experimental literature<sup>25–28</sup> and with first-principles calculations, in particular when both disorder and spin-orbit coupling are included.<sup>29</sup>

We first discuss the ST-FMR spectra measured on rectangular stripes to determine the variation of the magnetodynamics with the Fe



**FIG. 1.** (a) Schematic layout of nanoconstriction devices made of  $\text{Ni}_{100-x}\text{Fe}_x/\text{Pt}$  bilayers. This shows the direction of the applied field and the dc current. (b) Scanning electron microscope image of a device with a width of 140 nm. (c) Anisotropic magnetoresistance and resistance (inset) of the devices as a function of Fe content.

content. The ST-FMR measurements were performed by connecting the devices to a pulse-modulated signal generator to excite the dynamics and to a lock-in amplifier to detect the mixing voltage, which arises in resonance due to the synchronism of the time-varying magnetoresistance with the excitation current. The devices are placed in a uniform magnetic field applied in-plane at an angle of  $30^\circ$  with respect to the x-direction. The measurements were performed by sweeping the magnetic field at a fixed frequency to reduce the nonmagnetic background. The measurements were performed over a broad range of frequencies.

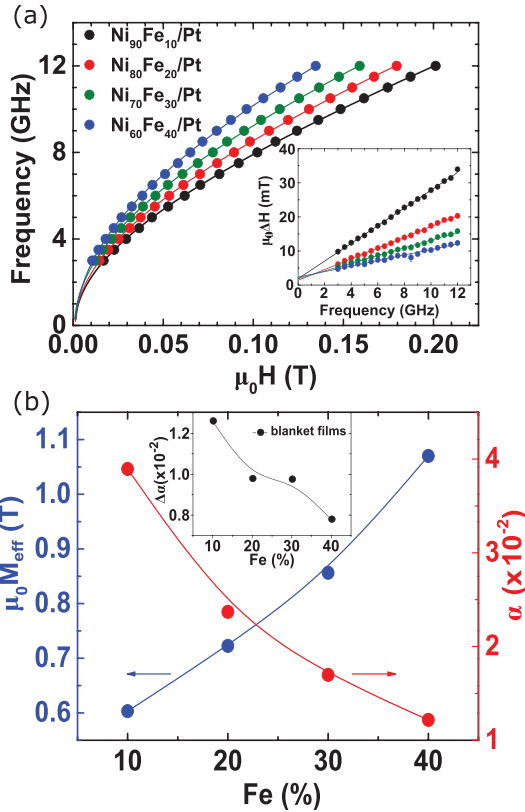
We observe that the magnitude of the dc voltage around the resonance field depends on the Fe percentage composition. The shape of the measured dc voltage originates from two processes that can be distinguished by their symmetry with respect to the field. The asymmetric part originates from the anisotropic magnetoresistance (AMR), while the symmetric part comes from the spin torque and inverse spin Hall effect voltage.<sup>30</sup> We, thus, fit the dc voltage to a single Lorentzian function, which has symmetric and antisymmetric components,

$$V = V_0 + V_S \frac{\Delta H^2}{\Delta H^2 + 4(H - H_r)^2} + V_A \frac{4\Delta H(H - H_r)}{\Delta H^2 + 4(H - H_r)^2}, \quad (1)$$

where  $V_0$  is the background voltage,  $V_S$  and  $V_A$  are the symmetric and antisymmetric Lorentzian coefficients, respectively,  $H_r$  is the resonance field, and  $\Delta H$  is the full width at half maximum. From the fitting, we extract  $H_r$  and  $\Delta H$  at each frequency  $f$ , as plotted in Fig. 2(a), to extract the magnetodynamics of these devices. By fitting  $f$  vs  $H_r$  to the in-plane Kittel equation,  $f = \frac{\mu_0 \gamma}{2\pi} \sqrt{(H + H_k)(H + H_k + M_{\text{eff}})}$ , where  $\gamma/2\pi$  is the gyromagnetic ratio and  $H_k$  is the in-plane anisotropy, the effective magnetization ( $\mu_0 M_{\text{eff}}$ ) of the device can be extracted. A clear reduction in  $\mu_0 M_{\text{eff}}$  is measured for Ni-rich films, as shown in Fig. 2(b), as the magnetic moment per atom decreases in Ni-rich films. We found only small values of  $\mu_0 H_k$  (Table I). We measure the saturation magnetization ( $\mu_0 M_s$ ) separately on blanket films, using Alternating Gradient Magnetometry (AGM) (Table I).

Gilbert damping ( $\alpha$ ) can be extracted from the slope of  $\mu_0 \Delta H$  vs  $f$  using  $\mu_0 \Delta H = \frac{2\alpha}{\gamma} (2\pi f) + \Delta H_0$ , where  $\Delta H_0$  is the inhomogeneous broadening. The results for the  $\alpha$  vs Fe composition are shown in Fig. 2(b). Several mechanisms determine the measured value of  $\alpha$ . An intrinsic contribution with a similar increase in the Gilbert damping has been reported in bulk Ni-Fe alloys.<sup>29,31</sup> In Ni-rich films, the larger values of  $\alpha$  can be understood in terms of a conductive-like behavior that can be explained by intraband scattering.<sup>32</sup> In addition to the intrinsic origin of  $\alpha$  in our devices, the spin pumping contributes substantially to the measured damping of each device due to the strong spin-orbit coupling of Pt. We compare the damping of  $\text{Ni}_{100-x}\text{Fe}_x$  with and without the Pt layer on similar blanket films, where we estimated the spin pumping contribution to the damping  $\Delta\alpha = (\alpha_{\text{FM}/\text{Pt}} - \alpha_{\text{FM}/\text{Cu}})$ . We measure a decrease in  $\Delta\alpha$  with the Fe content as shown in the inset of Fig. 2(b), which suggests that bilayers containing Fe-rich films have lower spin pumping.

Next, we discuss the correlation between the spin Hall efficiency and the Fe composition. We carried out ST-FMR measurements on rectangular stripes of spin Hall devices at  $f = 5$  GHz with an injected power of  $P = +15$  dBm. We examined the variation of  $\mu_0 \Delta H$  with an applied dc current ( $I$ ) in the range of  $+8$  mA to  $-8$  mA for all devices. The ST-FMR signal reverses its sign, as we sweep the direction of the

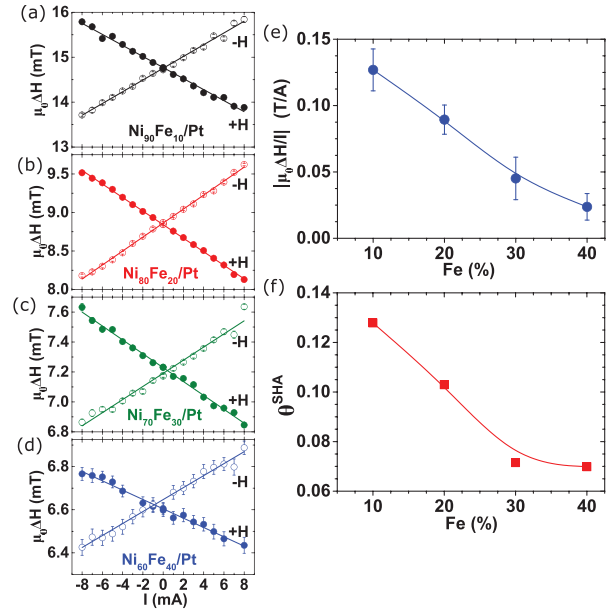


**FIG. 2.** Magnetodynamics of rectangular stripes. (a) Variation of frequency vs field and linewidth vs frequency (inset) for  $\text{Ni}_{100-x}\text{Fe}_x/\text{Pt}$  devices. (b) shows the magnetization (blue circles) and the Gilbert damping (red circles) as a function of the Fe content. Inset, variation of  $\Delta\alpha$  vs Fe content, the difference in damping of  $\text{Ni}_{100-x}\text{Fe}_x$  films with and without the Pt layer, measured on similar blanket films using broadband-FMR. The solid curves are given as guides to the eye.

field or the current direction to the opposite polarity. Figures 3(a)–3(d) show the variation of the extracted  $\mu_0\Delta H$  vs  $I$  for devices measured at  $+H$  (closed circles) and  $-H$  (open circles). A behavior common to these measurements is that, for a positive field,  $\mu_0\Delta H$  decreases with  $+I$  and increases with  $-I$ . We observe the opposite behavior under negative fields, as expected from the symmetry of the spin Hall effect. This is due to the spin torque exerted by the pure spin currents generated from the SHE in Pt and injected into the FM layer. The spin torque is aligned either parallel or antiparallel to the Gilbert

**TABLE I.** Summary of the magnetodynamics for  $\text{Ni}_{100-x}\text{Fe}_x/\text{Pt}$  rectangular stripe devices.

Samples	$\mu_0 M_s$ (T)	$\gamma/2\pi$ (GHz/T)	$\mu_0 M_{\text{eff}}$ (T)	$\mu_0 H_k$ (mT)	$\alpha$ ( $10^{-2}$ )	$R_{\text{FM}}$ ( $\Omega$ )
$\text{Ni}_{90}\text{Fe}_{10}/\text{Pt}$	0.67	30	0.673	1.1	$3.89 \pm 0.1$	98.5
$\text{Ni}_{80}\text{Fe}_{20}/\text{Pt}$	0.82	30	0.819	1.2	$2.37 \pm 0.1$	185.6
$\text{Ni}_{70}\text{Fe}_{30}/\text{Pt}$	0.926	30	0.926	0.8	$1.69 \pm 0.1$	152.2
$\text{Ni}_{60}\text{Fe}_{40}/\text{Pt}$	1.18	30	1.074	1.7	$1.2 \pm 0.1$	93.3



**FIG. 3.** (a)–(d) The current induces a change in the linewidth  $\mu_0\Delta H$  as a function of the dc current for different Ni-Fe compositions. Closed and open circles show results of measurements performed under  $+H$  and  $-H$ , respectively. (e) and (f) Variation in  $|\mu_0\Delta H/I|$  and the spin Hall angle ( $\theta^{\text{SHA}}$ ) as a function of Fe composition. The solid curves are given as guides to the eye.

damping, which leads to an effective enhancement or reduction in the damping of the ferromagnetic layer. The linewidth analysis is a reliable method for measuring the spin Hall angle, as it originates only from the antidamping spin-transfer torque. In the linear regime, the current-induced spin torque could be modeled as<sup>30,33</sup>

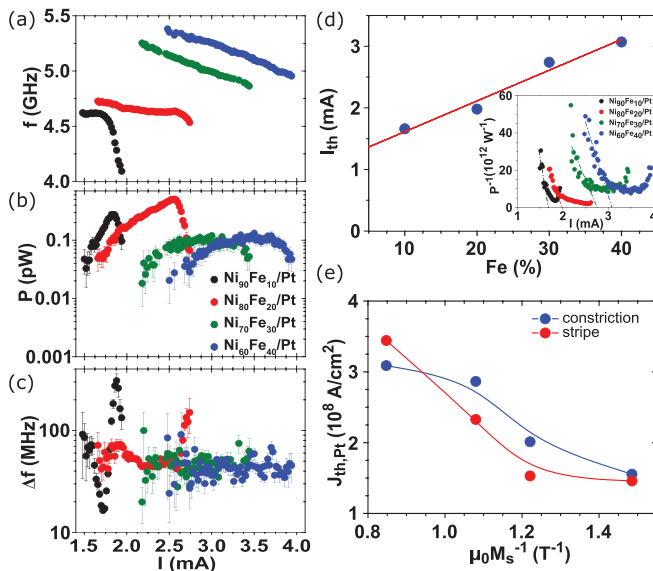
$$\mu_0\Delta H = \frac{2\pi f}{\gamma} 2\alpha + \Delta H_0 + \frac{2\pi f}{\gamma(H + 2\pi M_{\text{eff}})} \frac{\sin(\varphi)}{\mu_0 M_s t} \frac{\hbar}{2eA_c} \frac{R_{\text{FM}}}{R_{\text{FM}} + R_{\text{Pt}}} \theta^{\text{SHA}} I, \quad (2)$$

where  $R_{\text{FM}}$  and  $R_{\text{Pt}}$  are the resistance of the FM layer and Pt, respectively.  $A_c$  is the cross section of the device,  $\varphi$  is the in-plane angle, and  $\theta^{\text{SHA}}$  is the spin Hall angle of the FM/Pt bilayer that includes possible angular momentum losses at the interfaces. The first and second terms represent the linewidth in the absence of the dc current  $\mu_0\Delta H(I = 0)$ , while the third term on the right-hand side is the spin current-induced modification to  $\mu_0\Delta H$ .

The solid lines in Figs. 3(a)–3(d) are linear fits to Eq. (2), from which we extracted the slope  $|\mu_0\Delta H/I|$ . Figure 3(e) summarizes the results of the extracted  $|\mu_0\Delta H/I|$  for each device as a function of Fe composition. The impact of spin torque on the ST-FMR linewidth is found to be linear with a slope  $|\mu_0\Delta H/I|$  that decreases by a factor of six with the Fe content. We then calculated  $\theta^{\text{SHA}}$  from  $|\mu_0\Delta H/I|$  using Eq. (2) since the other factors are constants in our measurements. We measure  $R_{\text{Pt}} = 65 \Omega$ , and  $R_{\text{FM}}$  values are summarized in (Table I). Figure 3(f) shows  $\theta^{\text{SHA}}$  as a function of Fe content, where a reduction in  $\theta^{\text{SHA}}$  by a factor of two is measured for Fe-rich devices.  $\theta^{\text{SHA}}$  of  $\text{Ni}_{80}\text{Fe}_{20}/\text{Pt}$  is similar to the

reported values of Permalloy-based bilayers.<sup>30</sup> As the magnetization increases from 0.67 T for Ni<sub>90</sub>Fe<sub>10</sub>/Pt to 1.18 T for Ni<sub>60</sub>Fe<sub>40</sub>/Pt, roughly by a factor of two,  $\theta^{\text{SHA}}$  decreases by a factor two. This hints that the reduction in the spin Hall angle can be correlated primarily to the compositional effect: the spin Hall angle scales inversely proportional with the saturation magnetization. Note that the observed variation of the spin Hall angle with the increasing Fe content is also qualitatively consistent with the decrease in damping as a function of the Fe content, as shown in Fig. 2(b), and can be well explained in terms of the spin transport model given by Eq. (1) in Ref. 17. According to Eq. (1), the suppression of spin pumping due to increased  $\mu_0 M_s$  should result in lowering of the effective spin mixing conductance and, therefore, spin-torque efficiency, which can be seen as a monotonic decrease in the spin Hall angle with the increasing Fe content in Fig. 3(f). Other sources that may contribute to the reduction of  $\theta^{\text{SHA}}$  are (i) the transparency of the Pt-ferromagnet interface to the spin current<sup>24,34</sup> and (ii) part of the spin Hall efficiency that may also originate from the Ni<sub>1-x</sub>Fe<sub>x</sub> layer itself,<sup>8</sup> as the spin Hall angles of Ni and Fe have opposite signs.<sup>35</sup>

Next, we turn to discuss the compositional effect on the characteristics of auto-oscillations in nanoconstriction-based SHNOs. To study auto-oscillations, we replace the lock-in amplifier with a spectrum analyzer and a low noise amplifier with a +33 dB gain. The generated microwave power spectral density (PSD) was recorded as a function of a direct current under an in-plane magnetic field of  $\mu_0 H = 0.05$  T. The auto-oscillation frequencies, integrated power, and linewidth are extracted by fitting the peaks of all spectra with a Lorentzian function. Figures 4(a)–4(c) show the spectral characteristics



**FIG. 4.** (a) Frequency, (b) integrated power, and (c) linewidth of the microwave auto-oscillations as a function of current for different SHNOs. The applied field is  $\mu_0 H_{\text{ext}} = 0.05$  T. (d) Threshold current vs Fe concentration. The inset shows how the threshold is extracted from a linear fit of  $p^{-1}$  vs  $I$  at low currents (dashed line). (e) Threshold current density in Pt vs  $(\mu_0 M_s)^{-1}$  extracted from ST-FMR (red circles) and auto-oscillation (blue circles) measurements. The solid curves are given as guides to the eye.

of the microwave auto-oscillations. As the device is biased with a direct current, the emission of auto-oscillation starts once the spin torque overcomes the damping completely. We observe emission of auto-oscillation at a single frequency, below FMR, at the threshold current, and then at higher currents, a redshift in frequencies is detected. This behavior is expected for in-plane magnetic fields, where the active medium is characterized by a negative nonlinearity that makes the auto-oscillation frequency decrease with amplitude.<sup>36</sup> Note that the auto-oscillating mode starts to localize near one of the constriction edges where it, at higher currents, nucleates into a bullet mode, which is first more centrally localized in the constriction and then shrinks with increasing driving current.<sup>37</sup> As a result, we observe a drop of power at higher drive currents, as shown in Fig. 4(b). For the lowest Fe concentration device Ni<sub>90</sub>Fe<sub>10</sub>/Pt, at  $I = 1.75$  mA, we measure a sharp drop in the frequency, an increase in the linewidth, and a decrease in the power, which are signatures of the dominant spin-wave bullet mode. Our experimental data further suggest that the lower  $\mu_0 M_{\text{eff}}$  value for Ni-rich devices drives the nonlinearity in such a way that the nucleation of the bullet mode takes place at a lower drive current. Therefore, we observe a sharp decrease in frequency for Ni-rich devices. It is interesting to note that the onset auto-oscillation frequency differs between devices, i.e., the onset frequency shifts up for Fe-rich devices due to higher magnetization. The integrated power varies with the dc current, showing a bell shape of an amplitude around 1 pW. A qualitative comparison between the power of devices shows that Fe-rich devices have lower output power. This is understood as the readout of devices depends on AMR and as the AMR drops in Fe-rich devices the power follows. Finally, the measured auto-oscillations have linewidths around 50 MHz. The threshold current for auto-oscillations,  $I_{\text{th}}$ , is extracted from plots of  $p^{-1}$  vs  $I$ ,<sup>38</sup> as shown in the inset of Fig. 4(d). The dashed lines show the position of  $I_{\text{th}}$ ; obviously, for Fe-rich devices, higher currents are required to excite auto-oscillations as shown in Fig. 4(d). From  $I_{\text{th}}$ , we estimate the threshold current density in Pt,  $J_{\text{th,Pt}}$ , as plotted in Fig. 4(e) (blue dots). It is also interesting to compare  $J_{\text{th,Pt}}$  with that extracted from the ST-FMR measurements on stripes by extrapolating  $\mu_0 \Delta H$  vs  $I$  in Fig. 3(e), to zero. Results are shown in Fig. 4(e) (red dots). A perfect matching is measured between  $J_{\text{th,Pt}}$  extracted from both measurements. We observe an increase in  $J_{\text{th,Pt}}$  with the increase in the magnetization. The enhancement in threshold current densities for Fe-rich devices is a direct consequence of the reduction in the spin Hall angle. Our experimental results show that the dominant compositional effect is from the magnetization that plays a central role in determining the characteristics of the spin Hall devices.

In conclusion, we have investigated the impact of the alloy composition on the magnetodynamic properties of Ni-Fe-based spin Hall devices. Our results will be useful not only for fundamental studies of spin transport between Ni-Fe alloys and heavy metal layers but also in the engineering of new devices with controllable properties for spin torque and magnonic devices.

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#### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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