**INTRODUCTION**

The unidirectional propagation of signals is the key to modern electronic logic circuits and fundamental applications, such as diodes, isolators, gyrators, and circulators (1). Because nonreciprocity offers an opportunity to control the wave devices flowing only to a desirable direction, the nonreciprocity in waves has been extensively studied, and wave devices with nonlinear propagating properties are introduced in the fields of microwaves (2), acoustics (3, 4), and photonics (5, 6). A typical nonreciprocity ratio of nonreciprocal microwave devices (7) is $\sim 10^3$, and the nonreciprocity of acoustics (3) and photonics (5) is reported to be $\sim 10^4$.

In rapidly developing areas of magnonics, the spin waves (8–14) can be the information carrier for magnonic logic circuits (15–19). The specific property of the nonreciprocity might be a benefit for the enhancement of logic circuits based on magnetic waveguide. Therefore, the characterization and tuning of the nonreciprocity in magnonics have been intensively investigated (20–31), including the relation between the nonreciprocity and spin pumping (32) or Dzyaloshinskii-Moriya interaction (DMI) (33, 34). The nonreciprocity, due to its intrinsic characteristic in the magnetostatic surface wave (MSSW) mode (35), together with Joule-heat–free transport in gigahertz frequencies (30) and compatibility with the spin Hall current system (36, 37) makes spin wave one of the promising candidates for future data carriers.

However, the suitability of nonreciprocal spin waves for the above applications is limited to a small nonreciprocity parameter, $\kappa$. For example, it was reported that a typical value of $\kappa$ in MSSW is 0.6 to 0.9 in a permalloy (Py) strip by using time-resolved spin wave spectroscopy (see fig. S1) (23, 26) and 4 to 5 by using spatially resolved Brillouin light-scattering spectroscopy (22). The value of $\kappa$ in an yttrium iron garnet film, which has a low saturation magnetization, is higher compared to that in Py (27). However, a large value of $\kappa$ in a typical $3d$ ferromagnet, such as Py, is essential for the realization of spin wave–based applications, because Py is compatible with the prevailing microfabrication technology.

Here, we report giant nonreciprocal emission of spin waves in a Ta/Py bilayer with out-of-plane bias fields, with $\kappa$ values up to 14 and 60 in time and frequency domains, respectively. An optical image of the spin wave device used in this study is shown in Fig. 1A. A pulse inductive technique was used for measurements, in which an electric pulse was applied to the right asymmetric coplanar strip (ACPS) for the excitation of the spin wave in a ferromagnetic thin film (20–nm Py) on top of various thicknesses ($t_{Ta}$) of Ta from 1.2 to 8.9 nm. The spin wave propagating along the $x$ direction is detected using a sampling oscilloscope at the left ACPS. An out-of-plane bias field ($|H_0|$ < 490 mT) is applied to the sample during the measurements. All measurements were performed at room temperature.

The nonreciprocity in MSSW is due to either the interference between the spin waves produced by the $x$ and $z$ components of $\mathbf{rf}$ local excitation fields (antenna–spin wave coupling) or the local concentration of the spin waves at the top and bottom surfaces of the magnetic film. In our case, the amplitude of spin waves across the Py thickness can be assumed to be uniform because $kt_{Py} \ll 1$ (22, 35), where $k$ (2$\pi/\lambda$), the wavelength of spin waves) is the wave vector and $t_{Py}$ (20 nm) is the thickness of Py. Therefore, the basic mechanism of nonreciprocity in our system is the antenna–spin wave coupling. In addition, we discuss two reasons for the observed high nonreciprocity arising at the Ta/Pt interface. The first reason is the tilted out-of-plane anisotropy induced by the Ta underlayer, and the second one is the spin pumping effect at the Ta/Pt interface. The vibrating sample magnetometry (VSM) and anisotropic interface magnetoresistance (AIMR) measurements indeed show the presence of an interfacial anisotropy at the Ta/Pt interface. Taking this anisotropy into account, the micromagnetic simulations support the observed experimental results.

**RESULTS**

Figure 1B shows the contour plot of the field-dependent measurements in the time domain for the device with $t_{Ta}$ = 6.1 nm. A representative
spin wave packet measured at $H_z = 25.8$ mT is shown in Fig. 1C for different propagation directions ($\pm k$). The shape of this wave packet closely resembles that of the MSSW. The fast Fourier transform (FFT) of time domain signals in Fig. 1D shows that the traveling spin wave resonance frequency ($f_0$) increases with increasing $|H_x|$ for the device with $t_{Ta} = 6.1$ nm. Figure 1D also shows that the FFT intensity of spin wave packets is stronger for positive $H_x$, as compared to that for negative $H_x$, demonstrating nonreciprocal emission of spin waves depending on the polarity of the applied out-of-plane fields. For a comparison, the MSSW (in-plane bias fields), the magnetostatic backward volume (MSBV), the magnetostatic forward volume (MSFV), and the MSSW-like (out-of-plane fields) mode of this study are shown in Fig. 1E.

The amplitude of wave packets for devices with different $t_{Ta}$ is shown as a color plot in Fig. 2A. The maximum value of the spin wave amplitude was observed at $t_{Ta} = 6.1$ nm for the positive $H_x$ and at $t_{Ta} = 3.3$ nm for the negative $H_x$. Figure 2B shows that the amplitude at $H_z = -25.8$ mT decreases as $t_{Ta}$ increases, whereas the amplitude at $H_z = +25.8$ mT initially increases (up to $t_{Ta} = 6.8$ nm) and then decreases, as summarized in Fig. 2C. The first regime ($t_{Ta} < 6.8$ nm) is explained by the tilted anisotropy induced by the Ta layer, and the second regime ($t_{Ta} > 6.8$ nm) is attributed to the spin pumping effect, as discussed later. The spin wave nonreciprocity ($\kappa$) is defined as the ratio of spin wave amplitudes at positive and negative fields: $\kappa = A(+H_x)/A(-H_x)$. The maximum value of $\kappa$ in the time domain is $\sim 14$ for the device with $t_{Ta} = 8.2$ nm, as shown in the inset of Fig. 2C, and the $\kappa$ reaches up to 60 in the frequency domain (the details of $\kappa$ in the time and frequency domains are documented in sections S2 and S3).

We propose that the effect of the Ta underlayer is to induce a canted interfacial magnetic anisotropy, which affects the nonreciprocity factor. To confirm the presence of interfacial anisotropy induced by Ta underlayers, we conducted the out-of-plane AIMR measurements on all the devices, and the results are shown in Fig. S4C. Because of AIMR, the in-plane electrical resistance in the perpendicular anisotropy materials depends on the out-of-plane component of the magnetization (38). In Fig. 3, $\Delta R_{OP}$ at $+H_z$ and $-H_z$ are plotted versus the Ta underlayer thickness, where $\Delta R_{OP}$ is the maximum change in the resistance of the device when the magnetic field is swept along the $z$ direction. $\Delta R_{OP}$ is a measure of AIMR that originates from the Ta/Pt interface, due to anisotropic magnetic scattering of electrons (38). $\Delta R_{OP}$ changes its polarity across $t_{Ta} \sim 3$ nm, which suggests a change in anisotropy direction in the devices. It is consistent with the trend observed in a polarity change of spin wave amplitudes in Fig. 2B.

For further confirmation of the presence of induced anisotropy in our sample, VSM measurements were conducted, and a weak perpendicular anisotropy in the $z$ direction was indeed observed. The anisotropy was verified from an inner loop from VSM hysteresis as shown in Fig. S5, which is in line with a recent report (39). The $z$ component of the magnetization shows a finite remanence value ($M_r$) at $H_z = 0$. The measured $M_r$ with rotating the sample around the $x$ axis shows the minimum at a positive angle for $t_{Ta} = 1$ nm and at negative angles for $t_{Ta} = 3, 5,$ and 10 nm,
indicating a change in the canted angle of perpendicular anisotropy for different $t_{Ta}$. For the thicker $t_{Ta}$, the uniaxial anisotropy is canted to the $y$ axis with the projected $M_y$ along the $-y$ direction with $H_z > 0$, whereas the $M_y$ is along the $+y$ direction for the thinner $t_{Ta}$ with $H_z > 0$.

To better understand the relation between anisotropy and observed nonreciprocity, we performed micromagnetic simulations (see fig. S6). Simulations were carried out on a bilayer structure, with the bottom layer having a perpendicular uniaxial anisotropy. The spin wave amplitude obtained from simulations shows a qualitative agreement with the measured data as shown in Fig. 2D. From the investigation of magnetization configurations in simulated results, we found that the spin wave nonreciprocity is correlated with the polarity of the $y$ component of the magnetization. Because the magnetization has the $y$ component, the observed spin waves are MSSW-like. The tilting direction of the aforementioned anisotropy depends on the $t_{Ta}$ at fixed $\pm H_z$. When the applied $H_z$ varies, the $y$ component of the magnetization changes its sign, leading to a change in relative amplitude of spin waves at $\pm H_z$ ($21-23, 31$). In addition, for a better understanding of the antenna–spin wave coupling, we spatially and temporally analyzed our micromagnetic simulation results with the relation between the excitation magnetic field from the antenna and the local magnetization at the bottom of the antenna (see fig. S7).

Although a change in the anisotropy direction depending on $t_{Ta}$ can explain the trend of the nonreciprocity, a high nonreciprocal factor especially at thicker $t_{Ta}$ ($t_{Ta} \geq 6.8$ nm) cannot be fully explained using the above model. This discrepancy between the results from the micromagnetic simulations and the experimental observation suggests that a mechanism other than the interfacial anisotropy is also in operation and becomes more dominant at thicker $t_{Ta}$ layers. To further understand the origin of high nonreciprocity, the loss mechanisms for spin wave energy should be considered. Apart from the usual energy loss mechanisms in a ferromagnet, which are quantified by the Gilbert layer having a perpendicular uniaxial anisotropy. The spin wave amplitude obtained from simulations shows a qualitative agreement with the measured data as shown in Fig. 2D. From the investigation of magnetization configurations in simulated results, we found that the spin wave nonreciprocity is correlated with the polarity of the $y$ component of the magnetization. Because the magnetization has the $y$ component, the observed spin waves are MSSW-like. The tilting direction of the aforementioned anisotropy depends on the $t_{Ta}$ at fixed $\pm H_z$. When the applied $H_z$ varies, the $y$ component of the magnetization changes its sign, leading to a change in relative amplitude of spin waves at $\pm H_z$ ($21-23, 31$). In addition, for a better understanding of the antenna–spin wave coupling, we spatially and temporally analyzed our micromagnetic simulation results with the relation between the excitation magnetic field from the antenna and the local magnetization at the bottom of the antenna (see fig. S7).

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damping $\alpha$, a heavy metal Ta underlayer can also absorb the energy from magnetization dynamics via spin pumping (32). Spin pumping is the process in which the magnetization precession in a ferromagnet (Py) generates spin current density $J_{sp} \propto \dot{m} \times (\partial \dot{m}/\partial t)$ into the adjacent paramagnetic layer (Ta) (40–42). Because of high spin-orbit coupling in Ta, this spin current is converted to a transverse charge current $J_C \propto J_{sp} \times \dot{m}$ via inverse spin Hall effect (ISHE), which can be measured as a transverse electromotive force across the device.

We performed spin pumping–induced ISHE measurements to investigate energy loss from spin waves in Py to Ta. A schematic of the spin pumping measurement is shown in Fig. 4A. In the geometry of our experiment, a spin pumping–induced ISHE signal ($V_{sp}$) can be observed only if there exists a nonvanishing $y$ component of the magnetization at the interface, which accounts for the observation of MSSW-like wave packets in Fig. 2B. The ISHE signals measured at an rf frequency of 1.3 GHz corresponding to the spin wave resonance frequency at $|H_{rf}| = 25.8$ mT are shown in Fig. 4B. The amplitude of spin pumping signals exhibits an abrupt jump at $t_{Ta} = 6.8$ nm as shown in Fig. 4C, which can be related to the spin diffusion length in Ta (~7 nm) (43). The amplitude of $V_{sp}$ is a measure of loss of the total spin wave energy that is absorbed into the Ta layer (44). Thus, above $t_{Ta} > 6.8$ nm, the spin wave amplitude should decrease because of the strong spin pumping effect. In Fig. 2C, this decrease in the spin wave amplitude is clearly observable for both positive and negative applied fields for $t_{Ta} \geq 6.8$ nm.

If the spin pumping changes damping equally for the $+H_Z$ and $-H_Z$ spin waves, both wave amplitudes will be reduced proportionately, leading the nonreciprocity to be unchanged. Therefore, the spin pumping must unequally affect the spin waves for two field directions ($\pm H_Z$), to explain the observed giant nonreciprocity, especially at thicker Ta ($t_{Ta} \geq 6.8$ nm). We propose a plausible mechanism of the spin wave amplitude–dependent damping, in which the spin pumping–induced Gilbert damping of large-amplitude spin waves at $+H_Z$ is smaller than that of small-amplitude spin waves at $-H_Z$ (see section S8). Consequently, a large-amplitude (small-amplitude) spin wave experiences less (more) attenuation when propagating, resulting in an increased non-reciprocity at thicker Ta.

To confirm the spin pumping effect, we performed the ferromagnetic resonance (FMR) measurements with the same devices of the spin pumping measurements. Taking the spin pumping effect into account, the presence of Ta layer enhances the damping parameter of Py above the intrinsic value (45, 46)

$$\alpha_{eff} = \alpha_0 + \Delta \alpha = \alpha_0 + \frac{g_{MB}}{4\pi M_s f_{FM}} g_{eff}^\uparrow$$

where $\alpha_0$ is the intrinsic damping, $\Delta \alpha$ is an additional damping because of the spin pumping effect, $g$ (2.2) is the $g$-factor, $\mu_B$ is the Bohr magneton, $f_{FM}$ (20 nm) is the thickness of the Py film, $M_s$ is the saturation magnetization, and $g_{eff}^\uparrow$ is the effective spin-mixing conductance. We extracted the additional damping constant ($\Delta \alpha$) from the FMR spectra as shown in Fig. 4C. The plotted damping enhancement as a function of the Ta thickness shows good agreement with spin pumping signals.

**DISCUSSION**

From the first principles calculation, the 5d orbital of Ta, which has a large spin-orbit coupling, is strongly hybridized with the 3d orbital of Fe,
giving rise to strong perpendicular magnetic anisotropy at the interface (47). The anisotropy properties, such as an axis or strength, are sensitive to intermixing, roughness, oxidation, and hybridization at the interface, which are affected by different $t_{\text{Ta}}$. The origin of the canted perpendicular anisotropy to the $y$ axis could be a change in those parameters depending on $t_{\text{Ta}}$. The canting anisotropy only occurs at the Ta/Py interface, and the anisotropy direction becomes closer to the $x$ axis away from the interface because of the shape anisotropy. It has been reported earlier that a Ru seed layer on the top of NiFe can induce surface/interface anisotropy (48), and Ta or Ru underlayer with CoFeB can result in a perpendicular anisotropy (49). In our experiment, the nonreciprocity is found to change with different $t_{\text{Ta}}$ whereas the thickness of Ru is kept constant at 3 nm. Therefore, we conclude that the Ta underlayer induces a progressive change in the spin wave properties.

Finally, we investigated the possible role of DMI in our devices. DMI is observed in a broken inversion symmetry (50, 51) and at the interface or surface of magnetic multilayers (52, 53) and could be expected to have an opposite effect in the inverted sample structure. Therefore, we performed spin wave measurements in devices with the Ta layer on the top of the Py layer (see fig. S9). It turns out that the nonreciprocal ratio in the inverted devices show a similar trend as that of Ta underlayer structures. In our case, we conclude that the structural sequence between the Ta and Py layers does not affect the nonreciprocal emission of spin waves.

It must be noted that our observations are different from the earlier reports on spin wave nonreciprocity studied only in the MSSW mode (22, 23, 26, 28). To compare our results with the MSSW mode, we have also measured the nonreciprocity value with sweeping in-plane magnetic field, $H_y$. It is found that a small value (~0.7) of nonreciprocity is observed regardless of the Ta thickness, similar to previous reports (54) (see fig. S1).

The observed very large nonreciprocity of spin waves in the Ta/Py bilayer can be attributed to the combined effects, such as the polarity change in the in-plane magnetization due to the canted out-of-plane anisotropy and the spin pumping process at the Ta/Py interface. Utilizing an intimate coupling between the interfacial spin configuration and spin pumping effect, even larger nonreciprocity can be achieved. In addition, noncollinear spin configuration due to DMI may enhance the nonreciprocity ratio (33). Our intriguing findings will be useful for future spin wave–based applications, such as spin wave information transports and logic devices because of the high nonreciprocal ratio.

**MATERIALS AND METHODS**

**Sample preparation**

Si/SiO$_2$ substrates/Ta ($t_{\text{Ta}}$ nm)/Ni$_{81}$Fe$_{19}$ (20 nm)/Ru (3 nm) was patterned in a rectangular shape ($120 \times 340$ $\mu$m$^2$). Magnetic fields were not applied during the sputtering. A wafer was being rotated during sputtering to avoid a field deposition under any stray field effects from the magnetron sputtering gun or the chamber. The SiO$_2$ (35 nm) insulating layer was deposited to isolate an ACPS from the Py layer. The ACPS consists of Ta (5 nm)/Au (100 nm). The signal line width is 10 $\mu$m. The gap (edge to edge) between the signal line and the ground conductor is 10 $\mu$m, and the distance between the signal lines of two ACPSs is 10 $\mu$m as shown in Fig. 1A.

**Measurement setup**

We used pulsed inductive microwave magnetometry to generate and measure the spin wave signals. Spin waves were generated by applying a 1.8-V electric pulse to the excitation ACPS to produce local rf magnetic fields around the signal line. The rise and fall times of the electric pulse were ~70 and ~80 ps, respectively. The pulse width was ~100 ps, and the pulse cycle frequency was 100 MHz. The excited spin waves propagated through the Py strip and were inductively detected by the detection ACPS (8, 55). The detected spin wave signals were then amplified using a low-noise amplifier (29-dB gain) and subsequently passed into a sampling oscilloscope. The waveforms were averaged 10,000 times by the oscilloscope to improve the signal-to-noise ratio, followed by subtraction of reference signals to obtain the pure wave packets. To obtain the additional damping constant, vector network analyzer–based FMR measurements were performed by a frequency–swept technique with the same devices used in the spin pumping measurements.

**Simulations**

We used Object-Oriented Micromagnetic Framework developed by M. J. Donahue and D. G. Porter (see http://math.nist.gov/oommf/) to simulate spin waves by solving the Landau-Lifshitz-Gilbert equation. The magnetic film structure used in our simulations consists of two Py layers (top and bottom layer) with different anisotropy axes. The anisotropy axis was tilted 0.4° away from the $z$ axis toward the $−y$ direction for thick $t_{\text{Ta}}$ and toward the $+y$ direction for thin $t_{\text{Ta}}$. The value of the uniaxial anisotropy constant ($K_u$), saturation magnetization ($M_s$), and exchange stiffness constant ($A$) were 7.0 $\times$ 10$^5$ J/m$^3$, 8.0 $\times$ 10$^5$ A/m, and 1.3 $\times$ 10$^{-11}$ J/m, respectively. The Gilbert damping constant ($\alpha$) and the gyromagnetic ratio ($\gamma$) in our simulations were set to 0.01 and 2.32 $\times$ 10$^8$ m/(A s), respectively. The dimensions of the top and bottom layers were $8000 \times 200 \times 16$ nm$^3$ and $8000 \times 200 \times 4$ nm$^3$, respectively, and the simulation cell size was $4 \times 200 \times 4$ nm. To excite spin wave dynamics, a magnetic field pulse of amplitude 3 mT is applied to a volume of $200 \times 200 \times 20$ nm$^3$ at a distance of 1 $\mu$m from one end of the Py strip, and the propagating spin waves were detected by the summing of the dynamic magnetization over a volume of $800 \times 200 \times 20$ nm$^3$ at 5 $\mu$m away from the excitation source. The out-of-plane bias field was varied from −500 to +500 mT during our simulations.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/2/7/e1501892/DC1

section S1. Nonreciprocal ratios of surface spin wave mode with in-plane fields

section S2. Nonreciprocity ratios from spin wave amplitudes in the time domain

section S3. Nonreciprocity from spin wave intensity in the frequency domain

section S4. Anisotropic magnetoresistance measurements

section S5. VSM measurements

section S6. Simulations: Out-of-plane hysteresis and nonreciprocity

section S7. Antenna–spin wave coupling

section S8. Spin wave amplitude–dependent damping

section S9. Ta on the top of the Py layer

section S10. Dependence of nonreciprocity on different directions of field sweep

section S11. Dependence of nonreciprocity on the sign of the wave vector

section S12. Effect of antenna-to-antenna distance on nonreciprocity

fig. S1. Nonreciprocal spin wave emission in MSSW mode.

fig. S2. Nonreciprocity ratios in the time domain.

fig. S3. FFT of spin wave signals.

fig. S4. AMR measurements.

fig. S5. VSM measurements.

fig. S6. Out-of-plane hysteresis and nonreciprocity from simulations.

fig. S7. Spatial and temporal magnetization excited by the antenna.

fig. S8. Ta thickness dependence of spin wave resonance frequencies.

fig. S9. Nonreciprocity in devices with an Ta overlayer.
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