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## Spin-Current-Driven Permeability Variation for Time-Varying Magnetic Metamaterials

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2	Magnetic Metamaterials
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#### Abstract

We study permeability  $(\mu)$  variation of a lithographically-prepared magnetic meta-atom consist-15 ing of Ta/Py/Pt trilayers by means of spin-torque ferromagnetic resonance (ST-FMR). With an 16 injection of a direct current up to  $\pm 20$  mA together with an alternating current in GHz frequencies, 17 the meta-atom under external magnetic fields shows significant changes in position and width of 18 ST-FMR signals due to a massive spin-current injection. This leads to the spin-current driven 19  $\mu$  variation, which is verified by analytical calculation based upon the experimentally obtained 20 resonance field and Gilbert damping parameter. The present study paves a way to time-varying 21 spintronic metamaterials with a high modulation frequency for realizing microwave sources toward 22 the post-5th generation mobile communication system. 23

#### 24 I. INTRODUCTION

Metamaterials are man-made structures, exhibiting exotic optical properties unavailable 25 in natural materials. The most paradigmatic examples are metamaterials with negative re-26 fractive indices [1] and for invisible cloaks [2]. These are referred to as space-varying metama-27 terials because the refractive indices are modulated in space. Very recently, a paradigm for 28 wave generation and manipulation has emerged using time-varying metamaterials, whose 29 refractive indices are modulated in time [3]. The broken time-translation symmetry at 30 the temporal boundary brings about the change in the refractive index while leaving the 31 wavevector unchanged. As a consequence of the symmetry breaking, frequencies of electro-32 magnetic waves have to change. Therefore, a long-term vision of time-varying metamaterials 33 is achievement of the frequency leap by nonlinear up/down-conversion due to high modula-34 tion frequencies and large efficiencies. 35

Temporal modulation of the refractive index has been investigated so far using a variety of techniques; for example, commercially available electro-optical modulators, variable capacitance (varactor) diodes, photoexcited carrier generation in epsilon-near-zero materials [4, 5], plasma mirrors [6], photonic crystal nanocavities [7], structural-dispersion switching of waveguides [8], and microelectromechanical systems [9]. These techniques are based on

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electric permittivity ( $\varepsilon$ ) modulation. Similarly, modulation of magnetic permeability ( $\mu$ ) is anticipated because  $\mu$  is the counterpart of  $\varepsilon$  in the refractive index  $n = \sqrt{\varepsilon}\sqrt{\mu}$ . Nevertheless,  $\mu$  modulation for time-varying metamaterial is lacking. The simultaneous modulation of both  $\varepsilon$  and  $\mu$  is essential for realizing intriguing physical phenomena such as Fresnel drag effects for light without actual material motions [10].

In an electrical circuit analogue,  $\varepsilon$  modulation is assigned to a change in the capacitance, 46 while  $\mu$  modulation corresponds to an inductance variation. As inductance is relevant to 47 turn numbers, length, and cross section of coils, time-varying  $\mu$  intuitively seems to be 48 a tough challenge. However,  $\mu$  can be varied using magnetic materials. In particular, 49 ferromagnetic materials have frequency dispersion of  $\mu$  at a high frequency, i.e., in the 50 vicinity of ferromagnetic resonance (FMR) frequencies in the GHz region. Moreover, a large 51 magnetization, which originates from microscopic electron spins coupled in the ferromagnetic 52 materials, causes a large variation in  $\mu$ . Therefore, a significant  $\mu$  modulation at GHz 53 frequencies is achieved if the FMR conditions are modified somehow. 54

The FMR frequency is determined by effective magnetic fields and the gyromagnetic ra-55 tio  $\gamma$ , which is governed by the g-value [11–15]. Whereas  $\gamma$  is dependent on the magnetic 56 materials and structures, the effective magnetic fields are controllable and changed exter-57 nally. A key issue to be addressed is how the effective magnetic fields are changed in time. 58 A possible way is a straightforward one with time-varying external magnetic fields applied 59 by an electromagnet. However, the external magnetic fields is unlikely to be switched on 60 and off at GHz frequencies. An alternative option is the utilization of magnetic fields due 61 to a spintronic phenomenon, named spin-current, realized by the spin-Hall effect in heavy 62 metals with large spin-orbit interaction, for example, tantalum (Ta) and platinum (Pt). The 63 spin-current injection at GHz frequencies from Pt into ferromagnetic metal, like permalloy 64  $(Fe_{80}Ni_{20}; Py)$ , results in a change in the magnetic fields for FMR, that is to say, spin-torque 65 FMR (ST-FMR). In ST-FMR, the FMR frequency is varied by the spin-current amplitude. 66 Moreover, the spin-current injection is likely to cause a change in the FMR lineshape, repre-67 sented by the phenomenological Gilbert damping parameter  $\alpha$ . The ST-FMR has intensively 68 been investigated in the field of spintronics [16–21]. Contrastingly,  $\mu$  variation due to an 69 effective magnetic field change by the spin-current injection has yet to be explored. 70

Here, we experimentally study spin-current driven  $\mu$  variation in lithographically-prepared magnetic microstructures consisting of Ta/Py/Pt trilayers as magnetic meta-atoms, which

correspond to elementary components of magnetic metamaterials and determines primarily 73 the metamaterials properties. An alternating (AC) current at GHz frequencies is injected 74 to the microstructure under direct current (DC) magnetic fields to launch ST-FMR in the 75 Py layer. In addition, a DC current is injected to the microstructure to modify the effective 76 magnetic fields and the resonance condition. As the DC current increases, the ST-FMR sig-77 nal shows a significant shift and narrowing due to massive spin-current injection. The shift 78 and narrowing lead to the spin-current driven  $\mu$  variation, which is verified by analytical 79 calculation. These results represent a major step forward in the integration of metamaterials 80 and spintronics. Furthermore, when magnetic meta-atoms are assembled into metamaterials 81 and the DC current is replaced by yet another AC current at GHz frequencies, time-varying 82  $\mu$  metamaterials can be realized. The present study thus paves a way to time-varying spin-83 tronic metamaterials with a high modulation frequency in realizing frequency translation of 84 microwave for the post-5th generation (post-5G) mobile communication system. 85

#### **11. THEORY OF PERMEABILITY VARIATION**

In the following, letters in bold font styles denote vectors and normal style letters denote scalars. Consider magnetization M of a spherical ferromagnet with an effective magnetic field  $H_{\text{eff}}$  in the magnet. The equation of motion of M is expressed as

$$\frac{d\boldsymbol{M}}{dt} = -\gamma \mu_0 \boldsymbol{M} \times \boldsymbol{H}_{\text{eff}} + \frac{\alpha}{M_{\text{s}}} (\boldsymbol{M} \times \frac{d\boldsymbol{M}}{dt}), \qquad (1)$$

where  $\mu_0$  is the permeability of vacuum, and  $M_s$  is the saturation magnetization [22]. Equation (1) is referred to as the Landau-Lifshitz-Gilbert equation. Suppose that an external DC magnetic field  $H_{\text{ext}}$ , enough to saturate magnetization and obtain  $M_s$ , is applied to the sphere. Because there is no demagnetization fields in the sphere,  $H_{\text{eff}}$  is identical to  $H_{\text{ext}}$ . The solution of Eq. (1) indicates that the magnetization precesses around the  $H_{\text{ext}}$ -field vector with a precession angular frequency,  $\omega_0 = \gamma \mu_0 H_{\text{eff}} = \gamma \mu_0 H_{\text{ext}}$ , called the Larmor frequency. The precession is damped in a period determined by  $\alpha$ .

The magnetically saturated ferromagnet is then interacted with a weak AC (microwave) magnetic field  $H_{AC}$ ;  $H_{eff} = H_{ext} + H_{AC}$ , where  $|H_{AC}| << H_{ext}$ . The  $H_{AC}$  has a frequency  $\omega$  and a direction perpendicular to the  $H_{ext}$ -field axis,  $H_{ext} \perp H_{AC}$ . Such a field causes a forced precession at  $\omega$  of the magnetization around the  $H_{ext}$ -field axis. When  $\omega$  is swept in



FIG. 1. Schematic illustration of magnetic meta-atom of Ta/Py/Pt trilayer with spin-torque ferromagnetic resonance measurement setup. Inset: photograph of lithographically-prepared magnetic meta-atom.

the GHz region, the energy of a microwave having  $\omega = \omega_0$  is absorbed by the ferromagnet, giving rise to a resonance behavior referred to as FMR.

If the ferromagnetic material is not a sphere,  $H_{\text{eff}}$  is affected by demagnetization fields  $H_{\text{demag}}$  inside the material;  $H_{\text{eff}} = H_{\text{ext}} + H_{\text{demag}} + H_{\text{AC}}$ . The  $H_{\text{demag}}$  brings about a shift in the FMR angular frequency from  $\omega_0$  to  $\omega_{\text{FMR}}$ . Figure 1 illustrates the structure of a magnetic meta-atom in the present study. The meta-atom consists of a ferromagnetic Py thin film sandwiched by heavy metal Ta and Pt thin films. For the ferromagnetic thin film,  $\omega_{\text{FMR}}$  is described by the Kittel's equation [23] as

$$\omega_{\rm FMR} = \gamma \sqrt{\mu_0 H_{\rm FMR} (\mu_0 H_{\rm FMR} + \mu_0 M_{\rm eff})},\tag{2}$$

where the effective magnetization  $\mu_0 M_{\text{eff}}$ , which is similar to  $\mu_0 M_{\text{s}}$ , corresponds to the demagnetization field. In the present experiments,  $\mu_0 H_{\text{ext}}$ , not  $\omega$ , is swept so that FMR is observed at  $\mu_0 H_{\text{ext}} = \mu_0 H_{\text{FMR}}$ .

Frequency dispersions of complex AC magnetic susceptibility  $\chi(\omega)$  with magnetic losses is written using real  $\chi'(\omega)$  and imaginary  $\chi''(\omega)$  parts [24] as

$$\chi(\omega) = \chi'(\omega) - j\chi''(\omega), \qquad (3)$$

where

$$\chi'(\omega) = \gamma M_{\rm eff} \frac{\omega_{\rm FMR}(\omega_{\rm FMR}^2 - \omega^2) + \omega_{\rm FMR}\omega^2\alpha^2}{[\omega_{\rm FMR}^2 - \omega^2(1 + \alpha^2)]^2 + 4\omega_{\rm FMR}^2\omega^2\alpha^2},\tag{4a}$$

$$\chi''(\omega) = \gamma M_{\text{eff}} \frac{\alpha \omega [\omega_{\text{FMR}}^2 - \omega^2 (1 + \alpha^2)]}{[\omega_{\text{FMR}}^2 - \omega^2 (1 + \alpha^2)]^2 + 4\omega_{\text{FMR}}^2 \omega^2 \alpha^2},\tag{4b}$$

and -j is the imaginary unit. To relate H and magnetic flux density B, we introduce complex relative magnetic permeability with magnetic losses,  $\mu_{\rm r}(\omega)$ , and have

$$B = \mu_0 [1 + \chi(\omega)] H = \mu_0 \mu_r(\omega) H.$$
(5)

In this way,  $\mu_r(\omega)$  is written using real  $\mu'_r(\omega)$  and imaginary  $\mu''_r(\omega)$  parts as

$$\mu_{\mathbf{r}}(\omega) = 1 + \chi'(\omega) - j\chi''(\omega) = \mu_{\mathbf{r}}'(\omega) - j\mu_{\mathbf{r}}''(\omega).$$
(6)

In the present experiments in the following, we vary  $\mu_{\rm r}(\omega)$  by injecting DC current into 117 the meta-atom. The DC current modifies  $\omega_{\text{FMR}}$  and  $\alpha$  in Eq. (4). Using Eq. (2),  $\omega_{\text{FMR}}$  is 118 evaluated from  $\mu_0 H_{\text{FMR}}$ , which is experimentally obtained from the ST-FMR measurements. 119 Additionally, the ST-FMR measurements give values of  $\alpha$  and  $\mu_0 M_{\text{eff}}$ . Therefore complex 120  $\mu_{\rm r}(\omega)$  is analytically calculated using  $\mu_0 H_{\rm FMR}$ ,  $\alpha$  and  $\mu_0 M_{\rm eff}$  with Eqs. (4) and (6). More 121 importantly, the present study demonstrates that the spin-current injection induced by the 122 DC current modifies effective magnetic fields in the Py layer, leading to variations in  $\mu_0 H_{\text{FMR}}$ 123 and  $\alpha$ . From these variations, we evaluate how much  $\mu_{\rm r}(\omega)$  is varied by the spin-current 124 injection. 125

#### 126 III. EXPERIMENTAL METHOD

Heavy metal layers of Ta and Pt, and a ferromagnetic metal layer of Py are deposited 127 onto non-doped Si substrates at room temperature using magnetron sputtering with an 128 argon (Ar) gas pressure of 6  $\times 10^{-1}$  Pa. A 3 nm thick Ta layer as a buffer layer is deposited 129 on the substrates first, after which a Py layer of 2 nm and a Pt layer of 5 nm thickness are 130 subsequently deposited. The Ta buffer layer is mainly for better adhesion and flatness of 131 the Py layer on the substrate. The magnetization of the trilayer film is measured using a 132 vibrating sample magnetometer (VSM). The trilayer film is then patterned into a rectangular 133 shape of 5 µm width and 60 µm length using electron beam lithography (EBL) and Ar ion 134 etching. The contact electrodes consisting of 5 nm thick Cr and 200 nm thick Au layers 135

are prepared on the both sides of the rectangle using EBL and lift-off processes as shown in
Fig. 1. The inset in Fig. 1 shows a photograph of the prepared magnetic meta-atom. The
meta-atom length between the electrodes is 24 µm.

Figure 1 shows also a schematic illustration of the ST-FMR measurements [16]. An in-139 plane external DC magnetic field  $H_{\text{ext}}$  is applied using an electromagnet with a relative angle 140  $\theta = 45^{\circ}$  to the y-axis. An AC current  $I_{\rm AC}$  at microwave frequencies in a range between 2 and 141 9 GHz is injected from a signal generator into the meta-atom along the y-axis. The signal 142 generator's power of  $I_{\rm AC}$  is -1 dBm. The  $I_{\rm AC}$  induces an oscillating Oersted magnetic field 143  $H_{\rm AC}$  with the Ampere's law, primarily driving FMR of the Py magnetization. Moreover, 144 the  $I_{\rm AC}$  flowing in Pt and Ta layers generates vertical AC spin currents by the spin-Hall 145 effect. 146

The direction of the spin-current is expressed by  $J_{\rm spin} \parallel \theta_{\rm SH}(\hat{\boldsymbol{\sigma}} \times \boldsymbol{J}_{\rm charge})$ , where  $J_{\rm spin}$  is 147 the spin-current density,  $\theta_{\rm SH}$  is the spin-Hall angle,  $\hat{\sigma}$  is the Pauli spin matrix, and  $J_{\rm charge}$ 148 is the charge current density. This equation indicates that the direction of  $J_{\rm spin}$  induced by 149  $J_{\text{charge}}$  depends on  $\theta_{\text{SH}}$ . Because  $\theta_{\text{SH}}$  of Pt and Ta have the opposite signs [25], identical spin 150 moments are injected to the Py layer from both the top Pt and bottom Ta layers. The  $J_{\rm spin}$ 151 from the Pt layer is, however, much larger than that from the Ta layer because of large  $J_{\text{charge}}$ 152 in the Pt layer due to small electrical resistance compared to the Ta layer [26, 27]. Moreover, 153 as the Ta layer deposited on the substrate is likely to be oxidized, the electrical resistance of 154 the Ta layer is much larger, bringing about tiny  $J_{\text{charge}}$  in the Ta layer. Thus the spin-current 155 injection only from the Pt layer is considered in the following. The spin angular momentum 156 injected from the Pt layer is transferred to the in-plane Py magnetization, exerting a field-157 like torque (FLT) that secondary drives magnetization precession and damping-like torque 158 (DLT) that enhances or reduces magnetization relaxation. In this way,  $I_{\rm AC}$  gives rise to an 159 oscillating  $H_{\rm AC}$ , FLT, and DLT, which drive ST-FMR in the Py layer. 160

<sup>161</sup> Under the ST-FMR, the magnetization precession causes a resistance oscillating at the <sup>162</sup> frequency of  $I_{AC}$  due to the anisotropic magnetoresistance (AMR) of Py. By mixing  $I_{AC}$ <sup>163</sup> and the oscillating AMR, a time-independent longitudinal DC voltage  $V_{AMR}$  arises along <sup>164</sup> the *y*-axis. We measure  $V_{AMR}$  as a function of  $\mu_0 H_{ext}$  using a bias tee [16, 28] to observe <sup>165</sup> magnetization dynamics by ST-FMR and obtain  $\mu_0 H_{FMR}$  and  $\alpha$ .

Variations in  $\mu_0 H_{\rm FMR}$  and  $\alpha$  is achieved by injecting a DC current  $I_{\rm DC}$  together with  $I_{\rm AC}$  into the meta-atom. The  $I_{\rm DC}$  direction is parallel to that of  $I_{\rm AC}$ . The  $I_{\rm DC}$  leads to

an time-independent Oersted DC field  $H_{\text{Oe}}$  along  $\pm x$ -axis. Furthermore,  $I_{\text{DC}}$  generates a 168 vertical DC spin-current in the Pt layer, giving rise to time-independent FLT and DLT on 169 the Py magnetization as shown in Fig. 1. The FLT and DLT are regarded as internal 170 magnetic fields  $H_{\rm spin}$  [29, 30]. The  $H_{\rm eff}$  acting on the magnetization can thus be given by 171  $H_{\text{eff}} = H_{\text{ext}} + H_{\text{demag}} + H_{\text{Oe}} + H_{\text{spin}}$  as depicted in Fig. 1 when  $H_{\text{AC}}$  and crystalline magnetic 172 anisotropy are ignored. The  $I_{\rm DC}$  injection causes variations in  $H_{\rm Oe}$  and  $H_{\rm spin}$ , bringing about 173 a change in  $\mu_0 H_{\text{ext}}$  for FMR, i.e., a shift in  $\mu_0 H_{\text{FMR}}$ . Furthermore, the time-independent 174 DLT influences  $\alpha$ . When DLT has the same direction with the magnetization precession 175 damping, the effective  $\alpha$  is enhanced. Contrastingly, the effective  $\alpha$  becomes small when the 176 DLT direction is opposite to the damping. In the present experiments,  $V_{\rm AMR}$  signals with 177 various  $I_{AC}$  from 2 to 9 GHz and  $I_{DC}$  from -20 mA to +20 mA are measured as a function 178 of  $\mu_0 H_{\text{ext}}$  in a range between +150 mT and -150 mT. All measurements are carried out at 179 room temperature. 180

#### 181 IV. RESULTS AND DISCUSSION

#### A. Spin-torque ferromagnetic resonance signals with alternating electric current

Figure 2(a) shows a typical  $V_{\text{AMR}}$  signal by ST-FMR measured with 6 GHz  $I_{\text{AC}}$  and zero  $I_{\text{DC}}$ . The red open circles correspond to measurement results. The  $V_{\text{AMR}}$  signal in thin films is expressed as  $V_{\text{AMR}} = V_{\text{S}} + V_{\text{A}}$  [16, 17, 28], where

$$V_{\rm S} = S \frac{(\mu_0 \Delta_{\rm FMR})^2}{(\mu_0 H_{\rm ext} - \mu_0 H_{\rm FMR})^2 + (\mu_0 \Delta_{\rm FMR})^2},$$
(7a)

$$V_{\rm A} = A \frac{\mu_0 \Delta_{\rm FMR} (\mu_0 H_{\rm ext} - \mu_0 H_{\rm FMR})}{(\mu_0 H_{\rm ext} - \mu_0 H_{\rm FMR})^2 + (\mu_0 \Delta_{\rm FMR})^2}.$$
 (7b)

The  $\mu_0 \Delta_{\text{FMR}}$  is the half width at half maximum of the FMR signal. The S in Eq. (7a) is 183 a symmetric Lorentzian coefficient that is proportional to DLT, while A in Eq. (7b) is an 184 anti-symmetric Lorentzian coefficient that is proportional to the oscillating  $H_{\rm AC}$  and FLT. 185 The measured  $V_{\text{AMR}}$  is fitted using Eqs. (7). The green and blue solid lines in Fig. 2(a) 186 correspond to  $V_{\rm S}$  and  $V_{\rm A}$ , respectively. The sum of  $V_{\rm S}$  and  $V_{\rm A}$  is represented by the black 187 solid line, which reproduces well the experimental data. After the fitting, the  $V_{\rm S}$  signal as 188 well as  $V_{\rm A}$  signal gives the resonance field  $\mu_0 H_{\rm FMR} = 58.4$  mT and signal width  $\mu_0 \Delta_{\rm FMR} =$ 189 10.3 mT. 190



FIG. 2. (a) Measured spin-torque ferromagnetic resonance signal,  $V_{AMR}$ , as a function of external magnetic field,  $\mu_0 H_{ext}$ , at 6 GHz without direct electric current (red circles). Green and blue solid lines represent fitting results with symmetric ( $V_S$ ) and anti-symmetric coefficient ( $V_A$ ), respectively. The black solid line corresponds to the sum of  $V_S$  and  $V_A$ . (b) Red circles: microwave frequency plotted as a function of resonance magnetic field,  $\mu_0 H_{FMR}$ . Solid line: fitting curve by the Kittel equation [Eq. (2)]. Inset: trilayer's saturation magnetic flux density,  $\mu_0 M_s$ , measured by VSM plotted as a function of Py thickness.

<sup>191</sup> The frequency dependence of the  $V_{AMR}$  signals is acquired by keeping  $I_{dc} = 0$  but changing <sup>192</sup> a  $I_{AC}$  frequency from 2 GHz to 9 GHz with intervals of 1 GHz. Figure 2(b) highlights the  $I_{AC}$ <sup>193</sup> frequency versus  $\mu_0 H_{FMR}$  (red open circles). The black solid line in Fig. 2(b) corresponds <sup>194</sup> to a fitting curve by the Kittel equation [Eq. (2)]. Given that the *g*-factor of the Py film is <sup>195</sup> 2.1 [17], the fitting by Eq. (2) gives  $\mu_0 M_{eff} = 658$  mT. As in the inset of Fig. 2(b), VSM <sup>196</sup> measurments show that the Ta/Py/Pt trilayer with 2 nm thick Py has saturation magnetic



FIG. 3. Spin-torque ferromagnetic resonance signals with 6 GHz alternative electric currents measured at various direct electric currents,  $I_{\rm DC}$  from -20 mA to +20 mA with intervals of 2 mA

flux density  $\mu_0 M_s$  of 805 mT. This value is similar to  $\mu_0 M_{eff}$  of 658 mT obtained from 197 the ST-FMR measurements. The small difference between  $\mu_0 M_{\text{eff}}$  and  $\mu_0 M_{\text{s}}$  is probably 198 caused by the Py thickness discrepancy in the two samples. This result indicates that the 199  $V_{\rm AMR}$  signal originates from the uniform precession of electron spins in Py, referred to as the 200 Kittel-mode FMR. Moreover, the inset of Fig. 2(b) shows trilayer film's  $\mu_0 M_s$  plotted as a 201 function of Py thickness of 1, 2, 5, and 10 nm. Note here that  $\mu_0 M_s$  decrease significantly 202 at the trilayer with 1 nm thick Py. The origin of the decrease is most likely the magnetic 203 dead layer at the Py/Pt and Py/Ta interfaces [31]. 204

### B. Spin-torque ferromagnetic resonance signals with alternating and direct electric currents

Figure 3 shows  $V_{\text{AMR}}$  signals of ST-FMR with 6 GHz  $I_{\text{AC}}$  measured with various  $I_{\text{DC}}$ from -20 mA to +20 mA at intervals of 2 mA. A black solid line corresponds to a signal

with  $I_{\rm DC} = 0$  mA, a part of which is already shown in Fig. 2(a). A peak is seen at 70 mT 209 in the  $V_{\text{AMR}}$  signal with  $I_{\text{DC}} = 0$  mA. As  $I_{\text{DC}}$  increases from 0 to +20 mA, the peak at 70 210 mT is getting narrower. Contrastingly, the peak becomes broader as  $I_{\rm DC}$  decreases from 0 211 to -20 mA. When the direction of  $H_{\text{ext}}$  is reversed, the peaks change to dips because of the 212 reversal in the relative angle between  $I_{AC}$  and  $H_{ext}$ . The variation of the dip lineshape at 213 -70 mT is also inverted; the dip becomes broader as  $I_{\text{DC}}$  increases from -20 to +20 mA. 214 The lineshape change depends on both the amplitude and direction of  $I_{\rm DC}$ , indicating that 215 the origin is the Joule heating as well as the spin-current injection and DC Oersted field. 216

The  $V_{\rm AMR}$  signals at each  $I_{\rm DC}$  between -20 to +20 mA are obtained by varying  $I_{\rm AC}$ frequency from 2 to 9 GHz. The signals are analyzed using Eqs. (7) to evaluate  $\mu_0 H_{\rm FMR}$ and  $\mu_0 \Delta_{\rm FMR}$ . In Fig. 4,  $\mu_0 \Delta_{\rm FMR}$  evaluated at various  $I_{\rm DC}$  is plotted as a function of  $I_{\rm AC}$ frequency. Red open circles in Fig. 4 correspond to  $\mu_0 \Delta_{\rm FMR}$  versus  $I_{\rm AC}$  frequency at  $I_{\rm DC}$ = 0 mA. The frequency dependence of the signal linewidth gives the damping parameter  $\alpha$ . The  $\mu_0 \Delta_{\rm FMR}$  is expressed as

$$\mu_0 \Delta_{\rm FMR} = W + (\alpha/\gamma)\omega, \tag{8}$$

where W represents inhomogeneous linewidth broadening due to sample imperfections and is independent of the frequency of  $I_{\rm AC}$ . A red solid line in Fig. 4 is a fitting curve of experimentally evaluated  $\mu_0 \Delta_{\rm FMR}$  at  $I_{\rm DC} = 0$  mA by Eq. (8). Equation (8) indicates that a slope of the fitting curve corresponds to  $\alpha$ . The fitting at  $I_{\rm DC} = 0$  mA gives  $\alpha = 0.045$  $\pm 0.001$ . Figure 4 highlights that, as  $I_{\rm DC}$  increases from -20 to +20 mA, the fitting curve slope becomes smaller, indicating that a larger  $I_{\rm DC}$  brings about a smaller  $\alpha$ .

#### 229 C. Direct electric current dependence of damping parameter and resonance field

In Fig. 5(a), the evaluated  $\alpha$  (open circles) indicated from the left vertical axis is plotted as a function of  $I_{\rm DC}$ . The  $\alpha$  is 0.082 at  $I_{\rm DC} = -20$  mA. As  $I_{\rm DC}$  increases from -20 to +20mA,  $\alpha$  decreases monotonically and reaches the minimum value of 0.025 at  $I_{\rm DC} = +20$  mA. This result clearly shows that  $\alpha$  is controllable by the spin-current as reported previously in Refs. [16–18]. The variation in  $\alpha$  is approximately 0.057, which is comparable to that reported in Ref. [19].

Open squares in Fig. 5(a) also shows a resonance field shift  $\mu_0 H_{\rm FMR}^{\rm shift}(I_{\rm DC})$  by the  $I_{\rm DC}$ 



FIG. 4. Ferromagnetic resonance signal line width,  $\mu_0 \Delta_{\text{FMR}}$ , at various direct electric currents,  $I_{\text{DC}}$ , from -20 mA to +20 mA as a function of  $\omega/2\pi$ . Solid and dashed lines are fitting curves.



FIG. 5. (a)  $\alpha$  (open circles, left axis) and  $\mu_0 H_{\text{FMR}}^{\text{shift}}$  (open squares, right axis) are plotted as a function of  $I_{\text{DC}}$ . (b)  $\mu_0 H_{\text{intrinsic}}^{\text{shift}}$  (open triangles) is plotted as a function of  $I_{\text{DC}}$ .

injection as indicated from the right axis. The  $\mu_0 H_{\rm FMR}^{\rm shift}(I_{\rm DC})$  is defined as  $\mu_0 H_{\rm FMR}(I_{\rm DC}) - \mu_0 H_{\rm FMR}(0)$ , where  $\mu_0 H_{\rm FMR}(I_{\rm DC})$  corresponds to  $\mu_0 H_{\rm FMR}$  with non-zero specific  $I_{\rm DC}$  and  $\mu_0 H_{\rm FMR}(0)$  corresponds to  $\mu_0 H_{\rm FMR}$  with zero  $I_{\rm DC}$ . A positive value of  $\mu_0 H_{\rm FMR}^{\rm shift}$  represents a shift to a higher field. Figure 5(a) demonstrates that  $\mu_0 H_{\rm FMR}^{\rm shift}(I_{\rm DC})$  is  $-2 \,\mathrm{mT}$  at  $I_{\rm DC} = -20$ 

<sup>241</sup> mA and 8 mT at  $I_{\rm DC} = 20$  mA.

As already mentioned,  $\mu_0 H_{\rm FMR}^{\rm shift}(I_{\rm DC})$  is caused by the Joule heating, spin-current injection, and DC Oersted field. The enhanced fluctuations in magnetization by the Joule heating is a major factor in  $\mu_0 H_{\rm FMR}^{\rm shift}$  [32]. The influence of the Joule heating is dependent on the electric current intensity, but independent of the current direction. On the other hand, the influence of the spin-current injection and Oersted field is a odd function of the  $I_{\rm DC}$  direction. The Joule heating contribution can thus be removed by

$$\mu_0 H_{\text{intrinsic}}^{\text{shift}} = \frac{\mu_0 H_{\text{FMR}}^{\text{shift}}(I_{\text{DC}}) - \mu_0 H_{\text{FMR}}^{\text{shift}}(-I_{\text{DC}})}{2},\tag{9}$$

and intrinsic resonance field shift,  $\mu_0 H_{\text{intrinsic}}^{\text{shift}}$ , due to the spin-current injection and Oersted field is obtained.

Figure 5(b) shows  $\mu_0 H_{\text{intrinsic}}^{\text{shift}}$  as a function of  $I_{\text{DC}}$ . The  $\mu_0 H_{\text{intrinsic}}^{\text{shift}}$  increases linearly with 250 an increase in  $I_{\rm DC}$ . The slope is approximately 0.27 mT/mA. Using current density in the 251 sample, this value is transfromed to  $6.25\times10^{-12}~\rm mT/(A/m^2),$  which is similar to  $4.24\times10^{-12}$ 252  $mT/(A/m^2)$  in Ref. [17]. However, the amount of the shift obtained in this study is much 253 larger than that in the previous studies [17, 29, 30, 33]. Thanks to the well-insulated non-254 doped Si substrate with better thermal conductivity [34, 35], we can inject massive  $I_{\rm DC}$  up 255 to 20 mA, which causes the large shift variation of approximately 5 mT while the previously 256 reported one is less than 0.7 mT. 257

#### 258 D. Evaluation of permeability variation

In order to evaluate  $\mu'_{\rm r}(\omega)$  and  $\mu''_{\rm r}(\omega)$  at a specific  $I_{\rm DC}$  value from Eq. (6),  $\chi'(\omega)$  and  $\chi''(\omega)$ 259 should be calculated from Eq. (4) with  $\omega_{\text{FMR}}$ ,  $\alpha$ , and  $M_{\text{eff}}$  experimentally obtained at the 260  $I_{\rm DC}$  value. First,  $\mu'_{\rm r}(\omega)$  and  $\mu''_{\rm r}(\omega)$  of the meta-atom under DC magnetic field of 58.4 mT at 261  $I_{\rm DC} = 0$  mA are calculated from Eqs. (4) and (6). We use  $\omega_{\rm FMR}/2\pi = 6$  GHz,  $\mu_0 M_{\rm eff} = 658$ 262 mT obtained in Fig. 2, and  $\alpha = 0.045$  obtained in Fig. 4. Then, spin-current injection by 263 non-zero  $I_{\rm DC}$  values causes variations of  $\omega_{\rm FMR}/2\pi$  from 6 GHz at  $I_{\rm DC} = 0$  mA as shown in 264 Fig. 5(b) through variation in  $\mu_0 H_{\text{FMR}}$ . The spin-current injection also leads to variations 265 of  $\alpha$  from 0.045 at  $I_{\rm DC} = 0$  mA as shown in Fig. 5(a). The  $I_{\rm DC}$  dependence of  $\alpha$  in Fig. 266 5(a) is thus fitted by a linear function. Similarly, another linear function is acquired by a fit 267 of  $I_{\rm DC}$  dependence of  $\mu_0 H_{\rm intrinsic}^{\rm shift}$  in Fig. 5(b). These fitting functions give  $\alpha$  and  $\mu_0 H_{\rm FMR}$  at 268



FIG. 6. 2D plot of (a)  $\mu'_{\rm r}$  and (b)  $\mu''_{\rm r}$  evaluated using experimentally obtained  $\alpha$  and  $\mu_0 H_{\rm FMR}$  of meta-atom under DC magnetic field of 58.4 mT as a function of  $\omega/2\pi$  and  $I_{\rm DC}$ . Black dashed lines indicate  $\omega/2\pi = 5.74$  GHz. (c)  $I_{\rm DC}$  versus  $\mu'_{\rm r}$  (red solid line) and  $\mu''_{\rm r}$  (blue dashed line) at 5.74 GHz indicated by black dashed lines in (a) and (b).

a specific  $I_{\rm DC}$  value. Given that  $I_{\rm DC}$  do not influence the Kittel formula of Eq. (2),  $\mu_0 M_{\rm eff}$ = 658 mT, and  $\gamma = 1.87 \times 10^{-11}$  Hz/T,  $\mu_0 H_{\rm FMR}$  is converted to  $\omega_{\rm FMR}$  at each  $I_{\rm DC}$  from Eq. (2). Finally, from Eqs. (4) and (6) using  $\omega_{\rm FMR}$ ,  $\alpha$ , and  $M_{\rm eff}$ ,  $\mu'_{\rm r}(\omega)$  and  $\mu''_{\rm r}(\omega)$  at each  $I_{\rm DC}$ are analytically obtained.

Figures 6(a) and 6(b) respectively show 2D plots of  $\mu'_{\rm r}$  and  $\mu''_{\rm r}$  of the meta-atom under DC magnetic field of 58.4 mT as a function of  $\omega/2\pi$  and  $I_{\rm DC}$ . In Fig. 6(a), red color in the plot is assigned to a positive value of  $\mu'_{\rm r}$ , whereas blue is assigned to a negative value. In Fig. 6(b),  $\mu''_{\rm r}$  varies from 0 (blue) to 70 (red). The vertical cross-sections of the 2D plots correspond to frequency dispersions of  $\mu'_{\rm r}$  and  $\mu''_{\rm r}$  at a specific  $I_{\rm DC}$  value. Figures 6(a) and 6(b) shows that a larger  $I_{\rm DC}$  brings about a larger variation in  $\mu'_{\rm r}$  and  $\mu''_{\rm r}$ .

Figure 6(c) highlights  $I_{\rm DC}$  versus  $\mu'_{\rm r}$  (red line) and  $\mu''_{\rm r}$  (blue line) at 5.74 GHz, which is

indicated by black dashed lines in Figs 6(a) and 6(b). In Fig. 6(c),  $\mu'_{\rm r}$  at 5.74 GHz shows a small value of 0.4 at  $I_{\rm DC} = -20$  mA, increases with  $I_{\rm DC}$ , and exhibits a maximum value of 20.0 at  $I_{\rm DC} = +8.5$  mA. After the maximum value,  $\mu'_{\rm r}$  slightly decreases to 17.8 at  $I_{\rm DC}$ = +20 mA. Contrastingly,  $\mu''_{\rm r}$  at 5.74 GHz has a large value of 21.2 at  $I_{\rm DC} = -20$  mA. The  $\mu''_{\rm r}$  shows a maximum value of 23.3 at  $I_{\rm DC} = -12$  mA, and decreases down to 3.6 at  $I_{\rm DC} =$ +20 mA. The large variation in  $\mu'_{\rm r}$  and  $\mu''_{\rm r}$  is primarily caused by the massive  $I_{\rm DC}$  up to  $\pm$ 20 mA.

Let us consider a spin-current driven frequency up-conversion of a microwave having 5.74 287 GHz by the time-varying magnetic meta-atom under DC magnetic field of 58.4 mT. An AC 288 current at 5.74 GHz as a carrier wave flows through the meta-atom. Instead of the DC 289 current, another AC current having an amplitude of 20 mA and a frequency that is lower 290 than 5.74 GHz, for example, 1 GHz, is also injected to generate a magnetic field by the 291 spin-current injection for modulation. If a simple phase modulation is supposed, the 5.74 292 GHz carrier microwave in the meta-atom "feels" time-varying  $\mu'_r$  between 0.4 and 17.8 at a 293 modulation frequency of 1 GHz, leading to a creation of a modulated microwave having 6.74 294 GHz. The up-converted 6.74 GHz microwave can be radiated from an antenna connected to 295 the meta-atom. Furthermore, in addition to the phase modulation, amplitude modulation 296 due to time-varying  $\mu_{\rm r}''$  between 3.6 and 21.2 is likely and may realize a large modulation 297 amplitude. Because the variation in  $\mu'_{\rm r}$  and  $\mu''_{\rm r}$  is not a linear function of  $I_{\rm DC}$  as in Fig. 6(c), 298 a modulation frequency at 1GHz gives rise to  $\mu'_r$  and  $\mu''_r$  variation at a much higher frequency. 299 Last but at least, it is worth mentioning that natural antiferromagnets, for example, NiO 300 [36], and artificial antiferromagnetic structures, for example, Co/Ru multilayers, are possible 301 candidates for permeability modulation at much higher frequencies in millimeter wave and 302 THz light regions. 303

#### 304 V. CONCLUSION

We experimentally demonstrate spin-torque ferromagnetic resonance (ST-FMR) of a lithographically-prepared magnetic meta-atom consisting of Ta/Py/Pt trilayer thin films on non-doped Si substrates. The ST-FMR in the Py layer is performed by injecting a large direct electric current up to  $\pm 20$  mA together with an alternating electric current at GHz frequencies to the meta-atom under DC magnetic fields. Thanks to the well-insulated

substrates with a better thermal conductivity, the massive spin-current injection gives rise 310 to significant changes in damping parameter  $\alpha$ , and FMR field  $\mu_0 H_{\rm FMR}$ ;  $\alpha$  becomes ap-311 proximately 1/3 and the maximum amount of shift of  $\mu_0 H_{\rm FMR}$  is 5 mT. From analytical 312 calculation based on the experimentally obtained  $\alpha$  and  $\mu_0 H_{\rm FMR}$ , we verify large variations 313 in the real part of permeability from 0.4 to 17.8 and the imaginary part of permeability 314 from 3.6 to 21.2 at 5.74 GHz for the meta-atom under DC magnetic field of 58.4 mT. The 315 spin-current driven magnetic permeability variation can be realized by combining a mag-316 netic Py layer with heavy-metal Pt and Ta layers, thus highlighting a major step forward 317 in the integration of metamaterials with spintronics. The present study opens a door for an 318 avenue of time-varying spintronic metamaterials with a GHz modulation frequency. 319

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