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独立行政法人 日本学術振興会 理事長　殿

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（氏名は必ず自署すること）

海外特別研究員としての派遣期間を終了しましたので、下記のとおり報告いたします。
なお、下記及び別紙記載の内容については相違ありません。

記

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2. 研究課題名（和文）※研究課題名は申請時のものと違わないように記載すること。
反強磁性体/強磁性体ヘテロ接合におけるスピンダイナミクスの測定とその制御

3. 派遣期間：平成　29　年　4　月　14　日　～　平成　30　年　9　月　7　日

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5. 所期の目的の遂行状況及び成果…書式任意　**書式任意（A4 判相当 3 ページ以上、英語で記入も可）**
（研究・調査実施状況及びその成果の発表・関係学会への参加状況等）
（注）「6. 研究発表」以降については様式 10－別紙 1～4 に記入の上、併せて提出すること。

1. Background

Spintronic logic circuit utilising pure spin currents have been proposed and attracting much attention because of its advantageous characteristics such as nonvolatility, low power consumption, and high-speed operation. Because spin currents carry information in such a device, a medium in which spin currents propagate without dissipation is highly desired. Recently, antiferromagnetic NiO thin films have been intensively studied as media for the effective propagation of spin currents.[1,2] In such previous studies, precessional spin pumping has been used to generate spin currents, while spin currents are detected mostly by the inverse spin Hall effect (ISHE). Although those studies demonstrated spin current propagation through NiO thin films, the study of spin current within NiO is still incomplete because the ISHE has only been used to probe the “dc” spin current propagating through the NiO, while the “ac” spin current has not been measured. The ISHE may not be able to directly detect the ac spin current due to an electric parasitic effect. Another technique is required to study an ac spin current within NiO.

We propose to use the X-ray detected ferromagnetic resonance (XFMR) technique to study the propagation of ac spin current within NiO. XFMR, combining X-ray magnetic circular dichroism (XMCD) and ferromagnetic resonance (FMR), is a unique probe to study element-specific spin dynamics. In a typical XFMR measurement, the magnetisation of a ferromagnet is pumped by an rf field to generate a precessing moment (i.e., FMR) and then probed by the XMCD effect, where one can extract amplitude and phase of spin precession in a ferromagnet. This allows a spin current to be detected via the spin transfer torque exerted upon a ferromagnet in a ferromagnet/spacer/ferromagnet multilayer structure.[3] In fact, we previously detected an ac spin current propagating through a nonmagnetic metal spacer in CoFe/Cu/NiFe and NiFe/CuMn/Co multilayer structures by XFMR measurement.[4,5]

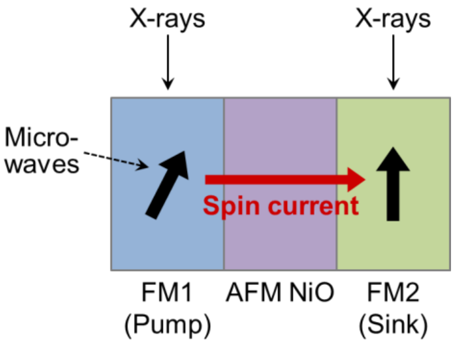

Fig. 1. Schematic of XFMR measurement of a ferromagnet/antiferromagnetic NiO/ferromagnet (FM/AFM NiO/FM) multilayer structure. FM1 (FM2) is a pump (sink) layer.

In this study, time-resolved element-specific XFMR measurements were performed upon a ferromagnet/antiferromagnetic NiO/ferromagnet (FM/AFM NiO/FM) multilayer structure. As shown in Fig.1, our scenario how to detect the spin current through NiO is as follows: (1) Microwave excites FMR in FM1 (pump layer); (2) FM1 injects an spin current into AFM NiO; (3) AFM NiO mediates the spin current towards FM2 (sink layer); (4) The resulting STT excites an off-resonance precessional response in FM2; (5) During these processes, the response of both FM is quantified by the XMCD effect. In accordance with this scenario, we performed the XFMR measurements upon a CoFe/NiO/Fe/NiFe multilayer.

2. Experimental method

Epitaxial MgO(001) substrate/MgO (5)/$Co_{25}Fe_{75}$ (5)/NiO (4, 6, 12)/Fe (1)/$Ni_{80}Fe_{20}$ (25)/MgO (3) (thicknesses in nm), structures were grown prior to the beamtime experiments by molecular beam epitaxy. The Fe/NiFe bilayer corresponds to a pump layer (FM1 in Fig. 1), while the CoFe layer does to a sink layer (FM2 in Fig. 1). The surface structure of each layer was monitored by low energy electron diffraction (LEED) in the MBE chamber. The samples were either in-situ magnetised along a cube edge of the CoFe before the NiO growth or ex-situ field-cooled from above the Néel temperature of NiO (252°C) with the field applied in this direction. We represent this direction as H_M in this study. Depending upon the preferred relative alignment of magnetic moments at the FeCo/NiO and NiO/Fe interfaces, these two methods may not necessarily yield the same results, because in one case only one of the ferromagnetic layers is present during the training of the NiO. However, in this study, both methods resulted in very similar hysteresis loops of the samples. Thus, we believe that all the samples are comparable in the magnetisation alignment. We carried out XFMR measurements on beamline 4.0.2 at the Advanced Light Source and beamline (ALS) and I10 at the Diamond Light Source (DLS). Figure 2(a) and 2(b) show a block diagram and the sample geometry, respectively, for the XFMR measurement.[3,5] Microwaves of 4 or 6 GHz frequency, and variable phase relative to the X-ray pulses, were supplied through a coplanar waveguide (CPW) so as to generate an

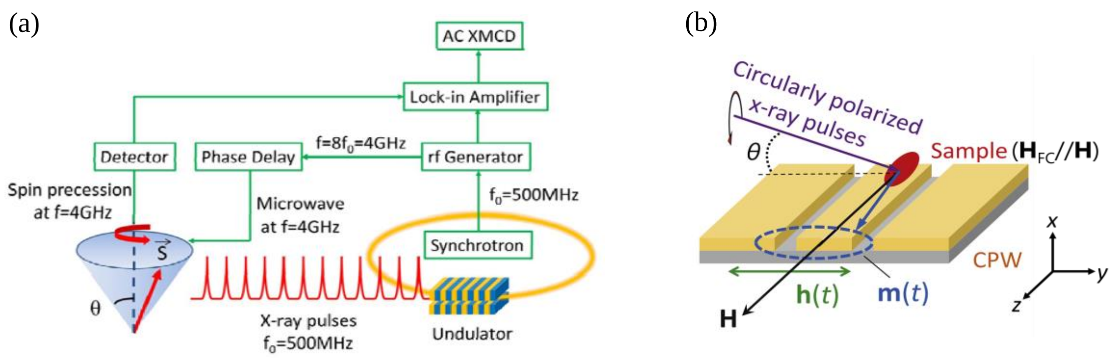

Fig. 2. (a) Block diagram and (b) sample geometry of XFMR measurement. In (b), H, $h(t)$, $m(t)$, and θ are the static bias field, rf field, dynamic magnetisation, and grazing angle, respectively. (a) and (b) were adapted from Ref. 4 and 3, respectively.

in-plane rf magnetic field at the overlaid sample, with direction orthogonal to the axis of the CPW, while an in-plane static magnetic field of magnitude H was applied parallel to the axis. X-ray pulses with grazing angle of 35° to the sample surface were incident through a hole in the CPW, so that X-rays transmitted through the sample stack can generate luminescence in the MgO substrate that was detected with a photodiode. All measurements were performed at room temperature.

3. Experimental results

Figure 3(a) shows an estimated epitaxial relationship between the CoFe and NiO layers in the sample. The NiO lattice rotates in plane by 45° with respect to the CoFe lattice so as to reduce the lattice mismatch. Because of the similarity in the lattice constants, the MgO (Fe) layer shows the same crystalline structure as that of the NiO (CoFe) layer: The epitaxial relationship between each layer in the multilayer structure should be MgO [100]//CoFe [110]//NiO [100]//Fe [110]. Figure 3(b)-3(e) show LEED patterns from the MgO buffer layer to the ultrathin Fe layer in the sample with 4-nm-thick NiO. The clear spots in the LEED patterns confirm the single crystalline structure of each layer up to the Fe layer in the sample. The epitaxial FM/AFM NiO/FM multilayer structure was successfully prepared, which should be important for efficient spin transport through the FM/AFM interfaces. Note that the NiFe layer was polycrystalline. This is not a problem in this experiment since the NiFe layer was grown on the Fe layer in order to reduce the Fe layer's FMR frequency while the NiO/Fe interface is epitaxial as mentioned above.

Then we measured the static X-ray absorption spectra (XAS) and XMCD across the Co and Ni $L_{2,3}$ edges. Note that Co and Ni atoms are contained within the CoFe sink layer and the NiFe pump layer, respectively. The nonzero XMCD signals at the Co and Ni $L_{2,3}$ edges clearly identify the ferromagnetic state of the CoFe and NiFe layers (not shown here). The X-ray energy was then tuned to the Co and Ni L_3 edges in order to measure the element-specific hysteresis loops, as shown in Fig. 4(a) and 4(b) for the sample with 4-nm-thick and 6-nm-thick NiO layer, respectively. Note that the experimental condition is not the same for these two samples: The external magnetic field was applied orthogonal (parallel) to the H_M direction in the sample with 4-nm-thick (6-nm-thick) NiO layer. This is just due to the limit of beamtime.

In the sample with 4-nm-thick NiO layer, the Co and Ni spins, i.e., the CoFe and NiFe layers showed almost the same switching behaviours. This indicates that there is a strong interlayer exchange coupling between the CoFe and NiFe layers through the NiO layer. In the sample with 6-nm-thick NiO layer, the NiFe layer showed a typical square hysteresis loop. The NiFe magnetisation directly reverses from one direction to the opposite direction at around ±50 Oe. On the other hand, the CoFe layer showed plateaus after abrupt changes at around ±50 Oe. This behaviour can be interpreted as follows. When the applied

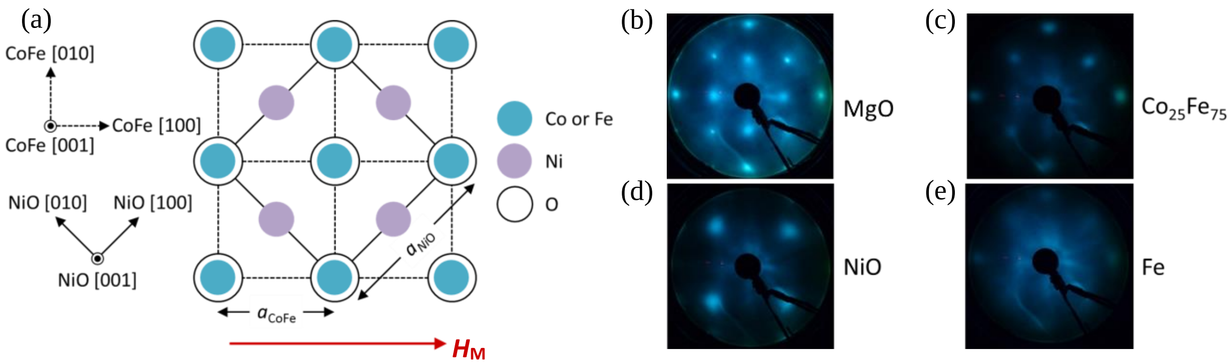

Fig. 3. (a) Epitaxial relationship between CoFe and NiO lattices. (b)-(e) LEED patterns of each layer from the MgO buffer layer to the ultrathin Fe layer in the sample.

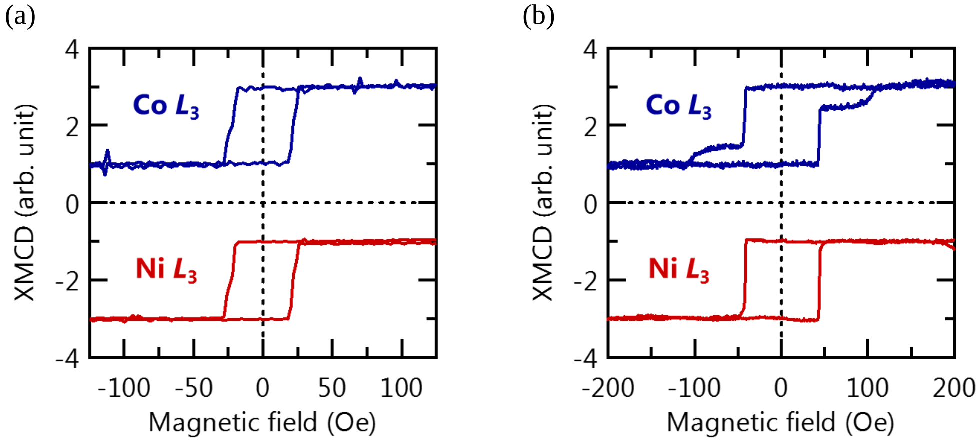

Fig. 4. Element-specific hysteresis loops measured at the Co and Ni L_3 edges for the sample with (a) 4-nm-thick and (b) 6-nm-thick NiO layer. H was applied orthogonal (parallel) to the H_M direction for the sample with 4-nm-thick (6-nm-thick) NiO.

magnetic field value becomes equal to the NiFe coercivity, the CoFe magnetisation rotates towards an intermediate direction that has a finite angle with respect to the magnetic field direction. In other words, a canted state of the CoFe magnetisation exists within the plateau regions. The canting angle can be estimated from the height of the plateau to be ~60° relative to the magnetic field direction. This canted state gradually collapses with increasing applied magnetic field, until finally the CoFe magnetisation aligns parallel to the magnetic field. These findings indicate that there exists an exchange coupling between the CoFe and NiFe layers through the antiferromagnetic NiO layer, which favours the magnetisations to align at a finite angle. In fact, a 90° interlayer coupling has been reported in a similar Fe/NiO/Co multilayer structures.[6] We believe the present structures also possess such an interlayer coupling through the NiO layer. The sample with 12-nm-thick NiO layer showed very similar loops to those of the sample with 6-nm-thick NiO layer (not shown here). This indicates that the CoFe and NiFe layers are coupled even through the 12-nm-thick NiO layer in this sample.

Figure 5 shows the XFMR delay scans acquired at the Co and Ni L_3 edges for the sample with 4-nm-thick and 12-nm-thick NiO layer. Note that microwaves of 4 GHz (6 GHz) were supplied to the sample with 4-nm-thick (12-nm-thick) NiO layer in the measurements. This is because of two reasons as follows: (1) The XMFR for 4-nm-thick NiO was measured at ALS where spin dynamics up to 4 GHz is detectable. (2) The

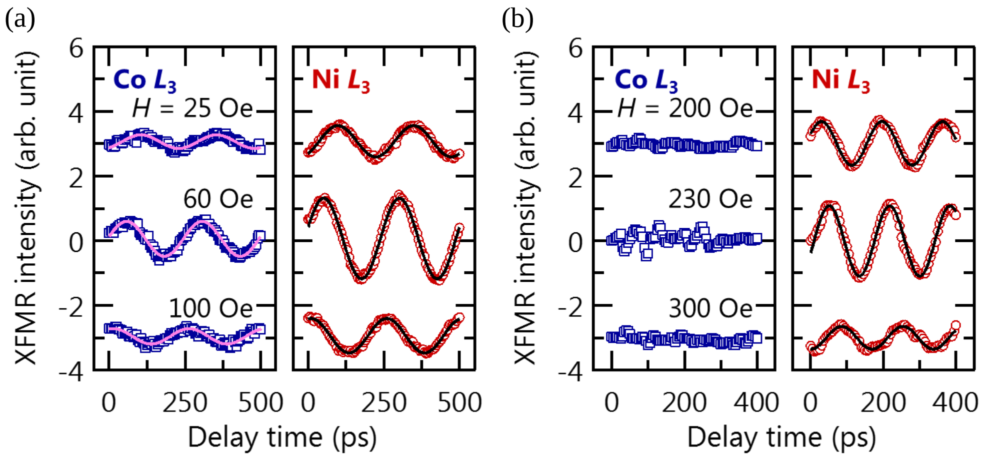

Fig. 5. XFMR delay scans measured at various H for the Co and Ni L_3 edges for the sample with (a) 4-nm-thick and (b) 12-nm-thick NiO layer. Microwaves of 4 GHz (6 GHz) were supplied to the sample with 4-nm-thick (12-nm-thick) NiO layer in the measurements. Sine curves are fits to the experimental data.

XFMR for 12-nm-thick NiO was measured at DLS where spin dynamics up to 10 GHz is detectable, while this sample's lowest FMR mode lies above 5 GHz. In the sample with 4-nm-thick NiO layer, the Co and Ni spins, i.e., the CoFe and NiFe magnetisations showed a periodic oscillation corresponding to the microwave frequency of 4 GHz. This shows that the rf field excites spin dynamics in the multilayer structure that are then detected by the XMCD effect. The amplitudes and phases of the magnetisation precession were extracted by fitting sine curves to the data acquired at the different values of H, as shown in Fig. 6. Both the CoFe and NiFe layers showed typical FMR behaviours, in which the amplitude shows a Lorentzian-like behaviour and the phase changes by 180° across the FMR condition. The FMR field was close to 60 Oe for both, and the precession was found to have very similar phase for both the layers. These results indicate that the CoFe and NiFe layers are strongly coupled and exhibit an acoustic precessional mode. Since the intrinsic FMR frequency of the CoFe layer is as high as 10 GHz, which we confirmed in a control sample of CoFe/NiO bilayer by VNA-FMR measurement (not shown here), it is not possible to solely excite FMR in the CoFe sink layer by microwaves of 4 GHz frequency. The magnetisation precession of the CoFe sink layer could be attributed to an ac spin current that is generated by the NiFe pump layer and then propagates through the NiO layer. We determined the spin current as "ac", because an ac spin current exerts a torque on the FM so that the magnetisation precesses at the frequency of the ac spin current, while a dc spin current excites the magnetisation precession of a FM at its intrinsic FMR frequency. Considering that we found the acoustic precessional mode, the XFMR results could be interpreted as a signature of ac spin current mediated by a quasi-uniform mode in the NiO layer. The sample with 6-nm-thick NiO layer showed very similar results to those of the sample with 4-nm-thick NiO layer (not shown here). This indicates that an ac spin current can propagate through the 6-nm-thick NiO layer at most. In the sample with 12-nm-thick NiO layer, on the other hand, the NiFe magnetisation showed a precession corresponding to the microwave frequency of 6 GHz, whereas the CoFe magnetisation did not show such an oscillation. This suggests that any precession of the CoFe magnetisation is much smaller than that in the sample with 4-nm-thick NiO layer since a spin current should be attenuated as it flows through the NiO. Thus, it is interesting that the sample with 12-nm-thick NiO layer showed a strong static coupling (as mentioned in the previous paragraph) but a weak dynamic coupling. We are not sure how this is possible at this moment.

Here, let us discuss other possible origins of the coupling between the CoFe and NiFe layers. It may be possible that either pinholes within the NiO layer allow the CoFe and NiFe layers to directly couple to each other, or else the NiO layer contains oxygen deficiencies and metallic (ferromagnetic) Ni components mediate the coupling. As for the possibility of the pinholes, we can exclude it by considering that the CoFe

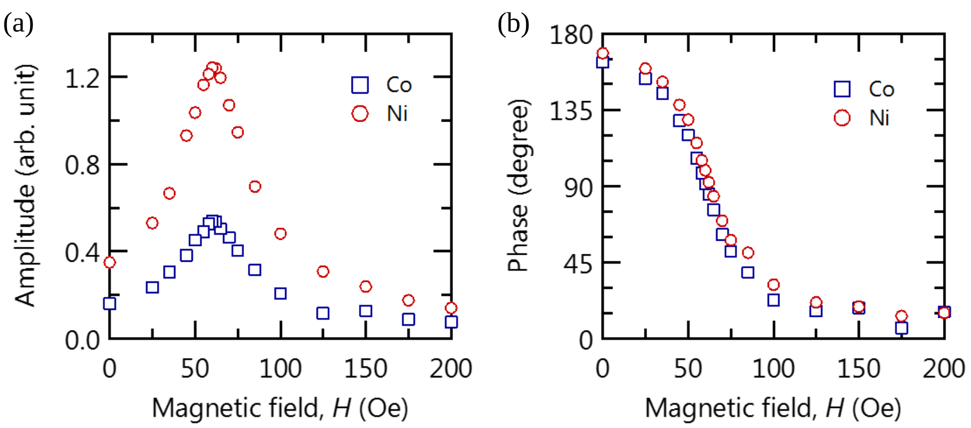

Fig. 6. (a) Fitted amplitudes and (b) phases extracted from XFMR delay scans at the Co and Ni L_3 edges for the sample with 4-nm-thick NiO layer.

layer showed the canting state which is attributed to the exchange coupling through the NiO layer, as discussed in Fig. 4. We can exclude the possibility of the metallic Ni components as well, because no XMCD signal was observed in the control sample of CoFe/NiO bilayer (not shown here). In addition, the control sample showed an XMLD (X-ray magnetic linear dichroism) signal which detects the Néel vector of an AFM. Thus, we believe that such defects in the NiO layer do not contribute to the coupling clarified in this study. Finally, let us discuss the consistency of our experimental results with a theory. Previously, Khymyn *et al.* theoretically studied a mechanism of spin current propagation through an AFM adjoining a FM.[7] It was demonstrated that a thin AFM can conduct spin current through the excitation of a pair of evanescent AFM spin wave modes. One can interpret that the evanescent spin wave modes correspond to an ac spin current, because they are defined as a time-dependent magnetisation component. In addition, they calculated the effective penetration depth of spin current for NiO as about 5 nm. Our XFMR results showed that an ac spin current propagates through the 4-nm-thick and 6-nm-thick NiO layer, which supports the calculated value. Thus, we believe that our experimental results are consistent with the theory proposed by Khymyn *et al.*

Here, let us show another evidence of spin current propagation through NiO. Another sample set, MgO(001) substrate/MgO (5)/$Co_{25}Fe_{75}$ (5)/Ag (5)/NiO (0, 2, 4, 6)/Fe (1)/$Ni_{80}Fe_{20}$ (25)/MgO (3), was prepared in the same manner as the previous samples in this study. The Ag layer was inserted in order to decouple the CoFe and NiO layers. The idea to is look for a bipolar phase variation of the CoFe sink layer as evidence of spin current propagation through the Ag and Ag/NiO layers, as such a phase behaviour was found as a signature of ac spin current in our previous studies.[4,5] Figure 7 shows the XFMR delay scans acquired at the Co and Ni L_3 edges for the sample without NiO layer and 2-nm-thick NiO layer. Microwaves of 4 GHz were supplied in the measurements. For these two samples, the CoFe and NiFe magnetisations showed a periodic oscillation corresponding to the microwave frequency of 4 GHz. On the other hand, another two samples with 4-nm-thick and 6-nm-thick NiO layers did not show considerable magnetisation precession of the CoFe layer (not shown here). Perhaps the sample with the 4-nm-thick NiO layer might show a quite small CoFe magnetisation precession only around the FMR field of the Fe/NiFe pump layer. As shown in Fig. 7, the amplitudes and phases of the magnetisation precession were extracted by fitting sine curves to the data acquired at the different values of H, as shown in Fig. 8. For both samples, the NiFe magnetisation showed a typical FMR behaviour as was expected, whereas the CoFe magnetisation showed an obviously different behaviour. The amplitude increases and the phase decreases around the FMR field of the NiFe layer, which is different from the results of the previous samples where the CoFe and NiO layers

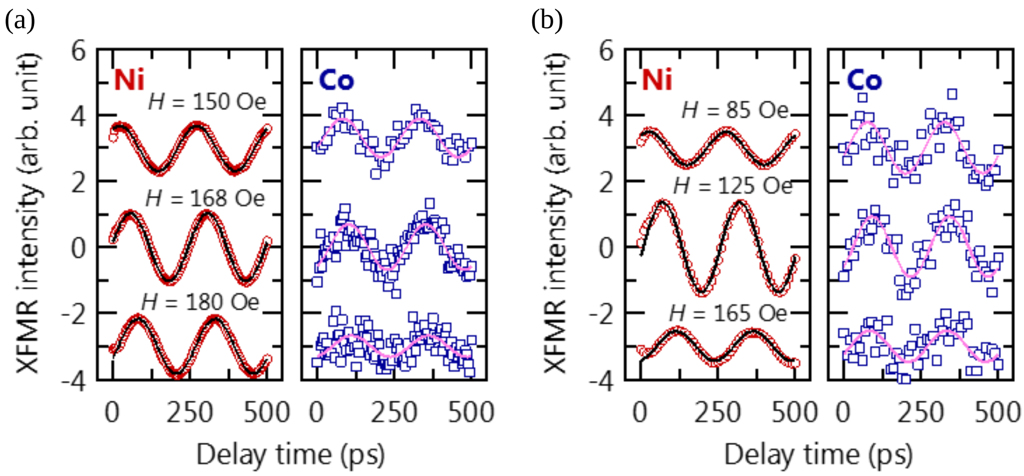

Fig. 7. XFMR delay scans measured at various H for the Co and Ni L_3 edges for the sample (a) without NiO layer and (b) with 2-nm-thick NiO layer. Sine curves are fits to the experimental data.

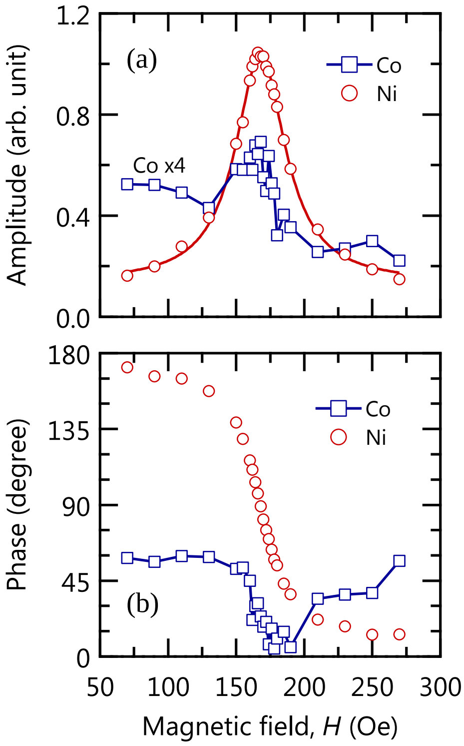

Fig. 8. Fitted amplitudes and phases extracted from XFMR delay scans at the Co and Ni L_3 edges for the sample (a)(b) without NiO layer and (c)(d) with 2-nm-thick NiO layer. Red lines in (a) and (c) are Lorentzian fits to data.

directly contact. The phase variation did not show a bipolar behaviour which we expected to find, but even so, these XFMR results indicate ac spin current propagating through the Ag and Ag/NiO layer, because the magnetisation precession of the CoFe layer was excited at the microwave frequency. In other words, these findings, especially the results of the sample with 2-nm-thick NiO layer, are another evidence of ac spin current propagating through antiferromagnetic NiO. Although the detailed mechanism of the spin current propagation is not clear up to now, a very simple picture we imagine is as follows: In the sample with 2-nm-thick NiO layer, an ac spin current could be pumped by the Fe/NiFe pump layer, mediated by magnons in the NiO layer, converted into spin-polarised electrons at the Ag/NiO interface, and transported into the CoFe sink layer. The difference in the XFMR results between the samples with and without the Ag layer could originate from the difference in the carriers of spin current, i.e., magnons or spin-polarised electrons.

4. Summary

We performed time-resolved XFMR measurements on CoFe/NiO/Fe/NiFe multilayer structures in order to detect an ac spin current propagating through the NiO layer. Element-specific hysteresis loops revealed the CoFe and NiFe layers are strongly coupled through the NiO layer. The XFMR delay scans indicate that the CoFe and NiFe layers are dynamically coupled and exhibit an acoustic precessional mode for 4 and 6 nm NiO thickness. The XFMR results could be interpreted as a signature of ac spin current mediated by a quasi-uniform mode in the NiO layer.

5. References

[1]C. Hahn, G. De Loubens, V. V. Naletov, J. Ben Youssef, O. Klein, and M. Viret, A Lett. J. Explor. Front. Phys. 108, 57005 (2014).

[2]H. Wang, C. Du, P.C. Hammel, and F. Yang, Phys. Rev. Lett. 113, 097202 (2014).

[3]G. van der Laan, J. Electron Spectros. Relat. Phenomena 220, 137 (2017).
[4]M.K. Marcham, L.R. Shelford, S.A. Cavill, P.S. Keatley, W. Yu, P. Shafer, A. Neudert, J.R. Childress, J.A. Katine, E. Arenholz, N.D. Telling, G. Van Der Laan, and R.J. Hicken, Phys. Rev. B 87, 180403(R) (2013).
[5]J. Li, L.R. Shelford, P. Shafer, A. Tan, J.X. Deng, P.S. Keatley, C. Hwang, E. Arenholz, G. Van Der Laan, R.J. Hicken, and Z.Q. Qiu, Phys. Rev. Lett. 117, 076602 (2016).
[6]J. Wu, J. Choi, A. Scholl, A. Doran, E. Arenholz, Y.Z. Wu, C. Won, C. Hwang, and Z.Q. Qiu, Phys. Rev. B 80, 012409 (2009).
[7]R. Khymyn, I. Lisenkov, V.S. Tiberkevich, A.N. Slavin, and B.A. Ivanov, Phys. Rev. B 93, 224421 (2016).

6. Achievements

These results have been presented at several conferences as follows:

[1] T. Nakano, M. Dabrowski, Q. Li, M. Yang, C. Klewe, D. Burn, P. Shafer, Z. Q. Qiu, G. van der Laan, E. Arenholz, and R. J. Hicken, "Time-resolved X-ray detected ferromagnetic resonance measurements of a CoFe/NiO/Fe/NiFe multilayer structure," Workshop on Emerging Applications of Spin Transfer Torque, Exeter, 2018.6.

[2] T. Nakano, M. Dabrowski, Q. Li, M. Yang, C. Klewe, D. Burn, P. Shafer, Z. Q. Qiu, G. van der Laan, E. Arenholz, and R. J. Hicken, "Time-resolved X-ray detected ferromagnetic resonance measurements of a CoFe/NiO/Fe/NiFe multilayer structure," IEEE International Conference on Microwave Magnetics, Exeter, 2018.6.

[3] T. Nakano, M. Dabrowski, Q. Li, M. Yang, C. Klewe, D. Burn, P. Shafer, Z. Q. Qiu, G. van der Laan, E. Arenholz, and R. J. Hicken, "Time-resolved x-ray detected ferromagnetic resonance measurements in a CoFe/NiO/Fe/NiFe multilayer structure," Magnetism, Manchester, 2018.4.

[4] T. Nakano, M. Oogane, T. Furuichi, and Y. Ando, "Magnetic tunnel junction using perpendicularly magnetized synthetic antiferromagnetic structure for wide-dynamic-range magnetic sensors," York-Tohoku-Kaiserslautern Research Symposium on New-Concept Spintronics Devices, York, 2017.6.

[5] T. Nakano, M. Dabrowski, Q. Li, C. Klewe, D. Burn, P. Shafer, Z. Q. Qiu, G. van der Laan, E. Arenholz, and R. J. Hicken, "Time-resolved x-ray detected ferromagnetic resonance measurements in a CoFe/NiO/Fe/NiFe multilayer structure," The 65th Japan Society of Applied Physics Spring Meeting, Tokyo, 2018.3.