Study of magnetic properties at the interface in ultra-thin CoFeB films using a high sensitivity VNA-FMR spectrometer

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Dynamical properties of ultra-thin magnetic films are attracting increasing attention with the advent of various MTJ based spintronic devices such as spin transfer torque magnetic random access memory (STT-MRAM) or voltage torque magnetic random access memory (VT-MRAM), in which the free layer is a CoFeB film with a typical thickness of approximately 1 nm or even less. It has been observed that many magnetic properties are dependent on the film thickness in the nanometer scale [1]. In addition, VT-MRAM takes advantage of the change of perpendicular magnetic anisotropy (PMA) by the application an electric field at the CoFeB/MgO interface [2]. Thus, these devices require thorough understanding of the interfacial magnetism.

Ferromagnetic resonance (FMR) is the most commonly used measurement for the study of magnetization dynamics. FMR provides various magnetic properties, some of which, such as the damping parameter or Lande g factor, cannot be obtained by other characterization techniques. It is highly desirable to perform broadband FMR measurement to characterize a magnetic thin film. However, such a measurement on an ultra-thin film has been difficult, mainly due to the lack of sensitivity or broadband measurement capability in conventional FMR spectrometers.

We have developed a high sensitivity vector network analyzer ferromagnetic resonance (VNA-FMR) spectrometer as shown in Fig. 1 to overcome this difficulty. In this system, a low frequency modulation field is applied to the sample in addition to a DC bias field (H_B) while a VNA measures the S-parameter, and the modulation frequency component of the S-parameter is extracted by a numerical data processing equivalent to lock-in detection, which significantly enhances the sensitivity while maintaining the broadband coverage.

Using this system, we measured FMR on a series of ultra-thin CoFeB films with thicknesses ranging from 1.5 nm down to 1.1 nm under H_B applied along the out-of-plane (OOP) direction. Fig. 2 is the FMR spectra on a 1.5 nm thick CoFeB film as a function of frequency and H_B , which shows a clean Kittel mode FMR signal. The gyromagnetic ratio that contains Lande *g* factor can be determined from the slope of the peak frequency vs H_B plot. Fig. 3 shows the value of *g* as a function of CoFeB thickness. The value of *g* increases as the thickness decreases, which suggests that the orbital angular moment is not completely quenched at the interfaces due to broken symmetry, which could be the cause of the increase of PMA or damping parameter previously reported in [1].

In the presentation, we will first introduce the high sensitivity VNA-FMR developed in this work, then report the FMR measurement results on CoFeB films over wide range of thicknesses and deposition conditions to explore the change of magnetic properties at the interface.

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Fig. 1, Block diagram of the VNA-FMR with field modulation detection developed in this work.



Fig. 2, FMR spectra on a 1.5 nm thick CoFeB film under OOP H_B .



Fig. 3, Lande *g* factor as a function of CoFeB thickness.

Magnonic band gaps of metallic one-dimensional magnonic crystals

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Magnonics is one of the most fascinating research fields in spintronics. Spin waves (SWs), also called magnons, have attracted special attention because of their potential application as information transport and processing for novel spin devices. In recent our work, some important characteristics on spin waves were reported, such as nonreciprocity [1, 2] and conductivity effect [3]. Among magnonics, magnonic crystals (MCs) are one of the most attractive topics for both fundamental physics and future applications. MCs, which are analogous to photonic crystals, are defined as artificial media with spatially periodic variation of some of their magnetic parameters. As a result, MCs show modified spectra compared with plain films. SWs with certain frequency ranges cannot be allowed to propagate in MCs and forms rejection bands, that is, band gaps. The formation of band gap can be used to not only the SW filters but also control of group velocity, for example, generation of the slow magnon. So far, research on MC using YIG have been the mainstream because of their ease of sample preparation. Recently, there were some reports about metallic MCs, but the most of the metallic MCs had structural periods ranging from 0.1 to 1 µm because metals have a shorter propagation length of SW (10 µm order) than that of YIG (mm order) and ones try to make a lot of periodic structures in a limited length. The wave numbers affected by them, however, are too high to access using antenna method. It is important to detect the influence of MCs on SWs by all electrical measurement method for development of novel spin wave devices. In this study, metallic 1D-MCs consisting of Py strips with periodic grooves were investigated. We measured the propagation properties of the magnetostatic surface wave (MSSW) in the Py MCs by an antenna method and demonstrated the electrical detection of the magnonic bandgaps.

Py strips with a width of 100 μ m and a thickness *d* of 50 nm were prepared on high-resistivity Si/SiO₂ substrates. The periodic grooves with a depth of 25 nm were formed by Ar⁺ ion milling. The grooves are aligned perpendicular to the propagation direction. The lattice constant (*D*) was 2.0, 3.0, and 4.0 μ m, and the grooves with a width of *D*/2 were separated by *D*/2 (see Fig. 1). We also fabricated an unstructured film for comparison. A 80-nm-thick SiO₂ layer were deposited for isolation and a pair of coplanar waveguides (CPWs) of signal (S)-ground (G) type was formed for excitation and detection antennae. The widths of S, G lines, and the SG-gap were 1.0, 50, and 1.0 μ m, respectively. A vector network analyzer and a microprobe station with an electromagnet was used for spin wave transmission measurement.

Micromagnetic simulation of spin-wave propagation were performed using the Object Oriented Micromagnetic Framework (OOMMF) software package based on the Landau-Lifshitz equation. The excitation field of the SG type antenna was separately calculated using MATLAB, and a Gaussian pulse excitation with a pulse width of 50 ps was applied in OOMMF using the calculated field profile. The SW spectra can be obtained by Fast Fourier Transformation (FFT).

Figure 2 (a) shows the spectra mapping as a function of distance x from an excitation antenna for the Py MC with D = 2 µm. The dispersion relation was also obtained by FFT along x (Fig. 2(b)). It has some pronounced dispersion branches and the clear magnonic band gaps can be observed at $k = \pi/D$ and $2\pi/D$. The spectrum at detection point (Fig. 2(c)) shows some dips at corresponding the band gap frequency. Thus, the introduction of a periodic structure yields rejection frequencies in spin wave spectrum.

Figure 3 shows the experimental (red) and calculated (blue) results of normalized spin-wave spectra for an unstructured film and MCs under $\mu_0 H = 20$ mT. In the unstructured film, there is no dip in the spectra of both experimental and calculated results. The shapes of the spectra for the MCs are obviously different from that for the unstructured film and have obviously large dips (denoted as first dips) at the specified frequencies. The calculated spectra for the MCs have obvious first (large) and second (small) dips. In experimental spectra of MCs, second dips are not so clear because of the small oscillations due to interference. The oscillation around here is, however, distorted compared to the unstructured film, and the frequencies of the dips for the experimental and calculated spectra are almost equal. Additionally, they depend on *D*. Therefore, we judged that it is the influence of existence of 2nd dip. Thus, we succeeded in the electrical detection of magnonic band gaps for Py 1D-MCs with periodic grooves utilizing an antenna method.

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Fig. 1 SEM photograph and schematic illustration of a Py MC sample.



Fig. 2 Simulated intensity distribution, dispersion relation, spin wave spectrum of a Py MC with $D = 2 \mu m$.



Fig. 3 Spin wave spectrum for an unstructured film and MCs of experimental and simulated results.

Imaging of microwave electric- and magnetic-fields by optical indicator microscopy

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High-resolution imaging techniques of microwave E-field and H-field can be powerful tools for visualization of the electromagnetic (EM) field distributions of materials and devices. Using the suggested method based on the optical microscopy one can visualize the E-field or H-field and temperature distributions through the thermo-elastic effect utilizing optical indicator films. Depending on the absorption properties of the indicator films, the system can visualize either the distributions of the temperature, E-field or H-fields.

The optical indicator microscopy technique uses a polarized light microscope method. Figure 1(a) shows the illustration of the visualization system. Probing green light is modulated to be circularly polarized by using a linear polarizer (0°) and quarter waveplate (45°). The reflected light passed through the stressed medium changes the polarization from circular to elliptical due to the photo-elastic effect in the glass substrate. The EM signal generated by the device under test (DUT) interacts with the optical indicator heating up ITO thin film. The detected heat distribution corresponds to the initial EM field distribution of the DUT. Finally, by using the analyzer (linear polarizer sheet) a CCD camera recorded the temperature or EM field distribution images¹.

Figure 1(b) demonstrates the DUT representing a microwave lowpass (LP) filter and the optical indicator microscopy system and the representative measurement results of the microwave H-field and E-field distribution images at 4 GHz. We used ITO glass for the H-field visualization owing to the rather good conductive properties of the ITO ensuring a strong interaction with the H-field of the DUT. As a result, the ITO film heats up due to the surface currents induced by the H-field and the corresponding thermal distribution appears. On the contrary, for the E-field visualization, we used an indicator consisting of aluminum nanoparticles coated by a poly (methyl methacrylate) (PMMA) thin film since it is well known that metal nanoparticles embedded in glass and polymer give a large increase in the dielectric loss.

The present technique provides a practical approach for high-resolution visualization of the E-field and H-field, as well as thermal distributions, which are valuable in investigations related to the influence of thermal variations on the properties of microwave electronic devices or materials.

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Figure 1. (a) Illustration of measurement principle of the TEOIM system. (b) The top shows the schematic of the tested lowpass filter and the optical indicator. The bottom images depict the visualized microwave H-field and E-field distribution images of the DUT at 4 GHz.

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Observation of magnon polarization through neutron scattering

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Spin current, a flow of the spin degree of freedom in condensed matter, is a fundamental concept in spintronics research. Many experimental and theoretical investigations are devoted to developing creation/annihilation and control methods of the spin current. As one of the creation methods, the spin Seebeck effect (SSE) attracts much attention recently due to possible applications to thermal spin generators for driving spintronics devices. The SSE is observed via the induced spin current in ferromagnets and ferrimagnets with an application of a temperature gradient. Driving spin currents thermally could lead to the manufacturing of a compact spin current source without using an electric current or magnetic field.

Deep into microscopic views, the spin current in magnetic insulators is carried by the transverse component of spin-waves (quantized magnons). Magnons can be polarized, and the magnon polarization, i.e., the direction of the precessional motion of the electronic spin, affects the thermodynamics of magnetic materials, governing the magnitude and sign of the SSE. However, the magnon polarization of magnon modes has eluded experimental observation. We here show our recent results of the first direct observation of magnon polarization through polarized neutron scattering experiment¹). Our results describe electromagnetic responses of magnons in THz regime that are spanned over a wide (Q, ω) space.

Target compound, the iron-based garnet $Y_3Fe_5O_{12}$ (YIG), is a ferrimagnetic insulator with a complex structure and is an essential material for microwave and optical technologies and also for basic research in spintronics, magnonics, and quantum information. One reason is that it has the highest quality magnetization dynamics among known magnets—resulting in long magnon lifetimes. There exist major two magnon modes, and the gap separating optical and acoustic modes is of the order of the thermal energy at room temperature. A maximum of the spin Seebeck signal in YIG near room temperature²⁾ has been interpreted in terms of the competition between magnon modes with opposite polarization. Our experimental findings are well accounted for by atomistic spin dynamics calculations. The observation of both signs of magnon polarization in YIG (Fig.) also gives direct proof of its ferrimagnetic nature.

The research was conducted through a collaboration with J. Barker, T. Kikkawa, Y. Shiomi, M. Fujita, G. E. W. Bauer, E. Saitoh, and K. Kakurai.

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Fig. Illustration of magnon polarization for two magnon modes in YIG¹). The positive polarization acoustic (left) and negative polarization optical modes (right) are shown.

Efficient terahertz frequency conversion in a Dirac semimetal Cd₃As₂ and terahertz anomalous Hall effect in a Weyl antiferromagnet Mn₃Sn

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Recently nonlinear light-matter interaction in terahertz (THz) frequency has attracted considerable attention for fundamental interests in physics and photonics and also for finding novel functionalities of materials bridging electronics and photonics. Here, we introduce our recent works using THz spectroscopy for topological semimetals^{1,2)}, *i.e.*, a Dirac semimetal and a Weyl semimetal with the massless energy dispersion, which may pave a new route for high-speed electronic and spintronics.

High harmonic generation (HHG) has been one of the central issues in modern photonics since it produces coherent high-energy photons by multiplication of an incident photon energy, which has been developed in gaseous media for attosecond science and also utilized for high-resolution angle-resolved photoemission spectroscopy. More recently, HHG in semiconductors with mid- or near-infrared laser pulse has been also reported³⁾, raising the possibility for realizing stable and compact soft X-ray sources. For much lower frequency around 1 THz, however, HHG has been still very difficult due to the lack of intense light source and suitable materials. We have found efficient THz third harmonic generation in a superconductor originating from the resonance with the Higgs amplitude mode⁴⁾. If such an efficient THz harmonic generation is realized at room temperature, it would be a key technology for frequency conversion and mixing in high-speed electronics as well as for sensitive detection of the cosmic microwave background. Here we demonstrate room-temperature efficient THz harmonic generation with 3f=2.4 THz in 3D Dirac semimetal Cd₃As₂ thin films¹⁾. Our pump-probe spectroscopy for Cd₃As₂ reveals that the nonlinearity originates from intraband acceleration of massless electrons across the Dirac node as theoretically anticipated. The unprecedentedly efficient harmonic generation in the 3-dimensional material would open an avenue for developing a novel THz frequency convertor.

We also investigated the THz response of a Weyl antiferromagnet Mn_3Sn . Spin motion in antiferromagnets is as fast as in THz frequency far beyond that in the ferromagnets and therefore it has been expected as a candidate for high-speed data processing in spintronic devices. But the readout of antiferromagnetic spin order is difficult due to the small net magnetization. The noncollinear antiferromagnet Mn_3Sn shows a large anomalous Hall effect comparable to ferromagnets in spite of the vanishingly-small net magnetization⁵), owing to broken time-reversal symmetry by the cluster octupole moment on Kagome bilayer. Therefore, the detection of the anomalous Hall current in Mn_3Sn at THz frequency is essential for high-speed readout of the spintronic device based on the antiferromagnet. Here we report observation of the THz anomalous Hall effect in a Mn_3Sn thin film by a broadband polarization-resolved THz spectroscopy up to 6 THz²). We found that the non-dissipative large anomalous Hall current appears up to around ~1 THz, while at higher frequency the dissipative part of the Hall current grows up possibly due to the interband transition across the Weyl node. Our observation of a large THz response in the antiferromagnets paves the way for ultrafast readout of antiferromagnetism with THz current on device.

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Designing spin textures and topological transports in insulating antiferromagnets

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Insulating antiferromagnets are now being recognized as one of the important platforms of spintronic devices¹). Several advantages over ferromagnets include a relatively high resonance frequency accelerating the writing speed for a memory storage device, and the absence of stray fringing fields important for microfabrication. However, controlling the magnetic structures of antiferromagnets, even detecting the orientation of the ordered moments, is not an easy task. Here, we show that for general antiferromagnets one can theoretically design and control their basic magnetic properties which are tightly bound to magnetotransport effects — an emergent spin texture in reciprocal space^{2,3}, nonreciprocity of magnon bands^{4,5}, and topological antiferromagnons that carry heat current⁴).

In noncentrosymmetric crystals, the spin-orbit (SO) coupling supported by the broken global inversion symmetry generates Rashba and Dresselhaus types of momentum-dependent electronic spin textures in metals. Although one may expect similar types of phenomena in insulating magnets, the magnons in ferro/ferrimagnets do not have up/down "spin degrees of freedom" like electrons which can couple to their spatial momentum k. In this context, the theoretical highlight of antiferromagnets is the two magnetic sublattices and corresponding two species of magnons, which serve as fictitious up/down "spin degrees of freedom". This allows us to design several types of pseudo-SO couplings and related properties; the Dzyaloshinskii-Moriya (DM) interaction that twists the spins work to mix the motion of up/down magnons in a spacially antisymmetric manner, and gives rise to the Rashba and Dresselhaus-types of spin textures of magnon bands. This texture can be easily controlled by a small magnetic field and might hopefully be detected by the microwave measurements. An anomalous thermal Hall (ATHE) effect of magnons are also predicted for the two-dimensional square lattice antiferromagnet with easy-axis anisotropy, when a small out-of-plane magnetic field is applied. The origin of this ATHE carried by the SU(2) magnons⁴⁾ is qualitatively different from the typical thermal Hall effect (THE) of ferro/ferrimagnets in pyrochlore lattices carried by U(1) magnons⁶⁾. We also show that the pseudo-SO coupling can be generated for other inversionsymmetry-unbroken cases, e.g. Kitaev antiferromagnets, where the spatially variant interactions are transformed in a way to mix the kinetic motion of two species of magnons³⁾. The basic framework of how to design or predict these phenomena by a sketch of crystal lattices, spin orientations, and the species of interactions, and the magnetic field are shown^{2,3,7)}.



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Circularly polarized microwave measurements for condensed matter physics

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A cavity resonator in the microwave band is indispensable technology in condensed matter physics. The extremely high Q value of the microwave resonance mode enables us sensitive detection of the electron spin resonance of a small sample [1]. More recently, such a method has been utilized in spintronics to investigate the spin transport and in quantum information to control quantum qubits. As a microwave is an electromagnetic wave, it has polarization such as linear polarization and circular polarization. However, only linearly polarized microwaves are mainly used in condensed matter physics.

In optical and terahertz regime, a circularly polarized electromagnetic wave has been used for the various purpose: the creation of spin accumulation, investigation of circular photogalvanic effects, circular dependent spin pumping with an antiferromagnetic material. This is because a circularly polarized electromagnetic wave has a net spin of ± 1 , which is suitable for investigating electron spin-related phenomena.

If we can control the circularly polarized microwave in a cavity resonator, this technique will stimulate efforts towards further research in condensed matter physics. Therefore, we recently established a technique for selectively exciting left and right circularly polarized microwaves and irradiating them to a sample with a cylindrical cavity resonator [2] (Fig. 1). To demonstrate the performance of the cavity resonator, we measured the ferromagnetic resonance of Yttrium Iron Garnet (Fig. 2) and estimated the polarization of left- and right-handed microwave excitation as more than 80%.

In this symposium, I will present the overview and potential of the circularly-polarized cavity resonator method. I will also present our recent results obtained by using this method.

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Fig. 1 Schematic image of the circularly-polarized microwave excitation. A rotating magnetic field of right-(left-) handed circularly-polarized microwave are excited by S21(S12), respectively.



Fig. 2 Mmeasured ferromagnetic resonance of Yttrium Iron Garnet as a function of f and B. Red(Blue) rejoin corresponds to the right-(left-) handed circularly-polarized mode, respectively.