Renaissance of Ferromagnetic Semiconductors and Spintronics Applications（強磁性半導体ルネサンスとスピントロニクスへの応用）

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Ferromagnetic semiconductors (FMSs) with high Curie temperature (T_C) are strongly required for spintronics device applications. So far, the mainstream study of FMSs is Mn-doped III-V FMSs; however they are only p-type and their T_C is much lower than 300 K. In this study, we present a new class of FMSs with high T_C, Fe-based narrow-gap III-V FMSs. Because Fe atoms are in the isoelectronic Fe^{3+} state in III-V, the carrier type can be controlled independently and thus both n-type and p-type FMSs are obtained. Using low-temperature molecular beam epitaxy, we have successfully grown both p-type FMS [(Ga,Fe)Sb [1], (Al,Fe)Sb [2]] and n-type FMSs [(In,Fe)As [3], (In,Fe)Sb [4]]. The most notable feature in these Fe-based FMSs is that the T_C value increases monotonically as the Fe content increases; and there is a tendency that T_C is higher as the bandgap is narrower, which contradicts the prediction of the mean-field Zener model. Intrinsic room-temperature ferromagnetism has been observed in $(Ga_{1-x},Fe_x)Sb$ with $x \geq 23\%$ [1] and $(In_{1-x},Fe_x)Sb$ with $x \geq 16\%$ [4], which are promising for practical spintronic devices operating at room temperature.

We also present our findings on new magnetotransport phenomena in heterostructures containing these Fe-doped FMSs. In an Esaki diode composed of a 50 nm-thick n-type FMS (In,Fe)As (6% Fe) / 250 nm-thick p^+ InAs:Be, we found that the magnetic-field-dependence of the current flowing through the pn junction (magnetoconductance, MC) can be largely controlled, both in sign and magnitude, with the bias voltages V [5,6]: The diode shows small positive MC (~0.5%) at $V < 450$ mV, but the MC changes its sign and magnitude at $V >$ 450 mV, reaching -7.4% (at 1T) at $V = 650$ mV. This bias-controlled MC originates from the change in the band components of (In,Fe)As that participate in the spin-dependent transport.

Furthermore, we found that the current flowing in a nonmagnetic n-type InAs quantum well (QW) that is interfaced to an insulating p-type (Ga,Fe)Sb layer (20% Fe, $T_C > 300$ K) exhibits a giant change of approximately 80% at high magnetic field and that its magnitude can be controlled by ten-fold using a gate. The mechanism for this large magnetoresistance is attributed to a strong magnetic proximity effect (MPE) via the *s-d* exchange coupling at the InAs/(Ga,Fe)Sb interface. It was found that a spin splitting in the InAs QW is induced by MPE, which can be varied between 0.17 meV and 3.8 meV by the gate voltage [7]. Other studies on ferromagnetic semiconductor heterostructures are underway and novel phenomena and properties are being investigated [7-9]; these new properties of the Fe-doped FMS-based materials and devices provide novel functionalities for future spin-based electronics.

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Room-temperature germanium spintronics developed by atomically controlled heterointerfaces

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Semiconductor (SC) spintronics is expected for the achievement of novel logic and memory architectures with low power consumption in future electronics[1)]. In particular, because of the compatibility with CMOS technologies and optical communication on the silicon platform (Si-photonics), germanium (Ge)-based spintronic technologies have so far been developed[2)]. To operate Ge spintronic devices with nonvolatile memory effect above room temperature, it is essential to obtain sufficiently large local two-terminal magnetoresistance (MR) signals. Unfortunately, the value of room-temperature MR ratio in *n*-Ge-based lateral spin-valve devices has been less than 0.001 % [3,4)].

In this talk, we introduce a new method for enhancing room-temperature MR ratio in Ge spintronic devices. Here, we utilize an atomic-layer termination technique in addition to our unique technology with ferromagnetic (FM) Heusler alloy/Ge Schottky-tunnel contacts on Si substrates[2)]. When we insert five-six Fe atomic layers between the Heusler-alloy spin injector and the Ge layer, the quality of the Heusler alloy near the interface is significantly improved [5)], As a result, even at room temperature, we can obtain a large MR ratio of 0.04 % [5)], two orders of magnitude larger than those in previous works[3,4)]. For obtaining the highest MR ratio, we can reduce the electric power down to ~ 0.12 mW, one order of magnitude lower than that (~ 1.15 mW) in Si-based devices with MgO tunnel barriers[6)]. Because the MR ratio at 8 K reaches 0.43 % for above devices [5)], we also explore the degradation mechanism of the MR ratio with increasing the temperature (T). From the analyses based on the standard one-dimensional spin-diffusion model[7)], we can verify the temperature dependence of the FM/Ge interface spin polarization (P). As consequences, the decay mechanism of the FM/SC interface P with increasing temperature can be interpreted in terms of the $T^{3/2}$ law meaning a model of the thermally excited spin waves in the FM electrodes[8)]. Also, we confirm that the temperature-dependent magnetization of the ultra-thin FM layer just on top of Ge is strongly related to the degradation of the MR ratio[8)]. Therefore, the strong ferromagnetism of the FM layer near the interface is essential for high-performance Ge spintronics devices above room temperature.

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Spintronics using local angular momentum of surface acoustic wave

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Spin-vorticity coupling (SVC), which is one of the general relativistic effects in rotating body, enables us to convert mechanical angular momentum with magnetization, i.e. spin and/or orbital angular momentum of electrons. Since a novel type of spin current (SC) generation via SVC in a surface acoustic wave (SAW) was theoretically predicted by Matsuo et al.[1)], experimental studies on the SVC had been reported by several groups. Recently, we have succeeded to demonstrate a spinwave resonance (SWR) excitation via alternating SC generated in a NiFe(20 nm)/Cu(200 nm) bilayer deposited on piezoelectric $LiNbO_3$ substrate when the Rayleigh-type SAW is applied[2)]. However, there are still some open questions. First, there is no clear evidence that a gradient of SAW vorticity needed for SC generation exists in 200-nm-thick Cu film. Second, to understand the SVC quantitatively, we must evaluate the alternating SC in Cu as a function of amplitude of lattice deformation. Third, the theory expects two different sources for spin accumulation. One is the time derivative of local angular momentum Ω, and the another is Ω itself. We must examine which is dominant contributor for the SC generation. Moreover, there is a renormalization factor in the analytical expression of SC which is hard to be determined theoretically. It is significant to quantify this factor because the magnitude determines the conversion efficiency between the local angular momentum of the SAW and spin angular momentum. To improve the understanding in the microscopic mechanism of the angular momentum conversion between microscopic electron spin and macroscopic angular momentum in the SAW, we need quantitative information on the magnitude and spatial distribution of the SAW in the bilayer system.

In this symposium, we will show our recent experimental studies on (i) highly nonreciprocal SWR excited using magnetoelastic coupling in Ni/Si bilayer[3)], (ii) reciprocal SWR excited using gyromagnetic coupling in NiFe single layer[4)], and (iii) electrical measurement of alternating SC in NiFe/Cu bilayer[5)]. The nonreciprocity of the SWR owing to a shear strain component was strongly enhanced by embedding the Ni far from the surface. From the variation of the nonreciprocity on the thickness of Si covered on the Ni, we can estimate the depth profile of the relative amplitude of the shear strain component with respect to the longitudinal strain component that gives the spatial distribution of the SAW. Moreover, a picometer order SAW amplitude averaged over the NiFe film was experimentally evaluated from the amplitude of SWR excited via gyromagnetic effect whose amplitude was simply given by the vorticity of SAW. Finally, from the comparison between the amplitude of the alternating SC in NiFe/Cu bilayer and the SAW amplitude evaluated, we found that the conversion efficiency of the angular momentum from the SAW to the electron spin was much larger than in the spin current generation using a vorticity of liquid metal[6)]. Theoretically, the conversion efficiency of the angular momentum from the lattice into electron spins becomes larger when the difference of the energy scales is smaller between lattice motion and spins. Consequently, the renormalization factor of the elastic system is much larger than that of the liquid-metal flow because the elastic motion of our setup is in the gigahertz range whereas the vorticity of the liquid-metal flow is in the kilohertz range.

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Controlling antiferromagnetic resonances

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In antiferromagnetic spintronics where manipulation of the antiferromagnetic spins is a central technological challenge[1)], it is important to understand the dynamic properties, especially their THz spin dynamics and the magnetic damping. While both experimental and theoretical investigations of the antiferromagnetic resonance began in 1950s[2)], they have been recently revisited with more advanced experimental techniques[3,4)] as well as with more rigorous theoretical treatments[5)] in the context of emerging antiferromagnetic spintronics. In the early stage of the investigations, the state-of-art spectroscopy with a rather inefficient and weak far-infrared source[1)] was employed to investigate various antiferromagnets, such as NiO, CoO, MnO, and Cr_2O_3. Although their high resonant frequencies have been experimentally confirmed, the experimental technique at the time was not sufficiently sensitive to withstand detail analyses of the spin dynamics and the magnetic damping. Moreover, importance of the magnetic damping in antiferromagnet was not seriously argued. However, thanks to the recent development of the THz technologies, frequency-domain THz spectroscopies with much better sensitivity than before has now became accessible and affordable for investigating in more detail the spin dynamics in antiferromagnets.

The talk will be based on our recent results on (1) frequency-domain THz spectroscopies of antiferromagnetic NiO and the detail analysis of the antiferromagnetic damping[6)], (2) observation of the THz spin pumping effect in NiO/Pt and NiO/Pd[7)], and (3) control of the antiferromagnetic resonance properties by cation substations of NiO[8)].

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Interfacial multiferroics with perpendicular magnetic anisotropy

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Multiferroic materials have a great potential for low-power mannipulation of magnetization orientation using an electric field, where the cross coupling between the ferroic orders such as magnetization, electric polarization, strain, etc. plays a critical role[1–3)]. In general, the ferroic phase transition temperatures of single-phase multiferroics is lower than room temperature, requiring alternative material systems with multiferroic properties for spintronic applications. In this study, we investigate electric field driven magnetization switching of ferromagnetic/ferroelectric (ferroelastic) interfacial multiferroics with perpendicular magnetic anisotropy (PMA), where the multiferroic properties appear at room temperature[4–7)].

The first example which we study is [Cu/Ni] multilayer/ferroelectric $BaTiO_3$(001) interfacial multiferroics with PMA. Since there are two types of ferroelectric domains, i.e., $a-$ and $c-$domains, in $BaTiO_3$ at room temperature, different misfit strains are exerted on the [Cu/Ni] multilayer on $a-$ and $c-$domains. Such strain gives rise to a change in the magnetization orientation on each domain through the magnetoelastic coupling. This enables to control the magnetization orientation of [Cu/Ni] multilayers by driving the ferroelectric $a-c$ domain wall in an electric field. With this approach, we demonstrate electric field control of the magnetization orientation of [Cu/Ni] multilayers between out-of-plane and in-plane. Also, X-ray magnetic circular dichroism measurements show that modulation of the orbital magnetic moments of Ni layers occurs in an electric field while no visible changes in the spin magnetic moments are seen. These results clearly indicate that the orbital magnetic moment that could be manipulated by electric field induced strain is responsible for the magnetization switching in [Cu/Ni] multilayer/$BaTiO_3$ interfacial multiferroics.

Another example of interfacial multiferroics with PMA is [Cu/Ni] multilayer/ferroelectric $0.7Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.3PbTiO_3$(001) (PMN-PT) heterostructures. PMN-PT has 8 equivalent $\langle 111 \rangle$ crystallographic orientations, along which the ferroelectric polarization favors to align. When an electric field is applied across the PMN-PT substrate, either 71°, 180°, or 109° switching of ferroelectric polarization occurs, thereby interfacial strain transferred from PMN-PT to [Cu/Ni] multilayer could trigger the magnetization switching. In this work, we demonstrate electric field modulation of the magnetic domain structures of [Cu/Ni] multilayer/PMN-PT using Kerr microscopy. When the out-of-plane magnetic field is swept from the positive saturation to a small negative magnetic field (~ -45 Oe), partial reversal of the magnetization occurs, thereby a maze-type domain structure appears. As an electric field is applied at the small negative magnetic field, a clear evolution of the reversed magnetic domains is observed. The result is compatible with separate Kerr magnetometry experiments, where multilevel magnetization states can be seen by an electric field. The underlying mechanism of the evolution of the magnetic domain structure will be discussed based on possible distinct interfacial strain for 71°, 180°, or 109° switching of ferroelectric polarization in PMN-PT.

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Electric operation of magnetic skyrmions

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A magnetic skyrmion is a topological spin texture that originated from the competition between the exchange interaction and Dzyaloshinskii-Moriya interaction [1-4]. Skyrmions in bulks can be driven by the electric current through the spin-transfer torque with the extremely low threshold current density of 10^6 A/m^2 [5] compared to that for the domain walls of 10^{10}–10^{12} A/m^2 . In addition to their small domain size ranging from several nm to 1 μm, this outstanding property offers new spintronics applications, including the non-volatile magnetic memories and current-driven shift resistors. From an application point of view, ultrathin magnetic heterostructures are favorable systems rather than bulk magnets because of their compatibility with existing spintronic technologies. Intensive studies related to the skyrmion observation, driving, and manipulation have been reported in Co-based and CoFeB-based heterostructures [6-8].

Here we demonstrate the current-driven skyrmion motion in Ir/Co/Pt thin films and MgO/CoFeB/W thin films. In the Ir/Co/Pt system, skyrmions can be observed under the hysteresis for a magnetic field, indicating that the skyrmion phase is thermally stable. Skyrmions segregate in the transverse direction to the current flow via the skyrmion Hall effect, which shows scalability for current density and wire width [9]. We also demonstrate several new findings: the significant material dependence of skyrmion dynamics, multiplication of skyrmions at the non-linear regime, and non-local accumulation of nonequilibrium skyrmions over several tens μm [10]. These results suggest the importance of skyrmions' collective nature, while only the behaviors of a single skyrmion have been discussed in previous studies. On the contrary, in the MgO/CoFeB/W systems, skyrmions can be observed as the transformation from the stripe domains by the current pulses, indicating that the skyrmion phase is metastable. Besides, the skyrmion Hall effect is much smaller than that in the Ir/Co/Pt system. The result suggests the difference in skyrmions' current-driven mechanism between MgO/CoFeB/W and Ir/Co/Pt systems.

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