MM-P01 (Poster)

In-magnetic-field Annealing Effects on the Phase Growth of Mn-Bi-Sn Ternary System

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Effect of magnetic field on the crystal growth and phase transformation have been studied for magnetic materials [1]. Since ferromagnetic (FM) phase is stabilizes by magnetic energy, its reaction and phase equilibrium can be controlled by magnetic fields. In particular, reaction from non-FM to FM phase is enhanced by magnetic field [2].

In the reaction of multi-elements system such as ternary or quaternary system, many equilibrium phases exist. If a magnetic field is applied during the synthesis reaction of the multi-elements system, selective reaction to FM phase and control of the reaction route can be expected. In this study, the in-magnetic-field reaction for Mn-Bi-Sn ternary system was performed for investigating the magnetic-field-induced phase growth from non-FM elements to the magnetic phases such as MnBi, Mn₃Sn₂, Mn₃Sn and MnSn₂.

The pellet-shaped samples were prepared from powder Mn (3N), Bi (4N) and Sn (4N). In-magnetic-field annealing was performed in a magnetic field of 5 T at 573 K.

Fig. 1 shows BSE images of the (a) 0 T-48 h and (b) 5 T-48 h samples In both samples, MnSn₂, Mn₃Sn₂ and Mn₃Sn phases were observed in addition to the unreacted Mn and Bi. MnBi phase was not detected with or without magnetic field. The Mn-Sn compounds were found to be more stable than the FM MnBi even when 5 T field was applied. The phase fraction of FM Mn3Sn2 decreased with application of 5 T, and antiferromagnetic MnSn₂ became main phase. As a result, it was found that Bi does not contribute to the reaction of Mn-Bi-Sn mixture, and the reaction rate of Mn-Sn is changed bymagnetic field.



FIG. 1. The BSE images of Mn-Bi-Sn annealed at 573 K in (a) 0 T and (b) 5 T.

Keywords: Magnetic field, Zeeman energy, Ferromagnetic

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MM-P02 (Poster)

The crystal structure and magnetoelectronic properties in Mndoped YCu₃Fe₄O₁₂

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The quadruple perovskite oxides $ACu_3Fe_4O_{12}$ (A = Ca, Sr, La, Y) [1–4] (Fig. 1 left) undergo various electronic phase transitions because of the instability of the unusually high valence states of Fe, where the types of phase transitions are closely related to charge transfer/disproportionation. $CaCu_3Fe_4O_{12}$ exhibits charge disproportionation $(2Fe^{4+} \rightarrow Fe^{3+} + Fe^{5+})$ at 210 K, whereas the nominally isoelectronic $SrCu_3Fe_4O_{12}$ displays a negative thermal expansion in the temperature range of 200–270 K. $LaCu_3Fe_4O_{12}$ shows a discontinuous volume expansion at 360–400 K, which is derived from an intersite charge transfer between Cu and Fe ($3Cu^{2+} + 4Fe^{3.75+} \rightarrow 3Cu^{3+} + 4Fe^{3+}$). In contrast, $YCu_3Fe_4O_{12}$ undergoes a ferrimagnetic transition and metal-tosemiconductor transition at 250 K simultaneously with a charge disproportionation ($Fe^{3.75+} \rightarrow 5/8Fe^{3+} + 3/8Fe^{5+}$). In this study, we investigated the crystal structure and magnetoelectronic properties of Mn-doped $YCu_3Fe_4O_{12}$.

YCu₃Fe_{4-x}Mn_xO₁₂ (x = 0-4) samples were successfully synthesized at 12 GPa and 1273 K. The lattice constant *a* at room temperature monotonically decreased with increasing *x*. The Y–O and (Fe, Mn)–O bond lengths also monotonically decreased, while the Cu–O bond lengths monotonically increased. Figure 1 (right) displays the X-ray absorption spectra for the Cu K-edge of YCu₃Fe_{4-x}Mn_xO₁₂ at room temperature. The absorption edge shifted monotonically from the higher to lower energy side with increasing *x*, estimating the reduction in Cu valence from Cu^{-2.33+} (x = 0) to Cu⁻²⁺ (x = 4). In the presentation, the magnetoelectronic properties will be discussed.



FIG. 1 (left) Crystal structure of quadruple perovskite oxide $ACu_3Fe_4O_{12}$ (right) X-ray absorption spectra of $YCu_3Fe_{4-x}Mn_xO_{12}$ at room temperature.

Keywords: quadruple perovskite oxides, high-pressure synthesis, charge transfer

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Magnetic Properties of Ni₂MnAl by Prepared in High Magnetic Field

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 $L2_1$ -Ni₂MnAl is a ferromagnetic Heusler alloy with magnetic moment of 4.02 μ B/f.u. and Curie temperature *T*C of 361 K [1-2]. Ni₂MnAl shows order-disorder transformation between $L2_1$ phase and *B*2 phase at 774 K. Ordering from the *B*2 phase to the $L2_1$ phase takes a long annealing time of about one month at relatively lower temperatures below 774 K [2]. This means that a very long-term annealing at lower temperature is required to obtain highly ordered $L2_1$ -N₂MnAl, which has made the investigation of the ordering mechanism in Ni₂MnAl difficult.

Meanwhile, recently, we found that $B2-L2_1$ ordering was accelerated by magnetic fields at 673 K [3]. It was pointed out that magnetic field influenced the reduction of critical radius of $L2_1$ phase in B2-matrix, resulting in the enhancement of the ordering. In this study, in order to clarify the ordering mechanism of Ni₂MnAl in a magnetic field, annealing treatment was carried out at 623 K under magnetic fields.

 B_2 -Ni₂MnAl were prepared by reactive sintering at stoichiometric compositions. The samples were annealed at 1373 K for 48 hours and then quenched. The obtained samples were annealed at 623 K in magnetic fields of 0 T, 10 T, and 15 T.

Figure 1 shows the thermomagnetization (M-T) curves for the sample prepared at 673 K in 15 T for 72 h [3] and prepared at 623 K in 15 T for 48 h. A single peak due to the TC of $L2_1$ phase was reported for the sample annealed at 673 K [3]. For the sample annealed at 673 K, two-peak separation was appeared at 318 K and 67 K, which were due to Néel temperature TN of B2-phase and TC of L21-phase, respectively. TC of 623 K-annealed sample was slightly higher than that of 673 K-annealed sample. These results suggest that the order degree and TC of $L2_1$ phase depend on the annealing temperature in a magnetic field. The growth of $L2_1$ phase in fields was explained by nucleation of $L2_1$ phase at certain order degree in B2 matrix.



FIG. 1 The thermomagnetization curve for the sample prepared at 673 K in 15 T for 72 h [3] and prepared at 623 K at 15 T for 48 h.

Keywords: Heusler alloy, Order-disorder transformation, ferromagnetic material, Magnetic field

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MM-P04 (Poster)

Crystal structure and magnetic property of magnetoplumbitestructured BaFe₁₂-xRh_xO₁₉

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Magnetoplumbite ferrites $AFe_{12}O_{19}$ (A = Ba, Sr, La) have been widely utilized as functional magnetic materials. Site-selective substitutions of Fe are desired to control the magnetic properties precisely [2], but it is generally difficult because of many crystallographic sites of 2*a*, 2*b*, 4*f*₁, 4*f*₂, and 12*k* of magnetoplumbite structure [Fig. 1(a)]. In this study, we attempted the site-selective substitution of Fe³⁺ by Rh³⁺ in BaFe₁₂O₁₉ by using high pressures up to 8.0 GPa and investigated the magnetic properties.

The BaFe12–*x*Rh_xO₁₉ phases (x = 1-6) were isostructural to the magnetoplumbite. With increasing *x* from 1 to 6, the amounts of impurity phases of Fe₂O₃ and Rh increased, indicating that the solubility limit of Rh was less than 50% in the synthesis conditions of the present study. Rietveld refinement using the synchrotron X-ray powder diffraction data confirmed that the Fe ions at 2*a*, 4*f*₂, and 12*k* sites with octahedral coordination were preferentially substituted by Rh ions, whereas the other sites were not substituted by Rh ions. This is consistent with the fact that Rh³⁺ ions usually occupy the octahedral sites.

Fig. 1(b) shows the isothermal magnetization curves of $BaFe_{12-x}Rh_xO_{19}$ (x = 0, 1, 2) at 300 K. The saturation magnetization drastically decreased with increasing x. This behavior indicates that the Rh^{3+} ions in the low-spin configuration (S = 0) substituting for the Fe³⁺ ions (S = 5/2) at 2a and 12k sites preferentially decreased the population of the majority spins. This finding proposes that the site-selective substitutions drastically change the magnetic properties of magnetoplumbite ferrites.



FIG. 1 (a) Schematic of crystal structure of $BaFe_{12}O_{19}$. The arrows represent the directions of the magnetic moments. (b) Isothermal magnetization curves of $BaFe_{12}-_xRh_xO_{19}$ (x = 0, 1, 2) at 300 K.

Keywords: magnetism, high-pressure synthesis, site-selective substitution

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MM-P05 (Poster)

High-pressure synthesis and oxygen evolution reaction activity of Fe⁴⁺-Mn⁴⁺-mixed perovskite oxide CaFe_{1-x}Mn_xO₃

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Perovskite oxides are known as efficient catalysts for oxygen evolution reaction (OER). A tetravalent iron oxide, CaFeO₃, exhibits high catalytic activity because of the preferable electronic structure with small charge-transfer energy [1]; however, high-pressure synthesis is needed to stabilize the Fe⁴⁺ ions. The Fe⁴⁺-Co⁴⁺-mixed perovskite oxide, café_{0.5}Co_{0.5}O₃, demonstrates higher catalytic activity than CaFeO₃ because of the synergistic effect between Fe⁴⁺ and Co⁴⁺ ions [2]. In this study, we performed high-pressure synthesis and characterization of café_{1-x}Mn_xO₃(x = 0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1) to study the synergistic effects between Fe⁴⁺ and Co⁴⁺ ions on OER catalytic activity.

OER catalytic activities in 0.1 M KOH aqueous solution were evaluated using a rotating disk electrode system in the same manner as the previous study [2]. Synchrotron X-ray diffraction revealed that $cafe_{1-x}Mn_xO_3$ at each studied composition crystallized in the orthorhombic perovskite structures. Lattice volume refined by the Rietveld method monotonically decreased with the increase in Mn amount (Fig. 1). As shown in Fig. 2, the OER overpotentials were compared with a linear interpolation of end points. The samples near the end points (x = 0.1 and 0.9) had the same overpotentials as the values derived from the linear interpolation, suggesting no synergistic effect. In contrast, substantial decreases in the overpotentials for x = 0.3, 0.5, and 0.7 were observed, suggesting the synergistic effect in the intermediate compositions (Fig. 2). We conclude that the synergistic effect is predominant in Fe⁴⁺-Mn⁴⁺-mixed oxides as well as the Fe⁴⁺-Co⁴⁺ system.



FIG.1 Refined lattice volume of CaFe_{1-x}MnxO₃ FIG

FIG.2 OER overpotential of CaFe_{1-x}Mn_xO₃

Keywords: Oxygen evolution reaction catalyst, Perovskite, high-pressure synthesis

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MM-P06 (Poster)

Brownian motion of depinned skyrmion under applying in-plane alternating electric current

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Magnetic skyrmions are topologically protected particle-like spin textures and have unique properties. One of the important properties is the Brownian motion at near room temperature [1-2]. In recent years, the application of skyrmions to novel computers such as Brownian / probabilistic computers is expected [3-4]. In order to apply skyrmions to these computers, deepening the understanding of skyrmion dynamics is essential. Various effects on skyrmion dynamics have been studied, such as increasing temperature [1], applying gate voltage [2], and magnetic field noise [5]. While direct-electric-current-driven skyrmion dynamics has been studied [6], skyrmion dynamics under alternating electric current (AC current) has not been studied. Therefore, in this study, we explore the effect of AC current on the skyrmion dynamics.

A skyrmion film consisting of Ta(5) $|Co_{16}Fe_{64}B_{20}(1.2)|$ Ta(0.2) |MgO(1.5)| SiO₂(3.0) (described by nm) were deposited by magnetron sputtering. The motion of the skyrmions was observed by magneto-optical Kerr effect microscope, and the diffusion coefficient of skyrmions *D* were calculated from the trajectories of skyrmions. Fig. 1 is the diffusion coefficient as a function of frequency of 0.336 mA AC current. Fig. 1 shows that D_{xx} and D_{yy} have the almost same value at 100 Hz, and then 100 Hz AC current dependence of the diffusion coefficient (Fig. 2) was obtained. We found out that 100 Hz AC current can exponentially increase the diffusion coefficient of both axes. This result is attributed to the depinning of the skyrmions by applying the AC current. This research was supported by ULVAC, Inc., JST CREST Grant number JPMJCR20C1 Japan and JSPS Grant-in-Aid for Scientific Research (S) Grant Number JP20H05666.





FIG. 1 Diffusion coefficient as a function of frequency of AC current of 0.336 mA (Root mean squared value)

FIG. 2 100 Hz AC current (Root mean squared value) dependence of the diffusion coefficient

Keywords: Skyrmion, Brownian motion, Diffusion coefficient, Alternating electric current

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Crystal structure and thermochromism of a high-pressure phase of Yin_{1-x}Mn_xO₃

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A Mn-doped YinO₃ (Yin_{1-x}Mn_xO₃) exhibits a bright blue color [1]. Yin_{1-x}Mn_xO₃ crystallizes in a hexagonal structure with the space group $P6_3cm$, where InO₅ units form five-coordinated trigonal bipyramids (Fig. 1a). Light of ~2 eV is absorbed by the d-d transition of thedoped Mn³⁺, resulting in its blue color.

YinO₃ is reported to undergo a structural phase transition to an orthorhombic perovskite structure under high pressures of 12–15 Gpa at room temperature (Fig. 1b) [2], whereas the detailed structure of the highpressure (HP) phase is not experimentally clarified. Since the trigonal bipyramid units are lost in the HP phase, color fading is expected. Hence, the phase transition from the HP phase to the ambient-pressure (AP) phase may exhibit a significant change in color. In this study, we aim to clarify the crystal structure and the thermochromism of HP phases of $Yin_{1-x}MnxO_3$ (x = 0, 0.1).

Synchrotron X-ray powder diffraction patterns confirmed that the HP phases of $Yin_{1-x}Mn_xO_3$ (x = 0, 0.1) crystallized in the GdFeO₃-type orthorhombic perovskite structures with the space group of Pnma (Fig. 2a). The volume per formula unit in the HP phase of YinO3 decreased by 8.15% from the AP phase. The HP phase of x = 1 displayed gravish-black due to the aforementioned lack of the trigonal bipyramidal structure, and annealing at 1200 °C in air resulted in the color change to bright blue (Fig. 2b). Thermogravimetry-differential thermal analysis supported the phase transition at 1100 °C. The blue-colored sample after the



FIG. 1 Schematics of crystal structures of YInO₃ in (a) AP and (b) HP phases



FIG. 2 (a) Synchrotron X-ray diffraction pattern and Rietveld refinement result for HP phase of YInO₃. The wavelength is 0.41996 +. (b) Color information from HP- to-AP phase of YIn_{1-x}Mn_xO₃

annealing was confirmed to be the AP phase by X-ray diffraction. The present study revealed that an irreversible thermochromism is realized in the HP-to-AP-phase transition of $Yin_{1-x}Mn_xO_3$.

Keywords: pigment, phase transition, thermochromic material, high-pressure synthesis

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MM-P08 (Poster)

V-V dimerization and its effect on magnetism in ilmenite-type CoVO₃

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The magnetic properties of transition-metal oxides can be explained by the interactions between isolated ions. The formation of a molecular orbital due to a closer metal-metal distance alters the electronic properties. The magnetism of such a compound is not sufficient to consider only the interaction between isolated ions. One of the most famous examples is the Peierls transition in VO₂. Formation of direct V-V bond results inan insulator (non-magnetic) to metal (paramagnetic) transition.

We have focused on ilmenite-type $CoVO_3$. In this compound, the Co^{2+} and V^{4+} form honeycomb lattices, respectively, and are stacked alternately (Fig. 1). The V-V dimers are expected to be formed in the vanadium layers. In this study, we reveal the existence of V-V dimers by crystallographic studies and the magnetic properties of $CoVO_3$ [1].

CoVO₃ samples were synthesized at high-pressure and high-temperature conditions of 8 GPa and 1100°C. The crystal structure analysis revealed the existence of V–V dimers below 550 K. The divalent cobalt ion in CoVO₃ shows an S = 3/2 state, whereas a *J*eff = 1/2 state was reported in ilmenite-type CoTiO₃ [2]. The reduction of structural symmetry by V–V dimerization could change the magnetic ground state.



FIG. 1. Crystal structure of $CoVO_3$ at 300 K viewed along the *c* axis and *b** direction. The V-V dimensional direction of the two structures of t

Keywords: dimer, crystal structure transition, magnetism, transition metal oxides

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MM-P09 (Poster)

Magnetic and Structural Properties of Cr_xMn_{1-x}ZnSb with Tetragonal Cu₂Sb-type Structure

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Mn-based ternary ferromagnets with a tetragonal Cu₂Sb-type crystal structure (MnAlGe, MnGaGe, and MnZnSb) are paid attention because of the high uniaxial magnetocrystalline anisotropy [1]. It was reported that Cr-substitution for MnZnSb enhanced Curie temperature $T_{\rm C}$ [2]. On the other hand, when Mn was completely replaced by Cr, CrZnSb also crystallized with the tetragonal Cu₂Sb-type structure. However, CrZnSb had a quite small magnetic moment and did not show ferromagnetic properties. Therefore, in this study, for optimizing magnetic properties of (MnCr)ZnSb, magnetic and structural properties of Cr_xMn_{1-x}ZnSb ($0 \le x \le 1$) were investigated.

 $Cr_xMn_{1-x}ZnSb$ ($0 \le x \le 1$) samples were prepared by reactive sintering. From X-ray diffraction experiments, all samples showed the tetragonal Cu₂Sb-type structure. Magnetization measurements were carried out by a SQUID magnetometer.

Figure 1 shows the thermomagnetization curves of $Cr_X Mn_{1-x} ZnSb$ at x = 0, 0.2, 0.5, and 0.8. *T*_C increased with increasing *x* from 0 to 0.2, and rapidly decreased for x > 0.2. It was found that the maximum value of *T*_C was obtained at 371 K at x = 0.2, which was 15.6% higher than *T*_C of MnZnSb (x = 0).



FIG. 1. Thermomagnetization curves of $Cr_xMn_{1-x}ZnSb$ at 0.1 T.

Keywords: Ferromagnetic compounds, Magnetic properties

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MM-P10 (Poster)

Negative thermal expansion in Mn-doped CaCu₃Fe₄O₁₂

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Quadruple perovskite oxides $ACu_3Fe_4O_{12}$ (A = Ca, Sr, Cd, rare-earth metals, Bi; see crystal structure in Fig. 1) exhibit various functional properties such as negative thermal expansion [1], thermal energy storage, and oxygen evolution reaction catalysis. $CaCu_3Fe_4O_{12}$, whose valence state at room temperature is represented as the ionic model of $CaCu^{-2.4+}$ Fe^{-3.7+} O, undergoes a charge disproportionation transition below 210 K simultaneously with a Fe-to-Cu electron charge transfer ($3Cu^{-2.4+} + 4Fe^{-3.65+} \rightarrow$ $3Cu^{-2.2+} + 4Fe^{-3.8+}$) [2, 3], leading to the discontinuous lattice shrinkage. In the present study, we synthesized $CaCu_3Fe_4-_xMn_xO_{12}$ (x = 0-4) under high-pressure and hightemperature conditions of 12 GPa and 1273 K, and investigated the effect of Mn doping on the structures and electronic properties.

Synchrotron X-ray powder diffraction confirmed that $CaCu_3Fe_{4-x}Mn_xO_{12}$ at all the compositions crystallized in the cubic quadruple perovskite structures. The temperature dependence of the lattice constant is shown in Fig. 2. A negative thermal expansion (NTE) was observed in x = 0.5 at temperatures between 230 and 280 K, obtaining a large negative linear thermal expansion coefficient $\alpha L = -2.28(8) \times 10^{-5} \text{ K}^{-1}$. The Cu–O bond lengths decreased on cooling in the NTE range, whereas the (Fe, Mn)–O bond lengths increased as observed in SrCu₃Fe₄O₁₂ [1]. The direction of the charge transfer between Cu and Fe in CaCu₃Fe₄O₁₂ is inverted by the Mn doping, as indicated by the Cu–O bond shrinkage and (Fe, Mn)–O bond elongation in the NTE temperature range on cooling.



FIG. 1 (left) Schematic of crystal structure of quadruple perovskite *A*Cu₃Fe₄O₁₂.

FIG. 2 (right) Temperature dependence of the normalized lattice constant for CaCu₃Fe_{4-x}Mn_xO₁₂ (x = 0, 0.5). The data for x = 0 were taken from the reference [3]

Keywords: negative thermal expansion, quadruple perovskite oxides, high-pressuresynthesis

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MM-P11 (Poster)

Stabilization of α-FAPbI₃ by addition of dopamine hydrochloride

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Perovskite solar cells using organic-inorganic hybrid perovskite crystals ABX₃ (A: organic cation, B: inorganic cation, X: inorganic anion) as light absorbing materials were developed by Miyasaka et al. in 2009 [1]. MAPbI₃, where MA is a methylammonium ion, i.e., CH₃NH ⁺, is widely studied at an early stage, but its volatility of the organic cation is an issue to overcome for the commercial application. Recently FAPbI₃, where FA is a formamidine ion CH(NH) ⁺ has been actively studied as an absorbent material to improve stability at high temperature. Although FAPbI₃ has improved thermal tolerance compared to MAPbI₃, non-active δ -FAPbI₃ appears as the stable phase at room temperature in air, rather than the photoactive phase, α -FAPbI₃,

which is stable at high temperature. Therefore, maintaining α-FAPbI₃ at room temperature in air is a major challenge. It was reported that introduction of dopamine the hydrochloride (hereafter DA) as an additive to MAPbI₃ can improve energy conversion efficiency and isolation from the external environment [2]. In the present study, DA is introduced as an additive to FAPbI₃, and the effect of DA addition on the phase stability of FAPbI3 at room temperature in air is evaluated using X-ray diffraction (XRD) measurements. Observed XRD patterns are shown in Fig. 1, which shows that pure FAPbI₃ (DA:0 mM) undergoes a phase transition to δ -FAPbI₃ after one day from preparation, while the one with DA (20 mM) can maintain α -FAPbI₃ for about a month after preparation.





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MM-P12 (Poster)

Evaluation of structural stability of quadruple perovskites $RMn_3Al_4O_{12}$ (R = Ce, Pr, Eu, Dy, Y, Yb)

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Quadruple perovskite oxides (general formula: AA'3B4O12) are one of the cationordered perovskites, in which the original A-sites are divided into icosahedral (A) and pseudosquare (A') coordinated sites. Unlike simple perovskite of ABO3, the three distinctive crystallographic sites of A-, A'-, and B-sites inhibit the rational evaluation of the structural stabilities solely from the tolerance factor. The global instability index [1], G, calculated by the bond valence sums can evaluate the structural stability, regardless of its structure type. In the previous study, G of RMn3Al4O12 series (R = rare-earth metals) were predicted by using SPuDS software, obtaining relatively stable compounds with G < 0.2 v.u. (R = Dy, Y, Yb), although those with higher G (R = La) could not be synthesized [2]. In the present study, we investigated the structural stability of RMn3Al4O12 (R = La, Ce, Pr, Eu, Dy, Y, Yb) using G obtained from the experiment.

RMn3Al4O12 (R = Ce, Pr, Eu) were successfully obtained at high pressures up to 12 GPa. The powder X- ray diffraction (XRD) patterns were indexed with the cubic quadruple perovskite structure as well as the reported RMn3Al4O12 (R = Dy, Y, Yb). The structure parameters were refined by Rietveld analysis of the synchrotron XRD data calculating the experimentally determined G (Gexp). The Gexp demonstrated а parabolic dependence on ionic radii of the R ion as well as RCu3Fe4O12 series [3]. We found that the Gexp is related to the lower limit of the synthesis pressure (PL), which was determined by synthesis experiments at various pressures from 3 to 12 GPa. Figure 1 depicts the results of synthesis as a function of Gexp for RMn3Al4O12. The PL increased almost linearly with Gexp, indicating that higher pressures are needed



FIG. 1 The results of synthesis of $RMn_3Al_4O_{12}$. Open circles indicate that the quadruple perovskite phases were successfully obtained, whereas solid triangles denote that samples that the quadruple perovskite phases could not be obtained. The dashed line represents the lower limit of thesynthesis pressure

to stabilize the compounds with severe instability with larger Gexp. This finding provides a valuable guide to designing the optimum synthesis conditions.

Keywords: quadruple perovskite, structural stability, high pressure

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MM-P13 (Poster)

Magnetic and Hyperfine properties of MnCoGe-MnFeGe system

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MnCoGe-based compounds exhibit giant magnetocaloric effect due to first order magnetic phase transition accompanied by structural phase transition. Mn1-*x*FexCoGe and MnCo1-*x*FexCoGe show magnetostructural phase transition around room temperature¹. However, magnetostructural transition is affected by both Fe content and the heat treatment condition greatly. Noguchi *et al* reported that the structural phase transition temperature changed at different heating temperature². In this study, to clarify the microscopic information of Fe substitution effect on MnCoGe, ⁵⁷Fe Mössbauer spectroscopy measurements, X-ray diffraction experiments, and magnetization measurements were performed for MnCo1-*x*FexCoGe ($0.2 \le x \le 1.0$).

The hyperfine property of Mn-site and Co-site was evaluated by ⁵⁷Fe Mössbauer spectroscopy at 15 K. Figure 1 shows the Fe content *x* dependence of isomer shift *IS* (a) and quadrupole splitting QS (b). Increase of *x* influences effectively for isomer shift of Mn-site rather than Co-site. Meanwhile, sign of QS at only Co-site changed. Therefore, it is suggested that Fe-substitution effectively changed electron distribution at Mn-site, and electric field gradient around Co-site.



FIG. 1 Fe content x dependence of isomer sift IS (a) and quadruple splitting QS (b) of $MnCo_{1-x}Fe_xGe$ (0.5 $\leq x \leq 1.0$)

Keywords: Mössbauer spectroscopy, MnCoGe, magnetic measurement

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Transition metal (TM) luminescent centers have been played an essential role in lightemitting devices such as LEDs and laser devices. The partially occupied 3d or-bitals of TM ions lead to exhibit many electron-configurations and transitions between these configurations (multiplets) observed in visible light region. In order to efficiently design new light-emitting materials, the multiplet energy levels should be predicted by firstprinciples calculations. However, first-principles calculations usually refer to the ground state properties. It is thus necessary to develop a first-principles method for ex- cited state properties without semi-empirical parameters nor specific models.

To go beyond usual local density approximation in density functional theory, we employed quasiparticle self-consistent GW (QSGW) method implemented in ecalj package developed by Kotani et al[1]. Specifically, we used QSGW80 method to quantitatively reproduce band gap energy of various semiconductors[2]. The localized 3d electron in TM luminescent centers are described using local atomic model Hamiltonian consisted of crystal-field (CF) terms and effective Coulomb interaction terms. This model Hamiltonian can be described by CF parameters and Slater-Condon parameters. These parameters are determined so that eigenvalues of the model Hamiltonian with Hartree-Fock approximation reproduce the QSGW80 TM 3d band structure as exactly as possible. As a confirmation, we first estimated Slater-Condon parameters of free TM ions. We found that our method gave comparative values compared with experimental ones. Next, we focus on Cr3+ in α -Al2O3. Our method also gave reasonable



FIG. 1 (a) Tanabe-Sugano diagram, (b) multiplet structure by QSGW80, (c) multiplet structure by experimental analysis [3] of Cr^{3+} in α -Al₂O₃

values of parameters in comparison with experiment. Furthermore, we show the Tanabe-Sugano diagram in FIG. 1. This diagram shows CF dependence on multiplet energy levels. The multiplet structure obtained by QSGW80 qualitatively reproduces that of experimental analysis[3] in the low energy region (FIG. 1(b) and (c)). Finally, we obtained the chemical trend of TM ions in α -Al₂O₃. It shows that our method reproduces the trend of experimental works.

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MM-P15 (Poster)

Site selectivity of Cu or Fe in Cu-doped or Fe-doped MnCoGe

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With decreasing temperature, MnCoGe alloys undergo a structural transition from Ni2In-type hexagonal structure ("Hex") to TiNiSi-type orthorhombic structure ("Orth") at Tstr = 420 K and a magnetic transition from paramagnetic state to ferromagnetic one at TC = 355 K [1,2]. Partial substitution of Cu or Fe for Mn or Co in MnCoGe alloys brings about stabilization of "Hex" and results in a decrease in Tstr [3-5].

Our results of DFT calculations for $(Mn_{1-x}X_x)CoGe$ and $Mn(Co_{1-x}X_x)Ge$ (x = 0.125) have shown that "Hex" is more stable for $(Mn_{1-x}Cu_x)CoGe$. This fact is not in agreement with experimental results [5]. In the case of $Mn(Co_{1-x}Cu_x)Ge$, we cannot determine whether "Hex" or "Orth" is more stable. In the case of Fe substitution, "Orth" becomes more stable in both cases, which is not consistent with the experimental result [3] (Fig. 1). In order to investigate the reason for this disagreement between experiment and theoretical results, we have performed DFT calculations for structures containing Cu or Fe at both Mn and Co sites.



FIG. 1 Change of ΔE ($\Delta E = E_{\text{Hex}} - E_{\text{Orth}}$) with Cu or Fe substitution for Mn or Co site

Keywords: Cu-doped, Fe-doped, MnCoGe, Ni₂In-type, TiNiSi-type

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MM-P16 (Poster)

Structures and Magnetic Properties of Novel Quadruple Perovskite Oxides LaMn₃Ru₂Mn₂O₁₂ and LaMn₃Ru₂Fe₂O₁₂

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A-site ordered quadruple perovskite oxides, AA'3B4O12, exhibit various functional properties such as giant dielectric constants and room-temperature magnetoresistance, most of which are related to their structural feature with two distinct crystallographic sites of A'- and B-sites for transition metals (Fig. 1). Further cationic ordering at B-sites may be achieved in selected couples of B-site ions, as represented in the formula AA'3B2B'2O12, which leads to intriguing physical properties. LaMn3Cr4O12 has A'-site Mn and B-site Cr ions coupled antiferromagnetically at each site,¹ whereas LaMn3Ni2Mn2O12 has antiferromagnetically ordered A'-site Mn ions and ferromagnetically ordered B-site Ni and Mn ions in an orthogonal spin order.² The LaMn3B2B'2O12 family is composed of only 3d metals, forming independent A'- and B-site magnetic sublattices. The effects of 4d or 5d metal ions on the magnetic properties in LaMn3B2B'2O12 have not been clarified yet. In this study, we aimed to elucidate the magnetic interactions of new 3d-4d hybridized quadruple perovskite oxides of LaMn3Ru2M2O12 (M = Mn, Fe), which are synthesized under high-pressure and high-temperature conditions.

The synchrotron X-ray diffraction patterns of LaMn3Ru2M2O12 (M = Mn, Fe) confirmed that the main phases were crystallized in a cubic quadruple peroxskite structure without *B*-site ordering between Ru and *M* ions. X-ray absorption spectroscopy revealed that their valence states are interpreted as the ionic models of LaMn³⁺ Ru³⁺ M^{3+} O (M = Mn, Fe). Both compounds exhibited no long-range magnetic ordering (Fig. 2). We conclude that the competition between ferromagnetic and antiferromagnetic interactions in *B*-site ions prevented the formation of long-range magnetic ordering of A'-site Mn ions which are originally independent in the distinct magnetic sublattices.





FIG. 1 Schematic of crystal structure of $AA'_{3}B_{4}O_{12}$ -type quadruple perovskite oxides

FIG. 2 Isothermal magnetization curves for $LaMn_3Ru_2M_2O_{12}$ (M = Mn, Fe) at 5 K

Keywords: Magnetism, Quadruple perovskite oxides, High-pressure synthesis

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MM-P17 (Poster)

The epitaxially grown ferroelectric Hf_{0.5}Zr_{0.5}O₂ thin film using pulsed laser deposition method

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The ferroelectric random acces memory (FRAM), one of the nonvolatile memories, has powerful advantages like a fast access time and a low power consumption which allows the FRAM to be a promising technology in memory field. Most researches associated with the advance of the FRAM technology are focused on PbZr_{0.52}Ti_{0.48}O₃ (PZT) due to its large remnant polarization and low coercive voltage. However, scailing down a thickness of PZT under 50 nm cause significant leakage current in a FRAM device which is triggered by low bandgap and sidewall problem. HfO₂-related materials are considered to be emerging ferroelectric materials for FRAM devices due to their excellent compatibility with CMOS process. In this study, pulsed laser deposition (PLD) method is used to deposit 10-nm-thick Zr-doped HfO₂ (HZO) films and La_{0.67}Sr_{0.33}MnO₃ (LSMO) bottom electrodes on the single crystalline SrTiO₃ (STO) substrates. The peaks from X-ray diffraction (XRD) demonstrate that the ferroelectric HZO film with orthorhombic phase is epitaxially grown on the STO substrate. The HZO thin film with very flat surface reveals large remnant polarization of up to 22µC/cm² measured by using positive-up negative- down (PUND) pulsed method. Therefore, ferroelectric properties of HZO thin film provide an opportunity to overcome the scaling down problems in the FRAM technology.

Keywords: PLD, ferroelectric, HZO, heterostructure



MM-P18 (Poster)

Effect of annealing on the exchange interaction between magnetic phases and the exchange bias effect in Co/CoO nanocomposite

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In this study, Co particles were synthesized by a polyol method and a post annealing in air was performed to prepare Co/CoO nanocomposite. The sample was characterized by scanning electron microscopy, X-ray diffraction, and magnetic property measurements. To determine the exchange bias field, the sample was cooled in a magnetic field of 50 kOe and measured hysteresis loops at 5 K. The effect of annealing on the exchange interaction between the Co ferromagnetic phase and CoO antiferromagnetic phase, which in turn affects the exchange bias effect in this material, was investigated. It showed that annealing enhanced the exchange interaction between two phases leading to an improvement of the exchange bias field (Heb) in comparison to the non-annealed sample. A value of $H_{eb} = 155$ Oe was obtained with annealing at temperature of 300 °C in 90 minutes. The dependence of H_{eb} on the exchange interaction between phases is discussed.





FIG. 1 The exchange bias field of samples annealed at 300 °C in different annealing time

MM-P19 (Poster)

Thermodynamic properties of bilayer honeycomb spin lattice

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Magnetic order of the few layers honeycomb spin lattice films shows the thickness and field dependence. For example, the monolayer CrI3 is ferromagnetic (FM) while the double layer one is antiferromagnetic (AF) as shown by the magneto-optical Kerr- effect measurement [1]. In this research, the thermodynamic properties of the honeycomb spin lattice consisting of two FM layers interacting by AF exchange interaction are investigated using the Heisenberg XZ-model in the longitudinal and transverse field [2], which is an extension of the previous transverse Ising model [3].

Expressions for the temperature and field dependent free energy, spin wave spectra (Magnon), magnetization are obtained and analyzed numerically. Some consequences like the magnetization process in the field perpendicular to the bilayer film plane is discussed and compared with experiment.

Keywords: thermodynamics, films, magnetization, magnon

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Design and Experimental Investigation of Piezoelectric Ceramic Element PZT Application for High-Power Ultrasonic Welding Transducer

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Currently, ultrasonic welding technology is developing rapidlyand is widely applied in many fields: science and technology, agriculture, environment, medicine, industry, and especially applied in industrial transmission lines non-woven fabric industry. In the world, the application of piezoelectric ceramic materials in ultrasonic welding has many manufacturers and traders of piezoelectric ceramic elements, and many scientific works have published different types of piezoelectric ceramics depending on the purpose of the application. In this research, we have researched and fabricated piezoelectric ceramic materials described by the general formula PZT. Thepiezoelectric ceramics were fabricated by solid-state reactiontechnique, sintering was carried out for 2 hours at temperatures ranging from 1100oC to 1200oC. The existence of sintering temperatures on microstructures and hysteresis rings of ceramic systems has been investigated. The crystalline phase and microstructureofthesintered compositions were investigated in detail using X-ray diffraction (XRD) analysis and scanning electron microscopy (SEM). Dielectric properties, such as dielectric constant (ϵr) and dielectric loss (tan δ) have been measured. The room temperature hysteresis loops of all unpolarized sintered samples show similar behavior to the "hard" PZT ceramic. The phase transition temperature (Tc) and piezoelectric properties such as electromechanical coupling coefficient (Kp), quality factor (Qm), and piezoelectric strain constant (d33) have been studied and presented in detail.

Keywords: PZT, piezoelectric ceramic, ultrasonic welding transducer.



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MM-P21 (Poster)

Study of Metal - Insulator transition in complex perovskite for sensingapplication

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Metal-insulator materials get great interest by its applicability in sensing application [1], [2]. Complex ferromagnetic-ferroelectric perovskites ([3],[4]) possess the multiferroic behavior and the metal-insulator transition (MIT), which can be adjusted for report study sensing application. In this we MIT behavior of $[La_{0.7}Sr_{0.3}Mn_{0.98}Co_{0.02}O_3]_{(1-1)}$ $_{x)}[(La_2NiO_4)_{0.9}(BaTiO_3)_{0.1}]_x$ or (LSMCO)_{1-x}(LNBT)_x perovskite with x = 0.0; 0.1; 0.2; 0.3; 0.4. Samples are prepared by the ceramic technology method with high energy ball milling. LNBT is prepared firstly by solgel method and has core-shell structure. The thermomagnetic curves are measured by PPMS system. Ferroic behavior is clearly observed in x=0.4 sample. Temperature dependence of resistivity is recorded by four probe method and conducting mechanism is discussed using the two components: small Polaron and large Polaron concept. Potential for sensing application of MIT in some samples is pointed out.





FIG. 1 Thermomagnetic curves of $(LSMCO)_{(1-x)} LNBT_x$ with x = 0.0, 0.1, 0.3, 0.4. Inset shows the curve of x = 0.2 sample

FIG. 2 The dependence of resistivity on temperature of x = 0 sample

Keywords: Ferroic, small polaron, large polaron.

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MM-P22 (Poster)

Tuning Independently The Exchange Bias and Coercivity in Toppinned and Bottom-pinned Co/Pd Multilayers by FeMn Film

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The interlayer coupling between a ferromagnet (FM) and an antiferromagnet (AFM) as well as the spin structure at such FM/AFM interface play an important role in magnetic properties, giving rise to various intriguing phenomena [1, 2] therefore being the key elements of novel spintronic systems. In this study we investigated perpendicular exchange bias (H_{EB}) and coercivity (HC) in Co/Pd multilayers (MLs) with an FeMn film at the bottom or on top. These MLs were fabricated by DC magnetron sputtering, the structural and the magnetic properties were characterized by X-ray diffraction (XRD) and Vibrating sample magnetometer (VSM). The results clearly shows that magnetic properties (e.g. H_{EB}, HC) of the top- and bottom-pinned systems are remarkably different. The large perpendicular H_{EB} is observed for the top-pinned [Co/Pd]₅/Co/FeMn system but not for the bottom-pinned FeMn/[Co/Pd]₅ system. However, in term of the HC, the HC of the bottom-pinned FeMn/[Co/Pd]₅ system is nearly two times higher than that of the top-pinned [Co/Pd]₅/Co/FeMn system which somehow is almost unchanged in comparison with the [Co/Pd]₅ system. Some physical models were used for visualization of the FM and AFM interaction as well as the spin configuration in these systems [3-6]. This approach opens up the way for tuning the magnetic coupling in top-pinned and bottom-pinned Co/Pd MLs by FeMn in order to tune their H_{EB} and HC independently.

Acknowlegdement

This work was financial supported by Vietnam Academy of Science and Technology (VAST) under Project NCXS01.04/22-24.

Keywords: [Co/Pd]₅ multilayers, FM/AFM interaction, FM/AFM surface, perpendicular exchange bias, coercivity

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MM-P23 (Poster)

Correlation between structure and electromagnetic properties of some high permeability amorphous Fe and Co based alloys

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Several high permeability soft magnetic materials from the iron and cobalt based alloys $Fe_{73.5}Si_{13.5}$ (BNbCu)₁₃ [1], $Co_{65.5}Fe_{3,5}$ (CrBSi)₃₁ [2] systems have been produced by the rapid quenching method with the temperature cooling rate about 10⁶ °C/s. X-ray analysis showed a complete amorphous structure of the as cast ribbons. The crystallization activation energy and crystallization process of the samples are determined using DSC 8231 equipment (Fig.1). DSC analysis results show that in the Iron based magnetic ribbons, α -Fe crystallization phase appears in the temperature range of 500-575°C. The α -Fe phase crystallization activation energy has been determined about 420 kJ/mol according to the Kissinger method. A significant reduction in the resistivity value ρ was observed due to crystallization upon thermal annealing, leading to the formation of a nanocrystal state in the alloys. The dynamical properties of the samples are also investigated. Magnetic permeability of the as- cast cobalt-based ribbon changes a little in the frequency range up to 100 kHz (Fig.2), while initial permeability reaches about 1.7×10⁴.



FIG. 1: DSC curve of the typical as-cast C065.5Fe3.5 (CrBSi)31 ribbons.

FIG. 2 Frequency dependence of the magnetic permeability of the typical as-cast Co_{65.5}Fe_{3.5} (CrBSi)₃₁ ribbons.

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Keywords: amorphous ribbons, DSC, permeability

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MM-P24 (Poster)

Tunable Electronic Properties of Novel 2D Janus MSiGeN₄ (M = Ti, Zr,Hf) monolayers by strain and external electric field

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Since the recent successful experimental synthesis of MoSi₂N₄ [1], the "MA₂Z₄ family" has been of particular interest to the scientists in the field of materials science due to its outstanding physical properties [2]. In addition, the vertical symmetry breaking in the two-dimensional (2D) layered nanostructures has given rise to many novel properties that do not exist in the symmetry materials [3–5]. Hence, we propose novel 2D Janus MSiGeN₄ monolayers (M = Ti, Zr, and Hf) and investigate their structural, elastic, and electronic properties using the first-principles calculations. The calculations of phonon spectra indicate that monolayers MSiGeN₄ are dynamically stable and can be experimentally synthesized. The obtained Young's modulus and Poisson's ratio of the Janus structures MSiGeN₄ are much larger than that of other binary 2D materials and meet the mechanical stability criteria suggested by Born and Huang. In the calculations using either PBE or HSE06 functionals, the Janus MSiGeN₄ structures exhibit indirect semiconductor characteristics with larger band gaps than that of similar septupleatomic-layer materials, such as MoSiGeN₄ and WSiGeN₄. In addition, the influences of biaxial strain and external electric field on the electronic structure of $MSiGeN_4$ are investigated. It is found that the biaxial strain tunes the electronic characteristics more significantly than the external electric field. The obtained results could provide insights into novel Janus monolayers with potential applications in electronic devices.

Keywords: 2D Janus structures, Electronic properties, First-principles calculations

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MM-P25 (Poster)

Critical exponents of La2/3Ca1/3MnO3 nanoparticles

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It is known that a La_{2/3}Ca_{1/3}MnO₃ (LCMO) perovskite-type compound shows the colossal magnetoresistive (MR) and magnetocaloric (MC) effects associated with a first-order phase transition. Upon fabricating nanostructures and doping a transition/rare-earth metal in it, one can remarkably improve the effects for application aspects [1,2]. Apart from the MR and MC related changes, it has also been observed the magnetic-order transformation from the first type to the second one. However, the variation tendency of critical exponents around the ferromagnetic-paramagnetic transition is less taken into account. This work presents a detailed study on the variation tendency in value of critical exponents (β and γ) of LCMO nanoparticles as changing the crystallite size t from 40 to \sim 100 nm. We have found LCMO showing a critical size (tc) of the first-to-second-order transition with $t_c \approx 70$ nm, corresponding to tricritical-point exponents. Decreasing t smaller than t_c , we have found the value change of the exponent parameters towards the 3D-Heisenberg and mean-field models. This proves the establishment of long-range magnetic order as decreasing t. Analyses of X-ray absorption spectra and electron microscopy images have indicated local structural distortions and the presence of magnetic-dead layer. These are thought to be important factors that have directly influenced magnetic order of nanoparticles.

Keywords: Perovskite manganite; Nanoparticles; Critical behavior

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MM-P26 (Poster)

Electric Field Control of Magnetization in Artificial Microporous Magnetic Structure Based Multiferroics

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Straintronics is currently a new bullet research area in solid state physics, at which strain-induced effects in solids are employed to establish next generation gadgets for information, energy transducer, and sensor. Achieving the comprehension of physical mechanisms is always desirable and in high demand for future straintronic devices and research standpoints. This letter will present an unconventional route to tune the magnetic anisotropy of the multiferroic systems of PZT/FeCSi by modulating the applying electric field. Thanks to extremely stress-sensitive characteristic of the microporous structure in FeCSi alloys, under the electric fields up to 20 kV/cm applied across the PZT substrate, the extrinsic magnetic properties purposely, i.e., magnetic susceptibility, magnetization, coercive force, and magnetic energy density can be easily tuned. The dependence of mentioned magnetic properties as a function of the electric field is then confirmed by phase-field simulation. Both experimental and phase-field simulated results shows the stress-activated domain process, where, the degree of electric field response strongly depends on the dimensions of microporous. This work will provide new pathways and additional degrees of freedom in understanding and tailoring the straintronics phenomena for future artificial straintronic devices.



FIG. 1. Micromagnetic simulation of magnetic domain in FeCSi microporous structure (a) and magnetization loops measured at various applied electric fields (b).

Keywords: Straintronics, Magnetoelectric, Multiferroics, Microporous structure, Magnetic domain.

MM-P27 (Poster)

Micro-patterned Configuration Controlling GMI Effectin Magnetic Artificial System

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Giant magnetoimpedance (GMI) effect has drawn intensively attention to the magnetic field sensing technology owing to outstanding sensitivity, resolution, and accuracy. Inspired by the artificial structure, the GMI effect in triangular spiral systems with various branch-widths (*w*) can be tuned and improved thanks to the shape anisotropy field optimization accompanied by the magnetic domain properties. The GMI effect is enhanced up to 250% under applied magnetic field of 100 Oe in the sensor with $w = 60 \ \mu m$, much higher than that of 5% in $w = 250 \ \mu m$ sensor. The magnetic characterizations demonstrate that the improvement of magnetic susceptibility at low field when the width is reduced is correlated to the shape anisotropy properties. The simulation shows that depending on the applied field directions, various closure magnetic domain configurations can be formed owing to the competition of anisotropy and Zeeman energies followed in the Stoner-Wohlfarth model. These results agree well with the augmentation of GMI effects assisted the transverse domain formations at critical dimension for micro-patterned magnetic system. These make the GMI spiral sensors a promising candidate for miniaturization of sensing magnetic applications.



FIG. 1. Micro-patterned triangle spiral magnetic systems (a), micromagnetic simulation of domain formation (b), and characterised with giant magnetoimpedance effect.

Keywords: Magneto-impedance, magnetic sensor, micro-coils, magnetic domain

MM-P28 (Poster)

Fundamentally Different Magnetoresistance Mechanisms in Related Co/Pd and Co/Pt Multilavers for Spintronic Applications

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The mechanisms of electron and spin transport in 3d ferromagnet (FM)/5d(4d) heavy metal (HM) multilayers (MLs) are of great interest for spintronics due to the high potential of such structures for magnetic recording [1]. Among them are Co/Pt and Co/Pd MLs with strong and tunable perpendicular magnetic anisotropy, which are widely studied for magnetic memory applications. Despite their high similarity, these MLs have been found to show completely different magnetoresistance (MR) mechanisms. The magnon MR, which dominates in the Co/Pd MLs [2], is almost completely suppressed in the Co/Pt-based structures. Inversely, strong anomalous and spin Hall effects determine the spin-dependent electron transport in the Co/Pt MLs, while they are definitely less significant for the MR of the Co/Pd MLs. Additional intriguing mechanisms appear only in the Pt-containing MLs, which are related to the enhanced contribution of the Lorentz-like MR mechanism at room temperature, as well as to the manifestation of anisotropic MR in a magnetic field rotating perpendicularly to the current. The origins of such differences between the MR mechanisms of the Co/Pt and Co/Pd MLs, which are supposed to lie in their interface structure, as well as in band structure of HM, have not yet been clearly elucidated. A comprehensive analysis of the MR and Hall voltage of both MLs in a wide range of temperatures T =3-300 K, as well as the values and orientations of magnetic field B = 0.9 T, was carried out in correlation with the structural analysis to clarify the principles of the emerging mechanisms.

Acknowledgment

This work was financial supported by Vietnam Academy of Science and Technology under Project OTBY01.02/23-24 and Belarusian Republican Foundation for Fundamental Research (project no. F23V-003).

Keywords: multilayered films, perpendicular magnetic anisotropy, magnetoresistance, magnons, spin Hall effect

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MM-P29 (Poster)

Spin Dynamics in Ferromagnetic Thin Films

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Spin dynamics over wide timescales from ultraslow to ultrafast regions has attracted much attention due to possible application for ultrafast-operating and long-term stable spintronic devices. In this talk, several examples of spin dynamic phenomena will be introduced such as unusual minor hysteresis loop behaviors involved with domain wall creeping behavior and nonequilibrium spin dynamics on femtosecond timescale linked with mutual dynamics among spin, electron, and lattice in the ferromagnetic materials. THz emission behavior involved with ultrafast spin dynamics will be also covered.



FIG. 1. THz emission profiles from Ni, Fe, and Co films with variation of pump fluences. [1]

Keywords: Magnetism, THz emission, Ultrafast spin dynamics

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