Seebeck and peltier effect pdf file free online version

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## Seebeck and peltier effect pdf file free online version

1. SEEBeck EFFECT & PETLIER EFFECT PRESENTED BY: VISHAL CHATURANI BSc(Engg.) MECHANICAL ROLL NO.- 114089 S No.-105 2. Thermoelectric phenomena: • the Seebeck effect, • the Peltier effect, and • the Thomson effect. 3. SEEBECK EFFECT Differential Thermocouple Vab  $\alpha = \Delta T 4$ . SEEBECK EFFECT • The Seebeck effect is the conversion of temperature differences directly into electricity • It is named after German physicist Thomas Johann Seebeck • in 1821 he discovered that a compass needle would be deflected by a closed loop formed by two metals joined in two places, with a temperature difference between the junctions. • This was because the metals responded differently to the temperature difference, creating a current involved, so he called the phenomenon the thermomagnetic effect. Danish physicist Hans Christian Ørsted rectified the mistake and coined the term "thermoelectricity". 6. Cause of seebeck effect • As we know that no of free electrons per unit volume and their average velocity varies from one metal to another . • At the junction of two dissimilar metals, electrons migrate in one direction across the junction. • It sets up an opposing electric field . • As a result a fixed potential difference is developed . 7. • This effect is called thermo e.m.f • The current which flows along the closed circuit is called thermo electric current 8. Cause (Contd.) • Since a migration of charges creates an electric potential, the buildup of charged carriers onto the cold side eventually ceases at some maximum value since the electric field is at equilibrium. • An increase in the temperature difference resumes a buildup of charge carriers on the cold side, leading to an increase in the thermoelectric voltage, and vice versa. 9. The voltage created by this effect is on the order of several micro volts per Kelvin at room temperature. S is positive when the direction of electric current is same as the direction of thermal current 10. Thermoelectric series • For a given temperature range, the thermo e.m.f induced is different metal combinations. • Seebeck arranged 35 metals in a series in such away when any two form a thermocouple:- • Current will flow through the cold junction from the one that occurs first in series to the other that occurs later in series • Between 0oC & 1000C the thermoelectric series is: Antimony, Fe, Cd, Zn, Ag, Au, Rb,Mo,Cr,Sn,Pb,Hg,M n,Cu,Pt,Cob, 11. Variation of thermoelectric E.M.F. with temperature(constant for a given thermocouple) • Ti =inversion temperature(constant for a given thermoelectric E.M.F. with temperature(constant for a given thermoelectric E.M.F. with temperature) • Ti =inversion temperature(constant for a given thermocouple) • Ti =inversion temperature(constant for a given thermoelectric E.M.F. with temperature) • Ti =inversion temperature(constant for a given thermocouple) • Ti =inversion temperature(constant for a given thermoelectric E.M.F. with temperature) • Ti =inversion temperature(constant for a given thermocouple) • Ti =inversion temperature) • Ti =inversion temperature(constant for a given thermocouple) • Ti =inversion temperature) • Ti =inversion temperature(constant for a given thermocouple) • Ti =inversion temperature(constant for a given temperature(c the junction of the thermocouple is opened and a third metal is inserted, the e.m.f. for the couple AB remains the same temperatures t1 and t2 is equal to the sum of e.m.fs (seebeck) for any number of successive steps into which the given range of temperatures may be divided. Thus  $e^{\theta}1^{\theta}n = e^{\theta}1^{\theta}2 + e^{\theta}3^{\theta}4 + \dots + e^{\theta}n^{-1}n^{$ Current starts flowing, making the junction hot. Temperature difference is obtained. Open K1 and close K2. Deflection is shown by galvanometer, thus confirming the seebeck effect 15. Peltier Effect Electric Current Difference in Material A Heat Absorbed or EF between Expelled Materials A and B Material B 16. Peltier effect • In 1834, a French watchmaker and part time physicist, Jean Peltier found that an electrical current would get hot while the hot junction w would get cold. 17. Cause of peltier effect When a current flows across the junction of two metals, it gives rise to an absorption or liberation of heat, depending on the direction of the current. i.e. Applying a current (e- carriers) transports heat from the warmer junction. 18. As an example of the Peltier effect, consider the circuit shown here. Under these conditions, it is observed, as indicated in the diagram, that the right-hand junction is heated. It shows, in other words, that electrical energy is being transformed into heat energy at the left one. 19. • The rate of evolution of or absorption of heat is proportional to the current & thus the peltier effect is also reversed. • The energy absorbed or evolved at one of the junctions of the two dissimilar metals when one ampere of current is changed then the peltier coefficient , denoted by 1 20. In this experiment, two rods of bismuth and antimony are joined as shown. A differential air thermometer is placed with its two bulbs at junctions A and B. 21. When no current flows through the thermocouple , the junctions A & B are at same temperature. Also Hg(mercury) pellet in the thermometer lies symmetrically at the centre. When large current is passed through the thermocouple, then there will be: • Evolution of heat at junction A. • Absorption of heat at junction B. • Hg pellet moves in the opp. Direction indicating that now A has become cold and B has become hot. 22. Applications Seebeck effect • The Seebeck effect is used in the thermoelectric generator, which functional power (a form of energy recycling). • in automobiles as automotive thermoelectric generators (ATGs) for increasing fuel efficiency. generators with the same mechanism but using radioisotopes to generate the required heat difference. 23. Peltier effect • The Peltier effect • The Peltier effect are frigerators are useful in applications where their advantages out weigh the disadvantage of their very low efficiency. 24. Applications Water/Beer Cooler Cryogenic IR Night Vision Si bench TE Electronic Cooling 25 25. Temperature measurement • Thermocouples and thermopiles are devices that use the Seebeck effect to measure the temperature difference between two objects, one connected to a voltmeter and the other to the probe. • The temperature of the voltmeter, and hence that of the material being measured by the probe. 26. THANK YOU when two metal wires of different materials were joined to form an electrical current was made to flow through the circuit, one junction would become cold and the other junction would become hot.indicated in the diagram, that the right-hand junction is heated. It shows, in other words, that electrical energy is transformed into electrical energy at the left junction, thereby causing it to be cooled. . If the current happens to flow in the same direction as the current that is produced by the Seebeck Effect at the hot junction (T2), heat will be absorbed, whereas at the cold junction (T1) heat will be liberated. The effect 1., but is less bulky, has no moving parts, and is typically more expensive and less efficient. can be measured separately using cold junction compensation techniques. DOI:
10.1039/D1EE00667C (Paper) Energy Environ. Sci., 2021, 14, 3480-3491 Received 3rd March 2021 , Accepted 28th April 2021 The spin Seebeck effect (SSE) refers to a direct energy conversion of heat to electricity that exploits magnon current in a ferromagnet (FM)/normal metal (NM) heterostructure. Since SSE is a transverse thermoelectric effect, it can be employed to realize transverse thermoelectric devices, which could potentially overcome the inherent limitations of conventional Seebeck effect-based devices. Yet, the widespread use of SSE is currently hindered by its low conversion efficiency. The thermoelectric efficiency of SSE can be improved through an optimization of bulk FM as well as FM/NM interface. A bulk FM can enhance the thermoelectric efficiency by introducing a 'phonon-crystal' concept, wherein scattering centers selectively scatter the heat-carrying phonons while not affecting the magnons that contribute to SSE. A high-quality FM/NM interface can boost the spin pumping across the interface as well as the electrical transport in NM. Here, by adopting polycrystalline nickel-ferrite (NFO)/Pt as a platform, we demonstrate the successful optimization of bulk FM together with FM/NM interface via a simple heat treatment scheme. The phase separation of NFO via such heat treatment results in a distinctive hierarchical microstructure of nano-sized NFO embedded in micro-sized NiO precipitates; this structure selectively scatters phonons while barely affecting magnons, leading to reduced thermal conductivity without variation of spin Seebeck coefficient. The simultaneous formation of a clean and smooth NFO/Pt interface provides efficient spin pumping at the interface and high electrical conductivity of Pt. As a result, the energy conversion efficiency of SSE in NFO/Pt system can be applied to design highly efficient oxide-based SSE devices. Thermoelectric (TE) conversion offers a carbon-free power generation from waste heat, which can contribute to mitigating both energy and climate challenges. The current TE power generation relies on the charge Seebeck effect, which requires a complex device geometry. It is possible to make the device structure much simpler if the TE device can be constructed by utilizing a transverse TE effect where a charge current is possible to make the device structure much simpler if the TE device structure much simpler if the terms at the structure much simpler structure much sin structure much simpl generated in the direction perpendicular to the applied temperature gradient. The spin Seebeck effect (SSE) is a suitable candidate for realizing such a transverse TE devices is still 3 to 4 orders of magnitude lower than that of the conventional TE devices. Since the figure of merit of an SSE device depends on multiple factors, optimizing it has been considered quite challenging. Here, using a polycrystalline nickel-ferrite (NFO)/Pt bilayer as a platform, we successfully demonstrate a simultaneous optimization of the magnon and phonon transport in bulk NFO and the NFO/Pt interface quality, two crucial factors that determine the figure of merit, via a simple heat treatment scheme. Our study paves a new route towards designing efficient transverse TE devices based on SSE. Introduction The daunting global climate challenges urge deployment of carbon-neutral technologies in each industrial sector. As for the energy sector, an energy conversion process is vital to produce any useful form of energy, e.g., electricity, but at the same time has been a significant source of carbon emission.1 As one potential way to realize a carbon-free power generation from waste heat, thermoelectricity has long been researched over the past century. Thermoelectric power generation is based on the Seebeck effect, wherein a direct conversion of heat-toelectricity happens through an all solid-state device. Thermoelectric devices do not involve any moving parts, working fluids, or chemical reactions, which allows reliable, noiseless, and carbon-free operation.2,3 All of these advantages place the thermoelectric conversion as one of promising next-generation technologies. A typical structure of conventional thermoelectric devices comprises many pairs of n and p-type semiconducting materials connected thermally in parallel and electrically in series. Since the Seebeck effect is a longitudinal thermoelectric effect wherein a charge current is generated in the direction parallel to an applied temperature gradient, the use of both n and p-type materials is required to return the induced charge current to an isothermal plane. Such a longitudinal geometry is inherently complex and thus less manufacturing-friendly. In addition, the requirement of parallel heat and charge flows makes overall performance largely dependent on intrinsic material properties rather than device design parameters. However, an optimization of thermoelectric material is challenging due to the counter-indicated nature of thermoelectric devices.4,5 These inherent limitations for longitudinal devices will persist regardless of any future research progress in conventional thermoelectrics. Recently, a concept of transverse thermoelectric energy conversion has been emerging as an alternative to overcome the limitations of the longitudinal counterpart.6-8 In a transverse thermoelectric device, a charge current is generated in the direction perpendicular to the applied temperature gradient, which allows that the device performance scales with extrinsic dimensions. Specifically, the output voltage (power) can be enhanced by simply increasing the device length (surface area) perpendicular to the temperature gradient. This means that the thermoelectric performance can be improved by manipulating device designs, even in the absence of improvement to material properties. In addition, transverse devices do not require the use of separate n and p-type materials, since the plane in which the output voltage arises is always isothermal. Thus, transverse devices can have significantly less complex structure and manufacturing than those of longitudinal devices. Realizing such a transverse device requires to employ a suitable transverse thermoelectric effect that guarantees a sufficiently large conversion efficiency. The spin Seebeck effect (SSE) refers to a heat-to-electricity energy conversion process via spin current in magnetic material. In general, the SSE requires a simple ferromagnet (FM)/normal metal (NM) bilayer structure,9–16 wherein the following conversion process occurs: first, the spin current of either magnons (when FM is an insulator) or spin-polarized electrons (when FM is a conductor) are generated in the FM/NM junction via the exchange coupling, i.e., spin pumping, and are subsequently converted into an electric field through the inverse spin Hall effect (ISHE) in the NM layer.17 Experimentally, the electric field, a length-normalized voltage difference, is measured perpendicular to the temperature gradient and magnetic field, which makes SSE suitable for transverse thermoelectric generation. Over the past years, fundamental aspects of SSE have been studied using yttrium iron garnet (YIG)/Pt platform.18-24 From an engineering perspective, however, the thermoelectric efficiency of SSE, largely determined by the dimensionless figure of merit ZSSET, is still 3 to 4 orders of magnitude lower than that of the conventional thermoelectric counterpart.16 In addition to ZSSET, the thermoelectric efficiency of SSE devices also depends on the thickness ratio between FM and NM as discussed in ref. 16. Therefore, novel approaches should be developed to optimize the both parameters in order to design highly efficient SSE devices for practical application. A promising approach to improve ZSSET is to reduce thermal conductivity KFM of the FM layer as well as to increase both spin Seebeck coefficient as the FM/NM system and electrical conductivity on the FM/NM system and electrical conduc layer while  $\alpha$ SSE is affected by magnons in the FM layer. At the same time,  $\alpha$ SSE and  $\sigma$ NM are also largely dependent on the FM/NM interface condition. Therefore, one valid approach towards improving ZSSET would be to develop strategies to separately control phonons and magnons in the FM layer and concurrently to optimize the FM/NM interface condition. Such strategies can be developed by using a bulk polycrystalline FM/NM system as a platform (Fig. 1). First, the separation of phonon and magnon currents could be achieved by inducing selective scattering mechanisms in the bulk FM; the heat-carrying phonons are effectively scattered while the transport of magnons is not hampered, so  $\alpha$ SSE2/kFM can be improved (i.e., lower kFM or raise  $\alpha$ SSE). Examples of such scattering sources are nano-particles (or -precipitates), grain boundaries, atomic defects, etc. In that regard, one attempt has been made to introduce nano-sized grains in polycrystalline YIG. Miura et al.23 observed 41% reduction of kFM in the nanostructured YIG compared to that of single-crystalline YIG, whereas they noticed more significant reduction of aSSE by 79%. The SSE signal in bulk YIG at 300 K is dominantly driven by long-wavelength magnons with spin diffusion lengths up to 10 µm, 18, 24 several orders of magnitude longer than the mean free path of phonons. It is likely that the scattering of those long-wavelength magnons by the nano-sized grains is spin-flipping, significantly reducing the magnon spin diffusion length. The larger than that of phonon mean free path in this system. Fig. 1 Strategies to optimize bulk FM and FM/NM interface properties for improving the figure of merit (ZSSET) of polycrystalline FM/NM systems (FM: ferromagnet, NM: normal metal): introduction of the FM/NM interface morphology. Such selective screening between different energy carriers has been a popular topic in conventional thermoelectric research, wherein the main objective is to separately
control phonons and electrons, realizing a phonon-glass electron-crystal system. Although many successful cases have been reported in SSE-based thermoelectrics yet. In SSE-based thermoelectrics, a 'phonon-glass magnon-crystal' concept has to be developed, which requires a careful design of the scattering mechanisms by considering the spectral nature of phonons and magnons. The previous work by Miura et al.23 suggests that nano-sized grains are not an appropriate candidate at least for YIG, as they strongly suppress the propagation of long-wavelength magnons. Since it is difficult to predict whether the scattering nature of a given scattering centers that simply less scatter the long-wavelength magnons than the heat-carrying phonons. Below we demonstrate that nano-precipitates with the size of tens of nanometers effectively realize the phonon-glass magnon-crystal in polycrystalline NiFe2O4 (NFO). Next, the optimization of FM/NM interface condition can be achieved by deliberately controlling the FM/NM interface morphology before NM layer deposition. A clean and smooth FM/NM interface generally leads to more efficient spin pumping at the FM/NM interface as well as high-quality NM layer deposition, each resulting in a higher aSSE and  $\sigma$ NM (i.e. reduced Joule loss), respectively, and thus an improved aSSE2 $\sigma$ NM. For example, Aqeel et al.29 reported that the magnitude of SSE is strongly enhanced by mechanically polishing the surface of YIG single crystal before Pt deposition. Saiga et al.30 investigated the effect of annealing YIG in air at 1073 K before Pt deposition increases the spin Seebeck voltage by 2.8 times that of a non-annealed sample and attributed the increase to the improved interface crystallinity. We also recently conducted a systematic study on the effect of polishing force and annealing temperature on the SSE signal in polycrystalline NFO/Pt, and revealed that mesoscale defects (cracks, pores, and grain grooves) and roughness of interface strongly affect the SSE signal.31 Despite the independent efforts to realize each of the above-mentioned two optimization strategies in a single SSE platform. In this study, we use polycrystalline NFO/Pt as our platform to demonstrate a facile realization of both strategies. First, a temperature-dependent phase separation in NFO by a simple heat treatment scheme allows an intriguing hierarchical microstructure to be generated within the bulk NFO matrix. We identify that the hierarchical microstructure consists of many nano-sized NFO precipitates embedded in a micro-sized Fe-doped NiO precipitate. Detailed analyses of microstructure and transport properties reveal that the hierarchical microstructure indeed works as selective scatterers for phonons rather than magnons, leading to the favorable αSSE2/κFM. Next, we also find that the simple heat treatment simultaneously leads to a clean and smooth NFO/Pt interface; the high-quality interface guarantees an enlarged active area fraction and a small roughness of the NFO surface. So, those surface characteristics enable efficient spin pumping at the interface as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high-quality Pt deposition, resulting in the increased as well as high US Nano Inc.) were uniaxially pressed into three different pellets (12.7 mm in diameter) at 155 MPa (Hydraulic press, Carver Inc.). Then, two-step sintering processes were applied on the three pellets with different temperature profiles (Fig. S1 in ESI<sup>†</sup>) to control a size and amount of precipitates in the NFO matrix. For comparison, we henceforth label the three samples with their final sintering temperature (1623, 1673, and 1723 K), i.e., NFO1623, NFO1673, and NFO1723. After the sintering processes, each pellet was cut by a diamond wire saw (STX-202A, MTI Korea) into a rectangular slab (2.4 mm × 3.8 mm × ~0.5 mm) for SSE measurements and a 2 mm-thick disk for thermal diffusivity measurements. Surface treatment of NFO slabs and Pt growth The fabricated NFO slabs were subjected to the optimized surface treatment, which was reported in our previous work31 and can be summarized as follows. First, all NFO slabs were deliberately polished using an auto polisher (MetPrep 4, Allied High Tech.) with abrasive SiC papers (P400-P4000) and diamond suspensions (3, 1 and 0.25 µm). During the polishing process, polishing force remained constant at 13 N with rotation speed of 150 rpm for 30 min. Then, the polished slabs were post-annealed at 573 K for 3 h to improve their micronized or amorphized surface. After all surface treatments, an e-beam evaporation system (Korea Vacuum Tech.) was used to deposit a 10 nm Pt layer (99.9%, TASCO) on the NFO surface. Thermal conductivity measurements Thermal diffusivity D, density p, and specific heat Cv of NFO samples according to  $\kappa = D\rho Cv$ . Here, D was measured using a laser flash instrument (LFA467, Netzsch), while ho was estimated from the Archimedes' method (XS105, Mettler Toledo) and Cv was taken from the literature.32 SSE measurements The SSE signal was measured using a customized liquid nitrogen cryostat system (Lake Shore Cryotronics, Inc.). Prior to the SSE measurements, two copper electrode leads (bare wires with 0.001-in. diameter, SPCP/SPCI-001-50, Omega Engineering, Inc.) were attached on edges of the Pt layer with silver epoxy (H20E, EPO-TEK). Then, the NFO/Pt samples was sandwiched between two insulting BeO pads with Apiezon H grease to improve thermal contact for each measurement. The BeO pads were used as heat spreaders because they disperse heat effectively due to their high thermal conductivity (~370 W m-1 K-1).33 Then, two 120 Ω resistive heaters were put on the epoxy and connect in series with each other. The assembled sample block was tied up with a nylon thread on the cryostat platform to ensure mechanically solid contact. Here, we assumed that heat leakage through the applied temperature gradient, and magnetic field were perpendicular to each other. The temperature gradient was estimated from the measured heat flux and thermal conductivity of NFO. During the measurements, the cryostat platform was kept under high vacuum (1473 K) sintering by referring to the Ni-Fe-O2 phase diagram41 (Fig. S2 in ESI†). The oxygen vacancy generation in NFO at elevated temperatures and charge neutrality requirement could be a major driving force to form the rock-salt Fe-doped NiO, wherein the average oxidation number of cations is lower than that in the spinel NFO. Next, we also observed the detailed microstructure inside the Fe-doped NiO precipitate. Interestingly, the HAADF-STEM images of NFO1723 (Fig. 4a and b) show that many square nano-precipitates with different size (up to tens of nanometers) are uniformly distributed inside the Fe-doped NiO precipitates is clearly identified as the spinel structure, which was estimated from the difference in superlattice reflections of the corresponding FFT patterns (marked as red circles in insets of Fig. 4b). The spinel nano-precipitates are also distinguished from the rock-salt NiO precipitates, i.e., the ratio of Ni, Fe and O counts, is similar to those of NFO matrix (Table S1 in ESI<sup>+</sup>). So, we can conclude that the nano-precipitates were generated in the Fedoped NiO precipitate during the sintering process. This result is in accordance with the previous report about precipitation of nano-sized NFO from 4 at% Fe-doped NiO material after cooling (100 °C h-1) it from above solvus temperature of 1473 K.42 Thus, the observation thus far suggests that our NFO samples possess an intriguing hierarchical microstructure of NFO matrix - micro-sized Fe-doped NiO precipitates. Fig. 4 Microstructure analysis inside Fe-doped NiO precipitates. (a) Low-magnification of HAADF-STEM image of a near-edge region of the NiO precipitates. (b) Nanosized NFO precipitate embedded in the micro-NiO/nano-NFO interface. (d) Enlarged region of the micro-NiO/nano-NFO interface. Yellow dashed circles indicate Ni/Fe disorder region. An interface region (Fig. 4c) between the nano-NFO and its surrounding Fe-doped NiO was magnified (Fig. 4d) for close examination of strain between the two
phases; Ni and Fe ions are indicated by green and brown circles. Lattice of the two phases; Ni and Fe ions only exists at the nano-NFO/micro-NiO interface. Accordingly, no significant effect of the strain was observed at the interface from high-resolution strain mapping image (Fig. S9 in ESI<sup>+</sup>). Discussion on the thermal conductivity reduction in the NFO samples by scattering phonons, leading to the reduction of KNFO observed in Fig. 2a. Here, we assume that phonons dominantly contribute to the heat transport because our NFO samples are electrically insulating. Although magnons conduct heat as well, their contribution is usually negligible at 300 K and above,43 the temperature range under consideration in this work. The influence of the precipitates on κNFO can be closely examined by the phonon frequency ω such that 44 (2) Here, C is the phonon heat capacity, vg is the phonon group velocity, and τtot is the total phonon relaxation time.  $\tau$  tot can be obtained following the Matthiessen's rule by taking the reciprocal of the sum of the scattering by nano-precipitates ( $\tau$ np-1), and phonon scattering by boundaries ( $\tau$ b-1):  $\tau$ tot-1( $\omega$ ) +  $\tau$ b-1.(3)We exclude phonon scattering by nano-precipitates ( $\tau$ np-1), and phonon scattering by boundaries ( $\tau$ b-1):  $\tau$ tot-1( $\omega$ ) +  $\tau$ b-1.(3)We exclude phonon scattering by nano-precipitates ( $\tau$ np-1), and phonon scattering by boundaries ( $\tau$ b-1):  $\tau$ tot-1( $\omega$ ) +  $\tau$ b-1.(3)We exclude phonon scattering by boundaries ( $\tau$ b-1). dislocations or strain fields as we did not find any evidence for those from the STEM analysis (Fig. S9 in ESI<sup>+</sup>). First, the Umklapp contribution is described by 45,46 (4) where parameter  $\varphi$  and Debye temperature TD are measures of anharmonicity of lattice, respectively. Slack and Galginaitis 45 showed that  $\varphi$  can be approximately obtained from the formula (5) where h is the reduced Planck constant, y is the Grüneisen parameter, NA is the Avogadro number, and ma is the average molar mass of solid. They found that the  $\varphi$  obtained from eqn (5) agrees well with the one obtained by fitting experimental thermal conductivity data plotted against T/TD. Following their approach, we can estimate  $\varphi$  for our NFO samples. First,  $\gamma$  can be estimated by  $47\gamma = \alpha KT/Cv$  where  $\alpha$  is the thermal expansion coefficient, KT is the isothermal bulk modulus, and Cv is the heat capacity at constant volume of solid. For NFO,  $\alpha = 1.56 \times 10-5 \text{ K}-1.32 \text{ KT} = 180 \text{ GPa}, 48 \text{ and } \text{Cv} = 2.83 \times 106 \text{ J} \text{ m}-3 \text{ K}-1.32 \text{ which give } \gamma = 0.99$ . Also, vg can be approximated using by the Debye theory 44 where E is the Young's modulus and  $\rho$ th is the theoretical density of solid. With E = 99.4 GPa49 and  $\rho$ th = 5.368 g cm-3 for NFO, we find vg = 4303 m s-1. Plugging those  $\gamma$  and vg values with ma = 234.38 g mol-134 into eqn (5) yields  $\varphi$  = 9.03 × 10-17 s. Our experimental fit to the measured  $\kappa$ NFO for the three NFO samples gives the  $\varphi$  values that are close to each other (Table 1), suggesting the lattice anharmonicity is similar between them. The average  $\varphi$  and  $\varphi = 9.03 \times 10-17$  s within 20%; this agreement supports that our experimental fitting procedure provides reliable estimates of φ as well as τu-1 and τnp-1 (see below for details). Table 1 Experimentally measured thermal conductivity κNFO, fitting parameter φ, Umklapp scattering rate τnp-1, and the relative change of τnp-1 to total scattering rate τtot-1 of each NFO sample at 300 K κ NFO (W m-1 K-1)  $\phi$  (s)  $\tau$  u -1(s-1)  $\tau$  np -1 (s-1)  $\tau$  np -1(s-1)  $\tau$  np -1/ $\tau$ tot-1 NFO1623 12.3 1.12 × 10-16 1.02 × 109 3.22 × 108 0.80 NFO1723 10.4 1.16 × 10-16 1.06 × 109 5.51 × 109 2.20 × 108 0.81 The nano-precipitates contribution is described by 50,51 where θ is the scattering cross-section for spherical nano-precipitates and N is the number density of nano-precipitates. Here, θ is given as (7) where R is the radius of precipitates and χ is the size parameter (8) for the phonon wavelength Λ. For simplicity, we treat our nano-NFO precipitates as spheres although they originally have a cube-like shape. Eqn (7) basically describes the relation between  $\theta$  and  $\omega$  at two extreme regimes ( $\chi \ll 1$  and  $\chi \gg 1$ ), where  $\omega$  is linked with  $\Lambda$  in eqn (8) through a dispersion relation for given material. So, when  $\chi \ll 1$  (Rayleigh scattering regime),  $\theta$  becomes independent of ω. For the NFO samples, we can estimate θ by investigating which regime is more influential compared to the other, and thus closely examine τnp-1 as follows. In the long-wavelength limit ( $\chi \ll 1$ ), it has been shown51–53 that mass density and force constant differences between nano-precipitates and host matrix play an important role in determining  $\theta$ . Our nano-NFO precipitates have almost identical structure and composition with those of the NFO matrix. So, the mass and force constant differences can be assumed to be negligible, leading to  $\theta \rightarrow 0$  in this limit. However, in the short-wavelength limit ( $\chi \gg 1$ ), eqn (7) yields  $\theta \rightarrow \pi R^2$  regardless of  $\omega$ . Therefore, the nano-NFO precipitates may barely scatter long-wavelength phonons whereas effectively scatter the short-wavelength phonons with  $\tau np-1 \sim vg\theta N \sim vgR2N$ . Here, we assume vg to be constant by the Debye theory. So, we can expect  $\tau np-1$  to be increased with the sintering temperature because R and N of the nano-NFO precipitates may increase accordingly with the sintering temperature (Fig. S10 in ESI†). These numbers are far larger than the mean free path of phonons at 300 K;56,57 therefore, not much contribution of  $\tau b-1$  is expected. In addition, although the larger grain size of NFO1723 is still expected to lead to higher  $\kappa L$ , the lowest  $\kappa L$  of NFO 1723 among the three samples suggests that the other scattering mechanisms mentioned above play more dominant roles in the phonon scattering. Concerning the micro-sized precipitates, as their size is a few µm in all three samples (Fig. 3b), we may presume that those micro-sized precipitates, as their size is a few µm in all three samples (Fig. 3b), we may presume that those micro-sized precipitates behave such as the grains in matrix. Therefore, likewise a relatively small contribution to phonon scattering is expected. By fitting the experimental  $\kappa$ NFO data in Fig. 2a with the theoretical estimated  $\tau u-1$ ,  $\tau np-1$  and  $\tau np-1$ ,  $\tau np-1$ tot-1 for each sample are summarized in Table 1. It is noted that the contribution of nano-NFO precipitates, τnp-1, turns out to be the most dominant in all three samples, whereas that of grain boundary, τb-1, is one order of magnitude smaller than the other two mechanisms as predicted above. In-between three samples, τnp-1 clearly shows an increasing behavior as the sintering temperature rises, due to the associated growth of R and N of the nano-precipitates. As for  $\tau u - 1$ , no clear trend is observed between the samples, suggesting that the degree of phonon anharmonicity is comparable in all samples. Finally, the relative change of  $\tau n p - 1/\tau tot - 1$  agrees well with that of  $\kappa NFO$  between the three samples. With the phonon scattering analysis thus far, we may summarize that the phonon heat transport in our NFO samples is dominantly affected by the presence of nano-NFO precipitates, and the observed significant reduction of KNFO for the NFO1723 sample can be explained by its larger size (R) and higher number density (N) of the nano-NFO precipitates than the other samples. Surface characteristics of NFO and electrical conductivity of Pt We examined the surface morphological characteristics of each sample before the Pt deposition with the BSE and AFM images (Fig. 5). The deliberately controlled surfaces have the uniformly distributed pores with a size of several micrometers (Fig. 5a-c). Among the three samples, NFO1723 has the most compact surface (Fig. 5c), which can be attributed to its highest sintering process. The pore characteristics on the surface was quantitatively evaluated by measuring the pore area fraction in 100 µm × 100 µm area for each sample, which helped to quantify inactive regime for spin pumping. The active area fraction (%), gradually increases with the sintering temperature (Fig. 5d); this result agrees well with the bulk relative density change of each ranging from 93.3% (NFO1623) to 95.6% (NFO1723). As for the average surface roughness analysis (Fig. 5e-g), we limited our AFM measurements to 2 µm × 2 µm area wherein no pores existed. So, only nanoscale scratches on the surface were included while excluding large-scale defect structures from evaluating the surface roughness. The average surface roughness of all samples falls within the range of 0.55 nm (NFO1623) (b) NFO1623) (c) NFO1623) (Fig. 5h). This range is enough to drive efficient spin pumping at the FM/NM interface, as previously reported.29,31 Fig. 5 Surface morphological characteristics analysis before Pt deposition. (a–c) BSE images of (a) NFO1623, (b) NFO1673, and (c) NFO1723) (d) Active area fraction versus sintering temperature. (e-g) AFM images of (e) NFO1623, (f) NFO1673, and (g) NFO1723. (h) Average surface roughness can largely alter  $\sigma$ Pt of the deposited 10 nm-thick Pt layer because the interface morphology condition influences the quality of Pt growth. Generally, surface with small porosity and roughness leads to high oPt due to suppressed electron scattering temperature results in the reduced porosity and surface roughness; thus, we expect oPt to be increased with the
sintering temperature, which was experimentally confirmed in Fig. 2b. Analysis of the spin Seebeck coefficient Now we turn our attention to the analysis of aSSE. It is known that aSSE is determined by multiple parameters, which can be categorized into three different groups according to each of relevant physical domains. The first domain is the NM layer wherein the ISHE-induced electrical field EISHE under a given  $\nabla Tz$  leads to the experimentally estimated  $\alpha SSE = |ISHE/\nabla Tz| = |(\theta SH\rho NM)\hat{s} \times (s/\nabla Tz)|, (9)$  where  $\theta SH$  and s are the unit vector along electron-spin polarization in the NM layer and the spatial direction of spin current (with the magnitude of spin current density js) induced by SSE, respectively. The second domain is the FM/NM interface where the spin pumping determines js pumped into the NM layer and in turn affects αSSE as given in eqn (9). The last domain corresponds to the FM layer wherein the magnon current is generated by VTz and propagates to the FM/NM interface. It has been shown19,61 that the estimated αSSE in YIG/Pt system is proportional to the amount of magnon accumulation, we attempt to interpret the observed trend of αSSE among the three NFO samples. We do so by correlating the surface and bulk property changes with relevant parameters that affect  $\alpha$ SSE. First, we discuss how the surface morphology-induced change in  $\sigma$ Pt influences  $\alpha$ SSE through the rewritten eqn (9) as follows:62–64 (10)where  $\lambda$ NM and dNM are the spin diffusion length and thickness of NM layer, respectively, j0s is the spin current density at the interface. In eqn (10), it is noted that all the parameters except for j0s/VTz are relevant to the ISHE efficiency of NM, whereas j0s/VTz, as discussed below, is determined by the combined contribution of spin pumping efficiency of NM, whereas j0s/VTz are relevant to the ISHE efficiency at the FM/NM interface and magnon accumulation in the bulk FM. So, by plugging in appropriate values for those parameters in eqn (10), we can estimate the sole contribution of ISHE efficiency to  $\alpha$ SSE, represented by . For our three NFO samples, we calculated using  $\theta$ SH = (1600  $\Omega$ -1 cm-1)/ $\sigma$ Pt,  $\lambda$ NM = (0.61 × 10-15  $\Omega$ m2) ×  $\sigma$ Pt,65,66 and dNM = 10 nm. The estimated decreases as the sintering temperature increases (Fig. 6a) mostly due to the increased  $\sigma$ Pt. This estimated decreases as the sintering temperature increases (Fig. 6a) mostly due to the increased  $\sigma$ Pt. result suggests that the improved surface morphology by the high-temperature sintering leads to the diminished Pt contribution to asse. Although not investigated in the current work, it is worth to note that the optimization of dNM is also important as it can influence both asse. 300 K. (b) Spin pumping efficiency  $\xi$  norm of the NFO/Pt interface at 300 K. (c) (j0s/ $\nabla$ Tz)/ $\xi$  norm representing bulk magnon contribution to  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experimentally determined spin Seebeck coefficient  $\alpha$ SSE at 300 K. (d) Experime FM/NM interface, we introduce a new dimensionless parameter ξnorm that represents qualitative spin pumping efficiency accounted by the interface morphology condition. By referring to our previous work,31 we can infer that ξnorm is linearly proportional to the active area fraction of NFO surface, regardless of the change in surface roughness (see Fig. S11 in ESI† for details). For our current samples, the active area fraction increases with the sintering temperature with little variance in the surface roughness; thus, we can estimate that ξnorm at 300 K accordingly increases from 1.06 (NFO1623) to 1.36 (NFO1723) (Fig. 6b). This result indicates that the latter sample has the highest spin pumping efficiency due to its largest active area fraction. Lastly, how much magnons accumulate at the FM/NM interface in the bulk FM largely determines the pumped magnon current density j0s/VTz and thus αSSE. It has been shown 19,61,67 that such magnon accumulation at the interface induces temperature difference between magnons in FM and electrons in NM, and the difference works as driving force for spin pumping across the interface. Therefore, the normalized (j0s/VTz)/ξnorm is necessary to qualitatively account for the sole contribution of bulk magnons in FM to j0s/VTz by excluding the contribution of ξnorm. For our NFO samples, (j0s/VTz)/ξnorm hardly changes with the sintering temperature (Fig. 6c), suggesting similar degrees of magnon accumulation at the interface of all three samples This result is quite intriguing, considering that the phonon contribution to KNFO is significantly suppressed in the NFO samples that were sintered at higher temperature. It suggests that the nano-NFO precipitates, which work as the dominant scatterers for heat-carrying phonons, barely affect the magnon transport that contributes to SSE. This discrepancy can be understood by comparing the relevant length scales of phonons and magnons, and the relative effects of nano-NFO precipitates mostly scatter the phonons in the short-wavelength limit at 300 K. As for magnon scattering, the nano-NFO precipitates are likely to work in the same way as they do for the phonons; only the short-wavelength limit is relevant, because their magnetic and lattice properties are identical with those of the matrix, making  $\theta \rightarrow 0$  in the long-wavelength limit. This means that the nano-precipitates likewise preferentially scatter short-wavelength magnons; the intensity of scattering for them should vary between the samples with the different nano-NFO contents. In that sense, the observed negligible variation in the magnon contribution to a SSE (Fig. 6c) strongly suggests that the magnons; the intensity of scattering for them should vary between the samples with the different nano-NFO contents. In that sense, the observed negligible variation in the magnon contribution to a SSE (Fig. 6c) strongly suggests that the magnons; the intensity of scattering for them should vary between the samples with the different nano-NFO contents. nano-precipitates and may play a dominant role in inducing SSE. In the case of YIG/Pt system, there is an increasing consensus that SSE at 300 K is mediated primarily by sub-thermal magnons with the energies below ~40 K and corresponding long-wavelengths.18,43,68 Our observation indicates that the SSE in NFO/Pt may also be largely driven by the similarly long-wavelength magnons to those in YIG. On the other hand, we do not exclude the possibility that the short-wavelength magnons make a considerable contribution to SSE in NFO/Pt. Further experimental study such as low temperature and high magnetic field measurements of SSE could provide more detailed information regarding the relevant energy and wavelength of magnons in NFO.18,20 Other than the nano-NFO precipitates, another possible mechanism for the magnon scattering is the grain boundary scattering. It has been shown24 that the relevant propagation length scale of magnons, that determines a SSE, is the spin diffusion length in bulk FM, while the magnon energy relaxation length of thermally excited magnons in NFO has been reported to be 2 to 3 µm at 300 K.69,70 Since the average grain size of our NFO samples ranges between 13.8 µm and 23.0 µm, much larger than the spin diffusion length of NFO, no significant scattering of magnons off the grain boundaries is expected as in the case of phonons. In addition, it has been suggested that the magnon propagation characteristics can be estimated by referring to the critical exponents of temperature dependence of SSE.23,71 The critical exponents for the three NFO samples were calculated by fitting the temperature dependence of (j0s/∇Tz)/ξnorm (see ESI† for details), which reveal that all samples show comparable critical exponent values. of nano-precipitates or grain boundaries. Now we can understand the observed trend in
αSSE in the three NFO samples at 300 K (Fig. 6d) by considering the above discussed three contributions. The Pt contributions and the spin pumping efficiency (ξnorm) show their compensating dependences on the sintering temperature. In contrast, the bulk magnon contribution (j0s/∇Tz)/ξnorm barely changes with the sintering temperature. As a result, αSSE exhibits very small variation between the three samples, proving its robustness against the scattering mechanisms that significantly suppress κNFO. FM/NM interface for ZSSET improvement. Experimentally, the approach is executed at ease by simply adjusting the heat treatment conditions of the polycrystalline NFO/Pt platform. The resulting phase separation in NFO matrix enables the controlled generation of distinctive hierarchical microstructure within the bulk NFO. The nano-precipitates in the hierarchical microstructure selectively scatter heat carrying phonons while barely affecting magnon-crystal system with an improved aSSE2/kFM. In-depth analyses of experimentally measured kFM and asset magnon-crystal system with an improved asset magnon-crystal system with asset magnon-crystal system with asset magnon-crystal system with asset magnon-crystal system with asse may originate from the large discrepancy in the relevant length scales in the two systems. Such a phonon-glass magnon-crystal concept can also be useful for realizing a high-performance spin Peltier device, since SSE and the spin Peltier device, since SSE and the spin Peltier device. simultaneously lead to clean and smooth NFO/Pt interfaces. The well-controlled interface morphology enables efficient spin pumping at the interface as well as high-quality growth of the Pt layer, which results in an improved aSSE20NM. Our study provides an efficient way to increase the thermoelectric efficiency of SSE by combining proper heat treatments and temperature-dependent phase separation in oxide systems. The proposed approach may be applied to various oxide-based SSE structures and contributions M. Y. Kim: methodology, validation, formal analysis, investigation, writing – original draft preparation, writing – review & editing, S. J. Park: methodology, validation, writing – review & editing, G.-Y. Kim: formal analysis, funding acquisition, supervision, writing – review & editing, S.-Y. Choi: formal analysis, funding acquisition, supervision, supervision, writing – review & editing, G.-Y. Kim: formal analysis, funding acquisition, supervision, writing – review & editing, S.-Y. Choi: formal analysis, funding acquisition, supervision, writing – review & editing, S.-Y. Choi: formal analysis, funding acquisition, supervision, supervision, writing – review & editing, S.-Y. Choi: formal analysis, funding acquisition, supervision, writing – review & editing, S.-Y. Choi: formal analysis, funding acquisition, supervision, supervision, writing – review & editing, S.-Y. Choi: formal analysis, funding acquisition, supervision, supervi writing - review & editing. Conflicts of interest There are no conflicts to declare. Acknowledgements M. Y. K., S. J. P. and H. J. acknowledge the support of the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (NRF-2020R1C1C100429112) and the Ministry of Education, Science and Technology (NRF-2020K1A4A7A0209543811). G.-Y. K. and S.-Y. 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