

1 **Magneto-thermal resistance effect in a Co₅₀Fe₅₀/Cu multilayer studied via** 2 **analysis of electron and lattice thermal conductivities**

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15 **ABSTRACT.** This study investigates the giant magneto-thermal resistance (GMTR) effect in a fully-bcc epitaxial
16 Co₅₀Fe₅₀/Cu multilayer through both experimental and theoretical approaches. The applied magnetic field results in a
17 giant change of the cross-plane thermal conductivity ($\Delta\kappa$) of 37 W m⁻¹ K⁻¹, which reaches 1.5 times larger than the
18 previously reported value for a magnetic multilayer and record the highest value at room temperature among the other
19 solid-state thermal switching materials working on different principles. We investigated the electron thermal conductivity
20 for exploring the remarkable $\Delta\kappa$ by the two-current-series-resistor model combined with the Wiedemann-Franz (WF) law.
21 However, the result shows the electron contribution accounts for only 35% of the $\Delta\kappa$, indicating the presence of additional
22 spin-dependent heat carriers. Further investigation of the lattice thermal conductivity, which is expected to be spin-
23 independent, using non-equilibrium molecular dynamics (NEMD) simulations suggests a striking contrast: the additional
24 spin-dependent heat carrier contribution is significantly enhanced in the parallel magnetization configuration but nearly
25 negligible in the antiparallel configuration. These findings provide a fundamental insight into the origin of large GMTR
26 effect and highlight its potential of active thermal management technologies for future electronic devices.

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30 **I. INTRODUCTION**

31 Thermal switching devices are pivotal for advancing thermal management technologies, which are essential
32 for enhancing the efficiencies in modern electronic devices [1,2]. By controlling heat flow through a dynamic change in
33 thermal conductivity of solids, these devices address critical challenges in managing heat dissipation across nanoscale to
34 macroscale regimes. To realize practical thermal switching, it is necessary to achieve a significant change of thermal
35 conductivity, a wide operating temperature range, and seamlessly integration with existing electronic systems. Various
36 mechanisms yielding the thermal conductivity change in solids have been extensively investigated so far, including metal-
37 insulator transition [3,4], electrochemical reaction [5,6], voltage-controlled ferroelectricity [7,8], superconductors [9,10]
38 and the magnetoresistance (MR) effect [11]. Among them, the MR effect in magnetic materials offers distinct advantages;
39 it enables noncontact, high-durability and high-scalable thermal conductivity switching by applying an external magnetic
40 field and/or controlling a magnetization of magnetic materials. However, the magnetic-field-induced change for the
41 thermal conductivity in single magnetic materials is much smaller than other mechanisms at room temperature, hindering
42 the practical application of MR-based thermal switching [11].

43 In magnetic multilayers consisting of alternatively stacked ferromagnetic metal (FM) and non-magnetic metal
44 (NM) layers, their electrical conductivity depends on the relative magnetization angle of the adjacent ferromagnetic layers,
45 a phenomenon known as the giant magnetoresistance (GMR) effect [12,13]. The GMR effect is one of the most familiar
46 phenomena in spintronics and has been widely investigated for various applications, such as read heads for hard disk
47 drives and magnetic sensors. Similar to electrical conductivity, thermal conductivity is influenced by the magnetization
48 configuration in these multilayers, leading to a phenomenon called the giant magneto-thermal resistance (GMTR) effect
49 [14-16]. The GMTR effect enables a large change of the thermal conductivity from the parallel (P) and antiparallel (AP)
50 magnetization configurations. The performances of the GMR and GMTR effects are commonly characterized by the MR
51 ratio and magneto-thermal resistance (MTR) ratio, defined as $(\sigma_{AP} - \sigma_P)/\sigma_{AP}$ and $(\kappa_P - \kappa_{AP})/\kappa_{AP}$, respectively. Here,
52 $\sigma_{AP(P)}$ and $\kappa_{AP(P)}$ represent the electrical conductivity σ and the thermal conductivity κ in the AP state (P state),
53 respectively. It is known that σ and κ in the metallic materials and systems are often well connected through
54 Wiedemann-Franz (WF) law, i.e., the linear relationship between the electron thermal conductivity and the electrical
55 conductivity. Additionally, thermal conductivity has lattice contribution. Therefore, when only these two contributions
56 are assumed, the MTR ratio should be smaller than the MR ratio because the lattice component should be independent of
57 magnetization direction. Previous studies also suggested that in the presence of inter-spin and spin-conserving inelastic
58 scattering in metallic magnetic multilayers where the whole thermal transport is explained by the contribution of electrons,
59 the WF law does not hold and the MTR ratio can be further reduced [17,18]. However, Nakayama et al. recently reported
60 that the MTR ratio (~150%) is significantly higher than the MR ratio (~60%) in a fully-bcc epitaxial $\text{Co}_{50}\text{Fe}_{50}$ (3 nm)/Cu
61 (1.6 nm) multilayer, in which Cu forms a metastable bcc structure at room temperature [19,20]. To explain this behavior,
62 they investigated the applicability of the WF law in the case where the density of state shows a steep change near the
63 Fermi energy [21] and performed the first-principles calculation of the spin-dependent ballistic electron transmittance to
64 analyze the GMTR effect. Although their calculated MTR ratio can be larger than MR ratio at the certain temperature
65 range, this range does not include room temperature and the difference between MTR and MR ratios was too small to

66 fully explain the measured MTR ratio. The experimentally observed large MTR ratio holds promise for future thermal
67 management applications and raises fundamental interest in uncovering additional contribution of thermal transport. In
68 this respect, the direct comparison between MTR and MR ratios in the cross-plane direction is essential to understand the
69 electrical contribution. However, and such comparison have so far been limited to the fully-bcc epitaxial $\text{Co}_{50}\text{Fe}_{50}$ (3
70 nm)/bcc-Cu (1.6 nm) multilayer system. Furthermore, the quantitative analysis of the contribution of the lattice thermal
71 conductivity arising from phonons, one of essential heat carriers, remains unexplored in such a magnetic multilayer.

72 In this study, we observed the GMTR effect in a $\text{Co}_{50}\text{Fe}_{50}$ /metastable bcc-Cu multilayer with a thicker $\text{Co}_{50}\text{Fe}_{50}$
73 layer (5.1 nm) and compared the behavior to that with a thinner $\text{Co}_{50}\text{Fe}_{50}$ layer (3.0 nm) used in the previous study [19].
74 Notably, the change in the cross-plane thermal conductivity of the present sample $\Delta\kappa = \kappa_{\text{P}} - \kappa_{\text{AP}}$ and MTR ratio were
75 $37 \text{ W m}^{-1} \text{ K}^{-1}$ and 108% respectively; the obtained $\Delta\kappa$ exhibits the highest value among GMTR reported so far. We
76 compared the observed MTR ratio with the MR ratio evaluated by the two-current-series-resistor (2CSR) model [22] in
77 the cross-plane direction to gain insight into the electrical contribution. In addition, we analyzed the atomic resolution
78 micro-structure of $\text{Co}_{50}\text{Fe}_{50}$ and bcc-Cu using high-angle annular dark field scanning transmission electron microscopy
79 (HAADF-STEM) and calculated the phonon contribution using a nonequilibrium molecular dynamics (NEMD)
80 simulation [23,24] based on the observed microstructure. Our quantitative analyses revealed that approximately 42% of
81 the κ_{P} value and 65% of the $\Delta\kappa$ value were arising from other spin-dependent contributions that are neither the
82 contributions of pure electron nor phonon transports based on the conventional theories.

83 II. METHODS

84 The magnetic multilayer film with the structure of $\text{Co}_{50}\text{Fe}_{50}/[\text{bcc-Cu}/\text{Co}_{50}\text{Fe}_{50}]_{20}$ was deposited at room
85 temperature onto a [001]-oriented MgO single crystalline substrate using an automated ultra-high-vacuum magnetron
86 sputtering system (hereafter, $\text{Co}_{50}\text{Fe}_{50}$ and bcc-Cu are referred to as CoFe and Cu for simplicity.) The CoFe thickness was
87 designed to be thicker than in the previous study [19], and the Cu thickness was fixed at 2.0 nm to achieve an AP
88 magnetization configuration at zero field via the anti-ferromagnetic interlayer exchange coupling (IEC) between the CoFe
89 layers through the Cu layers. The surface of the multilayer was covered by an Al layer without breaking the chamber
90 vacuum. The Al layer serves as a transducer for the thermal conductivity measurement using the time-domain
91 thermoreflectance (TDTR) method. The actual thickness values of each layer will be shown later. Prior to the deposition
92 of the bottom CoFe layer, the MgO substrate was heated up to 600°C in the sputtering chamber for the surface cleaning.
93 After the deposition of all layers, the sample was annealed at 250°C with applying a constant magnetic field of 300 mT
94 for 1 h [20].

95 The crystal structure of the multilayer film was characterized by the x-ray diffraction (XRD) with the Cu K_{α}
96 radiation and HAADF-STEM with the acceleration voltage of 200 kV. The GMR effect in the current-in-plane
97 configuration and the magnetization were measured using a standard dc four probe method and a vibrating sample
98 magnetometer, respectively, by applying a magnetic field parallel to the in-plane direction. To investigate the GMTR
99 effect in the $\text{CoFe}/[\text{Cu}/\text{CoFe}]_{20}$ multilayer film, we performed the TDTR measurement [25,26], one of the optical pump-

100 probe methods, in the front-heating and front-detection configuration. In this method, ultrafast pump laser pulses heat the
101 surface of the Al transducer layer on the CoFe/[Cu/CoFe]₂₀ multilayer film, while probe laser pulses irradiated with
102 controlled delay time detect the transient response of the surface temperature via thermoreflectance, i.e., the temperature
103 dependence of the reflectivity, which enables the quantitative determination of thermal transport properties of thin films.
104 Further details on the TDTR experiment are found in Refs [27-31]. All the TDTR measurements were performed at
105 ambient temperature under air atmosphere.

106 III. RESULTS

107 An out-of-plane XRD pattern of the sample is shown in Fig. 1(a). The diffraction peak with the fringe around
108 66° corresponds to the 002 peaks of bcc-CoFe, confirming the [001]-oriented growth of bcc-CoFe on the MgO substrate
109 and the formation of the smooth interfaces with a small roughness. Since the peaks from a normal fcc-phase of Cu were
110 not observed, Cu layer was expected to grow in bcc-structure with the [001]-orientated growth in between CoFe layers in
111 our multilayer. Because the lattice constants of bcc-CoFe and bcc-Cu are almost the same, the 002 diffraction peaks
112 originated from bcc-CoFe and Cu cannot be distinguished. Moreover, the 011 peak from the Al layer is overlapped with
113 the 002 peak from the MgO substrate. Figure 1(b) shows a schematic of the fabricated multilayer film with the actual
114 thicknesses confirmed by the microstructure analysis using a transmission electron microscopy (TEM): 5.1 nm for CoFe
115 layers, 2.0 nm for Cu layers, and 44 nm for the Al layer. We confirmed each CoFe layer was thicker than that in the
116 previous study (3.0 nm). The TEM analysis also confirmed that the total thickness t_{total} of the CoFe/[Cu/CoFe]₂₀
117 multilayer was 147 nm. Figures 1(c)-1(f) show the cross-sectional HAADF-STEM images captured along the [100] zone
118 axis of the MgO substrate. Figure 1(c) indicates that the 20-period CoFe/Cu multilayer structure maintained the interfacial
119 flatness consistently from the bottom to the top layers. To investigate the interfacial atomic lattice matching at different
120 regions [upper, middle, and lower regions in Fig. 1(c)] of the CoFe/[Cu/CoFe]₂₀ multilayer, high-magnification HAADF-
121 STEM images and inverse fast Fourier transform images reconstructed from 110 peaks are separately shown in Figs. 1(d)-
122 (f). We directly observed the metastable bcc-Cu that formed in the multilayer from the HAADF-STEM images. Besides,
123 very smooth CoFe/Cu interfaces was observed across all regions. Therefore, we concluded that the nearly complete lattice
124 matching and the atomically flat interfaces were formed in the CoFe/[Cu/CoFe]₂₀ multilayer throughout the entire
125 structure. These microstructure analyses validate that our multilayer can be regarded as the homogenous single medium
126 to evaluate its effective thermal conductivity by the TDTR measurements and analyses.

127 Figures 2(a) and 2(b) respectively show the external magnetic field H dependence of the current-in-plane MR
128 ratio and the normalized magnetization M/M_s , where M_s represents the saturation magnetization. Clear resistance
129 plateaus observed in the range of $|\mu_0 H|$ from 2 mT to 15 mT indicate that the AP state was realized by IEC within this
130 magnetic field range, where μ_0 is the vacuum permeability. At $|\mu_0 H|$ increases beyond 15 mT, the resistance notably
131 drops, and both the MR ratio and magnetization showed the saturation behaviors, indicating the formation of the P state.
132 It is worth noting that the gradual changes in the magnetizations in the field range corresponding to the plateau of the MR
133 curves are likely caused by the formation of magnetic domains which affect the magnetization but not MR ratio, as the

134 AP state is locally formed in each magnetic domain.

135 TDTR signals were recorded as a function of delay time between pump and probe laser pulses. As shown in
136 Fig. 2(c), the TDTR signal was significantly changed from the AP state ($\mu_0 H = 0$ mT) to the P state ($\mu_0 H = \pm 75$ mT),
137 suggesting the large magnetization-configuration-dependent thermal conduction. By analyzing TDTR signals using the
138 one-dimensional heat diffusion model, we experimentally estimated the effective thermal conductivity of the
139 CoFe/[Cu/CoFe]₂₀ multilayer (κ_{exp}) (see Supplementary Material [27]). The fitting curves [solid curves in Fig. 2(c)]
140 showed good agreement with the experimental results. Figure 2(e) summarizes the H dependence of κ_{exp} for the
141 CoFe/[Cu/CoFe]₂₀ multilayer. We found that the CoFe/[Cu/CoFe]₂₀ multilayer exhibited a drastic κ_{exp} change in the
142 range from -25 mT to 25 mT, while κ_{exp} remains almost constant at $|\mu_0 H| > 25$ mT, following the changes in the
143 magnetization configuration (Fig.2(b)). The field range exhibiting the large change in κ_{exp} corresponds to the range
144 where the transition from the minimum to the maximum resistance is observed in the MR curve [Fig. 2(a)]. This indicates
145 that κ_{exp} reaches its minimum and maximum values in the AP and P state, respectively, although the number of data
146 points was limited. Although the observed MTR ratio (108%) was a smaller than the reported one (150%) in the previous
147 study [19], the measured change in the thermal conductivity ($\Delta\kappa_{\text{exp}} = 37 \text{ W m}^{-1} \text{ K}^{-1}$) was not only approximately 1.5 times
148 larger than that reported in the previous study ($\Delta\kappa_{\text{exp}} = 25 \text{ W m}^{-1} \text{ K}^{-1}$) but also the record-high value among solid-state
149 thermal switching materials. In the previous report [19], the used Al thickness of 5nm may be insufficient for absorbing
150 pump-laser energy, requiring the implementation of a bidirectional heat conduction model in TDTR analyses. In contrast,
151 the present Al thickness (44 nm) was thick enough to assume the one-directional heat diffusion model. Despite different
152 models used, both present and previous multilayers [19] exhibited giant MTR ratios, suggesting the robustness of the
153 GMTR effect.

154 From the perspective of practical applications of thermal switching devices, achieving both low and high
155 thermal conductivity states under zero external magnetic field (non-volatile bistability) is crucial. In the present multilayer,
156 a clear hysteresis appears in both MR and magnetization curves [Figs. 2(a) and 2(b)], whereas no such hysteresis was
157 observed in previous study [19]. The observed hysteresis probably originates from the increased crystalline magnetic
158 anisotropy energy of CoFe due to the thicker CoFe layer thickness compared to the previous study. By utilizing this
159 hysteretic behavior, we demonstrated the bistable control of the thermal conduction. As shown in Figs. 2(d) and 2(e), by
160 adjusting $\mu_0 H$ to 10 mT with changing the sweeping direction of H , the TDTR curve and estimated κ_{exp} value
161 reproduced both data at the AP and P states at the same $\mu_0 H$ value. This bistability was also realized in negative field
162 region at -6 mT. Although detailed mechanisms of non-volatile bistability are to be investigated, this result suggests that
163 non-volatile bistability could be also obtained by stabilizing P and AP states at zero magnetic field, potentially through
164 the exchange bias effect instead of IEC, exhibiting the feasibility of a nonvolatile thermal switching device based on the
165 GMTR effect in the future.

166

167 **IV. DISCUSSION**

168 Next, we analyzed the component of the electron thermal conductivity by simulating the RA (electrical
 169 resistance-area product) and MR ratio in the current-perpendicular-to-plane (CPP)-configuration using the generalized
 170 2CSR model [Fig. 3(a)]. The 2CSR model [32-34] is the well-established theoretical approach that predicts the resistance
 171 change in the CPP-GMR structure by considering the individual series resistors for majority- and minority-spin channels.
 172 In the simulations, the CPP-GMR structure was modelled as Cu (1 nm)/[CoFe (5.1 nm)/Cu (2.0 nm)]₂₀/CoFe (5.1 nm)/Cu
 173 (1 nm), where the 1-nm-thick bottom and upper Cu layers considered as the electrodes. The simulation requires several
 174 parameters including the bulk resistivity (ρ), bulk spin-scattering asymmetry coefficient (β), spin diffusion length (λ),
 175 thickness of each layer (t), interfacial resistance area product (r), and interfacial spin-scattering asymmetry coefficient
 176 (γ) [35,36]. Reasonable parameters, such as ρ of CoFe and Cu; β of CoFe; λ of CoFe and Cu, were adopted from
 177 previous studies [35,36], which were summarized in Table I. We used the ρ and λ reported for fcc-Cu in place of those
 178 for bcc-Cu as there is no reported values for bcc-Cu. It should be noted that these parameters in bulk region of Cu have
 179 negligibly weak effect on the simulated results because the thickness of Cu (2.0 nm) is much shorter than λ . However, it
 180 is necessary to obtain values of r ($r_{\text{CoFe/Cu}}$) and γ at the metastable CoFe/Cu interface ($\gamma_{\text{CoFe/Cu}}$) since the previous
 181 study has reported that the metastable bcc-Cu has a significantly different interfacial electronic band matching with CoFe
 182 compared to fcc-Cu [20]. In this study, we quantitatively analyzed $r_{\text{CoFe/Cu}}$ and $\gamma_{\text{CoFe/Cu}}$ based on the experimental data
 183 of the [CoFe(3 nm)/bcc-Cu]_N multilayer CPP-GMR devices [19]. By analyzing the measured RA_{AP} for $N = 3, 5, \text{ and } 7$
 184 [19] using the series circuit model consisting of the bulk and interfacial resistance area products, we determined $r_{\text{CoFe/Cu}} =$
 185 $0.27 \text{ m}\Omega \mu\text{m}^2$. We also attempted to obtain the $\gamma_{\text{CoFe/Cu}}$ by comparing the experimentally measured resistance change-
 186 area products (ΔRA) for $N = 3, 5, \text{ and } 7$ [19] with the simulated ΔRA as a function of $\gamma_{\text{CoFe/Cu}}$ [Fig. 3(b)]. As a result,
 187 we found $\gamma_{\text{CoFe/Cu}}$ to be 0.76 ± 0.03 . By adopting these $r_{\text{CoFe/Cu}}$ and $\gamma_{\text{CoFe/Cu}}$ in the analysis based on the 2CSR model,
 188 we finally estimated $RA_{\text{AP}} = 47.2 \text{ m}\Omega \mu\text{m}^2$, $RA_{\text{P}} = 29.4 \text{ m}\Omega \mu\text{m}^2$, and obtained CPP-MR ratio of 60% for the present
 189 CoFe/[Cu/CoFe]₂₀ multilayer with the 5.1-nm-thick CoFe layers. The obtained MR ratio was much smaller than the
 190 experimentally observed MTR ratio, approximately half of the MTR ratio of 108%, whose tendency is consistent with
 191 the previous report [19]. Based on the WF law, the electron thermal conductivity of conventional metals is described as
 192 σLT , where σ ($= t_{\text{total}}/RA$) is the electrical conductivity, L is the Lorenz number, and T is the absolute temperature.
 193 Assuming that the WF law and L value of free electron ($L_0 = 2.44 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$) can be applied for the present
 194 multilayer structure, the electron thermal conductivity values for the AP and P states (κ_e^{WF}) were estimated to be 22.6 and
 195 $36.2 \text{ W m}^{-1} \text{ K}^{-1}$ at $T = 297 \text{ K}$, respectively, using the simulated RA_{AP} and RA_{P} (see Table II). The calculated their
 196 difference $\Delta\kappa_e^{\text{WF}}$ of $13.6 \pm 0.5 \text{ W m}^{-1} \text{ K}^{-1}$ is only about 35% of the experimentally observed $\Delta\kappa_{\text{exp}}$ ($37 \pm 10 \text{ W m}^{-1} \text{ K}^{-1}$).
 197 Thus, $\Delta\kappa_{\text{exp}}$ should include the additional spin-dependent contribution $\Delta\kappa_{\text{add}} (= \Delta\kappa_{\text{exp}} - \Delta\kappa_e^{\text{WF}})$, which is calculated to
 198 be very large $24 \pm 10 \text{ W m}^{-1} \text{ K}^{-1}$. It should be also mentioned that the L value in ferromagnetic materials can be smaller
 199 than L_0 due to several factors (e.g., spin-dependent scattering mediated by exchange interaction and high localized
 200 density of states near Fermi energy). For instance, the experimentally reported L values in Ni and Co at room temperature
 201 are slightly smaller than L_0 [11,37]. It was also reported that L value for magnetic Co/Cu multilayer is smaller than L_0
 202 due to the predominant electron-phonon scattering [15]. By considering the reduction of L from L_0 in present CoFe/Cu
 203 magnetic multilayer suggested from these earlier studies, intrinsic electron thermal conductivity may be smaller than κ_e^{WF} ,

204 respectively. Therefore, it is expected that the deviation of L more likely result in an underestimation of the $\Delta\kappa_{\text{add}}$
 205 component, rather than overestimation. Another potential factor in correcting electron thermal conductivity is the Seebeck
 206 effect, expressed as $\sigma S^2 T$, where S is the Seebeck coefficient. However, assuming the S values of our multilayer in the
 207 AP and P states are the same as those measured in the previous study [38], the correction on the electron thermal
 208 conductivity is estimated to be less than $1 \text{ W m}^{-1} \text{ K}^{-1}$ in both the AP and P states, which cannot explain the observed large
 209 $\Delta\kappa_{\text{add}}$.

210 Here, we explore the lattice component of thermal conductivity in CoFe/Cu multilayers. In actual CoFe layers,
 211 Co and Fe atoms are expected to be randomly alloyed, as annealing at 250°C is insufficient to form the ordered $B2$
 212 structure of CoFe [39]. Even though the layer thicknesses are on the nanometer scale, phonons are expected to experience
 213 significant alloy scattering. Furthermore, considering the Debye temperatures of the constituent elements [40], the
 214 quantum effect should have a minimal effect. Therefore, we employed NEMD simulations to evaluate the lattice thermal
 215 conductivity. As illustrated in Fig. 4(a), the CoFe/Cu multilayer was modeled by stacking a bcc-based CoFe/Cu slab along
 216 the z direction. The coordinates of Co and Fe atoms in each layer were randomly distributed while maintaining an atomic
 217 fraction of 1:1. Furthermore, their coordinates in different slabs were set to be uncorrelated [Fig. 4(b)]. The lattice
 218 constants of both CoFe and Cu slabs were set to 2.86 \AA , as determined from the lattice spacing analysis of TEM images.
 219 The interatomic interactions among the Co, Fe, and Cu atoms were described by the embedded atom method potential
 220 with the parameterization of Zhou et al. [41]. In the NEMD simulation, a system with a length (l) and a cross-sectional
 221 area (A) was sandwiched by hot and cold thermostats with the length of $l/2$. The periodic boundary condition was applied
 222 to the cross-sectional direction. The temperatures of the thermostats were controlled by the Langevin thermostat. Denoting
 223 inflow and outflow of the energies in the thermostats at a time interval Δt as $\Delta Q_H(\Delta t)$ and $\Delta Q_C(\Delta t)$, the heat flux in
 224 the system was given by the average; $q(t = k\Delta t) = \sum_j^k [\Delta Q_H(j\Delta t) - \Delta Q_C(j\Delta t)] / 2Ak\Delta t$, where k is the total time steps
 225 of the NEMD simulation. We first performed a 5 ns-long NEMD simulation to ensure the system reached a non-
 226 equilibrium steady state more smoothly. Subsequently, we conducted an additional 5 ns-long NEMD simulation to sample
 227 $q(t)$ and evaluated its mean value \bar{q} was evaluated after a nonequilibrium steady state was established. An effective
 228 lattice thermal conductivity of the CoFe/Cu multilayer system was calculated by $\kappa_L^{\text{NEMD}} = \bar{q}l/\Delta T$, where ΔT denotes a
 229 temperature difference between the hot and cold thermostats. κ_L^{NEMD} was finally evaluated through the ensemble average
 230 for five different atomic configurations. For the comparison with measurements, l was set to approximately 143 nm. In
 231 addition, A was set to $2.29 \times 2.29 \text{ nm}^2$, which ensures the convergence of κ_L^{NEMD} . To fix the translational and rotational
 232 motions of the entire system, the CoFe/Cu slabs with frozen internal degrees of freedom were attached to both edges of
 233 the system. All NEMD simulations were performed by the LAMMPS package [42] with a time step of 0.5 fs. As shown
 234 in the Fig. 4(c), a steady state was achieved at 2.5 ns to 5.0 ns, κ_L^{NEMD} was evaluated from the heat flux in the time
 235 range. Although small fluctuations appeared in the obtained temperature profile for $\Delta T = 50 \text{ K}$ [Fig. 4(d)], a nearly
 236 linear temperature profile was established, confirming the homogeneity of the CoFe/Cu multilayer. The resulting
 237 κ_L^{NEMD} of the CoFe/Cu multilayer was $5.7 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature. The κ_L^{NEMD} value remained independent
 238 of the magnitude of ΔT (see Supplementary Material [27]). Note that the embedded atom method-type potential

239 used in this calculation is not fully optimized for modeling heat conduction in the present multilayer system.
 240 Furthermore, this potential may not accurately capture phonon transport mechanisms in certain pure metals [43].
 241 However, this obtained value exhibits magnitude similar to previous superlattice calculations [44,45]. Since it is
 242 reasonable to assume the lattice thermal conductivity is independent of the magnetization configurations, we put the same
 243 κ_L^{NEMD} for AP and P states and $\Delta\kappa_L^{\text{NEMD}} = 0$ in Table II.

244 Finally, we quantitatively compared the value of κ_{exp} , κ_e^{WF} , κ_L^{NEMD} , and explored the additional contribution
 245 of the thermal conductivity ($\kappa_{\text{add}} = \kappa_{\text{exp}} - \kappa_e^{\text{WF}} - \kappa_L^{\text{NEMD}}$) as summarized in Table II. The quantitative analysis of κ_L^{NEMD}
 246 has led to an important finding: κ_{exp} in the AP state is nearly equal to the summation of κ_e^{WF} and κ_L^{NEMD} , i.e., $\kappa_{\text{add}} =$
 247 $5 \pm 4 \text{ W m}^{-1} \text{ K}^{-1}$, whereas in the P state, it is significantly larger than their summation, i.e., $\kappa_{\text{add}} = 30 \pm 9 \text{ W m}^{-1} \text{ K}^{-1}$. This
 248 result suggests that the contribution of additional heat carriers in AP state could be negligibly small, although our analysis
 249 still contains a certain level of the error bar. Notably, the present quantitative analysis shows that almost 42% of κ_{exp} in
 250 the P state and 65% of $\Delta\kappa_{\text{exp}}$ originate from κ_{add} that is neither the contributions of electron nor phonon calculated
 251 based on the WF law and the NEMD simulation, respectively. A possible additional carriers contribution to the GMTR
 252 effect is magnon, collective dynamics of localized magnetic moment in magnetic materials. The magnetization-
 253 configuration-dependent magnon transport, known as the magnon valve effect, has been demonstrated in magnetic
 254 multilayer structures, which highlights the importance of magnon–magnon interactions between adjacent magnetic layers
 255 [46,47]. Although the magnon contribution in the thermal conduction is usually discussed at the low temperature [48-50],
 256 recent theoretical calculation has shown that the magnon thermal conductivity also depends on the magnetization
 257 configuration even at room temperature [51]; in the trilayer consisting of two magnetic insulator layers sandwiching a Cu
 258 interlayer, the large magnon-driven MTR ratios of up to 40% have been predicted. However, even if magnons contribute
 259 to κ_{exp} in the P state, their thermal conductivity in typical ferromagnetic metals is around $\sim 10 \text{ W m}^{-1} \text{ K}^{-1}$ at room
 260 temperature [31], which is much smaller than the estimated κ_{add} . Therefore, the conventional sole magnon contribution
 261 is unlikely to be a major factor in our observations. Thus, we should also consider the possibility of magnon-electron and
 262 magnon-phonon interactions [52-55]. An example of a phenomenon to which such interactions contribute is the magnon-
 263 drag effect in the thermoelectric conversion [52,53]. However, the reported modulation of thermopower by the magnon
 264 origin was very small in ferromagnetic metals, such as CoFe- and NiFe-based system, at room temperature [38,53]. If the
 265 magnon-drag-induced change in the thermal conductivity in our CoFe/Cu multilayer is comparable to that in the
 266 thermopower, its contribution is negligible. Regarding phonons, there is a possibility that they are influenced by the
 267 external magnetic field through spin-lattice interaction [55,56]. However, changes in phonon thermal conductivity have
 268 been observed only at the low temperature and under the strong magnetic field, which are completely different from the
 269 conditions in this study. Moreover, these changes were on the order of $0.1 \text{ W m}^{-1} \text{ K}^{-1}$, making their impact relatively small.
 270 We cannot entirely rule out the possibility of that some of these magnon-related interactions may still be significant, as
 271 we have not explicitly evaluated their effects in our sample. To clarify the microscopic mechanism of giant MTR ratio
 272 and κ_{exp} , further experiments and calculations from a series of multifaceted perspectives are necessary.

273 V. CONCLUSIONS

274 In summary, we investigated the GMTR effect in the CoFe/Cu magnetic multilayer with 5.1 nm-thick CoFe
275 layers. We achieved the giant thermal conductivity change of $37 \text{ W m}^{-1} \text{ K}^{-1}$, not only surpassing the previous study [19]
276 but also marking the highest value in the solid-state thermal switching materials. The MTR ratio of 108% is found to be
277 greater than the MR ratio for the CPP configuration, reproducing the tendency of previous study for 3 nm-thick CoFe. By
278 comparing the value of κ_{exp} , κ_e^{WF} , and κ_L^{NEMD} , we identified a significant contribution from additional components of
279 the thermal conductivity to the GMTR effect although this contribution has not been uncovered yet in this study.
280 Interestingly, the quantitative analysis of the lattice thermal conductivity suggests that the contribution of this additional
281 heat carriers is remarkable in P state but negligibly small in AP state. This could provide an important clue to elucidate
282 the origin of the unconventional thermal transport in the magnetic multilayers. Furthermore, this study demonstrated the
283 bistability of low and high thermal conductivity states at the same magnetic field due to the hysteretic behavior of the
284 magnetization. These findings highlight the potential of active thermal management technologies using the GMTR effect
285 for electronic devices.

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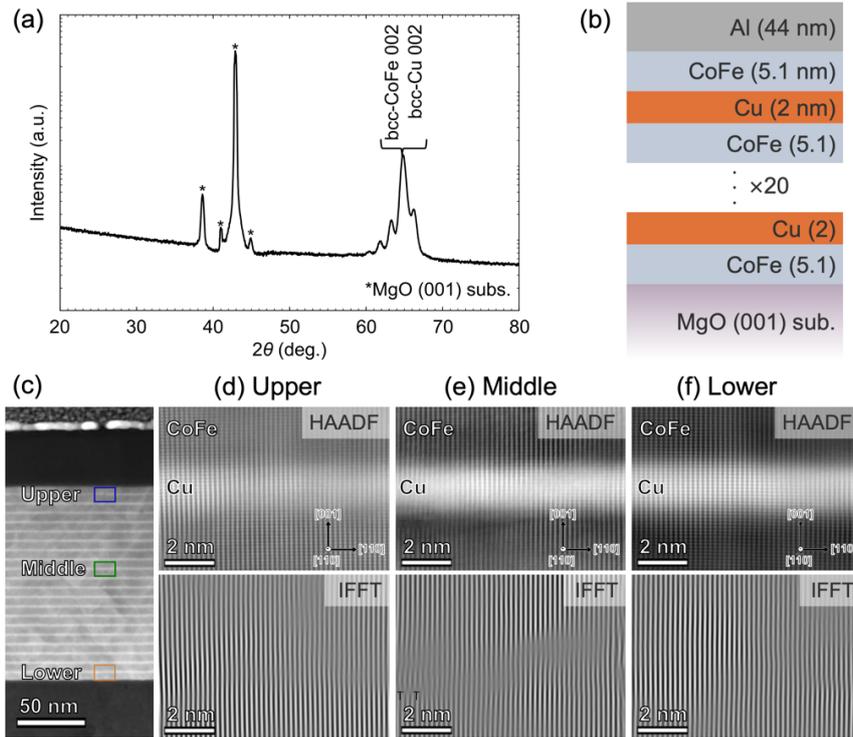
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297 **DATA AVAILABILITY**

298 The raw data are available in the Zenodo repository [57]

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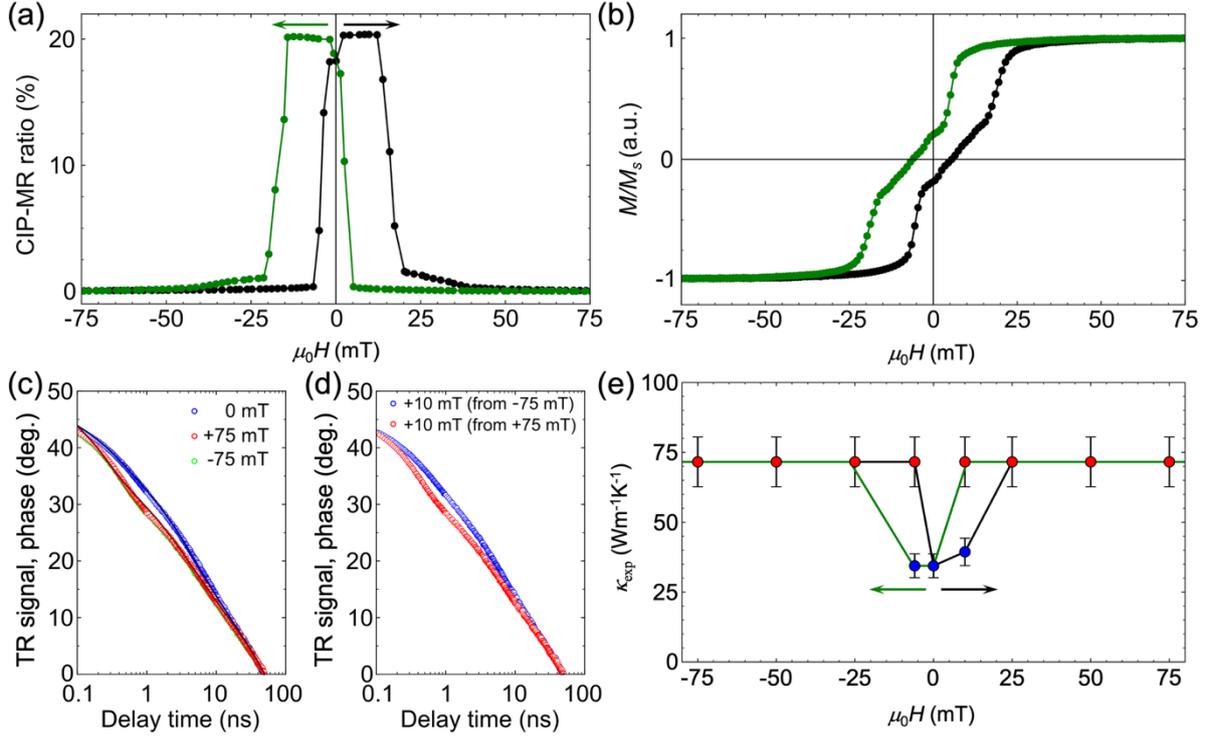
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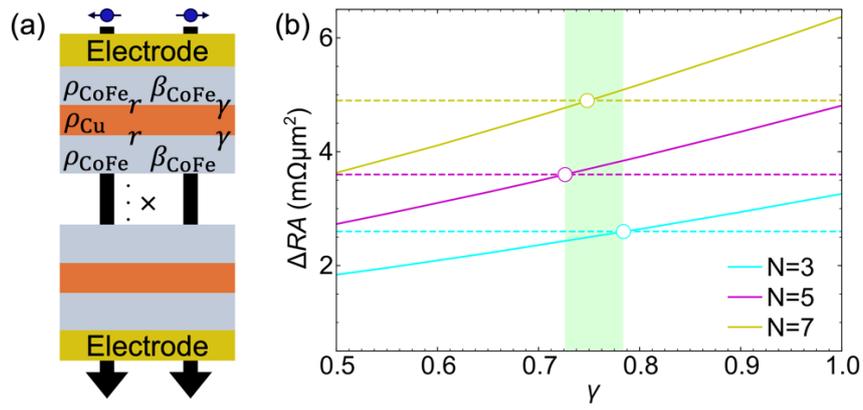
FIG.1. (a) Out-of-plane x-ray diffraction curve of the multilayer film. (b) Schematic structure of the CoFe/[Cu/CoFe]₂₀ multilayer film deposited on the MgO (001) single crystalline substrate. (c) Low-magnification high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image of the multilayer film. (d)-(f) High-magnification HAADF-STEM images and inverse fast Fourier transform (IFFT) images reconstructed from 110 peaks of (d) Upper, (e) Middle, and (f) Lower parts indicated by the rectangular regions in (c), respectively.



308

309 FIG.2. (a) Magnetic field H dependence of current-in-plane magnetoresistance (CIP-MR) and (b) normalized
 310 magnetization for the CoFe/[Cu/CoFe]₂₀ multilayer film with antiferromagnetic coupling via Cu spacer. (c) Temporal
 311 response of thermoreflectance (TR) signals, i.e., TDTR signals, for the CoFe/[Cu/CoFe]₂₀ multilayer film with applying
 312 the magnetic field $\mu_0 H$ of 0 and ± 75 mT. The black solid curves represent the best fits for each measurement. (d) TDTR
 313 signals for the CoFe/[Cu/CoFe]₂₀ multilayer film at $\mu_0 H$ of 10 mT applied with adjusting the field sweep direction. (e)
 314 H dependence of κ_{exp} of the CoFe/[Cu/CoFe]₂₀ multilayer film. Black and green arrows in (a) and (e) indicate the sweep
 315 direction of the magnetic field

316



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 318 FIG.3. (a) Schematic of two-current series-resistor (2CSR) model. (b) Simulated spin interfacial asymmetry γ
 319 dependence of resistance change-area product ΔRA (solid curves). Dashed lines show experimentally measured ΔRA of
 320 the $\text{CoFe}/[\text{Cu}/\text{CoFe}]_N$ multilayer with repeating number N of 3, 5, and 7 [15]. Open circles show the γ values obtained
 321 from the agreement between experimental and simulated ΔRA .

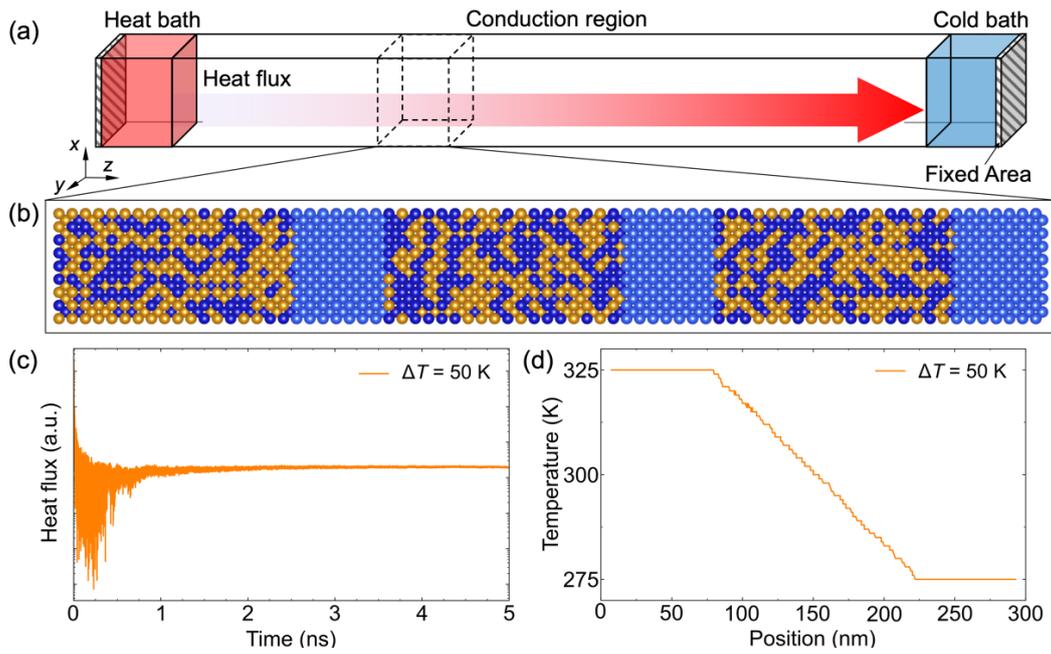
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323 TABLE I. Used parameter for the generalized two-current series-resistor (2CSR) model

	ρ [$\mu\Omega$ cm]	β	λ [nm]	t [nm]	r [$m\Omega \mu m^2$]	γ
CoFe	19.1 [22]	0.62 [23]	15 [22]	5.1	0.27	0.76 ± 0.03
Cu	7.0 [22]	0	100 [22]	2.0	0.27	0.76 ± 0.03

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327 FIG.4. (a) Schematic of the model for the nonequilibrium molecular dynamics (NEMD) simulations. (b) Stacking
 328 structure for NEMD. The atomic position is determined so as to be different from the adjacent CoFe layers. (c) Time
 329 dependence of the heat flux for temperature difference, ΔT of 50 K. (d) The obtained temperature profile for ΔT of 50
 330 K and the liner temperature approximation used to calculate the temperature gradient.

331

332 TABLE II. Summary of the experimentally observed thermal conductivity κ_{exp} , the theoretically analyzed electron
 333 thermal conductivity κ_e^{WF} and effective lattice thermal conductivity κ_L^{NEMD} and the evaluated additional component of
 334 thermal conductivity κ_{add} in AP and P states.

	κ_{exp} [$\text{W m}^{-1} \text{K}^{-1}$]	κ_e^{WF} [$\text{W m}^{-1} \text{K}^{-1}$]	κ_L^{NEMD} [$\text{W m}^{-1} \text{K}^{-1}$]	κ_{add} [$\text{W m}^{-1} \text{K}^{-1}$]
AP state	34 ± 4	22.6	5.7 ± 2.0	5 ± 4
P state	72 ± 9	36.2 ± 0.5	5.7 ± 2.0	30 ± 9
$\Delta\kappa$	37 ± 10	13.6 ± 0.5	0	24 ± 10

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