Band match enhanced current-in-plane giant magnetoresistance in epitaxial Co$_{50}$Fe$_{50}$/Cu multilayers with metastable bcc-Cu spacer
Band match enhanced current-in-plane giant magnetoresistance in epitaxial Co$_{50}$Fe$_{50}$/Cu multilayers with metastable bcc-Cu spacer

Cite as: APL Mater. 7, 111106 (2019); doi: 10.1063/1.5119370
Submitted: 10 July 2019 • Accepted: 15 October 2019 • Published Online: 15 November 2019

Kresna B. Fathoni, 1,2 Yuya Sakuraba, 1,a) Taisuke Sasaki, 1 Yoshio Miura, 1 Jin W. Jung, 1 Tomoya Nakatani, 1 and Kazuhiro Hono 1,2

AFFILIATIONS
1 Research Center for Magnetic and Spintronic Materials, National Institute for Materials Science (NIMS), Tsukuba 305-0047, Japan
2 Graduate School of Pure and Applied Sciences, University of Tsukuba, Tsukuba 305-8571, Japan

a) Author to whom correspondence should be addressed: SAKURABA.yuya@nims.go.jp

ABSTRACT

Although current in-plane giant magnetoresistance (CIP-GMR) is widely used as various magnetic field sensors, a higher magnetoresistance (MR) ratio is still required to improve their sensitivity and detectivity for certain applications. Here, we report dramatic enhancement of the MR ratio up to 26.5% in a spin valve device and 40.5% in an antiferromagnetically coupled trilayer device using fully epitaxial Co$_{50}$Fe$_{50}$/Cu/Co$_{50}$Fe$_{50}$ structures with metastable bcc-Cu spacer layers. Transmission electron microscopy analysis indicated that the metastable bcc-Cu had a perfect lattice match at the bcc-Co$_{50}$Fe$_{50}$/bcc-Cu interfaces. First-principles calculations showed good electronic band matching that induces a large spin asymmetry of the electron transmittance in the in-plane direction. The combination of this substantial lattice match and electronic band match is attributed to the large MR ratio, suggesting that exploring the use of metastable structure in ferromagnetic/nonferromagnetic multilayers will lead to further enhancement of CIP-GMR.

© 2019 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5119370

I. INTRODUCTION

Since the discovery of current in-plane giant magnetoresistance (CIP-GMR) in 1988, an enormous number of the studies have been performed on this phenomena and various applications have been developed.1-4 The GMR effect was first found in ferromagnetic (FM)/nonmagnetic (NM) multilayers, in which the ferromagnetic layers magnetically interact with each other by interlayer exchange coupling through the nonmagnetic layer. The discovery of a large exchange bias effect from antiferromagnetic (AFM) materials to pin the magnetization of the ferromagnetic (FM) layers led to the development of spin valve (SV) type CIP-GMR,5 which allows the FM layer to switch at a small external magnetic field to be suitable for magnetic field sensor applications, including earth magnetic field sensors, speed-rotation-position sensors in automobiles,5 and detectors of magnetic beads used in biomedical applications.6 Here, magnetic encephalography and cardiography, which require much lower noise, would be promising applications because CIP-GMR has intrinsically small 1/f noise and the device resistance is tunable.7,8 For all these magnetic sensing applications, a large magnetoresistance (MR) ratio is always beneficial for improving sensitivity and detectivity.

Based on various experimental studies and theoretical treatments using ab initio calculations for electronic structures and the classical Boltzmann equation for spin-dependent transport,9-13 it has been recognized that the most important factor for enhancing the MR ratio is the lattice and electronic band matchings between the ferromagnetic (FM) layer and nonmagnetic layer. For example, in the case of Fe/Cr multilayers, Fe and Cr both have a body centered cubic (bcc) structure with a very small lattice mismatch of 0.63%,14 which minimizes the additional spin-independent scattering at the interface caused by structural strain and dislocations. In addition, the band dispersions of the minority-spin
electrons in Fe are Cr similar, while those of the majority-spin electrons are not, which give rise to spin asymmetry of electron scattering at the Fe/Cr interface. In particular, MR ratios of 220% at 1.5 K and 42% at room temperature have been reported in Fe/Cr multilayers. Similarly, the good lattice matching and band matching of majority-spin electrons explains the large MR ratio in CIP-GMR devices with Co/Cu and Co$_{50}$Fe$_{50}$/Cu multilayers. Co$_{50}$Fe$_{50}$/Cu multilayers have been used in read heads for HDDs because of the soft magnetic properties of this Co$_{50}$Fe$_{50}$. However, after the CIP-GMR read head was replaced by the tunneling MR (TMR) read head, the research activity for CIP-GMR has been diminished. Since then, little progress has been made in increasing the MR ratio of CIP-GMR because no other combination of conventional FM and NM materials showing superior lattice and band matchings has been found.

As a motivation of the present work, we noticed an interesting study reporting a highly lattice matched all-bcc-Fe/Cu/Fe layered film with a metastable bcc Cu spacer mimicking the structure of bcc Fe. Although such a metastable bcc Cu is expected to grow on any Co$_{50}$Fe$_{50}$ having a bcc structure ($x \geq 0.25$), the CIP-GMR properties of all-bcc Co$_{1−x}$Fe$_x$/Cu/Co$_{1−x}$Fe$_x$ have not been investigated systematically. Thus, we fabricated fully epitaxial CIP-GMR spin valve devices with Co$_{1−x}$Fe$_x$ layers of varying $x$ to investigate the relationship between metastable bcc growth of Cu on Co$_{1−x}$Fe$_x$ and the MR properties. Interestingly, we observed the largest MR ratio ever reported for CIP-GMR at room temperature in the devices and the Cu spacer. The speciments for the (S)TEM observations were prepared with the lift-out technique using a dual-beam focused ion-beam/scanning electron microscope (FIB/SEM; FEI Helios G4).

### II. METHODS

#### A. Experimental details

CIP-GMR spin-valve films consisting of Co$_{1−x}$Fe$_x$ (6 nm)/Cu ($t$ nm)/Co$_{1−x}$Fe$_x$ (6 nm)/IrMn (8 nm) were deposited on top of the MgO (001) substrate. The deposition was done using magnetron sputtering in an ultrahigh vacuum system with a base pressure lower than 5 $\times$ 10$^{-7}$ Pa. The surface of the MgO substrate was etched by Ar ion milling in the sputtering chamber before depositing the Co$_{1−x}$Fe$_x$ film to obtain (001)-oriented epitaxial growth. Different Co$_{1−x}$Fe$_x$ alloy targets ($x = 0.10, 0.25, 0.5, 0.67,$ and $1$) were used to study the CoFe composition dependence. All films had the same CoFe compositions in the top and bottom layers. The Cu thickness $t$ was varied from 0 to 5 nm by making a wedge shape structure. To make samples showing a larger MR ratio, a trilayer structure with Co$_{50}$Fe$_{50}$ (3 nm)/Cu (1.6 nm)/Co$_{50}$Fe$_{50}$ (3 nm)/MgO (2 nm) and a multilayer with the structure of [Co$_{50}$Fe$_{50}$ (3 nm)/Cu (1.6 nm)]$_{5}$Co$_{50}$Fe$_{50}$ (3 nm)/MgO (2 nm) were fabricated under the same conditions as the previous samples. The Co$_{50}$Fe$_{50}$ thicknesses were reduced from 6 nm to 3 nm to minimize current shunting through Co$_{50}$Fe$_{50}$ layers which will increase the MR ratio. All samples were annealed in a 239 kA/m constant magnetic field at 250$^\circ$C, holding this temperature for 1 h. Photolithography and physical etching were used to pattern the film into wires. The magnetoresistance properties of each wire were measured using the standard dc four probe method with the applied external magnetic field parallel to the current direction.

The microstructure of the spin-valves with Fe, Co$_{50}$Fe$_{50}$, and Co$_{50}$Fe$_{50}$ compositions was compared by transmission electron microscopy. An aberration-corrected (scanning) transmission electron microscope ([S]TEM; FEI Titan G2 80-200) was used to analyze the microstructures of the samples, especially the Cu spacer and interfaces between the Co$_{1−x}$Fe$_x$ and the Cu spacer. The specimens for the (S)TEM observations were prepared with the lift-out technique using a dual-beam focused ion-beam/scanning electron microscope (FIB/SEM; FEI Helios G4).

#### B. First-principles calculation details

The transport calculations were performed with the QUANTUM espresso code using the generalized gradient approximations for the exchange-correlation energy. The number of k points was $15 \times 15 \times 1$, and a broadening parameter of 0.01 (Ryd) was used. A B2-CoFe(or Fe)/Cu/B2-CoFe(or Fe)(001) trilayer was constructed in a tetragonal supercell, where the in-plane lattice parameter of the supercell was fixed at 2.86 Å. We prepared a supercell, consisting of a multilayer containing 9 atomic layers of Cu spacer and 15 atomic layers of CoFe (or Fe) for the Co- and Fe-terminated interfaces, in which the atomic positions were fully optimized. Furthermore, we prepared the fcc-Co$_{50}$Fe$_{50}$/fcc-Cu/fcc-Co$_{50}$Fe$_{50}$(011) trilayer with 9 atomic layers of fcc-Cu and 15 atomic layers of fcc-Co$_{50}$Fe$_{50}$, where the in-plane lattice parameters are fixed to that of fcc-Cu lattice constant, 3.61 Å and $3.61 \times \sqrt{2}$ Å. For the transport calculations, we considered an open quantum system consisting of a scattering region corresponding to Cu spacers and a junction with CoFe (or Fe) attached to left and right semi-infinite electrodes corresponding to bulk. The transmittance was obtained by solving the scattering equation with infinite boundary conditions in which the wave function of the scattering region and its derivative were connected to the Bloch states of each electrode. The potential in the scattering equation was obtained from self-consistent electronic structure calculations for the supercell containing a left and a scattering region. Since our system was repeated periodically in the $xy$ plane, propagating states were assigned an in-plane wave vector $k_{||} = (k_{x}, k_{y})$ index.

### III. RESULTS AND DISCUSSION

A diagram illustrating the stacking of the fabricated films and x-ray diffraction (XRD) patterns of the respective samples are shown in Figs. 1(a) and 1(b). As expected, the films with Co$_{75}$Fe$_{25}$, Co$_{50}$Fe$_{50}$, Co$_{25}$Fe$_{75}$, and Fe having a bcc structure all had a peak around 66°, i.e., the (002) peak from bcc Co$_{1−x}$Fe$_x$, indicating (011)-oriented growth of CoFe and Fe on the MgO substrate. The film with Co$_{50}$Fe$_{50}$ did not have this particular peak; it is known to have a stable fcc phase.

Figures 2(a) and 2(b) show the Cu spacer thickness dependence of the MR ratio and CoFe composition ratio dependence of the MR ratio at each MR maximum and at $t_{Cu} = 2.5$ nm. The MR ratio was much larger in the devices with bcc Co$_{50}$Fe$_{50}$, Co$_{25}$Fe$_{75}$, and Co$_{75}$Fe$_{25}$ than in fcc Co$_{50}$Fe$_{50}$. Co$_{50}$Fe$_{50}$ had the largest MR ratio, 26.5%, at room temperature, which is about 2.6 times larger than the 10.3% of Co$_{50}$Fe$_{50}$, a material often used in magnetic sensors. There was a dramatic enhancement in the MR ratio from 5.4% for...
pure Fe to 23.5% for Co$_{33}$Fe$_{67}$, even though the Fe and Co$_{33}$Fe$_{67}$ had the same (001)-oriented bcc structure. The sheet resistances are 13.6, 13.5, 9.3, 8.8, and 13.3 Ω for Fe, Co$_{33}$Fe$_{67}$, Co$_{50}$Fe$_{50}$, Co$_{75}$Fe$_{25}$, and Co$_{90}$Fe$_{10}$, respectively, at each MR maximum. In all cases shown in Fig. 2(a), the MR ratio increased as the thickness of the Cu spacer decreased until the Cu threshold thickness was reached where the FM layers became ferromagnetically coupled and the GMR signal was lost because of the absence of the antiparallel (AP) state. Because a clear/nearly AP state appeared in the MR curve in the devices with the Co$_{1-x}$Fe$_x$ layers above the Cu threshold thickness [inset of Fig. 2(b)] and the observed MR ratio in Fe/Cu/Fe is close the value in previous report, we concluded that the large enhancement of MR ratio from Fe to Co$_{33}$Fe$_{67}$ is attributed to an intrinsic mechanism.

Figures 3(a)–3(c) show cross-sectional high angle annular dark field STEM (HAADF-STEM) images as well as bright field TEM images of the Co$_{50}$Fe$_{50}$, Co$_{90}$Fe$_{10}$, and Fe CIP-GMR SV samples with Cu spacer layers. Note that the images were taken along the [100] zone axis of the MgO substrate. An fcc Co$_{90}$Fe$_{10}$ layer grew on the MgO(001) substrate with the orientation relationship (110)$_{\text{Co}_{90}\text{Fe}_{10}}$/[001]$_{\text{MgO}}$, [111]$_{\text{Co}_{90}\text{Fe}_{10}}$/[010]$_{\text{MgO}}$ with a few misfit dislocations at the interface. Figure 3(a) indicates that the Co$_{90}$Fe$_{10}$/Cu/Co$_{90}$Fe$_{10}$ grew epitaxially with rather smooth Co$_{90}$Fe$_{10}$/Cu interfaces. Its diffraction pattern indicates that the Co$_{90}$Fe$_{10}$ and Cu layers had fcc structures textured with a cube-on-cube orientation relationship. The [110] zone axis is parallel to the electron beam, indicating that the spacer layer consisted of the equilibrium fcc Cu phase. The bright-field image shows misfit dislocations at the Co$_{90}$Fe$_{10}$/Cu interfaces, which came from the finite lattice mismatch between the Co$_{90}$Fe$_{10}$ and Cu (∼2% mismatch).

The HAADF-STEM images for the Co$_{50}$Fe$_{50}$ and Fe-based SVs [Figs. 3(b) and 3(c)] also show the epitaxial growth with atomically smooth Co$_{50}$Fe$_{50}$/Cu and Fe/Cu interfaces. The diffraction patterns of the Co$_{50}$Fe$_{50}$ and Fe layers indicated (002)-plane oriented epitaxial growth. The diffraction patterns of the Cu spacer layer were identical to those of the respective ferromagnetic layers in the Co$_{50}$Fe$_{50}$ and Fe-based SV, suggesting that the Cu spacer grew in similar fashion as the lower Co$_{50}$Fe$_{50}$ and Fe layers, forming a metastable bcc structure instead of a thermal equilibrium fcc structure, as reported in a previous study on Fe/Cu/Fe. No dislocations can be seen at the Co$_{50}$Fe$_{50}$/Cu and Fe/Cu interfaces in the STEM images, because the lattice spacing of bcc Cu was reported to be 2.88 Å, which is very close to that of the lower Co$_{50}$Fe$_{50}$ and Fe layers. The bright-field images from both samples show very coherent Co$_{50}$Fe$_{50}$/Cu and Fe/Cu interfaces.
FIG. 3. TEM images of spin valve devices with Co$_{90}$Fe$_{10}$, Co$_{50}$Fe$_{50}$, and Fe showing the highest MR ratio in each case. The images include HAADF-STEM images, nanobeam electron diffraction patterns for ferromagnetic and spacer layers, and bright-field images of (a) Co$_{90}$Fe$_{10}$, (b) Co$_{50}$Fe$_{50}$, and (c) Fe-based spin valve samples.

without any visible misfit dislocations throughout a wide region, suggesting that there was almost no lattice mismatch at the interfaces. Additional TEM images taken from a different zone axis direction of Co$_{50}$Fe$_{50}$ (Fig. S1) confirmed the formation of bcc Cu. Therefore, we conclude that the nearly complete in-plane lattice matching in both Co$_{50}$Fe$_{50}$/Cu/Co$_{50}$Fe$_{50}$ and Fe/Cu/Fe arises from the formation of a metastable bcc Cu spacer. We also took TEM images of a Co$_{50}$Fe$_{50}$/Cu/Co$_{50}$Fe$_{50}$ sample with a thicker Cu region (~4.3 nm) in which the MR ratio decayed to 15% (see Fig. S2). The metastable bcc Cu structure remained even at this thickness, but we found the misfit dislocations at the Co$_{50}$Fe$_{50}$ interface and distortion of the lattice of the Cu spacer. Such deterioration of the interfacial lattice matching with growing Cu thickness seems to be the cause of the rapid reduction in the MR ratio against Cu thickness in Co$_{33}$Fe$_{67}$, Co$_{50}$Fe$_{50}$, and Co$_{75}$Fe$_{25}$.

We investigated the interlayer exchange coupling between two Co$_{50}$Fe$_{50}$ layers through a bcc Cu spacer to see if we could get a further enhancement of the MR ratio by making a multilayer CIP-GMR structure. The investigation of the Cu thickness dependence of the magnetization curve for a device with a MgO-subs./Co$_{50}$Fe$_{50}$(3 nm)/Cu/Co$_{50}$Fe$_{50}$(3 nm)/MgO capping layer structure revealed a clear antiferromagnetic coupling with a coupling field of 12 mT for a 1.6-nm-thick Cu spacer, as shown in Fig. 4(a). The MR ratio of CIP-GMR for the corresponding sample reached 40.5%, which is nearly twice that of SV-type CIP-GMR, and the largest value at room temperature ever reported for trilayer CIP-GMR devices (reported MR ratios in CIP-GMR is summarized in Fig. S3). This improvement relative to the SV type can be explained by the specular reflection of the conductive electrons at the Co$_{50}$Fe$_{50}$/MgO capping layer interfaces.\(^7\) Both MgO substrate and MgO capping layer interfaces reflect electrons back into the FM/NM layers without losing kinetic energy, leading to additional spin-dependent scattering; this increases the MR ratio. Note that we observed smaller MR ratio of about 30% in the another device with 6 nm-thick bottom and top Co$_{50}$Fe$_{50}$ layers, which suggests the contribution of the specular reflection from MgO on the enhancement of the MR ratio. A [Co$_{50}$Fe$_{50}$(3 nm)/Cu(1.6 nm)]$_{3}$/Co$_{50}$Fe$_{50}$ multilayer structure was also fabricated. The M-H curve showed almost zero magnetization at zero field, which clearly indicates an antiparallel magnetization configuration [Fig. 4(b)]. The MR ratio was up to 73.3% at room temperature. The HAADF-STEM image in Fig. 4(c) shows that the Cu had a well-grown epitaxial bcc structure throughout the sample. This huge multilayer structure maintains the interfacial coherency from the bottom to the top layer through the metastable growth of bcc Cu, which contribute to the large MR ratio.

Finally, to understand the mechanism behind the behavior of the MR ratio, we performed first-principles calculations of the electronic band dispersion of Co$_{50}$Fe$_{50}$, Fe, and bcc Cu (see the supplementary material, Fig. S3). The dispersive 4s bands that would hold the conduction electrons at the Fermi level in bcc Cu are unoccupied in both the majority- and minority-spin bands for Fe. In contrast, only the majority-spin band in Co$_{50}$Fe$_{50}$ has a similar occupied 4s band at $E_F$ because the position of the Fermi level in Co$_{50}$Fe$_{50}$ is higher than in Fe. To elucidate the effect of this difference in band matching between Fe/Cu and Co$_{50}$Fe$_{50}$/Cu on
spin-dependent transport, we performed electronic transport calculations on magnetic junctions having the structures observed in the TEM analysis, i.e., (011)-oriented all fcc-CoFe/Cu/fcc-CoFe, (001)-oriented all bcc CoFe/Cu/fcc-CoFe, and Fe/Cu/Fe. Note that we considered B2-ordered CoFe for the transport calculation of CoFe/Cu/fcc-CoFe. Figures 5(a)–5(c) show the in-plane wave vector ($k_x$, $k_y$) dependence of the majority-spin and the minority-spin transmittance for each junction in the parallel magnetization configuration. As shown in Figs. 5(a)–5(c), there are clear differences at the boundary edge of the two-dimensional (2D) first Brillouin zone ($k_x$, $k_y$) = (±0.5, ±0.5) and ($k_x$, $k_y$) = (−0.5 to 0.5, ±0.5)[2π/a]. In the majority-spin transmittance, fcc-CoFe/Cu/fcc-CoFe and B2-CoFe/Cu/fcc-CoFe show large transmittances around the zone boundaries, the four corners of ($k_x$, $k_y$) = (±0.5, ±0.5), and over a wide range of $k_x$ ± 0.5 and $k_y$ ± 0.5. In the ballistic transport calculation based on the Landauer formula, the $z$ components of the wave-vector of incident electrons perpendicular to the plane are determined by the $k_z$-band crossing points at the Fermi level of the electrode material. Furthermore, the total incident energy of conductive electrons is given by $E = h^2 k^2/(2m)$, where $k^2 = k_x^2 + k_y^2 + k_z^2$. Thus, the majority-spin transmittance with a large in-plane wave vector ($k_x$, $k_y$) corresponds to that of the incident electrons with a glancing angle to the plane. In the CIP-GMR effect, the momentum vectors of conductive electrons are almost parallel to the plane, indicating that the transport properties of electrons with a glancing angle are very important. Therefore, the large majority-spin transmittance $G_1 > 2G_0$ and the small minority-spin transmittance $G_1 < 0.5G_0$ around the boundary edge of the 2D Brillouin zone of fcc-CoFe/Cu/fcc-CoFe and B2-CoFe/Cu/fcc-CoFe provide high spin asymmetry to conductive electrons parallel to the plane, resulting in a large CIP-GMR ratio. On the other hand, the majority-spin transmittance of Fe/Cu/Cu/Fc abruptly decreases to zero around the edge of the 2D Brillouin zone, leading to low spin asymmetry in the conductive electrons parallel to the plane. These calculated results are consistent with the experimental results. There are two possible reasons for higher MR ratio in B2-CoFe/Cu/fcc-CoFe than that in fcc-CoFe/Cu/fcc-CoFe: the first is the formation of dislocation-free perfectly coherent interfaces that suppress spin-independent electron scattering at the interfaces. Another is the much smaller transmittance of the minority-spin electron in the wide region of ($k_x$, $k_y$) plane that can be seen by comparing Fig. 5(e) with Fig. 5(d), suggesting larger spin-asymmetry of interface transmittance in CoFe/Cu/fcc-CoFe.

Now, let us examine the band dispersion in the $k_x$ direction along [001] for $k_{y} = 0.44, 0.46, 0.48, 0.50[2π/a]$ at $k_{x} = 0.3[2π/a]$ in bcc-Cu, bcc-CoFe, and bcc-Fe [indicated by the white lines in Figs. 5(b) and 5(c)] in order to reveal the transport properties around the boundary edge of the 2D Brillouin zone. As shown in Fig. 5(h), there are two broad dispersive $s$–$d$ bands at $k_{x} = 0.44$ in CoFe: one crosses the Fermi level; the other starts from the origin around $E − E_F = 0.5$ eV. With increasing $k_x$, the upper $s$–$d$ band falls and the lower $s$–$d$ band rises toward the Fermi level. At $k_x = 0.5$, these two bands degenerate and cross the Fermi level. Therefore, the number of conductive channels of the CoFe electrode at the Fermi level increases from one to two around $k_x = 0.46$. Similar behavior can be observed in the band dispersion of bcc-Cu along [001] for $k_{x} = 0.44−0.50$ at $k_{y} = 0.3$ [Fig. 4(g)], which results in a sudden increase in the majority-spin transmittance by a factor of two for $k_{y} = 0.44−0.50$ in B2-CoFe/Cu/fcc-CoFe. On the other hand, the $k_z$-direction band dispersions of bcc-Fe shows different behavior from those of CoFe [Fig. 5(i)]. This is because there are fewer valence electrons in Fe than in CoFe and the Fermi level of bcc-Fe is located near the bottom of the lower $s$–$d$ band. The lower $s$–$d$ band rises with increasing $k_x$, and finally, at $k_x = 0.48$, this band does not cross the Fermi level. Thus, the conductive channel of bcc-Fe decreases from one to zero around $k_x = 0.48$, leading to poor band matching between bcc-Fe and bcc-Cu around the boundary edge of the 2D Brillouin zone. Note that we also fabricated a current-perpendicular-to-plane (CPP) GMR nanopillar device using...
the bcc-Co$_{50}$Fe$_{50}$/Cu/Co$_{50}$Fe$_{50}$ film to confirm the validity of this analysis (shown in the supplementary material, Fig. S4). The CPP-GMR device had a much smaller MR ratio (about 4% with the value of $\Delta R_A$ is 1.8 m$\Omega$ $\mu$m$^2$ at RT) than that of the CIP-GMR device. This suggests that a large spin-asymmetry of the transmittance cannot be obtained for electrons propagating in the perpendicular direction, as predicted by the calculations illustrated in Figs. 5(b) and 5(e). These results also suggest that first-principles calculations of ballistic transmittance in a 2D Brillouin zone can be used to predict whether various stacking structures intended for exploiting CIP-GMR will have a high MR ratio.

**IV. CONCLUSIONS**

In conclusion, we thoroughly investigated the CIP spin-dependent transport properties through microstructure analyses and first-principles calculations of Co$_{1-x}$Fe$_x$ layers with a metastable bcc Cu spacer layer. A perfectly lattice-matched metastable bcc Cu

---

**FIG. 5.** In-plane wave-vector ($k_x$, $k_y$) dependence of majority- and minority-spin transmittance at the Fermi level for (a) and (d) (011)-oriented fcc Co$_{90}$Fe$_{10}$/Cu/Co$_{90}$Fe$_{10}$, (b) and (e) (001)-oriented bcc Co$_{50}$Fe$_{50}$/Cu/Co$_{50}$Fe$_{50}$, and (c) and (f) (001)-oriented bcc Fe/Cu/Fe in the parallel magnetization configuration. The majority-spin band dispersion of (g) bcc-Cu, (h) bcc-Co$_{50}$Fe$_{50}$, and (i) bcc-Fe for $k_y = 0.44, 0.46, 0.48, 0.50 \ [2\pi/a]$ at $k_x = 0.3 \ [2\pi/a]$. The corresponding position in the ($k_x$, $k_y$) plane for (g)–(i) is indicated by the white line in (b) and (c).
layer was grown on both Co\textsubscript{50}Fe\textsubscript{50} and Fe layers without any notable misfit dislocations; however, we found a large difference in the MR ratio between the spin-valve CIP-GMR device with Co\textsubscript{50}Fe\textsubscript{50} layers (26.5\%) and ones with Fe layers (5\%). First-principles calculations for the spin-dependent transmittance at Co\textsubscript{50}Fe\textsubscript{50} indicated a strong enhancement in the transmittance of majority-spin electrons for bcc-Co\textsubscript{50}Fe\textsubscript{50}/bcc-Cu/Co\textsubscript{50}Fe\textsubscript{50} but a rapid drop in transmittance to zero in bcc-Fe/bcc-Cu/Fe near the boundary of the two-dimensional Brillouin zone. This result clearly explains the difference between the observed MR ratios between the Co\textsubscript{50}Fe\textsubscript{50} and Fe-based devices. It further suggests that a calculation of ballistic transmittance in a GMR stack can predict the MR ratio of CIP-GMR devices, which would be a guiding principle for further enhancement of the MR ratio in these devices. Accordingly, we achieved the MR ratio of 40.5\% in an antiferromagnetically coupled trilayer bcc-Co\textsubscript{50}Fe\textsubscript{50}/bcc-Cu/Co\textsubscript{50}Fe\textsubscript{50} including a specular reflection layer, which is the highest MR ratio ever reported for the trilayer CIP-GMR. However, although the MR ratio is large, current bcc-Co\textsubscript{50}Fe\textsubscript{50}/bcc-Cu/Co\textsubscript{50}Fe\textsubscript{50} cannot be used as a magnetic field sensor as is. Further study is required to increase the sensitivity of the device, for example, by using a softer ferromagnetic layer having a similar bcc structure as Co\textsubscript{50}Fe\textsubscript{50} to achieve more linear response instead of a jump. More exploration in bcc-Co\textsubscript{50}Fe\textsubscript{50} and bcc-Cu devices using an industrially more viable polycrystalline structure with Si wafer substrates also have to be considered for future magnetic sensor application. Finally, we conclude that although CIP-GMR devices are the original spintronics form that was thoroughly studied from its discovery, there is still a room for enhancement of their MR properties by exploring new materials including metastable structures such as bcc-Cu to achieve a large spin asymmetry at interfaces and that such development may lead to a new class of sensitive magnetic sensors.

SUPPLEMENTARY MATERIAL

The supplementary material for the additional STEM result, the first-principles calculation of band structures, and the comparison with CPP-GMR and MR ratio roadmap is available online.

ACKNOWLEDGMENTS

The authors thank Hiroyasu Nakayama for help in making the multilayer GMR structure and Natsuko Kojima and Kyoko Suzuki for their technical support. This work was supported by a Grant-in-Aid for Scientific Research (S) (Grant No. 17H06152) from the Japan Society for the Promotion of Science (JSPS).

REFERENCES

1. M. N. Baitich, J. M. Broto, A. Fert, F. N. Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. 61, 2472 (1988).
2. G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B 39, 4828 (1989).
3. P. Grünberg, R. Schreiber, Y. Pang, U. Walz, M. B. Brodsky, and H. Sowers, J. Appl. Phys. 61, 3750 (1987).
4. B. Dieny, V. S. Speriosu, S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B 43, 1297 (1991).
5. J. Loguies, D. Krauss, R. Kruppe, J. Rittinger, P. Taptimthong, A. Wienecke, L. Rissing, and M. C. Wurtz, Sensors 15, 28665 (2015).
6. S. Cardoso, D. C. Leitao, T. M. Dias, J. Valadeiro, M. D. Silva, A. Chicharo, V. Silverio, J. Gaspar, and P. P. Freitas, J. Phys. D: Appl. Phys. 50, 213001 (2017).
7. C. Zhang, K. Zhu, S. Cardoso de Freitas, J.-Y. Chang, J. E. Davies, P. Eames, P. P. Freitas, O. Kazakov, C. Kim, C.-W. Leung, S.-H. Liou, A. Ognev, S. N. Piramanayagam, F. Ripka, A. Samardak, K.-H. Shin, S.-Y. Tong, M.-J. Tung, S. X. Wang, S. Xue, X. Yin, and P. W. T. Pong, IEEE Trans. Magn. 55, 1 (2019).
8. K. B. Klaspers, J. Van Peppen, and X. Xing, IEEE Trans. Magn. 42, 108 (2006).
9. T. Todorov, E. Y. Tsymbal, and D. Pettitof, Phys. Rev. B 54, R12685 (1996).
10. A. Vedyayev, M. Chshiev, N. Ryzhanova, B. Dieny, C. Cowache, and F. Brouers, J. Magn. Magn. Mater. 172, 53 (1997).
11. E. Y. Tsymbal and D. G. Pettitof, J. Magn. Magn. Mater. 202, 163 (1999).
12. F. E. Erler, P. Zahn, and I. Merti, Phys. Rev. B 64, 094408 (2001).
13. E. Y. Tsymbal and D. G. Pettitof, Solid State Phys. 56, 113–237 (2001).
14. F. T. Parker, H. Oesterreicher, and E. Fullerton, J. Appl. Phys. 66, 5988 (1989).
15. R. Schad, C. D. Potter, P. Belien, G. Verbanck, J. Dekoster, G. Langouche, V. V. Moshchalkov, and Y. Bruynseraede, J. Magn. Magn. Mater. 148, 331 (1995).
16. X. Peng, A. Morrone, K. Nikolaev, M. Kief, and M. Ostrowski, J. Magn. Magn. Mater. 321, 2902 (2009).
17. M. A. Seigler, IEEE Trans. Magn. 43, 651 (2007).
18. H. Yuasa, Y. Kamiguchi, and M. Sahashi, J. Magn. Magn. Mater. 267, 53 (2003).
19. V. V. Ustinov, M. A. Milaev, and L. I. Naumova, Phys. Met. Metallogr. 118, 1300 (2017).
20. B. Heinrich, Z. Celinski, J. F. Cochran, W. B. Murr, J. Rudd, Q. M. Zhong, A. S. Arrott, K. Myrtle, and J. Kirschner, Phys. Rev. Lett. 64, 673 (1990).
21. I. Ohnuma, H. Enoki, O. Ikeda, R. Kainuma, H. Ohtani, B. Ogund, S. N. Piramanayagam, F. Ripka, A. Samardak, K.-H. Shin, S.-Y. Tong, and M.-J. Tung, J. Magn. Magn. Mater. 253, 10242 (1999).
22. A. Smogunov, A. Dal Corso, S. Gironcoli, and P. GEAZZOLI, https://www.quantum-espresso.org/.
23. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
24. H. Joon Choi and J. Ihm, Phys. Rev. B 59, 2267 (1999).
25. A. Smogunov, A. Dal Corso, and E. Tosatti, Phys. Rev. B 70, 045417 (2004).
26. T. L. Monchesky, B. Heinrich, R. Urban, and M. Klaua, Phys. Rev. B 60, 10242 (1999).
27. K. Yuge, A. Seko, K. Kobayashi, T. Tatsuoka, S. R. Nishitani, and H. Adachi, Materials Transactions 45, 1473 (2004).