

Searching for Cu-*X* Spacers Using a Half-Metallic Co₂FeGa_{0.5}Ge_{0.5} Electrode to Boost Magnetoresistance in CPP-GMR Devices: A First-Principles Study

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Current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) devices are promising candidates for next-generation hard disk drive (HDD) read heads due to their low resistance-area product (RA) and high-speed operation. This study investigates Cu-based binary alloys (Cu-*X*) as alternative spacers to conventional Ag spacer in Co₂FeGa_{0.5}Ge_{0.5} (CFGG)-based CPP-GMR devices using first-principles calculations. Majority-spin ballistic conductance, which is essential in the large spin-asymmetry of the interface resistance, was evaluated for CFGG/Cu-*X*/CFGG(001) trilayers in comparison with CFGG/Ag/CFGG(001) trilayer. The results show that several spacer materials have possibilities to enhance the MR ratio compared to Ag. In particular, we found that B2-CuZn will be the most suitable for CFGG electrode. The large majority-spin conductance of CFGG/CuZn/CFGG(001) is attributed to *p-d* hybridization between CFGG and Cu *s*-states near the Γ point, which enhances electron transport efficiency, while the thermal stability of the interface spin-moment at CFGG/CuZn(001) is slightly smaller than that of CFGG/Ag(001). Finally, we proposed that the insertion of Ag layer between the CFGG and CuZn interfaces will enhance not only the spin-asymmetry in the interface resistance but also the thermal stability of the interface spin-moment.

Keywords: First-principles calculation; Spin-dependent transport properties; Cu-based binary alloys; Half-metallic Co₂FeGa_{0.5}Ge_{0.5}; CPP-GMR

1. Introduction

Current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) devices are metallic multilayer structures composed of alternating ferromagnetic (FM) and non-magnetic (NM) layers. When an electric current flows perpendicular to the plane, the magnetoresistance (MR) effect arises from spin-dependent scattering in the bulk FM layers and at FM/NM interfaces, depending on the relative magnetization orientation in FM layers. CPP-GMR devices have emerged as promising candidates for next-generation read heads in hard disk drives (HDDs) with areal densities exceeding 2 Tbit/in². Their main advantage in applications over tunneling magnetoresistance (TMR) devices lies in their lower resistance-area (RA) product[1–3], a result of their fully metallic structure. Additionally, CPP-GMR sensors offer shorter read gaps and improved electrode contacts[2,4], making them well-suited for scaling magnetic recording heads to accommodate smaller media bit sizes[5–7]. However, a major challenge of CPP-GMR devices is their relatively lower MR ratio, typically less than 100% at room temperature[8–10], which is significantly lower than that of TMR devices[11,12]. To overcome this limitation, extensive research has focused on the incorporation of half-metallic Heusler alloys as electrode materials[13–16]. These alloys offer large spin polarization, high Curie temperature, and structural stability, which significantly improve MR ratios, as demonstrated in various studies[17–23]. These advances highlight the critical role of material innovation in optimizing CPP-GMR devices to meet the performance requirements of future HDDs with ultra-high areal densities.

The resistance-area product difference (ΔRA) between antiparallel and parallel magnetizations in CPP-GMR device, related to the magnetoresistance (MR) ratio, is described by the two-current series resistor (2CSR) model[24]. This model includes spin-dependent scattering and is characterized by two essential parameters: the bulk spin-asymmetry coefficient (β) and the interfacial spin-asymmetry coefficient (γ). The bulk spin-asymmetry coefficient, which pertains to the FM layers, is expressed as $\beta = \frac{\sigma_{\text{bulk}}^{\uparrow} - \sigma_{\text{bulk}}^{\downarrow}}{\sigma_{\text{bulk}}^{\uparrow} + \sigma_{\text{bulk}}^{\downarrow}}$ where $\sigma_{\text{bulk}}^{\uparrow}$ and $\sigma_{\text{bulk}}^{\downarrow}$ are the conductivities for majority and minority spin channels, respectively. Similarly, the interfacial spin-asymmetry coefficient is defined as $\gamma = \frac{R_{\text{int.A}}^{\downarrow} - R_{\text{int.A}}^{\uparrow}}{R_{\text{int.A}}^{\downarrow} + R_{\text{int.A}}^{\uparrow}}$ where $R_{\text{int.A}}^{\uparrow}$ and $R_{\text{int.A}}^{\downarrow}$ denote the resistance-area products for majority and minority spins at the FM/NM interface, respectively. Maximizing the

CPP-GMR effect, therefore, relies on optimizing both β and γ , necessitating the discovery of novel FM and NM materials.

High spin polarization of FM materials has already been well-studied[25–27] in terms of half-metallic FM with 100% spin polarization at the Fermi level especially for full Heusler alloys X_2YZ . In our previous works[28] using machine learning analysis and first-principles calculations that include finite temperature effects, we found that among full Heusler alloys X_2YZ , only those with Co at the X -site and Mn and Fe at the Y -site can be identified as highly spin-polarized ferromagnetic materials at room temperature. From an experimental perspective, half-metallic Heusler alloys have attracted much attention in spintronics due to their MR ratios[29–31]. Among these, Co-based full-Heusler alloys exhibit outstanding properties, which are supported by many theoretical studies[32–34]. CPP-GMR devices utilizing Co-based full-Heusler alloy electrodes, such as Co_2MnSi (CMS), $\text{Co}_2\text{Mn}_{0.6}\text{Fe}_{0.4}\text{Si}$ (CMFS), and $\text{Co}_2\text{FeGa}_{0.5}\text{Ge}_{0.5}$ (CFGG), combined with Ag or Ag-based spacers, have achieved MR ratios exceeding 30% at room temperature[35–38]. In particular, CFGG stands out with spin polarization values of up to 93% at 10 K and 84% at 290 K, outperforming other Co-based Heusler compounds[39,40]. These advantages make CFGG a promising FM electrode in this study.

Despite significant progress, there remains room to develop new suitable NM spacer materials to achieve higher MR ratio. NM spacer plays a critical role in CPP-GMR device performance and scalability directly through interfacial spin-dependent phenomena due to the spin-asymmetric band matching with FM electrodes. An ideal NM spacer should exhibit low spin-orbit coupling to ensure a long spin-diffusion length as well as electronic band matching, and should possess structural and lattice compatibility with FM electrodes to prevent defect and dislocation formation at the interfaces. Previous studies on CPP-GMR devices with Co-based Heusler electrodes have demonstrated a high MR ratio when Ag or Ag-based alloys are used as NM spacer materials[39,41–43]. This performance has been attributed to the strong spin-asymmetric density of states and proper lattice matching between Ag and the Heusler electrodes, combined with the long spin-diffusion length of Ag. However, Ag-based spacers face limitations in achieving MR ratios sufficient for practical read-head applications[44–46]. To address these challenges, the development of new spacer materials with improved interfacial band matching (γ parameter), high lattice compatibility, and long spin-diffusion lengths will be required.

In this study, we theoretically investigate Cu-based binary alloy (Cu- X) spacers as potential alternatives to Ag-based spacers. Cu- X spacers have several desirable properties, including high conductivity, low spin-orbit coupling (facilitating extended spin diffusion length) and a small lattice mismatch with the CFGG electrode. In addition, their cubic B2- or L1₂-type structures are highly compatible with the cubic L2₁ structure of CFGG, ensuring better symmetry and structural matching at the interface. Several experimental studies have been carried out on Cu- X spacers, such as CuZn[47], CuN[48], and CuIn[49]. Based on these findings, we explore a wider range of Cu- X spacer materials and evaluate their potential for spin-dependent transport, aiming to outperform the spin transport properties of conventional Ag spacers and improve the MR ratio.

In selecting alloying elements for Cu-based spacers, we focused on 3d transition metals with filled d -shells to ensure non-magnetic behavior, which is essential for preserving spin polarization in CPP-GMR junctions. Elements such as Mn and Ni, although abundant, are less suitable for this purpose. Mn tends to retain local magnetic moments even in alloyed states, potentially introducing spin-flip scattering and unwanted magnetic interactions at the interface[50]. Ni, being ferromagnetic, also contributes to significant spin-flip scattering due to its short spin diffusion length, which can reduce spin asymmetry[51,52]. In contrast, elements like Zn, with a fully filled 3d shell and no magnetic moment, enable the tuning of interfacial electronic properties while maintaining high spin transport efficiency. Therefore, our choice of Cu- X alloy systems is guided not only by cost and conductivity, but also by the requirement for structural compatibility and non-magnetic character to optimize spin-dependent transport.”

Furthermore, we predict the temperature dependence of the MR ratio in Heusler-based CPP-GMR devices through interface exchange stiffness, which is a key parameter in assessing spin transport efficiency through the interface. Finally, we conclude that optimizing Cu- X spacer to enhance γ through spin-asymmetric interface matching is crucial for achieving higher MR ratio in CPP-GMR devices.

2. Computational method

According to the 2CSR model[24], the spin-dependent interface scattering typically dominates both the resistance and magnetoresistance when the layer thickness is not excessively

thick[53,54]. Therefore, to understand CPP-GMR, underlying mechanisms of spin-dependent interface resistance need to be clarified[55]. However, in practical devices, the current is a combination of ballistic and diffusive mechanisms due to scattering at interfaces and material imperfections. The Landauer theory is particularly useful for analyzing ballistic transport in atomic-scale contacts, where interfacial resistance at FM/NM junctions plays a significant role in determining transport properties. Therefore, we performed first-principles ballistic transport calculations based on the Landauer formula[56] to investigate the majority-spin conductance in conventional Ag spacers and Cu-spacer materials. To confirm the half-metallic nature of $\text{Co}_2\text{FeGa}_{0.5}\text{Ge}_{0.5}$ (CFGG), we provide the calculated band dispersion and spin-resolved density of states in Figure S1 of the Supplementary Material. These results show a metallic majority-spin channel and a pseudo-gap in the minority-spin channel, supporting the high spin polarization at the Fermi level ($\sim 71\%$) used in our transport model.

The calculations were carried out using density functional theory (DFT) within the generalized gradient approximation (GGA) for the exchange-correlation energy[57]. We employed the plane-wave basis set and the ultrasoft pseudopotential method, implemented in the QUANTUM ESPRESSO code[58] in order to facilitate the structure optimization of FM/NM junctions. A $10 \times 10 \times 1$ k-point grid was used for Brillouin zone integrations. The wave function and charge density cutoff energies were set to 40 Ryd and 400 Ryd, respectively. To justify the choice of a plane-wave cutoff energy of 40 Ry, we performed convergence tests on total energy and conductance for the CFGG/CuZn/CFGG(001) junction with FeGa- and Cu-terminated interfaces. The cutoff energy was varied from 30 to 50 Ry, with the charge density cutoff set to 10 times the wavefunction cutoff. The total energy variation fell below 0.0005 Ry/atom beyond 40 Ry, and conductance changes were less than 0.0015 [e^2/h], confirming that 40 Ry provides sufficient accuracy for both energy and transport calculations, as shown in Figure S2. The ballistic transport calculations with semi-infinite boundary conditions were performed by the method proposed by Choi and Ihm[59,60]. The total conductance G is obtained by summing the transmission probability $T(k_{\parallel})$ of electrons at the Fermi level (E_F) for each in-plane wave vector $k_{\parallel}$ across the two-dimensional (2D) Brillouin zone (BZ) as follows:

$$G = \frac{e^2}{h} \sum_{k_{\parallel}} T(k_{\parallel}), \quad (1)$$

where e is the elementary charge and h is Planck's constant. The transmission probability was calculated using a 50×50 k-point grids in the 2D BZ integrations. Then, the interface resistance-area product ($R_{\text{int}}A$) value was calculated as A/G to evaluate the γ parameter, where A represents the cross-sectional area. We constructed tetragonal supercells for CFGG/Ag/CFGG(001) and CFGG/Cu- X /CFGG(001) junctions. The in-plane lattice parameters for all junctions were fixed at 4.06 Å, corresponding to $a_0/\sqrt{2}$ where a_0 is the lattice constant of bulk CFGG (5.740 Å). The lattice mismatch between the Ag spacer and the CFGG electrode was 1.691%, and various Cu- X spacers for B2-type and L1₂-type structures are listed in Table I and

Table II, respectively.

To further understand the impact of interface terminations on the stability of interface spin-moment at finite temperatures, we calculated the exchange stiffness constant at the interface. At 0 K, spin moments are perfectly aligned, resulting in a maximum MR ratio due to the collinear spin alignment. However, as temperature increases, thermal fluctuations disrupt spin alignment, inducing spin-flip scatterings of electrons through the junctions. These disturbances weaken spin-asymmetry of interface resistance, leading to reduction in the MR ratio at finite temperatures. Interface regions are particularly susceptible to spin fluctuations because of reduction of the exchange interactions between the ferromagnetic layer and the nonmagnetic spacer. To clarify this effect, we estimated exchange stiffness constants of interface spin moments for the most stable termination of CFGG/Cu- X (001) and CFGG/Ag(001)[61]. Using the Vienna *Ab initio* Simulation Package (VASP)[62,63], we calculated the total energy variation, $E(\theta)$, as a function of the tilting angle (θ) of local spin moment via constrained magnetization method[64]. The results were fitted to the expression $E(\theta) = B(1 - \cos \theta)$, where B represents the inter-atomic-layer exchange stiffness constant. Note that VASP was used to evaluate interfacial exchange stiffness due to its more robust

and efficient handling of constrained magnetization with non-collinear magnetism, which is not as practical or stable in Quantum ESPRESSO.

For the initial screening of Cu-*X* materials as a NM spacer layer, we used the Open Quantum Materials Database (OQMD)[65,66] to identify candidates based on their stability, characterized by a negative formation energy and a lattice mismatch with the CFGG electrode of less than ~10%. It is worth noting that bulk materials with positive formation energies are not necessarily unstable when forming thin-film junctions. In such cases, the stability of interfaces (i.e., interface formation energy) is an important factor in determining their suitability. Therefore, bulk materials with positive formation energies below 0.45 eV/atom were also considered in this study. While there are many B2- and L1₂-type Cu-*X* materials, evaluating all possible candidates of Cu-*X* would be computationally intensive and time-consuming, potentially limiting the depth of analysis for each candidate. To ensure a rigorous and detailed investigation, a systematic research approach was maintained in this study, prioritising materials with promising stability, interfacial compatibility, and technological potential, as indicated by their transport properties.

Table I In-plane lattice mismatch (%) between Cu-*X* spacers with B2-type structure and the Co₂FeGa_{0.5}Ge_{0.5} electrode.

B2-type structure	In-plane lattice mismatch (%)
CuAl	4.178
CuAs	9.678
CuAu	8.531
CuGa	6.320
CuGe	8.700
CuMg	10.152
CuPd	4.982
CuPt	5.813
CuRu	3.408
CuSi	4.872
CuZn	3.405

Table II In-plane lattice mismatch (%) between Cu- X spacers with L1₂-type structure and Co₂FeGa_{0.5}Ge_{0.5} electrode.

L1 ₂ -type structure	In-plane lattice mismatch (%)
Cu ₃ Ag	6.919
Cu ₃ Al	9.201
Cu ₃ Au	6.635
Cu ₃ Ga	8.799
Cu ₃ Ge	8.235
Cu ₃ Pd	8.348
Cu ₃ Pt	7.951
Cu ₃ Zn	9.268

Figure. 1 shows the supercell structure of CFGG/Ag/CFGG(001) and CFGG/Cu- X /CFGG(001), respectively, with possible interfacial terminations for the electrode and spacer noted. The figure illustrates an example with the FeGe termination of CFGG/Ag(001) and CFGG/Cu- X (001). The interfacial termination of the CFGG/Ag(001) interface varies depending on the electrode materials (Figure. 1(a)), resulting in three types of terminations: FeGa, Co, and FeGe terminations. For the CFGG/Cu- X (001), the interfacial termination depends on the specific structure of the spacer (Figure. 1(b)). Cu and X terminations are observed in the B2-type Cu- X spacer, while Cu and Cu X terminations are found in the L1₂-type Cu- X spacer. The supercell consists of 7 atomic layers of the spacer, 21 atomic layers of CFGG for the FeGa termination, and 17 atomic layers of CFGG for the Co and FeGe terminations (See details in Ref.[61]).

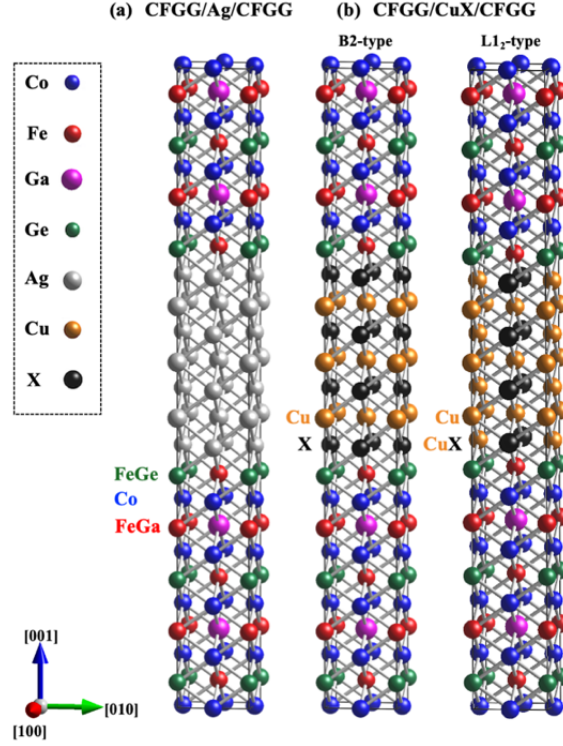


Figure. 1 Supercell of the (a) CFGG/Ag/CFGG(001) trilayers and (b) CFGG/Cu-X/CFGG(001) trilayers for B2-type and L1₂-type structure, respectively, visualized by VESTA [67] with various interfacial terminations.

3. Results and discussion

3.1. Structural stability comparison between CFGG/Cu-X/CFGG(001) and CFGG/Ag/CFGG(001)

To determine the stable interfacial configuration for each termination in CFGG/Cu-X/CFGG(001) and CFGG/Ag/CFGG(001) junctions, we minimized the total energy by relaxing atomic positions with changing the longitudinal supercell size. We then assessed the relative stability of each interface termination in CFGG-based magnetic multilayers with different Cu-X spacers based on the formation energies of various interface terminations, which were obtained the following equation:

$$E_{\text{form}}^{\text{ter}} = E_{\text{tot}}^{\text{ter}} - \sum_i n_i \mu_i. \quad (2)$$

$E_{\text{tot}}^{\text{ter}}$ is the total energy of the supercell for given junction termination, n_i is the number of atoms of species i (Co, Fe, Ga, Ge, Ag, Cu, Al, As, etc.), and μ_i represents their respective chemical potentials. The chemical potential of each constituent atom is assumed to be derived from the bulk total energy per atom of the pure element. Note that although not included in the present work, defect formation energies and site preferences in Cu-based binary alloy spacers will be explored in future studies to assess their thermodynamic stability and provide further insight beyond the transport-based analysis.

Figure. 2 shows the variation of formation energy for FeGe, Co, and FeGa terminated interfaces with each spacer termination in B2-type (Figure. 2(a-b)) and L1₂-type (Figure. 2(c-d)) structures. Among these terminations, the FeGa-terminated interface exhibits the lowest formation energy, indicating the highest interfacial stability. In contrast, the FeGe- and Co-terminated interfaces show comparable stability, although the difference in formation energy was smaller. These trends are consistent with previous calculations for the Ag spacer[61]. This enhanced stability in the FeGa-terminated interface arises from the better match in covalent bond radii between Fe (1.32 Å) and Ga (1.22 Å), which reduces interfacial strain and promotes stronger Fe–Ga bonding. In contrast, the slightly smaller covalent radius of Ge (1.20 Å) leads to bonding mismatches and greater local distortion in the FeGe interface. Additionally, Ga’s lower electronegativity (1.81 vs. Ge’s 2.01) favors more delocalized metallic bonding, further stabilizing the FeGa interface. The relatively higher formation energy of the Co-terminated interface is due to the equivalent positioning of Co atoms, which restricts their movement perpendicular to the interface. Conversely, in the FeGa(Ge)-terminated configuration, Fe and Ga(Ge) atoms are not equivalent, resulting in greater atomic relaxation along the perpendicular direction, which enhances the stability. For the B2-type structure, formation energies range from -0.155 J/m^2 to -0.025 J/m^2 for Cu termination and from -0.176 J/m^2 to -0.024 J/m^2 for X termination, suggesting that Cu interfacial termination is generally more stable than X termination. In contrast, the opposite trend is observed for the L1₂-type structure. The formation energy ranges from -0.147 J/m^2 to -0.061 J/m^2 for Cu termination and from -0.163 J/m^2 to -0.054 J/m^2 for Cu X termination, indicating that Cu X termination is more stable than Cu termination.

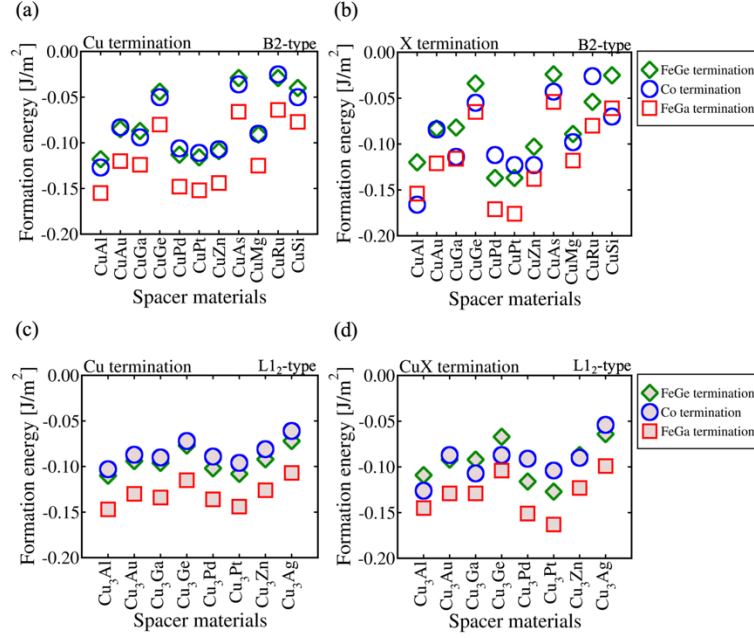


Figure. 2 Formation energies as a function of each spacer material and different interfacial terminations, including FeGe, Co, and FeGa terminations, for CFGG/Cu-*X*/CFGG(001) trilayers. Panels (a) and (b) depict Cu and *X* terminations for the B2-type structure, while panels (c) and (d) show Cu and Cu*X* terminations for the L1₂-type structure.

To evaluate the structural stability of CFGG/Cu-*X*/CFGG(001) trilayers relative to the conventional Ag spacer, we compared the formation energy differences between Cu-*X* and Ag spacers, as shown in Figure. 3. The formation energy difference is defined as:

$$\Delta E_{\text{form}} = E_{\text{form}}(\text{Cu-}X) - E_{\text{form}}(\text{Ag}) \quad , \quad (3)$$

where $E_{\text{form}}(\text{Cu-}X)$ and $E_{\text{form}}(\text{Ag})$ are the formation energies of the Cu-*X* and Ag systems, respectively. Both Cu-*X* and Ag spacers in CFGG-based junctions exhibit negative formation energies, indicating that all these supercell structures are theoretically stable.

In Figure. 3, a positive difference in formation energy indicates that the Ag spacer is more stable, while a negative difference means that the Cu-*X* spacer is more stable than the Ag spacer. Based on these results, the thermodynamic stability of B2-type Cu-*X* spacers relative to the Ag spacer can be classified into two main groups:

- (1) No improvement in stability: Cu- X spacers CuAs, CuGe, CuRu, and CuSi show a small difference in formation energy and comparable structural stability to the Ag spacer.
- (2) Enhanced stability: Cu- X spacers CuAl, CuAu, CuGa, CuMg, CuPd, CuPt, and CuZn exhibit larger negative formation energy differences, indicating improved structural stability over Ag spacers.

Furthermore, all L1₂-type Cu- X spacers show negative formation energy differences, suggesting that they are more thermodynamically stable than the Ag spacer across all terminations. These findings indicate that Cu- X spacers generally exhibit better structural stability than Ag spacers, and that stability is influenced by interface terminations, spacer materials, and crystal structures. Overall, the observed zigzag trend in formation energy as a function of X arises from variations in interfacial bonding strength, driven by differences in atomic size, electronegativity, and chemical bonding between the Cu- X spacers and the terminating atoms of the CFGG electrode.

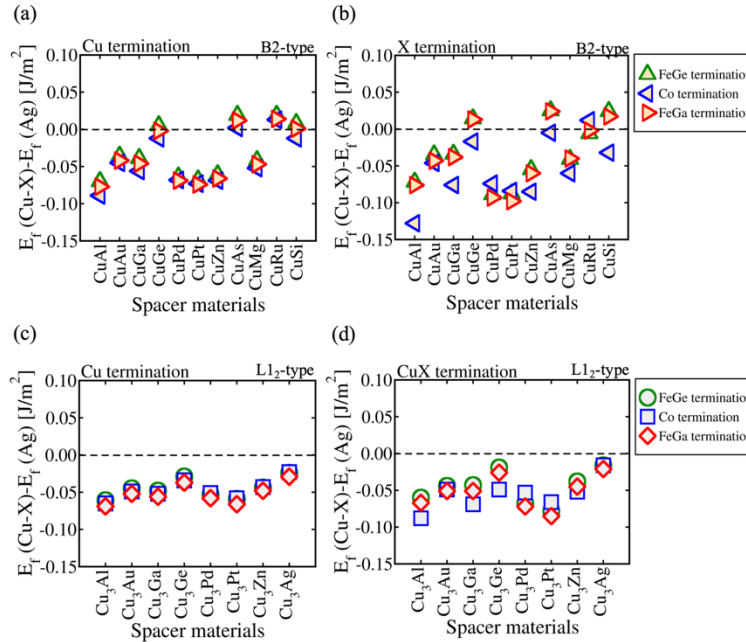


Figure. 3 Formation energy differences of various spacers compared to the Ag spacer, indicating the relative thermodynamical stability of CFGG/Cu- X /CFGG(001) trilayers with FeGe, Co, and FeGa terminations. Panels (a) and (b) depict Cu and X terminations for the B2-type structure, while panels (c) and (d) show Cu and Cu X terminations for the L1₂-type structure. The black dashed line represents the equivalent thermodynamical stability with the Ag spacer ($E_{\text{form}}(\text{Cu-X}) - E_{\text{form}}(\text{Ag}) = 0$).

3.2 Transport properties comparison between CFGG/Cu-*X*/CFGG(001) and CFGG/Ag/CFGG(001)

To investigate the physical factors influencing spin-dependent transport through CFGG electrode and Cu-*X* alloy spacers, we evaluate the majority-spin conductance at the Fermi level in the parallel magnetization configuration. Both B2-type and L1₂-type Cu-*X* structures with various interfacial terminations are considered in this study. To quantify conductance variation relative to the Ag spacer, we calculate the ratio of the conductance variation relative to the Ag spacer using:

$$\text{Conductance variation} = \left[\frac{G(\text{Cu-}X) - G(\text{Ag})}{G(\text{Ag})} \right] \times 100, \quad (4)$$

where $G(\text{Cu-}X)$ and $G(\text{Ag})$ denote the conductance with Cu-*X* and Ag spacers, respectively. Positive values indicate an enhancement in the parallel conductance, i.e., the interface spin asymmetry (γ), compared to the Ag spacer, while negative values signify a reduction in conductance and γ . Figure. 4 presents the normalized majority-spin conductance variation between Cu-*X* and Ag spacers. The conductance variation is strongly influenced by the interface termination, highlighting the critical role of interface electronic structures in spin-dependent transport.

For the B2-type structure (Figure. 4(a-b)), the Co termination consistently exhibits a larger conductance compared to the other terminations, particularly by considering Cu termination. In the Cu termination (Figure. 4(a)), CuAu, CuGa, CuMg, CuPt, CuSi, and CuZn show enhancements of conductance, whereas CuAl, CuAs, CuGe, CuPd, and CuRu result in reductions of conductance across all electrode terminations. In the Cu (B2-type) termination, CuZn achieves significant conductance enhancements of 22.17% for FeGe termination, 36.29% for Co termination, and 44.78% for FeGa termination. In the *X* termination (Figure. 4(b)), CuAu, CuPt, and CuZn exhibit conductance enhancements, while the remaining spacers lead to either comparable or reduced conductance across all electrode terminations. The significant conductance enhancements in *X* (B2-type) termination are observed for CuPt (11.65%) with FeGe termination, CuZn (31.63%) with Co termination, and CuZn (32.37%) with FeGa termination. These results indicate that CuZn consistently enhances the majority-spin conductance in the parallel magnetization, particularly for FeGa and Co terminations, making it a promising spacer candidate.

For the $L1_2$ -type structure (Figure. 4(c-d)), we can find the similar trend on the conductance variation. The Co and FeGa terminations show a significant increase in the conductance compared to the FeGe termination. Most spacer materials exhibit either comparable or improved conductance compared to the Ag spacer. Significant conductance enhancements in Cu ($L1_2$ -type) termination are observed for Cu_3Ge (24.66%) with FeGe termination, Cu_3Ga (29.45%) with Co termination, and Cu_3Ga (33.81%) with FeGa termination, while those in CuX ($L1_2$ -type) termination are observed for Cu_3Zn (7.71%) with FeGe termination, Cu_3Ga (37.02%) with Co termination, and Cu_3Ga (21.29%) with FeGa termination. However, Cu_3Pd and Cu_3Pt significantly exhibit negative conductance variations, with consistently lower conductance at all terminations, including both Cu-terminated (Figure. 4(c)) and CuX -terminated (Figure. 4(d)) interfaces. This suggests that Cu_3Pd and Cu_3Pt are less favorable spacer candidates due to their lower interface spin asymmetry (γ) compared to Ag spacer.

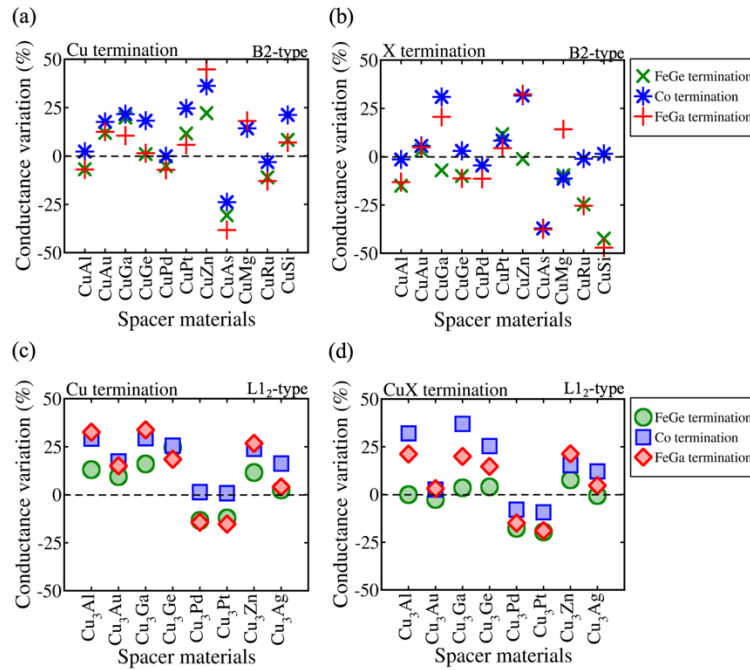


Figure. 4 Majority-spin conductance variation between Cu- X and Ag spacers as a percentage relative to Ag, including FeGe, Co, and FeGa terminations, for CFGG/Cu- X /CFGG(001) trilayers. Panels (a) and (b) depict Cu and X terminations for the B2-type structure, while panels (c) and (d) show Cu and CuX terminations for the $L1_2$ -type structure. Positive and negative values indicate improvements and reductions in conductance compared to the Ag spacer, respectively.

As a result, the B2-type Cu X spacers achieved conductance enhancements ranging from 12% to 45%, while the L1₂-type Cu₃ X spacers exhibited improvement between 8% to 37%. This indicates that the B2-type Cu X spacers are more effective than L1₂-type Cu₃ X spacers in enhancing conductance. It should be noted that in our study, for spacers containing Ag, Au, or Pt—which exhibit relatively strong spin–orbit coupling (SOC)—the effect of SOC on total conductance in the CPP direction is expected to be negligible. This is consistent with the approach adopted in previous studies on similar systems [68]. This negligible effect is attributed to the high spin polarization of the CFGG electrode and the ballistic nature of transport in our model. In such systems, spin-flip scattering is strongly suppressed, and the influence of SOC on spin-dependent transport becomes minimal, particularly in the ballistic regime where elastic scattering dominates. Similar conclusions have been reported for other Co-based Heusler junctions, where SOC was found to have minimal influence on electronic transmission [69] or was not considered [70].

These findings suggest that Cu- X spacers outperform the Ag spacer in CPP-GMR devices with CFGG by improving Fermi surface matching and enhancing interface spin asymmetry. This effect will be further examined in the next section.

3.3. Cu- X spacer potential candidates compared to traditional Ag spacer

To investigate and clarify the physical origin of better Fermi surface matching of Cu- X alloy spacers compared to conventional Ag spacer in CFGG-based magnetic multilayer systems, we study the interface resistance-area product ($R_{\text{int}}A$) and its orbital contributions to conductance. Furthermore, we estimated the exchange stiffness constant for each interfacial termination of CFGG/Cu- X (001) and compared it with that of CFGG/Ag(001) junctions. This parameter is critical for suppressing the reduction of MR ratio at room temperature, ensuring the stability of spin transport in CPP-GMR devices.

3.3.1 Interface resistance area product and its orbital contributions

Figure. 5 shows the relationship between $R_{\text{int}}A$ and majority-spin conductance for Cu- X spacers with various terminations. For comparison, the $R_{\text{int}}A$ and majority-spin conductance of the Ag spacer with each termination are also presented. The results are consistent for B2- and L1₂-type structures with comparable terminations. In Ag spacer, the $R_{\text{int}}A$ values at each electrode termination are 3.616 m Ω μm^2 for FeGe termination, 4.281 m Ω μm^2 for Co termination, and 3.691 m Ω μm^2 for FeGa termination. These values identify potential Cu- X spacer candidates with

a lower $R_{\text{int.}A}$ than Ag. As discussed earlier, the B2-type structure demonstrates a wider range of conductance enhancements compared to the L1₂-type structure. Among all candidates, CuZn spacer (B2-type structure) emerges as the most optimal Cu- X spacer, with the lowest $R_{\text{int.}A}$ value of 2.550 m Ω μm^2 at the FeGa- and Cu-terminated interface, which was identified as the most stable termination.

The promising spacer materials (CuZn, CuPt, Cu₃Ge, Cu₃Ga) were further analyzed by evaluating the majority-spin conductance at the Fermi energy as a function of the in-plane wave vector $k_{\parallel} = (k_x, k_y)$ across the 2D Brillouin zone (BZ) for each of the six interface terminations, as shown in Figure. 6(a-f). The results highlight the lowest $R_{\text{int.}A}$ value for each termination, which are consistent with the data in Figure. 5(a-f). A lower $R_{\text{int.}A}$ value corresponds to higher conductance in the $k_{\parallel}$ region. For the Cu-terminated interface (Figure. 6(a-c)), the CuZn spacer at FeGa termination has the lowest $R_{\text{int.}A}$ value, with a high conducting area along the diagonal region and near the center of the BZ. For the $X/\text{Cu}X$ -terminated interface (Figure. 6(d-f)), the CuZn at FeGa termination shows similar characteristics to the Cu-terminated interface, reinforcing its role as the most promising candidate to enhance conductance. Note that to verify that the spikes observed in the 2D conductance maps in Figures 6(a) and 6(c) are not numerical artifacts, we recalculated the conductance using a denser k-mesh (from 50 $\times$ 50 to 100 $\times$ 100). The results show that the spikes persist, indicating that they are intrinsic features of the electronic structure at the Fermi level, as shown in Figure S3.

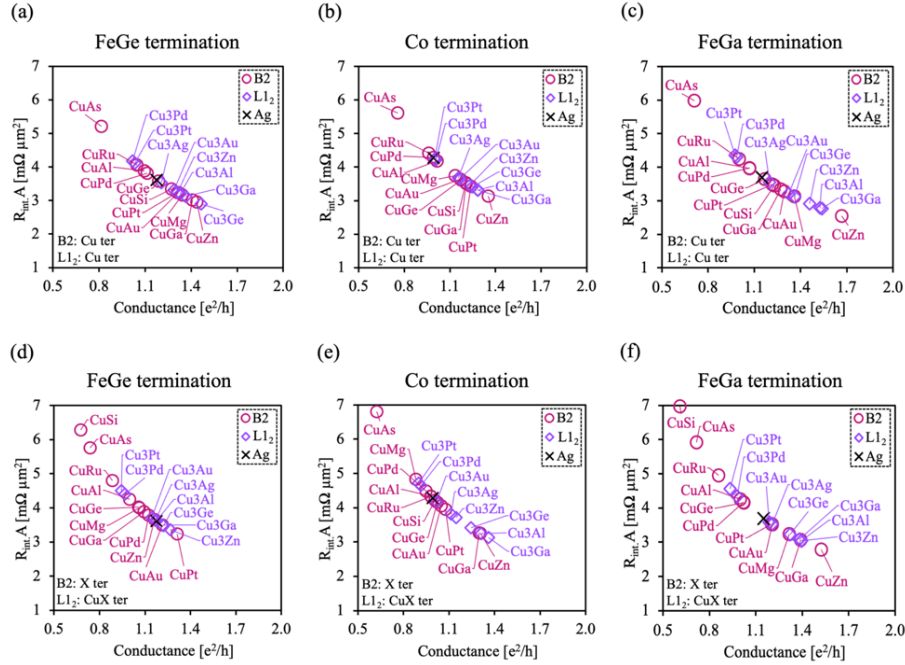


Figure. 5 Majority-spin resistance area product ($R_{int}.A$) as a function of majority-spin conductance for various Cu- X spacers and Ag spacers, using the CFGG electrode material. (a) Represents Cu termination for the Cu- X spacer with B2-type and $L1_2$ -type structure, along with each electrode termination: FeGe, Co, and FeGa, respectively. (b) Represents X and Cu X terminations for the Cu- X with B2-type and $L1_2$ -type structure, respectively, along with each electrode termination: FeGe, Co, and FeGa, respectively.

The difference in $R_{int}.A$ values can be attributed to variations in interface distance between FeGa-Cu and FeGa-Zn. Since the atomic radius of Cu (1.45 Å) is slightly larger than that of Zn (1.42 Å), the interface distance of FeGa-Cu (1.54 Å) is significantly smaller than that of FeGa-Zn (1.83 Å) and is closer to Cu's atomic radius. As a result, the overlap of the wave function of the FeGa-Cu interface is greater than that of the FeGa-Zn interface, leading to better band matching, and, consequently, lower interface resistance.

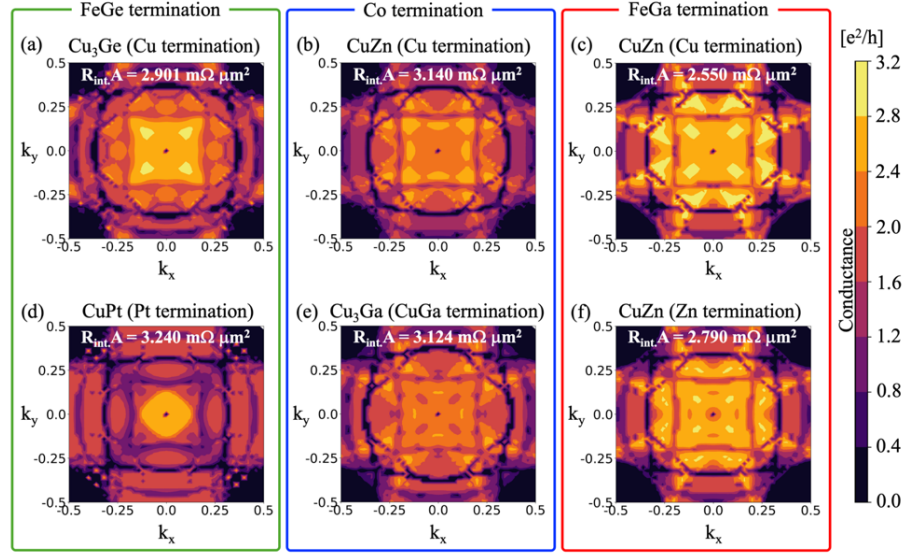


Figure. 6 In-plane wave vector $k_{\parallel} = (k_x, k_y)$ dependence of majority-spin conductance at the Fermi level in the parallel magnetization for CFGG/Cu- X /CFGG(001) at the lowest $R_{\text{int}}A$ value for each interfacial termination, as related to Figure. 5. (a) CFGG/Cu₃Ge/CFGG(001) at Cu termination, (b) and (c) CFGG/CuZn/CFGG(001) at Cu termination for FeGe, Co, and FeGa terminations, respectively. (d) CFGG/CuPt/CFGG(001) at Pt termination, (e) CFGG/Cu₃Ga/CFGG(001) at CuGa termination, and (f) CFGG/CuZn/CFGG(001) at Zn termination for FeGe, Co, and FeGa terminations, respectively.

The optimal CuZn spacer for the CFGG electrode was compared to the conventional Ag spacer. To further investigate their differences in the FeGa termination, the 2D $k_{\parallel}$ dependence of conductance in the BZ was analyzed for CFGG/Ag/CFGG(001) and CFGG/CuZn/CFGG(001) at the Fermi level, as shown in Figure. 7(a) and (b), respectively. It is found that there are significant differences in highly conductive regions and conductance distributions. For the Ag spacer, conductance is relatively small and the highly conductive region is confined near the BZ edge. In contrast, for the CuZn spacer, the conductance is more widely distributed, especially around the Γ point, which is a feature absent in the Ag spacer. Notably, the primary contribution of the conductance originates from the highly conductive region around the Γ point.

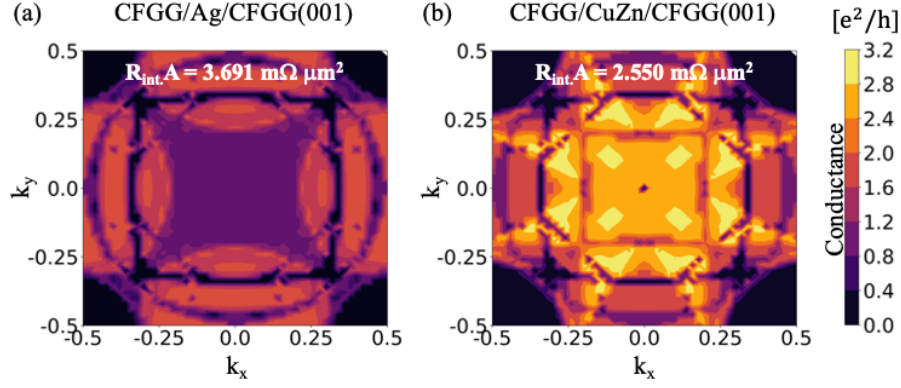


Figure. 7 In-plane wave vector $k_{\parallel} = (k_x, k_y)$ dependence of majority-spin conductance at the Fermi level in the parallel magnetization for (a) CFGG/Ag/CFGG(001) junction with FeGa termination and (b) CFGG/CuZn/CFGG(001) junction with FeGa and Cu terminations.

To better understand the orbital contribution to the lower $R_{\text{int}}A$ value and higher conductance, we analyzed the Fermi surfaces of CFGG electrode and Ag and CuZn spacers, which were decomposed into s , p , and d orbital components to determine their orbital contributions to the conductance as shown in Figure. 8. In the Ag spacer, hybridization occurs between the d -states of the CFGG electrode and the p -states of Ag at the BZ boundary. On the other hand, in the CuZn spacer, hybridization occurs between the d -states of CFGG and the p -states of CuZn in the diagonal region around the Γ -point of the BZ. This diagonal orbital hybridization in $k_{\parallel}$ opens the conductive channels across a wider BZ region, effectively reducing $R_{\text{int}}A$ compared to the Ag spacer.

In addition, hybridization between the p - and d -states of the CFGG electrode and the s -states of CuZn occurs around the Γ point. The spherical s -orbitals of CuZn effectively hybridize with the p - and d -orbitals of CFGG, creating highly delocalized states perpendicular to the plane. This hybridization at Γ further enhances conductance across the BZ, reducing the $R_{\text{int}}A$ in CuZn compared to Ag. Furthermore, there is the primary contribution of s -states around Γ originates from the Cu atom of CuZn, as shown in the left panel of Figure. 8. This suggests that Cu-based alloys will play a significant role in enhancing conductance in the parallel magnetization. Therefore, the improved Fermi surface matching and enhanced spin-dependent conductance

observed with Cu–*X* alloy spacers originate from their favorable electronic structure and atomic arrangement. Cu–*X* alloys exhibit *d*–*sp* hybridized states at the Fermi level that align more effectively with the spin-polarized states of CFGG, promoting stronger $k_{\parallel}$ -resolved conductance. This results in enhanced majority-spin conductance and interface spin asymmetry compared to the Ag spacer.

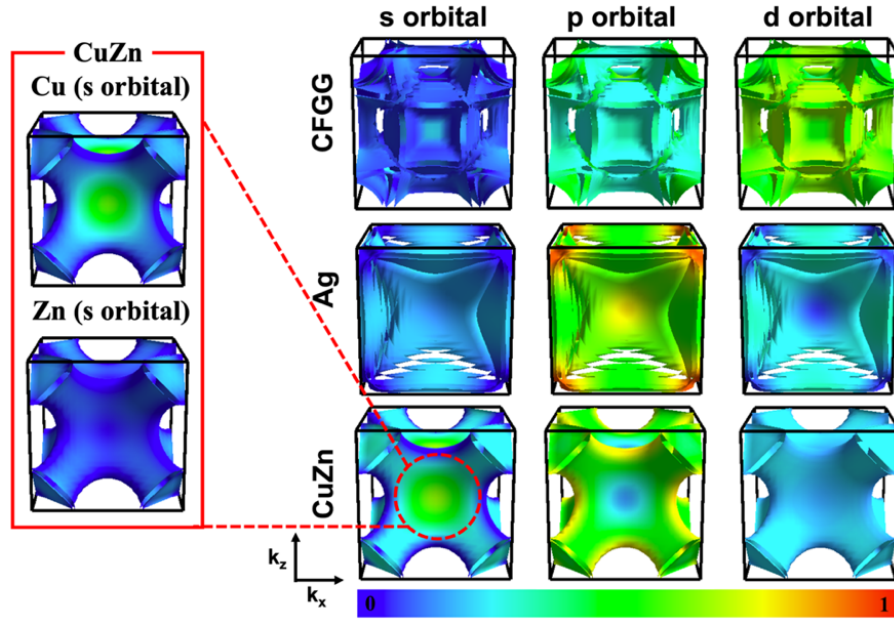


Figure. 8 Fermi surface separated into *s*, *p*, and *d* orbital compositions, where values of zero and one represent the ratio of each orbital contribution, for the tetragonal unit cell of L2₁-Co₂FeGa_{0.5}Ge_{0.5}, fcc-Ag, and B2-CuZn, visualized by FermiSurfer[71]. The *s* orbital of CuZn was further analyzed, separating the Cu-*s* orbital and Zn-*s* orbital. Around the Γ point ($k_x = k_y = k_z = 0$), the dominant *s* orbital contribution originates from the Cu-*s* orbital.

In addition, to understand the interfacial bonding between the CuZn spacer and the CFGG electrode, we analyzed the projected density of states (PDOS) for atoms in the bulk and interfacial regions of the CFGG/CuZn/CFGG(001) junction with FeGa- and Cu-terminated interfaces as shown in Figure S4 of the Supplementary Material. The PDOS confirms the preservation of CFGG's half-metallicity and reveals orbital hybridization between Fe *d*/Ga *p* states and Cu *sp*

states at the interface, indicating the formation of interfacial chemical bonds that enhance the system's electronic and structural stability.

3.3.2 Thermal fluctuation effects on interfacial spin moment stability

To evaluate the thermal stabilities of the spin moment at CFGG/CuZn interfaces, we analyzed the stiffness of the inter-atomic layer exchange interaction. The interface exchange stiffness constant (B) is determined from the increase in total energy when the interfacial spin moment is tilted with respect to the adjacent spin moment. A higher B value indicates stronger exchange interactions, suggesting that interfacial spin moments are more stable and less sensitive to thermal fluctuations. The estimated inter-atomic layer exchange stiffness constant for each interfacial termination at CFGG/Ag(001) and CFGG/CuZn(001) interfaces are summarized in Table III. As depicted in Figure. 9, the increase in total energy as a function of the tilting angle (θ) follows a quadratic trend ($\sim \theta^2$) up to $\theta = 15^\circ$. This consistency indicates that the error in estimating B using $E(\theta) = B(1 - \cos\theta)$ is negligible for the present system[72].

Table III Exchange stiffness constant B [meV/uca] fitted to the increase in energies, $E(\theta) = B(1 - \cos \theta)$, due to the noncollinearity of local spin moments for CFGG/Ag(001) and CFGG/CuZn(001) interfaces. The unit-cell area (uca) is approximately $33 \text{ \AA}^2$. The m_{spin} is the interfacial spin moment of Fe and Co for Fe(Ge/Ga) and Co termination, respectively.

B (meV/uca)	Interfacial termination of CFGG electrodes		
	FeGe (m_{spin} (μ_B))	Co (m_{spin} (μ_B))	FeGa (m_{spin} (μ_B))
CFGG/Ag(001) (Ref.[61])	134 (3.003)	115 (1.256)	129 (2.989)
CFGG/CuZn(001): Cu termination	115 (2.896)	124 (1.222)	106 (2.906)
CFGG/CuZn/(001): Zn termination	89 (2.778)	108 (1.034)	87 (2.756)

Overall, the CFGG/Ag(001) interface exhibits a larger exchange stiffness constant than the CFGG/CuZn(001) interface, as shown in Table III. This difference results from larger local spin moments for Fe and Co at the CFGG/Ag interface than at the CFGG/CuZn interface, as also listed in Table III. The larger spin moments at the CFGG/Ag interface enhance exchange interactions, thereby improving the stability of the magnetic configuration. However, an exception occurs with

the Co-terminated electrode and Cu-terminated spacer, where the CFGG/CuZn(001) interface exhibits a larger B value than its CFGG/Ag(001) interface, and the highest B value among all CuZn spacer terminations, as shown in Figure. 9. This exception can be attributed to the favorable interface bonding since the interface distance at the CFGG/CuZn interface (1.55 Å) is closer to the bulk CFGG interlayer distance (1.44 Å) than the CFGG/Ag interface (1.91 Å). This shorter interface distance can enhance exchange stiffness in certain configurations, particularly at the Co termination. For the most stable FeGa electrode termination with a Cu-terminated CuZn spacer, the CFGG/Ag(001) interface achieves a B value of 129 meV/uca, which is significantly larger than the 106 meV/uca observed for the CFGG/CuZn(001) interface. This higher B value suggests that the spin moment at the CFGG/Ag(001) interface is more stable against thermal fluctuations. Note that when comparing the exchange stiffness constant of CPP-GMR systems such as CFGG/Ag or CFGG/CuZn with that of the CMFS/MgO interface (typical of magnetic tunnel junction (MTJ) systems), where the electrode is half-metallic, the B value for CMFS/MgO is approximately two to three times higher—626 meV/uca for FeSi termination, 209 meV/uca for Co termination, and 223 meV/uca for MnSi termination—than those for the CFGG/Ag or CFGG/CuZn interfaces. This enhancement is attributed to the weaker hybridization between oxygen and Fe or Mn orbitals at the MgO interface compared to the stronger hybridization in metallic interfaces, resulting in more localized Fe and Mn spin densities and an increase in the exchange stiffness.

Consequently, the CFGG/Ag(001) interface is more optimal than the CFGG/CuZn(001) for suppressing thermal fluctuation of interface spin-moments. This suppresses the spin-flip scattering at the interface and reduces the MR ratio at finite temperatures. However, the majority-spin interface resistance-area product ($R_{\text{int}}A$) is significantly lower for the CuZn spacer compared to the Ag spacer (as discussed in Section 3.3.1). These results suggest that further improvements in CPP-GMR device performance can be achieved by controlling the multilayer interface structure, as explored in the next section.

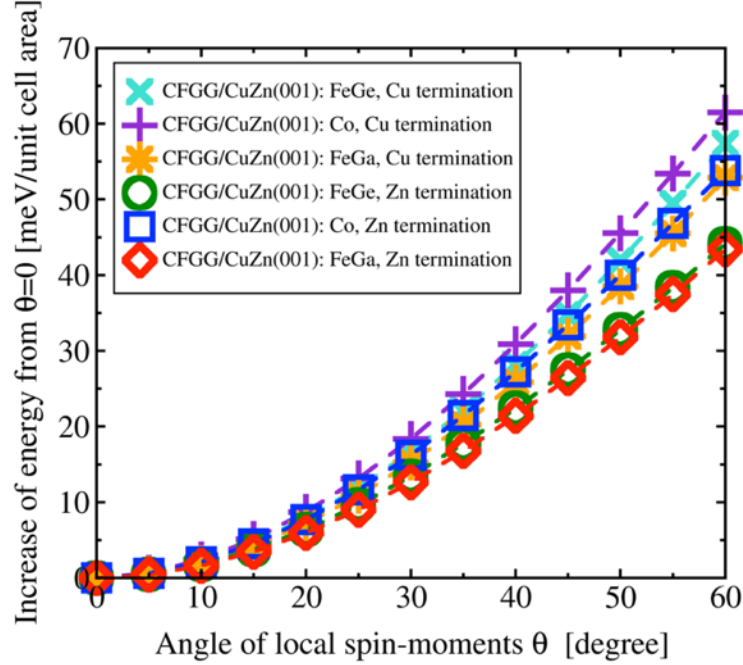


Figure. 9 Increase in the total energy relative to the collinear-spin system, $E(\theta)$, as a function of the angle of local spin moments, θ , for various interfacial terminations of CFGG/CuZn(001) junctions.

3.4. Beyond CuZn spacer material

We propose a new multilayer structure for CPP-GMR CFGG/Ag/CuZn/Ag/CFGG(001) with FeGa termination to maximize the advantages of the lattice matching, the thermal stability of the interface structure and the exchange stiffness of the interface spin-moment, and the Fermi surface matching.

- (1) Ag spacer at the CFGG interface: Connecting the Ag spacer to the CFGG electrode provides two important advantages. First, the lattice mismatch between the Ag spacer and the CFGG electrode is only 1.691%, compared to 3.405% for the CuZn spacer. This relaxes the lattice distortion in CuZn spacer, making the Ag/CuZn interface more structurally compatible with the CFGG electrode. Second, the CFGG/Ag interface exhibits larger exchange stiffness constant than the CFGG/CuZn interface, enhancing the stability of the spin moment against thermal fluctuation.
- (2) CuZn spacer at the Ag interface boundary: Making the Ag/CuZn/Ag multilayer spacer also offers two advantages. First, the lattice mismatch between Ag and CuZn is just

1.657%, which is significantly smaller than the lattice mismatch of 3.405% mismatch at the CFGG/CuZn interface. Second, the CuZn spacer has a low $R_{\text{int},A}$ value, which is about 31% lower than that of the Ag spacer. This will substantially enhance the interface spin asymmetry and the MR ratio.

Consequently, the CFGG/Ag/CuZn/Ag/CFGG(001) junction would be able to bring out the distinct advantages of each spacer material, achieving structural compatibility, spin stiffness, and high MR. This multilayer design opens new avenues for engineering advanced spacer materials to improve spintronic device performance.

To confirm the physical properties of CFGG/Ag/CuZn/Ag/CFGG(001) junction, we performed the DFT calculations of the formation energy and conductance for the junction inserting one monolayer of Ag spacer between CFGG and CuZn. Figure. 10(a) shows a comparison of formation energy (upper panel) and interface distance (lower panel) for the CFGG/spacer/CFGG(001) junction with FeGa termination. The CFGG/Ag/CuZn/Ag/CFGG(001) junction has a higher structural stability than the CFGG/Ag/CFGG(001) junction. This is indicated by the fact that both Ag-Cu (-0.113 J/m^2) and Ag-Zn (-0.096 J/m^2) terminations have lower formation energies than those of single Ag spacer, which are slightly higher than CFGG/CuZn/CFGG(001) junction (-0.144 J/m^2). The CFGG/spacer interface distance is around $1.50 \text{ \AA}$ to $1.75 \text{ \AA}$, while the single Ag spacer has a larger distance ($\sim 2.00 \text{ \AA}$). The majority-spin conductance at the Fermi level for parallel magnetization is presented in Figure. 10(b). The top panels show $k_{\parallel} = (k_x, k_y)$ dependence (heatmaps A and B) for Ag-Cu and Ag-Zn terminations, respectively. The bottom panel compares conductance values across different spacer configurations. Overall, the proposed CFGG/Ag/CuZn/Ag/CFGG(001) junction exhibits higher conductance than the CFGG/Ag/CFGG(001) junction, indicating improved Fermi surface matching compared to a single Ag spacer. In the proposed configurations, the Ag-Zn termination achieves higher conductance than the Ag-Cu termination (Figure. 10(b), lower panel), especially around the Γ point, as evidenced by the 2D in-plane wave vector ($k_{\parallel}$) dependence in Figure. 10(b) (upper panel).

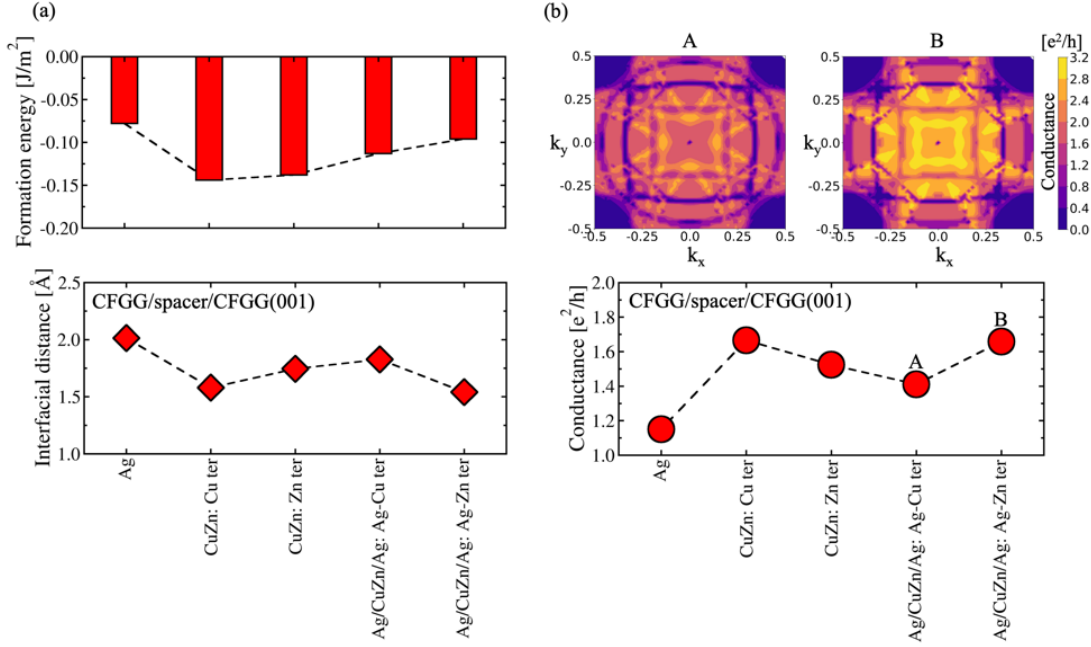


Figure. 10 (a) Formation energy (top) and interfacial distance (bottom) for the CFGG/spacer/CFGG(001) junction with FeGa electrode termination. Spacer materials include Ag, CuZn (with Cu- and Zn-terminated configurations), and Ag/CuZn/Ag (with Ag-Cu and Ag-Zn terminations). (b) Majority-spin conductance at the Fermi level for parallel magnetization: top panels show $k_{||} = (k_x, k_y)$ dependence (heatmaps A and B) for Ag-Cu and Ag-Zn terminations, respectively; the bottom plot compares conductance values across spacer configurations.

Furthermore, we estimated the exchange stiffness constant (B) for the CFGG/Ag/CuZn(001) interfaces with Ag-Cu and Ag-Zn terminations, which are about 106 meV/uca for both terminations. These results indicate improved stiffness of interface spin moment against the thermal fluctuation for the Ag-Zn termination compared to the CFGG/CuZn(001) interface at Zn termination (87 meV/uca). However, for the Ag-Cu termination, the stiffness constant hardly changes compared to the CFGG/CuZn(001) interface at Cu termination (106 meV/uca).

As a result, the proposed CFGG/Ag/CuZn/Ag/CFGG(001) junction with Ag-Zn termination exhibits large conductance comparable to that of the CFGG/CuZn/CFGG(001) junction with the Cu termination and increased interface exchange stiffness constant (from 87 meV/uca to 106 meV/uca). This finding suggests that the new multilayer design can improve

effective transport properties while enhancing spin moment stability against thermal fluctuations. Despite the structural complexity of the proposed junction, our design is inspired by experimentally realized CPP-GMR multilayers that incorporate ultrathin alloy and spacer layers to engineer interfacial properties. Similar strategies, such as the insertion of Ni layers[38] or NiAl layers[42] to improve MR performance in CFGG-based junctions, have been experimentally demonstrated. Therefore, the present findings offer valuable theoretical insights that can guide future fabrication of optimized multilayer spintronic devices using quaternary Heusler alloys and tailored alloy spacers.

4. Conclusions

This study evaluates Cu-*X* spacers as alternatives to Ag spacers for enhancing spin-dependent transport properties in CFGG-based CPP-GMR devices. We found that Cu-*X* spacers improve Fermi surface matching compared to Ag, for CuZn, CuMg, CuAu, CuGa, and CuPt in B2 structures, and for Cu₃Ga, Cu₃Al, Cu₃Zn, Cu₃Ge, Cu₃Au, and Cu₃Ag in L1₂ structures. All these spacer materials contribute to the enhancement of interface spin asymmetry (γ). Among these, the CuZn spacer is identified as the most optimal spacer for CFGG electrodes, achieving ~45% higher conductance and ~31% lower $R_{\text{int}}A$ compared to the Ag spacer.

For the optimal CuZn spacer, we clarified the origin of improved Fermi surface matching with CFGG, where hybridization between the *p*- and *d*-states of the CFGG electrode and the *s*-states of the CuZn spacer produces highly delocalized states, enhancing conductance. This coupling is mainly driven by the *s*-states of Cu, emphasizing the critical role of Cu-based alloys in improving electron transport.

Furthermore, we estimated the exchange stiffness constant of CFGG/CuZn(001) and CFGG/Ag(001), an important parameter for suppressing spin fluctuation and spin-flip scattering at finite temperatures. The FeGa-Cu termination exhibits a smaller exchange stiffness constant (106 meV/uca for CFGG/CuZn(001)) compared to 129 meV/uca for CFGG/Ag(001), indicating that spin moments in CFGG/CuZn(001) are more susceptible to fluctuations than in CFGG/Ag(001).

To exploit the advantages of both CuZn and Ag materials, we proposed a multilayer CFGG/Ag/CuZn/Ag/CFGG(001) junction, incorporating Ag spacers to enhance the exchange stiffness constant and CuZn spacers to improve Fermi surface matching. Our findings confirm that the CFGG/Ag/CuZn/Ag/CFGG(001) junction exhibits the highest conductance at the Ag-Zn termination, comparable to that of CFGG/CuZn/CFGG(001) junctions with Cu termination. This multilayer configuration addresses the concern of lattice mismatch by using thin Ag interlayers to accommodate strain and maintain coherent interfaces, while preserving the high transmission characteristics of CuZn. The Ag layers act as structural buffers, improving interface quality and potentially reducing defect formation. Therefore, this design not only enhances spin-dependent transport but also offers a practical route toward experimental realization with improved structural and interfacial spin moment stability.

These findings highlight the potential of Cu-*X* spacers, particularly in B2 structures, for advancing spin-dependent transport properties and optimizing the performance of spintronic devices. These results also emphasize the critical role of interfacial atomic structure, termination, and electronic band matching in determining spin polarization and conductance. By revealing how subtle variations in alloy composition and interface symmetry affect the MR ratio and exchange stiffness, this work contributes to a deeper understanding of structure–property relationships in magnetic multilayers. These insights can guide future experimental efforts in selecting and engineering spacer materials for high-efficiency, cost-effective CPP-GMR and other spintronic applications.

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Conflict of Interest

The authors have no conflicts to disclose.

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