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Phys. Rev. B **110**, L220408 — Published 18 December 2024

DOI: [10.1103/PhysRevB.110.L220408](https://doi.org/10.1103/PhysRevB.110.L220408)

Giant Rashba Spin-orbit Coupling on Light-Element Polar Superlattices

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(Dated: November 19, 2024)

We present a new strategy to amplify the spin-orbit coupling (SOC) effect in a simple metal-based polar superlattices with light elements to create a giant Rashba SOC up to $\alpha_R \approx 3.80$ eVÅ with negative sign. First-principles calculations demonstrate that the giant α_R originates from optimizing the internal electric field, which comes from the polarity, charge transfer, and superlattice systems. This strategy provides an effective way to design a next-generation spintronics device without the use of heavy elements.

The Rashba effect is a type of spin-orbit (SO) splitting that occurs on surfaces and interfaces where the inversion symmetry is broken[1, 2]. This effect can lead to a lifting of the spin degeneracy, which in turn can give rise to several interesting quantum phenomena and potential applications, such as the spin Hall effect[3], Majorana fermions in solids[4], and the spin field-effect transistor[5]. The effective Rashba Hamiltonian can be written as[1]

$$H_R = \alpha_R(\boldsymbol{\sigma} \times \mathbf{p}) \cdot \hat{z}, \quad (1)$$

where α_R is the Rashba coupling, $\boldsymbol{\sigma}$ is the Pauli matrix-vector, $\mathbf{p}$ is the momentum, and $\hat{z}$ is the built-in electric field unit vector that is perpendicular to the surface or interface and comes from a perpendicular potential gradient due to the structural asymmetry. The strength of the Rashba spin-orbit coupling (SOC) that induces spin splitting of the electronic bands is defined as $|\alpha_R| = 2E_R/k_R$, where E_R and k_R are the Rashba energy and momentum offsets, respectively. Initially, the Rashba effect was found on semiconductor heterointerfaces such as InAlAs/InGaAs[6], which exhibits $|\alpha_R| \approx 0.07$ eVÅ and the surface of Au(111) and Ag(111)[7–9], which exhibits $|\alpha_R| \approx 0.33$ eVÅ and 0.03 eVÅ, respectively.

The two main contributions to creating a large Rashba SOC are a strong atomic SOC interaction (λ) and a potential gradient along the surface/interface normal (E_z). The development of large Rashba-type SO splittings in materials was carried out by improving the strengths of λ and E_z . The strength of λ can be improved straightforwardly by introducing heavy elements, e.g., Bi, Sb, Te, Hg, Pt, and Pb[10–13]. Therefore, the SO splitting can be increased to $|\alpha_R| \approx 3.05$ eVÅ, which was found at the Ag(111)/Bi interface[14]. On the other hand, SO splitting can be further enhanced by increasing the strength of E_z by introducing a polar surface or inter-

face. An example is BeTeI for the polar surface and the Pb/Al₂O₃(0001) for the polar interface, which exhibits $|\alpha_R| \approx 4.30$ eVÅ and 4.14 eVÅ, respectively[15, 16].

Nevertheless, compounds containing heavy atoms tend to have weaker chemical bonds, where they are more likely to have native defects [17], leading to poorer stability for performing exotic functionalities. An example can be found in the topological insulator Bi₂Te₃, which is a conductor due to the heavy n-type self-doping [18, 19]. Furthermore, almost all heavy elements are rare-earth metals, which have limited availability and high production cost. Unfortunately, up until now, there has been no non-heavy-element interface or surface that exhibits giant spin splitting on par with a heavy-element-based interface or surface. A new strategy needs to be proposed to utilize the giant Rashba SOC on light element-based materials. A recent study reported that the surface-oxidized Cu film demonstrates a spin current generation as large as that of Pt by means of spin-vorticity coupling [20]. However, the possible appearance of Rashba SO interaction has not yet been explored. Transition-metal nitride or oxide, such as Cu₃N or Cu₂O, creates a polar layer structure. Utilizing this polar layer to create a huge internal electric field and breaking the inversion symmetry by considering an interface of the transition metal and its oxide or nitride, a possible large Rashba SOC at the interface might be expected.

In this Letter, we report on a new strategy to utilize a light element and a base metal to exhibit giant Rashba-based SOC by maximizing all possible internal electric fields E_z created in the electronic interface structure. The general idea is shown in FIG. 1. To obtain an optimal internal electric field E_z , a semiconductor composed of a stacking of polar layers in the z -axis direction is first considered. In a stacked polar-layer semiconductor, the

charge carrier experiences an internal electric field because of the arrangement of the polar structures of the atoms. A cumulative electric field is expected from the combined effect of the internal electric dipoles created by each polar layer. When the periodicity of the stacked polar layer is broken, creating an asymmetric surface or interface, an effective electric field will be created, as shown in FIG. 1(a).

Second, the metal-semiconductor interface is considered to create an electric field at the interface, as a result of charge transfer. When a metal and a semiconductor come into contact, there is a Fermi level difference that gives rise to the charge-transfer mechanism that creates an electric field at the interface. The direction of the built-in electric field is parallel to the direction of electron transfer, as shown in FIG. 1(b). When the concept of enhancing the strength of E_z is combined through polarity and potential difference at the metal/semiconductor interface, a huge built-in electric field is expected at the interface of the polar semiconductor/metal shown in FIG. 1(c).

The last enhancement of the internal electric field is

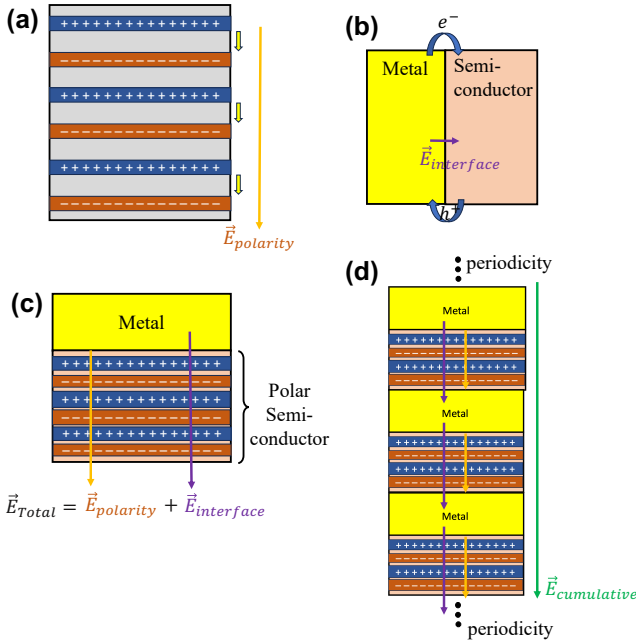


FIG. 1. The primary idea for enhancing the Rashba SOC in light-element based materials by optimizing internal electric fields involves following approaches: (a) utilizing polar layered materials to generate an internal electric field through electric dipoles ($\vec{E}_{polarity}$), (b) introducing a metal-semiconductor interface to create an internal electric field at the interface through charge transfer mechanisms ($\vec{E}_{interface}$), (c) combining the aforementioned contributions by creating a metal-polar semiconductor interface, and (d) implementing a superlattice system consisting of metal-polar semiconductor with asymmetric interfaces to generate a cumulative internal electric field across the interfaces of superlattice.

considered by introducing a metal/polar semiconductor superlattice system. Accumulating the internal electric field created from two previously explained concepts of the metal/polar semiconductor interface by repeating the interface in one system, a sum of huge internal electric field will be created, as shown in FIG. 1(d). A giant Rashba SOC is expected to be observed on the proposed superlattice system without introducing a heavy element or rare earth metal.

To implement our proposed concept, Cu/Cu₃N(001) superlattices as shown in FIG. 2 is introduced. This superlattice system consists of Cu slabs, which are a common metal, and Cu₃N slabs, which are a polar semiconductor having alternating layer polarity +1 in the Cu layer and -1 in the Cu-Cu-N (Cu₂N) layer, shown in FIG. 2(d). To validate our proposal, density functional theory calculations were performed on the proposed Cu/Cu₃N(001) superlattices (See Supplementary Materials Section I for details of the computational methods [21]).

When the Cu₃N slab interfacing with Cu slab in the superlattice system, to maintain the stoichiometry of Cu₃N slab, two type of interfacing was created. The interface formed between the Cu-layer termination of the Cu₃N slab and the Cu-layer of the Cu metal slab, as well as the interface formed between the Cu₂N-layer termination of the Cu₃N slab and the Cu-layer of the Cu metal slab. The structure at the interface between the Cu layer of Cu slabs and Cu₂N layer of Cu₃N slabs is most energetically stable when a strong hybridization is created, which is called type 3 in FIG. S1 [21]. From Table S1, it was shown that the type 3 interface has the lowest total energy. On the other hand, the second most energetically stable interface, which has a strong hybridization

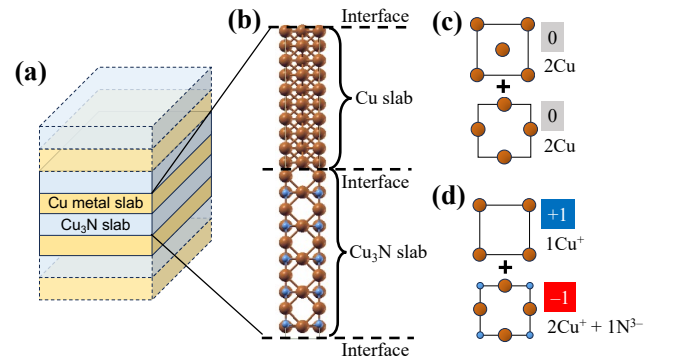


FIG. 2. (a) The superlattice structure Cu/Cu₃N(001) consists of Cu metal and Cu₃N slabs. (b) The atomic structure of Cu/Cu₃N(001) interface consists of 5-layer Cu and 5-layer Cu₃N with periodic boundary conditions on the x, y, and z-axes. The brown and blue color atoms correspond to Cu and N atoms, respectively. (c) and (d) corresponds to the atomic configuration on each layer and its polarity for Cu slab and Cu₃N slab, respectively.

between the Cu layer of Cu slabs and the Cu layer of Cu₃N slabs was shown in the type 2 interface configuration. Thus, the configuration of alternating type 3 and type 2 interfaces in infinite-layer superlattice was considered to be the most stable structure of Cu/Cu₃N(001) superlattice.

The band structure calculation for the proposed superlattice system revealed significant Rashba SOC splitting just below the Fermi energy near the M high-symmetry point (see FIG. S2 in Supplementary Materials Section II for the full-path band structure [21]). The Rashba SOC splitting shows $|\alpha_R| \approx 3.80$ eVÅ, with $k_R = 0.01$ Å⁻¹ and $E_R = 19$ meV, as shown in FIG. 3(b). This giant Rashba SOC is comparable to that found in rare-earth, heavy-element metal-based materials. Moreover, the band structure displays a $\mathbf{k}$ -linear Rashba SO splitting, which suggests that the effective magnetic field induced in momentum space is effectively "locked" perpendicular to the momentum. This leads to more precise control over the spin direction. This can be observed by examining the spin projection values of $\langle S_x \rangle$, $\langle S_y \rangle$, and $\langle S_z \rangle$. When considering the high-symmetry point path of $X \rightarrow M \rightarrow X$, only the spin component perpendicular to the momentum contributes to the projection. For example, along the k_x axis at the high-symmetry point M, the Rashba bands do not exhibit $\langle S_x \rangle$ and $\langle S_z \rangle$, while only the $\langle S_y \rangle$ projection is observed in the band structure (see FIG. 3(c)). Furthermore, the absence of $\langle S_z \rangle$ in the Rashba bands indicates that the spin polarization primarily lies in the plane. In addition, FIG. 3(d) shows the spin texture calculation at Fermi Energy, which confirmed the ideal spin precision on the xy plane of the momentum space. The spin texture of the system also shows a Rashba-type profile of spin precision with a negative sign of $|\alpha_R|$. Thus, our proposed Cu/Cu₃N(001) superlattice successfully creates a giant Rashba SOC.

The origin of the giant Rashba SOC was further investigated by analyzing the nature of the internal electric field that operates within the superlattice system. The layer responsible for the giant Rashba SOC was first identified. From the k -resolved local density of states of each layer shown in FIGs. S2 and S3, it is evident that the giant Rashba SOC primarily originates from the Cu₃N slab (see Supplementary Materials Section II for details [21]). An optimized E_z across the interface is expected within the Cu₃N slab.

The built-in electric field primarily originates from the polarity of the polar layers in the Cu₃N slab ($\vec{E}_{\text{polarity}}$). Table S4 presents the Bader charge analysis of the Cu₃N slab (see Supplementary Materials Section III for details [21]), and FIG. 4(a) shows the partial charge per layer in the Cu₃N slab. This analysis reveals an alternating polar layer pattern, with positive charges (+) from the Cu layer and negative charges (−) from the Cu₂N layer throughout the Cu₃N slab. The electric field $\vec{E}_{\text{polarity}}$ is generated across the Cu₃N slab, originating from each

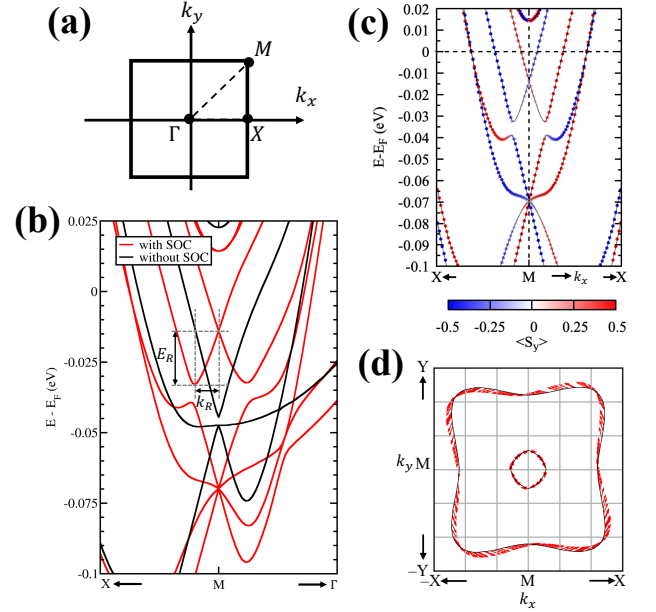


FIG. 3. (a) The 2D Brillouin zone of the Cu/Cu₃N superlattice and its high-symmetry points. (b) The band structure of Cu/Cu₃N superlattice with and without SOC interaction in the vicinity of the Fermi energy around the high-symmetry point M. Energy splitting (E_R) and momentum offset (k_R) of Rashba bands give a giant Rashba SOC with $|\alpha_R| \approx 3.8$ eVÅ. (c) Spinor projection of $\langle S_y \rangle$ in the k_x direction of the $X \rightarrow M \rightarrow X$ path. (d) Spin texture of Rashba splitting at Fermi energy that shows a negative sign of $|\alpha_R|$.

polar layer. Summing the contributions from all layers results in a net internal electric field in the $-z$ direction (from interface type 3 to type 2), as shown in FIG. 4(b).

The internal electric field arising solely from polarity in the Cu₃N thin film generates Rashba SOC band splitting. In the Cu₃N thin film, the internal electric field is nonzero due to inversion symmetry breaking at the surface, which creates a charge imbalance (see Fig. S3 in Supplementary Materials Section III for details [21]). This nonzero polarization in the Cu₃N thin film leads to significant Rashba SOC band splitting. This effect is demonstrated in FIG. 5(b), where Rashba SOC bands are observed near the M point. The Rashba band from X to M exhibits a typical Rashba profile with $|\alpha_R| \approx 1.42$ eVÅ. However, from X to Γ , the Rashba band appears twisted or intertwined. This effect may stem from variations in potential asymmetry along different crystallographic directions, causing variations in Rashba splitting along distinct high-symmetry paths. The anisotropic SOC can cause the spin-split branches to twist or intertwine, as observed from M to Γ .

Additionally, a built-in electric field is expected to arise from the metal-semiconductor interface. This field is created due to charge transfer at the Cu/Cu₃N interface, resulting from the difference in Fermi energy levels between

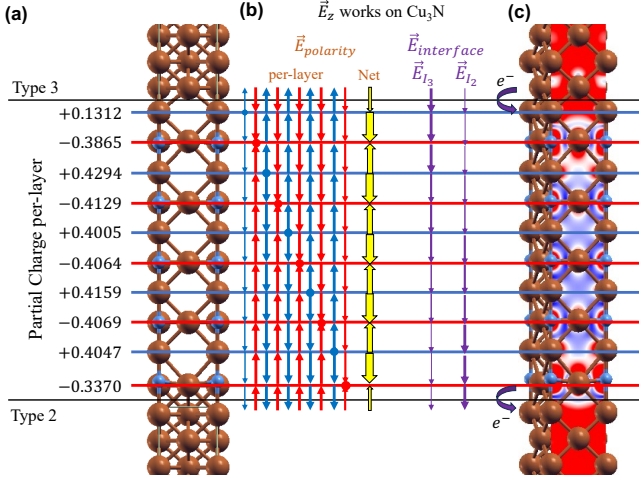


FIG. 4. (a) The partial charge corresponding to the polar charge of each layer of Cu₃N slab. (b) The internal electric field created across the Cu₃N slab, which consists of the electric field created from the polar charge ($\vec{E}_{polarity}$) and from the charge transfer at the interface ($\vec{E}_{interface}$). In $\vec{E}_{polarity}$, the electric field comes from each of the polar layers shown by a blue (red) color arrow, which originated from the positively (negatively) charged of Cu (Cu₂N) layer. The dots in $\vec{E}_{polarity}$ per layer represent the layer from which the electric field originated. The width of the arrow implies the electric field strength, which is less strong at the interface because of less polarity compared to the middle layer. The net electric field of polarity comes from the total contribution of the electric field from all layers. In $\vec{E}_{interface}$, the contribution comes from interface type 3 ($\vec{E}_{I_3}$) and type 2 ($\vec{E}_{I_2}$). (c) The integrated local density of states (ILDOS) from $E - E_F = -0.2$ eV to E_F . It shows the different charge transfer mechanisms between interface types 3 and 2. The red (blue) color background corresponds to the high (low) electron density.

the metallic Cu and the semiconductor Cu₃N. However, since Cu₃N is a polar material, the polarity of the layer at the interface plays a significant role in the charge transfer mechanism. When a positively (negatively) charged layer interfaces with the non-polar metal, the positively (negatively) charged layer tends to receive (donate) electrons, creating electron transfer from the non-polar metal (polar semiconductor) to the polar semiconductor (non-polar metal). This mechanism occurs because the positively (negatively) charged layer has a deficiency (excess) of electrons compared to its neutral state. Due to this deficiency (excess), the positively (negatively) charged layer readily accepts (donates) electrons to achieve a more stable, electron-filled state.

For the type 3 (type 2) interface, the Cu layer (Cu₂N layer) of Cu₃N has a positive (negative) polarity. As shown in FIG. 4(a), the partial positive (negative) charge of the Cu layer (Cu₂N layer) at the type 3 (type 2) interface is reduced due to electron transfer from the Cu layer of the Cu metal slab to the Cu layer of Cu₃N (from the Cu₂N layer of Cu₃N to the Cu layer of the Cu metal).

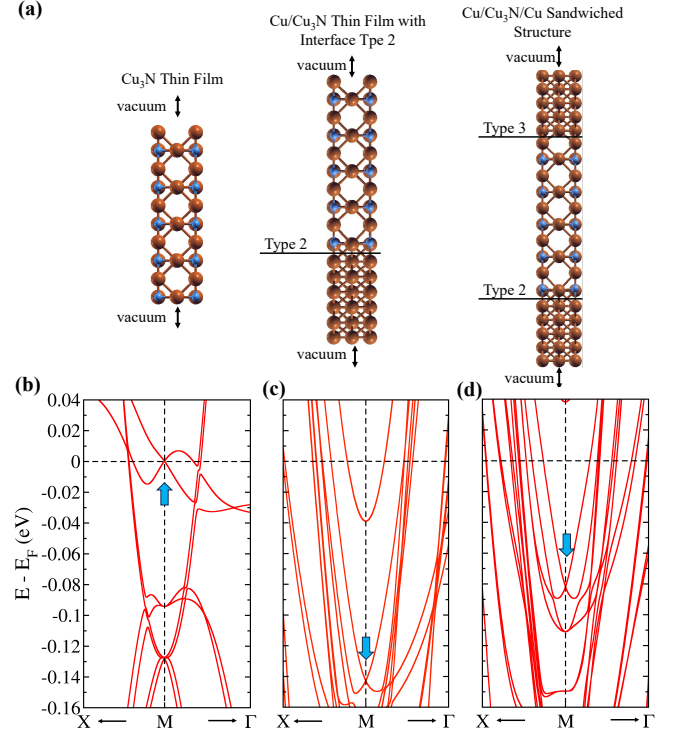


FIG. 5. (a) Crystal structures of the Cu₃N thin film, Cu₃N/Cu thin film with type 2 interface, and Cu/Cu₃N/Cu sandwiched structure containing both type 2 and type 3 interfaces. Band structures of (b) Cu₃N thin film, (c) Cu₃N/Cu thin film with type 2 interface, and (d) Cu/Cu₃N/Cu sandwiched structure near the Fermi energy around the high-symmetry M point. The Rashba bands in each system are indicated by blue arrows in the figure.

Simultaneously, since electrons transfer to the Cu layer of Cu₃N (Cu layer of the Cu metal), an internal electric field is created at the type 3 (type 2) interface, with direction from the Cu metal (Cu₃N slab) to the Cu₃N slab (Cu metal). Since the internal electric fields at interface types 3 and 2 are aligned, they create a constructive internal electric field across the Cu₃N slab, enhancing $|\alpha_R|$ in the Rashba SOC.

The constructive effect of the internal electric fields from both interfaces is evident when comparing the Rashba SOC splitting in the band structure shown in FIG. 5(c) and (d). In a thin film composed of Cu₃N and Cu slabs with only a type 2 interface, Rashba SOC splitting is observed with $|\alpha_R| \approx 1.38$ eVÅ, as shown in FIG. 5(c). When the type 3 interface is included, creating a Cu/Cu₃N/Cu sandwiched structure with both type 2 and type 3 interfaces, a Rashba SOC with $|\alpha_R| \approx 1.51$ eVÅ is observed, as shown in FIG. 5(d). The increase in Rashba SOC results from the constructive internal electric field generated by the type 3 and type 2 interfaces.

Additionally, interfacing the Cu₃N slab with the Cu metal layer can alter the local electric field at the interface. This alteration reduces the in-plane variation of the

electric field, resulting in a more uniform Rashba effect across different crystallographic directions, including the M- Γ path. This uniformity is evident when comparing the Rashba bands in the M- Γ path of Cu_3N thin film, $\text{Cu}/\text{Cu}_3\text{N}$ with type 2 interface, and the $\text{Cu}/\text{Cu}_3\text{N}/\text{Cu}$ sandwiched structure, as shown in FIG. 5. The twisted Rashba band in the Cu_3N thin film begins to unfold slightly in the $\text{Cu}/\text{Cu}_3\text{N}$ type 2 structure and crosses the Fermi energy, though it remains somewhat twisted near the Fermi level. When the $\text{Cu}/\text{Cu}_3\text{N}/\text{Cu}$ sandwiched structure is considered, the Rashba bands become consistent across the high-symmetry region around the M point. This consistency implies that interfacing the Cu_3N with the Cu metal creates a more homogeneous electric field, helping to maintain a well-defined Rashba band structure without twisting.

Although an enhancement in $|\alpha_R|$ is observed in the sandwiched structure compared to the Cu_3N thin film or the $\text{Cu}/\text{Cu}_3\text{N}$ type 2 structure, the $|\alpha_R|$ value in the sandwiched structure is still lower than that found in the superlattice system. This suggests that the infinite-layer superlattice structure further optimizes the internal electric field of the polar layers and the constructive internal electric field of the two interfaces by building a cumulative effect from multiple stacked $\text{Cu}/\text{Cu}_3\text{N}$ slabs. This optimization significantly enhances both $|\alpha_R|$ (from $\approx 1.51 \text{ eV\AA}$ to $\approx 3.8 \text{ eV\AA}$) and the Rashba energy splitting E_R (from $E_R \approx 0.0075 \text{ eV}$ to $\approx 0.0212 \text{ eV}$). Although an infinite-layer superlattice is considered, $|\alpha_R|$ saturates at approximately $3.8 \text{ eV\AA}$. This saturation occurs because the short-range nature of the internal electric field at the interfaces propagates through the Cu_3N slab, meaning that $|\alpha_R|$ depends on the thickness of the Cu_3N slab (see Supplementary Materials Section IV for a detailed quantitative comparison between Cu_3N slab thickness and $|\alpha_R|$ [21]).

The Supplementary Materials Section V demonstrated the importance of the polar interface by comparing the $|\alpha_R|$ between the $\text{Cu}/\text{Cu}_3\text{N}$ polar superlattice and a non-polar superlattices, such as Si/Al and Cu/Ru [21]. Furthermore, Supplementary Materials Section VI illustrated the significance of a charge transfer at the two types of interface in $\text{Cu}/\text{Cu}_3\text{N}$ polar superlattice in generating an optimum internal electric field by comparing the Rashba SOC in another polar superlattice, namely $\text{Cu}_2\text{O}/\text{Cu}$ [21]. This comparison shows that an efficient charge transfer mechanism at both interfaces is necessary to create a high internal electric field across the polar slabs for creating the giant Rashba SOC.

Finally, we summarize our work by referring to a comparison table on Rashba SOC for various types of materials in Table 1. Our proposed $\text{Cu}/\text{Cu}_3\text{N}$ polar superlattice shows giant Rashba SOC, comparable to the heavy-element rare-earth metal-based interface or surface. This giant Rashba SOC was created by optimizing all possible internal electric fields E_z created on Cu_3N slab in the su-

TABLE 1. The comparison of $|\alpha_R|$ in various materials.

Material	E_R (meV)	k_R ($\text{\AA}^{-1}$)	$ \alpha_R $ ($\text{eV\AA}$)	Ref.
Heavy-element doped non-polar surface:				
Bi/Ag(111)	200	0.13	3.05	[14]
Heavy-element Polar surface:				
BiTeI	108	0.05	4.30	[15]
Heavy-element monolayer on polar surface:				
Bi/ Al_2O_3	160	0.20	1.61	[16]
Pb/ Al_2O_3	266	0.13	4.14	[16]
Light-element polar interface superlattice:				
$\text{Cu}/\text{Cu}_3\text{N}$	19	0.01	3.8	This work

perlattice system. The optimal internal electric field E_z is obtained by combining three contributions, the electric field created from the electric dipole of polar layer Cu_3N slab ($\vec{E}_{\text{polarity}}$), the electric field from a charge transfer at $\text{Cu}/\text{Cu}_3\text{N}$ interface, where internal electric field at the interfaces types 2 and 3 exhibit a constructive effect ($\vec{E}_{\text{interface}}$), and the enhancement of the two aforementioned contributions by creating a cumulative electric field from an infinite-layer stacking superlattice system. Our study provides an effective way to achieve giant Rashba SOC by using simple metals (i.e., Cu).

Calculations were performed at the computer center of the RIKEN Hokusai Big Wave. JYY is supported by the Singapore National Research Foundation Investigatorship Program under Grant No. NRF NRFI08-2022-0009. BG is supported by the China National Natural Science Foundation (Grant No.12074378) and the Chinese Academy of Sciences (Grants No.YSBR-030, No.XDB28000000, and No.XDB33000000). KK is financially supported by the JSPS KAKENHI Grant numbers JP22K04864, JP21H01752, and JP19H00862. SM is financially supported by JST CREST (No.JPMJCR19J4, No.JPMJCR1874, and JPMJCR20C) and JSPS KAKENHI(No.20H01865 and No.24K00576) from MEXT Japan.

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