**Magneto-transport mechanism of individual nanostructures via
direct magnetoresistance measurement in situ SEM**

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Abstract

The accurate magnetoresistance measurement of individual nanostructures is essential and significant for not only enriching the fundamental knowledge of magneto-transport mechanism but also facilitating the designs of desired magnetic nanostructures for diverse technological applications. Herein, we report a deep investigation on the magneto-transport mechanism of single CoCu/Cu multilayered nanowire via direct magnetoresistance measurement by using our invented magneto-transport instrument *in-situ* scanning electron microscopy (SEM). Off-axis electron holography experiments combined with micromagnetic simulation prove that the CoCu layers in CoCu/Cu multilayered nanowires are formed a multi-domain structure, in which the alignment of magnetic moments is mainly determined by shape anisotropy. The MR of the single CoCu/Cu multilayered nanowire is measured to be only 1.14% when the varied external field is applied along nanowire length axis, which matches with the theoretical prediction of Granular Films model. Density functional theory (DFT) calculations further disclose that spin-dependent scattering at the interface between magnetic and nonmagnetic layers is responsible for the intrinsic magneto-transport mechanism.

1. Introduction

The trend of miniaturization in micro-electronics makes nanowires (NWs) more and more prominent. Magnetic multilayered nanowires (MNWs) are one group of the nanowire family, which own giant magnetoresistance (GMR) effect and are mainly grown by using template method with electrodeposition technique¹⁻³. Their magneto-transport behaviors are usually utilized for possible diverse nano-spintronic applications including magnetic memory, magnetic field sensor, logic devices, signal

processing device, current driven microwave oscillators⁴⁻⁵. The accurate characterization of individual NWs rather than groups is thus essential and critical for the functionality and reliability of the NW-component magnetoelectronics and magnetosensors. However, the most magnetoresistance (MR) measurements are performed on the array of multilayered nanowires which are still embedded into template channels due to technology limitations⁶⁻⁷, by which the MR signal of a single nanowire is then estimated by averaging. It is therefore not accurate and includes uncertain contact resistance of electrodes. There are also a limited work on the MR measurement of real single nanowire by making the electrodes on a single MNW via e-beam lithography (EBL) technique⁸. This method unavoidably involves contact resistance or chemical contaminations during the complex EBL processes, which affects the result accuracy.⁹⁻¹⁰ This is the main reason why the reported work show huge scatter of the electrical and magneto-electrical properties of individual metallic nanowires.

Nanomanipulators *in-situ* scanning electron microscope (SEM) could be a good solution to sort out the accurate measurements of magneto-transport properties of a single nanowire. This technique has been employed to measure the electrical transport properties of various nanowires¹¹⁻¹², which can select contact points at will and easily realize the test of length-resistance relationship. In addition, the whole test process is dynamically visualized and the contact resistance can be effectively avoided by punching probes into nanowire body. However, beside our very early prototyped demonstration¹³, the measurement of magneto-transport property of a single magnetic nanowire by nanomanipulators *in-situ* SEM has not been reported yet as far as we have known.

MR mechanism is considered to mainly generate from spin-dependent scattering of conducting electrons at interfaces between magnetic and non-magnetic layers.¹⁴ Magnetic state and external field direction also have an influence on the amplitude of MR effect. Therefore, the fully understanding of MR effect in individual magnetic multilayered NWs requires detailed information of interface, magnetic state and external field direction beside morphology, chemistry and geometry. Numerous experiments and model calculations have been already done to elucidate the impacts on the nanowire GMR.¹⁴⁻¹⁶ However, few studies focus on the interfacial structure at the atomic scale and magnetic state at nanoscale¹⁷⁻¹⁸ because of the requirement of extremely high spatial resolution. Aberration-corrected transmission electron microscopy (Cs-corrected TEM) and scanning transmission electron microscopy (Cs-corrected STEM) equipped with biprism and Lorentz lens provide the detection possibility.¹⁹⁻²¹ The image function of both Cs-corrected TEM and Cs-corrected STEM has recently achieved a surprisingly high spatial resolution below 0.5 Å and their chemical function (accessories of electron energy-loss spectroscopy (EELS) and energy-dispersive X-ray analysis (EDX)) has readily realized a direct imaging and electronic-state detection of individual atoms and even chemical bonds.²² The additional function of off-axis electron holography (EH)²³⁻²⁵ and Lorentz mode reaches a ~ 1 nm high-resolution visualization of electromagnetic field in an isolated nanostructure.

In this work, we choose CoCu/Cu MNW as an example sample to contribute an exploration of MR mechanism of single MNW via accurate characterization of its magneto-transport behavior in situ SEM by using our dedicated magneto-transport instrument (MTI) (see details in supplementary information S1). Off-axis EH method was employed to detect the magnetic state of the single CoCu/Cu MNW. Theoretical

three-dimensional (3D) distribution of magnetic moments was simulated by public-shared OOMMF software. Density functional theory (DFT) calculations on the base of experimental interface and magnetic state information were further carried out to disclose the intrinsic MR origin of the CoCu/Cu MNW. To further verify the correctness of the measurement results, Granular Films model is employed. Our work should not only provide a mature technique to accurately measure magneto-transport properties of single NW in situ SEM, but also disclose the MR mechanism of magnetic MNW.

2 Results and discussion

2.1 Structural and chemical information of the nanowires

The magneto-electrical properties strongly depend upon the microstructure of the nanowires such as the diameter, the thickness of the non-magnetic and magnetic layers, and interfacial structure between layers and nanoparticles as well as chemical composition. Therefore, the structural and chemical information of the nanowires are firstly characterized. Figure 1a shows a representative SEM image of CoCu/Cu MNWs, which were completely released from the channels of AAO template. Quantitative analysis discloses that the average diameter of the CoCu/Cu MNWs is approximately 52 nm, and that of length is about 30 μm . TEM image (Fig. 1b) shows that individual MNWs clearly have a bamboo-like morphology, in which the black contrast sections are CoCu layers and those of grey are Cu layers (see more analysis in supplementary information S2 and chemical analysis below). This result confirms that the periodic multilayered structure of CoCu/Cu nanowire has been successfully formed. Statistical analysis from numerous TEM images shows that the average thickness of the CoCu layers is about 124 nm, and that of non-magnetic Cu layers for

~ 14.2 nm. It is then calculated that the length-to-diameter ratio t/d of the CoCu layers (t and d represent the thickness and diameter, respectively) is about 2.16, which is larger than 1,²⁶ demonstrating that the magnetization direction should prefer the length axis. In addition, the thickness of non-magnetic layer is a key factor to study the magneto-electrical properties of MNWs²⁷, which is essential to be accurately measured. The thickness of the Cu layer (marked by yellow area) is thus detected under high-magnification TEM as shown in Fig. 1c, which gives an approximate 13.6 nm. This thickness reveals that the interlayer exchange coupling between neighbored magnetic CoCu layers should become very weak and their magnetic moments between neighbored magnetic CoCu layers are also uncorrelated.

Figure 1. The morphology, crystal structure, chemical composition of the CoCu/Cu MNWs analyzed by SEM and TEM. (a) SEM image of the nanowires dispersed on an SiO₂(200nm)/Si with free-standing formation after template dissolved; **(b)** TEM image of two nanowires with 55 nm uniform diameter; **(c)** A typical HRTEM of non-magnetic Cu layer; **(d)** HAADF-STEM image of MNW. **(e) - (g)** EDX elemental mappings of Co K α , Cu K α and

their combination, respectively. **(h)** EDX spectra measured from a bundle of CoCu/Cu, CoCu and Cu nanowires.

The chemical analysis was performed by using EDX and STEM mapping techniques. Fig. 1d shows the chosen single CoCu/Cu MNW, of which elemental maps of Co Ka and Cu Ka are presented in Fig. 1e and 1f, respectively. Figure 1g is their combination map, clearly showing a periodic structure of the blue Co and red Cu layers. Fig. 1e reveals that there is almost no Co element in the Cu layers, demonstrating a pure chemical composition. However, the signal of Cu element (Fig. 1f) exists in both Co and Cu layers. To quantify the proportion of the Cu impurity in the Co layers, additional single phase nanowires of Cu and CoCu were prepared (see supplementary information S3 for details). EDX spectra from the CoCu/Cu, CoCu and Cu nanowires were then measured to detect the chemical compositions as shown in Fig. 1h. As expected, Cu peaks appear in all spectra and Co peaks appear on both CoCu/Cu and CoCu nanowires, revealing a pure Cu chemistry in the Cu layers of the multilayered nanowires. Quantitative analysis of the EDX spectrum from the single phase CoCu nanowires indicates a 92.1:7.9 atomic ratio of Co:Cu, inferring a $\text{Co}_{92.1}\text{Cu}_{7.9}/\text{Cu}$ composition of the multilayered nanowires. As our recent report¹, the CoCu layers consist of 5 nm Cu grains and 12 nm Co grains, and both are fcc polycrystals with random orientations. Besides, the interface between Co and Cu grains is rough, which causes a large scattering with conduction electrons and reduces the magneto-resistance. Further investigation shows that the concentration of Co grains per unit volume is about 57.27% in the CoCu layers, of which detailed calculation is shown in supplementary information S4.

2.2 Resistance measurement of a single CoCu/Cu MNW

Electrical properties of the single CoCu/Cu MNW were measured by using dedicated MTI in situ SEM as illustrated in Fig. 2a. Figure 2b shows a representative SEM image of the experimental set-up, where two conductive nichrome probes pierced the nanowire body to achieve a good ohmic contact. The measured resistance (R_e) includes the intrinsic resistance of CoCu/Cu MNWs (R_i), the contact resistance (R_c) and the feed line resistance of circuitry (R_f). To remove the feed line resistance, the two tips were contacted to each other (Inset of Fig. 2c) and current vs applied voltage relationship was then recorded as shown in Fig. 2c. The inverse of the slope of this I-V curve gives a 60.3Ω feed line resistance.

Figure 2 Resistance measurement of single periodic CoCu/Cu MNW. (a) Schematic illustration of the measurement setup, in which two conductive nichrome probes are tightly pressed into the nanowire body; (b) A representative SEM image of the experimental testing; (c) I-V behavior of the tip-tip circuit, as shown in the top-left illustration; (d) - (i) A series of SEM images of six measurement positions (length), which are for obtaining R-L curve; (j)

The corresponding I–V behaviors at the 6 MNW lengths; **(k)** The plotted R–L curve, in which red dots are for the measured data and blue line is for the linear fit of the experimental data.

To further get the accurate values of R_i and R_c , resistance-length (R_e - l) relationship were detected via the changes of touching positions of nanomanipulator tip. Fig. 2d-2i show the measured positions and Fig. 2j shows the corresponding I–V curves of the six touching positions. Furthermore, R_e - l curve can be plotted according to these curves as shown in Fig. 2k. It is found that R_e is a linear function of l and thus the R_e - l curve is fitted with a linear function. The intercept b of this curve, which is $62.8\ \Omega$, is the sum of R_c and R_f . Since R_f has been obtained above, the R_c is then calculated to be $R_c = b - R_f = 2.5\ \Omega$. This value demonstrates that the contact resistance is very small and the intrinsic R_i can be obtained by subtracting the intercept value of $62.8\ \Omega$, which proves that our designed MTI in situ SEM is very accurate and reliable for detecting the electrical property of a single nanomaterial.

The resistivity ρ of the MNW is further analyzed. It is noted that the 52 nm diameter of our MNW is larger than the free electron mean as reported in previous work.^{11-12, 28} The apparent quantum confinement effect is negligible in the case of our CoCu/Cu MNW and the classical of resistance can be adapted as:

$$R_e = R_i + R_c + R_f = \frac{\rho}{A}l + b \quad (1)$$

where A is the MWN's cross-sectional area. ρ is then calculated to be $3.39 \times 10^{-7}\ \Omega \cdot \text{m}$, which is higher than that of the standard Co and Cu bulk ($6 \times 10^{-8}\ \Omega \cdot \text{m}^{29}$ and $1.75 \times 10^{-8}\ \Omega \cdot \text{m}^{30}$ at the room temperature, respectively). Based on the structural and chemical information of MWN, this high resistivity is deduced to originate from the rough interface structure between CoCu layers and Cu layers (Fig. 1c), which may lead to an increase of electron scattering at the interface. Besides, the nanocrystalline size, the

number of interface, and diameter of the nanowires also influence the resistivity.

2.3 Magnetoresistance measurement of a single CoCu/Cu MNW in situ SEM

Figure 3. Distribution of magnetic moments in CoCu/Cu MNW. (a) STEM image of the chosen single MNW; (b) Magnetic flux image displayed by cosine of phase ϕ_M which has been amplified 10 times ($\cos(10 \phi_M)$); (c) Color-coded $\nabla \phi$ where the color hue and brightness represent $\text{atan}(10 \nabla \phi)$ and $|\nabla \phi|$, respectively. (d) The 5 line profiles of magnetic phase marked by using white dotted line in (c); (e) Simulated 3D micromagnetic structures; (f) The corresponding 2D magnetic distribution.

MR is known to relate to the distribution of the magnetic moments and external field direction.²² In order to correctly characterize the origin of magnetic moment distribution in the CoCu/Cu MNW at the remnant magnetic state, traditional VSM measurement and off-axis EH method were employed. Fig. S4 shows two hysteresis loops measured under the external field perpendicular and parallel to the MNW length axis, revealing that the easy axis of the MNW is along the MNW length axis. Fig. 3a shows a chosen single CoCu/Cu MNW used for off-axis EH detection at room temperature and Fig. 3b presents its corresponding magnetic flux ($\cos(10\phi_{MAG})$) image. It is clearly observed that the magnetic lines are mainly parallel to the MNW length axis and bend at the ends of each CoCu layer, revealing that the length axis of the single MNW is easy axis and the dipolar interaction between CoCu layers is weak. This observation is consistent with above structural and chemical analysis. According to above and our previous reported¹ structural information, the CoCu/Cu MNW is polycrystalline and the magnetocrystalline anisotropy can be ignored. The magnetic flux image also demonstrates that the magnetic moment distribution of this CoCu/Cu MNW is determined by its shape anisotropy rather than its magnetocrystalline anisotropy and dipolar interaction. To further determine the physical reason of magnetic moments distribution in CoCu layers, the ϕ_{MAG} distribution in the forms of color-coded ($\nabla\phi_{MAG}$) and the profile of magnetic phase shift are carried out.³¹ Fig. 3c is the ϕ_{MAG} distribution in the forms of color-coded ($\nabla\phi_{MAG}$) where the color hue and brightness represent the direction ($\text{atan}(\nabla\phi_{MAG})$) and intensity ($|\nabla\phi_{MAG}|$) of the in-plane component of magnetic moment, respectively. It is observed that the magnetic moments mainly arrange along the nanowire length axis. However, the magnetic moments at some locations are disorderly arranged inside CoCu layers, revealing that the CoCu layers have a multi-domain structure. The profiles of phase shift as shown

in Fig. 3d coming from the five lines in Fig. 3c (numbered as 1-5) further enlarge the turbulence of magnetic flux (especially rectangle region), double revealing the feature of multi-domain structure. It is known that a single crystal particle is typically in a single domain state when its radius is smaller than critical single domain radius. Theoretically, a single domain state can be maintained in a 12 nm single-crystalline Co particle under the remnant magnetic state, in which the direction of magnetic moments is determined by magnetocrystalline anisotropy. The random orientations of Co grains with tiny size in the magnetic CoCu layers in this work therefore induce the formation of multi-domain structure. This result proves that shape anisotropy is a key factor to determine the micromagnetic configuration of the CoCu layers in the CoCu/Cu MNWs rather than magnetocrystalline anisotropy.

Micromagnetic simulations by using OOMMF software were further used to verify this experimental observation,³² in which the sample model was constructed on the base of experimentally morphological geometries, crystal structures and chemical compositions (see more details in supplementary information S6). Figure 3e and 3f respectively show the simulated three-dimensional (3D) and 2D magnetic induction maps of the CoCu/Cu MNW, clearly revealing that the magnetic moments of each magnetic layer are in a consistent state around MWN length axis and appear a curling state at both ends of each magnetic CoCu layer. This result further reveals that the shape anisotropy plays a dominant role in the alignment of magnetic moments when the length-to-diameter ratio t/d of magnetic layer is larger than 1.

The MR effect of individual CoCu MNWs was then measured by the dedicated MTI in-situ SEM (Fig. 4a). Fig. 4b shows a typical SEM image of practical setup, where two conductive nichrome tips stabbed into a 3.51 μm CoCu/Cu MNW and the MNW was immersed in a varied external field. Figure 4c shows the collected MR

signal (based on standard MR formula of $MR = [(R_H - R_{\max}) / R_{\max}] \times 100\%$) under 1500 Oe maximum field at room temperature. The MR value reaches the maximum value of 1.14% at about ± 270 Oe and minimum value at saturation field, indicating a 270 Oe coercivity. This value is smaller than that of reported CoCu/Cu MNWs arrays³³, which is deduced that the single MNW escapes the neighbored magnetic interactions that exist in MNWs array from group measurements. Besides, above off-axis EH experimental results and micromagnetic simulations reveal that shape anisotropy plays a decisive role in the distribution of magnetic moments in the single CoCu/Cu MNW, which also means that the MR of individual MNWs is dominated by anisotropic magnetoresistance (AMR). However, the detected MR effect of the single CoCu/Cu MNW is very small and the AMR should be almost zero when the direction of the applied magnetic field is parallel to the nanowire length axis³. Therefore, the MR of the single CoCu/Cu MNW should be governed by other mechanism. Density functional theory (DFT) calculations were employed to further understand the MR mechanism of the CoCu/Cu MNW. Figure 4d presents an atomic model of Co-Cu interface along [111] direction according to our recent report of atomic packing analysis by Cs-STEM¹, in which $4 \times 4 \times 1$ Cu unit cell (marked by blue balls) and $4 \times 4 \times 1$ Co unit cell (marked by red balls) are consisted of. Figure 4e and 4f show the 3D and 2D charge redistributions, respectively, defined as $\rho_{\text{Co/Cu}} - \rho_{\text{Cu}} - \rho_{\text{Co}}$, where $\rho_{\text{Co/Cu}}$ is the charge density of the Co/Cu system, ρ_{Cu} or ρ_{Co} is the charge density of the Cu or Co surface. The red color represents for the electron accumulation and the blue is for the electron depletion. The d orbital of Co located at the blue area and the p orbital of Cu at the red area in Fig. 4f indicate that the d orbital of Co loses electrons and the p orbital of Cu gains electrons at the interface. Due to the charge redistribution, Cu atoms at the interface have induced weak magnetic moments (see supplementary

information S7 for the calculated data in detail). As illustrated in Fig. 4g, the Cu and Co atomic layers at the interface have different values of magnetic moments and the opposite direction under zero external magnetic field, which cause a large scattering of conduction electrons and exhibit a state of high resistance. When the applied magnetic field is big enough along the nanowire length axis, the magnetic moments at the interfaces are parallel to align along the length axis and the spin-dependent scattering of the conducting electrons is reduced, which appears a state of low resistance. These physical interactions are similar with those of in granular films system¹⁴, therefore, the intrinsic MR mechanism of the single MNW comes from spin-dependent scattering at the interface between CoCu and Cu layer.

Figure 4. MR mechanism of single CoCu/Cu MNW by using MTI and DFT (a) Schematic of measurement MR inside a SEM. (b) A representative SEM image of the experimental setup; (c) MR effect of individual CoCu/CuCo MNWs with the applied magnetic field parallel to the nanowire length axis; (d) Interfacial structure of Co and Cu along [011] orientation with a good lattice matching; (e) - (f) 3D and 2D charge redistributions, respectively, defined as $\rho_{\text{Co/Cu}} - \rho_{\text{Cu}} - \rho_{\text{Co}}$; (g) Illustration of magnetic moments at

the interface; (**h**) MR as a function of the interface mean free path between Co and Cu grains.

In order to verify the correctness of our measurement, the Granular Films model was applied to analyze the intrinsic MR of the single CoCu/Cu MNW¹⁴. In order to simplify the model, the tilts of magnetic moments at both ends of each magnetic CoCu layer are not taken into account. The MR is governed by eqn. 2 (see detailed derivations in supplementary information S8):

$$\begin{aligned} \text{MR} &= -\frac{\rho_2^2}{\rho_1^2} \\ \rho_1 &= \frac{Ck}{ne^2} \left(\frac{1+P_b^2}{\lambda_m} + \frac{1-C}{C\lambda_{nm}} + \frac{3(1+P_s^2)}{r_m\lambda_s} \right) \\ \rho_2 &= \frac{Ck}{ne^2} \left(\frac{2P_b}{\lambda_m} + \frac{6P_s}{r_m\lambda_s} \right) \end{aligned} \quad (2)$$

where C is the concentration of Co grains per unit volume, 57.27% for the CoCu layer; d_m is the average diameter of CoCu layer; P_b and P_s are the spin-dependent ratios for scattering within the CoCu layers and at their surfaces, which are 0.2 and 0.52, respectively; $\lambda_m = 5$ nm is the mean free path for Co grains and $\lambda_{nm} = 25$ nm for the Cu matrix¹⁴. MR versus interface mean free paths (λ_s) is then plotted according to the eqn. 2 as shown in Fig. 4h. Thus, the MR of single CoCu/Cu is achieved for a small λ_s (small interface roughness), indicating that spin dependent scattering mainly derives from the interfaces between CoCu and Cu layer. The MR is 1.11% when $\lambda_{nm} = 9$ nm, which was close to the experimental value of 1.14 %. This result proves that the mechanism of magnetoresistance effect in individual CoCu/Cu MNWs is mainly derived from spin-dependent scattering of the interfaces between CoCu and Cu layers. Besides, it is demonstrated that our designing and manufacturing magneto-transport

instrument can be used to precisely and simultaneously measure the magnetic transport properties of single nanomaterial in situ SEM.

4 Conclusions

In conclusion, we have investigated the MR origin of single magnetic CoCu/Cu multilayered nanowires via precise measurement of magneto-transport property by our dedicated MTI in situ SEM. Structural and magnetic state analysis disclose that the polycrystalline CoCu layers composed of tiny grains form a multi-domain structure and the dipolar interaction between neighbored magnetic CoCu layers is very weak, whilst, the magnetic moments of each CoCu layer are mainly parallel to the nanowire length axis. The electron holography characterization further proves that shape anisotropy dominantly determines the magnetization configuration of magnetic layers in CoCu/Cu MNW when the length-to-diameter ratio of CoCu layers is larger than 1. Micromagnetic computational simulations double confirm the finding. The MR of individual CoCu/Cu MNW is experimentally measured to be 1.14 %, well matching with theoretical calculation of 1.11% by using Granular Films model. DFT calculations discover that the MR mechanism of the CoCu/Cu MNW originates from spin-dependent scattering at the interface between magnetic and non-magnetic layers. Our work should not only provide a visual evidence to understand the MR mechanism of magnetic MNW, but also demonstrate a novel method to precisely and simultaneously measure magneto-transport properties of single nanomaterial in situ SEM, which should be significant for the exploration of novel functional magnetic structures to satisfy the diverse applications in electronic nanodevices and nanosensors.

Methods

Specimen Preparation. CoCu/Cu MNWs were prepared by electrodeposition into anodic alumina (AAO) templates^{28, 34-35} with 50 nm diameter pores. In a typical process, a gold layer was sputtered onto one side of the template to serve as the working electrode in a standard three-electrode electrochemical cell. A saturated calomel electrode was used as reference electrode and graphite electrode was used as counter electrode. The electrodeposition electrolyte used to deposit CoCu and Cu was composed of 0.5 mol·L⁻¹ CoSO₄·7H₂O, 0.005 mol·L⁻¹ CuSO₄·7H₂O and 0.6 mol·L⁻¹ H₃BO₃. The electrodeposition potential was alternately pulsed between a constant -0.67 V for 30 s to deposit only Cu and -1 V for 6 s to deposit CoCu. The total deposition cycle numbers was 300. The entire electrodeposition process was the same as our previous reported paper¹⁻².

SEM and TEM Characterization. Magnetic CoCu/Co MNWs completely released from the AAO templates by using 0.1 M sodium hydroxide solution were dispersed on an SiO₂(100nm)/Si and holey carbon-film-coated grids for SEM and STEM analysis, respectively. The morphology, atomic structure and chemistry of the MNWs were analyzed by a scanning electron microscope (SEM, Tescan MIRA 3, Czech) and an aberration-corrected scanning transmission electron microscopy (Cs-STEM, FEI Cubed Themis Z, USA) equipped with energy-dispersive X-ray analysis (Super-X), electron energy lose spectroscopy (EELS, Gatan GIF 965, USA), Lorentz lens and biprism, in which the electron biprism for off-axis EH can achieve ~1 nm spatial resolution.

Magnetic Property Measurements and Electron Holography. The magnetic properties of the CoCu/Cu MNWs were carried out by using a vibrating sample magnetometer (VSM, Lakeshore, USA) and magnetic flux distribution was obtained

by electron holography. Off-axis electron holography was carried out in Cs-STEM using an electron biprism at 180 V, under which the amplitude and phase shift of electrons penetrated from sample were recorded. The phase shift is induced by the in-plane component of magnetic and electrostatic potential, of which the magnetic caused phase shift can be obtained from the half of the signal difference between the paired holograms on the positive and negative side of the samples. All off-axis EH experiments were operated at zero magnetic field at room temperature.

Calculations methods. First principles calculations were performed in the framework of density functional theory (DFT) as implemented in the Vienna ab initio simulation package (VASP)³⁶ with projector augmented wave (PAW) potentials and generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE). The energy cutoff for the plane-wave basis set was 520 eV, and total energy was converged to 10^{-5} eV. The crystal geometry was optimized without any symmetry constraints until all residual forces on each atom were less than $0.01 \text{ eV} \cdot \text{\AA}^{-1}$. The Brillouin zone sampling was done by using a Gamma point centered $5 \times 5 \times 1$ k -point mesh. A slab model using $4 \times 4 \times 1$ unit cell was constructed, which consists of 5 Cu layers and 5 Co layers in (011) direction (22 Å vacuum spacing).

Micromagnetic Simulations. The 3D magnetic moment distribution in single CoCu/Cu MNW was simulated by public OOMMF micromagnetic software. The geometrical parameters were defined on the basis of the measured results of STEM and TEM, and the magnetic parameters is obtained by the VSM measurement and the previous report.

MR Measurements. The MR measurements of the single CoCu/Cu MNW were done in situ SEM by using a dedicated MTI which was updated from our previous prototyped MTI. The electromagnet part in MTI was rotated to the nanowire long axis.

After two conductive nichrome probes were tightly punched into the bottom ends of the single CoCu/Cu MNW, a 0.2 V voltage was conducted to the MNW to measure its resistivity when a scanning magnetic field from 1500 Oe to -1500 Oe and opposite scan were applied to the single MNW.

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Author contributions

J.W.Z. designed the experiments, prepared the specimens, characterised, analysed and interpreted all data, and wrote the paper. Y.P. designed the experiments, provided overall guidance, supervised the TEM measurements, characterised, analysed and interpreted the data, and wrote the paper. H. M. grew the CoCu/Cu magnetic nanowires and measured the MR of single nanowire. X. Z. and X. D. performed the TEM measurements together. R. C. performed the DFT calculation. J. Z and Y. P. drafted the paper and all authors commented on the manuscript. S. F. Z., C. S. G., Y. H. and A. K. discussed the data. K.T. performed the DFT calculation and wrote the DFT part of the manuscript. X.X.Z. analysed the magnetic data and discussed the content. J. Z. and H. M. contributed equally to this research.

Reference

1. Zhang, J.; Chen, R.; Li, X.; Peng, Y.; Ma, H.; Hu, Y.; Zeng, X.; Deng, X.; Guan, C.; Hu, Y.; Zhang, M.; Karim, A.; Tao, K.; Zhang, X., Interfacial Roughness Facilitated by Dislocation and a Metal-Fuse Resistor Fabricated Using a Nanomanipulator. *ACS Applied Materials & Interfaces* **2020**.
2. Zhang, J.; Ma, H.; Zhang, S.; Zhang, H.; Deng, X.; Lan, Q.; Xue, D.; Bai, F.; Mellors, N. J.; Peng, Y., Nanoscale characterisation and magnetic properties of Co₈₁Cu₁₉/Cu multilayer nanowires. *Journal of Materials Chemistry C* **2015**, 3 (1), 85-93.
3. Ohgai, T.; Hoffer, X.; Fábíán, A.; Gravier, L.; Ansermet, J.-P., Electrochemical synthesis and magnetoresistance properties of Ni, Co and Co/Cu nanowires in a nanoporous anodic oxide layer on metallic aluminium. *Journal of Materials Chemistry* **2003**, 13 (10), 2530-2534.
4. Kozlov, S. N.; Skryabina, O. V.; Egorov, S. V.; Golovchanskiy, I. A.; Klimenko, A. A.; Napolskii, K. S.; Stolyarov, V. S., Magnetoresistance of a single polycrystalline nickel nanowire. *Journal of Applied Physics* **2019**, 125 (6), 063902.
5. Krimpalis, S.; Dragos, O. G.; Grigoras, M.; Lupu, N.; Chiriac, H., Magnetoresistance and spin transfer torque in electrodeposited NiFe/Cu multilayered nanowires. *Journal of Advanced Research in Physics* **2010**, 1 (2).
6. Tang, X.-T.; Wang, G.-C.; Shima, M., Layer thickness dependence of CPP giant magnetoresistance in individual Co Ni/ Cu multilayer nanowires grown by electrodeposition. *Physical Review B* **2007**, 75 (13), 134404.
7. Krimpalis, S.; Dragos, O.-G.; Moga, A.-E.; Lupu, N.; Chiriac, H., Magnetization processes in electrodeposited NiFe/Cu multilayered nanowires. *Journal of Materials Research* **2011**, 26 (9), 1081-1090.

8. Li, P.; Wu, W.; Wen, Y.; Zhang, C.; Zhang, J.; Zhang, S.; Yu, Z.; Yang, S. A.; Manchon, A.; Zhang, X. X., Spin-momentum locking and spin-orbit torques in magnetic nano-heterojunctions composed of Weyl semimetal WTe₂. *Nat Commun* **2018**, *9* (1), 3990.
9. Dubois, S.; Beuken, J.; Piraux, L.; Duvail, J.; Fert, A.; George, J.; Maurice, J., Perpendicular giant magnetoresistance of NiFe/Cu and Co/Cu multilayered nanowires. *Journal of magnetism and magnetic materials* **1997**, *165* (1-3), 30-33.
10. Huang, X.; Tan, L.; Cho, H.; Stadler, B. J., Magnetoresistance and spin transfer torque in electrodeposited Co/Cu multilayered nanowire arrays with small diameters. *Journal of Applied Physics* **2009**, *105* (7), 07D128.
11. Peng, Y.; Cullis, T.; Inkson, B., Accurate electrical testing of individual gold nanowires by in situ scanning electron microscope nanomanipulators. *Applied Physics Letters* **2008**, *93* (18), 183112.
12. Peng, Y.; Cullis, T.; Luxmoore, I.; Inkson, B., Electrical properties of individual CoPt/Pt multilayer nanowires characterized by in situ SEM nanomanipulators. *Nanotechnology* **2011**, *22* (24), 245709.
13. Elawayeb, M.; Peng, Y.; Briston, K. J.; Inkson, B. J., Electrical properties of individual NiFe/Pt multilayer nanowires measured in situ in a scanning electron microscope. *Journal of Applied Physics* **2012**, *111* (3), 034306.
14. Zhang, S., Theory of giant magnetoresistance in magnetic granular films. *Applied physics letters* **1992**, *61* (15), 1855-1857.
15. Prieto, A. G.; Fdez-Gubieda, M., Influence of the interface on the magnetic anisotropy of CoCu granular alloys. *Physica B: Condensed Matter* **2004**, *354* (1-4), 92-97.

16. Prieto, A. G.; Fdez-Gubieda, M. L.; Chaboy, J.; Laguna-Marco, M. A.; Muro, T.; Nakamura, T., -Interface effects on the magnetic moment of Co and Cu in CoCu granular alloys. *Physical Review B* **2005**, 72 (21).
17. Gan, W.; Chandra Sekhar, M.; Wong, D.; Purnama, I.; Chiam, S.; Wong, L.; Lew, W., Multi-vortex states in magnetic nanoparticles. *Applied Physics Letters* **2014**, 105 (15), 152405.
18. Bryan, M.; Bance, S.; Dean, J.; Schrefl, T.; Allwood, D., Transverse and vortex domain wall structure in magnetic nanowires with uniaxial in-plane anisotropy. *Journal of Physics: Condensed Matter* **2011**, 24 (2), 024205.
19. Beeli, C.; Doudin, B.; Ansermet, J.-P.; Stadelmann, P., Study of Co, Ni and Co/Cu nanowires: magnetic flux imaging by off-axis electron holography. *Journal of magnetism and magnetic materials* **1996**, 164 (1-2), 77-90.
20. Akhtari-Zavareh, A.; Carignan, L.; Yelon, A.; Ménard, D.; Kasama, T.; Herring, R.; Dunin-Borkowski, R. E.; McCartney, M.; Kavanagh, K., Off-axis electron holography of ferromagnetic multilayer nanowires. *Journal of applied physics* **2014**, 116 (2), 023902.
21. Cantu-Valle, J.; Barriga-Castro, E. D.; Vega, V.; García, J.; Mendoza-Reséndez, R.; Luna, C.; Prida, V. M.; Nielsch, K.; Mendoza-Santoyo, F.; Jose-Yacaman, M., Quantitative magnetometry analysis and structural characterization of multisegmented cobalt–nickel nanowires. *Journal of Magnetism and Magnetic Materials* **2015**, 379, 294-299.
22. Reyes, D.; Biziere, N.; Warot-Fonrose, B.; Wade, T.; Gatel, C., Magnetic configurations in Co/Cu multilayered nanowires: evidence of structural and magnetic interplay. *Nano letters* **2016**, 16 (2), 1230-1236.

23. Rodríguez, L. A.; Bran, C.; Reyes, D.; Berganza, E.; Vázquez, M.; Gatel, C.; Snoeck, E.; Asenjo, A., Quantitative nanoscale magnetic study of isolated diameter-modulated FeCoCu nanowires. *ACS nano* **2016**, *10* (10), 9669-9678.
24. Midgley, P. A.; Dunin-Borkowski, R. E., Electron tomography and holography in materials science. *Nature materials* **2009**, *8* (4), 271.
25. Gan, Z.; Gu, M.; Tang, J.; Wang, C.-Y.; He, Y.; Wang, K. L.; Wang, C.; Smith, D. J.; McCartney, M. R., Direct mapping of charge distribution during lithiation of Ge nanowires using off-axis electron holography. *Nano letters* **2016**, *16* (6), 3748-3753.
26. Tang, X. Magnetotransport and magnetization reversal of electrodeposited multilayer nanowires. Rensselaer Polytechnic Institute, 2007.
27. Kashi, M. A.; Ghaffari, M.; Torshizi, F., Structural and magnetic tunability of Co/Cu multilayer nanowires induced by electrolyte acidity and spacer layer thickness. *Journal of Alloys and Compounds* **2019**, 153087.
28. Peng, Y.; Cullis, T.; Möbus, G.; Xu, X.; Inkson, B., Nanoscale characterization of CoPt/Pt multilayer nanowires. *Nanotechnology* **2007**, *18* (48), 485704.
29. Datta, M.; Osaka, T.; Schultze, J. W., *Microelectronic packaging*. CRC press: 2004.
30. Hanaoka, Y.; Hinode, K.; Takeda, K. i.; Kodama, D., Increase in electrical resistivity of copper and aluminum fine lines. *Materials transactions* **2002**, *43* (7), 1621-1623.
31. Murakami, Y.; Tanigaki, T.; Sasaki, T.; Takeno, Y.; Park, H.; Matsuda, T.; Ohkubo, T.; Hono, K.; Shindo, D., Magnetism of ultrathin intergranular boundary regions in Nd-Fe-B permanent magnets. *Acta Materialia* **2014**, *71*, 370-379.
32. Carlton, D. B.; Emley, N. C.; Tuchfeld, E.; Bokor, J., Simulation studies of nanomagnet-based logic architecture. *Nano letters* **2008**, *8* (12), 4173-4178.

- 1
2
3 33. Wang, J.-Q.; Xiao, G., Transition-metal granular solids: Microstructure, magnetic
4 properties, and giant magnetoresistance. *Physical Review B* **1994**, *49* (6), 3982.
5
6
7
8 34. Schlörb, H.; Haehnel, V.; Khatri, M. S.; Srivastav, A.; Kumar, A.; Schultz, L.;
9 Fähler, S., Magnetic nanowires by electrodeposition within templates. *Phys. Status*
10 *Solidi B* **2010**, *247* (10), 2364–2379.
11
12
13
14 35. Wade, T. L.; Wegrowe, J.-E., Template synthesis of nanomaterials. *The European*
15 *Physical Journal-Applied Physics* **2005**, *29* (1), 3-22.
16
17
18
19 36. Kresse, G.; Furthmüller, J.; Hafner, J., Theory of the crystal structures of selenium
20 and tellurium: the effect of generalized-gradient corrections to the local-density
21 approximation. *Physical Review B* **1994**, *50* (18), 13181.
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