Electronic Supplementary Material (ESI) for ChemComm.

Supporting information

Chemical Conversion Synthesis of Magnetic $Fe_{1-x}Co_x$ Alloy Nanosheets with Controlled Composition

Desheng Pan,[a] Bin Xiao, [a] Qing Wang [b] and Hong Wang*[a]

[a] Department of Materials Science and Engineering and Shenzhen Engineering Research Center for Novel Electronic Information Materials and Devices, South University of Science and Technology, Shenzhen, 518055, China

[b] Department of Materials Science and Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802, USA

Experimental Details

Materials. Commercially available reagents including iron acetylacetonate ($Fe(acac)_3$, 98%), coblat acetylacetonate ($Co(acac)_2$, 97%), thiourea(99%), diethylenetriamine (DETA, 99%), trioctylphosphine (TOP, 90%), hexane (97%) and ethanol (99.7%) were purchased from Shanghai aladdin bio-chem technology Co., LTD. (China). All chemicals were used as received without any further purification.

Synthesis of $Fe_{1-x}Co_xS_{1.2}$-DETA hybrid nanosheets. $Fe(acac)_3$ and $Co(acac)_2$ with the molar ratio of 9:1, 7:3, 5:5, 3:7 and 8 mmol thiourea with a total metal/sulfur molar ratio of 1:1 were mixed in 120 mL DETA in a 250 mL three-neck flask. The mixture solution was heated to 100 ^oC and kept for 1 h under a nitrogen flow. Afterwards, the temperature was raised to 200 ^oC at a ramping rate of 5 ^oC min^{-1} and aged for 3 h, followed by cool down naturally. The precipitates were centrifuged at 5000 rmp min^{-1} and washed by 20 mL ethanol for three times. The products were dried in vacuum.

Conversion of $Fe_{1-x}Co_xS_{1.2}$-DETA hybrids to $Fe_{1-x}Co_x$ nanosheets. 500 mg $Fe_{1-x}Co_xS$-DETA powder mixed with 20 mL TOP was heated to 270 ^oC at a rate of 5 ^oC min^{-1} and kept for 2h under N_2 flow in a four-neck flask. Then the heating source was removed to cool down to room temperature naturally. The intermediates were obtained when the temperature reached 250 ^oC in the heating process. The intermediates and final products were centrifuged at 10000 rmp min^{-1} and washed by 20 mL ethanol and 20 mL hexane mixture for three times. The products were dried in vacuum and kept for further characterization.

Characterization. Powder X-ray diffraction (PXRD) was performed on a Rigaku diffractometer (D8 Eco) using the Kα line of Cu target. The transmission electron microscopy (TEM) and high resolution transmission electron microscopy (HRTEM) images were obtained by a Tecnai F30 transmission electron microscope. The scanning transmission electron microscopy (STEM), selected area electron diffraction (SAED), and energy dispersive spectroscopy (EDS) were carried out by a Talos F200X transmission electron microscope. Thermogravimetric analysis (TGA) was carried out on a STA 449 F5 thermal analyzer under N_2 flow with a heating rate of 10 $^{o}C \cdot min^{-1}$ between 30 ^{o}C and 600 ^{o}C. The magnetic hysteresis loops were measured by superconducting quantum interference devices (SQUID) equipped with a vibrating sample magnetometer (VSM).

Supplementary Figures

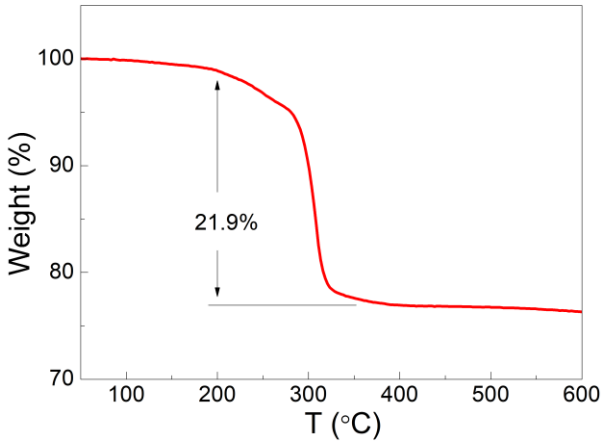

Figure S1 TGA curves of the $Fe_{0.9}Co_{0.1}S_{1.2}$-DETA hybrid nanosheets. The weight loss of 21.9 % over 200-400 ^{o}C is caused by the loss of DETA molecules intercalated in the hybrid structures.

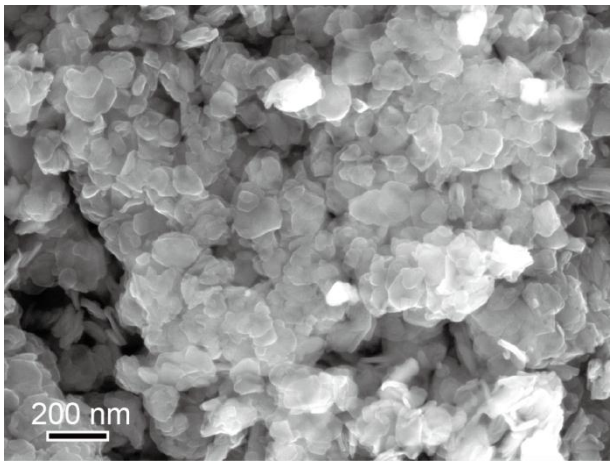

Figure S2 SEM image of the $Fe_{0.9}Co_{0.1}S_{1.2}$-DETA hybrid nanosheets.

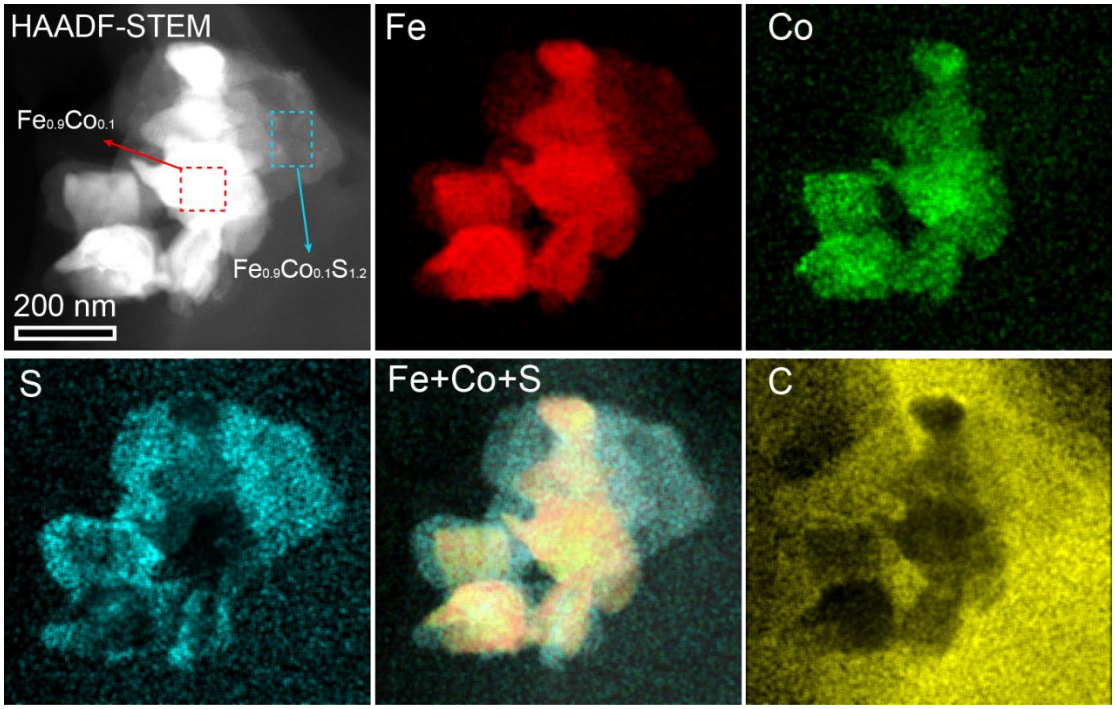

Figure S3 HAADF-STEM and EDX mapping of Fe, Co, S, overlay and C for the intermediates in the converting process. The inhomogeneous distributions of Fe, Co, and S indicate the intermediates are composed of $Fe_{0.9}Co_{0.1}$ and $Fe_{0.9}Co_{0.1}S_{1.2}$ nanosheets.

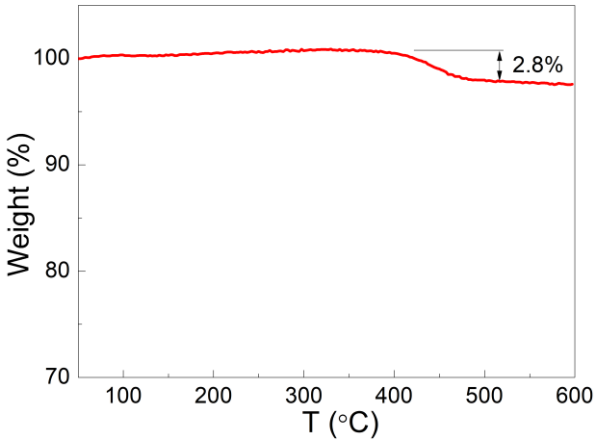

Figure S4 TGA curves of the converted $Fe_{0.9}Co_{0.1}$ magnetic alloy nanosheets. The observation of no weight loss below 400 ^{o}C indicates the complete decomposition of its $Fe_{0.9}Co_{0.1}S_{1.2}$-DETA hybrid precursors. When the temperature is higher than 400 ^{o}C, the weight loss of 2.8 % could be caused by the loss of TOP adsorbed on the surface of the alloy nanosheets.

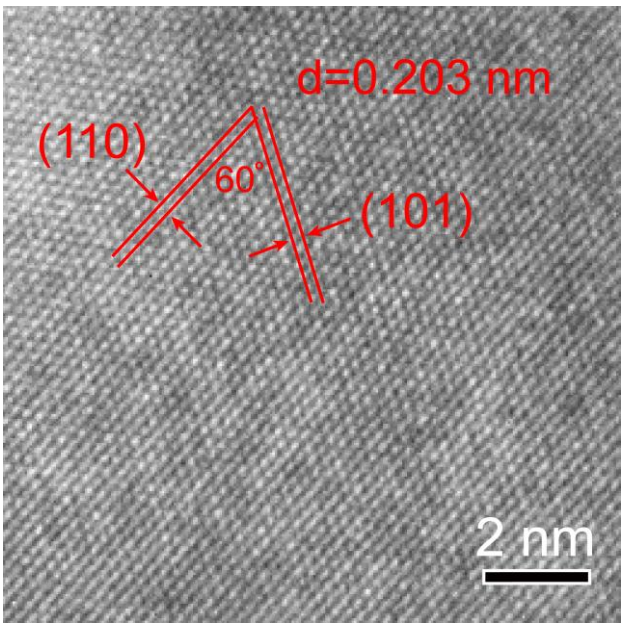

Figure S5 HRTEM image of the $Fe_{0.9}Co_{0.1}$ nanosheet. The HRTEM image shows the $Fe_{0.9}Co_{0.1}$ nanosheet is structurally single-crystal with the lattice spacing of 0.203 nm for the (110) and (101) facets, corresponding to the {110} planes of the cubic FeCo. The orientational relationship between (110) and (101) planes confirms that the alloy nanosheet is oriented along [-111] zone axis.

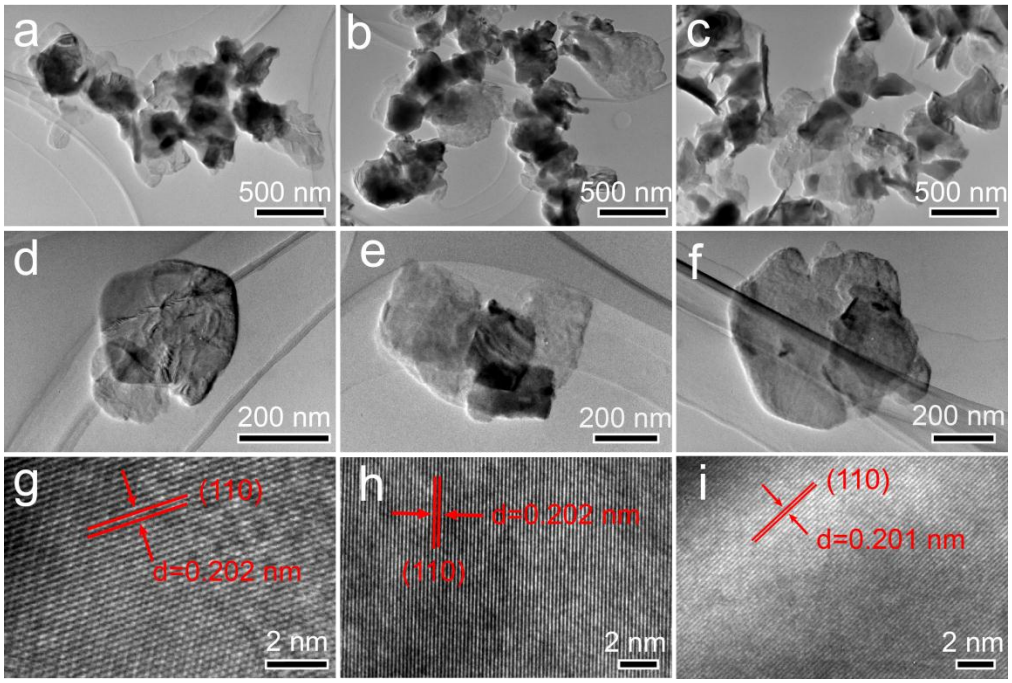

Figure S6 TEM and HRTEM images of (a, d, g) $Fe_{0.7}Co_{0.3}$, (b, e, h) $Fe_{0.5}Co_{0.5}$ and (c, f, i) $Fe_{0.3}Co_{0.7}$ alloy products. All the converted $Fe_{1-x}Co_x$ products share the morphology of 2D nanosheets. The (110) crystal faces are identified from the HRTEM images.

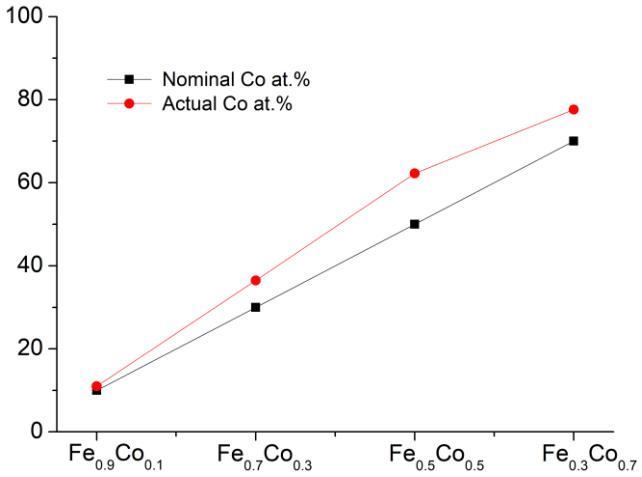

Figure S7 The nominal (black squares) and actual (red circles) Co at.% in the $Fe_{0.9}Co_{0.1}$, $Fe_{0.7}Co_{0.3}$, $Fe_{0.5}Co_{0.5}$, and $Fe_{0.3}Co_{0.7}$ products respectively.

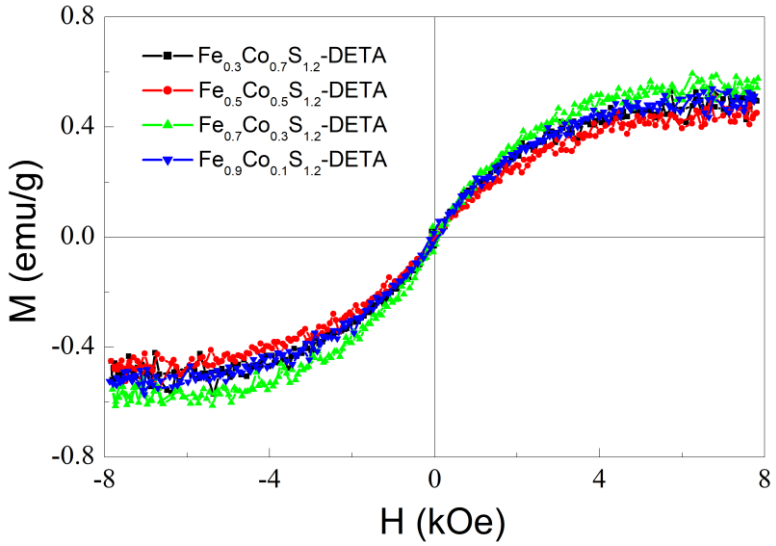

Figure S8 Room-temperature magnetic hysteresis loops of $Fe_{1-x}Co_xS_{1.2}$-DETA (x=0.1, 0.3, 0.5, 0.7) inorganic-organic hybrid nanosheets.