Tunable Ferromagnetism and Thermally Induced Spin Flip in Vanadium-Doped Tungsten Diselenide Monolayers at Room Temperature

Yen Thi Hai Pham, Mingzu Liu, Valery Ortiz Jimenez, Zhuohang Yu, Vijaysankar Kalappattil, Fu Zhang, Ke Wang, Teague Williams, Mauricio Terrones,* and Manh-Huong Phan*

The outstanding optoelectronic and valleytronic properties of transition metal dichalcogenides (TMDs) have triggered intense research efforts by the scientific community. An alternative to induce long-range ferromagnetism (FM) in TMDs is by introducing magnetic dopants to form a dilute magnetic semiconductor. Enhancing ferromagnetism in these semiconductors not only represents a key step toward modern TMD-based spintronics, but also enables exploration of new and exciting dimensionality-driven magnetic phenomena. To this end, tunable ferromagnetism at room temperature and a thermally induced spin flip (TISF) in monolayers of V-doped WSe₂ are shown. As vanadium concentration increases, the saturation magnetization increases, which is optimal at ≈ 4 at% vanadium; the highest doping level ever achieved for V-doped WSe₂ monolayers. The TISF occurs at ≈ 175 K and becomes more pronounced upon increasing the temperature toward room temperature. The TISF can be manipulated by changing the vanadium concentration. The TISF is attributed to the magnetic-field- and temperature-dependent flipping of the nearest W-site magnetic moments that are antiferromagnetically coupled to the V magnetic moments in the ground state. This is fully supported by a recent spin-polarized density functional theory study. The findings pave the way for the development of novel spintronic and valleytronic nanodevices and stimulate further research.

Y. T. H. Pham, V. O. Jimenez, Dr. V. Kalappattil, Prof. M.-H. Phan
Department of Physics
University of South Florida
Tampa, FL 33620, USA
E-mail: phamn@usf.edu
M. Liu, Z. Yu, Prof. M. Terrones
Department of Physics
The Pennsylvania State University
University Park, PA 16802, USA
E-mail: mut11@psu.edu
M. Liu, Dr. F. Zhang, Prof. M. Terrones
Center for Two Dimensional and Layered Materials
The Pennsylvania State University
University Park, PA 16802, USA

Z. Yu, Dr. F. Zhang, Prof. M. Terrones
Department of Materials Science and Engineering
The Pennsylvania State University
University Park, PA 16802, USA
Dr. K. Wang, T. Williams
Materials Research Institute
The Pennsylvania State University
University Park, PA 16802, USA
Prof. M. Terrones
Department of Chemistry
The Pennsylvania State University
University Park, PA 16802, USA

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There has been a growing interest in 2D van der Waals materials due to their potential role in ultralow-power and ultra-compact device applications.[1–5] 2D transition metal dichalcogenides (TMDs) with combined magnetic, electric, and optical features are particularly outstanding for the design and fabrication of multifunctional nanodevices.[5–7] Due to the isotropic Heisenberg exchange,[8] the Mermin-Wagner theorem predicts the absence of long-range magnetic order at finite temperatures in 2D materials. TMD monolayers are known to possess no inversion symmetry and their spin–orbit coupling results in a significant splitting of the Kramers degeneracy.[5,7] However, the 2017 breakthroughs regarding the discovery of 2D magnetism in monolayer semiconductor crystals (e.g., CrI₃) and the observation of layer-dependent magnetic phases open up new paradigms in both fundamental science and device technologies.[9–13] Since these discoveries, a large body of work has been devoted to the realization of long-range ferromagnetic order in monolayer
forms of bulk van der Waals materials (e.g., Cr$_2$Ge$_2$Te$_6$, Fe$_4$GeTe$_6$, Fe$_2$GeTe$_6$, VI$_4$).[14–17] While these systems hold enormous potential for sophisticated magnetoelectronics, as well as for the combination of logic and memory for high-performance computing, they are restricted to low temperature, well below 300 K.[9–17] 2D vanadium-based dichalcogenides such as VSe$_2$ monolayers have recently been reported to show room-temperature ferromagnetism.[18–20] However, the origin of the observed ferromagnetism has remained a subject of debate.[21–24] While decoupling the intrinsic magnetism from the defect-induced magnetism is still a challenging task,[20] the latter has been shown to contribute dominantly to the magnetism of this 2D system.[24] In addition, these VSe$_2$ monolayers are metallic, which cannot access the rich electronic and optical phenomena open to semiconductors. Since the most important technological applications demand magnetic ordering above 300 K, current research is driven by the creation of air-stable room temperature 2D magnets.[18,20,25]

An alternative strategy to induce long-range ferromagnetism (FM) in TMDs is through the introduction of small quantities of magnetic dopants to form a dilute magnetic semiconductor. Density functional theory (DFT) has predicted tunable ferromagnetism over a wide range of monolayer TMD semiconductors doped with magnetic transition metals, such as V-doped MoS$_2$,[26,27] V-doped WS$_2$,[26] and V-doped WSe$_2$.[29] Calculations by Gao et al. have shown an increase in the magnetization and Curie temperature in monolayer V-doped/alloyed MoS$_2$ with increasing V-concentration up to 9 at%.[27] Duong et al. have theoretically and experimentally shown the occurrence of room temperature long-range FM ordering in monolayer V-doped WSe$_2$ for a relatively low (0.1 at%) V-doping level.[29,30] Through a combined study of aberration-corrected high-resolution scanning transmission electron microscopy (AC-HRSTEM), magnetization, and first-principles calculations, we have recently demonstrated room-temperature FM optimization in monolayer V-doped WS$_2$ at an intermediate V-concentration of ≈2 at%, which is consistent with our ab initio calculations of a near-ideal random alloy in which magnetism is quenched due to orbital hybridization at too-close vanadium-vanadium distances.[31] While enhancing FM in these semiconductors is the key to developing TMD-based spintronics and quantum computing technology, it has also enabled the observation and exploitation of new and novel dimensionality-driven physical phenomena.[4–7,21]

Here, we show the enhancement and tunability of room temperature ferromagnetism in V-doped/alloyed WSe$_2$ monolayer films (optimal around 4 at%V) grown on SiO$_2$ substrates using a single-step powder vaporization method. Such strong magnetic-field- and temperature-dependent magnetization enables the observation of a novel magnetic crossover phenomenon associated with a thermally induced spin flip (TISF) that becomes prominent at room temperature. The origin of TISF is attributed to the magnetic-field- and temperature-dependent flipping of the nearest W-site magnetic moments that are antiferromagnetically coupled to the V magnetic moments. Our results are in good agreement with a recent spin-polarized DFT calculation.[29]

First, the structural properties of pristine and V-doped/alloyed WSe$_2$ (0.5–8 at%) monolayer films grown on SiO$_2$ substrates are discussed (sample synthesis is described in Experimental Section, followed the recently published works.[31,32]). Transmission electron microscopy (TEM) diffraction patterns of pristine and V-doped WSe$_2$ monolayers indicate that the V-WSe$_2$ monolayers are single crystals (Figure S1, Supporting Information). Atomic resolution high-angle annular dark-field STEM (HAADF-STEM) images show the high crystalline quality of the synthesized samples and reveal that V replaces W throughout the film (Figure 1a–d and Figure S2, Supporting Information). Vanadium aggregation is quite modest even at 8 at% V (Figure 1d). Average V–V spacing (d) is determined for all V-doped WSe$_2$ samples from the AC-HRSTEM images (Figure S3, Supporting Information). d decreases with increasing V concentration (see Table S1, Supporting Information). For example, d decreases from ≈4.95 nm for 0.5 at% V to ≈1.56 nm for 5 at% V. Such a change in d significantly alters the magnetism of V-doped WSe$_2$ monolayers.

Atomic force microscopy (AFM) images of pristine and V-doped WSe$_2$ monolayers are shown in Figure 1e–i,k. There appear some white particles and dark holes on the surfaces of the flakes, especially for highly doped samples (e.g., 8 at% V). While the white particles are most likely localized add-layers, the dark holes with bright edges could come from the excess salt and selenium precursor residues from the synthesis process. A detailed analysis of these features based on scanning electron microscopy images (Figure S4a,b, Supporting Information), the AFM height profile (Figure S4c, Supporting Information), and Raman spectra (Figures S4d and S5, Supporting Information) is provided in the Supporting Information.

The magnetic properties of the V-doped/alloyed WSe$_2$ monolayer films are characterized by magnetic force microscopy (MFM), magnetometry, and anomalous Hall effect (AHE). A method for extracting magnetic parameters (M$_s$ and H$_C$) from the magnetic hysteresis (M–H) loops of the V-WSe$_2$ monolayer samples is presented in the Supporting Information (Figure S6, Supporting Information). Unlike pristine WSe$_2$ monolayers that are almost diamagnetic in nature (Figure 1f), the vanadium introduction results in long-range ferromagnetic order at room temperature (see Figure 1h, and Figure S7, Supporting Information). These results are also in full agreement with the spin-polarized DFT calculation for a V-doped WSe$_2$ monolayer[29] and the MFM data for a 0.1 at% V-concentration.[30] It is worth mentioning that the magnetic hysteresis (M–H) loops for V-doped/alloyed WSe$_2$ monolayers show a distinct ferromagnetic signature at 300 K (Figure S7, Supporting Information) and a remarkable increase in saturation magnetization (M$_s$) when increasing the V concentration from 0.5 to 4 at% (Figure 2 and Figure S8, Supporting Information), thus agreeing well with the DFT calculations.[30,31] This indicates that the FM of V-doped WSe$_2$ monolayers can be further strengthened by optimizing the distribution of dopant–dopant (V–V) neighbor separations. For high V concentrations (e.g., 5 at%), the FM is partially quenched due to orbital hybridization at too-close V–V distances whose magnetic moments are antiferromagnetically coupled (Figure S7, Supporting Information). This is fully consistent with the absence of magnetic domains probed by the MFM for the 8 at% V-doped WSe$_2$ monolayers (Figure 1i). This trend is also similar to that observed previously for V-doped WS$_2$ monolayers,[33] the FM signal of this system is relatively weaker though. While room temperature...
Figure 1. a–d) Atomic-resolution HAADF-STEM images of pristine (a) and V-doped WSe$_2$ monolayers at 1 at% (b), 4 at% (c), and 8 at% (d) vanadium. The black dots indicate the presence of V atoms that replace W ones. e–l) AFM and MFM images of the pristine (e,f), and 1 at% (g,h), 4 at% (i,j), and 8 at% (k,l) V-doped samples, respectively. While the pristine WSe$_2$ sample shows no magnetic domains (f), the 1 at% and 4 at% V-doped WSe$_2$ samples show noticeable changes in magnetic domain (h,j), when the samples are magnetized with the same magnetic field (0.5 T). The change in magnetic domain becomes unobvious in the 8 at% V-doped WSe$_2$ sample (l), indicating a strong suppression of the FM due to orbital hybridization at too-close vanadium–vanadium spacings.

Figure 2. a,b) Magnetic hysteresis loops taken at 300 and 10 K for V-doped WSe$_2$ monolayers at 1 at% (a) and 4 at% (b) vanadium. c,d) Temperature dependence of saturation magnetization ($M_S$) and coercive field ($H_C$) for the 1 at% (c) and 4 at% (d) V-doped samples. e) Directions of V and W spins in the nearest and distant regions are shown by the arrows. While the nearest W-site (W1) spin couples antiferromagnetically with the V-spin, the distant W-site (W2, W3, W4, W5) spins couple ferromagnetically with the V-spin. The height of the arrow indicates the magnitude of the magnetic moment.
FM has been previously reported by the MFM in V-doped WSe₂ monolayers at very low V-doping concentrations (0.1–0.3 at%). Here, we show enhancement and tunability of ferromagnetism (\(M_S\) and \(H_C\)) at room temperature in V-doped/alloyed WSe₂ monolayers over a wide range of V concentrations (Figure S8, Supporting Information). Magnetic measurements were also performed on V-doped WSe₂ samples synthesized at different times and the reproducible results are obtained. The room-temperature FM nature is independently confirmed by AHE experiments (e.g., for the 4 at% V-concentration, see Figure S9, Supporting Information). In addition to the air-stability of the synthesized V-WSe₂ monolayer films (Figures S10 and S11, Supporting Information), the strong and tunable ferromagnetism enables the full exploration of these 2D magnets and their heterostructures for advanced spintronics and valleytronics. Our findings also assert the recent DFT predictions of long-range FM tunability in 2D TMD semiconductors by changing the vanadium concentration.

From Figure 2c,d, upon lowering the temperature, \(H_C\) increases considerably, while \(M_S\) decreases gradually in V-doped/alloyed WSe₂. The temperature evolution of \(M_S\) can be explained with spin-polarized DFT calculations that have recently been carried out for 1 at% V-doped WSe₂. The calculation results support the presence of an antiferromagnetic (AFM) coupling between V and its nearest W-site (W1) magnetic moments, in addition to the dominant ferromagnetic couplings between neighboring V magnetic moments, and between V and their farther W-site (W2, W3, W4, W5) magnetic moments. A simplified picture that describes the coupling of these spins is depicted in Figure 2e. Accordingly, it is suggested that at low temperatures, the AFM coupling between V-spin up and W1-spin down is strong and remains stable upon the application of an external magnetic field. However, as the temperature is increased, the AFM coupling becomes weak, and the application of a sufficiently high magnetic field will align the magnetic moment of W1 along the direction of the applied magnetic field. As a result, the total magnetic moment or \(M_S\) of V-doped WSe₂ monolayers increases with increasing temperature (Figure 2c,d). Another important fact that needs to be accounted for is the AFM coupling of V magnetic moments at too-close V–V distances due to orbital hybridization, arising from dopant aggregations that often occur in experimentally synthesized V-doped TMD monolayers. This AFM coupling becomes considerable when increasing V concentration (decreasing V–V distance) and competitive with the FM coupling between the moderately separated V-atoms. Upon increasing the temperature, this AFM interaction also becomes weaker, thus increasing the \(M_S\) of the sample. The decrease of \(H_C\) with increasing temperature is attributed to the increase in thermal fluctuations of the magnetic moments, a common feature also observed previously for monolayer V-doped WS₂ films.

Magnetometry measurements on the V-doped/alloyed WSe₂ monolayers also revealed the emergence of a new magnetic crossover between the ascending and descending branches of the \(M–H\) loop at and above a so-called crossover temperature (\(T_{cr} = 170–190\) K) (Figure 3 and Figure S7, Supporting Information). We define the magnetic field values at which the ascending and descending branches of the \(M–H\) loop cross at

![Image](https://example.com/image1.png)

**Figure 3.** a) Magnetic hysteresis loops indicating the presence of a magnetic crossover at high temperature (300 K) and its disappearance at low temperature (100 K) for the 1 at% V-doped WSe₂ sample. Critical fields \(H_{cr}\) and \(H_{cs}\) and difference in saturation magnetization \(\Delta M_S\) are shown. b,c) Temperature dependence of \(H_{cr}\) and \(H_{cs}\) for 1 at% (b) and 4 at% (c) V-doped WSe₂ samples. d) Temperature dependence of \(\Delta M_S\) for the 1 at% and 4 at% V-doped WSe₂ samples.
a low field and emerge at high field range to be \( H_{cr} \) and \( H_S \), respectively. These critical fields were observed to depend largely on temperature; \( H_{cr} \) decreased while \( H_S \) increased with increasing temperature from 175 to 300 K (Figure 3b,c). Over the low-temperature region (\( T < \approx 175 \) K), ferromagnetic and AFM spins are coherently coupled, and the film reveals a conventional magnetic hysteresis loop (inset (i) of Figure 3a). However, at high temperatures (\( T > 175 \) K), we can attribute the magnetic crossover of the ascending branch of the \( M-H \) loop to a thermally weakened AFM V-W1 coupling. At these temperatures, the W1 magnetic moments begin to follow the direction of the V magnetic moments when an external magnetic field exceeds a critical value (\( H_{cr} \)), which leads to the sharp increase in the magnetization (\( M \)) (Figure 3a and its inset (ii)). For the descending branch of the \( M-H \) loop, due to the Zeeman and hyperfine field effects, gradual rotation of the W1 magnetic moments for \( H < H_S \) occurs instead. The temperature \( T_{cr} \approx 175 \) K is considered to be the crossover temperature that separates the TISF state from the low-temperature spin coherent state. Paying close attention to the temperature dependence of the difference in saturation magnetization, \( \Delta M_S \), between the ascending and descending branches of the \( M-H \) loop, one observes \( \Delta M_S \) increases (Figure 3d) and \( H_{cr} \) decreases significantly as the temperature increases from 175 to 300 K (Figure 3b,c). Furthermore, both parameters can be tuned by changing the V concentration (Figure S12, Supporting Information). Therefore, these findings highlight a new and exciting perspective for simultaneously thermal and magnetic control of the magnetization state in atomically thin magnetic semiconductors at desirable ambient temperatures for wide-ranging applications in spintronics, ultrafast sensing, and high-speed information storage systems.\(^{[3-5]} \)

We note that TISF is unique to the V-doped WSe\(_2\) monolayer films, as it is absent in magnetically defect-induced monolayer semiconductors such as WSe\(_2\) and MoS\(_2\)^{[35,36]} as well as in metallic monolayer magnets such as VSe\(_2\) and MnSe\(_2\)^{[18,25]} The TISF is also absent in V-doped/alloyed WS\(_2\) monolayers,\(^{[31]} \) in which V and W1 magnetic moments are ferromagnetically coupled.\(^{[37]} \) Since TISF is present, but small, in bulk single crystals,\(^{[17]} \) our observation of a large TISF in monolayer V-doped/alloyed WSe\(_2\) points to a dimensionality-enhanced novel TISF phenomenon.

Finally, to probe the nature of the exchange coupling between FM and AFM spins in the V-doped WSe\(_2\) monolayers, we have proposed a novel measurement protocol that is typically employed to explore the exchange bias (EB) effect in an FM/AFM binary system.\(^{[38]} \) Briefly, the EB effect arises from exchange coupling between nearby FM and AFM spins at an FM/AFM interface. The protocol includes zero-field-cooled (ZFC) and field-cooled (FC) magnetization versus magnetic field measurements, and thus these were performed on the V-doped WSe\(_2\) samples, and the results are displayed in Figure S13, Supporting Information. For each FC \( M-H \) measurement, the sample was cooled down in the presence of a magnetic field (\( H_{FC} = 5 \) kOe) from 300 to 10 K, then the \( M-H \) loop was measured.

Figure 4. a–d) Field-cooled (FC) magnetic hysteresis (\( M-H \)) loops for eight successive measurement times and coercive field (\( H_{C} \)) as a function of measurement time (\( n \)) for 1 at\% (a,c) and 4 at\% (b,d) V-doped WSe\(_2\) samples. For FC \( M-H \) measurement, a magnetic field of 5 kOe was applied while cooling down the sample to 10 K from 300 K, and the \( M-H \) curve was taken at 10 K. The \( H_C(\( n \)) \) dependence has been fitted using the equation \( H_C = H_C^{1} + Bn + Cn^2 \), where \( H_C^{1} \) is the coercive field for \( n = 1 \), and \( B \) and \( C \) are the constants.
at 10 K. Relative to a symmetric ZFC $M$–$H$ loop, the FC $M$–$H$ loop shows a horizontal shift with a distinctly enhanced value of the coercive field ($H_C$ increases from 645 to 698 Oe for 1 at% V-doped WSe$_2$ and from 393 to 412 Oe for 4 at% V-allloyed WSe$_2$), indicating a small EB effect. The EB field of the V-doped WSe$_2$ monolayer films, defined as $H_{EB} = 0.5 (H^+ + H^-)$, was found to be $\approx 20$ Oe, where $H^+$ and $H^-$ are the coercive fields of the ascending and descending branches of the $M$–$H$ loop, respectively. The dynamics of the FM/AFM spins at the irreversible/reversible interface are examined through the training effect of the coercive field ($H_{CFC}$) obtained from FC $M$–$H$ measurements. The sample was also cooled in $H_{FC} = 5$ kOe ($H_{FC} > H_S \approx 3.5$ kOe) from 300 to 10 K, where the magnetic field was cycled a number of times (Figure 4a–c). As one can clearly observe from Figure 4c,d, $H_{CFC}$ remains almost unchanged for the first to third cycles of the magnetic field but decreases strongly with successive cycling. The decrease in $H_{CFC}$ is due to the rearrangement of spins occurring at the AFM/FM interfaces. The $H_M(n)$ dependence, where $n$ is the number of cycles, for the V-doped WSe$_2$ samples is fit using the equation $H_C = H_C^1 + Bn + Cn^2$, where $H_C^1$ is the coercive field for $n = 1$, and $B$ and $C$ are constants. The $H_M(n)$ dependence is remarkably different from previously reported observations of AFM/FM systems.$^{[38]}$ This can be attributed to a topological arrangement of the V/W spins in monolayer V-WSe$_2$ (Figure 2e), combined with low dimensionality effects.

In summary, we have demonstrated the enhancement and tunability of room temperature ferromagnetism in V-doped WSe$_2$ monolayers at 0.5 to 5 at% V concentrations. An intriguing and novel thermally induced spin flipping phenomenon has also been observed, which appears to be an intrinsic property of this 2D material. The novel magnetic switching, in conjunction with tunable ferromagnetism, now enables the direct magnetic control of valley splitting in V-WSe$_2$ monolayers at room temperature.$^{[31]}$ an arrangement in stark contrast to the interfacial magnetic exchange field induced by a ferromagnetic substrate (e.g., EuS, CrI$_3$) whose Curie temperature is limited to low temperature ($<100$ K).$^{[34,39]}$ Our findings provide an approach for the development of applications in van der Waals spintronics, valleytronics, and quantum computing devices.

**Experimental Section**

**Samples Synthesis:** In a typical process, a deionized water solution of ammonium metatungstate ((NH$_4$)$_6$H$_2$W$_12$O$_{40}$), vanadium oxide sulfate (VO(SO$_4$)$_2$), only for V-doped WSe$_2$ samples, and sodium chloride (C$_2$H$_5$NaO$_3$) was spin-coated onto SiO$_2$/Si substrates. The substrates were then placed in a quartz tube, along with an alumina boat containing selenium powder located upstream. With a mixture of Ar/H$_2$ (10% H$_2$) gas flushing at 80 sccm, the quartz tube was heated in a furnace at 825 °C for 10 min. After the reaction, the system was left cooling down naturally to room temperature. As compared to other work,$^{[32]}$ the successful incorporation of higher V-doping concentrations into V-WSe$_2$ monolayers was achieved in the present study, which was likely related to the choice of W and V precursors.$^{[31]}

**Materials Characterization:** The aforementioned pristine and V-doped monolayer WSe$_2$ samples were transferred onto Quantifoil R 2/1 200 mesh TEM grids. HAAADF-STEM was then performed on an FEI Titan G² 60–300 kV microscope with a cold field-emission electron source. The microscope was operating at 80 kV with double spherical aberration correction to obtain a high resolution of $\approx 0.7$ Å. A HAADF detector (collection angle 42–244 mrad, camera length 115 nm, beam current 45 pA, beam convergence 30 mrad) was used to acquire Z-contrast STEM imaging.

**Magnetic Measurements—Magnetic Force Microscopy:** The pristine and V-doped WSe$_2$ samples were magnetized with a strong magnet for $\approx 10$ s before the scans. Both AFM and MFM images were obtained on a Bruker Icon AFM system, in tapping mode for sample surface topography and lift mode for magnetic domain structure observation. A Bruker Sb-doped Si magnetic probe (225 x 35 x 2.8 µm$^3$, with tip radius $\approx 30$ nm) was used for the measurements. The force constant and the resonant of the tip were $3.0$ N m$^{-1}$ and 75 kHz, respectively.

**Magnetic Measurements—Vibrating Sample Magnetometry:** Temperature- and magnetic-field-dependent magnetization measurements were carried out in a Physical Property Measurement System (PPMS) from Quantum Design with a vibrating sample magnetometer magnetometer over a temperature range of 2–350 K and fields up to 9 T. All magnetic measurements were conducted with the applied magnetic field parallel to the film plane. For field-dependent magnetization measurements ($M$–$H$), the sample was cooled from 300 K in the absence of a dc field and M–H data was taken at each selected measurement temperature. To exclude unwanted effects on the magnetization versus magnetic field ($M$–$H$) loops that can arise from subtracting diamagnetic and paramagnetic backgrounds, the smoothed, as-measured $M$–$H$ loops, were presented at all measured temperatures and the saturation magnetization ($M_S$) and coercive field ($H_C$) were deduced directly from these loops (see Figure S6, Supporting Information).

**Magnetic Measurements—Anomalous Hall Effect:** The 4 at% V-doped WSe$_2$ monolayer film was etched into the shape of a Hall bar device using ion milling. Etching was done using a shadow mask which was fabricated lithographically. Electrical contacts were made using indium pressed dot and Au wires. An alternating current of 0.1 mA was applied for detecting the AHE. A measurement configuration is shown in Figure S5, Supporting Information, and the magnetic field ($H$) was applied perpendicular to the sample surface. AHE measurements were performed at 300 K inside the PPMS. The anomalous Hall resistance data was obtained after subtracting the linear contribution from the ordinary Hall effect.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.
Author Contributions

Y.T.H.P. and M.L. contributed equally to this work. M.L., Z.Y., and F.Z. conducted the synthesis experiments. M.L., K.W., and T.W. conducted the TEM/MFM characterizations. Y.T.H.P., V.O.J., V.K., and M.-H.P. performed the magnetic measurements and analyzed the magnetic data. V.K. measured and analyzed the AHE data. M.-H.P. and M.T. initiated the concept and supervised the whole work. All authors have participated in writing the manuscript and given an approval to the final manuscript.

Keywords

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