Dilute magnetic semiconductors (DMS), achieved through substitutional doping of spin-polarized transition metals into semiconducting systems, enable experimental modulation of spin dynamics in ways that hold great promise for novel magneto–electric or magneto–optical devices, especially for two-dimensional (2D) systems such as transition metal dichalcogenides that accentuate interactions and activate valley degrees of freedom. Practical applications of 2D magnetism will likely require room-temperature operation, air stability, and (for magnetic semiconductors) the ability to achieve optimal doping levels without dopant aggregation. Here, room-temperature ferromagnetic order obtained in semiconducting vanadium-doped tungsten disulfide monolayers produced by a reliable single-step film sulfidation method across an exceptionally wide range of vanadium concentrations, up to 12 at% with minimal dopant aggregation, is described. These monolayers develop p-type transport as a function of vanadium incorporation and rapidly reach ambipolarity. Ferromagnetism peaks at an intermediate vanadium concentration of ~2 at% and decreases for higher concentrations, which is consistent with quenching due to orbital hybridization at closer vanadium–vanadium spacings, as supported by transmission electron microscopy, magnetometry, and first-principles calculations.

Room-temperature 2D-DMS provide a new component to expand the functional scope of van der Waals heterostructures and bring semiconducting magnetic 2D heterostructures into the realm of practical application.
Intrinsic ferromagnetism has been confirmed in semiconducting monolayer CrI₃ and insulating few-layer CrₓGeₓTe₇ at cryogenic temperatures. Moreover, a transition from paramagnetic to ferromagnetic in vanadium diselenide has been recorded when this metallic material was isolated in monolayer form. Air sensitive monolayer VSe₂ has displayed ferromagnetic order even at and above room temperatures.

Monolayer samples provide compelling advantages in the characterization of atomic structure and integration into van der Waals heterostructures. Monolayer tungsten disulfide is a direct-gap semiconductor with high photoluminescence yield that can achieve a reasonably high on/off current ratio (>10⁷) in field-effect transistor geometries. Reliable substitutional cation doping of WS₂ and its sister material MoS₂ can induce degenerate n-type (rhenium[23]) and p-type (carbon[22] and niobium[23]) conduction. Beyond simply introducing charge carriers, a judicious choice of dopant may also introduce spin polarization. Scalable and controllable synthesis of single-phase monolayer DMSs with ferromagnetic ordering at room temperature could thus provide a new component for van der Waals heterostructures that express novel modes of magneto–electric and magneto–optical response.[24,25]

We report the single-step and atmospheric pressure deposition (via film sulfidation) of high-quality V-doped WS₂ monolayers exhibiting room-temperature ferromagnetism. Aberration-corrected high-resolution scanning transmission electron microscopy (AC-HRSTEM) and X-ray photoelectron spectroscopy (XPS) reveal substitutional vanadium concentrations up to 12 atomic percent (at%) without substantial structural deformation or degradation. Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation. Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation. Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation. Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation. 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Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation. Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation. Interestingly, vanadium doping (or alloying) reduces the optical bandgap and induces structural deformation or degradation.

Representative Raman spectra of pristine and V-doped WS₂ monolayers were obtained using excitation wavelengths of intermediate vanadium concentration (∼2 at%). First-principles calculations suggest that magnetism can be further strengthened by optimizing the distribution of dopant–dopant neighbor separations and also reveal how spin-polarized impurity levels break the valley degeneracy. These results now establish a promising route to room-temperature 2D spintronic devices.
Figure 1. a) One-step synthesis of monolayer V-doped WS₂, optical and electronic properties, as described schematically, b) yields a TEM/EELS spectrum with a prominent vanadium L₂,3 edge. c) A loss of double resonance in Raman (under 532 nm excitation) and d) pronounced change in photoluminescence response reflect a change of electronic structure as a function of V doping. e–h) Back-gated V-doped WS₂ field-effect transistors were fabricated on a 50 nm thick Al₂O₃ substrate with a Pt/TiN/p⁺ back-gate electrode for each doping level. Drain current (I_DS) versus back-gate voltage (V_BG) (obtained for drain voltages from 0.2 to 1 V in 0.2 V steps) show a steady shift in threshold voltage across different doping levels and achieve close-to-symmetric ambipolar conduction in heavily doped WS₂.

532 nm (Figure 1c) and 488 nm (Figure S5, Supporting Information). Pristine WS₂ monolayers exhibit the representative E′(Γ) and A₁′(Γ) first-order phonon modes at 355 and 417 cm⁻¹, respectively.[22] Both E′(Γ) and A₁′(Γ) blueshift as a function of vanadium concentration, which is consistent with previously reported spectra of V-doped MoS₂.[27] In V-doped WS₂ samples, the defect-activated longitudinal acoustic mode (LA(M)) gradually emerges as the vanadium concentration increases, indicating lattice disorder induced by V dopants.[28] A high-intensity 2LA(M) second-order double resonance peak involving two longitudinal acoustic phonons[29] characteristic for pristine WS₂ monolayers was progressively suppressed upon increasing vanadium concentration, indicating substantial changes in the WS₂ electronic structure, which drive the system out of resonance.[22,28] Pristine monolayers of WS₂ show an intense PL peak at 1.97 eV, corresponding to the A exciton[30] (Figure 1d). The optical gap decreases under increasing vanadium doping, while the PL peak broadens (likely due to lattice disorder from dopants possibly accompanied by vacancies) and drops in intensity. This evolution of the PL response is consistent with the Raman results discussed above.

Figure 1e–h shows the drain current (I_DS) versus back-gate voltage (V_BG) transport characteristics for 0, 0.4, 2, and 8 at% vanadium monolayers at various drain voltages (V_D), measured by back-gated FET geometry (detailed in Experimental Section). The 0 at% device shows unipolar electron conduction with no hole current, as expected for a pristine WS₂-based FET. A small hole branch emerges at 0.4 at% doping (Figure 1f), with increasing hole currents at higher doping levels as the threshold voltage shifts from 2.9 volts in the pristine system to 5.6, 6.3, and 11.3 volts in devices with progressively higher doping levels (we extract V_{TN} by the iso-current method for a current of 1 nA µm⁻¹ at V_D = 1 volt). The systematic threshold shift confirms p-type doping, with the most heavily doped sample demonstrating close-to-symmetric ambipolar transport. Strong hole current is not seen as p-type conduction in most 2D materials, including WS₂, is limited by a large Schottky barrier (SB) at the metal/WS₂ interface for hole injection, due to the phenomenon of metal Fermi level pinning. Thermal boundary conductance (h_K) measured at the Al/V-WS₂/SiO₂ interface using time-domain thermo-reflectance (Experimental Section, Figure S6, Supporting Information) reveals a significant improvement in heat dissipation under vanadium doping that could be helpful in device applications.

High-angle annular dark-field (HAADF)-STEM imaging confirms the presence of substitutional V atoms at W sites (written VW) in WS₂ and reveals surprisingly little dopant aggregation. Under Z-contrast imaging, vanadium is easily distinguished from much heavier tungsten, and its concentration can be extracted by statistical analysis of HAADF-STEM images (Figure 2a–d and Figure S7, Supporting Information) at multiple locations on each flake. Dopant aggregation is modest even at 12 at% vanadium. Occasional aggregations into stripes at high doping levels (Figure S3, Supporting Information) tend to align to the proximate outer edge of the flake, thus suggesting
Figure 2. a–d) Atomic resolution HAADF-STEM images and e–h) magnetization versus field loops at 300 K for pristine and vanadium-doped WS₂ monolayers at 0.4, 2, and 8 at% vanadium, scale bars are 2 nm. i) An expanded view of the hysteresis loop for the 2 at% sample and j) its temperature-dependent saturation magnetization \( M_S \) and coercivity \( H_C \).

Comparing experimental STEM images to image simulations (simulation details in Experimental Section, Figure 2a–d and Figure S7, Supporting Information) clearly reveals elemental identities by contrast differences \( W > 2S > V > 1S \). At vanadium concentrations of 8 at% or above (Figure S3e, Supporting Information), sulfur vacancies are more likely to be coupled to V atoms \( \text{written } VW^+ \), which is consistent with prior work on TMD alloys \cite{18,32} and first-principles calculations described below.

The magnetic properties of pristine and V-doped WS₂ monolayer samples were measured by a vibrating sample magnetometer. To exclude unwanted effects on the magnetization versus magnetic field \( M-H \) loops that can arise from the SiO₂ substrate and the double-sided carbon tape used to hold the sample (see Figure S8, Supporting Information) and from subtracting diamagnetic backgrounds (see Figure S9, Supporting Information) \cite{33,34} Figure 2e–h presents smoothed-as-measured \( M-H \) loops at 300 K, and we deduced the saturation magnetization \( M_S \) and coercive field \( H_C \) directly from these loops. The pristine WS₂ sample exhibits a weak ferromagnetic signal, which we tentatively ascribe to undercoordinated sulfur atoms at crystallographic defects (e.g., edges) \cite{35} on a diamagnetic background. Vanadium doping of 0.4 at% significantly increases the ferromagnetic signal \( (M_S \text{ and } H_C) \) with further strengthening at 2 at% doping and then a much weaker ferromagnetic response at 8 at% vanadium. The ferromagnetism observed in V-doped samples is too strong to originate from edge effects, and its dependence on vanadium concentration suggests an origin in local magnetic moments associated with unpaired electrons in vanadium d orbitals \cite{16,18,33}. The 2 at% WS₂ sample shows large, clear hysteresis loops at all temperatures from which \( M_S \) and \( H_C \) (Figure 2i) are extracted and plotted as a function of temperature (the \( M-H \) loops are close to square when rotated to account for the diamagnetic background). We performed magnetic measurements on three V:WS₂ samples for each concentration and obtained reproducible results (Figure S10, Supporting Information). The saturation magnetization and coercivity both increase with decreasing temperature (shown in Figure 2j), with an interesting non-monotonicity of both around 150–200 K. Since magnetic measurements using the Physical Property Measurement System (PPMS) are limited to 350 K, we cannot measure \( M-T \) or \( M-H \) curves at higher temperatures to determine Curie temperature \( T_C \) values of the V:WS₂ samples. In order to estimate the Curie temperature of the V:WS₂ monolayers (e.g., 2 at% V), we have fitted the \( M(T) \) and \( M_s(T) \) data to a theoretical expression: $M(T) = M(0) \times [1-(T/T_C)]^\alpha$ adapted for a ferromagnetic system (see Figure S11, Supporting Information). \cite{16,37} The value of $T_C$ that edge energetics/kinetics influence dopant aggregation. \cite{31}
Figure 3. DFT calculation results for V-doped WS₂ monolayers. All non-equivalent positions for the second V dopant are circled. The band structures of a) single vanadium and b–d) two vanadium atoms with the nearest and the farthest separations are plotted. Other, symmetry non-equivalent k-directions (due to low-symmetry dopants placements) look similar (Figure S12, Supporting Information). Red/blue indicates spin up/down polarization for states of V dz² character. The arrows in the supercell show the primitive-cell lattice vectors used to label dopant pairs in Table 1.

≈ 470 K fitted from the data sets is in line with the previous observation of ferromagnetic signals above 420 K from magnetic force microscopy (MFM) measurements carried out on V-doped WSe₂ monolayers. Our direct observation of tunable room-temperature ferromagnetism in the V-doped WS₂ monolayers from the magnetometry measurements is an important evidence towards establishing the existence of the ferromagnetic order in V-doped TMDs.

Estimates of the magnetization per formula unit arising from V doping must address uncertainties in the overall sample thickness, the presence of a diamagnetic background, and possible contributions from point and edge defects as seen in pristine material. For the most reliable such estimate, we selected the sample with the strongest ferromagnetic signal, at 2 at% V doping, and subtracted from its measured magnetization (= 0.252 × 10⁻⁵ emu) the highest measured Ms of pristine-but-defective WS₂ (= 0.241 × 10⁻⁵ emu). We assumed an average sample thickness of ≈ 1 nm (i.e., mostly monolayer) across the 3 × 4 mm sample area and estimated the number of formula units from the known mass density (7.5 g cm⁻³) and formula unit mass (245.3 g mol⁻¹) of WS₂ (vanadium does not significantly affect the mass density at this level of precision). The result is an Ms of ≈ 0.054 μB per formula unit or ≈ 2.7 μB per V atom; a similar analysis for other samples yields values ranging from 1.5 to 3.6 μB per V. The variation between samples and modest excess over the computational results (see below) may arise from uncertainties in film thickness (e.g., some multilayer portions on the single-layer film, possibly with interstitial V[39]), vanadium concentration (we use the concentration in the middle region of Figure S4, Supporting Information; the exact overall doping level is unknown) and those induced by background subtraction, modulated by the possibly distinct temperature dependences for different magnetic contributions. Considering the role of defects in these systems, it is important to consider evidence that may help distinguish direct contributions arising from the spin polarization of states associated with substitutional vanadium from indirect contributions which might arise from, for example, structural defects that could be induced by the presence of vanadium during growth. The nonmonotonicity in the magnetic response as a function of vanadium doping is helpful here; the return of the magnetic response at the highest vanadium doping levels to behavior similar to that of undoped samples is consistent with the quenching of magnetism observed in first-principles calculations (see below), but is more difficult to reconcile with an indirect mechanism mediated by structural defects.

Density functional theory calculations show local moments on substitutional vanadium atoms whose spin polarization and coupling are sensitive to the relative placement of dopants, a behavior similar to that seen in other computational investigations of doped TMDs[15,16,40]. A single vanadium dopant in a 7 × 7 supercell hosts a local moment of 0.67 μB with vanadium dz² character associated with a partially occupied spin-split defect level sitting ≈ 74 meV below the valence band maximum, as shown in Figure 3. Table 1 compiles the results of the interaction between two vanadium dopants in this supercell. At nearest and next-nearest neighbor separations, the vanadium defect states hybridize strongly, and local moments quench. At larger separations, a clear preference for a ferromagnetic alignment emerges.
Estimating the Curie temperature $T_C$ is beyond the scope of the current paper due to the sensitivity of the coupling parameter $J$ to the carrier doping level, quantitative limits of the DFT+U method, and the presence of sulfur vacancies (see Section S13, Supporting Information). In some cases, extracting an effective $J$ is not straightforward as one of the spin states is not a local minimum. A very large supercell is also necessary to cover longer range interactions and describe the geometrical frustration of antiferromagnetic interactions. Many-body interactions such as vanadium triples need to be considered carefully as well. These concerns require a separate computation/theory paper. In this paper, instead of finding a precise $T_C$, we provide a semiquantitative discussion about the energy scale of interactions. Considering that ferromagnetic order in the experimental system will benefit from additional neighbor interactions beyond the single pair alignment in the current model, the $\approx 10 \text{ meV}$ stabilization for parallel alignment over anti-parallel is reasonably consistent with the observed room-temperature ferromagnetic order. The origin of the non-monotonicity in $M_S$ and $H_C$ around 150–200 K, where the saturation magnetization drops just as the coercivity more rapidly increases, is less clear. Speculatively, it might reflect domains of competing anti-ferromagnetic order that reduce $M_S$ while increasing $H_C$ through exchange pinning. In spite of all these caveats on the correspondence of computation to experiment, the appearance of similar quenching phenomena even in calculations without spin-orbit coupling (Table S1, Figure S13, Supporting Information) lends substantial confidence to the quenching mechanism.

Spin-momentum locking at the valence band top of WS$_2$ interacts closely with the local moments of the vanadium dopants, yielding band crossings or avoided crossings respectively if the spin of a dopant band and the valence band extremum has the same or opposite direction, with the circumstance switching between these two cases as one proceeds from K to $-K$, which is visible in Figure S12, Supporting Information. For single vanadium (Figure 3a), the two types of spin-polarized band edges shift in opposite directions in energy such that the valence band maximum (VBM) has an energy difference of 12.6 meV between valleys while the original split in conduction band minima (CBM) collapses in one valley become nearly spin-degenerate. Increasing the doping level to two vanadium atoms in the supercell (with ferromagnetic order), the VBM difference increases to 19.5 meV, and the CBM shows a uniform spin direction. These band edge offsets arising from the ferromagnetic polarization correspond to an effective external magnetic field on the order of $\approx 100 \ T$.[41]

A statistical analysis of the expected fraction of magnetically polarized dopants in a random alloy (Section S11, Supporting Information) suggests that the optimal doping level to obtain maximum saturation magnetization is intermediate between the 2 and 8 at% samples examined experimentally. First-principles calculations indicate that the nearest-neighbor separation of a pair of vanadium dopants is 50–90 meV more stable than larger separations, meaning a modest energetic preference for dopant aggregation as is evidenced experimentally by occasional stripes at higher vanadium concentrations. This suggests that the choice of the precursor may be an important factor in atomic-scale dopant structure optimization (Figure S1, Supporting Information). The rare occurrence of V aggregation in moderately doped samples suggests that synthesis is far from equilibrium, presumably a key to incorporating V dopants into the WS$_2$ host in the first place. Finally, the observed spatial correlation between sulfur vacancies and vanadium dopants (Figure S3, Supporting Information) is consistent with computational results that sulfur vacancies bind to single vanadium by $\approx 0.04 \text{ eV}$ and a nearest-neighbor V–V dimer by $\approx 0.71 \text{ eV}$.

This study successfully develops a universal, scalable, and controllable synthesis route for V-doped WS$_2$ atomic layers as a dilute magnetic semiconductor, with intrinsic ferromagnetic ordering at room temperature. As the vanadium concentration increases, V-doped WS$_2$ monolayers exhibit a reduction of the optical bandgap and the emergence of p-type transport, reaching ambipolarity. The vanadium doping induces inherent ferromagnetic ordering at room temperature, with the strongest ferromagnetic signal for the moderately doped (2 at%) sample among grown samples. The non-monotonicity of the magnetization as a function of doping level is explained by a combination of atomic resolution TEM imaging and DFT calculations, which show how hybridization between dopant defect states quenches the magnetic moment. An effective Zeeman shift corresponding to $\approx 100 \ T$ is observed in the calculated band structure. Such dilute magnetic semiconductors based on magnetic-element-doped transition metal dichalcogenides exhibit great promise as future

### Table 1. Net moments and energies for vanadium dopant pairs. The energies are relative to that of the pair at the nearest distance. Dopant pairs are labeled by their separation in lattice coordinates and colored in reference to the supercell in Figure 3. Systems were initialized with either parallel or anti-parallel local moments around the two dopants. Moments after self-consistent iterations are perpendicular to the plane except for the (0, 2) separation, which is 76° away from this axis. For the closest and next-closest dopant separations (●), the lowest energy state examined has no spatially resolvable spin texture. “—” means that both parallel and anti-parallel initial spin textures converge to the same self-consistent state.

<table>
<thead>
<tr>
<th>Dopant pair in lattice coordinates</th>
<th>Pair separation [Å]</th>
<th>Energy of the most stable spin texture [meV]</th>
<th>Net magnetic moment $\mu_B$</th>
<th>Energy of competing spin texture [meV]</th>
<th>Moment of competing spin texture $\mu_B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-1, 0$</td>
<td>3.19</td>
<td>0 (●)</td>
<td>0.00</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>$1, 1$</td>
<td>5.52</td>
<td>63.9 (●)</td>
<td>0.00</td>
<td>67.5 (●)</td>
<td>0.14</td>
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<tr>
<td>$0, 2$</td>
<td>6.38</td>
<td>48.2 (●)</td>
<td>0.93</td>
<td>—</td>
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</tr>
<tr>
<td>$1, 2$</td>
<td>8.44</td>
<td>71.1 (●)</td>
<td>1.18</td>
<td>84.4 (●)</td>
<td>0.00</td>
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<tr>
<td>$0, 3$</td>
<td>9.57</td>
<td>86.8 (●)</td>
<td>1.22</td>
<td>95.4 (●)</td>
<td>0.03</td>
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<tr>
<td>$2, 2$</td>
<td>11.02</td>
<td>88.8 (●)</td>
<td>1.24</td>
<td>93.6 (●)</td>
<td>0.07</td>
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<tr>
<td>$1, 3$</td>
<td>11.49</td>
<td>86.5 (●)</td>
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<td>93.9 (●)</td>
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spintronic/valleytronic devices, with novel magneto–electric and magneto–optical responses. Furthermore, they constitute a new set of atomically thin layers that could be integrated into multifunctional van der Waals heterostructures.

**Experimental Section**

**Synthesis of Pristine and Vanadium-Doped WS$_2$ Monolayers:** 0.05 g ammonium metavangstate (NH$_4$H$_2$W$_{12}$O$_{40}$xH$_2$O, AMT) and 0.2 g sodium cholate (C$_{24}$H$_{39}$NaO$_5$·H$_2$O) powders were dissolved in 10 mL water to form a tungsten precursor solution. 0.05 g vanadyl sulfate (VO(SO$_4$)$_2$) powder was dissolved in 10 mL deionized water to form vanadium precursor (1 x 10$^{-2}$ mol L$^{-1}$). The W and V solutions with different concentrations were controlled to form solution-based cation precursors. The precursor solution was drop-casted onto an SiO$_2$/Si substrate, followed by spin-coating for 1 min with 3000 rpm. The film sulfidation process was carried out at atmospheric pressure in a quartz reactor tube (1" inner diameter) with sulfur powder (400 mg) heated upstream at low temperature (220 °C, heated up using a heating tape), and the cation precursor spin-coated on the SiO$_2$/Si substrates, at the high temperature (825 °C) zone. Ultrasil purity argon was employed as the carrier gas. The furnace was then allowed to cool to room temperature naturally after 15 min synthesis.

**Materials Characterization:** A Renishaw InVia microscope with a Coherent Innova 70C argon-krypton laser at the excitation of 488 nm and a LabRAM HR evolution (Horiba) equipped with a 532 nm laser were used for acquiring the Raman and photoluminescence (PL) spectra using a backscattering configuration and an 180° detector. X-ray diffraction (XRD) was taken with PANAnalytical Empyrean X-ray diffractometer with a Cu source. X-ray photoelectron spectroscopy (XPS) experiments were performed using a Physical Electronics VersaProbe II instrument. The binding energy axis was calibrated using sputter cleaned Cu foil (Cu 2p$_{3/2}$ = 932.7 eV, Cu 2p$_{3/2}$ = 75.1 eV). UV–vis absorption spectra were recorded using an UV–vis spectrophotometer (V-750). The total conductance of the Al/doped WS$_2$/SiO$_2$ interface was measured via time-domain thermoreflectance. The specific analyses can be found elsewhere.[512] In the implementation, the 808.5 nm output of a Ti:Sapphire oscillator was spectrally separated into high-energy pump and low-energy probe paths. The pump was electro-optically modulated at 8.4 MHz and created a frequency-dependent heating event at the sample surface. The probe was mechanically delayed in time, and the change in reflectivity due to two pump-induced heating events (i.e., thermoreflectivity) was monitored as a function of delay time. Both the pump and probe were concentrically focused through a 10x objective, yielding 1×2× diameters of 14 and 11 μm, respectively. The data were fit to the radially symmetric heat diffusion equation to extract the conductances at the Al/doped WS$_2$/SiO$_2$ interface.

**DFT Calculations:** Spin-orbit-coupled DFT calculations were implemented in the Vienna Ab-initio Simulation Package (VASP).[534–538] A 7 x 7 supercell of WS$_2$ was tested with different V doping levels. The z-axis cell dimension was 15 Å to isolate a layer from its periodic images. The exchange-correlation was treated under GGA PBE approximation[46] with PAW method.[47] The energy cutoff in all calculations was 700 eV, and the k-point sampling was set as 4 x 4 x 1 centered at Γ. The WS$_2$ unit cell lattice constant calculated as 3.188 Å matched with previous work[48] and was fixed for doped WS$_2$ since the doping level was not high enough to change the lattice constant significantly. The residual force after relaxation was smaller than 0.01 eV Å$^{-1}$ for all atoms. All visualizations were done with VESTA[59] and pymatgen.[10]

As the experimental distribution of vanadium dopants was irregular and covered a wide range of pairwise separations, the system was not modeled with a regular array of dopants at uniform mutual separations but instead examined a dopant pair hosted within a large 7 x 7 supercell across a range of separations so that their interactions could be elucidated on a pairwise basis. Two dopants within this supercell were closer to each other than to any periodic replicas. Both ferromagnetic (parallel) and antiferromagnetic (anti-parallel) initial spin configurations were considered for two vanadium dopants in a supercell, with the system also being able to converge self-consistently into an unpolarized state in the case of moment quenching.

**Electronic Device Fabrication:** Pristine and V-doped WS$_2$ triangles were transferred from the growth substrate (Si/SiO$_2$) to a 50 nm thick and atomic layer deposition grown Al$_2$O$_3$ substrate with Pt/TiN/p$^+$ Si as the back-gate electrode. All FETs were fabricated with a channel length of 1 μm with 40 nm Ni/30 nm Au as the source/drain contact electrodes defined using a standard electron-beam lithography process.

**Thermal Transport Measurements:** To examine the thermal boundary conductances ($h_{bd}$) of devices contingent on the use of doped WS$_2$, a nominally 80 nm Al film was deposited via electron beam evaporation. The total conductance of the Al/doped WS$_2$/SiO$_2$ interface was measured via time-domain thermoreflectance. The specific analyses can be found elsewhere.[512] In the implementation, the 808.5 nm output of a Ti:Sapphire oscillator was spectrally separated into high-energy pump and low-energy probe paths. The pump was electro-optically modulated at 8.4 MHz and created a frequency-dependent heating event at the sample surface. The probe was mechanically delayed in time, and the change in reflectivity due to two pump-induced heating events (i.e., thermoreflectivity) was monitored as a function of delay time. Both the pump and probe were concentrically focused through a 10x objective, yielding 1×2× diameters of 14 and 11 μm, respectively. The data were fit to the radially symmetric heat diffusion equation to extract the conductances at the Al/doped WS$_2$/SiO$_2$ interface.

**Supporting Information**

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

**Acknowledgements**

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

F.Z., T.Z., and M.L. conducted the synthesis experiments. F.Z. collected the TEM data; and F.Z., K.F., L.M., and N.A. performed TEM data analyses. Optical characterization was performed by F.Z. and T.Z.; B.Z., Y.W., and V.C. carried out with the computational calculations. Y.T.H.P., V.K., V.O.J., and M.H.P. performed magnetic measurements and analyzed the magnetic data. A.S., R.P., and S.D. integrated the devices and conducted electronic transport measurements. D.H.O. and P.E.H. carried out with thermal transport measurements. K.F., Y.L., and A.L.E. helped with data analysis, discussion, and guidance about doping precursors and experimental parameters. M.T., M.H.P., and V.C. supervised the work. The manuscript was written through contributions from all and all authors have given approval to the final version of the manuscript.

Keywords

2D ferromagnets, dilute magnetic semiconductors, room-temperature ferromagnetism, tungsten disulfide, vanadium doping

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