Light-Controlled Room Temperature Ferromagnetism in Vanadium-Doped Tungsten Disulfide Semiconducting Monolayers

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1. Introduction

Dilute magnetic semiconductors (DMSs) offer an alternative path towards the realization of cutting-edge spintronic devices.[1–6] The use of light to control magnetism in these semiconductors has the added advantage of being able to control both charge and spin simultaneously, which supports the demands of multifunctional (smart) sensing devices, information storage, and quantum computing technologies.[7–10] So far, it has been reported that a carrier-mediated ferromagnetic interaction between the Mn ions in p-type (In,Mn)As/GaSb semiconductor has been enhanced by the illumination of light through the generation of excess holes in the (In,Mn)As layer.[7] Unfortunately, this effect is limited to temperatures (<50 K), well below ambient temperature, while the most important technological applications are required to operate at room temperature (RT).[11,12]
It has recently been theoretically and experimentally shown that the introduction of a magnetic transition metal atom into semiconducting 2D transition metal dichalcogenide (TMD), such as V-doped WSe$_2$ (V-WSe$_2$) and V-doped WS$_2$ (V-WS$_2$), permits long-range ferromagnetic order that can be induced at RT.\cite{13-16} Currently, the Ruderman–Kittel–Kasuya–Yosida (RKKY) mechanism is believed to be responsible for the long-range ferromagnetic order in these TMD systems, where free holes are the medium that support the interaction between V atoms.\cite{13–15} In particular, we have recently demonstrated that p-type V-WS$_2$ monolayers have strong and tunable RT ferromagnetism.\cite{16} By replacing W, having six valence electrons, with V, having five valence electrons, an electron deficiency is created in V-WS$_2$ that eventually becomes a p-type dominant semiconductor. Unlike diamagnetic pristine WS$_2$ (Figure S1a, Supporting Information), ferromagnetism is enhanced with V doping in monolayers of V-WS$_2$, which is found to be optimized at $\approx$2 at% V (Figure S1b, Supporting Information).\cite{16} The long-range ferromagnetic order in this optimally magnetic 2D DMS allows us to modify its magneto-electronic properties with external stimuli, like a magnetic field, an electric field, or as we show in this letter, with light. Photoluminescence (PL) reveals that, even after doping, strong photoluminescence is still present (Figure S1c, Supporting Information). These observations lead us to propose that the ferromagnetism in the V-WS$_2$ monolayer can be mediated by illumination with a laser of appropriate energy, that is, above the optical gap (see Figure 1). Electrons from photogenerated electron–hole pairs may be captured by the V atoms, thus creating an imbalance in the carrier population (i.e., the generation of excess holes) such that the ferromagnetism of the monolayer is modified.

While p-type (In,Mn)As/GaSb showed light-mediated ferromagnetism at temperatures below 50 K,\cite{7} Probing magnetism in atomically thin materials is extremely challenging when compared to bulk systems.\cite{17,18} While techniques such as vibrating sample magnetometry (VSM) and superconducting quantum interference devices (SQUID) are capable of measuring the magnetic moment of these materials,\cite{19,20} measurements in real time while applying external stimuli are not easily achievable. Methods that require electrical contact, such as transport measurements, have an extra layer of difficulty since monolayer films often do not span the surface continuously and the size of the electrical contacts are large compared to the surface area.\cite{21} Optical methods based on the magneto-optical Kerr effect (MOKE) have proven very successful in thin films and have had a measure of success with 2D materials such as CrI$_3$.\cite{17} However, local heating from high laser powers causes thermal instability, which is a significant source of noise in these measurements. Therefore, cryogenic temperatures are needed to reduce thermal and mechanical measurement noise, but for RT, this is not an option. Despite this, optical measurements remain a powerful practice that yield crucial insight into the spin dynamics of atomically thin materials.\cite{22,23} The shortcomings of these techniques motivate the development of a new approach to measure 2D magnetization in real time as external stimuli are applied.

In order to probe the light-induced magnetization of an atomically thin magnetic film such as V-WS$_2$ monolayers, we propose a new technique utilizing our recently developed magneto-LC resonance sensor with ultrahigh magnetic field sensitivity (pT regime).\cite{24,25} The sensing element of this device is a magnetic microwire wound into a coil driven with a frequency of 118 MHz, which is near the coil’s LC resonance (Figure S2, Supporting Information). The impedance of the coil is then measured with an impedance analyzer, from which we extract novel applications in spintronic devices that have not been yet realized.

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the reactance of the coil. The setup is depicted in Figure 1. The film is placed at the core of the coil and the reactance of the coil is measured. Theoretically, the reactance of the coil is written as:

\[ X_{\text{coil}} = \frac{\omega L (1 - \omega^2 L C_{\text{par}}) - C_{\text{par}} \omega R_{\text{par}}}{(1 - \omega^2 L C_{\text{par}})^2 + (\omega C_{\text{par}} R_{\text{par}})^2} \]  

(1)

where \( \omega \) is the angular frequency. Since the film is ferromagnetic, it will modify the relative permeability of the space within the coil, thus changing the magnetic flux through the coil and consequently the reactance of the coil. As the microwire itself is ferromagnetic, the magnetization of the film will also lead to a change in the effective permeability of the microwire. Therefore, the reactance of the sensor depends on this effective permeability, \( X = X(\mu_{\text{eff}}) \). Changes in the permeability of the film upon light illumination will influence the effective permeability of the coil, which can be accessed through the change in its reactance, \( \Delta X = X(\mu_{\text{eff, laser on}}) - X(\mu_{\text{eff, laser off}}) \).

3. Results and Discussion

Due to the presence of defects and edge effects in WS\(_2\) monolayers, a small magnetic moment may be present in these “pristine” films. Therefore, we first examine whether photogenerated electron-hole pairs have any effect on the magnetization of the film. Figure 2a shows the signal obtained from a WS\(_2\) monolayer film with and without laser excitation. In this figure, a small change in the signal, likely due to thermal fluctuations in the film, can be seen. Next, we performed the measurement on the 2 at\% V-doped WS\(_2\) monolayer (Figure 2b), where we observe a large change in reactance (magnetic permeability or magnetization) upon illumination with the same laser. The excitation wavelength is \( \lambda = 650 \text{ nm} \) (\( h \nu = 1.91 \text{ eV} \)) with the power of 5.1 mW cm\(^{-2}\) at the sample surface. The laser was operated for several minutes during these measurements, which were carried out at RT. Since prolonged laser exposure may heat up the magnetic coil and hence change the magnetic properties, coil heating effects due to laser exposure have also been studied. As shown in Figure S3, Supporting Information, the dot laser, which covers only a small area of the coil (0.11 cm\(^2\)), has a negligible effect on the magnetism of the coil. These findings indicate that the observed enhancement of the magnetization/permeability from the experimental setup, that is, illuminated V-WS\(_2\) monolayer (Figure 2b) within the coil, is not due to a laser/sample heating effect but originates from carrier-mediated ferromagnetism, similar to the case of a p-type (In,Mn)As/GaSb semiconductor. As shown in Figure S4, Supporting Information, the dot laser, which covers only a small area of the sample would affect the permeability/magnetization from the experimental setup, that is, illuminated V-WS\(_2\) monolayer (Figure 2b) within the coil, is not due to a laser/sample heating effect but originates from carrier-mediated ferromagnetism, similar to the case of a p-type (In,Mn)As/GaSb semiconductor. As shown in Figure S5, Supporting Information. It is worth mentioning that upon light illumination with comparable laser powers, the magnetization change in the illuminated (In,Mn)As/GaSb film (\( \lambda = 685 \text{ nm}, 6 \text{ mW cm}^{-2} \)) was only observed below 50 K, while enhanced magnetization is achieved at RT for the illuminated V-WS\(_2\) monolayer (\( \lambda = 650 \text{ nm}, 5.1 \text{ mW cm}^{-2} \)). This striking difference makes this atomically thin ferromagnetic semiconductor a promising candidate for use in light-controlled spintronics and other multifunctional nanodevices. The semiconducting nature of V-WS\(_2\) facilitates incorporation to current silicon-based technology and provides a platform for optoelectronic phenomena; combined with its FM properties we obtain a unique way to manipulate the spin states in the material by illuminating it with light.

It is of interest to determine how an increase in illumination area of the sample would affect the permeability/magnetization, so we illuminated the film with two different 650 nm lasers, labeled “dot” and “target” lasers, with 0.11 and 0.41 cm\(^2\) coverage areas, respectively. The two lasers have different spot sizes, but the same light intensity of 4.2 mW cm\(^{-2}\). In Figure 3a,b it is observed that increasing the coverage area increases the change in magnetization. Finally, we sought to determine the light intensity dependence of the change in magnetization. In Figure 3c,d we demonstrate this for the two 650 nm lasers, both of which show a similar trend. Initially, we see a sharp increase in magnetization with increasing light intensity, and at higher laser intensities the change in magnetization begins...
to saturate. Since higher laser powers bring about a considerable heating effect, which may damage the coil or the sample, in this study we restricted the laser intensity below 6 mW cm$^{-2}$ for light-induced magnetization experiments. Photon concentration was calculated as a function of laser intensity, for each laser, using the relation $E = n \hbar \nu$ (where $E$ is the energy and $\nu$ is the frequency, $n$ is the photon-generated carrier concentration, and $\hbar$ is Planck’s constant), assuming 2% absorption in the V-WS$_2$ layer[26] and assuming that only 1% of the electrons/holes created do not immediately recombine. We observe that by increasing the illumination area, a smaller photon concentration, compared to the “dot” laser, is required to achieve a change on magnetization. A smaller photon concentration is also necessary to achieve the saturation feature; using the “dot” laser a concentration of $\approx 3.1 \times 10^{12}$ photons/cm$^2$/s is necessary to approach saturation, while using the “target” laser saturation starts around $\approx 2.7 \times 10^{12}$ photons/cm$^2$/s. This points to a long range cumulative effect in which the enhanced magnetic moments in the illuminated area may couple with the moments surrounding it.

To elaborate on these findings, the band structures of the WS$_2$ and V-WS$_2$ monolayers were investigated by density functional theory (DFT) calculations and the results are shown in Figure S6, Supporting Information. In order to establish the change of the WS$_2$ band structure by a V atom, the valence band edge was used as the reference. The V atom induces two empty doping states: one is below the conduction band and the other is on the top of the valence band. These two doping bands are flat and localized, implying a weak interaction with W and S atoms. The latter atom plays the role of electron acceptor, where it accepts thermally excited electrons from the valence band and manifests the p-type characteristic of the V-WS$_2$ monolayer.[16] In addition, there are two strong hybridization bands between the V and W atoms, which induce $\approx 1\mu_B$ of the magnetic moment in the V-WS$_2$ monolayer. The energy of the ferromagnetic state is 0.24 eV, which is lower than that of the no-spin state. Similar hybridization bands have been reported in monolayers of V-doped WSe$_2$.[13,14] which also shows a lowering of the energy of a FM state with Curie temperature above RT. This points to V-WS$_2$ monolayers as another candidate for light mediated magnetism.

Generally, two main factors have been suggested to influence the magnetic moment of a DMS upon light illumination:[27–31] i) the population of the free excited carries in the conduction and valence bands;[29] and ii) the localized excited carriers trapped by magnetic doping states in the band gap of the host material.[25] The former effect is usually dominant in lightly doped samples (e.g., 1.1% of Mn in GaAs[29]), whereas the latter becomes significant in heavily doped samples (e.g., 10% of Mn in CdSe and HgTe).[27,28] in which the dopants form a new band

![Figure 3](image-url)
inside the gap of the host. Due to its single layer limit and V concentration of ≈2%, both free and localized excited carriers are expected to mediate the magnetization in the V-WS2 film when illuminated with an appropriate power laser. Figure 4a shows the distribution of the magnetic moments under varying carrier populations. Increasing the concentration of holes results in a more robust magnetic moment across the lattice, where W atoms far from the V site show an enhanced magnetic moment. Since long-range ferromagnetic interactions are mediated by free holes in V-WS2 and similar systems,[13] it is not surprising that optically injecting hole-carriers will lead to enhanced ferromagnetism. The evolution of the band structure is calculated under different carrier concentrations, as shown in Figure 4b. We find that while the Fermi level is shifted deeper inside the valence band with hole injection, it is shifted toward the conduction band edge with electron injection. The evolution of the exchange energy is also calculated, the result of which is presented in Figure 4c. The exchange energy becomes stronger with increasing hole concentration, so the magnetic moment must also change as the carrier concentration varies. Indeed, we observe an increase in the magnetic moment with increasing hole concentration (Figure 4c). This is consistent with our experimental findings (Figure 3c,d), where a higher light intensity resulted in a larger hole concentration and consequently an increased magnetic moment. At large hole concentrations the magnetic moment saturates, confirming the saturation feature we observed experimentally (Figure 3c,d). This is also consistent with another report,[32] in which hole injection into a V-WS2 monolayer increases the magnetic moment which also saturates at high hole concentrations due to screening of the charge carriers. We should note that both electrons and holes are populated in experiments whereas the simulation considers the separated effect for each type of carriers. It is interesting to consider how different V doping concentrations in WS2 will influence the light mediated magnetism effect. We have established that it is necessary for both magnetic and semiconducting properties to coexist, therefore higher doping concentrations are unlikely to show this effect since their semiconducting qualities are strongly suppressed. On the other hand, lower doping concentrations are very interesting to study, since semiconducting properties are enhanced but the magnetic moment is much smaller compared to the optimal 2 at%. It is possible that these samples may provide wider tunability of magnetism compared to the 2 at% samples. Further studies are necessary to understand the relation between doping concentration and light enhanced magnetism.

In summary, we have demonstrated that magnetism can be tuned with light in V-WS2 monolayers, by varying the light intensity or by changing the illumination area. As the film is illuminated, the absorbed photons generate electron–hole pairs and the electrons are captured by the electron deficient V sites, which generate an imbalance in the carrier population and, hence, a change in magnetic moment. We have shown that the carrier concentration can be tuned by changing the light intensity, allowing control over the magnetic moment of the film. Density functional calculations confirm that the magnetic moment of the V-WS2 monolayer can be enhanced by increasing the hole concentration. All of this is achieved at RT which has been a key obstacle to applied 2D spintronics. These findings highlight the potential for cutting-edge
applications of 2D DMS and other atomically thin magnetic semiconductors.

4. Experimental Section

Sample Synthesis and Characterization: The WS₂ and 2 at% V-doped WS₂ monolayer films were synthesized using a reliable single-step powder vaporization method. Scanning transmission electron microscopy (STEM)–energy dispersive X-ray spectroscopy (EDS) of the samples were performed by a FEI Talos F200X microscope with a SuperX EDS detector operating at 200 kV. PL spectra of the samples were recorded with a Coherent Innova 70C argon-krypton laser at 532 nm. Details of the samples’ synthesis and characterization have been reported previously.[10] High-angle annular dark-field (HAADF)–scanning transmission electron microscopy (STEM) images of pristine WS₂ and 2 at% V-WS₂ monolayers are shown in Figure S1a,b, Supporting Information, insets. This was used to confirm the V doping concentration of our samples.

Magnetic and Light-Induced Measurements: Magnetization versus magnetic field (M–H) measurements were carried out at RT in a Physical Property Measurement System (PPMS) from Quantum Design. The initial spin state was induced along the z direction and the total energy was optimized with 10 × 10 supercell, which corresponds to 1.6% of V atoms, was used with a 3 × 3 × 1 k-point grid. The structures of WS₂ and V-WS₂ were optimized until the convergence of force and energy were smaller than 0.0001 Ha and 0.001 (Ha bohr−1), respectively, without including spin-orbit coupling. The initial spin state was induced along z direction and the total energy was optimized with 10 × 10 Ha of convergence including spin-orbit coupling. To describe the strong correlation of the d electrons, the GGA+U method with U = 3 was used for the vanadium atoms.

Supporting Information

Supporting Information is available from the Wiley Online Library or from other authors. M.H.P. directed the research. V.O.J. and M.H.P. conceived the initial concept. V.O.J. performed light-induced magnetization experiments and analyzed the data with inputs from T.E. and B.M. Y. H.T.N. and V.K. performed magnetic measurements and analyzed the magnetic data. F.Z. and M.L. synthesized the films and characterized the structural and optical properties. D.L.D. performed the computational calculations. V.O.J. wrote the manuscript with inputs from other authors. M.H.P. directed the research.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

V.O.J. and M.H.P. conceived the initial concept. V.O.J. performed light-controlled magnetization experiments and analyzed the data with inputs from T.E. and B.M. Y.H.T.N. and V.K. performed magnetic measurements and analyzed the magnetic data. F.Z. and M.L. synthesized the films and characterized the structural and optical properties. D.L.D. performed the computational calculations. V.O.J. wrote the manuscript with inputs from other authors. M.H.P. directed the research.

Data Availability Statement

Research data are not shared.

Keywords

DFT, spintronics, ferromagnetism, semiconductors, sensors, transition metal dichalcogenides

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