Doping isolated one-dimensional antiferromagnetic semiconductor Vanadium tetrasulfide (VS$_4$) nanowires with carriers induces half-metallicity

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1 Figures

Figure S1: The high symmetry points of the first Brillouin zone for the band structure of the VS$_4$ bulk phase. This figure was created from http://materials.duke.edu/awrapper.html

Figure S2: Ferromagnetic (FM) and antiferromagnetic (AFM) states for (a) VS$_4$ bulk phases and (b) isolated VS$_4$ nanowires (NWs).

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Figure S3: (a) Phonon spectra and (b) total potential energy of NWs as a function of simulation time for the VS$_4$ bulk phase (conventional cell) and the isolated VS$_4$ NW by using \textit{ab-initio} molecular dynamics (300K). The inset shows the corresponding structure after the simulation for 9 ps.
\( T_N \) is defined as a maximum on the temperature dependent specific heat \( C_V \) curve,

\[
C_V = \frac{<E^2> - <E>^2}{T^2}
\]  

(1)

Where \( T \) is the temperature and \( E \) is the energy.

Figure S4: Specific heat calculated for the isolated VS\(_4\) NW with respect to the temperature.

Figure S5: (a) The schematic plot of transformation between the HMAF and AFM states under gate voltages of \( V_G < 0 \), \( V_G = 0 \) and \( V_G > 0 \), respectively. (b) The schematic plot of AFM spintronics device based on the isolated VS\(_4\) NW, together with the \( I - V_G \) relationship under the gate voltage. The switching of the spin current can be manipulated by gate voltages.
To evaluate the stability of the isolated VS\textsubscript{4} NW inside BN nanotubes of different sizes, the binding energy ($E_b$) for the unit cell is calculated as:

\[
E_b = E_{\text{total}} - E_{\text{NW}} - E_{\text{nanotube}}
\]

where $E_{\text{total}}$ stand for the total energies of the isolated VS\textsubscript{4} NW inside the ($m$, $m$) BN nanotube. $E_{\text{NW}}$ and $E_{\text{nanotube}}$ are the energies of the isolated VS\textsubscript{4} NW and the ($m$, $m$) BN nanotube. $m = 6, 7, 8$ and $9$, which are the direction of the vector.

Figure S6: (a) Binding energies $E_b$ of the isolated VS\textsubscript{4} NW inside various BN nanotubes are presented. (b) Spin polarization density of the nanocable. (c) The partial density of states of the nanocable at PBE + U level ($U = 3$). The gray shadow is the total density of states.
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Quasi one-dimensional (1D) vanadium tetrasulfide (VS₄) nanowires (NWs) are synthetic semiconductors which combine with each other through Van der Waals interactions to form bulk phases. However, the properties of these individual nanowires remain unknown. Nevertheless, our calculations of their stability indicate that VS₄ NWs can be separated from their bulk structures. Accordingly, we theoretically investigated the geometrical, electronic, and magnetic properties of bulk phase and isolated VS₄ NWs. Our results indicate that both bulk phase and isolated VS₄ NWs are semiconductors with band gaps of 2.24 and 2.64 eV, respectively, and that they prefer the antiferromagnetic (AFM) ground state based on DFT calculations. These calculations also suggested that isolated VS₄ NWs show half-metallic antiferromagnetism upon electron and hole doping because carrier doping splits the spin degeneracy to induce local spin polarisation. As a result, spin polarisation currents in isolated VS₄ NWs can be manipulated with locally applied gate voltage. Therefore, these 1D AFM materials have a high potential for advancing both fundamental research and spintronic applications because they are more resistant to magnetic perturbation than their 1D ferromagnetic counterparts.

1 Introduction

Research aimed at improving the performance of electronics has pushed the physical limits of these devices. However, further advances in information technology require developing alternatives to electronics 1–4. For this purpose, many new methodologies have been proposed, such as molecular electronics, nanoelectronics 5–7, spintronics 8–10, and valleytronics 11–13. Among these new developments, spintronics stands out for its compatibility with conventional electronics. Consequently, spintronics can be used to broaden the possibilities of conventional electronics 8,14. In contrast to electronics, in which electrical charges are manipulated to induce a current, spintronics aims to exploit the two spin polarisation of unpaired electrons to create a spin-polarised current, thus enhancing the performance of semiconductors, in terms of electrical conductivity and transport of binary information. Accordingly, spintronics enables us to improve the design of logic components, including memory boards and transistors 15,16.

Spintronics is based on the ability to control the intrinsic spin of the electron, which was first discovered 17 in ferromagnetic (FM) metals but has since been proved viable in other classes of FM materials, most notably in half-metallic ferromagnets 18. Unlike paramagnetic metals, FM metals have a different density of states for the two spins. Consequently, the latter have a neat magnetic momentum and exhibit a spin-polarised current at adequately low temperatures 17,19. Similarly, half-metallic ferromagnets have two spin channels (which guarantees a neat magnetic momentum), but only one of spin channels displays metallic character, whilst the other has a band-gap 20. FM materials with non-null magnetic polarisation have stray fields, which can induce interference between different elements, thus limiting the down-scaling of devices 20,21–23. For this reason, most studies have focused on antiferromagnetic (AFM) materials.

AFM materials have been primarily manipulated for spintronics by inducing half-metallic antiferromagnetism (HMAF) 24–26. HMAF materials were first proposed by H. van Leuken and R. A. de Groot 27, who showed that many Heusler compounds, more specifically CrMnSb, could be fully spin-compensated half-metallic materials. Heusler compounds are ternary compounds with two different magnetic centres at different sub-lattices, thus decoupling electronic from magnetic properties. Recently, these theoretical predictions have been experimentally confirmed in similar compounds, namely Half-Heusler: Mn₂RuX₆Ga and Mn₂Pt₆Ga alloys 28,29. These studies have mostly focused on bulk phases, while low-dimensional materials remain unexplored because their lower dimensionality requires different strategies to remove the spin degeneracy, either applying a bias voltage or using organic-inorganic materials, for example. Gong et al. showed that the bilayer 2H-VSe₂ becomes HMAF when applying proper electric fields 30, while Ai et al. designed a two-dimensional (2D) metal-organic HMAF (CoFePz), which paved the way to the development of organic HMAF 31. Among its potential applications of HMAF, spin field-effect transistors (FET) 30,32 stand out for their ability to control the spin current 33. Moreover, carrier doping enables us to manipulate the electronic and magnetic properties of low-dimensional materials, which are theoretically and experimentally accessible 34–36. Therefore, carrier doping is an effective strategy to control the spin current in low-dimensional magnetic materials. Few materials, for example NbSe₃ 37, can be isolated as true one-dimensional (1D) materials, which are joined in quasi-1D materials through Van der Waals forces. Some 1D compounds have been proposed as candidates for spintronic applications, including metal trihydride molecular nanowires 38 (NWs), 1D metal benzenetetramine coordination polymers 39, Co-dithiolene molecular wires 40, transition metal tribromide NWs 41, transition metal trichalcogenide NWs 42, and transition metal chalcogenide NWs 43. These materials have been theoretically investigated for their electronic and magnetic properties, but not as much for their spintronic properties, due to the lack of experimental reports.

The Vanadium tetrasulfide (VS₄) is found in nature as a mineral, and was discovered in 1906 44. Its linear chain-like structure is composed of two S₂⁻ moieties connecting V⁺⁺ centers. The different chains are bound together by Van der Waals forces to form nano-rods in quasi-1D compounds 45–47. VS₄ NWs have been studied for other applications, such as batteries, capacitors, and photocatalysts 48–51. The possible oxidation states of Vanadium ions induce different magnetic properties that have been demonstrated in different materials: Vanadium dichalcogenides (VX₂, X = S, Se) monolayers, MXenes (VX₂, X = C, N) 52,53, and Haeckelite VS₂ 54. However, the geometric, electronic and magnetic properties of VS₄ as a 1D NW are not clearly understood yet. This study aimed to assess how the magnetic properties of VS₄ are affected by its dimensionality and how VS₄ can be used for spintronic applications.

2 Methods

All calculations were performed within the density functional theory (DFT) as implemented in the Vienna ab-initio simulation package (VASP) 55,56. The structural properties have been determined using the Perdew Burke Ernzerhof (PBE) version of the generalised gradient approximation (GGA) 57, while the electronic and mag-
netic properties have been calculated with the hybrid HSE06\textsuperscript{58} functional. The dispersion forces have been included in the DFT-D3 method\textsuperscript{39}. An energy cut-off of 500 eV was employed to define the plane-wave basis sets, considering the wavefunction converged for total energy variations below $10^{-8}$ eV. The same parameters have been used for both PBE and HSE06 calculations. The structures were fully optimised to minimise the forces below $-0.02$ eV/Å. For both geometry relaxation and electronic properties in the Brillouin zone, this material was sampled using a Monkhorst–Pack \( k \)-point mesh of \( 5 \times 5 \times 5 \) and \( 1 \times 1 \times 5 \) for the bulk phase and isolated NWs, respectively. The isolated NW was simulated including a vacuum space of 15 Å in the \( x \) and \( y \) directions. The high symmetry points of the first Brillouin zone for the band structure of the VS\textsubscript{4} bulk phase are shown in Figure S1. We calculated phonon spectra using the finite differences method, as implemented in VASP. Post-processing and analysis have been performed using the software PhonoPy\textsuperscript{60}. For this task, we have used tighter convergence criteria, more specifically $10^{-8}$ eV for the wavefunction and $-0.001$ eV/Å for the forces. In addition, we address the kinetic stability of the structures performing a set of ab-initio molecular dynamics (AIMD) calculations, as implemented in VASP. These calculations have been completed using the Nose\textsuperscript{61} algorithm in the NVT ensemble at room temperature (300 K) for the duration of 9 ps. The formation energy (\( E_{\text{form}} \)) of each VS\textsubscript{4} unit formula is calculated as:

\[
E_{\text{form}} = E(\text{VS}_4)/n - E(V) - 4E(S) \tag{1}
\]

where \( E(\text{VS}_4) \) is the total energy of the VS\textsubscript{4} bulk phase or the isolated NW containing \( n \) unit formula, while \( E(V) \) and \( E(S) \) are the single atom energies from the bulk phases of cubic \( V \) and \( S \).\textsuperscript{62} Carrier doping was simulated by removing or adding electrons from the system with the homogeneous background charge to keep charge neutrality.

The magnetic properties have been analysed using a collinear model for simplicity. We considered FM and AFM states to calculate the preferred magnetic ground state structures of NW (Figure S2). To account for these effects, we used the Monte Carlo method to solve a simple Ising model\textsuperscript{63}:

\[
H = -\sum_{ij} J_{\text{intra}} M_i \cdot M_j \tag{2}
\]

where \( J_{\text{intra}} \) is the nearest-neighbour exchange-coupling parameter of intra-NWs and \( M \) is the spin magnetic moment per chemical formula. The \( J_{\text{intra}} \) value can be calculated by the exchange energy (\( E_{\text{ex}} = E_{\text{FM}} - E_{\text{AFM}} \)). The exchange coupling parameters have been used to calculate the Néel temperature (\( T_N \)) performing a Monte Carlo simulation on a 150 1D chain lattice with $10^{-5}$ steps for each temperature using the open-source software ALPS\textsuperscript{64}.

3 Results

3.1 Structural analysis

Figure 1a shows the primitive cell of VS\textsubscript{4} bulk phase (C2/c #15) in which each V atom is coordinated with eight S atoms forming four dimers. From the bulk phase, we have built the isolated NW unite cell (Figure 1b). The geometric data on the VS\textsubscript{4} bulk phase and on the isolated VS\textsubscript{4} NW are summarised in Table 1. Both the bulk and isolate NW phases of VS\textsubscript{4} are dynamically stable, as shown by the absence of an imaginary frequency in the phonon dispersion (figure S3a). The stability of these materials is also confirmed by AIMD simulation at room temperature, thus suggesting that individual NWs can be isolated (figure S3b).

Figure 1 (a) Primitive cell of the VS\textsubscript{4} bulk phase from two directions, that is, along (001) direction, on the left panel, and along (111) direction, on the right panel. (b) Isolated VS\textsubscript{4} NW. The repeating units are marked by the black lines.

3.2 Electronic and magnetic properties

We now focus on the electronic and magnetic properties of the VS\textsubscript{4} bulk phase and the isolated NW. Both the bulk phase and the isolated NW have an AFM ground state, and their stability is given by \( E_{\text{ex}} \), as outlined in Table 1. The magnetic momentum of V is similar in the bulk phase and in the isolated NW, with values of 1.15 and 1.17 \( \mu_B \), respectively. The bulk phase has an indirect band gap of 2.24 eV between the \( Y \) and \( Z \) points of the Brillouin Zone. The calculated electronic band gap largely overestimated the experimental optical band gap of VS\textsubscript{4} films (approximately 1.35 eV)\textsuperscript{45} which is usually observed in similar cases\textsuperscript{65–67}. This difference of band gaps could be attributed to the experimental conditions in which the optoelectronic properties depend on the sulfur partial pressure of synthesis and on the morphology of the sample\textsuperscript{45}. Most importantly, our model for the bulk phase assumes perfect crystallinity, which is unlikely to occur in the family of low-dimensional compounds.

The spin density (Figure 2b) shows the AFM G-type motif of the material\textsuperscript{68}. The valence band maximum (VBM) and the conduction band minimum (CBM) of the VS\textsubscript{4} bulk phase are formed by the overlap of S 2p orbitals with V 3d levels as shown in the partial density of states (PDOS) (Figure 2a). The V 3d orbitals split into a non-degenerate \( d_{xz} \) orbital and into two 2-fold degenerate \( d_{xy} \) and \( d_{yz}/d_{xz} \) orbitals. The local V 3d orbitals can induce an antiparallel spin arrangement on neighboring V via the double-exchange mechanism (Figure 2b). The VBM and CBM of the VS\textsubscript{4} bulk phase derive from the S 2p orbitals and V \( d_{xz} + d_{yz} \) orbitals (Figure 2c), in line with its PDOS.

The electronic structure of the isolated VS\textsubscript{4} NW reflects the lower dimensionality of the material, which induces a larger band-gap of 2.65 eV with a higher density of state (Figure 3a). The lack of interactions between different chains of VS\textsubscript{4} affects the nature of the bond. We can observe a larger contribution of VBM and CBM in the isolated VS\textsubscript{4} NW due to the S 2p states (Figure 3a). The VBM and the CBM of the V 3d orbitals of the isolated NW are different from those of the bulk phase. \( V d_{xz}/d_{yz} \) orbitals of the isolated VS\textsubscript{4} NW contribute to the VBM and CBM.

To further understand the change of electronic and magnetic properties between the VS\textsubscript{4} bulk phase and the isolated VS\textsubscript{4} NW, we plotted the electron localisation function (ELF) for the VS\textsubscript{4} bulk
Table 1 Structural parameters, electronic and magnetic properties of the bulk phase and isolated NW for VS$_4$. $E_{\text{form}}$ is the formation energy (eV). $L_{\text{S-dimer}}$ is the S dimer bond-length (Å). $L_{\text{V-V}}$ is the V-V bond length. $L_{\text{V-S}}$ is the V-S bond length. $E_g$ is the band gap. $E_{\text{ex}}$ is the exchange energy (eV). $\mu_B$ is the local magnetic moment of the V atoms.

| configurations | $E_{\text{form}}$ | $L_{\text{S-dimer}}$ | $L_{\text{V-V}}$ | $L_{\text{V-S}}$ | $E_g$ | $E_{\text{ex}}$ | $\mu_B$ |
|----------------|------------------|---------------------|-----------------|-----------------|------|----------------|--------|
| Bulk           | -2.59            | 2.03                | 2.77, 3.20      | 2.54, 2.39      | 2.24 | 0.961          | 1.15   |
| NW             | -2.06            | 2.03                | 2.77, 3.22      | 2.55, 2.39      | 2.65 | 0.948          | 1.17   |

Figure 2 (a) Band structures and PDOS and (b) spin polarised density of the VS$_4$ bulk phase, where spin-up and spin-down densities are shown in green and blue, respectively. (c) The valence band maximum (VBM) and the conduction band minimum (CBM) are shown in green. The isosurface is 0.005 e·Bohr$^{-3}$.

Figure 3 (a) Band structures and PDOS and (b) spin polarised density of the isolated VS$_4$ NW, where spin-up and spin-down densities are shown in green and blue, respectively. (c) VBM and CBM are shown in green. The isosurface is 0.005 e·Bohr$^{-3}$.

phase and for the isolated VS$_4$ NW (Figure 4). The ELF of S atoms in the VS$_4$ bulk phase is more localised than that of the isolated VS$_4$ NW due to Van der Waals interactions of NWs in the VS$_4$ bulk phase. Moreover, VS$_4$ NW has distinct characteristics of localised $d$ electrons of V atoms: more relatively itinerant V $d$ electrons in the isolated VS$_4$ NW than that in the VS$_4$ bulk phase. Itinerant $d$ electrons of V atoms can induce spin polarisation of neighbouring S atoms, e.g., an anti-parallel spin arrangement via a double-exchange mechanism. While the PDOS of $d$ orbitals of V atoms differ between the VS$_4$ bulk phase and the isolated VS$_4$ NW. This difference in electronic properties does not affect the AFM character of these materials with a double-exchange mechanism, which also depends on the V atoms.

3.3 Macroscopic magnetic properties

Magnetic order is more susceptible to temperature in materials with lower dimensionality$^{69}$ due to the lower number of interactions. As a result, magnetic fluctuation, which destroys the order, is more likely to happen in low-dimensional materials than in three-dimensional materials. DFT calculations enable us to directly determine the magnitude of such interactions, but the value of $T_N$ cannot be simulated since it emerges as a statistic average. Nevertheless, we employed exchange interactions (obtained ab-initio) to describe the Temperature dependence of our system using an Ising model solved with a Monte Carlo approach. For the isolated VS$_4$ NW, we obtained a $T_N$ of 210 K (Figure S4).

3.4 HMAF based FET

Previous studies have shown that carrier doping manipulates spin currents through the voltage gate$^{34,36}$. We analysed doping concentrations of 0.1, 0.3, and 0.5 electron (and hole) per unit cell of the isolated VS$_4$ NW (equivalent to 0.83, 2.50, and 4.17 $\times 10^6$cm$^{-1}$). The isolated VS$_4$ NW with carrier doping is HMAF, exhibiting complete spin-polarisation around the Fermi level (Figure 5). Because carrier doping shifts the Fermi level and spin polarisation of S and V atoms, PDOS for the isolated VS$_4$ NW with carrier doping show metallic states in the spin-up channel and semiconductor states (band gaps are over 2 eV) in the spin-down channel. These effects are detected as small perturbations of the small AFM, which can be considered preserved for practical applications. The main contributions to the metallic states around the Fermi level derive from V $d_{xy}/d_{x^2−y^2}$ orbitals and S $2p$ orbitals.

The magnetic momentum of V atoms is affected by carrier doping, and the magnetic momentum of each S atom is very small. When doping with 0.5 electron, the magnetic momentum of one V atom (marked 4) increases from 1.17 to 1.46 $\mu_B$ (Figure 6a). When doping with 0.5 hole, the magnetic momentum of one V atom decreases from 1.17 to 1.08 $\mu_B$ (Figure 6a). The isolated VS$_4$ NW retains its AFM state. The $E_{\text{ex}}$ is rapidly decreased under hole doping and increased under electron doping. Our results indicate that the AFM state of the isolated VS$_4$ NW is stable when injected with a low concentration of carriers. Moreover, we visualise the partial charge density (around Fermi level) of the isolated VS$_4$ NW at 0.5 electron and hole doping (Figure 6c). The participation of V and S orbitals also differs: formed by V $d_{xy}/d_{x^2−y^2}$ and
Our results indicate that the isolated VS$_4$ NW is an AFM semiconductor with a high $T_N$ of 210 K. This is an important result because ferromagnetic 1D materials, such as the quasi-1D organic ferromagnet ($T_C$ 0.56 K)$^{70}$, the tribromide NW of the half-metal VBr$_3$ ($T_C$ 80 K)$^{41}$, the trihydride molecular NW of the half-metal CoH$_3$ ($T_C$ 98 K)$^{38}$ and transition metal dihalide nanowires CuCl$_2$ ($T_C$ 14 K)$^{71}$, usually have much lower Curie temperatures ($T_C$). Furthermore, $T_N$ of the isolated VS$_4$ NW is well above the temperature of liquid nitrogen (77K) and is also higher than that of different 2D antiferromagnets, such as bilayer CrI$_3$ ($T_N$ 45 K)$^{72}$ or polyradical nanosheets (42.5 K)$^{73}$. An even better prediction $T_N$ could be obtained for the isolated VS$_4$ NW, but this prediction is a promising starting point for further experimental characterisation.

Moreover, the AFM character and large band gap of the isolated NWs prevent spin-polarised currents in VS$_4$. The local spin polarisation of ideal antiferromagnetism, which in turn allows spin-polarised currents, could be a technological solution.$^{36}$ Here, we consider a model where carriers are injected into our materials, a process commonly known as carrier doping in the literature, in analogy to chemical doping$^{74}$. A method for carrier doping consists of building a spin FET device (scheme shown in Figure S5) where the charges are injected by applying different gate voltages.

The influence of the environment on the transport properties and electrical contacts of NWs limit their applications. Therefore, protecting NWs by preserving the electronic and magnetic properties of the isolated VS$_4$ NW should be tested in spintronic applications. Nanotubes may protect NWs by preventing oxidation and maintaining electronic and magnetic properties unchanged$^{37,41}$, and nanocables, composed of insulating outer sheaths and a NW core, may guide the design of the models. Here, a designed nanocable on the isolated VS$_4$ NW and a $1 \times 5 \times 5$ (8, 8) boron nitride (BN) zigzag nanotube is constructed (Figure S6), where the lattice mismatch is approximately 4.5%. The isolated VS$_4$ NW has a slight strain. The distance between the NW and the wall of the BN nanotube is approximately 3.57 Å. The electronic and magnetic properties of the VS$_4$–BN nanocable is similar to that of the isolated VS$_4$ NW. Therefore, the spin polarisation of the nanocable mainly derives from the inner NW, while the outer nanotube has...
a negligible effect on the nanocable. Such a hybrid structure may also enable interesting applications in spintronics.

5 Summary
We presented herein a new strategy in VS$_4$ NWs towards AFM spintronics. The VS$_4$ NWs are bound together by Van der Waals forces to form nano-rods in quasi-1D compounds in experiments. In this context, the geometric, electronic and magnetic properties of the VS$_4$ bulk phase and isolated VS$_4$ NW were analysed. First, we investigated the stability of the VS$_4$ bulk phase and isolate VS$_4$ NW by formation energy, AIMD and phonon spectra. After confirming the stability of VS$_4$ NWs, we investigated the electronic and magnetic properties of VS$_4$ NWs. The magnetic ground states of the isolated VS$_4$ NW is AFM with a high Néel temperature (210 K). Detecting and manipulating the spin of AFM materials remains a major challenge due to spin degeneracy in the band structure. Nevertheless, carrier doping can separate the spin degeneracy to induce local spin polarisation. The isolated VS$_4$ NW represent the half-metallic antiferromagnet resulting from carrier doping, which can be achieved with gate voltages. Our results indicate that the isolated VS$_4$ NW is a promising 1D material for AFM spintronic applications. Carrier doping induces a rigid shift in the Fermi level into the valence or conduction bands, resulting in a complete spin-polarisation of carriers, which can be induced by applying a gate voltage. Thus, spin polarisation currents can be manipulated by applying a gate voltage, with a high potential for spintronic applications. We further considered the protection of NWs. BN nanotubes can provide protection by preventing oxidation and by preserving the electronic and magnetic properties of VS$_4$ NWs. Our results open up new opportunities for applying 1D NWs in AFM spintronics by inducing half-metallic antiferromagnetism.

6 Conflicts of interest
There are no conflicts to declare.

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