

## Supplementary Information

Proposal for Spin Tunnel Diode via Doping Engineering on VS<sub>2</sub> Monolayer  
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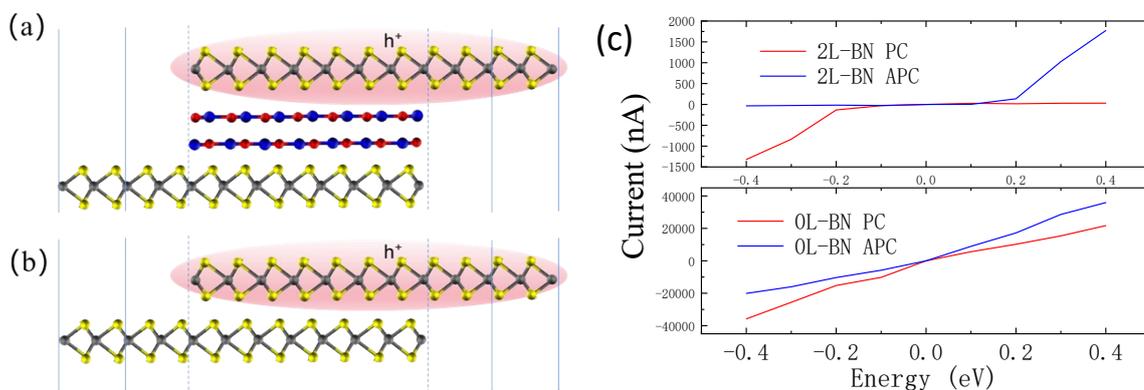
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### The effect of h-BN atomic layers

We investigated the effect of the insulating barrier on the spin transport properties of our STD with the right VS<sub>2</sub> monolayer by p-type doping of 0.05 e/atom. As shown in Fig 1(c), the STD structure with two atomic layers of h-BN demonstrated a desired PN diode behavior that the current increased exponentially with the incremental bias voltage after the threshold voltage of 0.2 V, both for PC or APC magnetic configurations of bilateral electrodes. Nevertheless, the almost linear current with the disappeared diode behavior is observed for the STD structure by removing the h-BN layers .

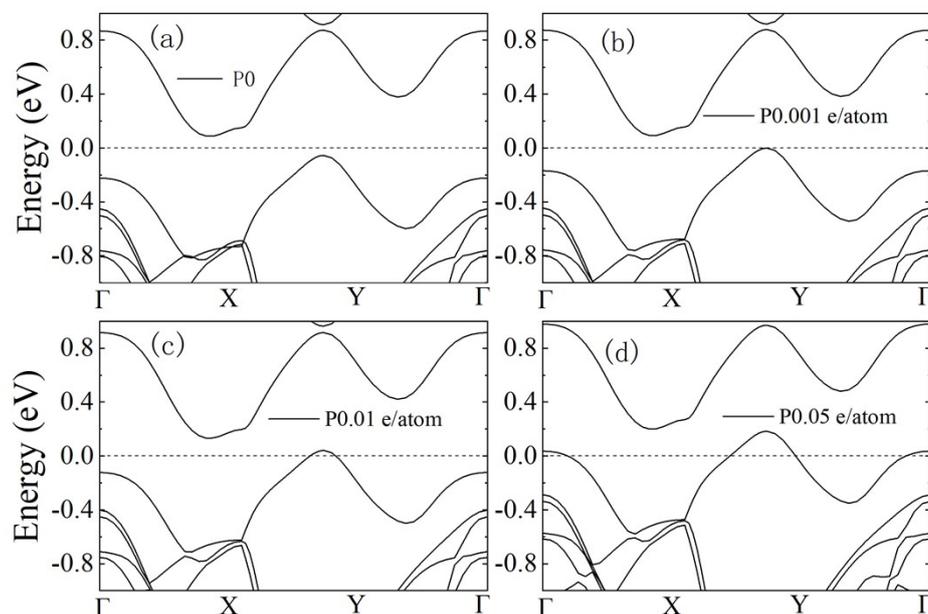


**Figure 1.** The device configuration for the spin tunnel diode (a) with the insulating tunnel barrier of two atomic layers of h-BN, (b) without h-BN. (c) The current-voltage characteristic.

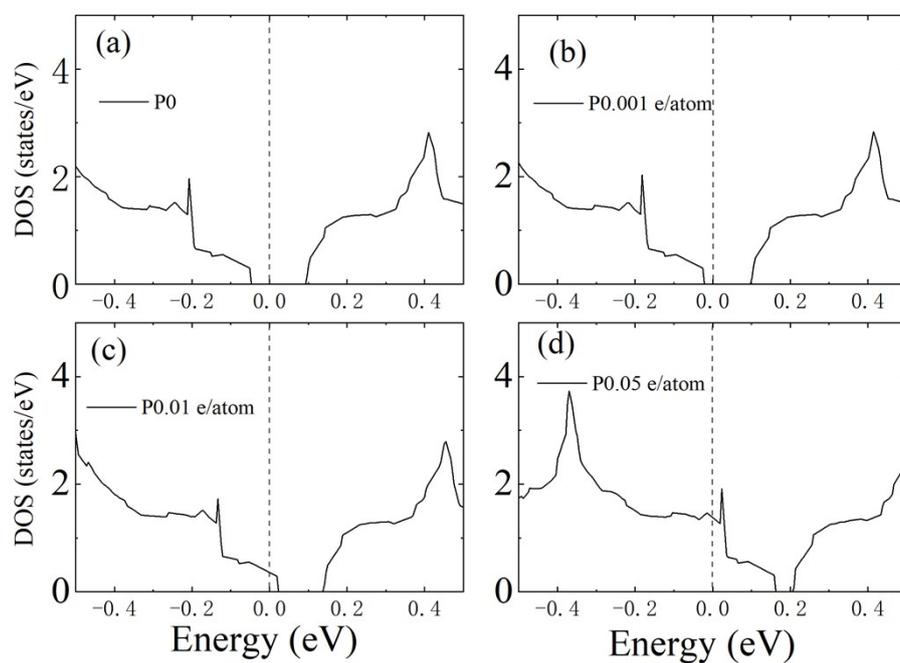
### The band structure and density of states with SOC

The stable FM can be ascribed to the spin-orbit coupling (SOC) of vanadium in VS<sub>2</sub> monolayer and thus the Mermin-Wagner theory does not function. We explore the band structure and DOS for VS<sub>2</sub> monolayer with p-type doping concentration of 0, 0.001 e/atom, 0.01 e/atom and 0.05 e/atom by GGA-PBE functional with SOC. The results are shown in Fig. 2. The spin-up and spin-down electrons could not be distinguished due to the mixture of their spin orientations by SOC effect. As compared to the band structure and DOS of VS<sub>2</sub> monolayer without including SOC, the band gap is reduced from 0.32 eV to 0.14 eV due to

the energy levels splitting. Secondly, we have also observed a phase transition from semiconductor to metal with doping density larger than 0.01e/atom.



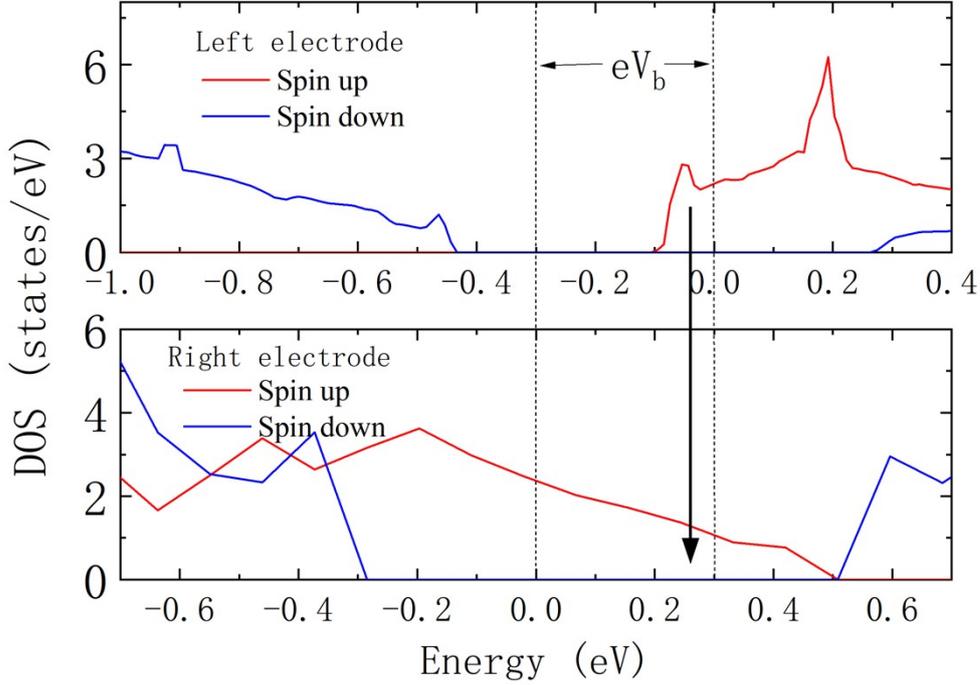
**Figure 2.** The band structure of VS<sub>2</sub> monolayer with p-type doping concentration of 0, 0.001 e/atom, 0.01 e/atom and 0.05 e/atom with SOC.



**Figure 3.** The DOS of VS<sub>2</sub> monolayer with p-type doping concentration of 0, 0.001 e/atom, 0.01 e/atom and 0.05 e/atom with SOC.

### The origin of new electronic states in transmission spectra

We noticed that there have been new electronic states which cross the Fermi level in Fig. 3c at positive bias. We made the DOS picture of left and right electrode, respectively, and bias voltage  $V_b=0.3$  V. It is noteworthy that there is a tunnel channel between the two electrodes, as denoted by the vertically downward black arrow in the figure below, indicating a non-zero transmission coefficient at this region.



**Figure 4.** The DOS of left and right electrodes at the bias voltage of 0.3 V. The vertically downward black arrow denotes the tunnel channel.

#### Mathematical fitting of Curie temperature

We probe the Curie temperature ( $T_C$ ) by mathematical linearly fitting of  $T_C$  curve by increasing the q-points to infinite. As shown in fig. 5b in the manuscript, we have demonstrated the dependence of Curie temperature on the reciprocal of  $N_q$ .  $T_C$  value is most accurate in the PRA approach with an average of the spin wave energy by increasing the sampled q-points to infinite in first Brillouin zone. By performing a linear fit, it is therefore possible to extrapolate the result to the limit of infinitely many q-points by evaluating the fit  $1/N_q=0$ , as shown in fig. 5b. The linear mathematical fitting equation are:

$$T_C(P0) = \frac{1376.7}{N_q} + 273.9$$

$$T_C(P0.001e/atom) = \frac{1376.7}{N_q} + 273.3$$

$$T_C(P0.01e/atom) = \frac{1369}{N_q} + 272$$

$$T_C(P0.05e/atom) = \frac{978}{N_q} + 208.2$$

Therefore indicating a Curie temperature value of 273.9 K, 273.3 K, 272 K and 208.2 K, for doping by 0, 0.001 e/atom, 0.01 e/atom and 0.05 e/atom systems, respectively.

### The origin of transmission dependence with the applied voltage

In the non-equilibrium Green's function (NEGF) method, the transmission coefficient may also be obtained from the retarded Green's function  $G(\varepsilon)$  and the broadening function of the left and right electrodes,  $\Gamma^L(\varepsilon)$  and  $\Gamma^R(\varepsilon)$ , by using:

$$T(\varepsilon) = G(\varepsilon)\Gamma^L(\varepsilon)G^\dagger(\varepsilon)\Gamma^R(\varepsilon)$$

The broadening function of the left electrode is given in terms of the electrode self energy complex matrix  $\Sigma(\varepsilon)$ , which described the effect of the electrode states on the electronic structure of the central region:

$$\Gamma^L = \frac{1}{i}(\Sigma^L - (\Sigma^L)^\dagger)$$
$$\Gamma^R = \frac{1}{i}(\Sigma^R - (\Sigma^R)^\dagger)$$

where  $\dagger$  is the conjugate transpose symbol, makes the self energy matrix in the following transformation:

$$(\Sigma^{L(R)})^\dagger = [(\Sigma^{L(R)})^*]^T$$

Actually, the electrode self energy  $\Sigma(\varepsilon)$  is related with the applied voltage since they describe the effect of the electrode states on the electronic structure of the central region. The applied voltage strongly affected the electron density matrix of left electrode  $D^L$  and right electrode  $D^R$  by tuning the Fermi function  $f$ :

$$D^L = \int \rho^L(\varepsilon) f\left(\frac{\varepsilon - \mu_L}{k_B T_L}\right) d\varepsilon$$
$$D^R = \int \rho^R(\varepsilon) f\left(\frac{\varepsilon - \mu_R}{k_B T_R}\right) d\varepsilon$$

where  $\rho^L(\varepsilon)$  and  $\rho^R(\varepsilon)$  denoted the density of states (DOS) matrix and the Fermi function  $f$  is affected by the applied voltage  $V_b$  by tuning the Fermi energy level  $\mu_L$  and  $\mu_R$ :

$$eV_b = \mu_R - \mu_L$$

### Calculation of transmission coefficient from the electronic Hamiltonian

In the non-equilibrium Green's function (NEGF) method, the transmission coefficient may also be obtained from the retarded Green's function  $G(\varepsilon)$  and the broadening function of the left and right electrodes,  $\Gamma^L(\varepsilon)$  and  $\Gamma^R(\varepsilon)$ , by using:

$$T(\varepsilon) = G(\varepsilon)\Gamma^L(\varepsilon)G^\dagger(\varepsilon)\Gamma^R(\varepsilon)$$

The broadening function of the left electrode is given in terms of the electrode self energy complex matrix  $\Sigma(\varepsilon)$ , which described the effect of the electrode states on the electronic structure of the central region:

$$\Gamma^L = \frac{1}{i}(\Sigma^L - (\Sigma^L)^\dagger)$$
$$\Gamma^R = \frac{1}{i}(\Sigma^R - (\Sigma^R)^\dagger)$$

The Green's function is only required for the central region and can be calculated from the Hamiltonian of the central region by adding the electrode self energies:

$$G(\varepsilon) = [(\varepsilon + i\delta_+)S - H - \Sigma^L(\varepsilon) - \Sigma^R(\varepsilon)]^{-1}$$

where  $\delta_+$  is an infinitesimal positive number. S and H are the overlap and Hamiltonian matrices, respectively, of the entire system. The bias voltage definitely change the electronic coupling of the VS<sub>2</sub> and BN layers, correspondingly change the electrode self energy  $\Sigma(\varepsilon)$ .