Magnetic Properties of NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$ Monolayers

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The recent demonstration of MoSi$_2$N$_4$ and its exceptional stability to air, water, acid, and heat has generated intense interest in this family of two-dimensional (2D) materials. Among these materials, monolayers of NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$ are semiconducting, easy-plane ferromagnets with negligible in-plane magnetic anisotropy. They thus satisfy a necessary condition for exhibiting a dissipationless spin superfluid mode. The Curie temperatures of monolayer VSi$_2$P$_4$ and VSi$_2$N$_4$ are determined to be above room temperature based on Monte Carlo and density functional theory calculations. The magnetic moments of VSi$_2$N$_4$ can be switched from in-plane to out-of-plane by applying tensile biaxial strain or electron doping.

Two dimensional (2D) layered materials with transition metals have been of interest for many decades due to the correlated phenomena and multiple polymorphs and phases that they can exhibit such as charge density waves,$^{1}$ superconductivity,$^{2}$ and magnetism.$^{3}$ The ability to exfoliate or grow single monolayers renewed the interest in these materials for possible electronic and optoelectronic applications$^{4,5}$ by both traditional transistor type devices$^{6,7}$ and by exploiting external control of their phase transitions.$^8$ The relatively recent demonstration of magnetism in single monolayer of CrI$_3$$^9$ and bilayers of Cr$_2$Ge$_2$Te$_5$,$^{10}$ has spurred intense experimental and theoretical activity to find other 2D magnetic materials with higher transition temperatures.$^{11-18}$

The most recent addition to the family of 2D materials are the transition metal silicon nitrides, phosphides, and arsenides with the chemical formulas MA$_2$Z$_4$, where M is the transition metal, A ∈ {Si, Ge}, and Z ∈ {N, P, As}$^{19}$ High quality multilayers and monolayers of MoSi$_2$N$_4$ were grown using chemical vapor deposition, and what was particularly notable was their stability to air, water, acid, and heat that was unprecedented among transition metal 2D materials.$^{19}$ This rather mundane property is highly desirable for manufacturing applications. While BN encapsulation is an effective solution for stabilizing reactive 2D materials for laboratory experiments,$^{20}$ it is less than ideal for manufacturing. Only MoSi$_2$N$_4$ was experimentally characterized in detail, WS$_2$Si$_2$N$_4$ was also grown, and 12 materials were simulated with density functional theory (DFT) and found to be stable. Among these 12, two of the nitrides, VSi$_2$N$_4$ and NbSi$_2$N$_4$, were identified as magnetic.

This work motivated immediate follow-on theoretical investigations of this material family both determining properties of the materials and extending the list of stable materials.$^{21-29}$ The most extensive theoretical survey found 32 thermodynamically and dynamically stable compounds of the form MA$_2$Z$_4$.$^{24}$ Of these, 6 were magnetic: VSi$_2$N$_4$, VSi$_2$P$_4$, NbSi$_2$N$_4$, VGe$_2$N$_4$, VGe$_2$P$_4$, and TaGe$_2$N$_4$. Based on the formation enthalpies, a Si based compound MSi$_2$Z$_4$ is approximately 3 times more stable than its equivalent Ge based compound MGe$_2$Z$_4$, and a silicon nitride compound MSi$_2$N$_4$ is also approximately 3 times more stable than its equivalent silicon phosphide compound MSi$_2$P$_4$.

Other recent works investigated MSi$_2$Z$_4$ bilayers (M = Ti, Cr, Mo; Z = N, P) for their sensitivity to vertical strain,$^{27,28}$ and MoSi$_2$N$_4$ and WSi$_2$N$_4$ for their sensitivity to biaxial strain and vertical applied electric field.$^{27}$ It was found that vertical strain can cause an insulator to metal transition;$^{28}$ biaxial strain can lead to an indirect to direct bandgap transition;$^{27}$ and that an applied electric field can result in an insulator to metal transition.$^{27}$ A theoretical investigation of 2D-2D contacts to monolayer MoSi$_2$N$_4$ using NbS$_2$ and graphene found ultralow p-type Schottky barriers with NbS$_2$ contacts and approximately equal n-type and p-type Schottky barriers with graphene contacts.$^{22}$ The graphene Schottly barriers were shown to be tunable with an applied vertical electric field.

In this letter, we theoretically investigate the magnetic properties of the three Si based magnetic materials: VSi$_2$N$_4$, VSi$_2$P$_4$, NbSi$_2$N$_4$. Values of the exchange constants and magnetic anisotropy energies are determined, and the Curie temperatures are calculated. The Curie temperatures of VSi$_2$N$_4$ and VSi$_2$P$_4$ are near or at room temperature and above, depending on the model used, whereas the Curie temperature of NbSi$_2$N$_4$ is low. Therefore, the primarily focus will be on the two vanadium

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FIG. 1. Structures and magnetic orientations of α$_1$-NbSi$_2$N$_4$, α$_1$-VSi$_2$N$_4$, and α$_1$-VSi$_2$P$_4$. Blue atoms are Si, white atoms are N, and pink atoms are P. The directions of the magnetic moments are shown on the transition metals Nb and V. All materials are easy-plane, semiconducting ferromagnets.
compounds. The magnetic anisotropy energies of VSi$_2$N$_4$ and VSi$_2$P$_4$ are calculated as a function of uniaxial and biaxial strain and electron and hole doping.

These materials can exist in a variety of hexagonal phases. Ref. [24] calculated the formation energies of 30 different phases and found that the lowest energy phase of NbSi$_2$N$_4$ and VSi$_2$N$_4$ was the $\alpha_1$ phase, which is the same phase as the lowest energy phase of MoSi$_2$N$_4$. The formation energy of the next higher energy phase ($\delta_2$) of NbSi$_2$N$_4$ was 13 meV per atom higher than the $\alpha_1$ phase, and the formation energy of the next higher energy phase ($\beta_2$) of VSi$_2$N$_4$ was 6 meV per atom higher than the $\alpha_1$ phase. For VSi$_2$P$_4$, the lowest energy phase was $\delta_4$ with a formation energy 0.3 meV below that of $\alpha_1$. Since the formation energy of the $\alpha_1$ phase of VSi$_2$P$_4$ is so close to that of the $\delta_4$ phase, and since the $\delta_4$ phase has been previously investigated$^{24}$, we will consider the $\alpha_1$ phase of all 3 materials: NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$.

We note that the piezoelectric and magnetic properties of the $\alpha_1$ phase of VSi$_2$P$_4$ have recently been investigated with DFT using the generalized gradient approximation (GGA).$^{21}$

DFT combined with Monte Carlo (MC) calculations are applied to determine the electronic and magnetic properties of these materials. The magnetic anisotropy energy (MAE) and exchange energy is evaluated from DFT calculations implemented in the Vienna ab initio simulation package (VASP).$^{30}$ The electron-core interactions are described by the projected augmented wave (PAW) potentials.$^{31}$ Electronic structure is calculated using three different functionals: GGA as parameterized by Perdew-Burke-Ernzerhof (PBE), PBE plus the Hubbard U correction (PBE+U), and the Heyd-Scuseria-Ernzerhof hybrid functional (HSE06).$^{32-35}$ The PBE+U calculation includes a Hubbard U correction term $U_{\text{eff}} = 1$ eV for the V atom where $U_{\text{eff}} = U - J$.$^{36,37}$ For calculations of the MAE, spin orbit coupling (SOC) must also be included. All plots shown for the MAE are the results from PBE(SOC)+U calculations. The cutoff energies for expanding the plane wave basis are 600 eV. Integration over the Brillouin zone uses a Monkhorst–Pack scheme with a $\Gamma$-centered $16 \times 16 \times 1$ k-point grid, an energy broadening parameter of 50 meV, and the total energy is converged to $10^{-6}$ eV.$^{38}$ Structures are relaxed until the forces are less than 0.001 eV/Å. A vacuum spacing of 20 Å is used in the direction normal to the 2D monolayer to eliminate the interactions from periodic images. The calculated lattice constants for NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$ are 2.96 Å, 2.88 Å, and 3.47 Å, respectively, and they are very close to previous reported results.$^{19,21}$

Uniaxial strain is applied along the $x$ axis corresponding to lattice vector $a_1$. The applied strain is evaluated using $\epsilon = (a - a_0)/a_0 \times 100\%$, where $a$ and $a_0$ are the lattice parameters of the strained and unstrained monolayer. Biaxial strain is applied by uniformly varying both in-plane lattice constants. For each strain, the atomic positions are relaxed using the DFT parameters described above. Spin-polarized self consistent calculations are performed with the relaxed structure for each strain to obtain the charge density. Using the charge densities, total energies are calculated in the presence of spin orbit coupling (SOC) for in-plane ($E_{\perp}$) and out-of-plane ($E_{\parallel}$) magnetization to find the magnetic anisotropy energy (MAE). The MAE is defined as $E_{\text{MAE}} = E_{\perp} - E_{\parallel}$. Positive magnetic anisotropy indicates that in-plane magnetization is favored. The values provided in meV are per magnetic atom, which for these 3 materials under consideration, are also per unit cell.

To investigate the transition temperatures, a nearest neighbor Heisenberg type Hamiltonian with magnetic anisotropy and long range dipole-dipole interactions is constructed,

$$H = -\frac{1}{2} J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + k_u \sum_i (S_i^z)^2 + H_{dd},$$

where $S_i$ is the spin for magnetic atom $i$, $\langle i,j \rangle$ are the indices of nearest neighbor magnetic atoms, $k_u > 0$ is the easy-plane magnetic anisotropy energy per magnetic atom, and $H_{dd}$ is the dipole-dipole interaction. The exchange energies ($J$) of these materials are calculated from the total energy differences between the antiferromagnetic ($E_{\text{AFM}}$) and ferromagnetic ($E_{\text{FM}}$) states.$^{38-41}$ With six nearest neighbours in the monolayer of 2D hexagonal lattice, the nearest-neighbor exchange energy is given by$^{40-42}$ $J = (E_{\text{AFM}} - E_{\text{FM}})/12$. In these monolayer materials all of the magnetic atoms are in the same plane. The second neighbor exchange energies were previously determined for VSi$_2$P$_4$ and found to be negligible compared to the nearest neighbor energies,$^{24}$ and, therefore, they are ignored. The Curie temperatures are determined from Monte Carlo (MC) calculations as implemented in the VAMPIRE software package using a $10 \times 10$ supercell.$^{43,44}$ The MC calculations incorporate the magnetic anisotropy energies, the magnetic moments, the exchange energies, lattice constants, and atomic positions determined from the DFT calculations, and they also include the long range dipolar interactions. This MC approach has been used to determine the Curie temperatures in other monolayer 2D materials such as VSi$_2$P$_4$, Vi$_3$, CrSBr, and CrSeBr.$^{24,45,46}$ We note that the Curie temperatures in other monolayer 2D materials determined from the MC approach have been shown to compare well to temperatures determined from renormalized spin wave theory (RSWT). For example, the MC/RSWT predicted Curie temperatures for monolayer CrSBr and CrSeBr were 168/150 K and 150/152 K, respectively.$^{46}$

The structures and the ground state orientations of the magnetic moments of NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$ are shown in Fig. 1, and the spin-resolved band structure for VSi$_2$N$_4$ and VSi$_2$P$_4$, calculated with the HSE06 and PBE+U functionals, are shown in Fig. 2. The band structure for all three materials calculated with PBE, PBE+U, and HSE06 are shown in the Fig. S3 of the Supplement, and the d-orbital resolved bands of VSi$_2$N$_4$ and VSi$_2$P$_4$ are shown in Fig. S2.
are spin-polarized, $d_{z^2}$ orbital bands centered on the transition metal atoms. Near K, these bands transition to $d_{x^2-y^2}$ at $\Gamma$, and it transitions to $d_{z^2}$ at K. The electronic structures from the HSE06 calculations show the following properties. In VSi$_2$N$_4$ and VSi$_2$P$_4$, the higher spin-up conduction band at $\Gamma$ crosses the lower spin-down band near K where the orbital composition of the two bands switch, $d_{z^2} \leftrightarrow d_{x^2-y^2}$. VSi$_2$N$_4$ is direct gap (0.78 eV) at K with a $d_{x^2-y^2}$ spin-up valence band and a $d_{z^2}$ spin-up conduction band. VSi$_2$P$_4$ is indirect gap (0.84 eV) with the $d_{z^2}$ spin-up valence band edge at $\Gamma$ and a mixed $d_{z^2} + d_{x^2-y^2}$ spin-down conduction band edge at M (the spin-up band at K is 40 meV higher). NbSi$_2$N$_4$ is indirect gap (0.54 eV) with the spin-up $d_{z^2}$ valence band at $\Gamma$ and a mixed orbital $d_{z^2} + d_{x^2-y^2}$ spin-down conduction band near M. The lower valence bands are primarily $p$-orbital bands which come from the N and P atoms. At the PBE level of theory, all 3 materials are semi-metals. Adding the Hubbard U correction creates a small gap at the Fermi level for VSi$_2$N$_4$ and VSi$_2$P$_4$. Adding a percentage of exact exchange with the HSE06 functional increases the gap substantially. We note that the HSE06 calculations match those reported in the Supplement of Ref. [24].

The equilibrium NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$ monolayers are easy-plane, semi-conducting ferromagnets. The calculated equilibrium magnetic moments, exchange energies, magnetic anisotropy energies, and Curie temperatures are shown in Table I for different levels of theory. The magnetic moments are comparable to those from prior studies. The positive MAE values indicate in-plane alignment of the magnetic moments. Within the energy resolution of our calculations (1 µeV), the total energy is independent of the angle of the magnetic moment within the plane of the monolayer. Since the monolayers are insulating, easy-plane FMs with extremely weak in-plane anisotropy, they satisfy the necessary conditions for exhibiting a dissipationless spin superfluid mode.37

The normalized magnetizations, determined from Monte Carlo calculations, are plotted as a function of temperature for VSi$_2$N$_4$ and VSi$_2$P$_4$ in Fig. 3. The solid lines show the best fits to the analytical expression given by Eq. (2).

\[ m(T) = (1 - T/T_C)^\beta. \] (2)

The fitted values of $T_C$ and $\beta$ are shown on the plots. At the PBE+U level of theory, both monolayer VSi$_2$P$_4$ and VSi$_2$N$_4$ have Curie temperatures above room temperature, 350 K and 452 K, respectively. The effect of the increasing levels of theory, PBE, PBE+U, and HSE06, is to successively increase the bandgap between the two spin-polarized $d_{z^2}$ bands, so that at the HSE06 level, the two bands are completely gapped which maximizes the spin polarization, magnetic moment, and the exchange constant. The predicted $T_C$ increases with the increasing gap, giving a maximum value of 506 K for VSi$_2$N$_4$ using the exchange constant determined from the HSE06 calculation. Since VSi$_2$N$_4$ has the same structure, surface chemistry, and formation energy as the experimentally characterized MoSi$_2$N$_4$, we expect VSi$_2$N$_4$ to also be an air/water-stable material with $T_C > 100^\circ$ C, which is a criterion for operation in a modern integrated circuit environment.

For context, we compare to several other 2D materials with similar predicted Curie or Néel temperatures. The calculated Curie-temperatures for monolayers of the

![FIG. 2. Spin resolved energy bands of VSi$_2$N$_4$ and VSi$_2$P$_4$ calculated with (a,b) HSE06 and (c,d) PBE+U. Spin up bands are red and spin down bands are blue. The two spin-split, narrow bands on either side of the Fermi level are $d$-orbital bands centered on the transition metal.](image)

![FIG. 3. Monte Carlo calculations of the normalized magnetization as a function of temperature for (a) VSi$_2$N$_4$ and (b) VSi$_2$P$_4$. The solid lines show the best fits to the analytical expression given by Eq. (2).](image)

| Materials (Theory) | Magnetic Moment (µB) | Exchange Energy ($10^{-21}$ J) | MAE (K) | $T_C$ (K) |
|--------------------|----------------------|-------------------------------|---------|-----------|
| NbSi$_2$N$_4$ (PBE) | 0.32                 | 0.064                         | 0.30    | 12        |
| VSi$_2$N$_4$ (PBE)  | 0.93                 | 1.50                          | 0.24    | 301       |
| VSi$_2$P$_4$ (PBE)  | 0.96                 | 1.11                          | 0.14    | 235       |
| VSi$_2$N$_4$ (PBE+U)| 1.05                 | 2.53                          | 0.25    | 452       |
| VSi$_2$P$_4$ (PBE+U)| 1.04                 | 1.77                          | 0.11    | 350       |
| VSi$_2$N$_4$ (HSE06)| 1.19                 | 2.80                          | ---     | 506       |

TABLE I. Magnetic moment, exchange energy per link, equilibrium magnetic anisotropy energy (MAE), and Curie temperature, calculated using different functionals for NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$. $^*T_C$ is calculated using the HSE06 exchange energy and the PBE(SOC)+U MAE.
FIG. 4. MAE as a function of uniaxial and biaxial strain calculated with PBE(SOC)+U. MAE versus uniaxial strain: (a) VSi$_2$N$_4$ and (b) VSi$_2$P$_4$. MAE versus biaxial strain: (c) VSi$_2$N$_4$ and (d) VSi$_2$P$_4$. Values are shown for the strain coefficients for different regions discussed in the text. The orientation of the magnetization of VSi$_2$N$_4$ rotates from in-plane to out-of-plane with 3.3% biaxial strain.

FIG. 5. MAE as a function of band filling calculated with PBE(SOC)+U. MAE as a function of excess electrons per unit cell for (a) VSi$_2$N$_4$ and (b) VSi$_2$P$_4$. Values are shown for the filling coefficients discussed in the text. The orientation of the magnetization of VSi$_2$N$_4$ rotates from in-plane to out-of-plane with electron doping of $2.0 \times 10^{14}$ cm$^{-2}$.

Transition metal dichalcogenides (TMDs) VS$_2$, VSe$_2$, and VTe$_2$ are 292 K, 472 K, and 553 K, respectively. The predicted monolayer Curie temperatures for the ternary 2D materials CrSeCl, VSeTe, and CrSe have 320 K, 350 K, and 360 K, respectively. Monolayers of ferromagnetic semiconductors TeCITe$_2$ have a predicted Curie temperature of 538 K. We note that Tc is radioactive, so that Tc compounds are unlikely to see magnetic applications. Monolayers of RuI$_3$, MnN, Co$_2$S$_2$, and 3R-MoN$_2$ have Curie temperatures of 360 K, 368 K, 404 K, and 420 K, respectively. We note that synthesis of layered 3R-MoN$_2$ requires high pressure, which is not amenable to thin film growth techniques, and the proposed graphitization synthesis route to achieve MnN monolayers has not yet been demonstrated. Within the list above, VSi$_2$N$_4$ stands out for its combination of air stability and relatively high Curie temperature.

The equilibrium values of the MAE are listed in Table I. The equilibrium values of VSi$_2$N$_4$ and VSi$_2$P$_4$ range from 0.11 to 0.25 meV/magnetic atom. For comparison, monolayers of CrCl$_3$, CrBr$_3$, and CrI$_3$ have equilibrium MAEs of 0.02 meV, 0.16 meV, and 0.8 meV, respectively. Two-dimensional FeCl$_2$, NiI$_2$, Fe$_3$P, and Fe$_3$GeTe$_2$ have equilibrium MAE values of 0.07 meV, 0.11 meV, 0.72 meV, and 1 meV, respectively in their monolayer limit. In general, the MAE values of VSi$_2$N$_4$, and VSi$_2$P$_4$ are on the lower end of values for other 2D magnetic materials.

The calculated values of the MAE of VSi$_2$N$_4$ and VSi$_2$P$_4$ as a function of uniaxial strain and biaxial strain are shown in Fig. 4. We quantify the sensitivity by defining a strain coefficient as $\alpha_e = dE_{\text{MAE}}/de$. For small strain of both types in both materials, the values for $\alpha_e$ are well below the value of 32 $\mu$eV/%strain recently calculated for a 1.1 nm slab of CrSb. For compressive uniaxial and biaxial strain in VSi$_2$N$_4$, $\alpha_e = -10$ and $-6 \mu$eV/%strain, respectively. For VSi$_2$P$_4$, $\alpha_e = -6 \mu$eV/%strain for uniaxial tensile strain, $-10 \mu$eV/%strain for biaxial tensile strain, and $17 \mu$eV/%strain for biaxial compressive strain. For large (3–4%) biaxial strain in VSi$_2$N$_4$, the magnitude of the sensitivity increases. At a strain of 3.3%, the MAE changes sign, and the spins rotate from in-plane to out-of-plane. This magnitude of strain should be experimentally accessible, since 2D materials can sustain large strain.

The effects of band filling on the MAE are shown in Fig. 5. The MAE is reduced in VSi$_2$N$_4$ and VSi$_2$P$_4$ for both electron and hole doping. To quantify the sensitivity of the MAE to charge filling we define the parameter $\alpha_n = dE_{\text{MAE}}/dn$ evaluated at zero filling. For electron filling in VSi$_2$N$_4$, $\alpha_n = -0.29$ meV/electron, and for hole filling in VSi$_2$P$_4$, $\alpha_n = 0.31$ meV. What is most interesting is that the sign of the MAE can be reversed in VSi$_2$N$_4$ at an electron filling of 0.28 electrons per unit cell corresponding to a sheet carrier concentration of $n_s = 2.0 \times 10^{14}$ cm$^{-2}$. This carrier density, the orientation of the magnetization of VSi$_2$N$_4$ rotates from in-plane to out-of-plane. These densities are high, however since the thickness of the monolayer is under 1 nm, such densities could be experimentally accessible using electrolytic gating.

In conclusion, NbSi$_2$N$_4$, VSi$_2$N$_4$, and VSi$_2$P$_4$ are easy-plane, semiconducting FMs with negligible in-plane anisotropy. Exchange constants extracted from PBE+U DFT used in MC calculations predict Curie temperatures for monolayer VSi$_2$P$_4$ and VSi$_2$N$_4$ of 350 K and 452 K, respectively. The effect of the increasing levels of theory, PBE, PBE+U, and HSE06, is to successively increase the energy gap between the two spin-polarized d$_z^2$ bands, so that at the HSE06 level, the two bands are completely gapped which maximizes the spin polarization, magnetic moment, and the exchange constant. The predicted $T_C$ increases with the increasing gap, giving a maximum value of 506 K for VSi$_2$N$_4$ using the exchange constant determined from the HSE06 calculation. Since VSi$_2$N$_4$ has the same structure, surface chemistry, and formation enthalpy as MoSi$_2$N$_4$, it is expected to also be air and water stable. The magnetic anisotropy energies of VSi$_2$N$_4$ and VSi$_2$P$_4$, determined by the difference in the
total energies resulting from out-of-plane versus in-plane alignment of the magnetic moments, are small, ranging from 0.11 to 0.25 meV/magnetic atom. Tensile biaxial strain of 3.3% in VSI₂N₄ causes the MAE to change sign such that the magnetic moments rotate from in-plane to out-of-plane. Band filling of 0.28 electrons per unit cell (2.0 × 10¹⁴ cm⁻²) also causes VSI₂N₄ to switch from in-plane to perpendicular magnetic anisotropy. **Supplementary Material:** The supplementary material contains plots of orbital projected band diagrams, comparisons of electronic bandstructures calculated with PBE, PBE+U, and HSE06 functionals, and MC calculations of the normalized magnetization as a function of temperature with exchange constants extracted from PBE DFT. It also contains an extended comparison of Curie temperatures of 2D materials.

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**Data Availability Statement:** The data that support the findings of this study are available within the article and its supplementary material.

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Monolayers: Supplementary Information

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Fig. S1 shows the orbital resolved band structure of VSi$_2$N$_4$. Fig. S2 shows the d-orbital resolved band structures of VSi$_2$N$_4$ and VSi$_2$P$_4$. Spin resolved PBE, PBE+U, and HSE06 band structures are shown in Fig. S3. Monte Carlo calculations using exchange constants extracted from PBE DFT are shown in Fig. S4.

**FIG. S1.** Orbital resolved PBE band structure of VSi$_2$N$_4$ at equilibrium: (a) s-orbital contribution, (b) p-orbital contribution, and (c) d-orbital contribution. The weight is given by the color bars at right. The two isolated, narrow bands near the Fermi level are d-orbital bands centered on the transition metal atoms vanadium (V). The higher valence bands are primarily p-orbital bands which come from the N atoms. The lower conduction bands are primarily d-orbital bands.

**FIG. S2.** d-orbital resolved PBE band structure of VSi$_2$N$_4$: (a) spin up bands, (c) spin down bands. d-orbital resolved band structure of VSi$_2$P$_4$: (b) spin up bands, (d) spin down bands.
FIG. S3. Spin resolved PBE band structure: (a) NbSi$_2$N$_4$, (b) VSi$_2$N$_4$, and (c) VSi$_2$P$_4$. Spin resolved PBE+U band structure: (d) NbSi$_2$N$_4$, (e) VSi$_2$N$_4$, and (f) VSi$_2$P$_4$. Spin resolved HSE06 band structure: (g) NbSi$_2$N$_4$, (h) VSi$_2$N$_4$, and (i) VSi$_2$P$_4$.

FIG. S4. MC calculations of the normalized magnetization as a function of temperature with exchange constants extracted from PBE DFT for (a) NbSi$_2$N$_4$, (b) VSi$_2$N$_4$, and (c) VSi$_2$P$_4$. Curie temperatures are calculated from Monte Carlo simulation using the VAMPIRE software.