Supplementary Information for: Antiferromagnetic Phase of Wurtzite Nickel Sulfide Monolayer

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I. THE CONVERGENCE OF THE HUBBARD U PARAMETER

The Hubbard U parameter for the $[NiS]_2$ monolayer is determined using the self-consistent linear response method [1]. In each iteration, the crystal structure is optimized using the Broyden-Fletcher-Goldfarb-Shanno algorithm, with a convergence threshold of 10^{-6} Ry for the total energy and 10^{-6} Ry/Bohr for atomic forces, using the Hubbard U parameter value obtained from the previous step. Iterating this process makes the value of the Hubbard U parameter converge to a constant value. Starting with a very small Hubbard U parameter, we find that the Hubbard U parameter converges to U = 7.58 eV.

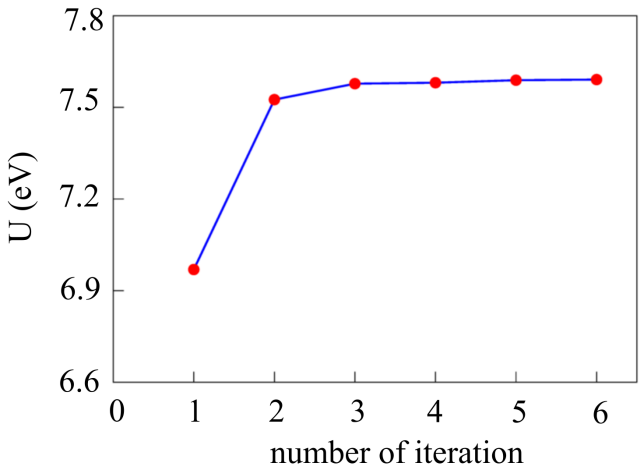

FIG. S1. The convergence of the Hubbard U parameter value calculated from the self-consistent DFT+U approach. (After the first iteration, the U is evaluated as U=6.9 eV.)

II. DERIVATION OF THE MAGNON DISPERSION RELATION FOR THE WURTZITE-ANTIFERROMAGNETS

A. Holstein-Primakoff transformation

Here, we perform a transformation of the anisotropic spin model to a bosonic Hamiltonian of spin-wave excitations. For this purpose, we use the first-order Holstein-Primakoff transformation (HP) of the spin operators. For the spin-up

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in the top sublattice,

$$\begin{aligned} S^{+}_{r,T} &= \sqrt{2S}a_{r,T}, \\ S^{-}_{r,T} &= \sqrt{2S}a^{\dagger}_{r,T}, \\ S^{z}_{r,T} &= S - a^{\dagger}_{r,T}a_{r,T}, \end{aligned} \tag{1}$$

and for the spin-down in the bottom sublattice,

$$\begin{aligned} S^{+}_{r,B} &= \sqrt{2S}a^{\dagger}_{r,B}, \\ S^{-}_{r,B} &= \sqrt{2S}a_{r,B}, \\ S^{z}_{r,B} &= a^{\dagger}_{r,B}a_{r,B} - S. \end{aligned} \tag{2}$$

The Heisenberg Hamiltonian can be rewritten as Eq. 1 in the manuscript,

$$\begin{aligned} H &= H_1 + H_2 + H_3, \\ H_1 &= J \sum_{\mathbf{r},\boldsymbol{\delta},\alpha\in T,B} \mathbf{S}_{\mathbf{r},\alpha} \cdot \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta},\alpha}, \\ H_2 &= J_\alpha \sum_{\mathbf{r},\boldsymbol{\delta}} \mathbf{S}_{\mathbf{r},T} \cdot \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta},B}, \\ H_3 &= -A \sum_{\mathbf{r},\alpha\in T,B} (S^{z}_{\mathbf{r},\alpha})^2. \end{aligned} \tag{3}$$

Before deriving each term of the Hamiltonian, we transform the real-space operators a and $a^{\dagger}$ to the reciprocal space by the Fourier transform,

$$a_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}.\mathbf{r}_i} a_{\mathbf{k}} \tag{4}$$

where N denotes the number of sites. The Fourier transformation yields,

$$\begin{aligned} S^{+}_{r,T} &= \sqrt{\frac{2S}{N}} \sum_{k} e^{ik.r_i} a_k, \\ S^{-}_{r,T} &= \sqrt{\frac{2S}{N}} \sum_{k} e^{-ik.r_i} a^{\dagger}_k, \\ S^{z}_{r,T} &= S - \frac{1}{N} \sum_{k,k'} e^{i(k'-k).r_i} a^{\dagger}_k a_k, \end{aligned} \tag{5}$$

for the spin-up, and

$$\begin{aligned} S^{+}_{r,B} &= \sqrt{\frac{2S}{N}} \sum_{k} e^{-ik.r_i} a^{\dagger}_k, \\ S^{-}_{r,B} &= \sqrt{\frac{2S}{N}} \sum_{k} e^{ik.r_i} a_k, \\ S^{z}_{r,B} &= \frac{1}{N} \sum_{k,k'} e^{i(k'-k).r_i} a^{\dagger}_k a_k - S, \end{aligned} \tag{6}$$

for the spin-down.

(1) Transformation of H_1:

The H_1 can be rewritten as,

$$\begin{aligned} H_1 =& J\sum_{r,\delta} \mathbf{S}_{r,T}.\mathbf{S}_{r+\delta,T} + \mathbf{S}_{r,B}.\mathbf{S}_{r+\delta,B} \\ &= S^z_{r,T}S^z_{r+\delta,T} + \frac{1}{2}(S^+_{r,T}S^-_{r+\delta,T} + S^-_{r,T}S^+_{r+\delta,T}) \\ &+ S^z_{r,B}S^z_{r+\delta,B} + \frac{1}{2}(S^+_{r,B}S^-_{r+\delta,B} + S^-_{r,B}S^+_{r+\delta,B}) \end{aligned} \tag{7}$$

Using the HP transformation as Eq. 5 and Eq. 6, and keeping only second-order terms in magnon operators, H_1 can be derived,

$$H_1 = 2JS\sum_k (6-\gamma_k)(a^\dagger_{k,T}a_{k,T} + a^\dagger_{k,B}a_{k,B}). \tag{8}$$

To obtain this equation, one can use $\sum_{\mathbf{r}_i} e^{-i\mathbf{k}-\mathbf{k}'.\mathbf{r}_i} = N\delta_{\mathbf{k},\mathbf{k}'}$. In the above equation, the structure factor is defined as, $\gamma_{\mathbf{k}} = cos\mathbf{k}.\delta$ and the number of the nearest neighbors of each atom within its sublattice is six.

(2) Transformation of H_2:
H_2 can be rewritten,

$$\begin{aligned} H_2 =& J_\alpha \sum_{\mathbf{r},\boldsymbol{\delta}} \mathbf{S}_{\mathbf{r},T}\cdot\mathbf{S}_{\mathbf{r}+\boldsymbol{\delta},B} \\ &= J_\alpha \sum_{r,\delta} S^z_{r,T}S^z_{r+\delta,B} + \frac{1}{2}(S^+_{r,T}S^-_{r+\delta,B} + S^-_{r,T}S^+_{r+\delta,B}) \end{aligned} \tag{9}$$

By applying Eq. 5 and Eq. 6,

$$\begin{aligned} H_2 = 2J_\alpha S\sum_k & 3(a^\dagger_{k,T}a_{k,T} + a^\dagger_{k,B}a_{k,B}) \\ &+ \gamma'_k(a_{k,t}a_{-k,B}). \end{aligned} \tag{10}$$

In Eq 10, γ'_k represents the structure factor and the number of the nearest neighbors of each atom in the adjacent sublattices is three.

(3) Transformation of H_3:
At last,

$$\begin{aligned} H_3 =& -A\sum_{\mathbf{r},\alpha\in T,B} (S^z_{\mathbf{r},\alpha})^2 \\ &= -A\sum_r S^2_{z,T} + S^2_{z,B} \end{aligned} \tag{11}$$

The Fourier representation takes the form,

$$H_3 = 2AS\sum_k a^\dagger_{k,T}a_{k,T} + a^\dagger_{k,B}a_{k,B} \tag{12}$$

Finally, after combining Eqs. 8, 10, and 12, we obtain the Hamiltonian of noninteracting magnons,

$$\begin{aligned} &H = H_k + H_{-k}, \\ &H_k = \sum_k\sum_\alpha \frac{A_k}{2}a^\dagger_{k,\alpha}a_{k,\alpha} + B_k a_{k,T}a_{-k,B} + H.c., \\ &A_k = S(2J(6-\gamma_k) + 6J_\alpha + 2A), \\ &B_k = 2J_\alpha S\gamma'_k. \end{aligned} \tag{13}$$

Since the Hamiltonian is still non-diagonal, an additional unitary transformation is required. To determine the magnon dispersion relation, we proceed with the derivation using the Bogoliubov transformation.

B. Bogoliubov transformation

As mentioned in the manuscript, the Bogoliubov Hamiltonian can be written,

$$H_{Bogoliubov} = \frac{1}{2}\begin{pmatrix} a_{k,T}^{\dagger} & a_{k,B}^{\dagger} & a_{k,T} & a_{k,B} \end{pmatrix} \Lambda_k \begin{pmatrix} a_{k,T} \\ a_{k,B} \\ a_{k,T}^{\dagger} \\ a_{k,B}^{\dagger} \end{pmatrix} \tag{14}$$

As a result, Eq. 13 can be written as $\Lambda_k e = \omega_k e$, where,

$$\Lambda_k = \begin{pmatrix} A_k & 0 & 0 & -B_k \\ 0 & A_k & -B_k & 0 \\ 0 & B_k & -A_k & 0 \\ B_k & 0 & 0 & -A_k \end{pmatrix}, e = \begin{pmatrix} u_T \\ u_B \\ v_T \\ v_B \end{pmatrix}. \tag{15}$$

to diagonalize the Hamiltonian and follow the commutation law of bosons,

$$\begin{aligned} &\sum_{\alpha} \left|u_{\alpha}^2\right| + \left|v_{\alpha}^2\right| \\ &\sum_{\alpha} \left|u_{\alpha}^2\right| - \left|v_{\alpha}^2\right|, \end{aligned} \tag{16}$$

must be preserved, respectively.

to derive ω_k, one can write Eq. 15 as,

$$\begin{aligned} A_k u_T - B_k v_B &= \omega_k u_T \\ A_k u_B - B_k v_T &= \omega_k u_B \\ B_k u_B - A_k v_T &= \omega_k v_T \\ B_k u_T - A_k v_B &= \omega_k v_B \end{aligned} \tag{17}$$

By squaring the above equation,

$$\begin{aligned} A_k^2 u_T^2 + B_k^2 v_B^2 - 2A_k B_k u_T v_B &= \omega_k^2 u_T^2 \\ A_k^2 u_B^2 + B_k^2 v_T^2 - 2A_k B_k u_B v_T &= \omega_k^2 u_B^2 \\ B_k^2 u_B^2 + A_k^2 v_T^2 - 2A_k B_k u_B v_T &= \omega_k^2 v_T^2 \\ B_k^2 u_T^2 + A_k^2 v_B^2 - 2A_k B_k u_T v_B &= \omega_k^2 v_B^2 \end{aligned} \tag{18}$$

After substituting, for example, the first and the third equations of Eq. 18,

$$A_k^2(u_T^2 - v_T^2) - B_k^2(u_B^2 - v_T^B) = \omega_k^2(u_T^2 - v_T^2) \tag{19}$$

As a result of Eq. 16, the magnon dispersion equation can be written as,

$$\omega_k = \sqrt{A_k^2 - B_k^2}. \tag{20}$$

III. THE ELECTRONIC BAND STRUCTURE

The electronic band structure of the 2D $[NiS]_2$ monolayer is plotted in ferromagnetic and antiferromagnetic phases. The Fermi level crosses with bands in both phases, indicating the metallic behavior of the monolayer.

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[1] I. Timrov, N. Marzari and M. Cococcioni, *Computer Physics Communications*, 2022, **279**, 108455.

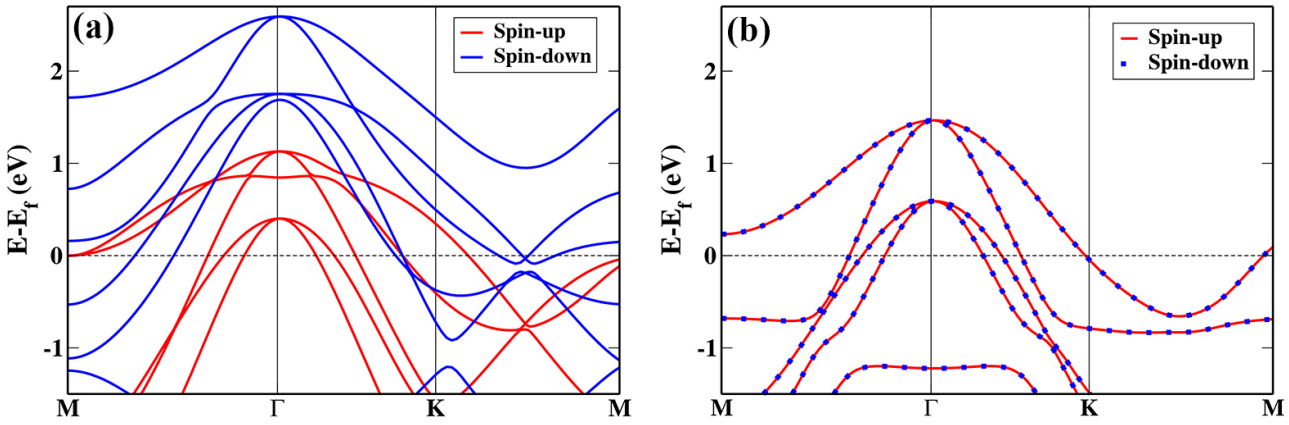

FIG. S2. The electronic band structure of $[NiS]_2$ monolayer in (a) FM and (b) AFM spin configuration. The blue and red line is shown as up and down spin, respectively. horizontal black line in (a), and (b) indicates the Fermi level energy which is set to 0 eV