Thickness dependence of electronic structure and optical properties of a correlated van der Waals antiferromagnet NiPS$_3$ thin film

Christopher Lane$^{1,2,*}$ and Jian-Xin Zhu$^{1,2,†}$

$^1$Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

$^2$Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

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We study the thickness dependence of the electronic, magnetic and optical properties of NiPS$_3$ thin film, an antiferromagnetic charge-transfer insulator. Utilizing state-of-the-art advanced density functionals, we find the antiferromagnetic zig-zag order, the band gap, and the main peaks in the dielectric tensor are all in good agreement with the corresponding experimental values. Upon thinning, the zig-zag antiferromagnetic order becomes virtually degenerate with a competing Neel order, consistent with the suppression of magnetic ordering observed by Raman spectroscopy. Other systematic changes in electronic dispersion and Kerr angle with thickness are discussed. Finally, an applied external electric field is found to precipitate an insulator-metal transition at a critical value of 0.7 eV/Å.

I. INTRODUCTION

Since the isolation of graphene in 2008, there has been explosion in the number of two-dimensional (2D) atomically-thin films predicted and synthesized using atomic species from across the periodic table. These 2D thin-films exhibit a wide variety of emergent novel phases including electronic,$^1$ excitonic,$^2$ valley,$^3$ and correlated physics$^4$ all under 2D confinement. Owing to their varied properties, 2D materials hold immense potential in a diverse spectrum of technological applications including optoelectronics,$^5$ single-molecule detection,$^6$ and energy storage and harvesting.$^7$–$^9$

The discovery of 2D materials exhibiting strong magnetic correlations and long-range order is nontrivial and has garnered substantial attention not only for their practical uses in advancing spintronics and quantum information sciences, but also for the fundamental questions they raise. Due to the 2D confinement, exotic quantum phases of matter have been predicted including 2D Kitaev spin liquids$^{10}$ and fractionalized charge states,$^{11}$ along with new elementary excitations such as Majorana fermions. Though a few candidate materials have been found, none have been confirmed for these exotic states.$^{10}$ Additionally, the high-temperature superconducting cuprate and iron-pnictide material families are composed of antiferromagnetic (AFM) 2D CuO$_2$ or Fe$Pn$ ($Pn$=As, P) planes interwoven with spacer layers comprised of rare-earth elements.$^{12,13}$ These 2D planes exhibit unconventional superconductivity and a myriad of other stripe, charge density wave, and correlated metal phases. Therefore, the discovery of other 2D materials displaying similar phase diagrams may provide valuable insights into the mechanism of unconventional superconductivity and the pseudogap regime.

In particular, the $M$PS$_3$ ($M$=Mn, Fe, Co, Ni) family of compounds are quasi-2D with their crystal structure analogous to graphene.$^{14}$ The transition metal $M$ is octahedrally coordinated by sulfur ligands within a single layer. The S atoms are connected to two P atoms located above and below the Ni plane producing a honeycomb lattice in the $ab$-plane. The atomically-thin films are then stacked in the $c$-direction in an $AB$ manner. Since two P atoms and six S atoms are covalently bonded among themselves, forming a ($P_2S_6$)$^{1−}$ anion complex, each transition metal carries a 2$^+$ ionic state.$^{15}$ Therefore, all members of the family display various long-range magnetic orders including ferromagnetic (FM), zig-zag, Neel, and stripy AFM on the transition metal sites. Moreover, the magnetic configurations follow the Ising, XY, and Heisenberg spin models, making this family of materials a unique platform for studying the phase diagram of these models under pressure, doping, and external fields.

NiPS$_3$ has recently gained attention for displaying strong charge-spin correlations$^{16}$ and a possible Mott metal-insulator transition under pressure$^{17}$ similar to that of the high-$T_c$ cuprates. Moreover, NiPS$_3$ is reported to follow the highly anisotropic XXZ Heisenberg model, where upon thinning of the film, magnetic fluctuations are found to dominate, suppressing the emergence of long-range order down to very low temperatures, in accord with the Kosterlitz-Thouless (KT) transition.$^{18,19}$ In light of these intriguing properties, there is currently no comprehensive study examining the electronic and magnetic structure as a function of thickness and the effect of applied electric field.

In this article, we present a first-principles investigation of ultra-thin films of NiPS$_3$, and systematically analyze the evolution of the ground state magnetic configuration and electronic structure with film thickness. The zig-zag AFM order is found to be the ground state for all values of the thickness. However, for a single layer, the zig-zag and Neel orders are virtually degenerate ($\Delta E \sim 0.2$ meV/Ni) facilitating large magnetic fluctuations, consistent with the suppression of magnetic ordering observed by a recent Raman spectroscopy study.$^{18}$ To verify our theoretically obtained electronic structure we calculate the energy dependent dielectric tensor and compare to the optical spectroscopy measured in a recent study.$^{16}$
The three prominent peaks in the observed optical spectra along with their line shape are in good agreement with theoretical spectra. Additionally, we find the leading edge of the complex Kerr parameters to provide a measure of thickness: discriminating between monolayer, bilayer and bulk NiPS\(_3\). Finally, we apply an external electric field along the \(z\)-axis and predict an insulator-metal transition by destabilizing the AFM order at a critical field of 0.7 eV/Å.

The outline of this paper is as follows. In Sec. II the computational details are summarized. In Sec. III the crystal structure is introduced along with a comparison of the various magnetic orderings. In Sec. IV the electronic structure and dielectric function of the Zig-Zag AFM ground state are presented. Sec. V discusses the effect of an external electric field on the electronic and magnetic state. Finally, Sec. VI is devoted to the conclusions.

II. COMPUTATIONAL METHOD

\textit{Ab initio} calculations were carried out by using the pseudopotential projector-augmented wave method\textsuperscript{20} implemented in the Vienna ab initio simulation package (VASP)\textsuperscript{21,22} with an energy cutoff of 300 eV for the plane-wave basis set. Exchange-correlation effects were treated using the strongly constrained and appropriately normed (SCAN) meta-GGA scheme.\textsuperscript{23} A \(8 \times 4 \times 1\) (\(8 \times 4 \times 8\)) \(\Gamma\)-centered k-point mesh was used to sample the slab (bulk) Brillouin zone. A denser mesh of \(12 \times 6 \times 1\) (\(12 \times 6 \times 12\)) was employed for the calculation of the dielectric tensor. Spin-orbit coupling effects were included self-consistently. The experimentally obtained atomic positions for the bulk C2/m (Space group number: 12) structure and the hexagonal D\(_{3d}\) monolayer were used throughout this work.\textsuperscript{24} A total energy tolerance of \(10^{-6}\) eV was used to determine the self-consistent charge density.

III. MAGNETIC GROUND STATE

Figure 1(a) shows NiPS\(_3\) in the bulk monoclinic structure, where the nickel atoms (teal spheres) are arranged in a honeycomb pattern and are octahedrally coordinated by six sulfur atoms (yellow spheres). The sulfur atoms are also bonded to two phosphorous atoms (pink spheres) which sit above and below the nickel layer. The NiPS\(_3\) layers are stacked along the \(c\)-axis such that a sulfur atom of one layer sits directly above the phosphorous of the next, resulting in a relative shift of the unit cell along the \(a\)-axis. The layers are weakly bound to each other by van der Waals interactions, enabling the exfoliation of atomically thin few-layer samples. Upon thinning, the monoclinic crystal symmetry is maintained until the monolayer, where the point group changes to the hexagonal D\(_{3d}\).

Figure 1(b) shows the three low-energy magnetic configurations of NiPS\(_3\) within the generalized honeycomb crystal structure of the monolayer. Magnetic moments are stabilized on the nickel atomic sites (red and blue arrows) oriented within the \(ab\)-plane, perpendicular to the \(a\)-axis, in accord with recent neutron diffraction.\textsuperscript{25} Our calculations show that the 15° out-of-plane spin tilt is energetically similar (\(\sim 1\) meV) from the 0° orientation in correspondence with the relatively weak spin-orbit coupling found in atomic nickel. The predicted value of the magnetic moment on nickel sites in the zig-zag phase is 1.504\(\mu_B\) which is in agreement with the average experimental value of 1.12\(\mu_B\).\textsuperscript{25} Our nickel magnetic moments are slightly enhanced compared to the experimental value due to the oversensitivity of the so-called iso-orbital in-
indicator (\(\alpha\)) used in SCAN to distinguish between various chemical bonding environments. Improvements in the SCAN functional in this connection,\textsuperscript{26,27} however are not likely to significantly change the conclusions of the present study. Curiously, this oversensitivity was not found in studies of the high temperature superconducting cuprates\textsuperscript{28–30} and 3\textit{d} perovskite oxides in general.\textsuperscript{31,32} Table I gives the magnitude of the magnetic moments along with the band gap and relative total energy of the various magnetic configurations.

**TABLE I.** Comparison of various theoretically predicted properties for the three possible antiferromagnetic ground states for bulk and monolayer NiPS\textsubscript{3}. The AFM orders presented for the bulk case follow the C-type stacking along the \(c\)-axis.

| Order     | Spin Orbital (\(\mu_B\)) | Orbital (\(\mu_B\)) | Total Gap (eV) | Relative Energy (meV/Ni) |
|-----------|---------------------------|----------------------|---------------|-------------------------|
| Bulk      |                           |                      |               |                         |
| Zig-Zag   | 1.434 0.069               | 1.504 1.719          | 0             |                         |
| Néel      | 1.438 0.069               | 1.508 1.623          | 1.12          |                         |
| Stripy    | 1.474 0.071               | 1.547 1.027          | 52.81         |                         |
| Monolayer |                           |                      |               |                         |
| Zig-Zag   | 1.435 0.069               | 1.504 1.827          | 0             |                         |
| Néel      | 1.438 0.069               | 1.508 1.905          | 0.19          |                         |
| Stripy    | 1.475 0.071               | 1.546 1.169          | 53.83         |                         |

Our \textit{ab initio} total energy calculations find the zig-zag type AFM order to be the ground state for all values of the film thickness in agreement with experimental observations.\textsuperscript{16,25} For the bulk crystal, the Néel and Stripy phases are found to lie at 1.12 meV and 52.81 meV above the ground state, respectively. Moreover, we also find the C-type AFM order along the \(c\)-axis to be marginally (\(\sim 1\) meV/Ni) more stable as compared to G-AMF, indicative of weakly bound van der Waals materials. Interestingly, as the number of layers of NiPS\textsubscript{3} is decreased below five layers, the relative energetic separation between zig-zag and Néel orderings decreases by an order of magnitude, resulting in a near degeneracy. The large fluctuations produced by the effective degeneracy of zig-zag and Néel configurations is consistent with the suppression of magnetic ordering observed Raman spectroscopy.\textsuperscript{18} This behavior resembles the pseudogap regime in the underdoped cuprates where competition between various magnetic ordering states suppress ordering down to low temperatures.\textsuperscript{53} Furthermore, a recent ARPES study of FeSe suggests Kosterlitz-Thouless physics may play a key role in the pseudogap regime, similar to NiPS\textsubscript{3}.\textsuperscript{34} Table I gives the magnitude of the magnetic moments along with the band gap and relative total energy of the various magnetic configurations.

**IV. ELECTRONIC STRUCTURE AND OPTICAL PROPERTIES**

Figure 2 shows the electronic band structure and atomic-site projected partial density-of-states for various values of film thickness of NiPS\textsubscript{3} in the zig-zag AFM...
phase. An AFM state stabilizes over the nickel atoms by splitting the up- and down-spin of the Ni-S antibonding states producing a gap in the Ni-$d$ bands. As a result of strong electron-electron interactions, the nickel dominated (82%) conduction states appear ‘mirrored’ at -4 eV, bookending the full bandwidth of Ni-S hybridized levels. In between, the valence states are mainly composed of sulfur-$p$ orbitals, accounting for 66% of the total atomic weight. This stacking-of-states follows the Zaanen-Sawatzky-Allen classification of a charge-transfer insulator.\textsuperscript{35} Therefore, in contrast to the Mott-Hubbard insulator, when a hole is doped into NiPS$_3$, the carrier would sit on the sulfur atomic sites rather than in the nickel sites.

![Graph showing the non-zero real (dashed lines) and imaginary (solid lines) components of the dielectric tensor of NiPS$_3$ in the monoclinic phase for bulk (black), monolayer (blue), bilayer (green), and trilayer (red) structures.](Figure 3)

The AFM order produces a 1.827 eV band gap in the monolayer case. The gap is indirect, with the lowest energetic transition occurring at ($\pm \frac{\pi}{2}, 0$) and ($\pm \frac{\pi}{2} + \delta k, 0$), for the valence and conduction bands, respectively. However, a phonon-assisted small momentum transfer of $\delta k \approx \pm 0.017 \times \frac{\pi}{2}$ is needed to connect valance and conduction band edges, allowing for efficient optical absorption and emission. Due to very weak spin-orbit coupling exhibited by the relatively light nickel atoms, spin splitting at $X'(Y')$ and $X(Y)$ is found to be very small ($\delta E \approx 0.3$ meV). Since the unit cell breaks four-fold symmetry in the $ab$-plane, $X$ and $Y$ directions in the Brillouin zone are inequivalent. This results in the states near $Y(Y')$ to be shifted away from the Fermi level to $-0.25$ eV.

As more layers are stacked along the $c$-axis, the band gap reduces converging to 1.719 eV in the bulk structure. Concomitantly, the valence band at the $X(X')$ point in the Brillouin zone shift to higher energies, pushing the leading valance band edge top away from $\Gamma$ towards $X(X')$ along the high symmetry line. This energetic shift in the valence band appears to be driven by an enhancement in the S-$p_z$ / Ni-$d_{xz}$ hybridized states concentrated around $X(X')$. This mechanism, is similar to the one found in MoS$_2$, where the interfacial Mo-$d_z$ and S-$p_z$ character states facilitate the direct-indirect transition.\textsuperscript{36} Interestingly, we find significant $k_z$ dispersion of the band structure in the bulk crystal of NiPS$_3$ resulting from appreciable interlayer coupling. The abrupt suppression of long-range ordering upon thinning suggests interlayer coupling plays a key role stabilizing the various magnetic states.

An important question in the physics of any material class is how their physical properties are linked to their electronic structure. A necessary step towards addressing this inquiry, is to be able to connect our theoretical ground state electronic structure to experimental measurements, that is, a manner by which to judge the quality of the theoretically obtained description. To this end, we calculate the dielectric tensor—a major ingredient in the interaction between light and matter— and compare the results to experimental observations. The imaginary part of the dielectric tensor within the independent particle approximation is determined by a summation over empty states using the expression

$$
\varepsilon^{(2)}_{\alpha\beta}(\omega) = \frac{4\pi^2 e^2}{\Omega} \int_{q=0}^{\delta k} \sum_{c} 2w_{ck}\delta(\varepsilon_{ck} - \varepsilon_{ek} - \omega) \times \langle u_{ck} + e_{\alpha} | u_{ek} \rangle^{*} \langle u_{ek} + e_{\beta} | u_{ek} \rangle
$$

where the indices $c$ and $v$ refer to the conduction and valence band states, respectively, $w_{ck}$ is the cell periodic part of the orbitals at crystal momentum $k$ in the irreducible Brillouin zone of weight $w_k$, and the vectors $e_{\alpha}$ are unit vectors for the three Cartesian directions.\textsuperscript{37} The real part of the dielectric tensor $\varepsilon^{(1)}_{\alpha\beta}$ is obtained by the usual Kramers-Kronig transformation

$$
\varepsilon^{(1)}_{\alpha\beta}(\omega) = 1 + \frac{2}{\pi} \mathcal{P} \int_{0}^{\infty} \frac{\varepsilon^{(2)}_{\alpha\beta}(\omega')\omega'}{\omega'^2 - \omega^2} d\omega'
$$

where $\mathcal{P}$ denotes the principal value.

Figure 3 shows the non-zero real (dashed lines) and imaginary (solid lines) components of the dielectric tensor for various thicknesses of NiPS$_3$. Two main transitions are distinctly seen at approximately 2 eV and 5 eV, along with a small peak at 3.5 eV in the imaginary part of the $xx$ and $yy$ tensor components for all thicknesses. Due to the layered nature of NiPS$_3$, the amplitude of the leading transition in $\varepsilon^{(2)}_{xx}$ is less than half of the corresponding peak in the in-plane tensor elements and is blue shifted by 0.5 eV. Combining the diagonal components on average, we find the peak energies in good accord with the $A$, $B$, and $C$ labeled peaks observed by Kim et al.\textsuperscript{16} Comparing to Fig. 2, transition $A$ is produced
by promoting an electron from the valance to conduction band edges along $X - M$ ($M' - X'$). The higher energetic transitions $B$ and $C$ originate from bands below the Fermi level connecting to the flat conduction bands along $Y' - \Gamma - Y$ and $X - M$ ($M' - X'$). Furthermore, our theoretically predicted electronic band gap is in very good agreement with the leading edge of the optical conductivity of Ref. 16. Upon thinning NiPS$_3$ from the bulk, the leading edge peak blues shifts by 0.08 eV, inline with the corresponding increase in the band gap. In $\varepsilon^{(2)}_{zz}$ a slight shoulder appears, mainly driven by the change in energy of the flat bands along $X - M$ ($M' - X'$).

When a linearly polarized light is reflected from a magnetic material, the reflected light is typically elliptically polarized. The angle through which the ellipse rotates is called the Kerr rotation angle. This phenomenon is exceptionally useful to mark structural phase transitions and to give direct insight into the local, microscopic magnetism in condensed matter systems. In this way, we track the changes in the Kerr parameters with layer number to see if they can be used as an indicator of film-thickness.

The Kerr parameters are related to the dielectric function by the following,

$$\Psi_k = \theta_K + i\gamma_K = \frac{-\varepsilon_{xz}}{(\varepsilon_{xx} - 1)\sqrt{\varepsilon_{xx}}}$$

for a polar geometry in the small angle limit. Here the photon propagates along the $y$-direction and describes a linearly polarized wave with the electric field along the $x$-direction. The off diagonal dielectric tensor element $\varepsilon^{(2)}_{xz}$ [Fig. 3] is non-zero and oscillates about zero following a clear amplitude enhancement with increased thickness, until saturation in the bulk.

Figure 4 shows the complex Kerr parameters from Eq. (3) as a function of energy for various thicknesses of NiPS$_3$. The values are small $\sim 0.1$, as expected for optical wavelengths. Interestingly, the leading edge of $\gamma_K$ near 2 eV follows a regular progression with layer number, where the spectra starts with a positive value and gradually reduces, until flipping sign for a film thickness larger than two layers. $\theta_K$ exhibits a similar behavior limiting to zero in the bulk case. Therefore $\theta_K$ and $\gamma_K$, though small, might hold promise as an optical descriptor of thickness in NiPS$_3$ and magnetic 2D materials in general.

V. EFFECT OF AN EXTERNAL STATIC ELECTRIC FIELD

Voltage control of magnetism has been intensely pursued during the past few decades not only due to the direct connection to application in the miniaturization of magneto-electronics needed for spintronics and quantum information technologies, but also for the rich fundamental physics at the heart of the interplay between charge, spin, orbital, and lattice degrees of freedom.

![FIG. 5. (color online) Evolution of the electronic band gap (crimson line) and the nickel magnetic moment (blue line) for monolayer NiPS$_3$ with external applied electric field. $E$ is taken positive along $z$-direction, and is shown schematically with the crystal structure of monolayer NiPS$_3$.](image-url)

Figure 5 shows the magnitude of the nickel magnetic moment for monolayer NiPS$_3$ as a function of electric field positively aligned along the $z$-axis. As the electric field strength increases the magnetic moments decrease. At an $E_z$ of 0.6 ($-0.7$) eV/Å a first-order transition occurs, quenching the magnetic moment. Additionally, the
maximum deviation of the magnetic moments before collapse is only 3%, allowing for clean high-to-low and low-to-high transitions needed in magnetic based switches. Simultaneously, the electronic band gap decreases with lapse is only 3%, allowing for clean high-to-low and low-

Figure 5 (inset) shows the charge density difference between the insulating-magnetic and metallic-non-magnetic phases where the positive (negative) charge clouds are colored yellow (light blue). A clear migration of charge from the sulfur atoms to the nickel atomic sites are half-filled. Due to significant on-site electron repulsion in the 3d transition metals, the spin degeneracy of the $e_g$ orbitals is split to lower the total energy of the system, producing a zig-zag AFM order across nickel atomic sites. When the electric field is applied, the system reorganizes to screen the external field, pushing the system towards metallicity – as indicated by the quickly reduced band gap – which in turn reduces the effect of the on-site Coulomb potential. Once the on-site Coulomb potential is sufficiently screened, the AFM order collapses.

VI. CONCLUDING REMARKS

By examining the ground state electronic structure of NiPS$_3$ as a function of the film thickness, we systematic changes in the electronic and magnetic structure. Key properties, including the competition of magnetic ordering states in the monolayer resemble the characterizing features of the cuprate high-temperature superconductors. In order to elucidate the connection between NiPS$_3$ and the cuprates, if any, further doping studies are needed to examine the evolution of the electronic and magnetic phases, and compare to the phenomenology of the cuprates. Additionally, the layer degrees of freedom gives us another knob, by which to tune the ground state, providing a platform to study the competition of various short-range magnetic and charge fluctuations.

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* laneca@lanl.gov
† jxzhu@lanl.gov

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Since we are employing a Meta-GGA density functional, the derivative of the cell-periodic part of the orbitals was computed using the finite difference scheme as implemented in vasp.