Defect-induced 4p-magnetism in layered platinum diselenide

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Platinum diselenide (PtSe$_2$) is a recently-discovered extrinsic magnet, with its magnetism attributed to the presence of Pt-vacancies. The host material to these defects itself displays interesting structural and electronic properties, some of which stem from an unusually strong interaction between its layers. To date, it is not clear how the unique intrinsic properties of PtSe$_2$ will affect its induced magnetism. In this theoretical work, we show that the defect-induced magnetism in PtSe$_2$ thin films is highly sensitive to: (i) the layer-thickness (ii) defect density, and (iii) substrate choice. These different factors dramatically modify all magnetic properties, including the magnitude of local moments, strength of the coupling, and even nature of the coupling between the moments.

We further show that the strong inter-layer interactions are key to understanding these effects. A better understanding of the various influences on magnetism, can enable controllable tuning of the magnetic properties in Pt-based dichalcogenides, which can be used to design novel devices for magnetoelectric and magneto-optic applications.

I. INTRODUCTION

The recent discovery of magnetism in two dimensional (2D) layered materials\cite{11, 12} has sparked renewed interest in one of the oldest and most well-studied emergent phenomenon within solid state physics: collective magnetism. Not only do these 2D magnets circumvent the Mermin-Wagner theorem, which forbids long-ranged order in low dimensions through magnetic anisotropy, but they also possess an interesting tunability of their magnetic properties that is facilitated by their 2D nature. For instance, magnetic ordering in CrI$_3$ thin films\cite{13} has been shown to depend on the number of layers. It was also shown that the nature of magnetic coupling in a CrI$_3$ bilayer\cite{14} can be tuned by electric fields (gating). Theoretical work on CrX$_3$ (X = Cl, Br, I) monolayers\cite{15} also indicates that strain can be used to tune magnetic properties. Hence, 2D magnets and their tunable properties offer opportunities to study the phenomena of local moment formation and collective magnetism in low-dimensions, as well as to design devices for spintronics, magneto-optics, quantum-information and -sensing applications. These 2D magnets can be: (i) intrinsic in nature, such as CrI$_3$, or (ii) extrinsic in nature. In the latter category, we include all 2D materials, otherwise non-magnetic, in which magnetism can be induced through: (i) defect engineering (vacancies, substitutionals, creating edges/nanoribbons)\cite{16, 17} (ii) intercalation between layers by magnetic species\cite{18, 19}, or (iii) proximity effects (2D crystal placed on a magnetic substrate)\cite{20, 21}. With only a few known intrinsic 2D magnets, there is a strong motivation to use the aforementioned strategies to induce collective magnetism in non-magnetic 2D crystals.

Amongst extrinsic 2D magnets, long-ranged magnetism was recently discovered in platinum diselenide (PtSe$_2$)\cite{22}. In a joint experimental-theoretical work, Avsar et al. attributed the observed magnetism to the presence of platinum vacancies (V$_{Pt}$)\cite{23} which had also been previously investigated using density functional theory (DFT)\cite{24}. Over the past few years, several other theoretical strategies have been proposed to induce a magnetic moment in PtSe$_2$ monolayers, such as: (i) combining strain with hole doping to induce magnetism\cite{25} (ii) Se-vacancy in the simultaneous presence of strain\cite{26} (iii) hydrogenation on Se sites\cite{27} and (iv) doping with transition metal elements\cite{28}. However, most of these theoretical works used freestanding monolayers of PtSe$_2$ placed in vacuum, ignoring the effects of layer-thickness, or the presence of a substrate. On the other hand, experiments indicate that the magnetic properties in 2D crystals are strongly affected by layer thickness and substrate\cite{29, 22}. The importance of taking layer-thickness and/or substrates into account was also recently highlighted in careful theoretical studies on different 2D materials\cite{30, 27}.

In this work, out of the different proposed ways of inducing magnetism in PtSe$_2$ thin films, we chose to use platinum vacancies, as these defects have been experimentally implicated in the observed ferromagnetism in PtSe$_2$\cite{22}. Platinum vacancies were also experimentally visualized as one of the defects present in PtSe$_2$ samples, which were synthesized using the chemical vapor transport method\cite{29}. We explored the dependence of the defect-induced magnetism on several factors: (i) vacancy concentrations, (ii) layer-thickness, by varying the number of layers up to 4 layers, and (iii) substrate choice. Using DFT-based calculations, we find that the magnitude of the magnetic moment and the exchange coupling between the moments are modified with vacancy concentrations. Moreover, the magnetic properties depend significantly on layer thickness/substrate due to the strong interlayer interaction between the layers in the presence of Pt-vacancies. Our results show that accounting for the “real-world conditions”, such as layer thickness or the presence of a substrate, is important when predicting the magnetic properties of 2D materials.

II. CALCULATION DETAILS

Our spin-polarized DFT calculations were performed using the Vienna Ab-initio Simulation Pack-
FIG. 1. Pristine PtSe\textsubscript{2} properties. (a) Structure (top view) of pristine T-phase PtSe\textsubscript{2} monolayer. The dashed area shows the primitive unit cell for PtSe\textsubscript{2}. Band structures for a PtSe\textsubscript{2} (b) monolayer and (c) bilayer, showing layer dependence of electronic structure properties. The change density difference plot, $\Delta\rho = \rho(\text{bilayer}) - \rho(\text{top-layer}) - \rho(\text{bottom-layer})$, for (d) PtSe\textsubscript{2} and (e) WSe\textsubscript{2} bilayers, respectively, showing larger interlayer interaction in the former structure versus the latter structure. The pink and yellow isosurfaces represent charge accumulation and depletion, respectively.

III. RESULTS AND DISCUSSION

Figure 1(a) show the optimized structure of the pristine PtSe\textsubscript{2} monolayer, for which the lowest energy phase is the octahedrally-coordinated T-phase. Before investigating defect-induced magnetism in 1T-PtSe\textsubscript{2}, it is instructive to consider the unique properties of the pristine structure, which set PtSe\textsubscript{2} apart from more extensively-investigated group-VI TMDs. Recent experimental\textsuperscript{32} and theoretical\textsuperscript{33,34} studies showed a dramatic change in its electronic structure properties as a function of thickness. This material, which prefers AA-stacking, is metallic in bulk, semi-metallic as a trilayer, and semiconducting as a bilayer or a monolayer. Figures 1(b) and (c) are plots of the band structures of a pristine monolayer and a bilayer, respectively, showing the layer-dependence of the indirect band gap, which changes from about 1.40 eV for the former structure to about 0.20 eV in the latter structure. This strong dependence of electronic structure properties on the sample thickness stems from a strong interlayer interaction, resulting in much smaller distances between the layers. For example, in the case of a PtSe\textsubscript{2} bilayer, the binding energy, $E_b$, is $-0.199$ eV/f.u. with $E_b = E_{\text{bilayer}} - E_{\text{top-layer}} - E_{\text{bottom-layer}}$. $E_X$ = total energy of system X, and formula unit abbreviated as f.u., and the calculated interlayer distance is about 2.30 Å. For comparison, the calculated binding energy for the WSe\textsubscript{2} bilayer is $-0.148$ eV/f.u., and
the interlayer distance is about 3.15 Å, where we have chosen WSe$_2$ as a representative material from group-VI TMDs. The strong interlayer interaction in PtSe$_2$ can also be seen from the charge density difference ($\Delta \rho$) plot for the PtSe$_2$ bilayer shown in Fig. 1(d). Here, $\Delta \rho = \rho$(bilayer) $- \rho$(top-layer) $- \rho$(bottom-layer), is a measure of the extent to which there is a charge redistribution upon formation of a bilayer, and hence, a measure of the interlayer interactions. In contrast, for the case of WSe$_2$-bilayer shown in Fig. 1(e), for which the interaction is mostly van der Waals in nature, the structure shows minimal charge-redistribution. In the subsequent discussion, we will show how the greater strength of interlayer interactions profoundly affect defect-induced magnetism in PtSe$_2$ layers.

A. Defect-induced magnetism in PtSe$_2$ monolayer

Once we remove a platinum atom from the matrix, we find that a total magnetic moment of 4 $\mu_B$ is introduced in the freestanding PtSe$_2$ monolayer, as was also reported in previous works.\cite{20,21} Most of the magnetic moment originates from the partially occupied 4$p$ orbitals of the six neighboring Se atoms ($\sim$ 0.46 $\mu_B$/per atom), as shown in Fig. 2(a). No such local moment is induced by, for example, a tungsten monovacency in WSe$_2$ monolayer.

In PtSe$_2$, the platinum vacancy results in a unique reconstruction around the defect, in which the nearest-neighbor Se atoms move outwards away from the vacancy. The Se-Se distance increases by $\sim$ 0.178 Å in-plane and $\sim$ 0.16 Å out-of-plane as compared to the pristine PtSe$_2$ monolayer. However, in the case of a WSe$_2$ monolayer, Se atoms surrounding the defect move closer to each other, with the in-plane distance decreasing by about 0.062 Å (as compared to a pristine WSe$_2$ monolayer). This inward movement of atoms leads to an increased overlap between the dangling bonds, resulting in a nonmagnetic structure. As a result, one does not associate metal vacancies in group-VI TMDs with induced magnetism, unlike the vacancies in PtSe$_2$. Figure 2(b) shows the band structures of defective PtSe$_2$ monolayers with three different defect concentrations. All three of the band structures show spin-splitting of the defect states into the spin-up (majority) and spin-down (minority) states. For the largest defect concentration (11.11%), the bandwidths of the defect states [highlighted in Fig. 2(b)] are larger due to the high defect concentration, which results in an increased interaction between the defects. This large dispersion can be seen in the widths of the highlighted defect states around the Fermi level in the density of states (DOS) plot for the largest defect concentration [see, Fig. 2(c)]. As a result of the large dispersion, there is a considerable overlap between the two spin channels, resulting in a smaller spin-polarization energy of 44.27 meV [see Fig. 2(d)]. Here, the spin polarization energy is defined as a difference in total energies of the magnetic and non-magnetic structures: $\Delta E_{sp} = E_{\text{non-magnetic}} - E_{\text{magnetic}}$. On the other hand, as can be seen in the the band structures and DOS plots for the smaller defect concentrations (6.25% and 4.00%) in Figs 2(b) and (c), respectively, the defect states are nearly dispersionless, resulting in an increased spin-splitting between the two spin channels. In turn, this results in larger spin polarization energies. The calculated values of spin-polarization energies for the 6.25% and 4.00% concentrations are 102.36 meV and 320.17 meV respectively [see Fig. 2(d)]. These are large energies (compared to room temperature ($k_B T \approx 25$ meV), implying that the local moments will survive at room temperature. Another interesting point to note is that for the largest defect concentration that we considered for freestanding monolayer, the structure is a half-metal; spin-up states show semiconducting behavior while spin-down states show metallic characteristic. For the lower defect-concentration (6.25% and 4.00%), both spin-up and spin-down channels show semiconducting behavior.

Formation of defect-induced local moments does not necessarily result in collective magnetism, with the latter being a result of the exchange interaction between the moments. In order to determine the strength and the nature of magnetic exchange coupling between the defect-induced moments, we determined the energy difference between the ferromagnetic (FM) and antiferromagnetic (AFM) alignments of the moments. In order to do so, we doubled the size of the supercells in the $x$-direction, and depending upon how we initialized the moments, we obtained either the AFM or the FM alignment of moments. The difference in total energies for the two stable magnetic structures, $\Delta E = E_{\text{AFM}} - E_{\text{FM}}$, is then mapped onto an effective Heisenberg spin model, which gives the distance-dependent values of the exchange coupling constant. According to the Heisenberg model, taking into account only nearest-neighbor interaction: $H = -J_0 \sum_{\langle ij \rangle} S_i \cdot S_j$, where $J_0$ is the nearest-neighbor exchange coupling constant and $S_i$ is the spin induced by the $i^{th}$ platinum vacancy. This yields the expression: $J_0 = \Delta E/8S^2$. The strength of the interaction between moments itself depends on the distance between the interacting moments and hence, on the distance between the defects. In this work, we considered three vacancy concentrations: 4.00%, 6.25%, and 11.11%. The corresponding distances between the two vacancies are 18.75 Å, 15.00 Å, and 11.25 Å. Figure 2(c) shows $J_0$ as a function of vacancy concentration (or equivalently, defect-defect distances). The figure shows that as the defect concentration decreases, the exchange coupling parameter becomes smaller, ultimately switching sign, implying that the AFM alignment of moments becomes the preferred state for very small concentrations.
FIG. 2. Properties of platinum vacancies in a PtSe$_2$ monolayer. (a) The relaxed atomic structure and the isosurface plot of spin density distribution for $V_{Pt} = 6.25\%$. (b) Band structure for different defect concentrations, showing overall increase in the bandwidths as well as increasing overlap of spin-split defect states (yellow highlight) with increasing defect concentration. (c) Density of states (DOS) for defective structures, showing the defect states (yellow highlight) around the Fermi level (used as the reference energy). (d) Spin polarization energy as a function of defect concentration, showing a decrease in spin polarization as the overlap between the spin-split defect-states increases with increasing defect concentrations. (e) The computed exchange coupling constant ($J_0$) as a function of defect concentration (hence, the defect-defect distance) for PtSe$_2$ monolayers.

B. Thickness dependence of defect-induced magnetism in PtSe$_2$

The defect-induced magnetic properties of PtSe$_2$ are expected to be affected by not only the vacancy concentration, but also the layer thickness. In this work, we considered up to 4 layer-thick PtSe$_2$ films. Due to the computational costs, we restricted this part of the study to only two $V_{Pt}$ concentrations: 6.25% and 11.11%. In each case, $V_{Pt}$ was created on the topmost layer only. This is because according to our calculations, a vacancy in one of the inner layers of a PtSe$_2$-stack does not yield a net magnetic moment, and the system remains non-magnetic. Fig. 3(a) shows the effect of PtSe$_2$ layer thickness on the magnetic moment for the considered systems. As discussed in the previous section, in the case of a monolayer, the value of the defect-induced magnetic moment (4.00 $\mu_B$) is independent of the $V_{Pt}$ concentrations [see Fig. 2(a)]. Beyond one-layer thick PtSe$_2$, the magnetic moment depends on the layer-thickness, and this dependence varies with changing defect concentration. In general, the magnetic moment decreases as a function of thickness. However, this decrease is more pronounced for lower vacancy concentration (6.25% $V_{Pt}$), where the system becomes non-magnetic for all thicknesses beyond a single-layer [see Figure 3(a)]. For the higher defect concentration of 11.11%, the structure remains magnetic, although the moment decreases with the number of layers. For example, the magnetic moment reduces from 4.00 $\mu_B$ in a monolayer to a value of 1.99 $\mu_B$ in a bilayer.

Figure 3(b) gives spin polarization energy as a function of layer-thickness for different concentrations of vacancies. For the smaller defect concentration of 6.25%, the structure becomes non-magnetic as thickness is increased beyond a single layer. This is reflected in the spin polarization values that reduce to zero [see the black curve in Fig. 3(b)]. The behavior of spin polarization energy values obtained for the higher defect concentration (11.11%) is more interesting. At this concentration, the calculated spin-polarization energy for a bilayer shows an unexpected increase in the value as compared to that obtained for a freestanding monolayer. The band structure for the bilayer with 11.11% defect concentration, plotted in Fig. 3(c), reveals the interrelated factors that explain the unexpected increase of the spin-polarization energy. It shows a large reduction in the bandwidth of the defect-induced states (within the band gap) as compared to those in the band structure for the freestanding monolayer [see Fig. 2(b)] with the same defect concentration. The resulting spin-splitting between the two spin channels is relatively larger, explaining the increase in spin polarization energy. The change in the bandwidth of the defect-states in the bilayer as compared to those in the free-standing monolayer [shown in Fig. 2(b)] can be attributed to the differences in structural distortions around the defect. In turn, this is due to the substrate
friction from the bottom layer, which changes the extent of relaxation around the defect in the defective top layer within a bilayer. Figure 3(d) is a plot of DOS for bilayers with 11.11% defect concentrations, projected onto the top (defective) and the bottom layers. As was also seen in the band structure for the system, the highlighted defect states [see Fig. 3(d)] around the Fermi level (used as the reference energy) are narrower as compared to those for the freestanding monolayer with the same defect concentration [Fig. 2(c)]. This again implies greater localization of the defect states in the bilayer as compared to the monolayer. In the case of the smaller defect concentration (6.25%), the DOS projected onto the top and bottom layers within a bilayer reveals a symmetric DOS in the two spin channels [Fig. 3(e)], and hence, the quenching of magnetism for lower defect concentrations.

To further understand how the presence of additional layers affects the magnetism in the top layer, we considered the spin density distribution, $\Delta \rho = \rho^\uparrow - \rho^\downarrow$. Figure 4(a) is a plot of the calculated $\Delta \rho$ for PtSe$_2$ bilayers with 11.11% defect concentration. It shows a reduction in magnetic moment ($\sim 0.11 \mu_B$) contributed by each of the Se-atoms that surround the defect and are located at the interface of the bilayer. On the other hand, the magnetic moment of Se atoms on the surface is $\sim 0.37 \mu_B$ [see Fig. 4(a)]. This is quite different from the spin density profile of the PtSe$_2$ monolayer [Fig. 2(b)], where all six Se-atoms surrounding the Pt vacancy contribute equally to the total moment. This near-quenching of the magnetism for the larger defect concentration, and its complete quenching for the bilayers with smaller defect concentrations, can be attributed to the following different yet inter-dependent factors: (i) changes in the structural distortion around the defect within a bilayer due to substrate-friction (here the substrate is another PtSe$_2$ layer), (ii) charge transfer from the bottom layer, and (iii) interface states due to strong interaction between the two layers. To investigate each of these effects sequentially, we followed the steps taken in Ref. 26 and conducted a series of tests on structures with 11.11% and 6.25% defect concentrations: (i) We first investigated the effect of the changed structural distortion in the defective layer, with these changes arising due to the presence of a neighboring layer. For this calculation, we considered a freestanding, defective PtSe$_2$ monolayer with the “frozen-in” atomic positions obtained from the defective top layer at equilibrium within a bilayer. The calculated magnetic moment for both concentrations considered here remains 4.00 $\mu_B$. These results indicate that the changes in the structure around a vacancy within a composite (as compared to a freestanding relaxed structure) do not play a role in reducing the magnetic moment. (ii) Another factor that can contribute to the reduction in moment is the calculated $n$-doping of the defective top layer by the bottom PtSe$_2$ layer. Using Bader charge...
The spin-split states induced by the defect are highlighted. (e) Effect of 2D and 3D substrates. At equilibrium, the Se-atoms of the top and bottom layers surrounding the V_{Pt} make bonds within the 2D PtSe_{2}/h-BN heterostructure. Within the mixed dimensional heterostructure consisting of PtSe_{2} on a Cu(111) substrate, the strong interaction leads to the formation of covalent bonds. For both composites, we observe a quenching of magnetism.

Analysis of the calculated charge transfer to the defective layer from its neighboring layer is 0.19 e/supercell when the defect concentration is 11.11%, and 0.17 e/supercell when the defect concentration is 6.25%. Again starting with the hypothetical monolayer described in the previous paragraph, we doped the structures with the appropriate charges depending on the defect concentration. Our calculations show a small reduction in magnetic moment due to charge doping, with 3.81 \( \mu_B \) for the 11.11% defect concentration, and 3.83 \( \mu_B \) for the smaller defect concentration. Therefore, we find that although n-doping of the top layer reduces the magnetic moment, this reduction is not significant.

(iii) The third factor that we investigated is the stronger interlayer interaction within a bilayer in the presence of a defect. Our calculations show that the binding energies for the bilayers with 11.11% and 6.25% defects are \(-0.287 \text{ eV/f.u.}\) and \(-0.264 \text{ eV/f.u.}\), respectively. These are significantly higher than the binding energy for a pristine bilayer \((-0.199 \text{ eV/f.u.})\). The importance and the strength of interlayer interaction can be seen in the charge density difference plots for the two concentrations [Figs 4(b,c)]. In both cases, there is an enhancement of charge rearrangement upon forming a bilayer when one of the PtSe_{2} layers contains a defect [cf. Fig. 4(d) for defect-free bilayer]. The structure with the lower defect concentration can be seen to have gone through a larger charge rearrangement [Fig. 4(c)], compared to the structure with the larger defect concentration [Fig. 4(b)]. This might explain the complete quenching of magnetism for the structure with 6.25% defect concentration. In order to further test the effect of strong interlayer interaction, we started with the fully relaxed structure of a defective PtSe_{2} bilayer with equilibrium inter-layer distance, \( d_{eq} \), and moved the two layers apart in the steps of 0.2 Å. Table 1 lists the magnetic moment as a function of this rigid shift, \( \Delta d \), for defective PtSe_{2} bilayers with different defect concentrations. The magnetic moment increases as a function of \( \Delta d \) and the system recovers the magnetic moment calculated for a freestanding layer (4.00 \( \mu_B \)) when the rigid shift \( \Delta d = 0.8 \text{ Å} \) for the vacancy concentration of 6.25%, and when \( \Delta d = 1.0 \text{ Å} \) for the defect concentration of 11.11%. These results indicate that the strong interlayer interaction in a bilayer is a major contributing factor responsible for the reduction in magnetic moment. This can also be seen from the Fig. 4(d), which shows the DOS projected onto the top defective layer (with 6.25% \( V_{Pt} \)) at different values of \( \Delta d \). As \( \Delta d \) increases, the spin-splitting reappears, and the defects states around the Fermi energy become narrower.
approaching the case of a freestanding monolayer.

| ∆d (Å) | Magnetic moment (µB) [6.25% V_{Pt}] | Magnetic moment (µB) [11.11% V_{Pt}] |
|--------|-----------------------------------|-----------------------------------|
| 0      | 0.00                              | 1.99                              |
| 0.2    | 0.54                              | 1.98                              |
| 0.4    | 0.83                              | 1.96                              |
| 0.6    | 2.92                              | 2.13                              |
| 0.8    | 4.00                              | 3.53                              |
| 1.0    | 4.00                              | 4.00                              |

C. Substrate Effects

Lastly, in most applications, the PtSe$_2$ layer(s) will be part of 2D or mixed-dimensional heterostructures. Therefore, we investigated the effect of using conventional 2D or 3D substrates on the magnetic properties of defective PtSe$_2$. We chose two representative substrates: h-BN, which is a 2D insulator and Cu(111), which is a 3D metal. The heterostructures are shown in Fig. 4(e). For both of the substrates studied here, their interactions with the defective PtSe$_2$ layer results in the quenching of magnetism. We found that even though the interlayer distance between PtSe$_2$ and h-BN is large (∼3.4 Å) as compared to PtSe$_2$ bilayers, h-BN strongly modifies the magnetic properties of PtSe$_2$ films. When PtSe$_2$ is placed on h-BN, the reconstruction around the defect in the PtSe$_2$ layer results in bonding between Se atoms from the top and bottom faces around the vacancy [see Fig. 4(e)]. When the PtSe$_2$ layer is placed on Cu(111), the monolayer interacts strongly with the copper substrate, which results in the formation of bonds between the two subsystems in the composite structure [see Fig. 4(e)]. Once again, the strong interlayer interaction results in quenching of the magnetism in the monolayer. We would like to note that due to the large computational cost of such calculations, our results for the heterostructures were obtained for a single defect concentration (6.25%) in a single layer of PtSe$_2$ placed on the substrate. In experiments, the outcome of using a substrate may change depending on not only the substrate-PtSe$_2$ interactions, but also a number of other factors, such as the defect concentration and the number of PtSe$_2$ layers in the heterostructure. Nevertheless, our proof-of-principle study for the 2D and mixed-dimensional heterostructures shows that a careful choice of substrates may be important to ensure survival of magnetism in the defective PtSe$_2$.

IV. CONCLUSIONS

Using first-principles calculations, we studied magnetism in PtSe$_2$ layers that is induced by platinum vacancies. In a monolayer, the presence of V$_{Pt}$ induces a net magnetic moment of 4µ$_B$, irrespective of the defect concentration. The dangling bonds around the defect are responsible for this local moment, with the largest contribution coming from the 4p-orbitals of Se-atoms. These orbitals are an unusual source of unconventional magnetism, which is typically associated with the unpaired electrons within the localized 2p-orbitals of the second row elements$^{[30],[35]}$. On the other hand, the 4p-orbitals give rise to highly dispersive bands and are not implicated in local moment formation in 3D solids. In PtSe$_2$, however, the 4p-derived defect states surrounding the vacancy were found to be sufficiently localized to give rise to a net magnetic moment. In fact, it was recently shown that the unpaired electrons in the 3p- and 4p-orbitals in 2D materials can indeed give rise to localized orbitals and hence, to a net magnetic moment due to the quantum-confinement effects in a 2D-structure$^{[39]}$. We showed that this magnetism in defective PtSe$_2$ layers is strongly affected by the vacancy concentration, thickness of the PtSe$_2$ layers, as well as, the choice of the substrates. These different factors dramatically modify all magnetic properties, including the magnitude of local moments, strength of the coupling between the moments, and even nature of the coupling between the moments. Our results explain the dependence of the magnetic properties on different factors. Pt-based dichalcogenides are important candidate extrinsic magnets that can be used to design novel devices for magnetoelectric and magneto-optic applications. A better understanding of the various influences on magnetism will be important for controllably tuning their magnetic properties.

V. ACKNOWLEDGEMENTS

This work is supported by National Science Foundation under Grant Number DMR-1752840. PD and PM acknowledge the computational support provided by the Extreme Science and Engineering Discovery Environment (XSEDE) under Project PHY180014, which is supported by National Science Foundation grant number ACI-1548562. For three-dimensional visualization of crystals and volumetric data, use of VESTA 3 software is acknowledged.

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