Reviewers' comments:

Reviewer #1 (Remarks to the Author):

This manuscript reports a study of the electronic and magnetic properties of a VSe_2 single layer grown on $NbSe_2$. The main experimental technique is STM/STS, and the results are compared with DFT calculations. The key issue of interest is the magnetic state of VSe_2. Some prior studies have suggested that single layer VSe_2 is ferromagnetic, while other studies have indicated otherwise. This study adds to the ongoing debate. PPMS measurements of a capped sample shows a magnetic signature, but the Ms value is much smaller than that in VSe_2/HOPG (Ref. 13). The authors' tunneling data do not match well with the calculations under different assumptions and are thus inconclusive. STM shows no evidence for a charge density wave (CDW), which differs from some prior studies. The authors suggested that the lack of CDW might be related to magnetic ordering. After reading through the relevant publications, it appears that the different conclusions from the various studies are not necessarily contradictory and could be simply related to the different sample configurations and environments. DFT calculations in the present work (and also in prior studies) show that the energy differences between the different states (CDW, magnetic, and nothing) are tiny and depend on the details of the calculations. Therefore, it seems reasonable that differences in the substrate choice (metals, semimetals, semiconductors, …), presence or absence of capping layer, and contamination by non-UHV processing conditions as in some prior studies could very well be the source of the divergent observations and opinions. The ferromagnetic state could be caused by "extrinsic effects" based on the majority of evidence in the literature.
Specifically, it is unclear that the PPMS data for a capped sample and the STM/STS data for a pristine sample in the present study can be directly correlated. The samples are just different, and they could be in different states. This should be emphasized in the manuscript. Ideally, one would want to perform PPMS measurement on a sample decapped under UHV, but it is probably a hard experiment. The authors' statement that "… lack of charge density wave signatures indicates magnetization of the sheet..." is simply a conjecture, as there is no direct proof that the UHV sample is in fact magnetic. CDW can be suppressed for a number of other reasons including charge transfer, lattice distortion, impurities, and electronic coupling to the substrate. The authors' strong statement is simply not justified. STS measurements of the electronic structure may in principle be able to settle this issue; if one sees exchange splitting of the bands that diminishes toward the Curie temperature, the case would be clear. Unfortunately, the STS data presented in the manuscript are not well correlated with the calculated density of states.
Another concern is sample quality, which could be an issue for many of the studies to date. The authors stated that the VSe_2 islands show sharper edges at higher growth temperatures, but at the same time the CDW modulation of the $NbSe_2$ substrate becomes weakened because of intercalation. One wonders if the CDW state of VSe_2 might be similarly affected. It is also curious that the authors annealed their cleaved $NbSe_2$ samples at 600 K for 1 h before film growth. Is the annealing step needed? Cleaved samples should have a pristine surface. Annealing could deplete Se and create defects.
A 3% lattice mismatch is large. Is the VSe_2 lattice constrained to that of the $NbSe_2$ in the calculations? This could affect the ground state. Please clarify the assumptions.
The tunneling current of STM is typically confined to a cone angle. As a result, STS typically measures the density of states with a strong weighting along the surface normal direction. The tunneling spectra might resemble more a one-dimensional density of states. Please clarify.
The introduction contains a good review of different kinds of composite quantum materials. Maybe it could be made more concise by focusing specifically on the VSe_2 case.
In summary, the experiments seem to be carried out with care. The analysis, interpretation, and discussion are not necessarily accurate and could be improved. The different sample conditions and environments for the different studies should be pointed out and considered as a possible reason for the different conclusions.

Reviewer #2 (Remarks to the Author):

This is an interesting paper on the in-situ growth and characterization of heterostructures of VSe2/NbSe2, with the experimental results being compared to DFT calculations. Interestingly, the VSe2 layer seems to host magnetism, which affects the superconducting gap of the underlying NbSe2 substrate.
The successful growth and results are interesting and deserve publication in Communications Physics. However, I would like to see some more discussion/information concerning these aspects:
1. The growth of atomically clean layers is an important aspect of this paper and shall serve as a recipe for future experiments. Therefore, more details on the growth procedure should be added to the methods section. This should include the growth rate, Se and V flux, and partial pressure during growth.
2. The VSe2 monolayer is assumed to be in the 1T phase. Do calculations for the 1H phase give a significantly different band structure that would not match de dI/dV spectra? Why was the 1T phase chosen?
3. What is the stacking sequence in the double layer of VSe2?
4. The specific structures should be well labeled in the figure caption. E.g. Fig. 3 does not state explicitly that the band structure is for a monolayer of VSe2 on NbSe2. The legend for the blue-yellow color scale in Fig. 3a is missing. How would the calculations look for a bi-layer?
5. Does the dI/dV signal vary across the islands?
6. Presumably, the magnetization measurements were done on the samples with mixed mono- and bilayer VSe2 coverage. The text only states "all the VSe2 samples". This could be mistaken for bulk samples. (In general, I suggest to check that the wording about the structures is precise throughout the manuscript, see also point 4.)
7. How large is the error margin for the superconducting gap in Fig. 5b?
8. The zero-bias peak at the islands' edges is tentatively assigned to a Kondo peak. This seems to be a rather vague interpretation. Did the authors try to measure the response in a magnetic field? Does the width or lineshape vary along the edge? Is there any correlation to the topography of the islands edge?
9. The authors may consider adding the band structure of NbSe2 to Fig. 2. It has been published many times, but would allow for a direct comparison of band structure and DOS.

Reviewer #3 (Remarks to the Author):

Authors report the electronic and magnetic properties of VSe2 monolayers on superconducting NbSe2 surface by combining STM/S, SQUID, and DFT calculations. The STM/S measurements demonstrate both absence of CDW and reduction of superconducting gap in VSe2 monolayers on NbSe2. The magnetic signature of the hybrid sample (ML VSe2 on NbSe2) was understood by means of theoretical modeling, which suggest ferromagnetic ground state in the hybrid sample. Here, I listed several comments and critiques.

1. For monolayer VSe2, there have been intense reports about its CDW and magnetism. So it is quite interesting to find out absence of CDW in ML VSe2 on NbSe2 bulk. It is noteworthy to deliberately explain the disappearance of CDW in terms of substrate, strain, and thickness, since in recent literature, the bilayer VSe2 turns out to become 1T' phase instead of showing CDW and the CDW of TiSe2 can be tuned by means of substrate. In the current manuscript, such discussion is lacking for understanding the missing CDW of VSe2 in accordance to the recent literature.

2. In case of VSe2, there have been many theoretical calculations to address the CDW and magnetic ground states, but they show diverse results for detailed ground states of VSe2 monolayers. However, I do not see how the current DFT calculation is compared to the previous ones. Detailed discussion is needed to address how the freestanding VSe2 monolayer and the hybrid structure are different from the other calculation literatures.

3. The main finding of this paper is about interaction between the magnetism of VSe2 monolayer and the superconductivity of NbSe2. However, the main message seems to be scattered among CDW, superconducting gap, and magnetism, so I wonder what is the key message to wide audience in condensed material field.

4. Although the authors present detailed experiments for magnetic properties of VSe2, the conclusion is not clear about magnetic origin among the VSe2 monolayer itself or its edge state. If the edge state is the major magnetic origin, then it is not clear whether the volume of edge portion is enough to give the macroscopic magnetic signal and whether edge magnetism is related to the superconducting gap reduction inside the VSe2 islands on NbSe2.

Reviewer #1

We thank the reviewer for his/her positive comments and for supporting the publication of our manuscript. The specific points are addressed in detail in the following.

Reviewer comment: *This manuscript reports a study of the electronic and magnetic properties of a VSe2 single layer grown on NbSe2. The main experimental technique is STM/STS, and the results are compared with DFT calculations. The key issue of interest is the magnetic state of VSe2. Some prior studies have suggested that single layer VSe2 is ferromagnetic, while other studies have indicated otherwise. This study adds to the ongoing debate. PPMS measurements of a capped sample shows a magnetic signature, but the Ms value is much smaller than that in VSe2/HOPG (Ref. 13). The authors' tunneling data do not match well with the calculations under different assumptions and are thus inconclusive. STM shows no evidence for a charge density wave (CDW), which differs from some prior studies. The authors suggested that the lack of CDW might be related to magnetic ordering.*

Author reply: The reviewer is correct in pointing out that there is a whole range of different results reported in the literature with samples grown on different substrates. Our bulk magnetization measurements show the presence of a ferromagnetic signal, but it is difficult to quantify it (as discussed in the SI Section IV). The value of the saturation magnetism from bulk magnetic measurements is large and the presence of a paramagnetic background signal makes it difficult to reliably estimate the magnetic moment per V atom. Similar difficulties probably also apply to the experiments on VSe_2 on HOPG referred to by the reviewer and the different saturation magnetization values do not necessarily reflect actual differences in the VSe_2 magnetization.

While the experimental STM spectroscopy does not fully match with any of the calculated spectra (see Fig. 3 and SI Section II), there are quantitative differences in the simulated results depending on, e.g., the choice of the DFT functional and the numerical value of the Hubbard U parameter as discussed at length in the SI Section II. We conclude that the measured spectra are most consistent with the simulations of the FM ground state. This is also consistent with the rest of the evidence pointing to ferromagnetic ordering: lack of CDW order (which is necessary but not a sufficient condition for magnetism) and the consistently and uniformly reduced superconducting gap of the $NbSe_2$ on the VSe_2 islands.

As a summary, while the large bias scale spectroscopy (Fig. 2) is maybe not sufficient to single out the FM ground state, the lack of CDW in the STM images and the effect of the VSe_2 on the $NbSe_2$ superconducting gap strongly suggests at least local ferromagnetic ordering.

Action: In addition to the other changes to the manuscript in response to all the referee comments (see below for details), the conclusion has been highlighted in the revised manuscript on p. 8 “Even though the magnetic response of VSe_2 is rather complex, taken together with the lack of CDW in the STM images, our STM and STS results are the most consistent with simulated response of the FM phase.”

Reviewer comment: *After reading through the relevant publications, it appears that the different conclusions from the various studies are not necessarily contradictory and could be simply related to the different sample configurations and environments. DFT calculations in the present work (and also in prior studies) show that the energy differences between the different states (CDW, magnetic, and nothing) are tiny and depend on the details of the calculations. Therefore, it seems reasonable that differences in the substrate choice (metals, semimetals, semiconductors, …), presence or absence of capping layer, and contamination by non-UHV processing conditions as in some prior studies could very well be the source of*

the divergent observations and opinions. The ferromagnetic state could be caused by "extrinsic effects" based on the majority of evidence in the literature.

Specifically, it is unclear that the PPMS data for a capped sample and the STM/STS data for a pristine sample in the present study can be directly correlated. The samples are just different, and they could be in different states. This should be emphasized in the manuscript. Ideally, one would want to perform PPMS measurement on a sample decapped under UHV, but it is probably a hard experiment. The authors' statement that "… lack of charge density wave signatures indicates magnetization of the sheet..." is simply a conjecture, as there is no direct proof that the UHV sample is in fact magnetic. CDW can be suppressed for a number of other reasons including charge transfer, lattice distortion, impurities, and electronic coupling to the substrate. The authors' strong statement is simply not justified. STS measurements of the electronic structure may in principle be able to settle this issue; if one sees exchange splitting of the bands that diminishes toward the Curie temperature, the case would be clear. Unfortunately, the STS data presented in the manuscript are not well correlated with the calculated density of states.

Author reply: The referee is pointing out that the energy differences between the different ground states (CDW, magnetism etc.) of the VSe_2 monolayer are small and minor perturbations due to the substrate, capping, sample quality, adsorbates etc. could be decisive in terms of the differing observed behavior. We agree that the balance between the different ground states is subtle and small extrinsic changes could tilt the balance in different directions. We have rephrased some parts of the revised manuscript to better reflect this.

While temperature-dependent STS experiments could in principle shed further light into how the electronic ground state changes as a function of temperature (e.g. through the possible Curie point), considering the wide temperature range that would have to be accessed and the fact that the STM experiment should be carried out at the same atomic location of the sample, this experiment is not feasible.

Finally, the bulk magnetisation measurements show a ferromagnetic signal even though the paramagnetic background makes it difficult to quantify the corresponding saturation magnetization. On the other hand, the STS experiments related to the reduction of the superconducting gap of the $NbSe_2$ substrate strongly suggest magnetism of the VSe_2 sheet, which is consistent with no major differences between the capped samples used in the PPMS measurements and the uncapped samples used in the STM experiments. The samples were grown under identical conditions using the same setup.

Action: We have removed the sentence in the abstract to remove the perceived causal link between lack of CDW and ferromagnetism: “Low temperature scanning tunneling microscopy (STM) measurements show an absence of the typical charge density wave on VSe_2 and demonstrate a reduction of the superconducting gap of $NbSe_2$ on the VSe_2 layer. This suggests magnetization of the VSe_2 sheet, at least on the local scale.”

Added sentence (page 9): “Furthermore, as indicated by the DFT calculations, the energy differences between the different ground states (CDW, magnetism etc.) of the VSe_2 monolayer are small. Minor perturbations due to the substrate, capping, sample quality, adsorbates etc. could cause variations in the observed behavior. However, local measurements (such as STM) should still probe the ferromagnetic phase suggested in the data above that corresponds to the DFT calculations.”

Reviewer comment: *Another concern is sample quality, which could be an issue for many of the studies to date. The authors stated that the VSe2 islands show sharper edges at higher growth temperatures, but at the same time the CDW modulation of the NbSe2 substrate*

becomes weakened because of intercalation. One wonders if the CDW state of VSe2 might be similarly affected. It is also curious that the authors annealed their cleaved NbSe2 samples at 600 K for 1 h before film growth. Is the annealing step needed? Cleaved samples should have a pristine surface. Annealing could deplete Se and create defects.

Author reply: The referee is correct in pointing out the importance of the sample preparation. When the substrate is held at a higher temperature, we do indeed observe significant vanadium intercalation. However, the VSe_2 lattice constant does not depend on the specific island and we do not observe sample to sample variations. In addition, vanadium intercalation does not seem to affect the lattice constant and we do not observe a charge density wave on VSe_2 in any of the cases. Fig. R1 shows atomically resolved images on two completely different sample preparations carried out at different substrate temperatures of 270 °C and of ca. 300 °C, respectively. Fig. R1a corresponds to the usual sample preparation conditions (the slightly visible superstructure matches the CDW of the underlying $NbSe_2$ substrate). Fig. R1b shows an image from a sample preparation carried out at a higher temperature, where we get significant vanadium intercalation (visible as the background contrast variation). In both cases, the VSe_2 lattice constant is 3.5±0.1 Å and we do not observe a CDW.

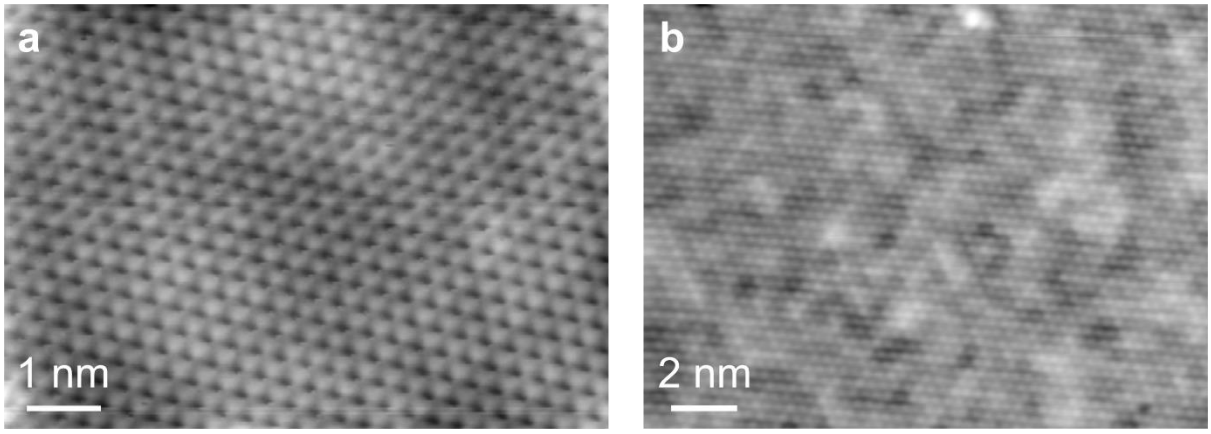

Figure R1. Atomically resolved STM images of the VSe_2 lattice. (a) Sample corresponding to the usual preparation conditions. (b) VSe_2 growth carried out at a higher temperature resulting in significant intercalation of vanadium.

As the referee points out, we do anneal our cleaved $NbSe_2$ samples at 573 K for 30 min. This is to outgas the epoxy glue used to attach the $NbSe_2$ samples onto the sample holder, which is important for the subsequent growth step of VSe_2 where the sample is also heated. We have checked the quality of the $NbSe_2$ surface after annealing and there are no discernible differences to samples that have only been cleaved.

Action: Data in Fig. R1 has been added to the Supplementary Information of the revised manuscript. We have rewritten the Methods section to give the details on the outgassing procedure and noted that it does not adversely affect the substrate quality.

Reviewer comment: *A 3% lattice mismatch is large. Is the VSe2 lattice constrained to that of the NbSe2 in the calculations? This could affect the ground state. Please clarify the assumptions.*

Author reply: The 3% lattice mismatch mentioned by the Reviewer was measured from STM images. The revB86b-calculated mismatch is about 4.5% and 1.5% without U parameter and with U=2 eV, respectively. Although the strain state of the VSe_2 in the experiment is not known accurately, it appears that the VSe_2 layer is mostly unstrained and that is why most of the presented results (energy differences in particular) are obtained from unstrained systems.

Only when determining the stacking sequence between $NbSe_2$ and VSe,, the heterostructure with strained layers was used.

The energy difference between the FM and CDW phases were given in Table S1 and calculated using the optimized lattice constant for each phase. If using PBE or revB86b functionals without U-parameter, the energy difference is very small (<~10 meV) and strain can certainly affect the ground state. When even a moderate U-parameter is used (e.g. 2 eV), the difference becomes fairly large (~160 meV) and it is unlikely that the ground state could be changed by strain.

Reviewer comment: *The tunneling current of STM is typically confined to a cone angle. As a result, STS typically measures the density of states with a strong weighting along the surface normal direction. The tunneling spectra might resemble more a one-dimensional density of states. Please clarify.*

Author reply: The reviewer is referring to the momentum distribution of the tunneling electrons and how they sample different (in-plane) momenta in the substrate Brillouin zone. While one would not expect strong weighting within the usual picture of STM, where the tip can be modelled as a localized wavefunction (Tersoff-Hamann model) and there is no momentum conservation, there are indeed examples where this effect does take place. These include experiments on graphene (on h-BN) by the Crommie group (Giant phonon-induced conductance in scanning tunnelling spectroscopy of gate-tunable graphene, Nat. Phys. 4, 627 (2008)). In addition, some experiments have demonstrated that the momentum distribution of the tunneling electrons can be sensitive to the microscopic tip apex (e.g. P. Leicht et al. In Situ Fabrication Of Quasi-Free-Standing Epitaxial Graphene Nanoflakes On Gold, ACS Nano 8, 3735 (2014)). However, we do not observe strong variations in the d*I*/d*V* spectra depending on the microscopic tip apex. In addition, the most prominent features on the VSe_2 spectra close to the Fermi level are predicted to arise from the high density of states from bands close to the Γ point. These would not be expected to be affected by momentum selection of the tunneling electrons.

Action: We have added a short note (page 8) on this effect in the revised manuscript.

Reviewer comment: *The introduction contains a good review of different kinds of composite quantum materials. Maybe it could be made more concise by focusing specifically on the VSe2 case.*

Author reply: Communications Physics does not have a strict length restriction on the articles it publishes. We feel that introduction motivating the interest in this type of systems is warranted.

Reviewer comment: *In summary, the experiments seem to be carried out with care. The analysis, interpretation, and discussion are not necessarily accurate and could be improved. The different sample conditions and environments for the different studies should be pointed out and considered as a possible reason for the different conclusions.*

Author reply: We thank the reviewer for his / her constructive criticism and have considerably revised the manuscript along these lines.

Reviewer #2

Reviewer comment: *This is an interesting paper on the in-situ growth and characterization of heterostructures of VSe2/NbSe2, with the experimental results being compared to DFT calculations. Interestingly, the VSe2 layer seems to host magnetism, which affects the superconducting gap of the underlying NbSe2 substrate.*

The successful growth and results are interesting and deserve publication in Communications Physics. However, I would like to see some more discussion/information concerning these aspects:

Author reply: We thank the reviewer for his/her positive comments and the specific points are addressed in detail in the following.

Reviewer comment: *1. The growth of atomically clean layers is an important aspect of this paper and shall serve as a recipe for future experiments. Therefore, more details on the growth procedure should be added to the methods section. This should include the growth rate, Se and V flux, and partial pressure during growth.*

Author reply: The referee is correct to point out the importance of the sample preparation procedure and we have added a detailed sample preparation procedure in the Methods section of the revised manuscript.

Action: Methods section has been rewritten**.**

Reviewer comment: *2. The VSe2 monolayer is assumed to be in the 1T phase. Do calculations for the 1H phase give a significantly different band structure that would not match de dI/dV spectra? Why was the 1T phase chosen?*

Author reply: VSe_2 is paramagnetic in bulk form and commonly crystallizes in the 1T structure. DFT calculations predict that either the 2H and 1T phases could be the lowest energy structure with the answer depending sensitively on the correlations of the V d-shell electrons incorporated through the Hubbard U parameter in DFT+U simulations (Phys. Chem. Chem. Phys. 18, 33047–33052 (2016)). Despite the theoretical prediction of the existence of the 2H-VSe_2, all of the recent experimental results using MBE report the formation of the 1T phase with no evidence of a successful growth of 2H phase.

We have added to SI results of calculations for the 2H-phase of VSe_2. In addition to the basic material properties, we show the band structures and the simulated STS of 2H-VSe_2 calculated using revB86b with U-parameter equal to 0, 2, and 5 eV (Figs. S6 and S14 and Table II).

2H phase of VSe_2 is semiconducting or semimetallic (depending on the value of the U parameter; for higher values of U, the indirect gap becomes negative) while the 1T phase remains metallic even in the monolayer limit. Both phases are predicted to have ferromagnetic ordering with in-plane spin alignment.

Action: We have added a note in the text on the possibility of the 2H phase and the details are given in the Supplementary Information.

Reviewer comment: *3. What is the stacking sequence in the double layer of VSe2?*

Author reply: The calculated binding energies for different stackings are collected in the table below, where we have adopted the same notation as in Table SIII and the first layer is always fixed at AbC.

	AbC	AcB	BcA	BaC	CaB	CbA
E_b (eV)	-0.226	-0.216	-0.205	-0.216	-0.136	-0.135

The AbC/AbC stacking, which also corresponds to the bulk AA-stacking is the most stable also in the case of bilayer.

Action: We have added this information in the revised version of the SI.

Reviewer comment: *4. The specific structures should be well labeled in the figure caption. E.g. Fig. 3 does not state explicitly that the band structure is for a monolayer of VSe2 on NbSe2. The legend for the blue-yellow color scale in Fig. 3a is missing. How would the calculations look for a bi-layer?*

Author reply / action: The calculations in Fig. 3 are for an unstrained monolayer VSe2 and the colors reflect the PDOS of vanadium (blue) and selenium (orange). We have now stated these explicitly in the figure caption and added a legend for the color scale.

We have added the band structure and density of states for the bilayer (in the lowest energy AA-stacking) to SI (Fig. S16). As expected, all bands are doubled and split by the interlayer interaction. In particular, density of states shows splitting of those peaks that contribute most to the STS, which also agrees well with the observed changes between the monolayer and bilayer spectra.

Reviewer comment: *5. Does the dI/dV signal vary across the islands?*

Author reply: We do not observe noticeable variation of the large bias scale d*I*/d*V* spectra (Fig. 2 in the main text) over the islands. In fact, the d*I*/d*V* spectra in Fig. 2a represent an average of a line spectra on the island. We also do not observe a variation of the superconducting gap over the island which can be seen in the supporting info (Fig. S19). However, the zero bias peak is inhomogeneously distributed along the edges of the VSe_2 islands and there are strong intensity variations as illustrated in Fig. 5d. In addition to the spatial distribution of the zero bias peak, its width is also strongly position dependent. We observe both zero bias peaks that are confined within the superconducting gap (e.g. Fig. 5c), but on some other locations (see Fig. S19), its width can be a couple of times larger than the superconducting gap width.

Action: The lack of variation in the middle of the VSe_2 islands is now explicitly stated in the revised manuscript (page 6).

Reviewer comment: *6. Presumably, the magnetization measurements were done on the samples with mixed mono- and bilayer VSe2 coverage. The text only states "all the VSe2 samples". This could be mistaken for bulk samples. (In general, I suggest to check that the wording about the structures is precise throughout the manuscript, see also point 4.)*

Author reply / action: The magnetization measurements were done on the samples with mixed mono- and bilayer VSe_2 coverage. In the particular sample shown in the manuscript, the total coverage of VSe_2 is 0.57 with 0.44 monolayer and 0.13 bilayer present. This is now also explicitly stated in the revised manuscript (Methods section).

Reviewer comment: *7. How large is the error margin for the superconducting gap in Fig. 5b?*

Author reply: We have estimated the error bar of the fits shown in the Supporting Information (Fig. S18). They are 0.02, 0.02, and 0.04 meV for the points shown in Fig. 5b and the error bars are shown in the revised figure.

Reviewer comment: *8. The zero-bias peak at the islands' edges is tentatively assigned to a Kondo peak. This seems to be a rather vague interpretation. Did the authors try to measure the response in a magnetic field? Does the width or line shape vary along the edge? Is there any correlation to the topography of the islands edge?*

Author reply: The zero bias peak is inhomogeneously distributed along the edges of the VSe_2 islands and there are strong intensity variations as illustrated in Fig. 5d. In addition to the spatial distribution of the zero bias peak, its width is also strongly position dependent. We observe both zero bias peaks that are confined within the superconducting gap (e.g. Fig. 5c main text), but on some other locations (see SI Fig. S19), its width can be a couple of times larger than the superconducting gap width. These characteristics would fit a Kondo peak arising from unpaired spins at the edges of the VSe_2 islands. The Kondo peak is typically spatially localized very strongly over the spin, which is in-line with the strong spatial fluctuations that we observe. The width of the Kondo peak is proportional to the Kondo temperature, which is determined by the exchange coupling of the isolated spin with the Fermi sea of the underlying metallic substrate. The exchange coupling depends very strongly on the atomic scale details (e.g. adsorption site) – again consistent with our experimental results.

We have performed a measurement of the zero bias peak under an external magnetic field of up to 2 Tesla (out of plane) and the results are shown in Fig. R2.

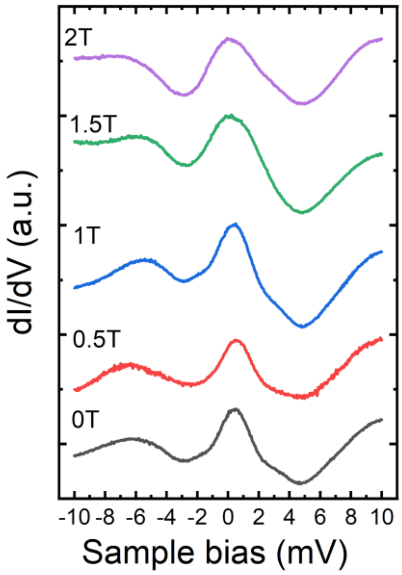

Figure R2. Zero bias peak response under an out-of-plane external magnetic field.

It can be seen in Fig. R2, the zero bias peak width gets broader as the external magnetic field increases which is typical for the Kondo peak under moderate magnetic field (the peak should split at sufficiently high field). Unfortunately, we do not have data at higher external field.

Reviewer comment: *9. The authors may consider adding the band structure of NbSe2 to Fig. 2. It has been published many times but would allow for a direct comparison of band structure and DOS.*

Author reply / action: We have realized this by adding a figure in the SI which combines the band structure with the calculated DOS and the experimental and simulated d*I*/d*V* spectra.

Reviewer #3

Reviewer comment: *Authors report the electronic and magnetic properties of VSe2 monolayers on superconducting NbSe2 surface by combining STM/S, SQUID, and DFT calculations. The STM/S measurements demonstrate both absence of CDW and reduction of superconducting gap in VSe2 monolayers on NbSe2. The magnetic signature of the hybrid sample (ML VSe2 on NbSe2) was understood by means of theoretical modeling, which suggest ferromagnetic ground state in the hybrid sample. Here, I listed several comments and critiques.*

Author reply: We answer the specific comments of the reviewer in the following.

Reviewer comment: *1. For monolayer VSe2, there have been intense reports about its CDW and magnetism. So it is quite interesting to find out absence of CDW in ML VSe2 on NbSe2 bulk. It is noteworthy to deliberately explain the disappearance of CDW in terms of substrate, strain, and thickness, since in recent literature, the bilayer VSe2 turns out to become 1T' phase instead of showing CDW and the CDW of TiSe2 can be tuned by means of substrate. In the current manuscript, such discussion is lacking for understanding the missing CDW of VSe2 in accordance to the recent literature.*

Author reply: We have addressed this issue by adding discussion and references to relevant literature on the possible role of the substrate for the disappearance of the CDW.

Action: We have added flowing sentences in the revised version of the manuscript (page 9) and cited relevant literature: "Based on the recent experimental works (*Nanoscale*, 2019, 11, 20096-20101; *Adv. Quantum Technol.* 2018, 1, 1800070), the absence of the CDW in our system might also be related to the underlying $NbSe_2$ substrate. Our DFT calculations also suggest that the stability of the CDW is sensitive to the strain, doping and amount of the defects in the system as shown in SI."

Reviewer comment: *2. In case of VSe2, there have been many theoretical calculations to address the CDW and magnetic ground states, but they show diverse results for detailed ground states of VSe2 monolayers. However, I do not see how the current DFT calculation is compared to the previous ones. Detailed discussion is needed to address how the freestanding VSe2 monolayer and the hybrid structure are different from the other calculation literatures.*

Author reply / action: Indeed, calculations have reported that the stable phase of VSe_2 can be H-phase FM or T-phase FM, AFM, or CDW (with different periodicities), depending on the choice of adopted functional, strain, doping. Experiments are assigned mostly to 1T-FM and 1T-CDW.

Summarizing findings from the DFT calculations:

1. LDA tends to favor CDW both in bulk and in monolayer.
2. PBE tends to favor FM both in bulk and in monolayer.
3. PBE+U further increases the magnetic moments and favors magnetic phases.
4. LDA+DMFT yields CDW bulk and FM monolayer. The magnetic moments are larger than in LDA but smaller than in PBE.
5. H-phase is more stable with PBE, but T-phase with PBE+U.

Since it is also known that tensile strain increases the magnetic moments and favors magnetic phases, it is possible that the difference between PBE and LDA originates from lattice constant

difference, i.e., larger lattice constant yields more localized d-orbitals. While it is generally expected that the hybrid functionals would improve the description of the local electronic correlations, hybrid functionals have not been employed widely in the literature. In agreement with Popov et al we find H-phase slightly lower in energy with HSE.

Clearly, it is difficult to choose the functional without comparison to experiments. Our experimental results show large lattice constant and no CDW, and, as mentioned, large lattice constant correlates with the existence of FM phase. We have thus adopted PBE+U functional that reproduce those findings. Obviously, previous studies where CDW were observed had chosen LDA since it reproduces those findings. It is possible that the real origin of the different behaviors originates from e.g. strain or doping, but when those are not known, we can only choose functional that best reproduces the experiments.

In the original version of the manuscript we did not want to delve deeply into these partially conflicting results in the literature, and simply mentioned that "there is a delicate balance between different competing interacting states and phases in monolayer transition-metal dichalcogenides, which may also depend on the nature of the substrate [35,36,39–43].", where most of the cited references contain DFT calculations.

In the revised version, we have included a bit more discussion of the previous calculations:
"According to previous DFT calculations the lowest energy phase of VSe2 can be H-phase or T-phase, in the FM or AFM configuration, with or without CDW, depending on the choice of adopted functional, strain and doping [35,39,42,43,47,48]. Despite the theoretical prediction of the existence of the 2H-VSe2, all of the recent experimental results using MBE report the formation of the 1T phase with no evidence of a successful growth of 2H phase. We also tested several different functionals, as shown in SI, and have chosen vdw-DF functional revB86b with +U parameter of 2 eV, since it reproduces the larger lattice constant and the lowest energy phase being ferromagnetic 1T-VSe2. In contrast, LDA would lead to CDW phase and PBE to H-phase, as in the previous reports [35,39,42,43,47]."

We feel that more detailed review of the computational results is not warranted here. We also added citation to Popov et al., Phys. Chem. Chem. Phys. 18, 33047–33052 (2016) to the manuscript.

Reviewer comment: *3. The main finding of this paper is about interaction between the magnetism of VSe2 monolayer and the superconductivity of NbSe2. However, the main message seems to be scattered among CDW, superconducting gap, and magnetism, so I wonder what is the key message to wide audience in condensed material field.*

Author reply: On a broad scale, our manuscript contributes to the efforts in designer materials that would realize electronic responses not found in naturally occurring materials. The idea is to create heterostructures combining different materials, where the desired physics emerges from the engineered interactions between the different components. Van der Waals heterostructures are ideally suited for this purpose as the different layers interact only through vdW forces and hence retain their intrinsic properties. This results in very high-quality interfaces and allows for a rational design of the emergent properties of the heterostructure.

Combining magnetism and superconductivity are the key ingredients in realizing topological superconductivity, which is the main motivation for our work. The standard "recipe" for topological superconductivity is through a combination of out-of-plane ferromagnetism, superconductivity and Rashba-type spin-orbit coupling. On the other hand, other schemes exist: it has been predicted that nodal topological superconductivity can be realized in $NbSe_2$ by fabricating nanostructured ferromagnets with an in-plane magnetization on the top surface

(S. Głodzik, T. Ojanen, Engineering nodal topological phases in Ising superconductors by magnetic superstructures, New J. Phys. 22, 013022 (2020)).

In order to test these kinds of proposals, we need to experimentally realize the heterostructure with sufficiently high-quality interfaces and edges, and to look for the presence of the proposed edge modes. In addition to this, we hope that our manuscript contributes to the understanding of the (complicated) magnetic response of VSe_2.

Action: We have revised the conclusions paragraph to better reflect the above considerations.

Reviewer comment: *4. Although the authors present detailed experiments for magnetic properties of VSe2, the conclusion is not clear about magnetic origin among the VSe2 monolayer itself or its edge state. If the edge state is the major magnetic origin, then it is not clear whether the volume of edge portion is enough to give the macroscopic magnetic signal and whether edge magnetism is related to the superconducting gap reduction inside the VSe2 islands on NbSe2.*

Author reply: We do not think the edge magnetism is a major contribution to the bulk magnetic signal of VSe_2. The STS experiments in the middle of the VSe_2 islands show very homogeneous gap reduction and there does not seem to be any kind of decay away from the edges. This suggests that the bulk of the VSe_2 islands have homogeneous magnetization. In addition, it is also clear that there is a magnetic signal related to the edges that is very localized to a few unit cells from the island edge.

Action: This has now been stated explicitly in the revised manuscript.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors have made a number of clarifications and improvements. The discussion is now more open to or more explicit about the lack of definitive proofs of the connection in regard to magnetism in different types of samples and the connection between CDW, magnetism, and superconductivity. This referee remains concerned: (1) the lack of CDW in VSe2 could be caused by many other issues including coupling to the substrate and interference from the CDW state in the NbSe2 substrate itself (HOPG and graphene do not have CDWs, by comparison). (2) The reduction of the superconducting gap as measured over the film, which is not naturally superconducting, is expected. The rate of gap reduction was taken by the authors as an indication of magnetic ordering, but this remains a conjecture. The case of Bi2Se3/NbSe2 does not provide a rigorous comparative test. The decay length of the gap can depend on many other things (like the density of normal carriers in the film).

The authors should take another look at Fig. 5b. The red dashed curve seems to have a decay length of about 1.4 or 1.5 nm, rather than 1 nm as labeled in the figure and stated in the text (1.1*e^-1 = 0.405). The blue dashed curve does not seem to correspond to a decay length of 2 nm at all. Please doubt check and correct as appropriate.

On balance, it seems OK to proceed with acceptance after the authors consider the comments above.

Reviewer #2 (Remarks to the Author):

The authors have adequately addressed all my remarks and incorporated the required changes into the manuscript. I recommend publication in its current form.

Reviewer #3 (Remarks to the Author):

To Authors,
The revised manuscript resolved the most of issues raised. I am satisfied with the current version of manuscript for the acceptance for publication to the Communication Physics.

Reviewer #1:

We thank the reviewer for his/her positive assessment of our revised manuscript. We answer his/her remaining concerns in detail below.

Reviewer comment: *"This referee remains concerned: (1) the lack of CDW in VSe2 could be caused by many other issues including coupling to the substrate and interference from the CDW state in the NbSe2 substrate itself (HOPG and graphene do not have CDWs, by comparison). (2) The reduction of the superconducting gap as measured over the film, which is not naturally superconducting, is expected. The rate of gap reduction was taken by the authors as an indication of magnetic ordering, but this remains a conjecture. The case of Bi2Se3/NbSe2 does not provide a rigorous comparative test. The decay length of the gap can depend on many other things (like the density of normal carriers in the film). "*

Author reply: Point (1) – the reviewer is correct and we already state in the manuscript (end of page 9) "Based on the recent experimental works[54,55], the absence of the CDW in our system might also be related to the underlying $NbSe_2$ substrate".

Point (2) – We have modified the sentence in the manuscript (middle of p. 12) from "The faster decay we observe in VSe_2 is most easily explained by magnetism of the VSe_2 layer." to "A possible explanation for the faster decay we observe in VSe_2 would be given by magnetism of the VSe_2 layer."

Reviewer comment: "*The authors should take another look at Fig. 5b. The red dashed curve seems to have a decay length of about 1.4 or 1.5 nm, rather than 1 nm as labeled in the figure and stated in the text (1.1*e^-1 = 0.405). The blue dashed curve does not seem to correspond to a decay length of 2 nm at all. Please doubt check and correct as appropriate.* "

Author reply: We thank the referee for pointing out this inconsistence. We had accidentally fitted our data with function of the form y = A1exp(-x/A2)+A3, i.e. with an unphysical offset term. Doing this correctly changes the numerical values to $\lambda = 1.3 \pm 0.1$ nm for VSe_2 and the literature data for Bi_2Se_3 to $\lambda = 4.3 \pm 0.3$ nm. This does not alter the conclusion that VSe_2 exhibits much faster decay than Bi_2Se_3.

Action: We have changed the fits in Fig. 5b and the corrected the values of the decay constants in the text.