Response to comment on "Manifolds of quasi-constant SOAP and ACSF fingerprints and the resulting failure to machine learn four-body interactions"

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It is uncontested that a machine learning scheme can not correctly reproduce physical properties that vary on a manifold in configuration space if the fingerprint, used as an input for the machine learning scheme, is constant on this manifold. In our original paper (Ref.[1]) we discovered manifolds of quasi-constant fingerprint for SOAP and ACSF fingerprints for test environments that contain 3 or 4 atoms around the central atom. Standard parameters were used for both fingerprints. We defined a quasi constant fingerprint as a fingerprint whose variation on the manifold is so small that the machine learning schemes behaves as if it was exactly constant. There is obviously a threshold for the variation of the fingerprint on the manifold above which machine learning will again become possible. In our original paper we found several manifolds whose fingerprint variation is sufficiently small to prevent machine learning based on a standard training scheme even if configurations on the manifold are included in the training set.

The authors of the comment claim that structures on our manifold of quasi constant fingerprint can be machine learned. While we used a standard training scheme where the structures used for training are chosen in the usual way according to physical criteria, the authors of the comment use an entirely non-standard training scheme that has never been used before for machine learning of potential energy surfaces. In this scheme the selection of the configurations to be fitted is dictated by the deficiencies of the SOAP fingerprint, requiring the calculation of a manifold of quasi constant fingerprint. An empirical weight coefficient for the terms in the loss function that contain manifold structures has then to be determined to give a larger weight to configurations on the manifold during the fitting process. Determining the manifold and the weights is a cumbersome process that is unlikely to be ever used in practice. Since this fitting scheme is constructed with the intention to compensate for the deficiencies of the SOAP fingerprint, we will refer in the following to this scheme as deficiency-adapted scheme.

For a given set of SOAP parameters one can construct many manifolds by starting from different initial structures. Even though manifolds that were constructed from identical initial configurations but with different parameters look similar by eye, they differ considerably in the central property, namely the variation of the fingerprint. In our publication we constructed a manifold, with the SOAP parameters $n_{max} = l_{max} = 16, \sigma = 0.3$ which are different from the parameters of $n_{max} = l_{max} = 8, \sigma = 0.2$ used in the comment. As shown in Fig. 1, the variation of our fingerprint is about an order of magnitude smaller than for the manifold used in the comment.

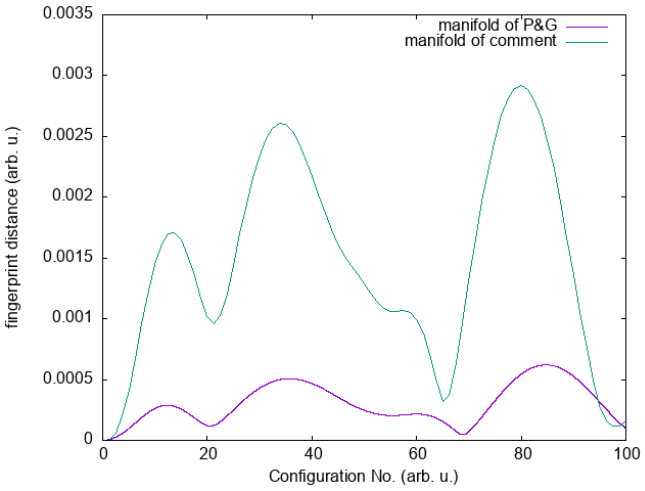
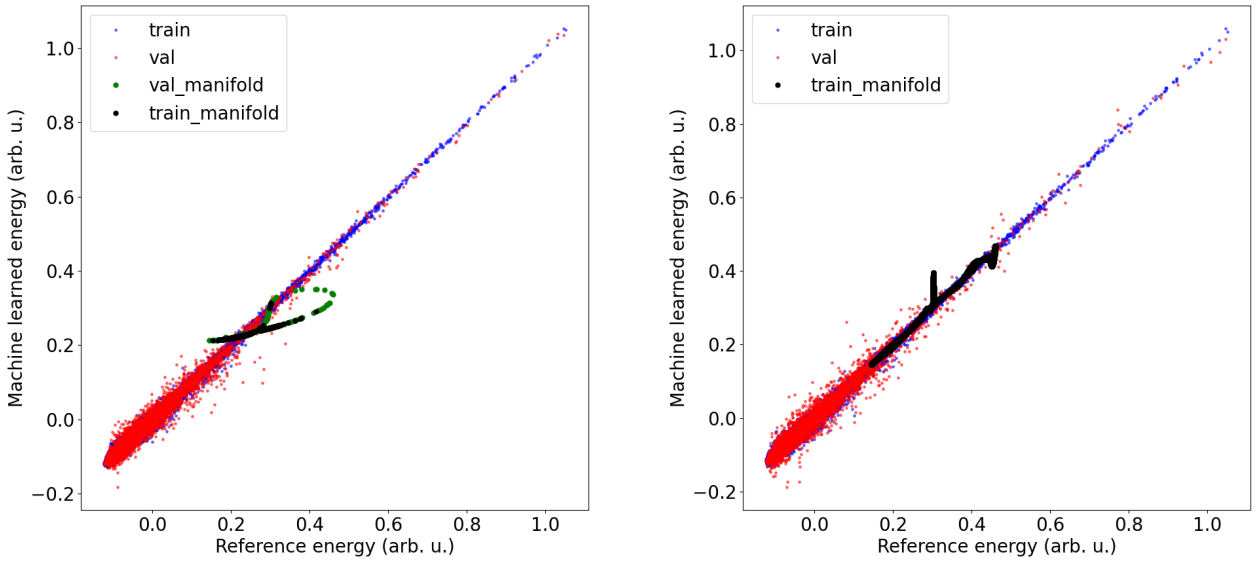


FIG. 1: The variation of the fingerprint for our original manifold[1] (P&G) compared to the variation of the fingerprint on the manifold of the comment. The variation along our original manifold is about an order of magnitude smaller than the one for the manifold of the comment.

In spite of these important differences, the comment gives the wrong impression that the manifolds are identical. So the phrase in the comment "However, contrary to what is claimed in Ref. 1, a SOAP-based model can approximate the energy along the 'quasi-constant' manifolds" is misleading. It was not to be expected that the authors of the comment obtain the same result as we did since they used a milder manifold in connection with a deficiency-adapted training scheme. Actually in reality the results of the comment confirm and extend our results. While we have shown that machine learning based on standard training fails for tight manifolds of small fingerprint variation, they have shown that it even fails for milder manifolds with a larger fingerprint variation.

The training of a neural network is a non-convex global optimization problem that does not have a unique solution. Relatively small changes in the methodology can therefore lead to different results. To investigate the purely academic question of whether structures on a quasi constant manifold can be machine learned if a sufficiently large effort is made, we therefore performed some additional tests using the Kernel Ridge Regression (KRR) method which gives a unique solution by solving a linear system of equations. For the construction of a Gaussian kernel matrix only two parameters are nec-



(a) (b)

FIG. 2: Machine learning results obtained with the deficiency-adapted training scheme for our original manifold[1]. 30'000 ordinary structures were obtained from a molecular dynamics run. Out of them 24'000 were used for the training set and 6000 for the validation set. In addition we created 16'000 structures that are lying on a single manifold. Panel a) shows that the manifold can not be learned when 1'000 of the manifold structures are added to the training set and the remaining 15'000 manifold structures are added to the validation set. This corresponds to a weight of 5 in the comment. Panel b) shows the result of adding all the 16'000 manifold structures to the training set, which corresponds to a weight of 80. Even in this case the deficiency-adapted method can only partially learn the manifold with the type of network training that we used in all previous investigations.

essary. So in contrast to neural network methods the parameter space can be fully scanned to find the optimal parameter set. The results based on such a scan are summarized in Fig. 3. Putting more and more weight on the manifold structures by increasing the number of manifold structures in the training set to very large values clearly improves the learnability of manifold structures.

Contrary to what a reader might think now, going through the painful process of determining a manifold and adapting the training scheme to it, does not cure the problem of applying the method to the calculation of potential energy surfaces. As pointed out in our publication, there exists a huge number of manifolds and machine learning one manifold does not mean that any of the other manifolds can be described correctly. An example of this is shown in Fig 4 where manifold 1 is the one from the authors of the comment. Manifold 2 was constructed from a different initial structure with a larger four body energy. One can clearly see that including one manifold does not at all improve the quality of the fit for the other manifold.

Since in a physical application one needs a machine learning model that describes correctly all the manifolds, our statement that the manifolds of quasi constant fingerprint prevent machine learning of four-body energies is true even in the case of mild manifolds with a relatively large variation of the fingerprint.

During the motion of the atoms on the manifold the H-H distances vary considerably. Hence the degeneracy will be lifted if the atomic environment fingerprints of all atoms are used to describe the molecule. So torsional energies might be represented as some long range pairwise interactions in machine learning schemes based on the SOAP fingerprint. It is at present not known how much such a compensation that is not in agreement with the standard chemical understanding of the origin of torsional energies will limit the accuracy of machine learned potential energy surfaces for systems where torsional motions are important.

While for the ACSFs it is clear that radial basis functions describe two-body terms and angular functions three-body terms and that therefore higher order terms are missing, such a connection can not be established for the SOAP fingerprint. Our analysis shows however that SOAP has the same intrinsic limitations to three body terms. Since the ACSF fingerprints are much shorter and much faster to calculate[2] they are probably for most applications the better compromise between efficiency and

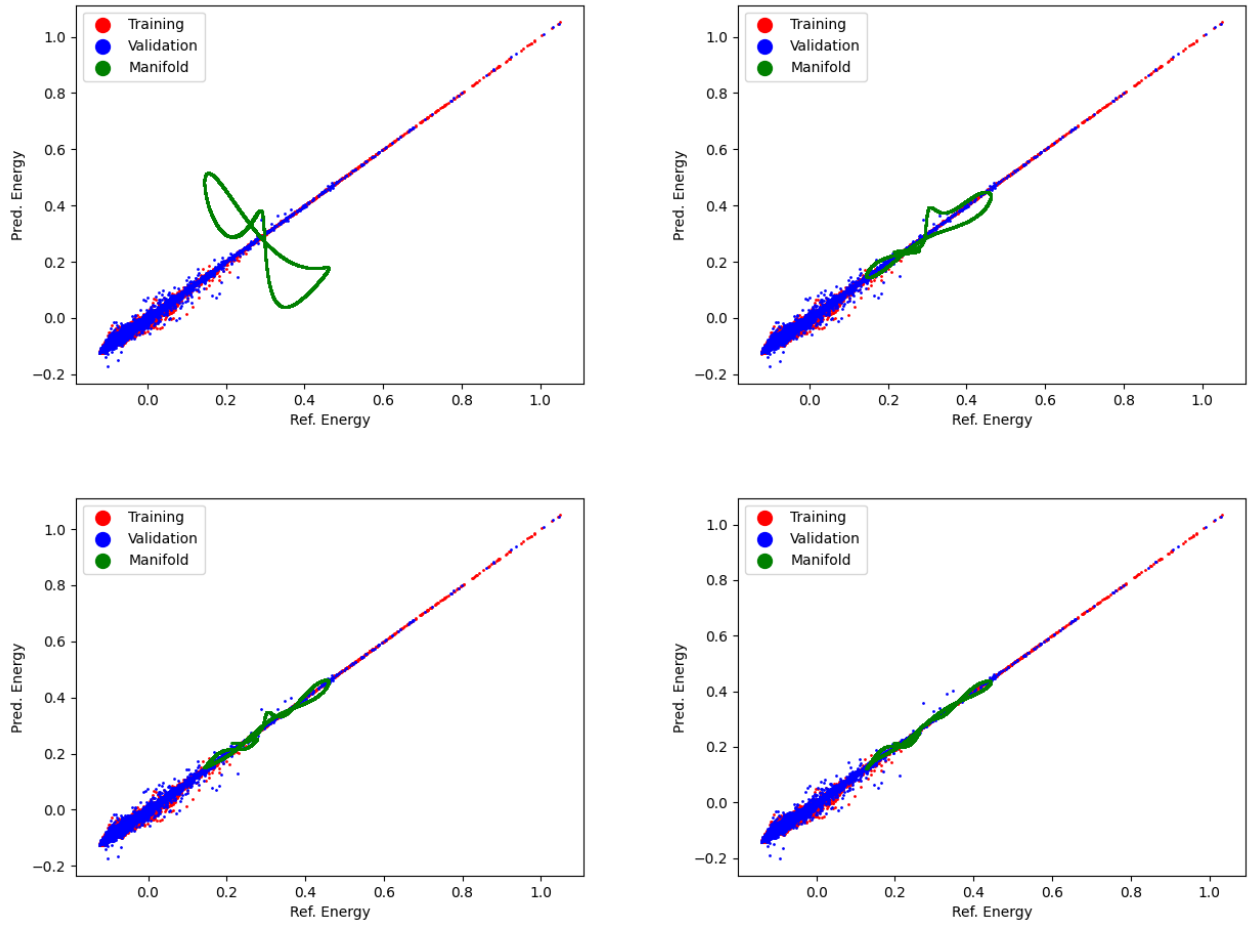


FIG. 3: Machine learning results obtained with the KRR method for the same data set as in Fig 2. Going from the left to the right panel and from the top to the bottom line, no structures, 16, 160 and 1600 structures on the manifold were included in the training set.

accuracy. If torsional terms are expected to be important, fingerprints[3,4] can be used that allow to include in a systematic way higher order terms.

The data and the codes on which our conclusions are based will be made available in the supplementary material.

[1]B. Parsaeifard and S. Goedecker, "Manifolds of quasi-constant soap and acsf fingerprints and the resulting failure to machine learn four-body interactions," The Journal of Chemical Physics **156**, 034302 (2022).

[2]Y. Zuo, C. Chen, X. Li, Z. Deng, Y. Chen, J. Behler, G. Csányi, A. V. Shapeev, A. P. Thompson, M. A. Wood, *et al.*, "Performance and cost assessment of machine learning interatomic potentials," The Journal of Physical Chemistry A **124**, 731–745 (2020).

[3]A. V. Shapeev, "Moment tensor potentials: A class of systematically improvable interatomic potentials," Multiscale Modeling & Simulation **14**, 1153–1173 (2016), https://doi.org/10.1137/15M1054183.

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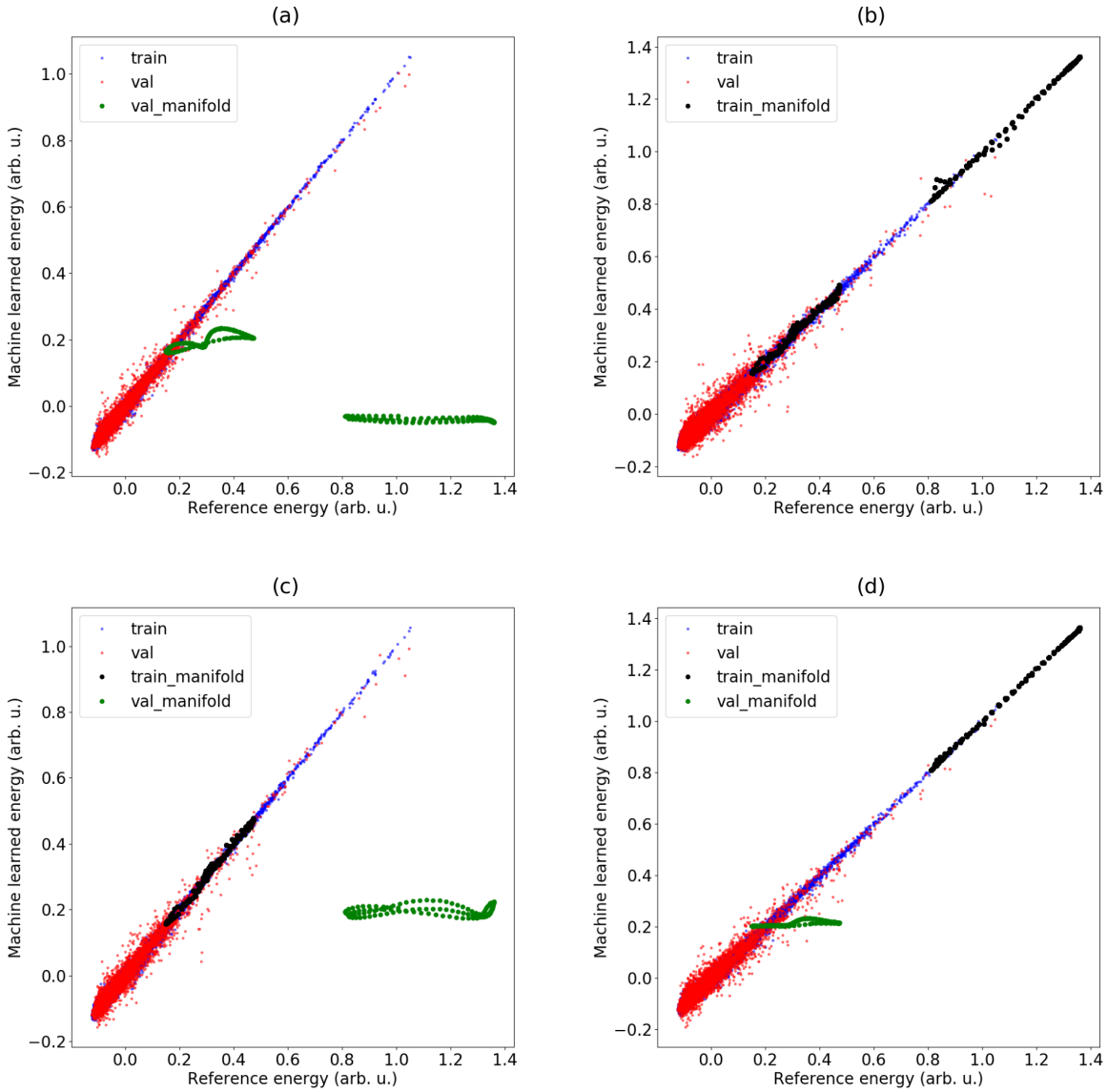

FIG. 4: Machine learning results using neural networks for the mild manifold of the comment (manifold 1 with reference energies of 0.2 to 0.4) in presence of another mild manifold (manifold 2 with reference energies of 0.8 to 1.4), both generated with the parameters of the comment. The red and blue points show the results for the same ordinary training and validation sets as in Fig. 2. In panel a) no structures from either manifolds were added to the training set and the manifolds can not be described. In panel b) structures from both manifolds were included in the training set. They were machine learned according to the deficiency-adapted procedure by adding 24'000 manifold structures (120 evenly distributed structures repeated 200 times) to the training set. In panel c) only structures from manifold 1 were included in the training. Manifold 1 can now be over-fitted but there is no improvement for manifold 2. In panel d) the opposite was done. Only the structures of manifold 2 were included in the training. In this case manifold 1 is not learned. So even though a single mild manifold can be learned with the deficiency-adapted procedure, the description of other manifolds is not improved.