Reply to SciPost referee reports in round 1 for "Learning crystal field using convolutional neural networks"

Reply to Report 1

> While not revolutionary, this is a simple yet useful example of how the machine learning approach can reduce a somewhat tedious task often performed manually in experimental labs. The paper is very clearly written, and is accompanied by an open source repository where the corresponding code can be directly used to compute Steven parameters from thermodynamic measurements. This clearly will be useful for future experiments.

> This paper can be accepted as it is, and I have only basic questions / suggestions.

We thank the referee for recommending our paper for publication. We address the referee's suggestions below and in the modified manuscript.

> To improve quality of the results for the hexagonal and tetragonal point group, it is suggested to add other input data such as a second magnetisation curve (obtained from a field aligned in a different direction). I guess this is quite easy to add in the training data and approach, and I would suggest to try it to see if it indeed improves the results. Same question for a larger temperature range.

We thank the referee for pointing this out. We have actually included magnetization and susceptibility data along two inequivalent crystalline directions ($[001]$ and $[100]$) in the training data sets for tetragonal and hexagonal systems (see Secs.4B and 4C), and in the newly added experimental example, $CeAgSb_2$ (see Sec.5A). Providing magnetization along different directions indeed results in more accurate CNN predictions of the CF parameters. In fact, to successfully predict the CF coefficients that describe the asymmetry between different crystal directions (e.g. ab versus c directions in a tetragonal system) requires such information. We have emphasized that point in the modified manuscript. Also, we chose to remove the entries in Table 2 and 3 that are not properly learned by the CNN (these entries were previously close to zero, which is the network's best prediction of a parameter that cannot be properly learned).

We have not systematically tested incorporating larger temperature and magnetic field ranges, mainly because we wanted to keep it the study as experimentally realistic as possible (i.e. we wanted to use data that can be easily acquired in the lab without going to special facilities, such as high-field laboratories). This can certainly be explored in future work.

We note that the magnetic field range for the newly added example $CeAsSb_2$ is going up to 50 T, and we find good agreement of the CNN prediction with the experimentally observed moment saturation values. Especially going to larger magnetic fields, where the higher energy levels mix into the B=0 ground state manifold provides useful information about the CF parameters.

> When comparing to experimental data, it would be interesting to see how good/bad is this approach when one of the experimental input is missing (as could happen in a lab, if e.g. specific heat measurements are not available).

It is generally true that more data is always better for the performance of the machine learning algorithm. Specific heat data is useful to constrain the splitting between the lowest two crystal field states. We have tested (for the data set for $CeAgSb_2$ explicitly) that the CNN can also converge to solutions with small MSE if we do not provide c_M data. However, generally constraining the results by more data is better as it reduces the number of solutions to the inverse problem. Most importantly, the data needs to contain sufficient information (e.g. about the anisotropy of the system or contributions from higher excited levels) to discriminate different CF parameters. Most notably, the CNN is unable to learn particular CF parameters (x_3 for hexagonal and $\{x_2, x_4\}$ for tetragonal) if the data set does not include information about magnetic anisotropy between ab and c directions (by providing both χ_c, M_c and χ_a, M_a). We emphasize that this is not a shortcoming of the machine-learning algorithm but just a property of the inverse problem. In the updated manuscript, we have emphasized this point in the captions of Tables 2 and 3.