GMLQC
Summer Semester 2024

Exercise Sheet 9

Due: 25.06.24 14h00

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General Regulations.

- Please hand in your solutions in groups of up to two people.
- Your solutions to theoretical exercises can be either handwritten notes (scanned), or typeset using LaTeX. In case you hand in handwritten notes, please make sure that they are legible and not too blurred or low resolution.
- For the practical exercises, always provide the (commented) code as well as the output, and don't forget to explain/interpret the latter. Please hand in an exported PDF of your notebook.
- Submit all your files in the Übungsgruppenverwaltung, only once for your group.

1 Paper reading

- KineticNet [1]: Please read II, III, IV and skim over the rest of the paper.
- M-OFDFT [3]: Please read 4.1 and 4.2.

Please briefly explain the differences in representations of the features inside the network in the case of KineticNet vs. M-OFDFT.

(4 pts)

2 Graph Neural Networks

In this exercise, you'll implement a simple Graph Convolutional Layer and train your model using real chemistry data. The aim is to try to predict the electronic ground state energy of diverse molecules using only the atom types and their "ball-and-stick" connectivity as input. You will use the `QM9` dataset, which consists of molecular structures with up to 9 heavy atoms, with labels calculated at DFT-level accuracy.

(a) Use the `pytorch-geometric` library to load the `QM9` dataset. Create a `DataLoader` object for training, validation, and testing. Use 2000 samples for training and 500 samples each for validation and testing. (3 pts)

(b) Implement the following graph convolutional layer (GCNConv) as a module. The features x_i on node i should update according to

$$x'_i = \sum_{j \in \mathcal{N}(i) \cup \{i\}} \frac{1}{\sqrt{d_i d_j}} A(x_j).$$

Here $\mathcal{N}(i)$ is the neighborhood of the node i, defined by the `edge_index` parameter inside the data samples. A is a linear layer and $d_i = |\mathcal{N}(i)| + 1$ is the number of neighbors of node i plus one for the self loop. You should be able to specify input and output size.

Hint: The `pytorch-geometric` MessagePassing class makes it easy to implement such modules.

(3 pts)

(c) Implement the entire network as described below:

```
1. GCNConv(input_size, hidden_channels)
2. ReLU
3. GCNConv(hidden_channels, hidden_channels)
4. ReLU
5. GlobalAddPool
6. Linear(hidden_channels, 1)
```

(3 pts)

(d) Write the training loop, including validation at each epoch and testing at the end. Use `MSELoss` as the loss function, mean absolute error for validation error, and Adam as the optimizer.

(3 pts)

(e) Train your model on the internal energy of the molecule at $0K$, with a learning rate of 0.0005 and `hidden_channels`$= 512$. You can adjust the number of epochs and batch size based on your available computational resources. Save the loss and validation error during training.

(2 pts)

(f) Plot loss and validation error. (2 pts)

(g) Now go wild! Within the realm of your computational resources, feel free to modify the input representation, architecture, hyperparameters, etc. and report the best validation error that you can achieve. The published state of the art for this task is around 3.9meV by Simeon et al. [2].

(**Bonus:** 5 pts)

References

[1] Roman Remme et al. "KineticNet: Deep learning a transferable kinetic energy functional for orbital-free density functional theory". In: *The Journal of Chemical Physics* 159.14 (Oct. 2023). ISSN: 0021-9606, 1089-7690. URL: https://pubs.aip.org/jcp/article/159/14/144113/2916356/KineticNet-Deep-learning-a-transferable-kinetic.

[2] Guillem Simeon and Gianni De Fabritiis. "Tensornet: Cartesian tensor representations for efficient learning of molecular potentials". In: *Advances in Neural Information Processing Systems* 36 (2024).

[3] He Zhang et al. "Overcoming the barrier of orbital-free density functional theory for molecular systems using deep learning". In: *Nature Computational Science* 4.3 (Mar. 2024), pp. 210–223. ISSN: 2662-8457. DOI: 10.1038/s43588-024-00605-8. URL: https://uebungen.physik.uni-heidelberg.de/c/image/d/vorlesung/20241/1883/material/zhang_24_overcoming.pdf.