Spectral learning for solving molecular Schrödinger equations

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Recently, there has been a significant research interest in using neural networks for solving partial differential equations (PDEs) in general \cite{E2018}, and Schrödinger equations in particular \cite{Hermann2020}. The use of neural networks was shown to mitigate, or even break the curse of dimensionality \cite{Grohs2018} encountered in standard numerical methods, such as finite-volume or spectral methods. However, standard neural networks for solving PDEs seem to be fragile \cite{E2020}, since they require a lot of engineering and show high sensitivity to hyperparameters. In the context of quantum mechanics, neural networks were shown to accurately approximate ground-states, i.e., eigenfunctions corresponding to smallest eigenvalues, of molecular systems, while scaling moderately with the dimension of the problem \cite{Hermann2020}. However, extensions to computing many excited states, i.e., eigenfunctions corresponding to larger eigenvalues, suffer from convergence issues and remain challenging \cite{Cuzzocrea2020}.

In this talk I introduce a neural-network based paradigm, where complex and rich families in $L^2$ are produced by pushing forward standard basis sets through non-singular measurable mappings. I show that a bijectivity assumption on the mapping is a necessary and sufficient conditions for the resulting families to be dense in $L^2$ \cite{Saleh2023}. This allows us to model these mappings using normalizing flows, an important tool from generative probabilistic modeling. I present a nonlinear variational framework to approximate molecular wavefunctions in the linear span of these flow-induced families. The framework allowed to compute many eigenstates of various molecular systems with orders-of-magnitude improved accuracy over standard linear methods \cite{Saleh2023}.

REFERENCES

\begin{itemize}
  \item \cite{E2020} E, W., Ma, C., Wojtowytsch, S., and Wu, L. (2020). Towards a mathematical understanding of neural network-based machine learning: what we know and what we don’t.
\end{itemize}