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 [2], and Schrödinger equations in particular [6]. The use of neural networks was shown to mitigate, or even break the curse of dimensionality [4] encountered in standard numerical methods, such as finite-volume or spectral methods. However, standard neural networks for solving PDEs seem to be fragile [3], 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 [5]. However, extensions to computing many excited states, i.e., eigenfunctions corresponding to larger eigenvalues, suffer from convergence issues and remain challenging [1].

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 [8]. 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 [7].

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