PYSEQM2.0: Machine Learnable GPU-based Semiempirical Mechanics for Ground and Excited States

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Recent strides in atomistic machine learning models have emerged into new toolkit and new data-centered research strategies in computational chemistry. However, surrogate approaches often face challenges when applied to unexplored regions of chemical space. Interatomic potential models, for instance, lack crucial electronic structure information, often leading to limited transferability. To address these limitations, our research group has been dedicated to developing PYSEQM (Pytorch-based Semiempirical Quantum Mechanics), a differentiable physics model that combines domain knowledge of semiempirical quantum mechanics with machine learning as a corrective tool. Paired with atomistic neural network backend, PYSEQM allows backpropagation through Hamiltonian, replacing atom-type dependent constants with structure-aware parameters generated on-the-fly.

This presentation will span the future release of PYSEQM2.0 which expands capabilities to excited states through Davidson eigensolver in Krylov subspace and dynamics beyond the ground state. To the best of our knowledge, this is the first implementation of batched mode introducing GPU data-parallelism. New developments, including Davidson algorithm, extend simulations to large organic molecules relevant to photovoltaics, organic solar cells and photoinduced processes.