Letter

Algorithmic Differentiation for Automated Modeling of Machine Learned Force Fields

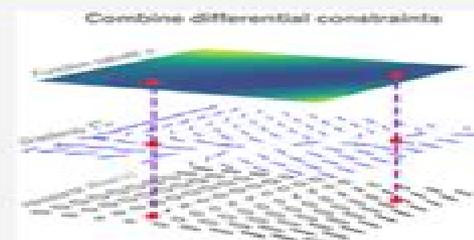Niklas Frederik Schmitz, Klaus-Robert Müller,* and Stefan Chmiela*

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ABSTRACT: Reconstructing force fields (FFs) from atomistic simulation data is a challenge since accurate data can be highly expensive. Here, machine learning (ML) models can help to be data economic as they can be successfully constrained using the underlying symmetry and conservation laws of physics. However, so far, every descriptor newly proposed for an ML model has required a cumbersome and mathematically tedious remodeling. We therefore propose using modern techniques from algorithmic differentiation within the ML modeling process, effectively enabling the usage of novel descriptors or models fully automatically at an order of magnitude higher computational efficiency. This paradigmatic approach enables not only a versatile usage of novel representations and the efficient computation of larger systems—all of high value to the FF community—but also the simple inclusion of further physical knowledge, such as higher-order information (e.g. Hessians, more complex partial differential equations constraints etc.), even beyond the presented FF domain.

Combine differential constraints

$\mathbf{M}$ost physical quantities are represented by differential equations (DEs)

$$\mathcal{L}u(\mathbf{x}) = \mathbf{f}(\mathbf{x}) \tag{1}$$

where $\mathcal{L} = \sum_{0 \leq i} a_i \mathcal{D}^i$ is a finite linear combination of differential operators $\mathcal{D}^i$ of order n.

Examples of eq 1 are found in electrodynamics, fluid dynamics, quantum mechanics, etc., always imposing strong constraints on the form of a physical solution $\mathbf{f}(\mathbf{x})$. For several years, machine learning (ML) has started to broadly contribute to physical modeling across its disciplines, for instance in particle physics,[1] atomistic simulations and force fields (e.g., see refs 2−6), fluid dynamics (e.g., see refs 7 and 8) or lattice gauge theories.[9−12] Note, however, that the standard procedure of ML models has been so far to learn from empirical data only. Recently, ML modeling has also started to incorporate physical constraints, be it as regularization terms (e.g., see refs 13 and 14) or by explicitly taking into account conservation laws, DEs such as eq 1, symmetries (e.g., see refs 3, 15, and 16), various operator response properties[17,18] or empirical correction terms, e.g. for long-ranged electrostatics.[19−23] Notably, the direct usage of DEs allows the method to greatly simplify modeling and to be highly data efficient since the known laws of physics do not need to be learned from empirical data anymore (e.g., see refs 3, 15, and 16).

Let us study the popular example of eq 1 force fields (FFs), namely, the negative gradient $\mathcal{L} = -\nabla_{\mathbf{x}}$, where $u(\mathbf{x})$ is a scalar energy potential mapped to a vector field of forces $\mathbf{f}(\mathbf{x})$. The unknown force field (FF) is sampled at several locations $\{\mathbf{x}_i\}_{i=1}^M$, and $\mathbf{f}(\mathbf{x}_i)$ are obtained from ab initio atomistic reference data.[5,15,23] The constraint

$$\mathcal{L} = -\nabla_{\mathbf{x}} \tag{2}$$

which is equivalent to both energy conservation and curl-free vector fields, enables the estimation of a faithful representation of true energy-conserving FFs as opposed to the estimation of general (unphysical) vector fields.[5,24,25]

This approach unfolds its usefulness well beyond the FF domain for higher-order differential operators and compositions thereof,[26−30] which characterize complex properties across all physics domains, e.g. Hessians or Laplacian operators.[31,32] Prior studies[13−15,34−46] have demonstrated the effectiveness of explicit DE constraints, but their adoption still remains low due to an inefficiency that hampered model development and training so far: (1) Differential operator transforms inflate model complexity, making their full algebraic derivation mathematically tedious and error-prone. (2) Training and evaluation of a model using DE constraints is often associated with significant time and memory costs. While there exist prior works with focus on efficiency improvement, they rely on manual derivations and are limited to first-order gradients and primitive kernels without physical descrip-

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