Multiscale simulation of high harmonic generation using artificial intelligence

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High harmonic generation (HHG) is a laser-matter process that allows to up-convert coherent infrared light into the extreme-ultraviolet or even soft x-rays upon highly nonlinear interaction in a gas or solid target (Fig. 1). The resulting radiation can be synthesized into laser pulses as short as tens of attoseconds, the shortest light pulses ever created, enabling unprecedented studies of ultrafast physics at the nanoscale.

A realistic simulation of HHG requires multiscale calculations from both the quantum microscopic and the macroscopic points of view. At the microscopic level (Fig. 2), the exact calculation of HHG is given by the solution of the time-dependent Schrödinger equation (TDSE), that describes the quantum laser-driven wavepacket dynamics in the vicinity of each atom. However, from the macroscopic point of view, this process has to be considered in all of the atoms involved in the experiment—trillions—and the already time-consuming TDSE has to be coupled with the Maxwell equations to account for propagation. Such calculation is extremely expensive computationally, well beyond the state-of-the-art capabilities, and approximations are required.

In this work we introduce the use of artificial intelligence (AI), and specifically Neural Networks (NN), to obtain complete TDSE-based macroscopic HHG calculations driven by structured laser beams in low density gas jets (Fig. 3). We have implemented a NN using Keras and TensorFlow in Python to predict the microscopic single-atom HHG response, and we have trained it with a dataset of 8×10^4 exact TDSE calculations, achieving a mean square error (MSE) of $\leq 10^{-5}$ (Fig. 4) . The dataset was generated with random amplitudes and phases of the driving infrared pulse, covering the whole input range of our simulations, by running a highly parallelized implementation of the TDSE in CUDA.

Once the NN is trained and validated, we integrate the predicted dipole acceleration into the macroscopic HHG calculation through the exact solution of the Maxwell equations [1]. The macroscopic calculation is parallelized with OpenMP and MPI to allow the prediction of the microscopic HHG in multiple atoms at once. Each OpenMP thread propagates the calculated dipole emission from each atom towards a far-field detector (Fig. 5). We have validated our AI-based HHG method with simulations that compute HHG from structured drivers (Fig. 6), an emerging field with many applications at the nanoscale [2].

Our results [3] demonstrate that AI applied to HHG provides a two-fold advancement: (i) it speeds up the calculations, providing a route towards *in-situ* strong-field simulations that can be performed in parallel to the experiments; and (ii) it allows to perform TDSE-based macroscopic simulations that can reveal hidden signatures neglected in the standard approximations, thus allowing for the exploration of new physics at the nanometer and attosecond scales.

Acknowledgment

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References

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[2] A. Forbes et al. Nature Photon. **15**, 253–262 (2021).
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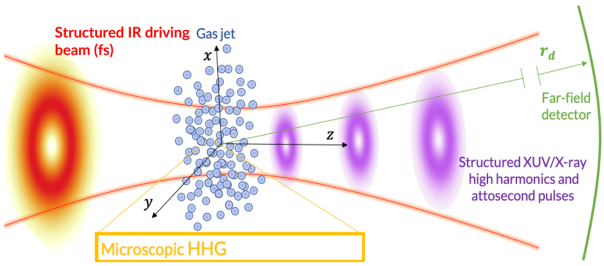

Fig. 1. Scheme of HHG from the macroscopic level. An ultrafast and ultraintense structured IR driving beam is focused in a gas jet. As a result of the highly nonlinear interaction, structured x-ray high-order harmonics, emitted in the attosecond timescale are emitted. The overall emission results from the coherent addition of the dipole acceleration emitted from each atom in the gas target.

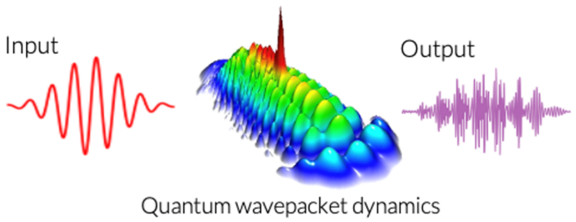

Fig. 2. Scheme of HHG at the microscopic level. An electronic wave packet is ionized by the intense infrared laser pulse. The electronic wave packet is then accelerated, and sent back to the original ionized atom or molecule, driven by the oscillating nature of the laser field. The energy gained by the electronic wave packet during the journey is emitted as high frequency harmonics at recollision. The exact calculation of such process is given by the TDSE.

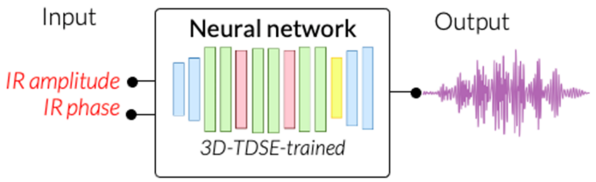

Fig. 3. We introduce the use of neural networks to predict HHG at the microscopic level. The NN is trained to predict the dipole acceleration taking as inputs the amplitude and phase of the driving laser pulse.

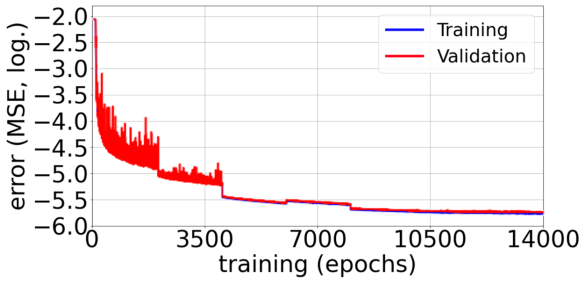

Fig. 4. Neural network training with 14000 epochs, increasing the batch size from 2^3 to the next power of 2 every 2000 epochs up to 2^{10}. The reached MSE of $\leq 10^{-5}$ allows us to get TDSE-based microscopic results in deciseconds instead of minutes-hours using inexpensive hardware

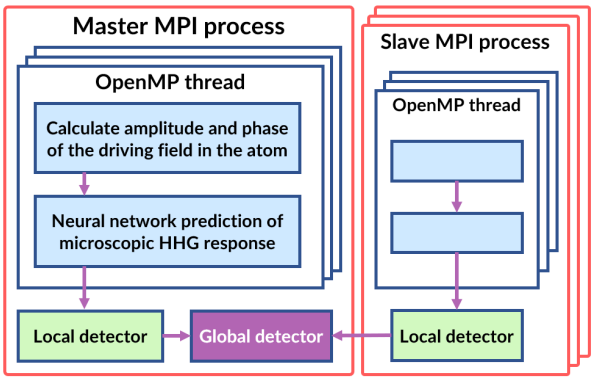

Fig. 5. Highly parallelized simulation platform implemented in C++ that takes advantage of our neural network, MPI and OpenMP to simulate macroscopic HHG. Local detectors with optimized synchronization code reduce synchronization overhead among OpenMP threads and decouples MPI processes until they are reduced in the master at the end of the simulation.

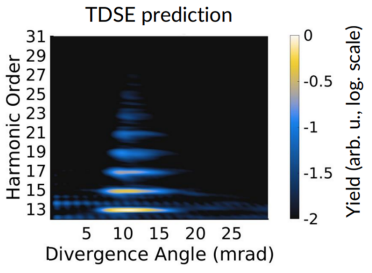

Fig. 6. Sample of macroscopic HHG results obtained with our TDSE-trained NN in combination with our simulation platform. The plot shows the intensity of the far-field high-order harmonics generated in atomic hydrogen as a function of the divergence. The driving laser beam is a combination of two infrared, intense vortex beams with different orbital angular momentum contributions.