Deep Learning for Retrieval of the Internuclear Distance in a Molecule with Fixed or Moving Nuclei from Photoelectron Momentum Distributions

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By using a convolutional neural network (CNN), we retrieve the internuclear distance in the two-dimensional (2D) H_2^+ molecule interacting with a strong laser pulse from the photoelectron momentum distributions [1]. We apply the transfer-learning technique to make the developed CNN applicable to distributions that correspond to parameter ranges the network was not explicitly trained for [2]. We study the effect of the carrier-envelope phase on the performance of the CNN and we also analyze the reconstruction of the internuclear distance based on the one-dimensional projected momentum distributions. The CNN is compared with alternative methods for the retrieval of the internuclear distance: the direct comparison of electron momentum distributions, decision trees, and support vector machines. The alternative approaches show very limited transferability. We use the occlusion-sensitivity technique to understand the features of the distributions that are used by a neural network to predict the internuclear distance [2].

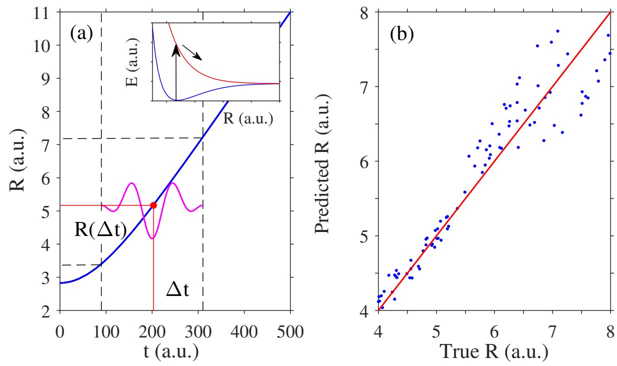

Figure 1: Retrieval of the time-dependent internuclear distance in the dissociating 2D H_2^+ molecule. (a) The internuclear distance as a function of time (blue curve) after the excitation by the pump pulse (see inset). The probe pulse arriving after a certain time delay Δt (magenta curve) ionizes the molecule at certain internuclear distance $R(\Delta t)$. (b) Plot of predicted *vs* true internuclear distances illustrating the performance of the CNN

Furthermore, we investigate the possibility of reconstruction of the time-dependent internuclear distance in a pump-probe scheme. A high-frequency pump pulse of moderate intensity initiates nuclear motion by excitation of the molecule to a higher electronic state. We assume that the atomic nuclei move classically along the Born-Oppenheimer potential surfaces. The molecule is ionized by an intense, short probe pulse acting after a certain time delay, which determines the internuclear distance at the time of ionization, see figure 1(a). For ionization by a two-cycle laser pulse, the CNN trained on momentum distributions for fixed internuclear distances predicts the internuclear distance with a mean absolute error of about 0.26 a.u., see figure 1(b). The application of a single-cycle probe pulse provides a mean absolute error of 0.21 a.u. These errors can be further decreased by application of the transfer-learning technique. Therefore, the CNN trained on distributions for fixed internuclear distances can be applied for retrieval of the time-dependent bond length.

References

[1] N I Shvetsov-Shilovski and M Lein, Phys. Rev. A **105**, L021102 (2022)

[2] N I Shvetsov-Shilovski and M Lein, Phys. Rev. A **107**, 033106 (2023)