



Unified theoretical description for thermal and nonthermal laser induced ultrafast structural modifications in materials

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Ab-initio approaches to solids irradiated by ultrashort laser pulses usually rely on the description of the excited hot electrons within the canonical ensemble and in terms of the Helmholtz/Mermin free energy, which also determines the potential energy surface (PES) for the ionic motion. On the other hand, the motion of the ions on that laser excited PES fulfills energy conservation if the electronic temperature T_e does not change and is therefore treated as a closed system by molecular dynamics (MD) simulations. Now, when the electron-phonon coupling is active, incoherent energy exchange between electrons and ions takes place, which leads to heating of the lattice. In this context, the mixed canonical-microcanonical scheme leads to incorrect results and cannot be used. In this talk, we present a unified theoretical description that effectively integrates the processes of ultrafast laser induced bond changes and electron-phonon coupling [1]. Our theory generalizes the two-temperature-model-molecular-dynamics (TTM-MD) method in a natural way and can be used either in ab-initio simulations or in large-scale MD simulations based on T_e -dependent interatomic potentials. Moreover, we will describe the development of accurate Machine-Learning interatomic potentials, trained and validated on data sets generated using ab-initio MD simulations [2,4]. In a first approach, a general interatomic potential is developed into polynomials[2,3], and their coefficients are trained using the data set. Our simulations closely match experimental observations, accurately reproducing the temporal evolution of the Bragg peaks in laser excited silicon films. The second approach is an extension of the concept of high dimensional neural network potentials (HDNNP) developed by Behler and Parrinello[5], to the laser excited state. We constructed, trained and validated a HDNNP for laser excited silicon and obtained good agreement with DFT simulations. We implemented the potential in our generalized TTM-MD scheme and described laser processing on a silicon surface[4]. Application of our approaches to the laser manipulation of diamond containing NV-centers (which can act as Qubits) is in progress[6]. Our method can be used for performing atomistic simulations of laser processing and functionalization of materials on experimentally relevant length and time scales.

References:

- [1] B. Bauerhenne and M. E. Garcia, Sci Rep 14, 32168 (2024).
- [2] B. Bauerhenne, V. P. Lipp, T. Zier, E. S Zijlstra and M. E. Garcia, Phys. Rev. Lett. 124, 085501 (2020).
- [3] TPH Sidiropoulos et al., Phys. Rev. X 11, 041060 (2021).
- [4] P. Plettenberg, B. Bauerhenne and M. E. Garcia, Commun Mater 4, 63 (2023).
- [5] J. Behler and M. Parrinello, Phys. Rev. Lett. 98, 146401 (2007).
- [6] M. Kempkes, T. Zier, K. Singer and M. E. Garcia, Carbon 174, 524 (2021).