Ab-initio and classical molecular dynamics simulations of ultrafast structural phenomena in laser excited solids

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Intense femtosecond-laser pulses are able to induce ultrafast nonthermal pase transitions in different materials along pathways that are inaccessible under thermodynamic conditions. In order to investigate the motion of atoms in the nonthermal transient state produced by femtosecond laser excitation we performed *ab-initio* molecular-dynamics simulations on laser-excited potential energy surfaces using our code CHIVES (Code for Highly excited Valence Electron Systems). We found surprising results, like excitation of squeezed thermal phonons, which constitute the precursor of nonthermal melting as a function of fluence[1], the presence of atomic fractional diffusion, as a transient state preceding the formation of a nonthermal liquid[2], and laser-fluence dependent anisotropic energy redistribution[3]. Also, experimentally measurable signatures of nonthermal melting were identified[4], and the possibility of controlling nonthermal melting by pulse shaping was considered. In this talk I will give an overview on the results of our ab-initio simulations. In order to extend our method for the study of nucleation phenomena, we developed an analytical interatomic potential for laser-excited silicon, which depends on the electronic temperature. Effects like bond softening in the presence of hot electrons are taken into account. With the help of this potential we were able to perform large-scale simulations and study nucleation dynamics during nonthermal melting.

In addition, we applied a hybrid atomistic-continuum model capable of capturing the essential mechanisms responsible for the laser-induced nanostructuring of gold surfaces upon excitation with spatially modulated ultrashort UV pulses. Our TTM-MD approach allowed us to perform calculations on experimental spatial and time scales and to a one-to-one comparison with experiments. Theoretical results are in excellent agreement with experiment. Our simulations show the presence of voids beneath the surface as a consequence of the laser excitation, explaining the formation of nanobumps and their sizes depending on the laser fluence[5]. I will also address these results in the present talk.

[1] E. S. Zijlstra, A. Kalitsov, T. Zier and M. E. Garcia, Squeezed Thermal Phonons Precurse Nonthermal Melting of Silicon as a Function of Fluence, Physical Review **X 3**, 011005 (2013).

[2] E. S. Zijlstra, A. Kalitsov, T. Zier and M. E. Garcia, Fractional Diffusion in Silicon, Advanced Materials **25**, 5605-5608 (2013).

[3] T. Zier , E. S. Zijlstra and M. E. Garcia, Quasimomentum-Space Image for Ultrafast Melting of Silicon, Phys. Rev. Lett. **116**, 153901 (2016)

[4] T. Zier , E. S. Zijlstra, A. Kalitsov, I. Theodonis and M. E. Garcia, Signatures of nonthermal melting, Structural Dynamics **2** , 054101 (2015).

[5] D. S. Ivanov, V. P. Lipp, A. Blumenstein, F. Kleinwort, V. P. Veiko, E. Yakovlev, V. Roddatis, M. E. Garcia, B. Rethfeld, J. Ihlemann, and P. Simon, Experimental and Theoretical Investigation of Periodic Nanostructuring of Au with Ultrashort UV Laser Pulses near the Damage Threshold, Phys. Rev. Applied **4**, 064006 (2015).