Squeezed phonons, fractional diffusion, structural phase transitions and control of nonthermal atomic motion in laser excited solids

M. E. Garcia

Institute of Physics, Universität Kassel, Germany

Intense femtosecond-laser pulses are able to induce ultrafast nonthermal melting of different materials along pathways that are inaccessible under thermodynamic conditions. However, little is known about such transient states. In order to investigate the nonthermal atomic motion we performed *ab-initio* molecular-dynamics simulations on laser-excited potential energy surfaces using our in-house code CHIVES (Code for Highly excIted Valence Electron Systems). We found surprising and fascinating phenomena. For instance, our simulations show that for silicon irradiated below the melting threshold room-temperature phonons become thermally squeezed, and that in such state the atoms move in the same directions as during the first stages of nonthermal melting. Our results demonstrate that the presence of squeezed thermal phonons announces complete atomic disordering as a function of fluence, and that this effect is not material specific [1].
Furthermore, by studying irradiated silicon for fluences above the melting threshold, we found that the atoms move successively superdiffusively and fractionally diffusively before becoming diffusive. We explain these transient behaviors by a laser-induced acceleration of atoms into structural voids and a subsequent deceleration, which we link to the time-dependent interatomic scattering rates. At a relatively low excitation density we find fractional atomic diffusion, not reported so far in materials, during more than 800 fs [2]. In this talk I will show, that combining the effects mentioned above, control of the nonthermal melting process by two pump pulses is possible. As further results, new mechanisms for the generation of coherent phonons in TiO_2 [3] and BN nanotubes [4] will be also presented.

In addition, I will show that both ultrashort laser pulses and XUV radiation can induce bond hardening in silver and copper, and that this effect can be used to achieve vacuum squeezing of particular phonon modes. For very intense XUV-pulses we were able to analyze the first steps in the formation of warm dense matter starting from a solid [5].

Finally, it will be shown that the combination of accurate time-resolved optical measurements with *ab-initio* calculation of atomic displacements from the knowledge of the reflectivity has clear advantages with respect to time-resolved crystallography and allows a detailed visualization and control of two-dimensional atomic motion in solids [6].

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) E. M. Bothschafter, A. Paarmann, E. S. Zijlstra, N. Karpowicz, M. E. Garcia, R. Kienberger and R. Ernstorfer, Physical Review Letters **110**, 067402 (2013).
4) B. Bauerhenne, E. S Zijlstra, A. Kalitsov and M. E Garcia, Nanotechnology **25**, 145701 (2014).
5) F. Cheenicode Kabeer, E. S. Zijlstra and M. E. Garcia, Road of warm dense noble metals to the plasma state: Ab initio theory of the ultrafast structural dynamics in warm dense matter, Physical Review **B 89**, 100301(R) (2014).
6) H. Katsuki, J.C. Delagnes, K. Hosaka, K. Ishioka, H. Chiba, E.S. Zijlstra, M.E. Garcia, H. Takahashi, K. Watanabe, M. Kitajima, Y. Matsumoto, K.G. Nakamura and K. Ohmori, All-optical control and visualization of ultrafast two-dimensional atomic motions in a single crystal of bismuth, Nature Communications **4**, 2801 (2013).