FROM ELECTRICAL CURRENT VIA NON-EQUILIBRIUM PHONONS TO FRENKEL DEFECTS

Dietrich E. Wolf, Dept. Physics, University of Duisburg-Essen, Germany
dietrich.wolf@uni-due.de

Key Words: Flash sintering, Frenkel pairs, Boltzmann equation, electron-phonon interaction, non-equilibrium molecular dynamics.

Early flash sintering experiments suggested that athermal amounts of interstitials and vacancies (Frenkel pairs) emerge in the bulk. Indeed, this would offer an explanation of the flash phenomenon. In a series of papers [1-3] we developed a theoretical picture, how this may happen. By solving coupled Boltzmann equations for electrons and phonons, describing an electrical current with momentum transfer to the crystal lattice [3], we found a strong proliferation of short wavelength lattice vibrations. The deviation from a thermal Bose-Einstein distribution is surprisingly strong, regardless of the details of the electron-phonon matrix elements. The electrical current drives the lattice vibrations out of equilibrium. With non-equilibrium molecular dynamics simulations [1], where a phonon mode is permanently excited with a certain rate, we demonstrated that Frenkel defects are generated in molar concentrations far above equilibrium, provided three conditions are fulfilled: The phonon mode must be near the Brillouin zone edge, the excitation rate must be within a window in the TeraHertz range, and the temperature must be above the Debye temperature, but sufficiently below the melting point. We postulate that these conditions can be achieved by a suitable electrical current. The proliferation of lattice vibrations near the Brillouin zone edge may be the reason for the generation of athermal Frenkel defects.The non-equilibrium molecular dynamics simulations were done for aluminum [1] and for rutile TiO_2 [2]. In the latter case, interstitial-vacancy pairs of Ti as well as of O are formed. In agreement with experiment we found that the mean-square displacements of the vibration amplitudes of the Ti and O atoms are specifically enhanced.

[1] Malte Jongmanns, Rishi Raj, Dietrich E. Wolf: Generation of Frenkel defects above the Debye temperature by proliferation of phonons near the Brillouin zone edge,
New J. Phys. 20, 093013 (2018).

[2] Malte Jongmanns, Dietrich E. Wolf: Element-specific displacements in defect-enriched TiO_2 : Indication of a flash sintering mechanism,
JACS 103, 589 (2020).

[3] Magdulin Dwedari, Lothar Brendel, Dietrich E. Wolf: Non-equilibrium phonon distribution caused by an electrical current, preprint (2022).