Simulation of a Femtosecond Laser Excitation in Antimony

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Project Term
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Clusters
Lichtenberg Cluster Darmstadt

Additional Software
CHIVES

University
University Kassel

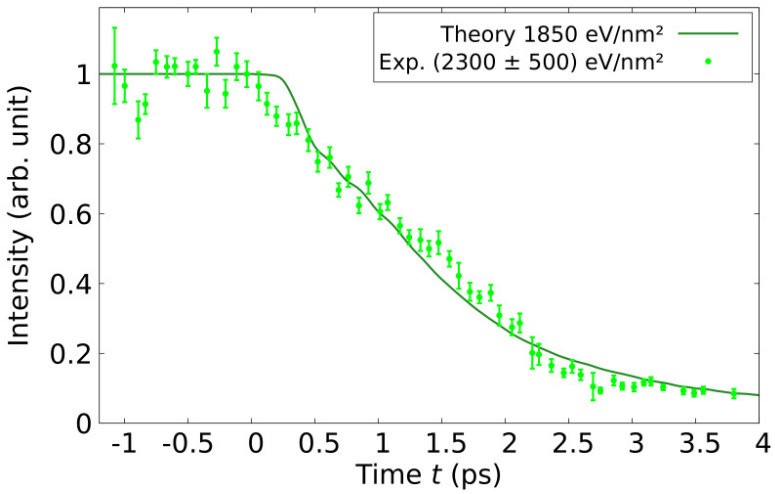

Figure 1: Time dependent intensity of the (200) Bragg peak measured from ultrafast x-ray diffraction and obtained from the simulations are shown. The incident fluence of the pump laser pulse was adapted in the simulations within the error bar of the experiment.

Introduction

If a femtosecond laser pulses excites matter, the electrons are excited to a high temperature whereas the ions remain cold. This state far away from thermodynamic equilibrium with hot electrons and cold ions exists for several picoseconds. During this state, strong forces act on the ions and many ultrafast phenomena occur. The measurement of the time-dependent intensities of several Bragg peaks provide insights into the atomic motions following a femtosecond laser excitation. But the full molecular movie can only be done by theoretical simulations, which are computational expensive and still under research. Within this project, we performed molecular dynamics simulations of a femtosecond laser pulse excitation in antimony from theoretical considerations and compared directly with experimental results.

Methods

We performed Molecular dynamics (MD) simulations using temperature dependent density functional theory using a small cell. We created a electronic temperature dependent interatomic potential from these simulations. We derived the optical properties of antimony from density functional theory. We performed large scale MD simulations using the new interatomic potential and analyzed the atomic coordinates.

Results

In the first year of the project, we performed molecular dynamics (MD) simulations of a thin antimony film using 384 atoms at various electronic temperatures Te using Te-dependent density functional theory. From the atomic forces and the structural energies of these simulations we derived a Te-dependent interatomic potential for antimony. We analyzed the obtained potential and found out that it reproduces the ab-initio data very well.
In the second year, we performed large scale MD simulations using the interatomic potential of a 30 nm thick antimony film at various laser intensities. For this, we derived the absorption coefficient and the reflectivity of antimony from density functional theory. The crystal structure of antimony consists of hexagonal planes, that are stacked on top of each other with alternating distances. The difference between this distances is described by the so called Peierls parameter.
A femtosecond laser excitation causes an oscillation of the Peierls parameter or an oscillation of the planes against each other. These oscillations cause a significant change of the optical properties during the pump laser pulse. Thus, we derived the optical properties as a function of the Peierls parameter. In our MD simulations, we also took the effects of the electron–phonon coupling into account. This effects is caused by incoherent electron-ion collisions that decrease the electronic temperature and increase the ionic temperature until both are equal. We derived the intensity behavior of the (200) Bragg peak from the atomic coordinates of the MD simulations and compared directly with the experimental results (see Figure 1).

Discussion

We found an excellent agreement with the experimental results which validates our approach to simulate the effects of a femtosecond laser excitation on matter. Since there are also experimental results for bismuth, we will repeat the steps done for antimony also for bismuth, which is much more complicated to describe theoretically due to the relevance of the spin-orbit coupling in contrast to antimony.

Publications

Bauerhenne, B., Lipp, V.P., Zier, T., Zijlstra, E.S., Garcia, M.E.: ”Self-Learning Method for Construction of Analytical Interatomic Potentials to Describe Laser-Excited Materials”, Phys. Rev. Lett., vol. 124, p. 085501, 2020 https://doi.org/10.1103/PhysRevLett.124.085501

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