I. Institute of Physics
Physics of New Materials

RWTH AACHEN UNIVERSITY

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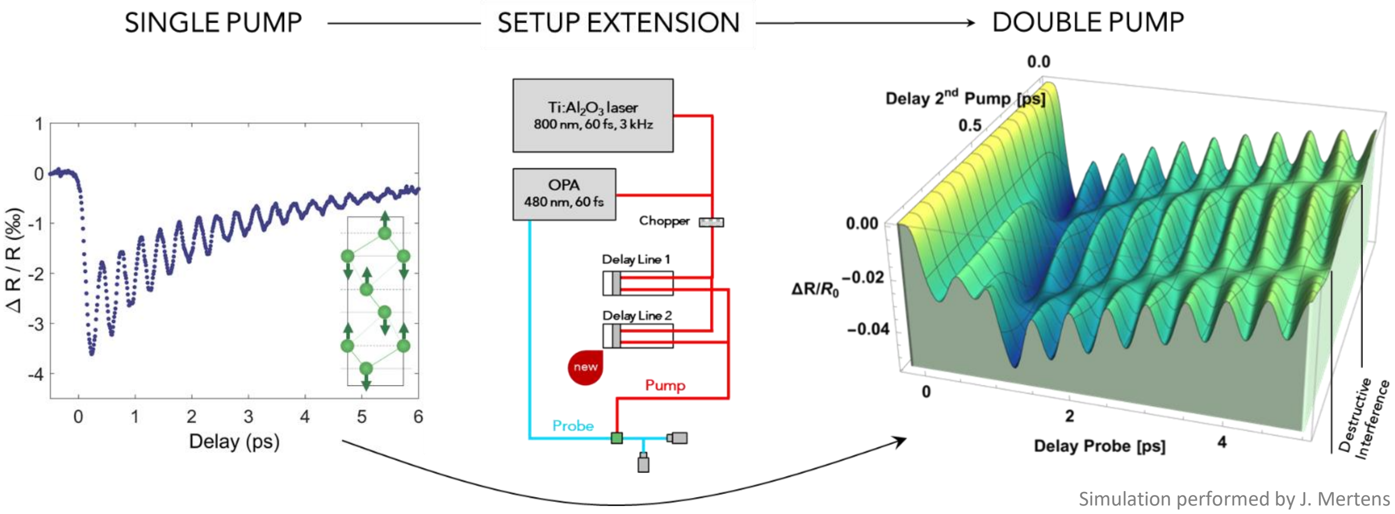
**Topic for a Bachelor or Master Thesis**

# Coherent Control of Optical Phonons by Degenerate Pump Probe Spectroscopy

Semimetals and Chalcogenides are known for their unique attributes as materials exhibiting Peierls distortion in their bulk form [1]. Their ability to undergo structural modifications easily via charge carrier dynamics renders them highly interesting for research, particularly in the context of optical pump-probe experiments. This project aims to enhance the existing pump-probe measurement scheme by introducing a second delay line into the pump beam path. A depiction of the measurement scheme is presented in the center of Figure **1**.

A single pump measurement of the transient reflectivity of a semimetal is displayed on the left side of Figure **1**. The pronounced oscillation pattern can be attributed to the $A_{1g}$ phonon mode of the material, illustrated in the inset featuring the material's unit cell. This phonon is initiated through a process known as displacive excitation of coherent phonons (DECP) [2], wherein a femtosecond laser pulse stimulates carriers in a manner that induces movement of the lattice to a new equilibrium position. The relaxation of this new equilibrium position is regulated by optical phonons over a timespan of several picoseconds.




Simulation performed by J. Mertens

Figure **1**: (**Left**) Transient isotropic reflectivity *ΔR/R* of a semimetal thin film. The measurement is performed at pump wavelength of 800 nm and a probe wavelength of 520 nm. The damped oscillation visible can be attributed to the excitation of a $A_{1g}$-symmetric coherent lattice vibrations. The oscillation motif of the unit cell is depicted as inset. (**Middle**) Extension of the pump probe setup with a second delay line. This extension leads to a degenerate pump-pump-probe measurement scheme with femtosecond time resolution. (**Right**) Simulations generated using the DECP model reveal an expansion in the measurement parameter space by one dimension upon the introduction of a second pump beam. Beyond configuring the pump-probe delay, an interpump delay can now be defined. As indicated by the two black lines on the right-hand side, modulation of the coherent phonon is feasible, exemplified in this instance by the occurrence of destructive interference.

Within this project, an additional delay line will be installed to extend the existing measurement scheme, shown in the middle of Figure **1**. This enhancement enables the adjustment of both the interpump delay and the pump-probe delay. Consequently, we gain the capability not only to measure the phonon dynamics, consisting amplitude, frequency, decay time, and phase, but also to actively manipulate and modulate these dynamics [3]. This is highlighted in the simulations presented on the figure's right side.

Various samples, including single crystals, MBE-textured thin films, sputter-deposited polycrystalline thin films, and amorphous chalcogenides, are subject to measurement. The lattice dynamics measurements are enhanced by Raman spectroscopy. The institute's numerous experimental setups contribute to an expanded understanding of the observed phenomena.

By combining materials science, MBE technology, and our advanced characterization methods, this project aims to shed light on the unique properties of semimetals and chalcogenides and their potential applications in emerging quantum technologies.

[1] Raty, Jean-Yves, and Matthias Wuttig. "The interplay between Peierls distortions and metavalent bonding in IV–VI compounds: comparing GeTe with related monochalcogenides." Journal of Physics D: Applied Physics 53.23 (2020): 234002.

[2] Zeiger, H. J., et al. "Theory for displacive excitation of coherent phonons." Physical Review B 45.2 (1992): 768.

[3] Cheng, Yu-Hsiang, et al. "Coherent control of optical phonons in bismuth." Physical Review B 96.13 (2017): 134302.

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