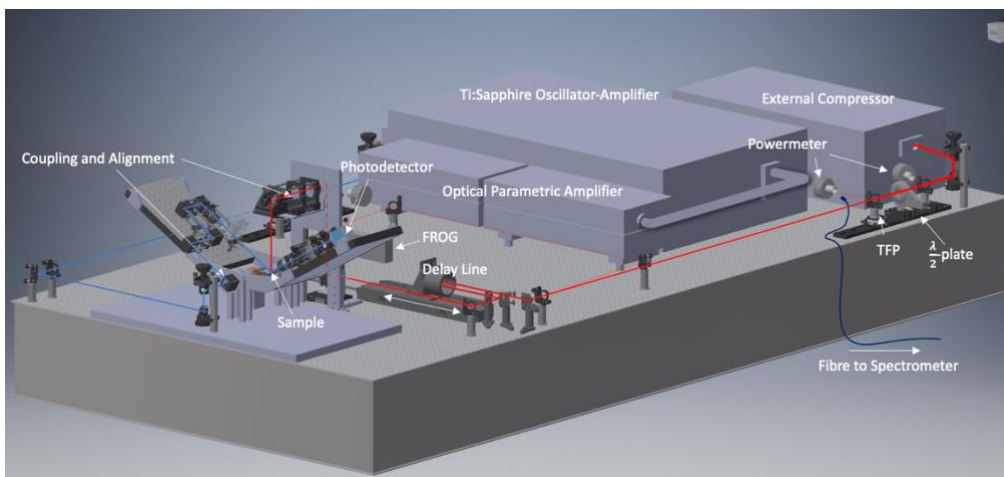


Topic for a master thesis

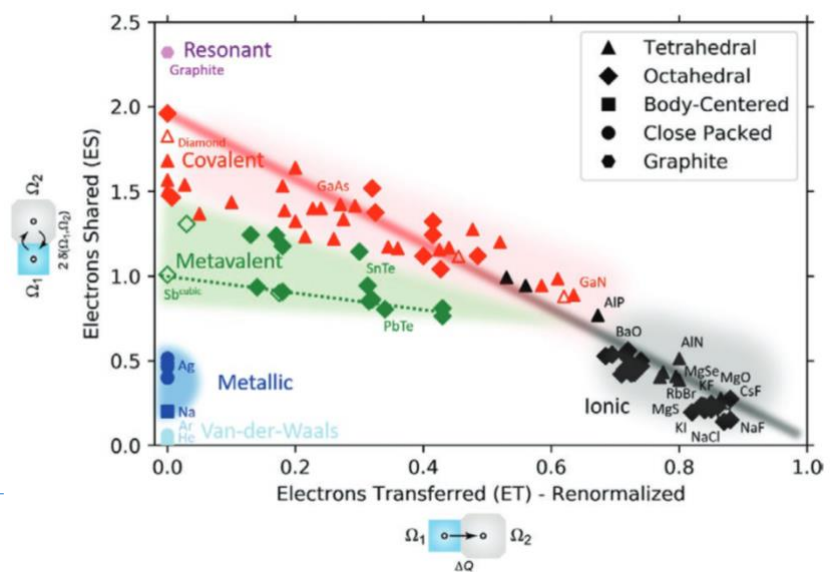
Investigating the ultrafast response of metavalently bonded materials performing an optical pump-probe experiment

Phase-Change Materials (short PCMs) have been widely used in optical data storage due to their remarkable properties among which the large optical contrast between a stable crystalline and a metastable amorphous phase may be the most fundamental one. Upon an intense laser pulse, both phases can be switched reversibly in a fast and reliable manner. Thus, a profound understanding of the extraordinary switching kinetics in these systems is crucial to further optimize their applicability in fast data storage devices. In the last year, *Persch et al.* found a deep link between the speed of crystallization and the underlying bonding mechanism for the material systems GeTe-GeSe, GeTe-SnTe and GeTe-Sb2Te3 [1]. In those investigations, the reflectivity contrast between the amorphous and the crystalline state was detected as a function of time with nanosecond resolution. It turned out that the speed of crystallization depends on the bonding mechanism in the crystalline state; more precisely: fast crystallizing materials could be allocated to systems where the competition between electron localization and delocalization plays a crucial role, i.e., in metavalently bonded systems. Interestingly, *Waldecker et al.* demonstrated, that the prototypical phase-change material GeSbTe225 reveals a large modification in the dielectric function upon reversible excitation with an ultrashort laser pulse ($\tau \sim 100$ fs), indicating that a large property contrast can be induced much faster than processes of atomic rearrangement [2]. This finding suggests the assumption, that a distortion of the metavalently bonded state can already lead to a significant change in the optical properties on femtosecond timescales, making those systems promising candidates for application in ultrafast photonic devices.



The **experimental setup** includes a Ti:Sapphire femtosecond laser generating pulses with a width of 100 fs for an optical pump-probe experiment. An intense 800nm pump-pulse irradiates the sample near to normal incidence while a weaker 400nm probe-pulse (generated in an optical parametric amplifier) is delayed relative to the pump-pulse to characterize the samples reflectivity under an adjustable angle of incidence. The reflected probe signal is detected either by a Si-photodiode or a CCD/CMOS-camera. Applying this scheme, changes in reflectivity upon optical excitation can be detected with a temporal resolution of around 100 fs.

Within the **scope of this thesis**, a materials map will be applied to investigate trends in the ultrafast response of optically excited metavalently bonded systems. The non-thermal manipulation of bonds in the initially crystalline state and its influence on the optical properties of the sample will be investigated with special attention to the displacive excitation of coherent phonons (DECP) driven by the ultrashort pump-pulse [3]. The excited optical phonon modes are in the range of a few THz and are visible as oscillations in the transient reflectivity traces. As the mechanism behind DECP is fundamentally linked to the Peierls-like distortion, which in turn is typical for metavalently bonded materials, a systematic investigation applying predictions from the materials-map would be promising. The pump-pulse fluency will be tuned to the reversible excitation regime of the sample material (increasing fluencies induce amorphization / ablation) allowing to investigate furthermore the electronic relaxation processes which are linked to the electron-phonon coupling strength.



[1] C.Persch et al. The potential of chemical bonding to design crystallization and vitrification kinetics, *nat. comm.* (2021) 12:4978

[2] L. Waldecker et al. Time-domain separation of optical properties from structural transitions in resonantly bonded materials, *nat. mat.* (2016) 14:991

[3] H.J. Zeiger et al. Theory for displacive excitation of coherent phonons, *Phys. Rev. B*, (1992) 45(2):768

M. Sc. Julian Mertens
M. Sc. Jonathan Frank
✉ mertens@physik.rwth-aachen.de
✉ frank@physik.rwth-aachen.de