Band-resolved relaxation of laser-excited gold

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When a noble metal is irradiated with a short-pulsed laser in the visible spectrum, sp- and d-electrons are excited into energetically higher free states while the phonons are not directly affected. This process increases the energy content of the electron system and induces non-equilibrium electron distributions including an imbalance in the band occupation. We investigate how this non-equilibrium evolves towards Fermi distributions. We apply full Boltzmann collision integrals for the excitation, electron-electron scattering and electron-phonon scattering. Our approach resolves the distributions in the sp- and d-bands and shows that temperatures are established at different rates.

After Fermi-distributions have been established, an occupational non-equilibrium of the considered bands can still persist [1]. Our kinetic calculations confirm that a transient under- or overpopulation of the sp-band can be controlled via the wavelength of the exciting laser. However, we find that both population mismatches can also be reached by varying the laser intensity for a given wavelength. We further investigate how an occupational non-equilibrium affects the electron-phonon coupling. We find a strong dependence of the coupling parameter on the band occupation reflecting features of the band-resolved density of states [2]. Our results demonstrate the importance of non-equilibrium electron distributions on the heating of the crystal lattice and subsequent phase transitions.

References:

[1] P. Ndione, S. T. Weber, D. O. Gericke and B. Rethfeld, Scientific Reports **12**, 4693 (2022).

[2] T. Held, S. T. Weber and B. Rethfeld, arXiv:2308.01067 (2023).