Femtosecond time resolved pump-probe spectroscopic ellipsometry – introduction and examples

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I will introduce the novel method fs-time resolved pump-probe spectroscopic ellipsometry (TSE),[1] as a mighty instrument to investigate the electronic structure and lattice properties of materials in great detail, going beyond many established experimental methods. TSE measures the transient complex dielectric function (DF, $\widetilde{\varepsilon}(\tau)$) after optical excitation of charge carriers, which gives us a powerful tool to probe band structure, joint-density of states and transition matrix elements in various regions of the Brillouin zone (BZ) as well as dynamic phenomena like carrier-carrier and carrier-phonon scattering, excitation, and relaxation. As all this properties and effects depend on the transient carrier density, analysis of TSE data provides vast knowledge about the electronic structure dynamics of materials, particularly valuable for fundamental theory approaches.

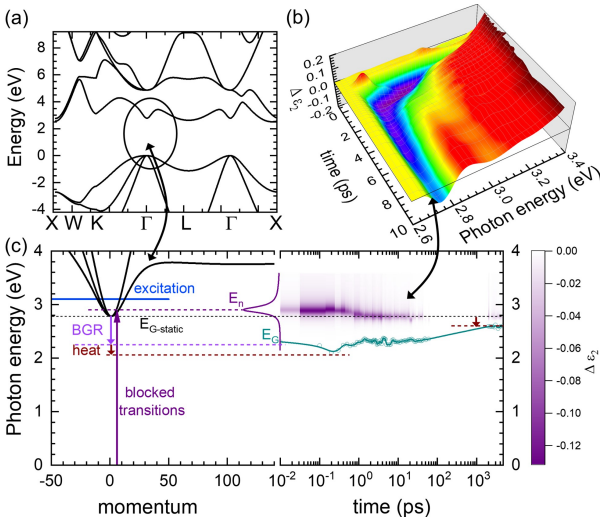

Fig. 1. (a) Band structure of GaP. (b) $\Delta\varepsilon_2(\tau)$ as function of delay time showing blocked and enhanced transitions. (c) Time evolution of the transition responsible for the strongest blocking seen in b) as violet feature: Energy-difference between the lowest CB and topmost VBs around Γ (left) and the assigned amplitude of the model function (violet shading and profile at $\tau = 0\,\mathrm{ps}$, right) as a function of time.[5]

After excitation with an intense pump-laser, electrons and holes are created in the conduction and valence bands, respectively. These excited carriers then can scatter within the BZ and interact with the lattice. This leads to dynamic carrier distribution changes in energy and momentum within time scales of fs up to ns or longer. The ellipsometry probe pulse then feels the actual energetic charge carrier distribution at a given delay time, as expressed in changes of the DF due to Pauli blocking or rising new transitions, energy shifts, as well as Drude response induced by these excess carriers in the respective states. When modelling the experimentally found transient DF with appropriate line shape model functions under mutual comparison with theoretically obtained data for the band structure and joint density of states, we can identify the positions of the charge carriers within the band structure in time, energy, and momentum. As an example, Fig. 1 shows band structure and the transient DF change $\Delta\varepsilon_2(\tau)$ for the classic semiconductor GaP.[5] The observed strong negative signal is related to absorption suppression due to excited carriers of the fundamental transitions at the Γ point. The dynamics of this was modelled with a Tauc-Lorentz line-shape function, whose amplitude and energy evolution is shown in (c).

In my presentation I will introduce technical details of the method TSE, discuss modelling strategies, and give examples of processes observed in various material systems as ZnO, Ge, Si, InP, GaP, ScN, and MoS_2.[2, 3, 4, 5]

References

[1] S. Richter, M. Rebarz, O. Herrfurth, S. Espinoza, R. Schmidt-Grund, and J. Andreasson, Rev. Sci. Instrum. **92**, 033104 (2021).

[2] S. Richter, O. Herrfurth, S. Espinoza, M. Rebarz, M. Kloz, J. A. Leveillee, A. Schleife, S. Zollner, M. Grundmann, J. Andreasson, and R. Schmidt-Grund, New J. Phys. **22**, 083066 (2020).

[3] C. Emminger, S. Espinoza, S. Richter, M. Rebarz, O. Herrfurth, M. Zahradník, R. Schmidt-Grund, J. Andreasson, and S. Zollner, Phys. status solidi: Rapid Res. Lett. **16**, 2200058 (2022).

[4] S. Espinoza, S. Richter, M. Rebarz, O. Herrfurth, R. Schmidt- Grund, J. Andreasson, and S. Zollner, Appl. Phys. Lett. **115**, 052105 (2019).

[5] R. Schmidt-Grund,N. Stiehm, M. Zahradnik, S. Espinoza, M. Rebarz, Y. Slimi,E. Runge, J. Andreasson, and S. Krischok, "Ultra-fast dynamics of the electronic system in GaP after laser excitation", submitted.