Ultrafast Structural and Optical Dynamics in Photoexcited Thin-Film and Layered Materials

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Understanding the transient response of photoexcited materials requires tracking both the evolution of the electronic structure and the accompanying lattice rearrangements on ultrafast timescales. Here, we highlight how complementary optical and structural probes provide access to these coupled processes in representative semiconductor systems. Our comparative approach combines time-resolved spectroscopic ellipsometry, which directly resolves transient changes in the dielectric function, with time-resolved X-ray diffraction, which reveals photoinduced structural distortions inaccessible to optical probes alone.

For zincblende cubic GaN thin films, femtosecond pump–probe spectroscopic ellipsometry captures pronounced carrier-induced modifications of the dielectric response near the band edge. The data reveal ultrafast shifts of the absorption onset driven by band filling and band-gap renormalization, followed by relaxation channels involving carrier cooling, recombination, and lattice heating [1]. For layered GaS, time-resolved X-ray diffraction provides direct evidence of laser-induced structural changes associated with transient and irreversible modifications of crystal anisotropy, including an elongation of the unit cell along the *c*-axis under femtosecond excitation [2].

Together, these case studies illustrate how correlated optical and structural measurements help disentangle nonequilibrium processes in photoexcited semiconductors across different dimensionalities and excitation regimes. Such a combined perspective is essential for understanding the microscopic origin of ultrafast functionality and for guiding the optical engineering of emerging materials under strong photoexcitation.

[1] Baron E., Goldhahn R., Espinoza S., Zahradnik M., Rebarz M., Andreasson J., Deppe M., As D. J. and Feneberg M. (2023) *J. Appl. Phys.*, 134, 075702 and 075703.
[2] Khakurel K. P., Clady R., Espinoza S., Chaulagain U., Ferre A., Andreasson J., Losurdo M., Juan D., Uteza O. and Gutierrez-Vela Y. (2025) *Opt. Mater. Express*, 15, 2534–2544.

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