Max-Planck-Institut für Struktur und Dynamik der Materie

Max Planck Institute for the Structure and Dynamics of Matter

Friday, December 8th 2017 - 11:30

CFEL Seminar room IV (Bldg. 99)

Michael Zürch

University of California at Berkeley

Photoexcitation dynamics in semiconductors and two-dimensional semiconductors observed by attosecond core-level spectroscopy

Investigation of the ultrafast photoexcited electronic response in semiconductors has provided invaluable insights into carrier dynamics. Germanium and its alloys with Si have promise for creating multi-junction solar cells with higher efficiency and mid-infrared optoelectronics. However, the dynamics are complicated by multiple energetically similar valleys, rendering an understanding of carrier thermalization and population inversion following photoexcitation difficult. Attosecond transient absorption spectroscopy (ATAS) has recently been employed to probe ultrafast electron and hole dynamics in germanium at the $M_{4,5}$-edge (~30 eV). In the experiment, a 5 fs VIS-NIR pump pulse excites carriers across the direct band gap and the dynamics are probed with a time-delayed broadband extreme ultraviolet pulse generated by high harmonic generation in xenon spanning ~20-45 eV. The observed transient absorption signal contains the energetic distribution of both carriers, electrons and holes, due to state blocking as well as spectroscopic features induced by bandshifts (e.g. due to band gap renormalization) and broadening (e.g. due to many body effects). By iterative procedures the measured signal can be successfully decoupled into these contributions resolving the carrier and band dynamics with excellent time and energy resolution. Hot carrier relaxation on a 100-fs time scale and carrier recombination on a 1-ps time scale were observed in nanocrystalline Germanium. Going from bulk semiconductor to two-dimensional layers, long-lived core-exciton states are observed at the $MoN_{2,3}$ edge between 32 and 35 eV in MoS_2. Comparing the XUV absorption spectra of bulk and monolayer MoS_2, a ~4 eV red-shift suggests a tightly bound core-exciton. The lifetime of the core-exciton states can be directly measured in the time domain. Furthermore, transient Stark shifts, coherences, and coherent population transfer between different core-exciton states are observed.

References:

[1] L. J. Borja, M. Zürch, C. D. Pemmaraju, M. Schultze, K. Ramasesha, A. Gandman, J. S. Prell, D. Prendergast, D. M. Neumark, and S. R. Leone, "Extreme ultraviolet transient absorption of solids from femtosecond to attosecond timescales," J. Opt. Soc. Am. B 33, 000C57 (2016).

[2] H.-T. Chang, M. Zürch, P. M. Kraus, L. J. Borja, D. M. Neumark, and S. R. Leone, "Simultaneous generation of sub-5 femtosecond 400 nm and 800 nm pulses for attosecond extreme ultraviolet pump-probe spectroscopy," Opt. Lett. 41, 5365 (2016).

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[4] M. Zürch, H.-T. Chang, P. M. Kraus, S. K. Cushing, L. J. Borja, A. Gandman, C. J. Kaplan, M. Hwan Oh, J. S. Prell, D. Prendergast, C. D. Pemmaraju, D. M. Neumark and S. R. Leone, “Ultrafast Carrier Thermalization and Trapping in Silicon-Germanium Alloy Probed by Extreme Ultraviolet Transient Absorption Spectroscopy,” Struct. Dyn. 4, in press (2017).

[5] S. K. Cushing, M. Zürch, P. M. Kraus, L. M. Carneiro, A. Lee, H.-T. Chang, C. J. Kaplan, and S. R. Leone, “Valley-Specific Hot Phonon and Carrier Relaxation Pathways in Si(100) Determined by Transient Extreme Ultraviolet Spectroscopy,” arXiv arXiv:1705.04393 (2017)

Host: Andrea Cavalleri

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