

Outlook on time resolved studies of photoexcitation in semiconductors by X-ray spectroscopy

Federico Boscherini

Department of Physics
University of Bologna, Italy

federico.boscherini@unibo.it



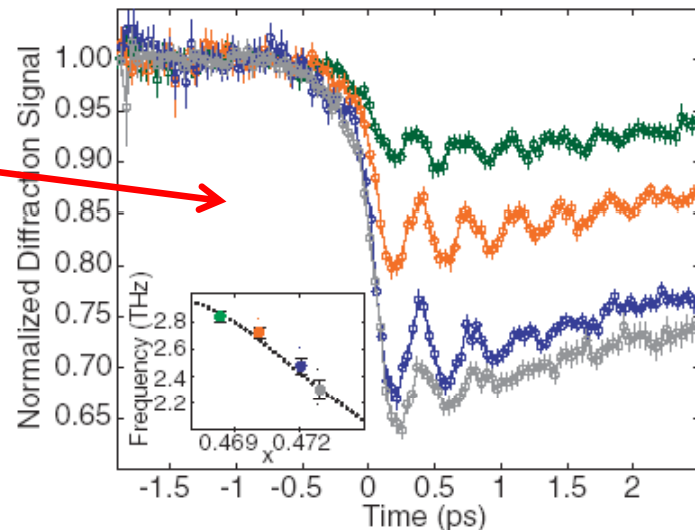
Plan

- PP structural studies in systems with strong electron – lattice coupling
- Deep centers in semiconductors
- Photoexcitation in semiconductors
 - PP optical spectroscopy
 - PP photoemission
- Opportunities to study structural relaxations following photoexcitation

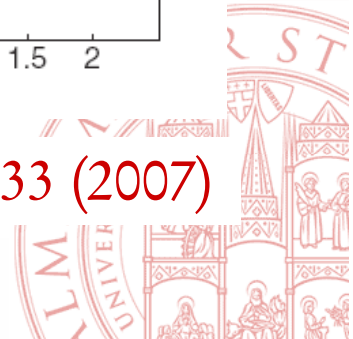


PP studies of strong electron - lattice coupling in condensed matter

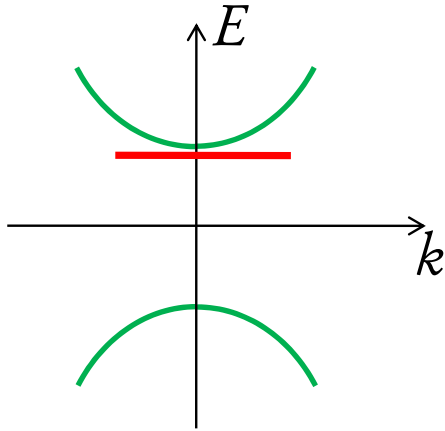
- Optical pump & X-ray probe experiments of particular interest when coupling between electron and lattice is strong
- Examples
 - Bi: Peierls distortion along trigonal axis
 - Perovskite superlattices (Bargheer et al)
 - Correlated oxides (van Loosdrecht et al)
 -



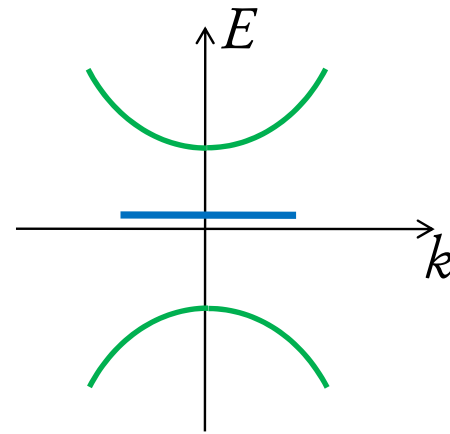
Fritz et al., Science 315, 633 (2007)



Dopants in semiconductors



“Shallow” dopants
energy levels near
band edges
(e.g. As in Si)

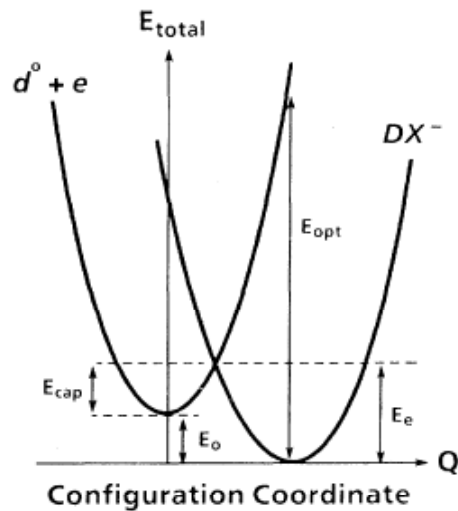


“Deep” centers
energy levels near midgap
(e.g. Si in AlGaAs)

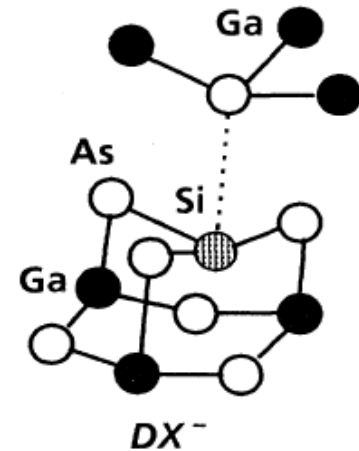


Deep centers in semiconductors

- Deep centers exhibit strong local structural distortions and strong electron – lattice coupling



DX^- center in AlGaAs
(Chadi & Chang, PRB 39, 10063, 1989)

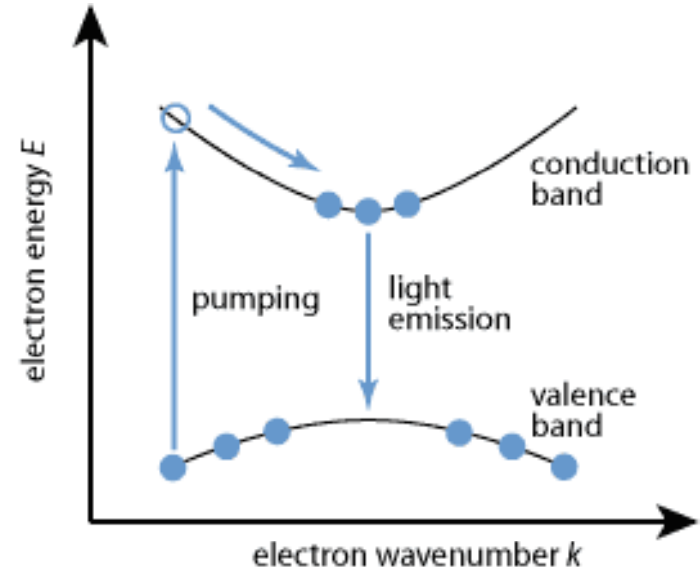


- Large difference between thermal and optical binding energies

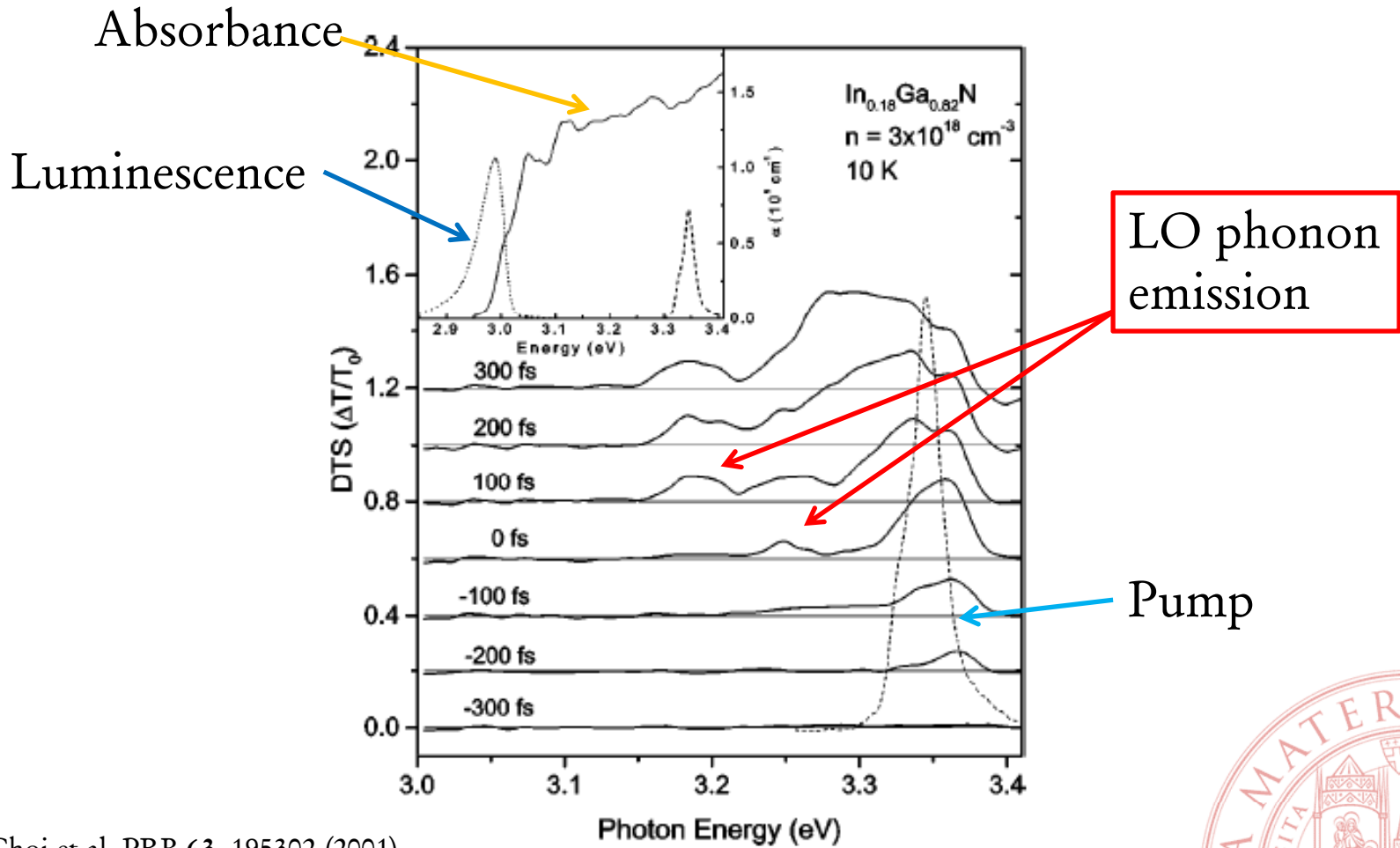


Photoexcitation in semiconductors

- After photoexcitation: thermalization
 - 100 fs – 1 ps: scattering & optical phonon emission
 - 1 – 100 ps: acoustic phonon emission
 - ~ 100 ps: exciton creation
 - e-h recombination
- These process now studied by pump – probe optical spectroscopy and photoemission



PP optical spectroscopy - InGaN



Choi et al, PRB 63, 195302 (2001)



PP photoemission – Si(001) surface

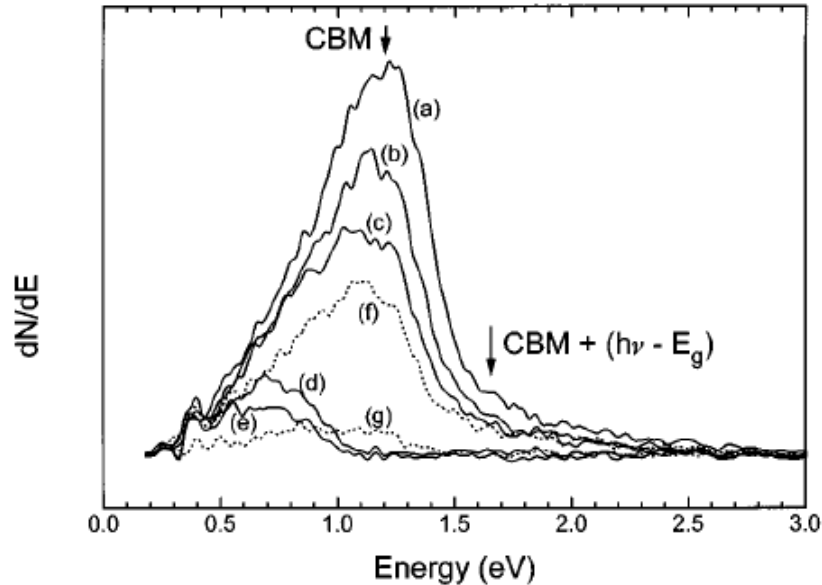
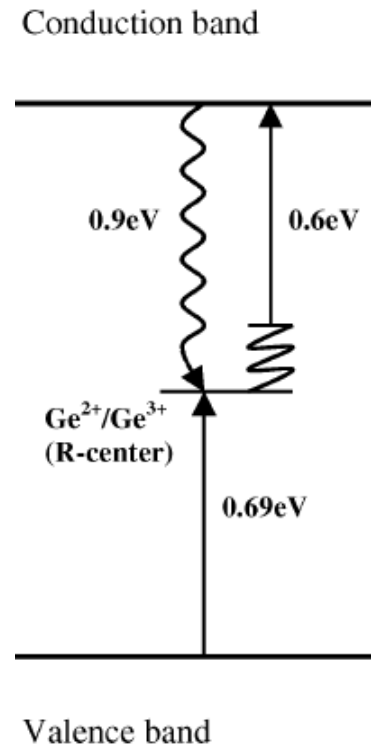


FIG. 3. Time-resolved spectra at different time delays: (a) $T=0$ psec, (b) $T=0.13$ psec, (c) $T=0.33$ psec, (d) $T=1.07$ psec, (e) $T=2.4$ psec, (f) $T=-0.27$ psec (dashed line), (g) $T=-0.47$ psec (dashed line).



Deep states in the gap, Ge:CdTe

- Ge forms a deep level in the gap of CdTe

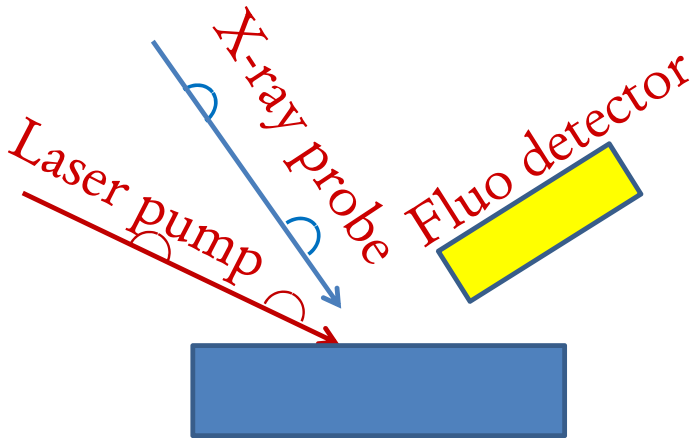
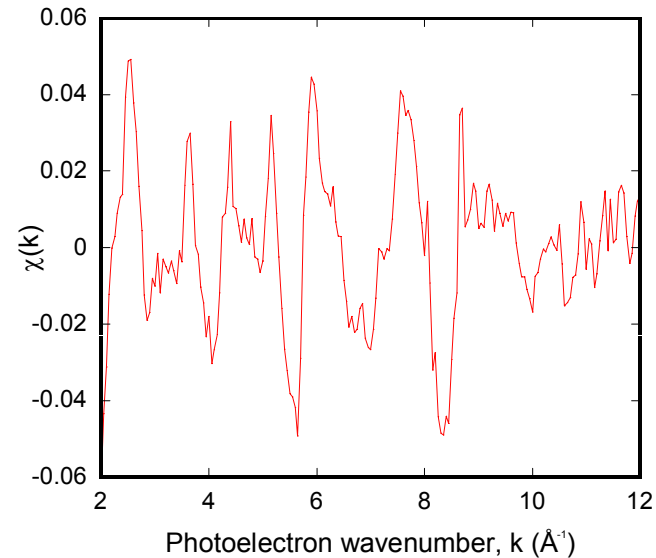


Fiederle et al,
J. Cryst. Growth 243, 77 (2002)



Probing excited states of Ge: CdTe

- Pump – probe scheme to
 - Excite Ge dopants (Ti: sapphire fs laser)
 - Probe structure with fs XAFS



XAFS spectrum
 10^{17} Ge cm^{-3} in CdTe
Recorded at ESRF / GILDA
with 10^{14} total photons
→ Feasible with not too many
pulses of EU-XFEL

fs XAFS of dopants

- Opportunity: follow in real time structural changes associated to photoexcitation of dopants and subsequent thermalization and recombination
- Relevance: provide structural basis for interpretation of photoexcitation phenomena in semiconductors



fs XAFS of dopants: issues 1

- Spot size: not important ($\sim 1 \text{ mm}^2$ ok)
 - Maybe better not to focus to avoid damage
- Divergence: not crucial
- I_f/I_0 normalization crucial (10^{-3} or 10^{-4})
- Energy
 - Range: 3 – 12 or 20 keV ok for important elements
 - Bandwidth: 10^{-4}
 - Scanning:
 - at least 50 eV around edge for XANES (many recent improvements in analysis!)
 - As much as 1000 eV for EXAFS



fs XAFS of dopants: issues 2

- Sample integrity: to be investigated; consider rotating & cooled sample
- Diagnostics: single pulse flux monitor
- Vacuum set – up : ok
- All dopant atoms are optically excited within x-ray absorption length
 - Dopants are dilute, matrix is transparent to below gap excitation: pump beam penetrates much more than probe beam
 - $\sigma_{\text{opt}} \gg \sigma_{\text{x-ray}}$
- Fluorescence detector should be optimized
 - Energy resolution, solid angle acceptance, linearity



Conclusions

- XFELs will allow to probe the structural changes associated to photoexcitation in semiconductors on the fs time scale
- This could open new exciting research topic
- Many opportunities and challenges and issues to be solved

