adamsb1.tex
Bernhard W. Adams: adams@mail.desy.de
Matthew F. DeCamp: decamp@umich.edu
Eric M. Dufresne: dufresne@umich.edu
David A. Reis: dreis@umich.edu
MHATT-CAT
Sector 7
Materials Science

Picosecond Laser-Pump, X-Ray Probe Spectroscopy of GaAs

[1]B. W. Adams, [2]M.F. DeCamp, [2]E.M. Dufresne, [2]D.A. Reis
[1]Argonne National Laboratory, Argonne, IL, USA
[2]University of Michigan, Ann Arbor, MI, USA

Introduction

The combination of ultrafast lasers with x-rays holds great promise in the study of ultrafast phenomena and a number of representative experiments have been done in recent years. Here, a laser-pump, x-ray-probe experiment on GaAs is described. Further development of the technique could eventually lead to a powerful spectroscopy of the bandstructure and carrier dynamics in semiconductors where the core level of the x-ray transition is an absolute energy reference. Other than in absorption and luminescence spectroscopy with only visible-light photons, the information obtained is direct and not convoluted over all pairs of states with a given energy difference.

Methods and Materials

In the present experiment [1, 2], conducted at station 7ID-D of MHATT-CAT, x-rays were monochromatized to match a transition from the Ga K level to the valence band of a GaAs sample and pulses of frequency-tripled light from a 1 kHz Ti:sapphire laser were focused onto the sample (see fig. 1). The photon energy of 4.6 eV corresponds to an absorption maximum in GaAs due to parallel branches in the valence and conduction bands. Being just short of the Ga K absorption edge, the x-rays suffer little absorption, and there is only a small Ga K_α fluorescence yield due to the tail of the lifetime-broadened K core level. When states in the valence band are vacated by laser-induced transfer of electrons to the conduction band, the Ga K absorption cross section goes up and the Ga K_α fluorescence increases accordingly. The x-rays do thus probe the valence holes as they are created and relax to the direct band gap, until they are eliminated by recombination. This is shown schematically in fig. 2.

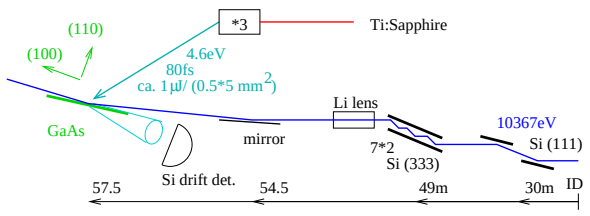

Figure 1: *A schematic of the setup with x-ray monochromator, channel cut (see text), focusing lens, harmonic suppressing mirror, and femtosecond laser system.*

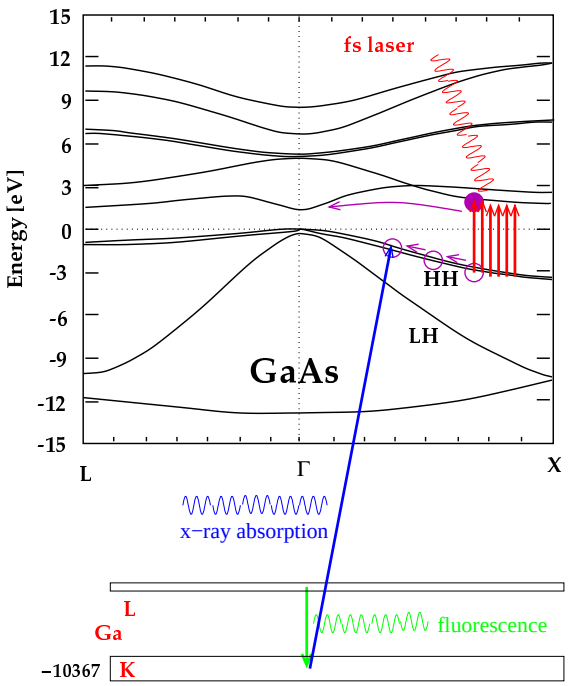

Figure 2: *Band structure of GaAs and core levels of Ga, with x-rays probing laser-vacated valence states.*

The x-ray monochromator was calibrated by plotting the Ga K_α fluorescence yield without laser excitation over the x-ray energy (see fig. 3), with the dots representing a simple monochromator scan, and the solid line showing a slight increase in contrast due to an additional channel cut Si crystal in the beam path. The remaining width of the

curve is mainly due to the K core hole lifetime broadening. Two energies on the lower end of the absorption edge, indicated by vertical lines in fig. 3, were then selected and the Ga K_α yield from the x-ray bunches that the laser was timed to, normalized by the yield from the other bunches, was measured in scans of the laser timing relative to the x-rays over ± 300ps.

Results

The results, shown in figs. 4 and 5, indicate an increase of the normalized Ga K_α fluorescence, following the laser excitation and persisting for ca. 250 ps. This time is much longer than the relaxation times of the photoexcited carrieres towards the direct band gap, and is consistent with the recombination time in GaAs. The results can thus be explained as an increase in the Ga K absorption cross section due to laser-created holes accumulating at the top of the valence band.

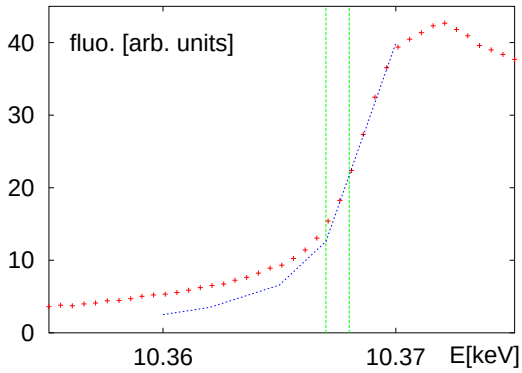

Figure 3: *Energy calibration with the Ga K_α fluorescence. Crosses: using only the main monochromator, solid line: with main monochromator and channel cut (see fig. 1). The vertical lines indicate the energies chosen for the measurement (see figs. 4 and 5).*

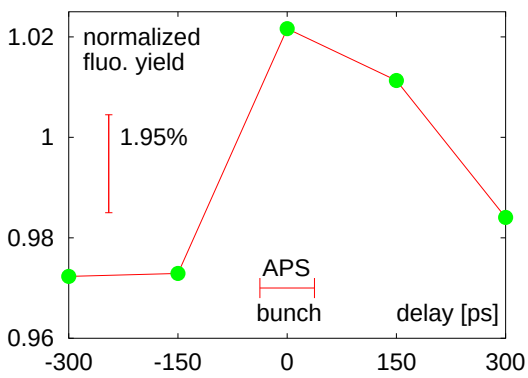

Figure 4: *Normalized Ga K_α fluorescence yield over laser timing at an incident energy of 10.367keV (left vertical line in fig. 3). Poisson statistics are indicated summarily.*

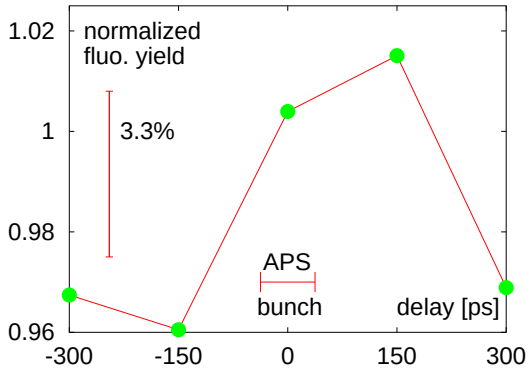

Figure 5: *Normalized Ga K_α fluorescence yield over laser timing at an incident energy of 10.367keV (right vertical line in fig. 3). Poisson statistics are indicated summarily.*

Discussion

To improve the energy resolution beyond the limits of the K shell lifetime broadening, an analyzer crystal for the fluorescent x-rays could be used. It would then become possible to resolve states within the band structure and follow the concentration of valence hole during relaxation after photoexcitation. Besides the spectroscopic application, the principle of this experiment could also be used for ultrafast laser, x-ray correlation [3]. The time resolution would be given by the time constants of hole creation (down to a few femtoseconds) and relaxation (ca. 100 fs with intra-band relaxation of deep valence states).

Acknowledgments

Operation of the MHATT-CAT sector 7 beamlines at the Advanced Photon Source was supported by DOE grant No. DE-FG02-99ER45743. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. Technical support by D.A. Arms is gratefully acknowledged.

References

[1] B. Adams, Rev. Sci. Instrum. **73**, 1632 (2002).

[2] B. Adams, M. DeCamp, E. Dufresne, and D. Reis, Rev. Sci. Instrum **(subm)**, (2002).

[3] B. Adams, Nucl. Instrum. Methods A **459**, 339 (2001).