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Femtosecond time-resolved photoelectron spectroscopy of annealed and sputtered GaP(1 1 0)

M. Leblans[a,b], R.K.R. Thoma[a], J.L. LoPresti[a,*], M. Reichling[c], R.T. Williams[a]

[a] *Physics Department, Wake Forest University, Box 7507 Reynolda Station, Winston-Salem, NC 27109, USA*
[b] *University of Antwerp (UIA), Physics Department, B-2610 Antwerp, Belgium*
[c] *Free University of Berlin, Physics Department, 14195, Berlin, Germany*

Abstract

The relaxation of photoexcited electrons and holes at surfaces of GaP(1 1 0) was studied by means of two- and three-photon photoelectron emission under 3.16 eV photon excitation. With 200 fs time resolution, only the relaxation of surface states within the bulk band gap could be resolved occurring with time constants of 0.3 and 2.5 ps.

Keywords: Photoelectron emission; Surfaces; Semiconductors; Ultrafast spectroscopy

1. Introduction and experiment

Because low-energy laser photoemission has excellent sensitivity for surface defect states in the gap [1, 2], it is particularly well suited for the investigation of surface dynamics and surface recombination of bulk carriers, including the influence of the surface preparation such as sputtering, laser annealing, and adsorption of gases.

We passed the second harmonic (3 eV) of a mode-locked Ti : sapphire laser having a repetition rate of 76 MHz through a Michelson interferometer to generate the excitation and delayed probe pulses, both 3 eV. Normally-unoccupied states were populated (and some occupied states are depopulated) with the first laser pulse following which photoelectron emission for the transient population is excited by the second pulse. Photoelectron spectroscopy was performed in a UHV chamber equipped with a double-pass cylindrical mirror analyzer (CMA) [1, 2]. The GaP(110) sample was cleaned by sputtering and annealing prior to measurement.

2. Results and discussion

A comparison was made between annealed and Ar^+ sputtered surfaces of GaP(1 1 0) . The intermediate state energies are displayed with respect to the surface Fermi level (0 eV), determined using the relation between the intermediate state energy and the electron kinetic energy as seen by the CMA [2] and assuming a two-photon probe process. For both the annealed and Ar^+ sputtered samples the power dependence of the photoelectron yield below and above 0 eV is quadratic and cubic, respectively, showing that both two- and three-photon processes occur. The energy distribution curve (EDC) of

* Corresponding author.
Tel.: [int + 1] 910 759 5515; fax: [int + 1] 910 759 6142;
e-mail:loprejl3@wfu.edu.

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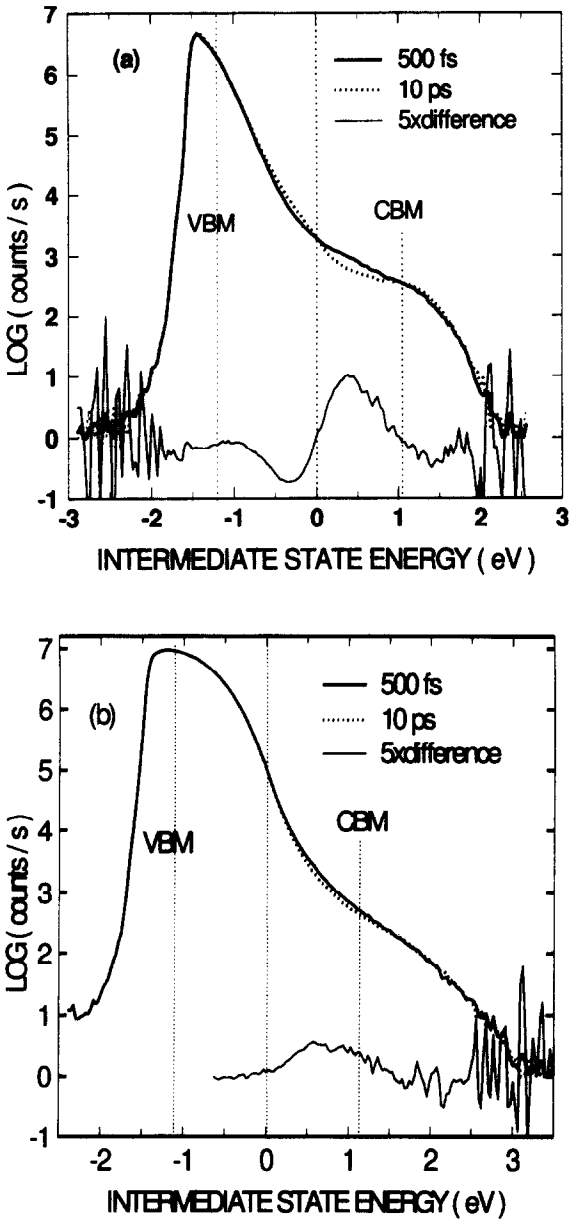

Fig. 1. Energy distribution curves at constant time delay on (a) annealed GaP(1 1 0) and (b) sputtered GaP(1 1 0).

annealed GaP(1 1 0) using fixed delays of 500 fs and 10 ps between the two 3 eV pulses of equal amplitude is shown in Fig. 1(a). For $E_i > 0$ eV, the intensity at 500 fs is higher than the constant background at long delays, while the opposite is true for $E_i < 0$ eV. On both samples the 500 fs and 10 ps EDCs cross each other at the Fermi level. Independently determined, this crossing point can be used as an alternative way to determine the surface Fermi level even in the presence of significant photovoltage shifts. On annealed GaP(1 1 0) the Fermi level is known to be located 1.2 eV above the valence band maximum (VBM) and the bulk band gap is 2.26 eV. These values define the position of the VBM and the CBM on Fig. 1(a). It is apparent that the difference between the EDCs at 500 fs and 10 ps disappears outside the bulk band gap. From time delay scans, the relaxation of the holes in the surface defect states ($E_i < 0$ eV) can be fitted to a single exponential with a time constant of 2.5 ps. In the relaxation curve of the excited electrons in the surface defect states ($E_i > 0$ eV) two different time scales can be distinguished (300 fs and 2.5 ps). The yield of photoelectrons from states in the gap increases by about an order of magnitude after sputtering. As can be seen in Fig. 1(b), the decay of photo-populated states above E_F is also observable on the sputtered sample but no decaying photo-depletion can be observed below E_F. The decay time of the photo-populated gap states above E_F are also in the range 0.3–3 ps on the sputtered GaP.

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