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# Optical anisotropy of Cs nanostructures on III–V(110) surfaces

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## Abstract

We present reflectance anisotropy spectra in the energy range 1.0–5.5 eV, measured for cleaved III–V surfaces after adsorption of caesium. In such a system Cs forms one-dimensional wire-like structures at low Cs coverages ( $\Theta \ll 1$  ML). The formation of these Cs wires leads to characteristic changes in the reflectance anisotropy spectra of the cleaved surface such as a minimum between the  $E_0$  and  $E_1$  bulk critical points of the substrate. For higher coverages the development of a two-dimensional layer can be optically monitored, as can the gradual transition to a three-dimensional disordered layer close to the saturation coverage.

## 1. Introduction

The study of one-dimensional structures, particularly nanowire formation, has intensified in recent years for scientific and technological reasons. In particular, self-assembled nanowires grown on different metal and semiconductor surfaces are believed to be of importance [1]. It was already shown in 1989 that caesium adsorption at room temperature leads to the formation of wire-like structures on the GaAs(110) surface [2, 3]. Later, similar structures were also found on InAs(110) and InSb(110) surfaces [4, 5].

From STM (scanning tunnelling microscopy) studies one knows that on all the III–V(110) cleaved ( $1 \times 1$ ) surfaces Cs forms isolated wires along the  $[1\bar{1}0]$  direction at small coverages ( $<0.3$  ML). Although the formation of Cs wires is a common feature, significant differences between the materials occur, which will be discussed later. In the coverage region around 0.3 ML (GaAs), Cs forms a closed two-dimensional adlayer. For InAs and InSb this 2D phase is completed at a coverage of 0.5 ML, which is one Cs atom per III–V(110) unit cell. At even higher coverages deposition of Cs at room temperature leads to the formation of a second Cs

layer which shows no local order. It is therefore often called the 3D disordered layer or Cs bilayer.

Both the 1D and 2D Cs covered surfaces were found to be insulating by STS (scanning tunnelling spectroscopy) and PES (photoemission spectroscopy) investigations [3, 6]. In the case of the 2D closed layer this result was rather surprising since a metallic surface was expected from the electron counting rule. Several models for this unusual behaviour were suggested, namely that the surfaces are Mott–Hubbard insulators or that bipolaron formation leads to a gap opening [6–9]. Therefore the caesium covered III–V(110) surfaces are not only ideal systems for studying the formation of nanowires but can also be used to study the states of correlated electrons of 1D and 2D Mott–Hubbard insulators. In this first study we focus on the formation of Cs wires and their influence on the spectra of the clean surfaces in the visible range of the spectrum. As we will see, we have found structures which can be directly correlated with the wire formation, showing that reflectance anisotropy spectra (RAS) are useful probes for monitoring the growth of self-assembled nanowires.

## 2. Experimental details

The samples were cleaved in UHV with a base pressure below  $8 \times 10^{-11}$  mbar. Cs was evaporated from SAES Getter alkali dispensers without any significant pressure rise. RAS and Auger spectra, as well as LEED images, were taken after each evaporation step for the first experiments, in order to calibrate the adsorption and link certain RAS to their corresponding LEED images and Auger spectra. The coverage was determined by Auger electron spectroscopy using a VG instruments LEED/Auger system. The Auger intensity of the Cs NOO line at 47 eV relative to that of the substrate Auger lines (As: LMM at 1230 eV, Ga: LMM at 1070 eV, In: MNN at 404 eV) saturates upon completion of the 3D layer. It is already known that for room temperature deposition, not more than one monolayer can be deposited [3]. In order to get an approximation of the coverage, we can therefore normalize the relative Auger intensities of Cs for each evaporation step with respect to the relative intensity at saturation. Please note that there have been some differences in the literature concerning the classification of one monolayer in the case of alkali adsorption. We use the definition of one monolayer as the number of atoms on the clean III–V substrate surface. The coverage calibration has to be done for each of the substrate materials since the sticking coefficient of Cs differs on the substrates investigated here. Using the same condition on the Cs cell we find saturation after 2 h for GaAs but after only 45 min for InAs (for minimized Cs flux). Since the evaporation rates for each Cs source are slightly different, a re-calibration is necessary for each individual dispenser. However, this was done using only the RAS as a reference, after each spectrum was already correlated with the corresponding coverage. The RAS presented here were taken during Cs evaporation using a minimized Cs flux without stopping for additional measurements in order to avoid any changes in the spectra introduced by otherwise necessary optical readjustments.

The RAS measurements utilized a system which was capable of measurements in the energy region from 0.8 to 6.5 eV. The set-up, following the original design of Aspnes [10], consists of a xenon lamp, a CaF<sub>2</sub> photoelastic modulator, MgF<sub>2</sub> Rochon polarizers, an InGaAs photodiode, a Si photodiode and a photomultiplier for the high energy region. The spectral region was limited by the InGaAs photodiode as well as the working range of the photoelastic modulator.

The reflectance anisotropy between two orthogonal crystal axes  $x$  and  $y$  is defined as

$$\frac{\Delta\tilde{r}}{\tilde{r}} = 2 \frac{\tilde{r}_x - \tilde{r}_y}{\tilde{r}_x + \tilde{r}_y}. \quad (1)$$

Since the bulk of zinc-blende III–V semiconductors is optically isotropic, the reflectance anisotropy arises from regions with broken symmetry, particularly surfaces and interfaces. To assign a specific structure of the RAS to a certain transition, one has to compare the measured spectra to calculated ones. For the system discussed in this work this is only possible for the clean surfaces, since calculations for Cs covered (110) surfaces currently do not exist. For details of the interpretation of RAS in general the reader is referred to [11, 12].

Until now, mostly surface state transitions and resonances to bulk states were discussed as the origins of structures in the RAS. For the low dimensional structures other possibilities might exist. In the case of the Mott–Hubbard insulators, as discussed here, transitions involving the Mott–Hubbard states of the surface might lead to RAS signatures. This is expected particularly for the 1D Mott–Hubbard insulator due to the anisotropic nature of the states. Two problems exist though. One is merely a technical one: since such transitions are expected at energies around the surface band gap, which is, for the 2D Cs layer on GaAs, at 0.65 eV [3], a different PEM and detector have to be used in order to access this spectral region. For measurements below 0.5 eV one additionally needs an alternative window for the UHV chamber, since the usual quartz windows will exhibit absorption in this energy range. Such a system was already used by some of us to study the anisotropy of the clean Si(111)-(2 × 1) surface [13] and will be applied to alkali adsorption in the near future. The second problem is of physical origin. If the real part for the RAS is expressed in terms of a three-layer model of a bulk, anisotropic surface layer of thickness  $d$  with a surface dielectric anisotropy  $\Delta\varepsilon = \varepsilon_x - \varepsilon_y$  and a vacuum overlayer, the RAS signal can be written as follows:

$$\operatorname{Re}\left(\frac{\Delta\tilde{r}}{\tilde{r}}\right) = \frac{4\pi d}{\lambda} \operatorname{Im}\left(\frac{\Delta\varepsilon}{\varepsilon_b - 1}\right). \quad (2)$$

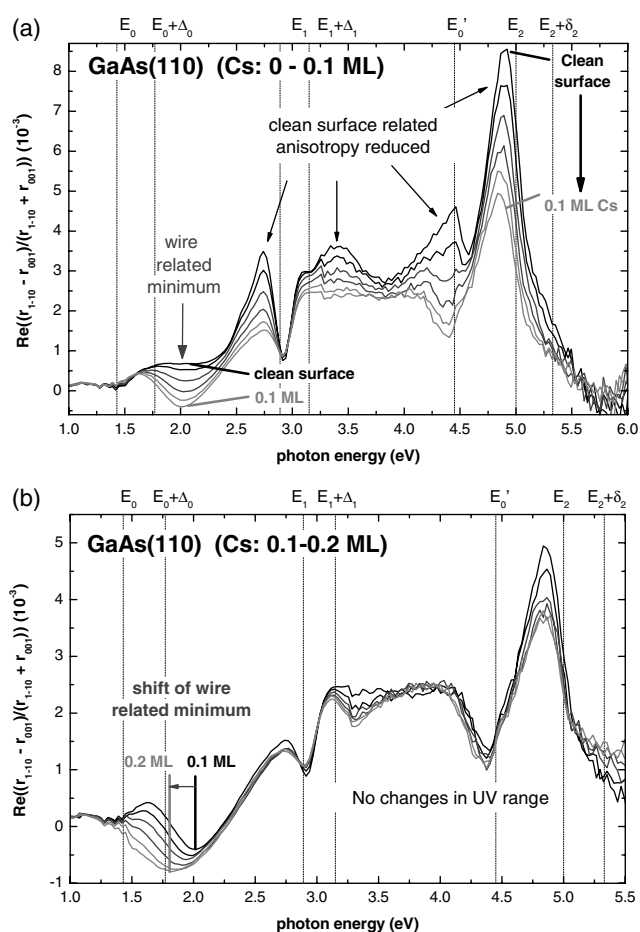
It is possible that the RAS signal in the infrared will be smaller than that in the visible range due to the larger  $\lambda$ . On the other hand, the bulk dielectric constant  $\varepsilon_b$  for most III–V semiconductors  $\operatorname{Im}(\varepsilon_b)$  is small and therefore might compensate the larger  $\lambda$  as is the case for the strong RAS signatures of the clean Si(111)-(2 × 1) [13]. Future measurements have to show whether infrared RAS is sensitive enough for directly measuring the transitions within the Mott–Hubbard gap or possible anisotropic intraband transitions for other, metallic nanowires. Particularly for clearly metallic nanowires, a large signal in the infrared is expected [14].

### 3. Cs on GaAs(110)

GaAs(110) was the first material investigated, since it is the most studied system so far. It was already known that wire formation occurs immediately after adsorption. For a coverage of 0.3 ML a 2D closed layer with a  $c(4 \times 4)$  symmetry was found using STM [3]. For even higher coverages a disordered second Cs layer forms. At the saturation coverage of 1 ML the surface is metallic.

The RAS of the clean GaAs(110) surface are shown in figure 1 (see also figure 6). In earlier studies of the GaAs(110) clean surface anisotropy it was found that the structure at 2.7 eV originates from transitions between surface states while the structure at 3.4 eV is related to transitions between surface and surface-perturbed bulk states [15]. The shoulder at 2.5 eV is attributed to a second surface state transition, which can be clearly resolved at low temperature [16]. The large structure at 4.9 eV was not seen in earlier measurements due to limitations in the spectral range of the RAS set-up [16, 17]. Nevertheless calculations already predicted a strong maximum in this energy region [15].

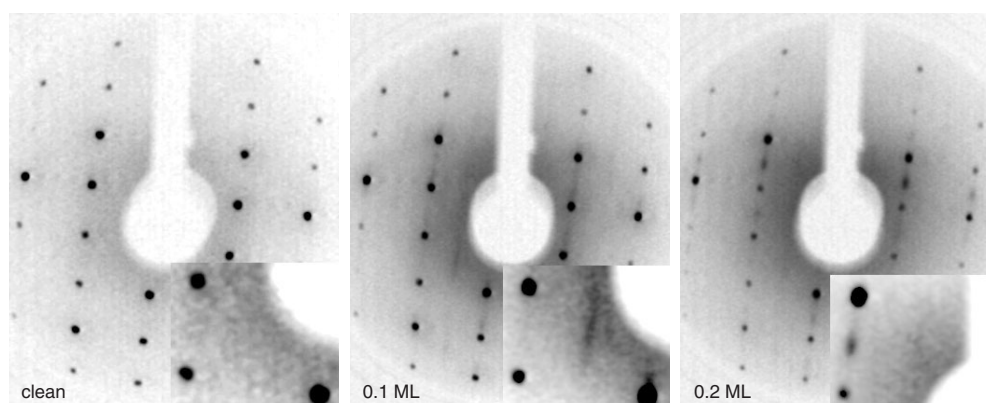
Upon Cs adsorption, structures related to surface states of the clean surface are quenched. Already at 0.15 ML the features at 2.7 and 3.4 eV are substantially reduced. The structure



**Figure 1.** RAS of the GaAs(110) surface with growing Cs coverage. (a) Shows the spectra from the clean surface to 0.1 ML coverage. The coverage difference between the individual spectra is 0.02 ML. At these low coverages a minimum at 2 eV emerges. (b) With growing coverage (0.1–0.2 ML) the minimum is shifted towards lower energies (to 1.75 eV). The coverage difference between subsequent spectra is again 0.02 ML. The energies of the bulk critical points are indicated by the dotted lines and are taken also for all the following materials from [18].

at 4.9 eV is quenched as well, though it can still be seen even at a fully saturated surface. We therefore conclude that this feature originates from transitions involving surface states and transitions involving only perturbed bulk states. This conclusion is consistent with a deconvolution of the calculated optical anisotropy where it is shown that both possible transitions are involved in this large structure [15, 19]. A similar interpretation has been recently proposed for the clean InAs(110) surface [20].

In addition to changes of RAS structures of the clean surface upon Cs adsorption a minimum at 1.9 eV evolves. Since the clean surface does not have a structure in this energy region and no GaAs bulk critical point is near 2.0 eV we attribute this structure directly to the presence of Cs wires. For the coverage range below 0.1 ML it was reported that a single zigzag chain of Cs is dominating the surface, while above that Cs triple chains are formed [2, 3]. In the visible energy range we are not able to observe features which could be directly related to such a change of wire geometry. In the coverage region below 0.1 ML the wire-related



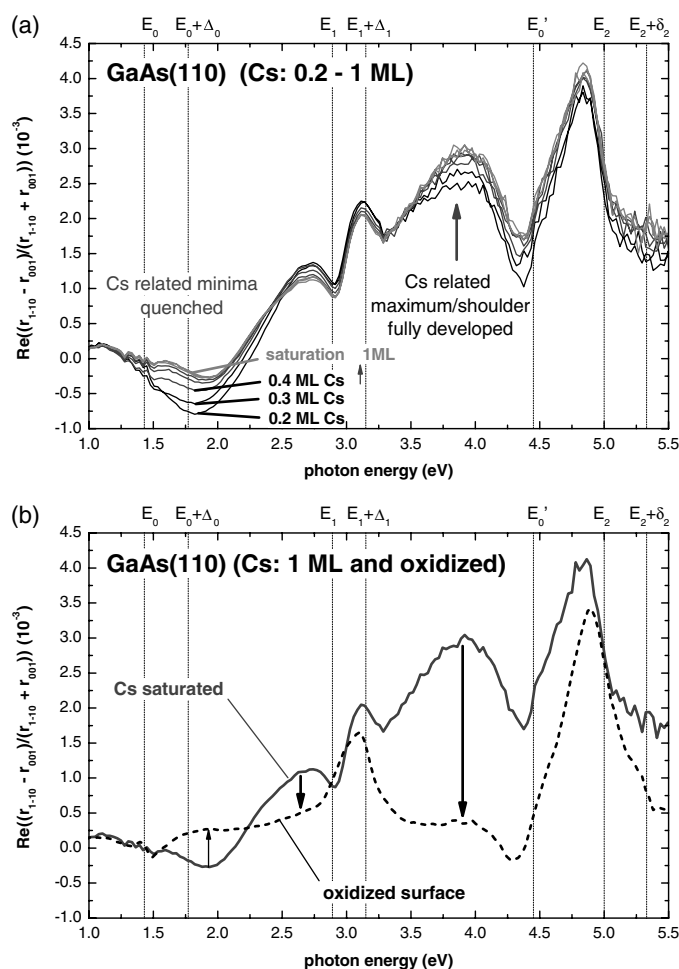
**Figure 2.** LEED images taken at 150 eV electron energy showing the clean surface (left), the streaky  $c(2 \times 2)$  at 0.1 ML (middle) and the  $(2 \times 1)$  symmetry at 0.2 ML (right). The inset in each case shows one  $(1 \times 1)$  unit cell with enhanced contrast.

minimum structure is not pronounced enough for observing possible differences due to the different wire types.

Another prominent change in the RAS is the ‘inversion’ of the peak-like structure at 4.5 eV (around the GaAs  $E'_0$ ) into a minimum. Since this structure is close to a bulk critical point of GaAs, it is not possible to interpret this finding directly as new optical anisotropy within the evolving Cs chain. The nature of this feature is better understood by comparison to other substrates. Consequently we will postpone the discussion to section 6.

Upon further Cs adsorption, exceeding 0.1 ML, the amplitude of the wire-related minimum at 2 eV still increases, but it is accompanied with a gradual shift of the structure to 1.75 eV at a coverage of 0.2 ML (figure 1(b)). It was already reported that the surface undergoes a structural transition into a  $c(4 \times 4)$  reconstructed surface [3]. We therefore attribute this shift to the formation of the closed 2D Cs layer. The shift is gradual since RAS are sensitive to the local bonding configuration but nevertheless integrate over the whole illuminated area of the sample. Parts of the sample are already in the  $c(4 \times 4)$  symmetry, while other parts still have wires. In this transition regime, a linear combination of the structures related to wires (at 2 eV) and to the  $c(4 \times 4)$  phase (at 1.75 eV) results in a gradual shift of the minimum in the intermediate coverage region. LEED images taken in this coverage range from 0.1 to 0.25 ML show a transition from a weak and streaky  $c(2 \times 2)$  symmetry (wires) to a  $(2 \times 1)$  symmetry (closed layer) at 0.2 ML (see figure 2), although the closed 2D layer was reported to be  $c(4 \times 4)$  reconstructed [3]. The latter symmetry was derived from STM images and might differ from the symmetry observed in LEED images. For other systems, namely Li on GaAs(110) [21] and Cs on InAs(110) [22], a  $(2 \times 1)$  symmetry in LEED for the 2D closed layer was also recently reported. We think that this strongly supports the correlation between the changes in RAS and the variation of the surface reconstruction upon the formation of the closed layer. At the moment, we are not able to explain the slight difference from STM data as regards the coverage where the 2D closed layer occurs (0.2 ML here, 0.3 ML in [5]). Although a difference in calibration of the coverage could be the origin, further STM/RAS experiments will clarify our assignment.

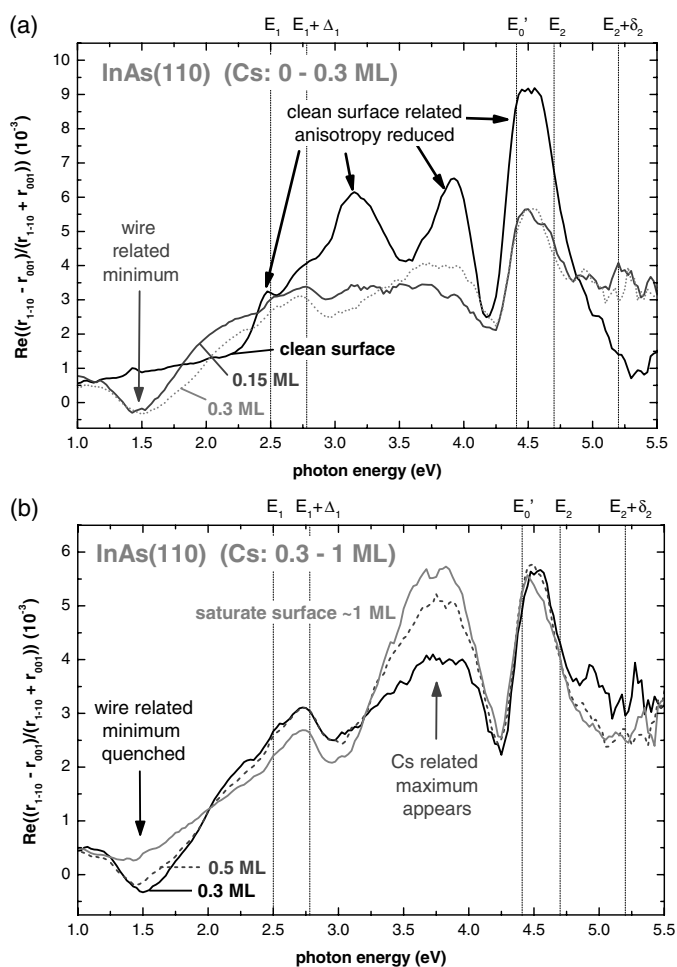
Further adsorption of Cs leads to a disordered second adlayer, which is also known as the 3D phase. Since the STM pictures suggest no local anisotropy in this adlayer [3], one expects a quenching of all RAS features which were attributed to the ordered 2D phase (minimum



**Figure 3.** RAS of the caesium covered GaAs(110) surface upon saturation (a). The minimum related to the two-dimensional phase vanishes, while the maximum at 4.0 eV is getting more pronounced. The spectra were taken during evaporation. The time difference between the individual spectra was 30 min. Since the coverage is not linear with time in this coverage region, there is no constant coverage difference between the spectra. (b) The Cs saturated and oxidized GaAs(110) surface.

at 1.75 eV). The disorder is also seen in LEED images, where we observe only weak spots. As one can see in figure 3(a), the quenching of RAS features related to the ordered phase (minimum at 1.75 eV) is clearly observed. The reduction of the minimum is accompanied with the development of a new structure at 4 eV. This structure is already present in the spectra of the 2D ordered layer, though less intense. To prove whether these structures originate from the Cs layer we oxidized the sample at normal atmospheric pressure for 5 min. The maxima at 4.0 and 2.6 eV and the rest of the minimum structure at 2 eV disappear completely upon oxidation (see figure 3(b)), while all the remaining anisotropies are close to the bulk critical points and are therefore probably related to surface modified bulk states. These structures are always present in the spectra, independently of the Cs coverage.

At this stage, we conclude that in the case of Cs adsorption on GaAs(110) surfaces we are able to attribute distinct features of the RAS to certain reconstructions or wire geometries.



**Figure 4.** RAS of different Cs coverages on the InAs(110) surface. (a) Clean surface, 0.15, 0.3 ML. (b) 0.3, 0.5 and  $\approx 1$  ML.

#### 4. Cs on InAs(110)

The adsorption of Cs on the InAs(110) surface has already been studied by STM and STS [3, 4]. Cs was found to form similar wire structures as on GaAs, although certain peculiarities were reported. In STM images two different wires are visible, distinguishable by their different widths (measured along a line perpendicular to the wire axis) [4]. The ‘narrow’ type (single-atom zigzag chains along the  $[1\bar{1}0]$  direction) occur at coverages below 0.2 ML, while the ‘broad’ type (1D triple lines of atoms along  $[1\bar{1}0]$ ) appear above 0.2 ML. In contrast to the case for GaAs(110), the formation of the 2D phase found at 0.5 ML is not accompanied with a reordering of the Cs, but consists of a closed package of the broader chain type. For larger coverages a similar disordered 3D phase was found [4].

From RAS we are able to distinguish all these phases as well. We see an initial quenching of all structures related to the clean surface (2.47, 3.15, 3.9 and 4.5 eV: see figure 4) similarly to the case for GaAs. A negative structure develops at low coverage at about 1.5 eV, which— analogously to the case for GaAs—we relate to the presence of wires. This minimum is



broadened to higher photon energy when we reach 0.3 ML. In this case, the two different line shapes of the minimum could be attributed to the presence of two different Cs wires at the surface. Since above 0.3 ML—apart from the gradual quenching of the minimum below 2 eV—no further variations are detected in this energy region, it is likely that there is no difference in the local Cs geometry going from the broad wires to the 2D layer (figure 4(b)). As for GaAs, an additional maximum develops at 3.75 eV for the closed 2D layer, becoming more pronounced after the 3D disordered phase is completed.

## 5. Cs on InP(110)

Although no other experimental results for alkali adsorption on InP(110) exist so far, we expected a similar Cs adsorption behaviour, since the atomic geometry and electronic properties of the InP(110) surface are similar to those of GaAs and InAs [23]. Even bulk critical point energies of InP are similar to GaAs, though with a smaller spin-orbit splitting [18].

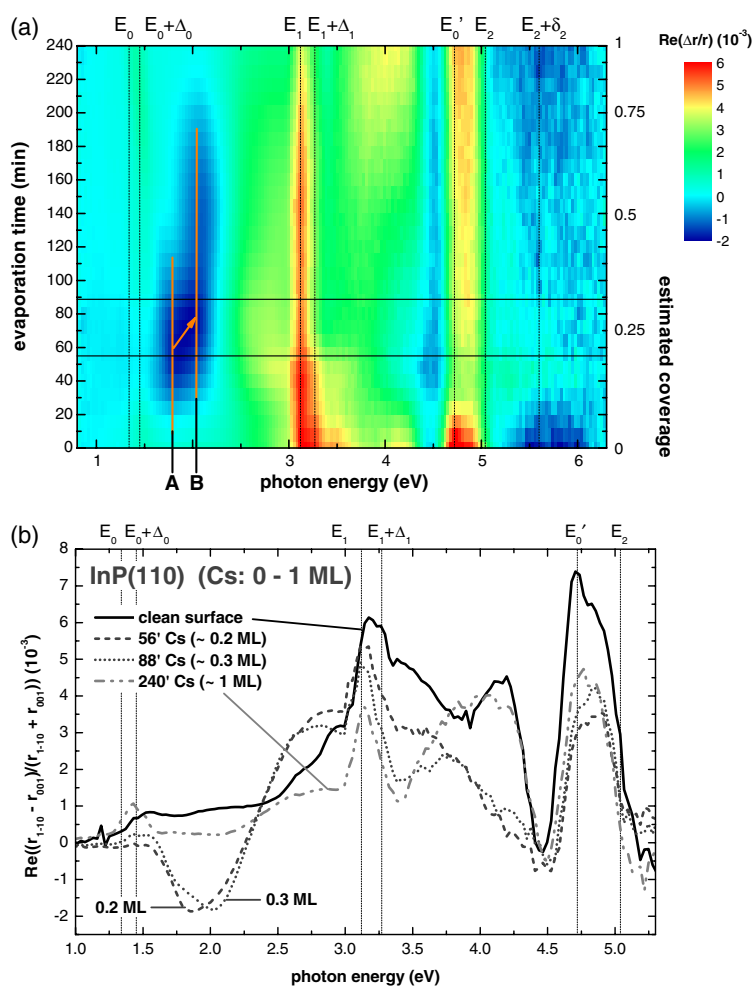
Indeed in RAS we observe changes that are analogous to those for GaAs and InAs. All structures related to the clean surface are quenched upon Cs adsorption: two broad maxima at 3.5 and 4.1 eV as well as two smaller structures at 2.6 and 2.9 eV, already seen in the first RAS measurements of this surface at low temperature [24]. Additionally, in close similarity to the cases for InAs(110) and GaAs(110) a minimum appears between the  $E_0$  and  $E_1$  bulk critical points. This minimum, very probably related to wires, initially occurs at 1.8 eV, and shifts to 2.1 eV above 0.1 ML (see figure 5(b)). We display all RAS in one graph by using colour coding (figure 5(a)). In such a graph one can see that the shift of the minimum is a gradual transition (arrow in figure 5(a)) from one adsorbate configuration to another (indicated by the vertical lines in figure 5(a)). One structure, leading to the minimum at 1.8 eV, can be found from the early stages of adsorption to 0.35 ML, while the other (minimum at 2.1 eV) occurs for coverages above 0.1 ML and vanishes only on formation of the disordered 3D layer. Although no experimental confirmation exists (e.g. from STM), it is likely that a similar transition from one wire type to another happens on InP(110) as on InAs. For higher coverages all changes are similar to those of InAs. At 0.5 ML the minimum at 2.1 eV is still present, with a shoulder at 2.7 eV just below the  $E_1$  critical point. Also a maximum around 4.0 eV starts to develop. At 1 ML the maximum is fully developed and the shoulder at 2.7 eV clearly visible, while the wire-related structure has disappeared. Individual spectra of the different coverage regimes can be also seen in figures 5(b), 6 and 7.

From comparison with Cs/InAs(110) and Cs/GaAs(110) we conclude that the adsorption of Cs on InP(110) also goes through distinct phases: from single isolated wires to a 2D closed layer (probably consisting of a dense package of wires) and finally to a disordered layer at saturation coverage. However, the assignment to certain coverages is more difficult in the case of InP due to the lack of additional data. Our estimation is based on the assumption that the saturated surface consists of 1 ML of Cs and that the 2D layer is complete at 0.5 ML, as in the case for Cs on InAs(110).

## 6. Comparison of the different substrates

In order to attribute the observed features in RAS to certain optical anisotropies coming from transitions within the surface band structure, we can compare the spectroscopic results presented in this work.

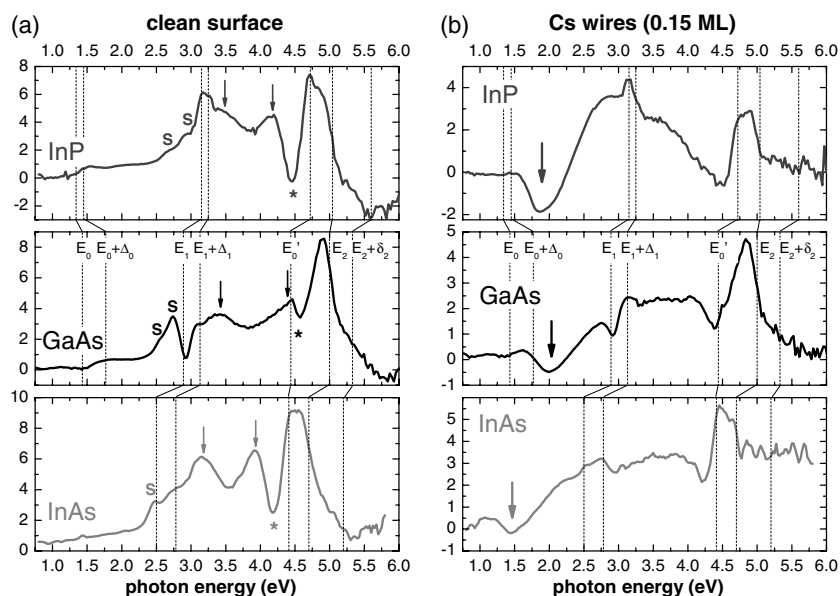
For the clean surface full *ab initio* calculations do exist, which makes assignment of certain features more conclusive. For GaAs(110), tight binding [17, 19] and DFT-LDA (density functional theory within local density approximation) calculations [15] were able



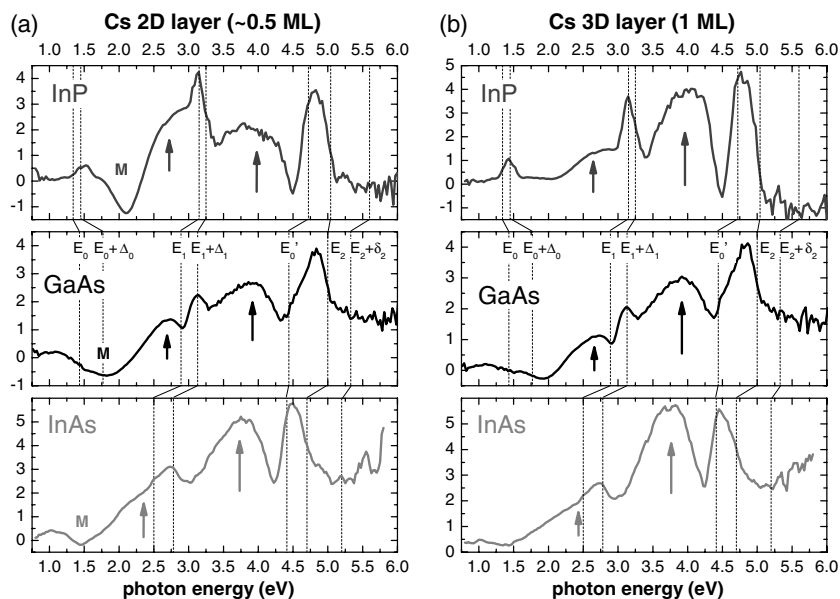
**Figure 5.** (a) RAS colour plot of caesium adsorption on InP. The RAS signal is colour coded (from  $-2$  to  $+6$  according to the legend) and all spectra from the clean surface to complete saturation are shown. Marked with two vertical lines are two minima (A and B) which are observed in the wire regime. (b) Spectra of the clean surface (solid), 0.2 ML (dashed), 0.3 ML (dotted) and 1 ML (dash-dotted). The spectra for the clean and saturated surface correspond to the first and last line in the colour coded image. The two coverages (0.2 and 0.3 ML) are marked with a horizontal line in (a).

(This figure is in colour only in the electronic version)

to supply an explanation of the measured spectra. Recently similar calculations were done for InAs(110) [20]. In the case of GaAs two prominent structures (at 2.5 and 2.7 eV), caused by surface states below the  $E_1$  critical point, are observed (S in figure 6(a)). Similar transitions between surface states in the case of InP and InAs are less evident at room temperature, but clearly seen in low temperature spectra [24]. We propose that for GaAs(110) another surface-related feature is nearly coincident with a negative minimum structure around  $E'_0$  which leads to an almost flat curve in this spectral region for the clean surface. Upon Cs adsorption and the quenching of surface states the minimum around  $E'_0$  appears, since only the surface-related term is reduced upon adsorption. This would lead to the apparent inversion of the



**Figure 6.** (a) Comparison of the RAS of clean III–V cleaved surfaces. The  $(1 \times 1)$  bulk truncated structure is characteristic for these surfaces and also the RAS are similar. Structures related to surface states are labelled S, structures due to surface to bulk transitions with an arrow and the minimum below the  $E'_0$  bulk critical point with \*, (b) The spectra of Cs wires at the different substrates. The main feature in this regime is the minimum (arrows) in the low energy region.



**Figure 7.** (a) Comparison of RAS of the 2D closed layer. The minimum related to the 2D phase is marked with M, other new structures with arrows. (b) Comparison of the fully saturated III–V(110) surfaces; the two structures related to this phase are again marked with arrows.

line shape around  $E'_0$  (see figure 1(a)). As an alternative explanation for such a reversal of sign we could consider strain [25], possibly modified upon Cs adsorption. However, since from theoretical calculations it is expected that Cs introduces different strain fields for GaAs and InAs [26], the fact that similar changes are observed in the RAS of the two surfaces challenges this explanation. Moreover, additional strain should affect the anisotropies at all critical points [25], while we only see changes around the  $E'_0$  bulk critical point. It is then not plausible that strain is responsible for the development of the RAS in this spectral region.

Considering GaAs(110), InAs(110) and InP(110) the RAS features evidently common to all III–V(110) clean surfaces are:

- (i) maxima related to surface state transitions below the  $E_1$  critical point (S in figure 6(a));
- (ii) maxima due to transitions between surface states and bulk-like states in the energy range between the  $E_1$  and  $E_2$  bulk critical points (marked with arrows in figure 6(a));
- (iii) a minimum below the  $E'_0$  bulk critical point (\* in figure 6(a)); and
- (iv) a maximum between  $E'_0$  and  $E_2$  where transitions from surface to bulk and from bulk to bulk states contribute.

Generally upon adsorption of Cs the structures involving states of the clean surface disappear. Structures at bulk critical points, even those visible for completely saturated or oxidized samples, remain. In all cases a minimum structure at low energies can be directly attributed to the Cs wires. It is evident that the energetic position of this minimum is substrate dependent (arrows in 6(b)). It is therefore unlikely that the minimum is caused by transitions between states localized at the Cs–Cs bonds, which are expected to be similar for all substrate materials [26]. For GaAs the minimum is located at 2 eV, which is exactly in the same energy region as a structure seen for Cs adsorption on GaAs(001) [27]. Although the atomic configuration is different in that case, this suggests the involvement of Cs–Ga or Cs–As bonds in the minima structure. The idea that such states are responsible for the minima around 2 eV is also strongly supported by an early combined photoemission and inverse photoemission study [28]. It was shown that upon Cs adsorption on GaAs(110) new surface states evolve, but all of them are above the conduction band minimum. Anisotropic transitions involving these states will therefore occur above the fundamental band gap of GaAs.

Another possibility would be that strain introduced by the Cs chains modifies bulk states, producing characteristic anisotropies. This would be consistent with the fact that the energetic position of the minimum scales with the bulk critical points of the different substrates. This model is supported by the fact that other adsorbates forming chains along the  $[1\bar{1}0]$  direction (such as Sb) lead to a minimum in the RAS around 2 eV as well [29, 30]. On the other hand, it is hard to understand why strain should effect the RAS in a region away from bulk critical points. We favour the assignment of the 2 eV minimum to an anisotropic transition from states localized at substrate atoms to states in the first Cs layer. One measurement possible for gaining new insight into the origin of the wire-induced anisotropy would be an adsorption experiment with Na or K. For the GaAs(001) surface no signal at 2 eV was reported [27], while for the (110) surface it is known that K and Na form similar wires but strain the underlying substrate in different ways to Cs [26].

As already described for the individual substrates the anisotropy changes upon completion of the first Cs layer are similar for all substrates. There is still a Cs-related minimum (M in figure 7(a)) and two new maxima (arrows in figure 7(a)) appear. The minimum is broadened and shifted in comparison to the minima of the wire regime. We attribute this to the change in geometry of the Cs adsorbates, namely the formation of the  $c(4 \times 4)$  reconstruction in the case of GaAs and the formation of broader wires for InAs and InP. The reason can again be either changes in the participating surface states or a change in strain due to different atomic

arrangements. As in the discussion about the wire-related minimum, the first explanation is favoured by the current data. As regards the new maxima, the assignment is similarly difficult. Since the energetic position is much more independent of the substrate, it is likely that transitions between states located at Cs–Cs bonds are involved. The structure at lower energies (2.7 eV for InP and GaAs) is already present in the wire regime, fully developed in the 2D phase and unchanged by the formation of the disordered Cs layer. For InAs it is not very well developed, possibly due to its coincidence with the InAs  $E_1$  bulk critical point. Since it is already present at the 2D layer it is reasonable to attribute it to Cs–Cs bonds within the surface plane, since they are present for all stages of evaporation.

The second and more pronounced maximum at 4.0 eV for InP, 3.9 eV for GaAs and 3.75 eV for InAs appears already in the 2D phase but is fully developed at 1 ML when the second disordered Cs layer is completed. Being independent of the substrate, it probably originates from Cs–Cs bonds between the first and the disordered second Cs adlayer. However, we cannot exclude the possibility that bulk-like states are involved, since they also vary only slightly in this energy region for the substrates used in this study [18]. This conclusion would be supported by recent results of Cs adsorbed on the GaAs(001) surface where a structure at 4 eV has been explained by transitions from bulk-like and surface states into states located in the Cs layer [27]. An additional experiment with InSb(110) could clarify this assignment, since for InSb the  $E_2$  bulk critical point is at a much lower energy than  $E_2$  for GaAs, InAs and InP. Finally, a word about the 4 eV maximum. For Cs/GaAs(110) the structure is already fully developed for the 2D closed surface while on InP and InAs it is clearly maximized upon formation of the 3D disordered layer. If one assumes that states localized at the caesium/substrate interface are involved, this might suggest that a rearrangement of the interface upon saturation occurs in the case of InAs and InP. It would be interesting to have calculations of the formation energies of different interface geometries as well as calculations of the optical anisotropy to explain these differences between the substrates.

## 7. Conclusion

We have shown that RAS are a powerful tool for monitoring the adsorption of Cs on different III–V(110) surfaces. We find similar signatures and changes in the optical anisotropy which we were able to relate directly to different phases of the Cs adsorbate. At low coverages one can monitor the formation of the Cs wires via the occurrence of a minimum structure in the optical anisotropy as well as via the quenching of RAS structures related to the clean surface. By comparing spectra of the different substrates we are able to give a physical explanation for the features occurring in the RAS, though for the wire-induced minimum two possibilities are discussed. New experiments or *ab initio* calculations will clarify whether adsorbate-induced strain or new surface states are responsible for this feature.

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