

**Cs-induced charge transfer on (2×4)-GaAs(001) studied by photoemission**O. E. Tereshchenko,<sup>1,\*</sup> D. Paget,<sup>2</sup> P. Chiaradia,<sup>3</sup> F. Wiame,<sup>4</sup> and A. Taleb-Ibrahimi<sup>5</sup><sup>1</sup>*Institute of Semiconductor Physics, Novosibirsk State University, 630090 Novosibirsk, Russia*<sup>2</sup>*Laboratoire de Physique de la Matière Condensée, Ecole Polytechnique-CNRS, 91128 Palaiseau Cedex, France*<sup>3</sup>*Dipartimento di Fisica, CNISM Unit and NAST, Università di Roma Tor Vergata, 00133 Roma, Italy*<sup>4</sup>*Laboratoire de Physico-Chimie des Surfaces, Ecole Nationale Supérieure de Chimie de Paris, 75231 Paris Cedex 05, France*<sup>5</sup>*Synchrotron SOLEIL, Saint Aubin, BP 48, 91192 Gif sur Yvette Cedex, France*

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Cesium adsorption on (2×4)-GaAs(001) was studied by photoemission and low-energy electron diffraction. The different Cs-induced changes of As 3*d* and Ga 3*d* core-level spectra show that charge transfer is almost complete for Ga surface sites, but is negligible to surface As at a coverage  $\Theta_{\text{Cs}} < 0.3$  ML. The situation becomes opposite for  $\Theta_{\text{Cs}} > 0.3$  ML, at which transfer occurs to As but no longer to Ga. Charge transfer to As atoms leads to surface disordering and destabilization and induces surface conversion from As-rich to Ga-rich (4×2)-GaAs(001) surface after annealing at a reduced temperature of 450 °C.

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**I. INTRODUCTION**

Adsorption of alkali metals on semiconductor surfaces has been investigated in most detail for silicon<sup>1,2</sup> and for the (110) cleavage face of GaAs (Refs. 3–5) for which the nature of chemical bonding, the amount of charge transfer, and the origin of the surface dipole have been essentially clarified. On the other hand, despite of technological applications, the fundamental aspects of adsorption on GaAs(001), including the difference in the adsorption mechanisms at the empty gallium-related and at the occupied arsenic-related dangling bonds is still far from being known. For the (2×4) As-rich surface, the early stages of Cs adsorption, characterized using both x-ray diffraction and theoretical calculations,<sup>6</sup> give rise to adsorption near both As dimers and Ga dangling bonds with a significant charge transfer only in the latter case. It was shown recently that Cs adsorption on the As-rich (2×4) surface produced a lowering by about 100 °C of the temperature of the transition to the Ga-rich (4×2) surface.<sup>7</sup> The microscopic mechanism for the decrease of the bond energy of surface As atoms remains unknown.

The present paper is an investigation of the microscopic nature of the bonding between Cs and (2×4)-GaAs(001) by using photoemission spectroscopy. From the modifications of the Ga 3*d* and As 3*d* core-level spectra (CLS) two regimes are revealed. For a coverage smaller than 0.3 ML, charge transfer mostly occurs at the Ga surface sites, related to second layer gallium atoms. At a larger coverage, the situation is opposite since the Ga 3*d* CLS does not change, while a significant modification of the As 3*d* CLS is observed. These results are interpreted and suggest the bond energy decrease of surface As atoms.

**II. EXPERIMENTAL**

The investigations were performed at the SU3 beamline of the SuperAco storage ring (Orsay, France), in a ultrahigh vacuum (UHV) chamber with a base pressure in the 10<sup>-11</sup> mbar range. The photoemission spectra were obtained using a hemispherical electron analyzer, at an overall energy

resolution on the order of 0.15 eV at photon energy of 100 eV. Clean reconstructed surfaces of epitaxial p-GaAs(001) were obtained by using a treatment by HCl in isopropyl alcohol under dry nitrogen atmosphere, followed by introduction into UHV and annealing to 420 °C.<sup>8</sup> Also available in the same chamber was a low-energy electron-diffraction (LEED) system. The measurements of the work-function changes were performed in an independent setup, also equipped with LEED, by a retarding field method using a LEED gun.<sup>9</sup> Several successive exposures to cesium (20, 30, and 50 min) were performed at room temperature (RT) using thoroughly outgassed dispensers in a vacuum not exceeding 10<sup>-10</sup> mbar. Using a previous Auger investigation,<sup>10</sup> we estimate that a deposition time of 50 min corresponds to a cesium coverage  $\Theta_{\text{Cs}}$  of about 50% of the saturation concentration, the latter concentration being taken as one monolayer (ML). Shown in Fig. 1 are the Cs 4*d* core-level spectra (CLS). As seen from this figure, these shapes consist of one relatively broad doublet, and do not significantly change with

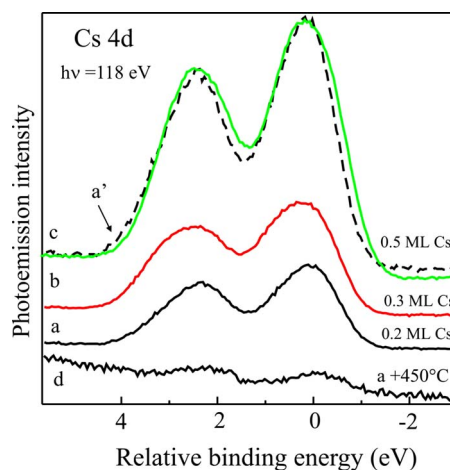


FIG. 1. (Color online) Cs 4*d* CLS at successive coverages of (a) 0.2, (b) 0.3, (c) 0.5, and after annealing to 450° (d). Curve a' is identical to Curve a, with an appropriate multiplication and perfectly corresponds with curve a. The CLS consist in one relatively broad doublet, and do not strongly change with coverage.

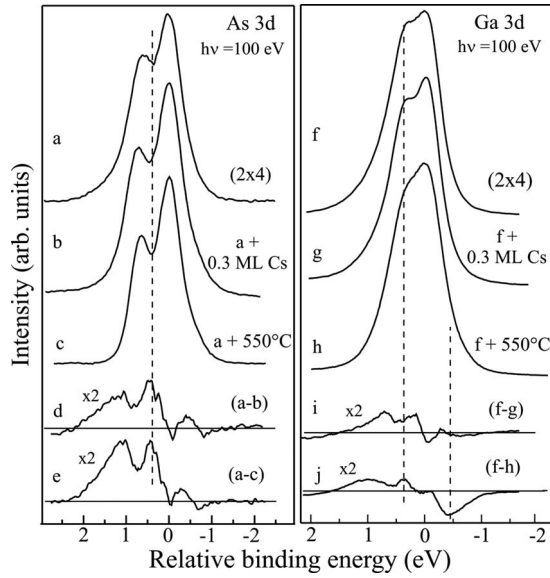


FIG. 2. The left panel shows the As  $3d$  CL spectrum of the clean  $(2 \times 4)$  As-rich surface (a), after 0.3 ML Cs deposition (b), and after annealing the clean As-rich surface to  $550^\circ\text{C}$ , shown to reveal the clean Ga-rich surface (c). The difference spectra a-b and a-c are almost identical, which demonstrates that Cs adsorption induces As desorption from the surface. The right panel shows similar spectra and differences for the Ga  $3d$  CL signal. All spectra are taken at RT.

coverage. Their intensities stay approximately proportional to the Cs exposure. These shapes are similar to those obtained for low coverage at low temperature.<sup>11</sup>

### III. RESULTS

Shown in curve a and curve b of the left panel of Fig. 2 are the As  $3d$  CLs before and after Cs adsorption, respectively. The Cs-induced effect, found from the difference a-b shown in curve d, corresponds to the disappearance of a doubletlike contribution lying at higher binding energy (BE). Also shown in the right panel of Fig. 2, are the corresponding Ga  $3d$  CLs, as well as in Curve i the difference showing the effect of Cs adsorption. This curve shows the decrease of a component at higher binding energy. Also shown in Curve c and Curve h are the CLs of the clean Ga-rich surface, obtained by annealing the clean As-rich one to  $550^\circ\text{C}$ , as well as the annealing-induced changes of the CLs (obtained from the differences between a and c and between f and h, respectively). Note that the annealing-induced change of the As  $3d$  CLs (curve e) is remarkably identical to the Cs-induced one (curve d).

Shown in Fig. 3 are the decompositions of the As  $3d$  and Ga  $3d$  CLs into bulk contributions and surface components (S for arsenic and S' for gallium), using the shapes and chemical shifts of surface components identified in Fig. 2. For Ga  $3d$  (As  $3d$ ) CLs the Lorentzian width was held constant at 0.16 eV (0.18 eV) and the Gaussian width was allowed to vary as a fitting parameter in the 0.40–0.45 eV (0.43–0.54 eV) range, while standard values of the spin-orbit splitting  $SO=0.44$  eV (0.69 eV) and of the branching ratio in the range  $R=1.50$ – $1.55$  were used.<sup>12</sup> The variations of the

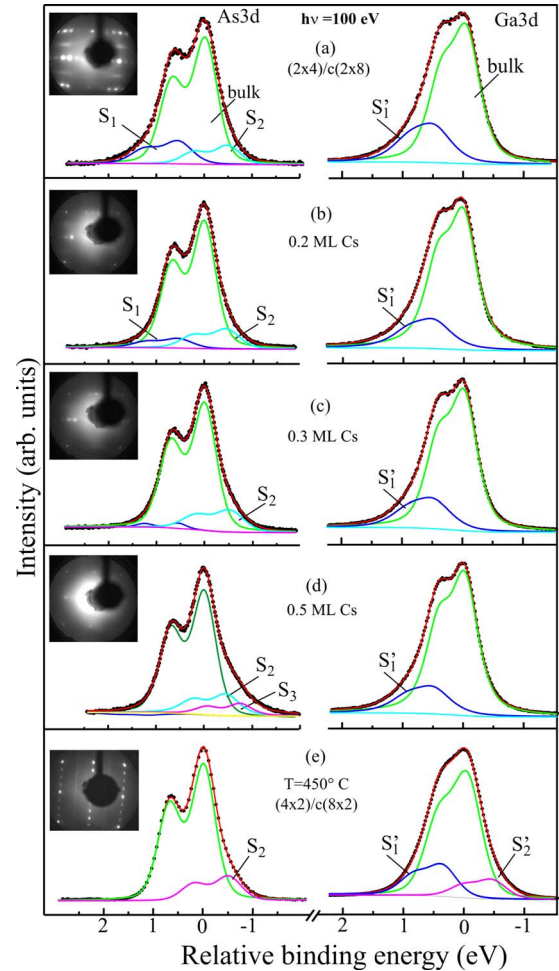


FIG. 3. (Color online) Decomposition of As  $3d$  (left panel) and Ga  $3d$  (right panel) using components identified from the differences in Fig. 2. The spectra are taken for the clean surface (a), after successive Cs exposures, of total coverage (b) 0.2, (c) 0.3, (d) 0.5, and after subsequent anneal to  $450^\circ\text{C}$  (e). Also shown are corresponding LEED patterns.

Gaussian widths and of the chemical shifts of the surface components, with respect to the bulk contribution, are summarized in Table I. For As  $3d$  at  $\Theta_{\text{Cs}} < 0.3$  ML, we have used two surface components. There is first a contribution  $S_1$ , shifted by about 0.60 eV to the higher BE side. Also present is a contribution at a chemical shift of  $-0.44$  eV, labeled  $S_2$ , and already observed before at similar chemical shift.<sup>12–18</sup> For the Ga  $3d$  CLs we used a surface component labeled  $S'_1$ , at a chemical shift of 0.49 eV, reminiscent of the signal in curve i of Fig. 2 and also reported before.<sup>15</sup>

The surface components can be given a simple interpretation. The  $S_1$  contribution is known to be caused by excess arsenic under the form of As dimers which are bonded to As of the second layer.<sup>13,14</sup> For interpretation of the other components, we use for illustration purposes the microscopic description of the  $\beta 2$  unit cell of the clean  $(2 \times 4)$  surface. This description is given by the ball-and-stick model of Fig. 4. As-related surface sites are essentially dimers (two As dimers at the top surface and one As dimer in the third layer). The most numerous gallium-related surface sites are the four  $T'_2$

TABLE I. Core-level binding-energy shifts (in eV), given with respect to the bulk GaAs component. Also shown is the total intensity ratio  $I(\text{As})/I(\text{Ga})$ , taken as unity for the clean GaAs(001)- $(2 \times 4)$  surface, and for the same surface after Cs adsorption and for gallium-rich GaAs(001)- $(4 \times 2)$  prepared by annealing a cesiated As-rich surface (according to Fig. 2).

	Ga 3d				As 3d					I(As)/I(Ga)	
	Shift (eV)		GW (eV)		Shift (eV)			GW (eV)			
	$S'_1$	$S'_2$	$S'_1$	$S'_2$	$S'_3$	$S_2$	$S_3$	$S_1$	$S_2$	$S_3$	
$(2 \times 4)$	0.49		0.43		0.62	-0.46		0.52	0.44		1
0.2 ML Cs	0.49		0.43		0.58	-0.46		0.54	0.46		0.87
0.3 ML Cs	0.49		0.43		0.55	-0.48		0.54	0.47		0.85
0.5 ML Cs	0.49		0.43			-0.49	-0.78		0.44	0.44	0.84
$(4 \times 2)$	0.41	-0.43	0.41	0.42		-0.49			0.43		0.75

sites near the empty dangling bonds of second layer Ga atoms. Also relevant at low coverage is the As-related  $T_3$  trench site, but since there is only one such site per unit cell, its occupation weakly influences the CLs shape at large coverage. The most reasonable interpretation of the  $S_2$  component of the As 3d CLS is in terms of dimerized As atoms because the positive sign of the chemical shift is in agreement with the excess electronic charge due to the occupied dangling bond of the top As atoms which are bonded to Ga of the underlying layer.<sup>13</sup> We attribute the  $S'_1$  component of the Ga 3d CLS to second layer Ga atoms for which the empty dangling bonds generate a shift to lower energy with respect to the bulk configuration.

#### IV. DISCUSSION

Summarized in Fig. 5 are the Cs-induced changes of the surface components, with respect to the intensity of the bulk contribution. Also shown are the changes of the work function under Cs deposition. The disappearance of the  $S_1$  component, due to excess arsenic, can be in principle caused by a Cs-induced chemical shift of the latter component, or by Cs-induced excess arsenic desorption. However, the latter explanation seems more realistic for two main reasons. First, the desorption energy of  $\text{As}_2$  molecules from the As-rich GaAs surface has been estimated to 1.84 eV.<sup>19</sup> In comparison, it is

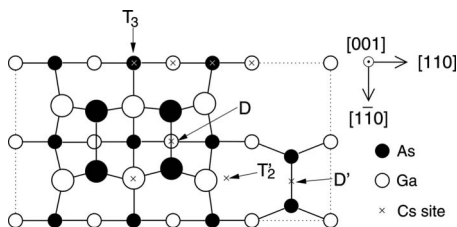


FIG. 4. Ball-and-stick model of the  $\beta_2$  unit cell of the  $(2 \times 4)$  reconstruction of GaAs(001), exhibiting two As dimers in the top layer and one As dimer in the third layer. Also shown are, among possible Cs adsorption sites (marked by crosses), the most energetically favorable ones at low coverage, such as the dimer ( $D$  and  $D'$ ) sites, the  $T_2$  sites near second layer gallium atoms and the  $T_3$  trench site.

known that adsorption of an individual Cs atom at GaAs releases an energy of the order of 3 eV at the initial stage of adsorption and about 1.5 eV at about half of saturation coverage.<sup>20</sup> Such energy can readily induce desorption of the weakly bonded excess arsenic atoms. Second, as seen in Table I, the decrease in the As surface concentration is also confirmed by a decrease of the As/Ga ratio by about 15% at the initial adsorption stage.

Other changes of surface components are different for As 3d and Ga 3d and reveal two distinct regimes, below and above  $\Theta_{\text{Cs}}=0.3$  ML (30 min Cs deposition), respectively. Up to 0.3 ML, the  $S_2$  component of the As 3d CLs stays constant while, for Ga 3d, the  $S'_1$  component decreases by almost 30%. The LEED pattern shows an increase of the background level, revealing some surface disordering but, at this coverage, the  $(2 \times 4)$  structure is still visible. The results of *ab initio* calculations<sup>6,21</sup> for the initial stage of adsorption (one Cs atom per unit cell) show that the Cs atoms can adsorb at both As-related and Ga-related sites. These sites, described in Fig. 4, are the dimer site  $D$  and the  $T_3$  trench site

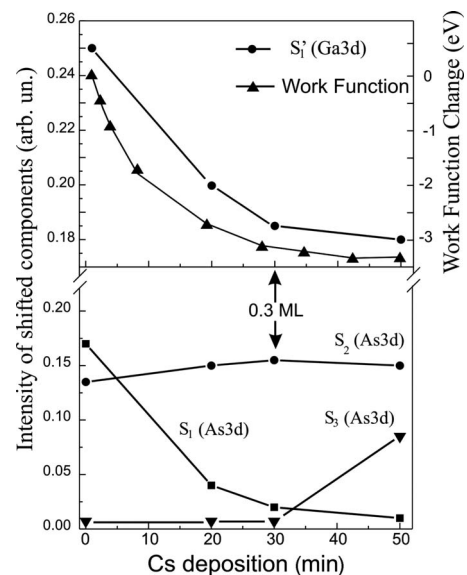


FIG. 5. Variation in the work function and the intensities of the surface components of the As 3d and Ga 3d CLs.



near surface arsenic and the site  $T'_2$  near the empty dangling bond of second layer gallium atoms. Cs adsorption only weakly modifies the surface geometry and the positions of surface atoms so that changes in surface components can only be due to Cs-induced modifications of the electronic environment of the relevant atoms. The constant value of the As dimer-related  $S_2$  component shows that charge transfer of Cs adsorbed at the As dimer site is negligible. Such fact is in agreement with theoretical predictions according to which charge transfer at the As-related dimer sites is smaller than 0.2 electron so that Cs adsorption at the latter sites weakly affects the  $S_2$  component.<sup>6</sup>

In contrast, the decrease of the Ga-related  $S'_1$  component reveals the change of the electronic environment of second layer Ga atoms due to a transfer of charge from Cs atoms adsorbed at the nearby  $T'_2$  site. Such interpretation is supported by three distinct arguments. (i) The latter charge transfer is in agreement with the theoretical predictions, according to which the charge transfer is of the order of  $\delta n_{\text{Ga}} \sim 0.75$  electrons,<sup>6</sup> and with the results of simple tight-binding calculations including the empty Ga dangling bond.<sup>10</sup> (ii) The decrease in the magnitude of the chemical shift of  $S'_1$ , induced by such transfer, is given at low coverage by  $\delta\Delta E = \Delta E_{\text{clean}} + \delta n_{\text{Ga}} \frac{e^2}{r_{\text{Ga}}} \left( \frac{1}{r_{\text{Ga}}} - \frac{1}{d_{\text{Cs-Ga}}} \right)$ , where  $\Delta E_{\text{clean}} = -0.49$  eV is the chemical shift of  $S'_1$  on the clean surface,<sup>4</sup>  $r_{\text{Ga}} = 0.126$  nm is the gallium covalent radius and  $d_{\text{Cs-Ga}} = 0.384$  nm is the bond length.<sup>6</sup> One finds that the two terms of the latter equation cancel each other so that  $\delta\Delta E \approx 0$ . As a result, the  $S'_1$  component should shift toward lower BE up to approximately the position of the bulk contribution, which explains that Cs adsorption induces a decrease of the  $S'_1$  component, without inducing the appearance of a new component. (iii) From the 30% decrease in the  $S'_1$  component obtained for  $\Theta_{\text{Cs}} = 0.3$  ML, we conclude that, in average, 1.3 of the four  $T'_2$  sites of the unit cell are occupied by Cs atoms. Such fact is in qualitative agreement with the estimated coverage of two Cs atoms per unit cell at this coverage<sup>22,23</sup> and with the joint occupation of the Ga dangling-bond sites together with the dimer sites.<sup>6</sup> As seen in Fig. 5, one observes in the same coverage range a decrease of the work function by as much as 3 eV. As shown by *ab initio* calculations, such decrease is partly due to the transfer of charge to Ga dangling bonds, and also to formation of a dipole in the region lying above the As dimers.<sup>6</sup>

For  $\Theta_{\text{Cs}} > 0.3$  ML, unlike the low coverage behavior, the Ga  $3d$  CLs is unmodified. For the As  $3d$  CLs, assuming a single component  $S_2$ , such component would become significantly broader, with a Gaussian width of 0.84 eV. Since there is no clear reason for such broadening, we conclude to the appearance of an extra component, labeled  $S_3$ , thus suggesting that surface arsenic becomes more negatively charged. Such effect could be due to a charge transfer from Cs to surface arsenic, but such explanation contradicts the fact that  $S_3$  appears only above 0.3 ML, whereas calculations show that Cs already adsorbs at dimer sites positions at lower coverages, with a very small charge transfer so that the bonding instead involves hybridization with states of the clean surface.<sup>6</sup>

Thus, the appearance of the  $S_3$  component can be interpreted as due to electron redistribution among the topmost

layers in which the top As atoms become more negatively charged. Such charge redistribution has been demonstrated by  $\text{He}^*$  scattering<sup>24</sup> and shown to be induced by the depolarizing field caused by the increased interaction between Cs atoms. Such effect is also revealed by the saturation of the work function, which, as shown in Fig. 3(a), is strongly correlated with the appearance of  $S_3$ . Depolarization effects also explain the constant value of  $S'_1$ , because of the increased interaction between surface dipoles of nearby Cs atoms results in an impossibility of charge accumulation at the  $T'_2$  site.

Since the bonding level and the dangling bonds of As dimers are occupied, such transfer very likely occurs to the empty antibonding level of As dimers, because of the decrease in the energy of this level in the electric field of Cs dipole layer. The resulting decrease of the dimer bonding energy induces surface destabilization and disordering. In agreement with the disordered nature of Cs on GaAs(001)-(2×4), as studied by scanning tunneling microscopy,<sup>25</sup> the (2×4) LEED pattern completely vanishes in the same coverage range. [In contrast, on the Ga-rich surface, the (4×2) pattern is observed up to 0.7 ML of Cs (Ref. 26).]

The destabilization of the top As layer is also revealed from the decrease of the temperature of the transition from As- to Ga-rich surface. Shown in the bottom panel of Fig. 2 are the LEED pattern and the CLs obtained at RT after annealing the cesiated surface ( $\Theta_{\text{Cs}} = 0.5$  ML) to 450 °C which, as shown in Curve d of Fig. 1, desorbs the large majority of Cs atoms. In agreement with a previous report,<sup>7</sup> such anneal induces a conversion to the *Ga-stabilized* surface, as seen from the clear  $c(8 \times 2)$  LEED pattern. The dominant CLs change lies in the appearance in the Ga  $3d$  CLs of a new component labeled  $S'_2$  at a chemical shift of  $-0.43$  eV. The latter component, characteristic of the Ga-rich surface,<sup>14,27</sup> can be attributed to dimerized gallium atoms in the second layer of the  $\varsigma$  model of the (4×2) unit cell.<sup>28</sup> In summary, the transition of the cesiated As-rich surface to the Ga-rich structure occurs at a temperature lower by 100 °C than the one necessary for the clean surface. Such result, as already observed in Si-H,<sup>29</sup> is found here to occur for  $\Theta_{\text{Cs}} > 0.3$  ML and reveals the Cs-induced weakening of the bonding energy of surface arsenic.

## V. CONCLUSION

Analysis of the Cs-induced modifications of the As  $3d$  and Ga  $3d$  CLS of (2×4)-GaAs(001) reveals: (i) Cs-induced desorption of excess As, (ii) For  $\Theta_{\text{Cs}} < 0.3$  ML, almost complete charge transfer to the second layer Ga sites, and (iii) For  $\Theta_{\text{Cs}} > 0.3$  ML, charge redistribution between top As dimers and underlying Ga atoms, leading to a substantial reduction (by  $\sim 100$  °C) in the temperature of transition from the As-stabilized to Ga-stabilized surface.

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