

Cesium-induced surface conversion: From As-rich to Ga-rich GaAs(001) at reduced temperatures

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We proved experimentally that Cs adsorption on the clean arsenic-rich GaAs(001)-(2×4)/c(2×8) surface followed by annealing at 450 °C–470 °C, induced conversion to the (4×2)/c(8×2) gallium-rich reconstruction. Thus, the conversion temperature is lower by ~100 °C than that required to produce the Ga-rich surface by a conventional ultrahigh vacuum annealing without preliminary Cs adsorption. This effect is monitored using low-energy electron diffraction, reflectance anisotropy spectroscopy, and photoreflectance spectroscopy. We propose that the conversion is due to Cs-induced weakening of arsenic bonds on the surface, which, in its turn, is supposedly caused by electron charge redistribution between Cs adatoms and GaAs surface atoms. Along with the transformation of surface stoichiometry and structure, Cs passivates intrinsic electronic surface states and thus leads to unpinned behavior of the Fermi level at the surface. In combination with the iodine-induced conversion from the Ga-rich to As-rich reconstruction, which was observed earlier, the reverse (As-rich to Ga-rich) low-temperature Cs-induced conversion presented here is promising for the development of atomic-layer etching of GaAs(001).

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I. INTRODUCTION

The physics of low-dimensional structures is essentially based on the developments of semiconductor epitaxial techniques. The advanced modifications of these techniques such as the migration enhanced and atomic layer epitaxies^{1,2} allow one to grow semiconductor structures in which the interface smoothness and the thickness of layers are controlled with the ultimate precision of one monolayer. Along with the atomic-layer growth, for modern nanotechnology it is important to develop techniques of atomic-layer “digital” etching. The atomic-layer etching consists in layer-by-layer removal of a semiconductor with monolayer resolution, keeping an atomically smooth surface morphology. The precision of conventional wet chemical and electrochemical etching techniques is determined by the diffusion and drift lengths of the reagents and products of the reaction and is usually restricted to a few monolayers. In principle, atomic layer etching with monolayer resolution can be realized on the polar faces of compound III-V semiconductors by using adsorbates which selectively react with the elements of III and V groups and, thus, allow selective removal of surface cations and anions. It was shown in Ref. 3 that iodine adsorbed on the Ga-rich GaAs(001) surface was bonded predominantly with gallium atoms. Low-temperature annealing of the iodine-covered surface led to the desorption of gallium iodides GaI_x and conversion to the As-rich surface with a (2×4)/c(2×8) reconstruction. Similar results were obtained for iodine adsorption on various III-V semiconductors.^{3,4}

The selective reactivity of halogens to group III elements of the III-V compounds suggests that, conversely, electropositive alkali metal adatoms may react selectively with group V atoms, weaken their bonds with the substrate and, thus, facilitate a reverse conversion from the anion-rich to cation-

rich surface by the removal of a monolayer of group V atoms at reduced annealing temperatures. The goal of the present paper is to demonstrate this reverse conversion on the Cs/GaAs(001) surface.

Gallium arsenide with adsorbed cesium is often considered as a model system for studying the interaction between metals and semiconductor surfaces and formation of the Schottky barrier, because under adsorption on GaAs, cesium does not form three-dimensional (3D) clusters and chemical compounds with the semiconductor components, does not diffuse into the bulk, and at room temperature forms an ultimately thin adsorption overlayer with a thickness of one monolayer.^{5–12} The major part of earlier studies were performed on the nonpolar GaAs(110) surface^{5–10} which has a simple (1×1) structure. The influence of Cs on the structure and electronic properties of the widely used GaAs(001) surface was studied in a number of papers.^{11–16} However, the data available in literature on the influence of Cs adsorption on various superstructures of this surface are not sufficient to investigate the opportunity of the Cs-facilitated low-temperature conversion from the As-rich (2×4)/c(2×8) to the Ga-rich (4×2)/c(8×2) reconstruction. The most relevant results are that on the GaAs(001) surface the adsorption of Cs followed by annealing at relatively low temperatures may improve the surface atomic order, as suggested in Ref. 11 from the improvement of the low energy electron diffraction (LEED) pattern, and may also reduce the density of surface states.¹⁷ In the present paper the Cs-induced changes in the long-range order studied by LEED are compared to the changes in the local order, stoichiometry, and electronic properties of the surface studied by reflectance anisotropy spectroscopy and photoreflectance spectroscopy. Preliminary results related to the LEED study of the Cs-facilitated structural transition were reported in Ref. 18.

II. EXPERIMENT

The experiments were performed on epitaxial *p*-GaAs(001) layers in order to exclude possible influence of defects, which are present with a larger concentration in the subsurface layer of bulk GaAs, on the stoichiometry and structure of the surface. The preparation of the clean surface included the removal of oxides by the solution of HCl in isopropyl alcohol (HCl-iPA) under dry nitrogen atmosphere, transfer to the ultrahigh vacuum (UHV) without air contact and subsequent annealing in vacuum.¹⁹ Cesium deposition was performed using dispensers thoroughly outgassed in a vacuum not exceeding 10^{-10} mbar. The surface composition and the long-range order in the surface structure were determined in the electron spectrometer ADES-500 by x-ray photoelectron spectroscopy (XPS) and by LEED, respectively. The local order on the surface was studied in an independent UHV system by reflectance anisotropy (RA) spectroscopy, which probes the surface optical transitions.^{20,21} This technique, which is strongly sensitive to surface stoichiometry, consists in measuring the relative reflectivity difference $\Delta R/R = (R_{[\bar{1}\bar{1}0]} - R_{[110]})/R$, where $R_{[\bar{1}\bar{1}0]}$ and $R_{[110]}$ are the reflectivities for light linearly polarized along the $[\bar{1}\bar{1}0]$ and $[110]$ directions of the (001) surface. Details on the UHV experimental setup used for RA spectroscopy can be found in Refs. 22 and 23. The evolution of the band bending, which is determined by the electronic surface states, was studied in a third UHV system, using photoreflectance (PR) spectroscopy.^{15,17,24,25} PR spectra were measured on special UP⁺ structures with thin 100-nm undoped GaAs cap layer grown on highly doped *p*-GaAs buffer layer.²⁴ These spectra contain Franz-Keldysh oscillations (FKO), which allow us to measure with high accuracy ($\sim 2\%$) the surface electric field and, thus, to calculate the band bending φ_s . As compared to the XPS technique,⁵⁻⁸ which allows one to determine the relative changes of φ_s , the absolute values can be determined by the PR spectroscopy. In all three UHV systems, accurate temperature measurements were performed optically, from the band-gap values, and the temperature values of the corresponding annealings did not differ by more than 10 °C.

III. RESULTS AND DISCUSSION

A. LEED study: Long-range order

It was shown earlier that the dose dependences of the amount of adsorbed cesium are similar and well described by Langmuir model with sticking coefficients close to unity for Cs deposition on both As- and Ga-rich GaAs(001) surfaces.¹⁸ Thus, cesium sticking on a GaAs surface does not depend on its stoichiometry and structure. To demonstrate the element-specific selectivity in the interaction of cesium with group III and group V elements of GaAs(001), we experimentally compared by LEED the evolution of the surface long-range order under Cs deposition on the As- and Ga-terminated reconstructions. The results are shown in Fig. 1. Figures 1(a) and 1(f) show LEED patterns of the initial clean surfaces with As-rich GaAs(001)- $(2 \times 4)/c(2 \times 8)$ and Ga-rich GaAs(001)- $(4 \times 2)/c(8 \times 2)$ reconstructions, which were obtained by HCl-iPA treatment and subsequent annealing at T

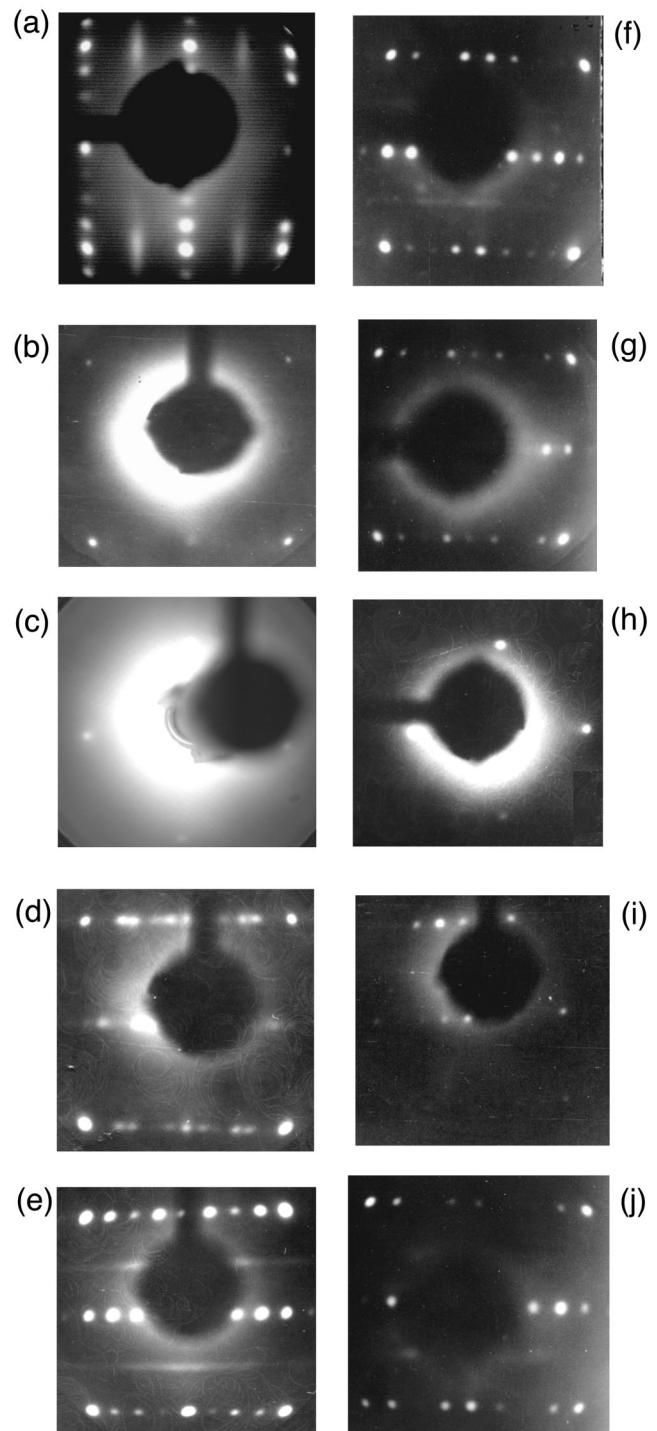


FIG. 1. LEED patterns of the clean As-rich GaAs(001)- $(2 \times 4)/c(2 \times 8)$ (a) and Ga-rich GaAs(001)- $(4 \times 2)/c(8 \times 2)$ (f) surfaces, under deposition of Cs on these surfaces [(b), (c) and (g), (h), respectively], and subsequent anneals after 1 ML Cs deposition [(d), (e) and (i), (j), respectively]. The Cs coverages θ , ML, (b), (g) 0.5 ML, (c) 0.7 ML, (h) 1 ML. The annealing temperatures T , (d) 400 °C, (e) 470 °C, (i) 200 °C, (j) 480 °C.

$=450$ °C and 560 °C, respectively. The deposition of Cs on the As-rich surface leads to a fast degradation of the diffraction pattern, which reveals itself in the increase of background (formed by diffusely scattered electrons) with respect

to the diffraction spot intensities by a factor of ~ 3 at small Cs coverage $\theta \sim 0.1$ ML and in the disappearance of the fractional spots at 0.3–0.5 ML [see Fig. 1(b)]. At higher coverages $\theta \sim 0.7$ ML some of the main diffraction spots disappear as well, while the intensity of the remaining spots becomes several times smaller than the diffuse background [Fig. 1(c)]. The observed degradation evidences the disordering of the surface. On the other hand, on the Ga-rich surface cesium is adsorbed more orderly, the $(4 \times 2)/c(8 \times 2)$ reconstruction is still clearly seen in the LEED pattern at $\theta \sim 0.5$ ML [Fig. 1(g)] and is preserved up to $\theta \sim 0.75$ ML. Under further Cs deposition up to $\theta \sim 1$ ML, fractional spots disappear, the LEED picture is converted to (1×1) pattern, and the diffuse background increases [Fig. 1(h)]. At all coverages the diffraction pattern on the Ga-rich surface with adsorbed cesium is considerably clearer than on the As-rich surface.

The observed degradation of the LEED pattern under Cs deposition and the formation of disordered Cs/GaAs(001) interfaces are in agreement with the results obtained earlier.^{11,14} A fast disordering of the As-rich GaAs(001)- $(2 \times 4)/c(2 \times 8)$ reconstruction under Cs adsorption was directly observed by STM.¹⁴ It was stated in Ref. 11 that the process of Cs-induced degradation is similar for the As- and Ga-rich surfaces. However, the data which allow direct comparison of the surface structure under Cs deposition on these surfaces were not available in literature. It follows from the results obtained in this work that there are significant differences in the action of cesium on the structure of the As- and Ga-rich surfaces. As the sticking coefficients for cesium on these surfaces are the same, the observed differences are due to fundamentally dissimilar mechanisms of adsorption.

The dissimilarity of Cs interaction with different reconstructions of GaAs(001) clearly follows from the experiments on heating cesium-covered surfaces. The results of these experiments are shown in Figs. 1(d), 1(e), 1(i), and 1(j). The most pronounced effect consists in the substantial Cs-induced reduction of the temperature at which the conversion to the Ga-rich reconstruction $(4 \times 2)/c(8 \times 2)$ takes place. This effect is demonstrated in Figs. 1(d) and 1(e). It is seen that deposition of ~ 1 ML of Cs on the As-rich surface at room temperature and annealing at ~ 400 °C results in the (8×1) reconstruction [Fig. 1(d)]. Subsequent annealing at higher temperatures $T = 450$ °C– 470 °C yielded a very clear $(4 \times 2)/c(8 \times 2)$ reconstruction characteristic of the Ga-rich surface [Fig. 1(e)]. These annealing temperatures are by ~ 100 °C lower than those required to obtain the same reconstruction by heating without preliminary Cs deposition.^{19,26} Figures 1(i) and 1(j) show LEED images obtained after deposition of ~ 1 ML of Cs on the Ga-rich surface and subsequent annealing at 200 °C and 480 °C, respectively. Contrary to the behavior of the Cs/GaAs(001)- $(2 \times 4)/c(2 \times 8)$ surface, the Ga-rich $(4 \times 2)/c(8 \times 2)$ structure, which was destroyed by Cs deposition, is restored by heating at temperatures $T \geq 200$ °C.

B. RAS study: Local order

The effect of low-temperature cesium-induced conversion from the As-rich to Ga-rich GaAs(001) surface were studied

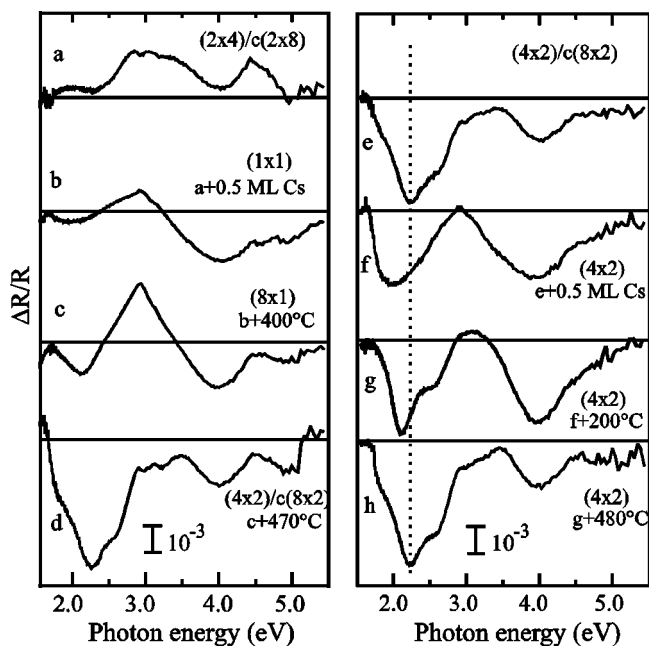


FIG. 2. RA spectra measured at room temperature on the clean As-rich GaAs(001)- $(2 \times 4)/c(2 \times 8)$ (a) and Ga-rich GaAs(001)- $(4 \times 2)/c(8 \times 2)$ (e) surfaces, under deposition of 0.5 ML of Cs on these surfaces [(b) and (f), respectively], and subsequent anneals [(c), (d) and (g), (h), respectively]. The annealing temperatures T , (c) 400 °C, (d) 470 °C, (g) 200 °C, (h) 480 °C.

also by the RA spectroscopy.^{20–22} The peaks in RA spectra were initially interpreted as being due to optical transitions between electronic states associated with gallium and arsenic surface dimers. However, the results of further experiments and theoretical calculations were explained in terms of a mixed bulklike and surface originated anisotropy both for the As- and Ga-terminated reconstructions.^{21,27–29} Irrespective of the exact microscopic origin of RAS peaks, for the present work it is essential that, first, RA spectra are highly sensitive to the surface structure, and there is a direct correspondence between the types of surface superstructures and the detailed shapes of RA spectra.²⁰ Second, in accord with the rule generally applicable to optical spectra of solids,³⁰ the major characteristics of the RA spectra such as the positions, amplitudes, and widths of RA spectral bands are mainly determined by the local electronic structure, and thus, by the local order of the atomic structure, in distinction with LEED which probes the long-range order.

The RA spectra measured on the clean As-rich GaAs(001)- $(2 \times 4)/c(2 \times 8)$ and Ga-rich GaAs(001)- $(4 \times 2)/c(8 \times 2)$ surfaces are shown in Figs. 2(a) and 2(e), respectively. These spectra are characteristic of the respective reconstructions²⁰ and contain two positive spectral bands around 3 eV and 4.5 eV [Fig. 2(a)], and a negative band peaked around 2.2 eV [Fig. 2(e)].^{20,28,29}

In conformity with a rapid Cs-induced destruction of the LEED pattern, the deposition of 0.5 ML of Cs on the As-rich surface radically changed the shape of the RA spectrum [Fig. 2(b)], both positive spectral bands disappeared, so that a negative Cs-induced band centered around 4 eV dominates the spectrum.³¹ Under further deposition of Cs up to ~ 1 ML,

the shape of the spectra (not shown) practically did not change. It is seen from Fig. 1(c) that heating of the Cs-covered $(2 \times 4)/c(2 \times 8)$ surface at 400 °C, which results in the (8×1) reconstruction, led to substantial changes in the spectral shape, the intensity of the negative 4-eV line diminished, a relatively sharp positive peak appeared at 3 eV, and a negative peak around 2 eV started to grow. Further annealing at higher temperature of 470 °C, which yields the conversion of the surface structure to the Ga-rich $(4 \times 2)/c(8 \times 2)$ reconstruction, led to a changeover of the RA spectrum to that characteristic of this reconstruction [see Fig. 2(d)], with the dominating negative band peaked at 2.3 eV and distinct additional shoulders on its low- and high-energy wings.

The right panel of Fig. 2 shows that upon Cs deposition on the Ga-rich surface and subsequent heating, the RA spectrum evolves much more gradually, in conformity with the relative stability of the $(4 \times 2)/c(8 \times 2)$ reconstruction and its reproduction after the heating. More specifically, the deposition of Cs up to 0.5 ML led to gradual broadening and low-energy shift of the dominating negative 2-eV line and to the increase of the Cs-induced negative band at 4 eV [Fig. 2(f)]. Upon further deposition of Cs up to ~ 1 ML the shape remained practically unchanged. Annealing at 200 °C, which removed ~ 15 –20% of cesium³² and partially restored the Ga-rich reconstruction [see Fig. 1(i)], led to narrowing of the negative 2-eV line and to its partial shift back to higher energies [Fig. 2(g)]. Finally, the annealing to 480 °C completely restored both the position and shape of the 2-eV line and diminished the intensity of the Cs-induced 4-eV band [Fig. 2(h)]. Thus, in addition to the effect of low-temperature Cs-induced conversion of the long-range order studied by LEED, the RA spectroscopy demonstrates transformation of the stoichiometry and local order of the GaAs(001) surface.

C. Mechanism of cesium-induced structural transformations on GaAs(001)

To elucidate the reasons for the cesium-induced disordering of the As-rich $(2 \times 4)/c(2 \times 8)$ reconstruction and the decrease in the temperature of the phase transition to the Ga-rich $(4 \times 2)/c(8 \times 2)$ reconstruction, we will compare these effects with the known results on the reverse transition from the cation to anion-terminated reconstructions on III-V semiconductor surfaces induced by adsorption of halogens, in particular, by iodine.^{3,4} The transformations of the surface structure under the action of both cesium and iodine are caused by the change of the surface stoichiometry due to a partial removal of the atoms of the upper layer, the cesium-induced transition is due to the removal of arsenic atoms, and the iodine-induced transition is due to the removal of gallium atoms. In turn, the adatom-induced removal of surface atoms depends primarily on whether the backbonds of these atoms become weaker or stronger upon the adsorption. It is known that the stabilization of the main surface reconstructions of III-V semiconductors and corresponding strengthening of bonds on the surface is attained through the transfer of electrons from the partially occupied dangling bonds of cations to the orbitals of anions, so that the orbitals of the anions

become fully occupied, while those of the cations become empty (“electron counting rule”).³³ Therefore, one can suggest that the destabilization of a reconstruction and the weakening of backbonds occur in the case when adsorption induces the back transfer of electron density from the anions to cations. Within this suggestion, the opposite action of cesium and iodine can be explained by the differences in the electronegativity of these elements. The adsorption of electropositive cesium atoms on the As-rich GaAs(001) surface may cause the return of electron density from arsenic to lower-lying gallium. This should lead to the weakening of arsenic backbonds and, as a result, to the disordering of the As-rich surface and to a decrease in the temperature of the transition to the Ga-rich surface, as was observed in this work. Analogous considerations for the electronegative adsorbate (iodine) lead to the conclusion that the cation backbonds become weaker upon adsorption of iodine on the cation-terminated surface, and this facilitates the conversion to the anion-rich surface upon the subsequent heating.^{3,4}

It should be stressed that the suggested explanation based on the charge transfer from adatoms to a semiconductor is only qualitative and does not take into account significant differences in the character of cesium and iodine interactions with the surface. In particular, it is known that chemically shifted components induced by chemisorption are distinctly visible in the photoemission lines of Ga and As on the I_2 /GaAs surface,³ while chemically shifted components are not observed in the XPS spectra of the Cs/GaAs(001) interface.³⁴

As compared to iodine and cesium, other halogens (F, Cl, and Br) and alkali metals (Na, K, and Rb) with smaller covalent radii lead to more rude effects, in particular, to steady etching of a surface by halogens³⁵ and to clusterization of alkali-metal adatoms^{8,10} due to their higher chemical activity in the interaction with the surfaces of III-V semiconductors. Cesium and iodine act in a more subtle and selective way, without intermixing with the bulk of a semiconductor, and, therefore, provide the possibility of low-temperature adatom-induced reconstruction transitions and atomic layer etching.

D. Cs-induced unpinning behavior of Fermi level on GaAs(001)

To study Cs-induced changes of the electronic surface states we used PR spectroscopy. PR spectra measured on the clean As-rich surface, after deposition of cesium and subsequent annealing are shown in Fig. 3. The increase of the FKO period under Cs deposition evidences the increase of the surface electric field and band bending due to charging of Cs-induced donorlike surface states.^{6,15} The evolution of the band bending under Cs deposition on the As-rich GaAs(001) surface is shown by circles in Fig. 4. After the deposition of ~ 0.5 ML of Cs, the surface was converted to Ga-rich by annealing at $T=500$ °C, and the dependence $\varphi_s(\theta)$ was measured again (triangles in Fig. 4). It is seen that for Cs deposition on the As-rich surface the band bending steeply increases at small coverages, goes through a maximum at $\theta \sim 0.1$ ML, and then saturates. Such “overshoot” behavior was observed earlier on the Cs/GaAs(110) surface^{5,7,8} and

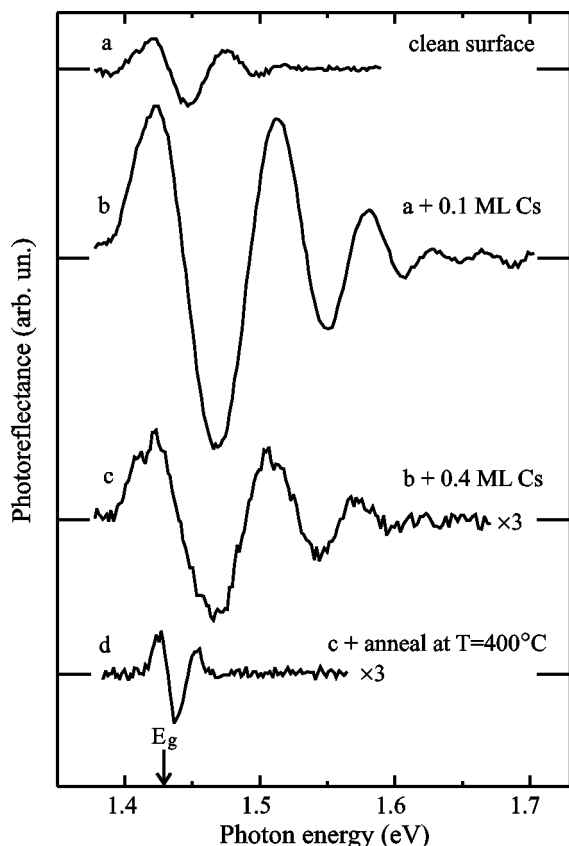


FIG. 3. PR spectra measured on the clean As-rich surface (a), after deposition of Cs [(b) and (c)], and subsequent anneal at $T=400^\circ\text{C}$ (d). The band gap E_g is indicated by the arrow.

can be qualitatively explained in terms of Fermi level pinning on the Cs-induced surface states and further evolution of the energy position of these states due to the dipole-dipole interaction between adatoms.³⁷ For the Ga-rich surface the dependence $\phi_s(\theta)$ is markedly different, the increase is less steep, the maximum is observed at $\theta \sim 0.4$ ML, and an additional shoulder is clearly seen at $\theta \sim 0.1$ ML. This difference reflects the differences in the atomic structure of these interfaces and can be tentatively ascribed to more favorable op-

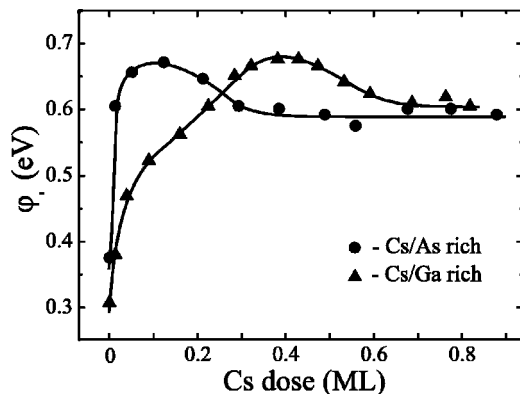


FIG. 4. The evolution of the surface band bending ϕ_s under Cs deposition on the As-rich surface (circles) and on the Ga-rich surface obtained by Cs-facilitated conversion (triangles).

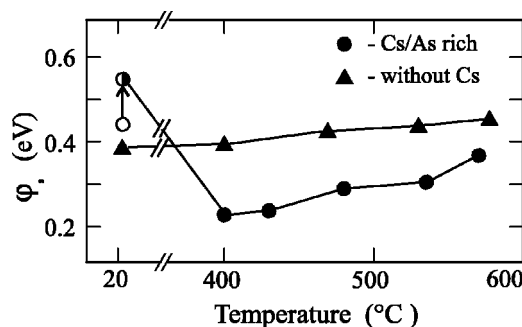


FIG. 5. The evolution of the surface band bending ϕ_s , measured on the HCl-iPA treated surface after annealing at $T=300^\circ\text{C}$ (the open circle), deposition of 0.5 ML of cesium at $T=20^\circ\text{C}$ (vertical arrow which points to the half-filled circle), and subsequent 10-minute anneals at increasing temperatures (filled circles). For comparison, similar evolution but without Cs deposition is shown by triangles.

portunities for Cs adatoms to occupy distinct adsorption sites on a more ordered Ga-rich surface.³⁸

An interesting effect consists in a substantial decrease of ϕ_s after deposition of 0.5 ML of Cs on the As-rich surface and subsequent annealing at $T=400^\circ\text{C}$.¹⁷ This effect is clearly seen from the comparison of FKO period in Figs. 3(a) and 3(d), and also from Fig. 5, where the evolution of the surface band bending under Cs deposition on the As-rich GaAs(001) surface and 10-minute anneals at successively increasing temperatures are shown by circles. Figure 5 shows that deposition of 0.5 ML of Cs led to an increase of the surface band bending indicated by the vertical arrow. Annealing of the Cs-covered surface at 400°C led to a drop of the band bending down to 0.23 eV, which is by ~ 0.2 eV lower than the band bending on the initial As-rich surface. The observed drop of the band bending proves that this anneal removed the defect-induced surface states, which pinned the Fermi level on the clean As-rich surface, and also the Cs-induced surface states localized in the midgap energy region. Under further annealing at higher temperatures the band bending gradually increased (by ~ 0.15 eV after annealing at 580°C), probably due to the increase of the concentration of thermal defects. For comparison, the open triangles in Fig. 5 show the dependence of the band bending on the annealing temperature measured on the clean As-rich surface, without preliminary Cs deposition. It is seen that, in agreement with Ref. 36, the Fermi level did not show the unpinned behavior, the band bending remained constant within ~ 0.1 eV, with a weak tendency for increase towards high temperatures.

The decrease of the midgap surface state density on the Ga-rich surface obtained by Cs-induced low-temperature conversion is in accord with the improved structural quality of this surface, which follows from the detailed analysis of the LEED patterns and RA spectra [compare Figs. 1(e), 1(f), 2(d), and 2(e)]. A possible reason for the observed structural improvement is smaller concentration of near-surface thermal defects due to annealing at lower temperatures.

IV. CONCLUSIONS

Experimental study of the atomic structure and electronic properties of GaAs(001) with adsorbed cesium overlayers by

means of low-energy electron diffraction, reflectance anisotropy spectroscopy and photoreflectance spectroscopy yielded two main results. First, for the As-rich and Ga-rich GaAs(001) surfaces, a qualitatively different behavior of the atomic and electronic structure was observed upon deposition of submonolayer amounts of Cs. The As-rich GaAs(001)-(2×4)/c(2×8) reconstruction is destroyed, and the RA spectrum is radically changed by deposition of small amounts (~0.1–0.3 ML) of Cs, while on the Ga-rich GaAs(001)-(4×2)/c(8×2) surface cesium adsorbs more orderly and causes more gradual evolution of the RA spectra. Second, we found a substantial reduction (by ~100 °C) in the temperature of the reconstruction transition from the As-rich to Ga-rich surface caused by preliminary deposition of cesium. These experimental results are explained by weakening the chemical bonds of arsenic atoms on the surface due to the charge transfer from cesium adatoms to the semiconductor.

The effect of the low-temperature cesium-induced conversion to the Ga-rich surface seems important, because a conventional method of preparing Ga-rich (4×2)/c(8×2) surface requires heating to high temperatures $T=560$ °C–600 °C, which are close to the temperature of incongru-

ent evaporation. This heating may lead to a generation of point defects in the near-surface region, deterioration of the GaAs morphology, and faceting of the surface in the presence of a submonolayer amount of contaminations. Also, along with the known phenomenon of a decrease in the bonding energy of Ga atoms caused by the adsorption of iodine on the Ga-rich GaAs(001) surface,^{3,4} this effect opens up possibilities for the low-temperature ($T \leq 450$ °C) atomic layer etching of polar faces of III-V compounds. The Cs-induced low-temperature conversion observed in this work is accompanied by the distinct improvement of the electronic properties. This improvement consists in Cs-facilitated passivation of the electronic surface states in the midgap region and, consequently, leads to the unpinned behavior of the Fermi level.

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