Correlation between the performance of double-barrier quantum-well infrared photodetectors and their microstructure: On the origin of the photovoltaic effect

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In this work we show clear evidences that silicon segregation out of quantum wells (QWs) is the mechanism responsible for the unexpected photovoltaic (PV) effect exhibited by AlGaAs/AlAs/GaAs double-barrier quantum-well infrared photodetectors. Our results are based on the combined analysis of the detectors' microstructure [obtained by transmission electron microscopy (TEM)] and their electro-optical characteristics (dark current and responsivity versus bias). A TEM image intensity analysis yields the result of an unintentional asymmetry between the two AlAs barriers adjacent to the QW attributed to the presence of segregated Si at the interface. Moreover, we find that the higher this *compositional* asymmetry, the higher the asymmetries in the electro-optical response of the detector. Additionally we show here direct evidences of how the growth-induced nonequivalence of the AlAs tunnel barriers can be ruled out as the origin of the PV effect. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2006990]

I. INTRODUCTION

Quantum-well infrared photodetectors (QWIPs) based on intersubband transitions have emerged as a viable technology for optical imaging arrays. QWIP structures based on the double-barrier quantum-well (DBQW) structure, AlGaAs/AlAs/GaAs, have shown considerable promise for detection within the technologically difficult 3–5-μm atmospheric window. However, this structure is affected by an unexpected and intriguing feature which must be taken into account for its practical operation, i.e., a remarkable photovoltaic (PV) effect observed in the detectors doped in the QW.[1–6] As a consequence, the QWIP detects infrared (IR) radiation at $V_{\mathrm{bias}}=0$ V, with the photoresponse disappearing at a certain applied voltage, $V_c \neq 0$ V. This outstanding PV response has been explained by the internal asymmetries arising during the growth process, where the two main contributions are the existence of structural dissimilarities in the layers[1,2,6,7] and the built-in fields caused by space-charge regions occurring due to silicon segregation towards the surface.[8,9] Although the latter is the most plausible explanation, the origin of the PV behavior is yet far-off to be fully explained. In a previous work, we have observed by transmission electron microscopy (TEM) that the structural properties of the layers (those related to the AlAs barriers, in particular) would seem to play a minor role in the origin of the PV response.[10] Our studies by TEM and high-resolution TEM (HRTEM) confirm this hypothesis and reveal that the existence of structural dissimilarities does not necessarily bring about an outstanding PV response. Moreover, a detailed analysis of the TEM micrographs and the QWIPs' electrical characteristics shows that Si segregation is the main mechanism responsible for the PV effect.

II. SAMPLE DESCRIPTION AND EXPERIMENTAL DETAILS

We examine three samples, which are nominally identical in structure but grown under different conditions. The detectors were grown on GaAs (001) by molecular-beam epitaxy (MBE). The growth procedure is reported elsewhere.[11] The active region comprises 25 periods of 55-Å GaAs QWs sandwiched between two 20-Å-thick AlAs inner barriers and further separated by 300-Å-thick $Al_{0.3}Ga_{0.7}As$ outer barriers. The central 35 Å of the GaAs QWs are symmetrically doped with Si up to $n=2\times10^{18}$ cm^{-3}. Sample A was grown at a substrate temperature $T_s=590$ °C.[11,12] The As beam equivalent pressure (BEP) at the AlAs barriers was $\mathrm{BEP_{As}}=2\times10^{-6}$ torr. Sample B was grown at the same temperature of 590 °C, but the $\mathrm{BEP_{As}}$ in the AlAs barriers was kept at 1×10^{-5} torr. Finally, in sample C the active region was grown at $T_s=550$ °C and under $\mathrm{BEP_{As}}=1.2\times10^{-5}$ torr.[13] In all cases the top and bottom GaAs:Si contact layers were grown at 580 °C. Prior to their

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characterization as photodetectors, the quality of the samples was assessed both by photoluminescence spectroscopy and high-resolution x-ray diffractometry. The QWIPs were processed into mesa photodiodes of 200 μm in diameter with ring-shaped ohmic contacts. The experimental setup for the responsivity (R) and dark current (I_d) measurements is as reported elsewhere.[14] In all measurements, the bottom contact was chosen as the ground.

III. RESULTS AND DISCUSSION

A. Electro-optical characterization

The three QWIPs considered, although grown under different conditions, exhibit a clear photoresponse at zero bias.[10] However, as deduced from the value of the compensating voltage V_c, i.e., the applied voltage that compensates the internal potential drop and quenches the photosignal, it seems that sample C (T_s=550 °C) would be affected by the lower PV effect: $V_{c,25\text{-K sample A}}=1.05$ V, $V_{c,25\text{-K sample B}}=1.1$ V, and $V_{c,25\text{-K sample C}}=0.65$ V. This result could be explained assuming Si segregation. It is well known that the low-temperature growth reduces Si segregation. In this respect, the temperature dependence of the Si incorporation coefficient (K) is described by[9]

$$K(T_s) = 2.4097 \times 10^{-6} \exp[0.7030(\mathrm{eV})/k_B T_s], \qquad (1)$$

where k_B is the Boltzmann constant and T_s the absolute growth temperature. Substituting the data we obtain $K_{\mathrm{A,B}}=0.030\,59$ for samples A and B (T_s=863 K); meanwhile for sample C (T_s=823 K), $K_{\mathrm{C}}=0.048\,22$. This means that Si incorporation is 37% higher (i.e., Si segregation is 37% lower) when reducing the growth temperature from 590 to 550 °C. This value agrees with the change in V_c for the detectors considered, estimated as 38%–40%. Note that, assuming that

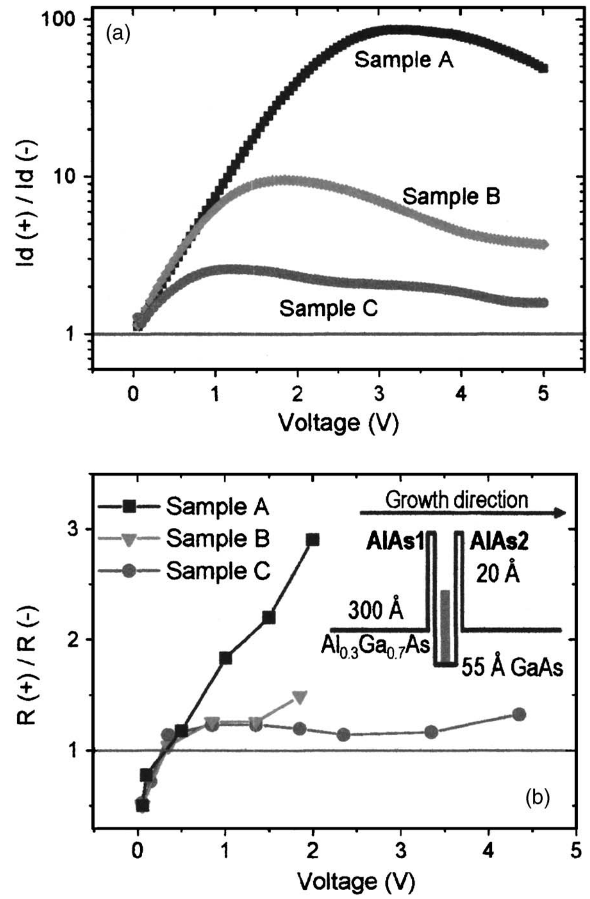

FIG. 1. Asymmetry in the electro-optical characteristics, (a) $I_d(+)/I_d(-)$ vs V at 77 K and (b) $R(+)/R(-)$ vs V at 25 K. In both cases notice the high asymmetry exhibited by sample A and the most symmetrical behavior of sample C.

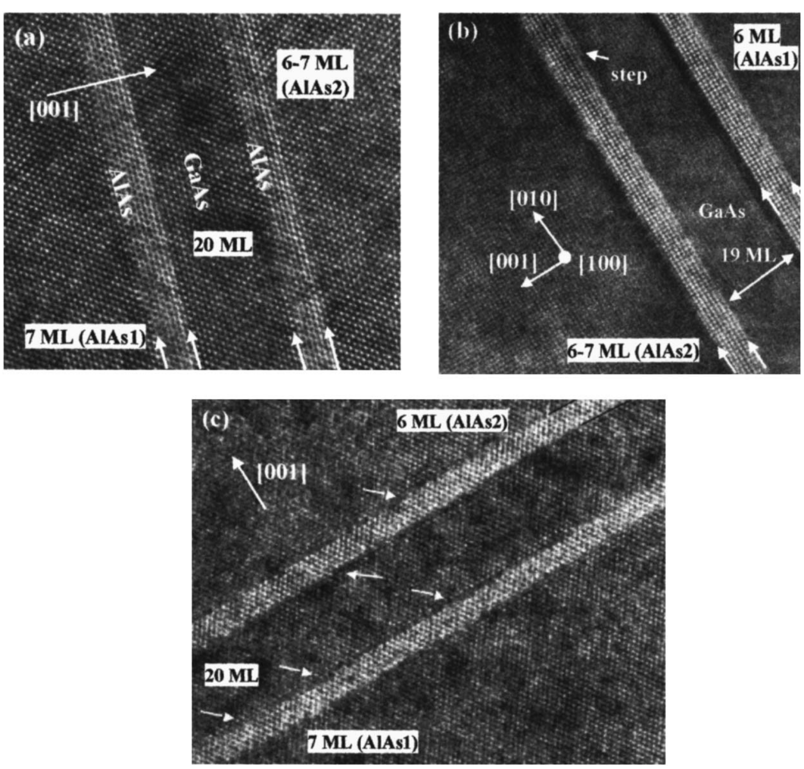

FIG. 2. Cross-sectional HRTEM images. The layer widths are also shown. (a) sample A and (b) sample B. In this case, the image is taken along [010], (c) sample C.

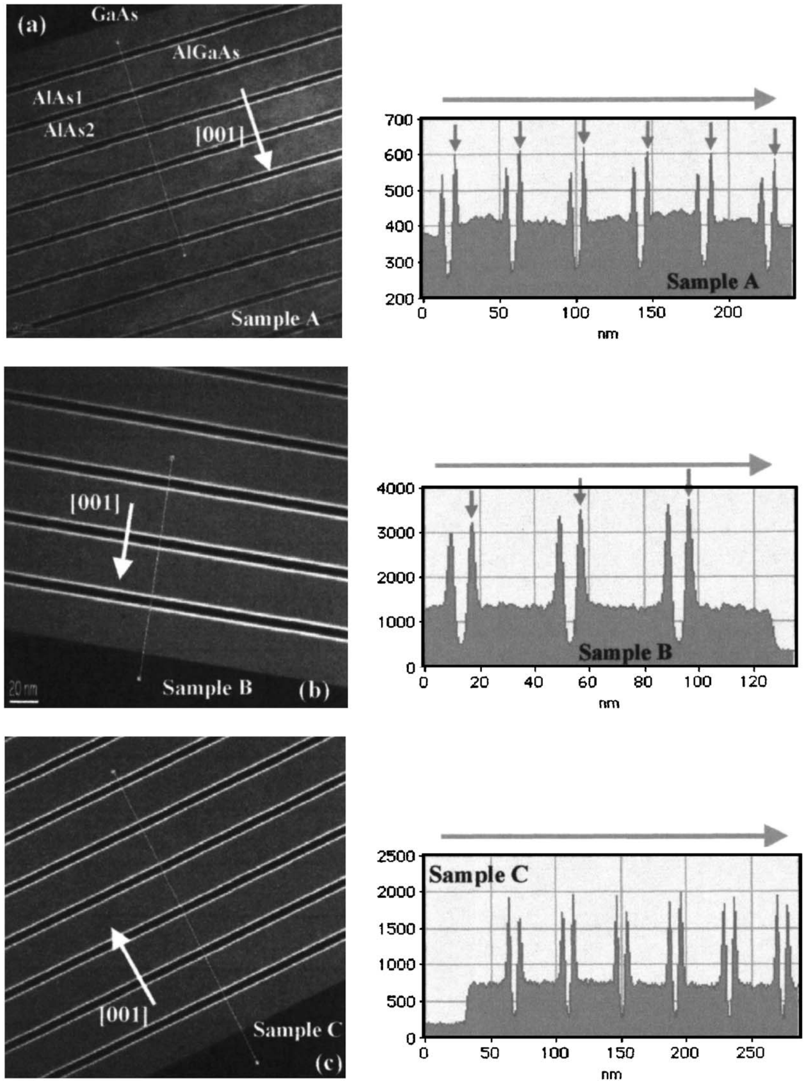

FIG. 3. Dark-field TEM micrographs obtained with **g** =(002). [(a) and (b)] The images' intensity reveals an unintended asymmetry between the two AlAs layers. (c) In sample C (T_s=550 °C) no asymmetry is detected. The arrow indicates the growth direction in each case.

the internal asymmetries and PV effect are caused by Si segregation, V_c may act as an indication of the *amount* of segregated Si. On the other hand, it is possible to compare the magnitude of the internal asymmetry in each detector through the analysis of their electrical characteristics I_d vs V and R vs V.[10,15,16] The asymmetry in the I_d and R data can be seen in Figs. 1(a) and 1(b), where we plotted the measured ratio of the forward to reverse current versus voltage and the ratio of the responsivity for negative and positive bias versus voltage, respectively. Notice that although the three detectors are nominally identical, the *unintended* internal asymmetries are different in each case. In both curves, the highest asymmetry is found in sample A, whereas sample C shows the most symmetrical plot. As before, assuming the asymmetries to be mainly due to Si segregation,[9,15] QWIP C behavior is in considerable agreement with the known fact that the low-temperature growth minimizes the Si segregation.[17]

B. Transmission Electron Microscopy

In order to study specifically the influence of the microstructure on the PV response and internal asymmetries, we have performed a study by cross-sectional TEM and HRTEM of the set of QWIPs. The analysis was carried out in a Jeol JEM 3010 microscope operating at 300 kV. Cross-sectional specimens were conventionally prepared including mechanical thinning and Ar-ion-beam sputtering. Samples A and C were examined along the [110] and [$\bar{1}$10] directions, while sample B was studied along the previous directions and, additionally, along [010]. Figure 2 shows the HRTEM images of the three detectors. Our first result is that, in all cases, the two AlAs barriers adjacent to the QW have the same width [±1 ML (monolayer), within the MBE growth uncertainty], ruling out the proposal of a nonequivalent AlAs layer thickness as the origin of the PV behavior, as suggested in the literature.[1] We find that for samples A and B the variation in the interface roughness between the inverted (GaAs/AlAs) and normal (AlAs/GaAs) interfaces seems to be very small [cf. Figs. 2(a) and 2(b)]; while the inverted interface is atomically abrupt, the normal shows a structural roughness of 1–2 ML. Despite that in these detectors the AlAs barriers have been grown under different BEP_{As}, both samples exhibit approximately the same amount of interface roughness, which do not justify the different "asymmetries" of the I_d and R curves.[10] In contrast, sample C is affected by a noticeable lateral inhomogeneity, which produces fluctuations in the layer thicknesses, especially in the AlAs barriers [Fig. 2(c)].

Additionally in this case, there is a high density of steps at the interfaces, with the GaAs/AlAs interface being rougher than the AlAs/GaAs. Notice that this feature is opposite to the interface properties of detectors A and B, where the rougher interface is AlAs/GaAs. Therefore, we find that the QWIP that exhibits the lowest PV effect and the more symmetric response in the I_d-V and R-V curves (sample C) exhibits the higher structural imperfections. It seems that the existence of any structural dissimilarity on the AlAs barriers (such as width fluctuation, interface roughness, and steps) is not the origin of the remarkable PV effect.

On the other hand, the analysis of many QWIPs' dark-field TEM micrographs under the diffraction condition $\mathbf{g}=002$ reveals some unexpected features that could be attributed to Si segregation at the interface. As shown in Fig. 3, the analysis of the images' intensity profile (in order to reduce the noise, the profiles are obtained with an integration width of 100) reveals a systematic difference in the relative intensity of the corresponding AlAs barriers for samples A and B. In particular, the intensity of the AlAs barrier that is closer to the surface (AlAs2) is *higher* than the corresponding AlAs barrier closer to the substrate (AlAs1). In the case of sample A, the intensity variation reaches the high value $\Delta I_{\mathrm{AlAs}}/I_{\mathrm{AlAs}} \sim 14\%$, while for sample B, $\Delta I_{\mathrm{AlAs}}/I_{\mathrm{AlAs}} \sim 5\%-6\%$. Notice that all the images have been obtained with imaging conditions ($\mathbf{g}=002$) that are sensitive to the alloy composition (Al,Ga)As,[18,19] and because both tunnel barriers are nominally composed of *only* AlAs no difference in the corresponding intensity should be found. Therefore the *unintended compositional* asymmetry that we find experimentally is not simply understandable. One possible explanation is the existence of segregated Si at the interface GaAs/AlAs. In the present case of study, we deal with very thin AlAs barriers (20 Å), so any contribution coming from the interfaces (an area of 2–5-Å thickness) can critically affect the contrast, due to the fact that these dimensions are close to the resolution of the (002) dark-field imaging (5 Å).

Our hypothesis on the presence of segregated Si is supported by the fact that the highest *compositional* asymmetry ($\sim$14%, for sample A) belongs to the QWIP with the highest electrical asymmetries. Moreover, in sample C (where, as mentioned above, the reduction in T_s is supposed to be highly efficient in minimizing Si segregation) the detailed analysis of the TEM images reveals no detectable asymmetry in the intensity of the AlAs barriers adjacent to the QW, as observed in Fig. 3(c). Concerning sample B ($T_s=590\ ^\circ$C, as sample A), the use of a high As flux during the growth of the AlAs barriers would reduce the amount of segregated Si at the interface, which would explain the asymmetry in the TEM images and electro-optical characteristics for this QWIP.[10] Although up to now all the results aim at Si segregation, further work is planned in order to detect the *real* presence of this element and its location.

In summary, we have demonstrated that the main mechanism responsible for the unexpected PV response of the double-barrier QWIPs doped in the QW is Si segregation and that it can be controlled by a careful choice of the MBE growth conditions.

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