



Polarization anisotropy in room temperature contactless electroreflectance spectrum of self-organized lateral compositional superlattices

SANDIP GHOSH[†], B. M. ARORA

SSE Group, Tata Institute of Fundamental Research, Colaba, Mumbai - 400 005, India

SEONG-JIN KIM, HAJIME ASAHI

ISIR, Osaka University, 8-1, Mihogaoka, Ibaraki, Osaka - 567, Japan

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We report the results of room temperature contactless electroreflectance measurements on $(\text{GaP})_2(\text{InP})_2$ superlattices grown on GaAs (001) substrates which show lateral composition modulation along the [110] direction in the (001) plane. The strong polarization anisotropy of some features in the spectrum has been explained by identifying them as arising from transitions in the lateral superlattice formed due to the composition modulation. The measured optical transition energies are considerably lower than theoretical estimates based on the random alloy model, suggesting the need for a better model to describe the electronic band structure of this system.

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1. Introduction

In recent years, self-organized growth as a route for growing high density semiconductor quantum structures for optoelectronic device applications has received considerable attention [1]. In this context, GaP/InP vertical superlattices grown on GaAs (001) substrate are an interesting class of strain-compensated short period superlattices, where, depending on the individual InP and GaP monolayer numbers, there occurs lateral composition modulation resulting in the formation of alternate lamellae of In- and Ga-rich regions parallel to the $[1\bar{1}0]$ direction in the (001) plane [2]. The cause of this composition modulation is still not well understood [1]. Transmission electron microscopy (TEM), energy-dispersed X-ray analysis (EDX) and polarized photoluminescence (PL) results have been put forth as evidence for the occurrence of composition modulation [3–5]. There is considerable interest in establishing the electronic band structure changes due to this composition modulation. Most of the reported results are based on PL measurements [5] which are very sensitive to defects, impurities and inhomogeneities in the sample. Unlike PL, contactless electroreflectance spectroscopy (CER) is largely insensitive to defects and can identify critical points lying at higher energy in the band structure

[†]E-mail: sangho@tifr.res.in

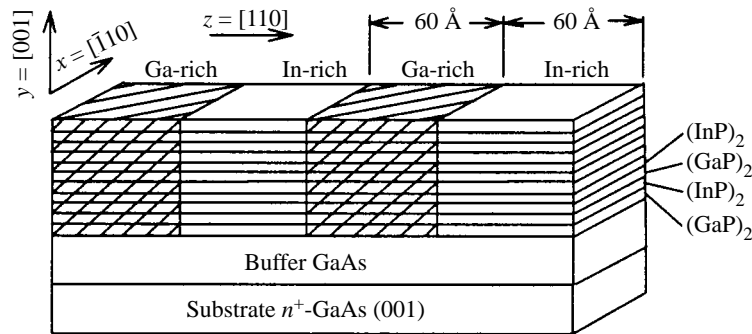


Fig. 1. Schematic of the structure of sample *S#1*. Here $(\text{GaP})_2(\text{InP})_2$ refers to the original vertical superlattice growth sequence which is modified during growth and gives rise to self-organized lateral composition modulation in the form of In-rich and Ga-rich regions.

even at room temperature [6]. In this paper we report polarization-dependent CER measurements on these samples, explain the origin of the strong polarization dependence of some of the features in the spectrum and give useful information about the electronic band structure of these samples.

2. Experiment

The two samples used in this study were grown by gas source molecular beam epitaxy. The first sample *S#1*, shown schematically in Fig. 1, consists of a $\approx 0.3 \mu$ layer of $(\text{GaP})_2(\text{InP})_2$ short period vertical superlattice grown on an n^+ -GaAs (001) substrate with a nonintentionally doped GaAs buffer in between. The details of the growth and structural characterization have been reported earlier [7]. Briefly, they are as follows. TEM pictures of the sample surface reveal alternating wire-like bright and dark patches oriented along the $[\bar{1}10]$ direction with a length typically around $0.3 \mu\text{m}$ and an average width of $\approx 60 \text{ \AA}$. The cross-sectional TEM pictures of the $(\bar{1}10)$ plane indicate that these patches extend vertically and nearly uniformly in the [001] direction all through the $0.3 \mu\text{m}$ thickness of the sample. The PL peak position of this sample ($\approx 1.68 \text{ eV}$ at 80 K) and its polarization characteristics (in emission) are identical to those reported by Pearah *et al.* [5]. EDX measurements performed on similar samples have shown that the bright regions in these structures are In-rich with the lower limit of In concentration being 59%, while the dark region are In-deficient with an upper limit of 41%. The second sample *S#2* was also a $(\text{GaP})_2(\text{InP})_2$ vertical superlattice but unlike *S#1*, was grown on a GaAs substrate with (411)A orientation. As such composition modulation is known to be sensitive to substrate orientation and structural characterization of the sample *S#2* revealed no regular composition modulation of the type seen in sample *S#1*. The details of the CER measurement technique have been reported elsewhere in the literature [8, 9]. In this study a polymer film polarizer was used to obtain the polarized probe beam.

3. Results and discussion

Figure 2 A shows the CER spectrum of the sample *S#1* at room temperature. In plot (i) the polarization of the incident probe beam was parallel to $[\bar{1}10]$ direction. The structures in this spectrum between 1.4 eV and 1.52 eV are Franz-Keldysh oscillations (FKO) arising from the buffer/substrate GaAs layers. Armelles *et al.* [10] have studied $(\text{GaP})_n(\text{InP})_m$ strained short period vertical superlattices previously using photoreflectance at 80 K and have identified the main feature at energies higher than the GaAs band gap as being the signature of these superlattices. In our spectrum we see not one, but two widely separated features at energies higher than the bandgap of GaAs. We shall refer to the feature between 1.55 eV and 1.66 eV as A1 and that between 1.72 eV and 1.83 eV as B. In plot (ii) the polarization of the incident probe beam was parallel to [110]. In this

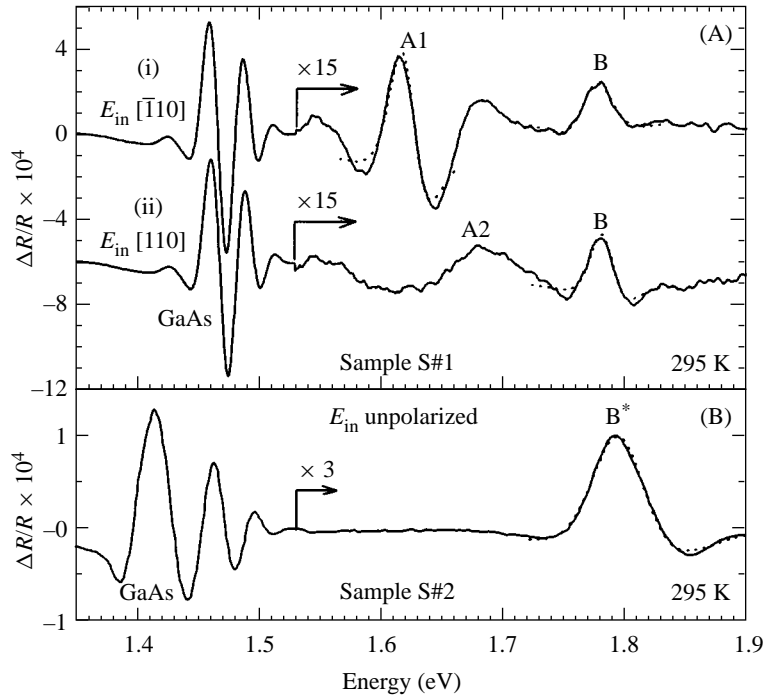


Fig. 2. (A) Contactless electroreflectance (CER) spectra of the sample *S#1*, grown on GaAs (001) substrate, for two polarizations of the incident probe beam. The plots are vertically shifted for clarity. (B) CER spectrum, with an unpolarized probe beam, of sample *S#2*, on a GaAs (411)A substrate which does not show lateral composition modulation of the type seen in sample *S#1*. The dotted lines represent the fitted lineshape.

spectrum the GaAs FKO and the feature B show negligible change. However, the feature A1, seen earlier with the $[\bar{1}10]$ polarized light between 1.55 eV and 1.66 eV, is no longer present and in its place there is a weak and much broader feature which we shall refer to as A2. This dramatic change in the spectrum is unusual because the two directions $[110]$ and $[\bar{1}10]$ are identical in the zinc blende structure. Note that there is a hump in the spectrum shown in plot (i), which occurs at the same energy where the feature A2 occurs in plot (ii). This indicates that the transition giving rise to the feature A2 is allowed for both polarizations of the incident probe light. To estimate the transition energies we fitted to these spectral features the following lineshape function:

$$\Delta R/R(E) = \sum_{j=1}^n \text{Re}[a_j \exp(i\theta_j)/(E - E_{0j} + i\Gamma_j)^m], i = \sqrt{-1} \quad (1)$$

where E_{0j} , Γ_j , θ_j and a_j are the transition energy, broadening parameter, phase factor and amplitude respectively, associated with the j -th transition. The above equation with $m = 3$, which mimics the case of a first derivative gaussian broadened lineshape function [6], fits our data best indicating inhomogeneous broadening of the critical point energies in these samples. The fitted curves are shown by the dotted lines in Figure 2. The values of E_0 and Γ for the main transition A1 were found to be 1.621 eV and 29 meV while the average values of those for feature B were found to be 1.78 eV and 30 meV, respectively. Although the presence of the feature A2 for light polarized along $[110]$ is unambiguous, its weak strength as compared with the background causes the parameter values obtained by lineshape fitting to be unreliable. Figure 2B shows the room temperature CER spectrum of sample *S#2* using an unpolarized probe beam. In this spectrum at energies higher than the

GaAs FKOs we see only one feature labelled B^* . This feature was found to be polarization-insensitive and its energy position was found to be 1.799 eV from lineshape fitting.

In sample *S#2*, which does not show composition modulation, the feature B^* is the signature of the vertical superlattice. This sample was grown on a (411)A substrate which makes an angle of 19.5° with respect to the (100) plane which is equivalent to the (001) plane on which sample *S#1* was grown. Thus, although the strain that the $(\text{GaP})_2(\text{InP})_2$ vertical superlattice would experience in sample *S#1* would be slightly different from that in sample *S#2*, it is not expected to drastically change the energy position of the fundamental gap of the $(\text{GaP})_2(\text{InP})_2$ superlattice. We therefore identify the feature B in sample *S#1*, which occurs at nearly the same energy as feature B^* in sample *S#2*, as the signature of the remnant $(\text{GaP})_2(\text{InP})_2$ vertical superlattice. This indicates the presence of two phases, one where composition modulation has taken place and the other where the original vertical superlattice still exists. In the present context the major difference between sample *S#1* and *S#2* is the absence of lateral composition modulation in the latter. The fact that the features $A1$ and $A2$ seen in sample *S#1* are absent in *S#2* indicates that these two features are associated with the lateral composition modulation seen in this sample. Thus, lateral composition modulation has resulted in a structure whose fundamental gap is lower than that of the original $(\text{GaP})_2(\text{InP})_2$ vertical superlattice by $\simeq 160$ meV at room temperature.

As such, the polarization dependence of the optical properties of III-V semiconductors with zinc blende crystal structure is related to the symmetry of the wavefunctions at the top of the valence band. In particular, the heavy hole wavefunction, unlike light holes, does not have a \hat{z} component so light polarized parallel to \hat{z} cannot excite a heavy hole transition but can excite a light hole transition [11]. Under normal conditions these states degenerate at the valence band edge and there is no preferred \hat{z} direction, therefore no optical anisotropy is observed. However, this degeneracy can be lifted in a number of ways. In the case of $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ quantum wells grown on (001) GaAs substrates, anisotropy in the electroreflectance (ER) signals between [110] and $[\bar{1}10]$ polarizations of the probe light has been induced by the application of large electric fields in the (001) direction [12]. However, the difference between the ER signals in the above case for the two polarizations of the incident light is nearly an order of magnitude smaller than the ER signal itself and that the difference exists only for the weak signal related to light holes. Therefore, an argument based on pre-existing surface electric fields in our samples cannot be invoked to explain the large anisotropy observed. Modification of the band structure due to strains arising from the presence of misfit dislocations can also induce polarization anisotropy [13]. This phenomenon does not give rise to new transitions but results in anisotropy at energies corresponding to existing critical point energies. Thus, if such a mechanism were applicable in our case, we would have seen features similar to $A1$ and $A2$ even in sample *S#2*, which we did not. In fact, although a multicomponent strain tensor arising from the composition modulation is expected to be present in our sample, it cannot by itself account for the complete vanishing of the feature $A1$ for the [110] polarization of the incident light. Degeneracy can also be lifted by CuPt ordering, as is found in the case of $\text{In}_x\text{Ga}_{1-x}\text{P}$ alloys. However, in such alloys, the main feature in the electroreflectance spectrum [14] associated with the ordered alloy is stronger for the [110] direction than for the $[\bar{1}10]$ direction, which is the exact opposite of what we observe. Furthermore, unlike our observations, the principal ordered-alloy related feature does not vanish completely for any polarization of the probe beam.

Thus, none of the above phenomenon can be invoked to explain the strong polarization anisotropy observed in our spectra. To understand the polarization dependence of the features $A1$ and $A2$ in the CER spectra of our samples consider the following. In the case of superlattices and quantum wells, quantum confinement splits the heavy hole and light hole states, and confinement direction defines the \hat{z} direction [11]. So if light is incident in a way such that it can be polarized both in the plane of the well and perpendicular to it then the relative intensities of various lines in the spectrum can change depending on the polarization [15]. In our case, in sample *S#1*, composition modulation results in the presence of alternate In- and Ga-rich layers oriented along [110] in the (001) plane forming a lateral superlattice (LS) along [110] with the In-rich regions as the wells and the Ga-rich regions as the barriers. The LS potential also defines the preferred \hat{z} direction as

[110]. Therefore, a heavy hole-related transition in the LS would not be excited for light polarization along [110], which lies in a plane perpendicular to the LS planes, while it would be excited for light polarized along $\bar{1}10$, which lies in the LS plane. This is exactly how the feature A1 behaves, we therefore identify it as arising from the lowest heavy hole-related transition in the LS. The weak feature A2 may be associated with a light hole-related transition in the LS as it is present even when the incident polarization is along [110], i.e. the \hat{z} direction. We do not see a signature of the LS barrier probably because in a superlattice, the barrier band gap does not correspond to a particularly large change in the joint density of states [11, 6]. The earlier identification of the feature B (in *S#1*) and B* (in *S#2*) as being the signatures of the $(\text{GaP})_2(\text{InP})_2$ vertical superlattice also explains their relative polarization insensitivity, because for this structure any polarization of a normally incident beam would always lie in the vertical superlattice plane.

In order to theoretically estimate the optical transition energies in the lateral superlattice Pearah *et al.* [5] originally used a model which considered the In-rich and the Ga-rich regions to be disordered $\text{In}_x\text{Ga}_{1-x}\text{P}$ alloys coherently strained with respect to the GaAs substrate. However, they found that by using an In concentration of 59% (lower bound on In concentration from EDX measurements) in the In-rich regions and 41% (upper bound) in the Ga-rich regions they could not match the lowest calculated transition energy with the PL peak at ~ 1.68 eV at 77 K (same as that seen in our sample *S#1*). To explain this discrepancy, it was suggested that the PL spectrum was dominated by emission from low band gap regions/defects resulting from inhomogeneities in the composition modulation. More recent calculations of Rich *et al.* [16], using a similar model, suggest that the strained In-rich well region of the LS has a band gap of 1.802 eV at room temperature so that the optical transition energy (effective band gap) is expected to be higher still when confinement energy is accounted for. Thus the 1.62 eV transition energy observed by us cannot be explained using the above model. As CER is largely insensitive to defects and measures band gaps accurately, our results suggest that an improvement in the theoretical model is required to describe the band structure of this material.

In the above context the possibilities are: (i) a much higher In composition contrast (79.5% /20.5%) [5] between the well and the barrier regions, which, however, seems unlikely considering the results of EDX measurements. (ii) Instead of considering the In-rich and the Ga-rich regions as completely disordered $\text{In}_x\text{Ga}_{1-x}\text{P}$ alloys, consider the vertical superlattice structure remaining essentially intact but with redistribution of Ga and In in the group III planes, forming a lateral superstructure of alternate Ga-rich $[(\text{GaP})_{2+\delta}(\text{InP})_{2-\delta}]$, and In-rich $[(\text{GaP})_{2-\delta}(\text{InP})_{2+\delta}]$ regions. The latter would be structurally closer to reality as observed in high resolution cross-sectional TEM pictures of the $\bar{1}10$ plane which show that the original vertical superlattice structure essentially remains intact with the InP layers being thicker in one region and the GaP layers being thicker in the adjacent regions [1, 17]. Moreover, the vertical order can provide for the lowering of the energy scale required to match the photoluminescence and electroreflectance measurements as the band gap of the $(\text{GaP})_2(\text{InP})_2$ vertical superlattice as measured by us (1.78 eV) is lower than that of the disordered $\text{In}_{0.49}\text{Ga}_{0.51}\text{P}$ alloy [5] by ~ 140 meV.

In conclusion, we have identified the features in the CER spectrum of $(\text{GaP})_2(\text{InP})_2$ superlattices on GaAs (001) substrates which show self-organized lateral composition modulation, explained their polarization dependence and determined the room temperature critical point energies. Our experimental results suggest that an improved model is required to describe the electronic band structure of these materials.

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