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Coherency of Interfacial Roughness in GaAs/AlAs Superlattices

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The presence of a *nonrandom* fractional number of atomic planes in each layer of a superlattice produces unusual diffraction patterns in which the peaks cannot be indexed in the usual fashion as due to a single series. The x-ray line broadenings are distinctly different from earlier measurements and calculations in which the interfacial roughness is due to random variations of the scattering function. Therefore, interfacial roughness encountered at a single interface may just be a consequence of controlled, but not random, roughness and that under proper growth conditions superlattices with atomically sharp interfaces may be produced. These results are in good agreement with experimental measurements.

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The structure and chemical composition of interfaces plays an important role in the physical properties of single and multilayered films.[1-4] In vapor-deposited multilayers, interfacial disorder may arise from a variety of factors including substrate roughness, rate fluctuations, island growth mode, interdiffusion, etc. In order to understand changes in the x-ray linewidths of GaAs/AlAs superlattices, the presence of *random* interfacial roughness was introduced[2-5] and a recent electron microscopy measurement has claimed the presence of substantial random interfacial roughness at these same interfaces.[6] This random roughness broadens diffraction lines in a well understood fashion.[2-5] In this paper, by introducing the concept of "*controlled* interfacial roughness," we are able to reconcile precisely controlled deposition rates with the roughness observed at one single interface. This type of peculiar roughness arises from the fact that with an accurate rate control, a noninteger number of atomic planes may be deposited in each layer. In this fashion, an unusual superlattice diffraction pattern arises with *sharp* diffraction lines, which can be related to the controlled interfacial roughness. This type of roughness cannot be uniquely established from local measurements which are performed at a single interface but rely on the long-range structural coherence of the superlattice.

In the interpretation of x-ray diffraction from superlattices, the diffraction peaks arise from a convolution of the diffraction by the multilayer stack with the finite-size-limited diffraction from a single layer of the constituent materials A and B.[5] The position of the allowed peaks is given by

$$q = 2\pi n/\bar{d} \pm 2\pi m/\Lambda , \tag{1}$$

where n and m are integers, Λ is the modulation wavelength, and $\bar{d}$ is the average lattice spacing defined by

$$\bar{d} = \Lambda/N = (d_A n_A + d_B n_B)/(n_A + n_B) , \tag{2}$$

where d_A, d_B and n_A, n_B are the lattice constants and number of atomic planes of materials A and B, respectively, and $N = n_A + n_B$ is the total number of atomic planes in a modulation wavelength. If N is an integer (as might seem necessary at a first glance), Eq. (1) can be rewritten as

$$q = 2\pi m'/\Lambda , \tag{3}$$

with $m' = \pm m + Nn$ an integer. In this approximation, the low-angle peaks (i.e., $n = 0$) merge perfectly with the high-angle peaks ($n = 1, 2, \ldots$), in a single well-defined series. It is generally assumed that interfacial roughness arises from fluctuations in the deposition process or island growth mode, thus giving rise to a variable N from layer to layer.[7-12]

Two types of deviations from a perfect crystalline structure can be envisioned: *random* and *controlled* roughness. A variety of random-type roughness models are available in the literature, including random continuous or discrete roughness in the layer thickness, random lateral roughness, etc.[5,7-12] All these models have the common feature that some property (layer thickness, interatomic spacing, etc.) varies in a random fashion. As a consequence of the randomness (because the randomness is cumulative),[10] the effect in all cases is to broaden the diffraction lines and to leave their position unaltered. Figure 1(a) shows the variation of the GaAs fraction in an interfacial plane as a function of interfacial plane

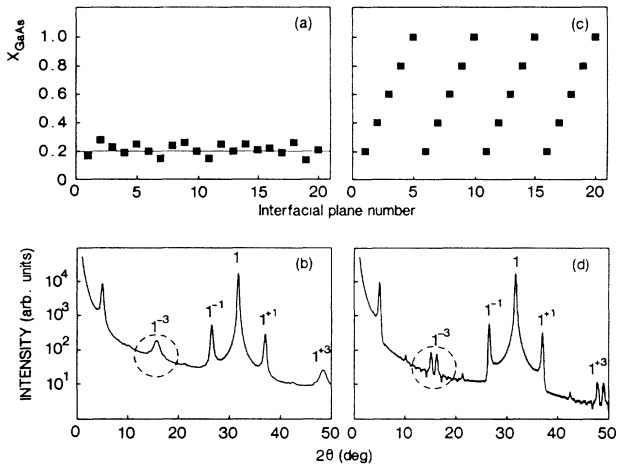

FIG. 1. (a) Variation of GaAs fraction in an interfacial layer (x_{GaAs}) as a function of interfacial plane number across a GaAs/AlAs superlattice with $\bar{N}=6.2$, and assuming *random* interfacial roughness. (b) Calculated x-ray intensity for the model in (a). Note that the third-order satellite 1^{-3} is considerably broader than the first-order one 1^{-1}. (c) Same as (a) but assuming *controlled* interfacial roughness. (d) Calculated intensities for the model in (c). Note that all peak widths are the same and that the peak 1^{-3} has an additional peak next to it.

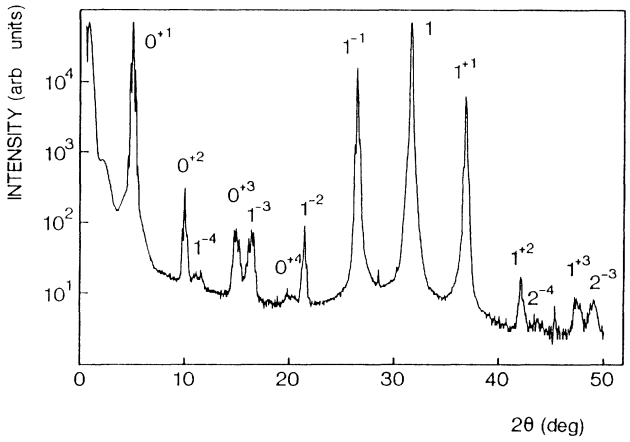

FIG. 2. Experimental x-ray diffraction from a GaAs(~9.15 Å)/AlAs(~9.15 Å) superlattice. The peaks are labeled $n^{\pm m}$ according to two series centered around the (000) and (002) ($n=0,1$) GaAs reflections and separated by $2\pi/\Lambda$ ($\Lambda\sim 18.3$ Å).

number across a GaAs/AlAs superlattice with an average $\bar{N}=6.2$. The GaAs fraction varies *randomly* from interfacial plane to interfacial plane, around a mean value of 0.2. As a consequence, the satellite linewidths increase with increasing order as shown in Fig. 1(b) (for instance, the linewidth of the peak labeled 1^{-3} is much broader than that of the central peak 1).

The type of interfacial roughness introduced in the present model is quite different in character. The amount of material deposited in each layer is well *controlled*, but does not correspond to an integer number of atomic planes. As a consequence, the interfacial layer consists of a mixture or alloy of GaAs and AlAs with the interfacial layer changing composition in a well-defined manner as shown in Fig. 1(c) for a layer thickness corresponding to an average $\bar{N}=6.2$, as in the previous random roughness model. This gives rise to a peculiar type of roughness which "advances" as the number of layers are increased (i.e., the proportion of material A in the interfacial atomic plane increases from layer to layer, in a well-defined manner). As a consequence, the linewidths do not increase with increasing satellite order and consequently the peak around $2\theta\sim 15°$ is split, with a separation given by the nonintegral fraction [see Fig. 1(d)].

The GaAs/AlAs superlattices used in this study were grown in a Varian GEN II molecular-beam-epitaxy system on (100)-oriented semi-insulating GaAs substrates. The superlattices were approximately 1 μm thick, grown on a 0.5-μm GaAs buffer, and terminated with GaAs. The growth rate for the GaAs was 1 μm/h, with a III/V flux ratio of 1:8. The AlAs was also grown at a rate of 1 μm/h with a III/V ratio of 1:20. The growth temperature of 680 °C was determined as an offset from the congruent sublimation point, as observed with reflection high-energy electron diffraction. The substrate was rotated at 25 rpm during growth.

The growth timing was controlled using an IBM-XT computer. The control software, written in-house, was designed to maintain the accuracy of the shutter timing to within 0.054 s. With the shutter hardware responding in less than 0.1 s, the shutter control has an absolute accuracy of approximately 0.15 s. This translates into ~0.15 monolayer at a growth rate of 1 μm/h. The relative precision of the layer thicknesses is significantly better than ~0.15 monolayer reported because much of the shutter actuation time is a systematic delay. The jitter in the shutter timing is substantially less than 0.1 s. Diffraction measurements were performed on a 12-kW rotating anode, Rigaku DMax II, diffractometer equipped with a flat pyrolitic Ge monochromator and Cu $K\alpha$ radiation of 1.54 Å wavelength. Extreme care was taken to assure the sample alignment using an in-house-built laser alignment system.

The experimental x-ray-diffraction spectrum obtained from a GaAs(~9.15 Å)/AlAs(~9.15 Å) superlattice is shown in Fig. 2. Note that the individual thicknesses of each layer are different from an integral number of atomic planes [$d_{(001)}$(GaAs)~$d_{(001)}$(AlAs)~2.82 Å]. As described above and predicted by Eq. (1), the spectrum consists of two series of peaks labeled in the figure as $n^{\pm m}$, where n and m indicate the order of the main reflection (n) and its satellite ($\pm m$). The separation between diffraction peaks within a given series yields $\Lambda=18.3$ Å with $N=6.5\pm 0.05$, the latter being definitely noninteger. It is important to notice in Fig. 2 that peaks 0^{+3} and 1^{-3} are located close together and

their separation is *not* given by $2\pi/\Lambda$, making it evidently clear that the two sets of peaks cannot be indexed on a single series. We should also point out that such a peak was observed earlier in Mo/Fe superlattices; however, its origin was not identified.[13] Moreover, the widths of the peaks are constant, independent of satellite order, and given by the instrumental resolution. We should also point out that the fine structure observed on the satellite peaks is real and reproducible and can be understood within the present model.

The structure factor for a sample with controlled roughness is given by

$$F(q)=\sum_{j=1}^{M} f_j(q)e^{iqjd}, \tag{4}$$

with M,j integers and $f_j=f_A$ or $f_j=f_B$ in the bulk of layers A and B, respectively, and at the interface $f_j=pf_A+(p-1)f_B$ with p the fraction of the layer occupied by A-type atoms [see Fig. 1(c)]. The results of such a calculation for a GaAs(3+w layers)/AlAs(3 layers) superlattice is shown in Fig. 3 for $w=0$, 0.2, 0.4, 0.6, 0.8, and 1.0 in going from Figs. 3(a) to 3(f). Figures 3(a) and 3(f) correspond to the commensurate superlattices with $N=6$ and 7, respectively; as such the peaks could be indexed on a single series according to Eq. (3). Furthermore, in Fig. 3(a), because the constituent layers have the same thickness, the even m' peaks are absent, except for the $m'=6$ peak ("central" peak) which is due to the expected AlAs(002) reflection. The calculations assuming *controlled* roughness with a fractional number of atomic planes [Figs. 3(b)–3(e)] correspond closely to the experimental observations (Fig. 2). There is a series of peaks separated by $2\pi/\Lambda$ centered around the (000) peak, labeled $n=0$ in our notation, and another series centered around the (002) peak, labeled $n=1$. Note that the position of this latter peak does not change as the fraction w is varied, and that it is not possible to label it as a higher-order reflection of the superlattice according to Eq. (3). Most importantly, unlike in the case of random roughness [Figs. 1(a) and 1(b)] the linewidths do not broaden and are only given by the instrumental resolution. *Moreover, unlike in all previous random cases, the roughness is characterized also by changes in the position of the peaks and not merely by intensities and linewidth changes.*

The effects described here were possibly not reported earlier, because their presence is not manifest in such a clear-cut way in thicker superlattices, which are the ones ordinarily studied. Moreover, extreme care should be taken in controlling shutter actuation and growth timing so that the layer thickness is precisely controlled.

The existence of fractional interfacial layers may have significant bearing on other physical properties: E.g., the luminescence from quantum wells will show broadened peaks corresponding to the superposition of two different thickness layers,[14] and in short-period superlattices, which exhibit extremely different subband structures depending on the number of atomic planes in each layer,[15] the optical properties could be affected dramatically. The experiments presented here imply that it is possible to vary the fractional layer thickness in a reproducible manner with the consequent control on the physical properties.

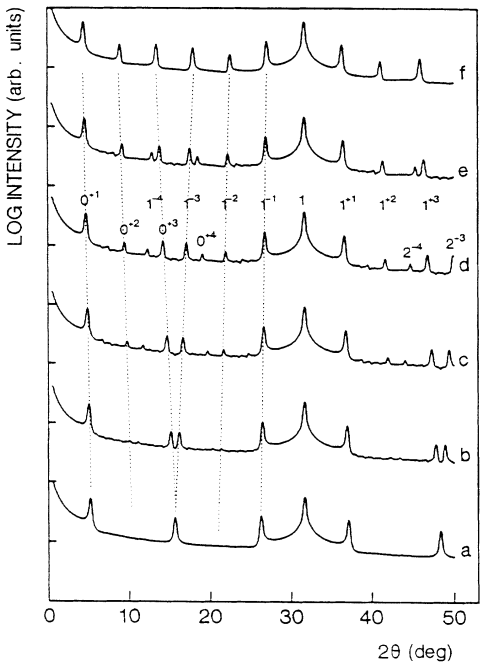

FIG. 3. Calculated x-ray diffraction from GaAs/AlAs superlattices with various periodicities: (a) 16.92 Å $=6.0d_{(200)}$; (b) 17.48 Å $=6.2d_{(200)}$; (c) 18.05 Å $=6.4d_{(200)}$; (d) 18.61 Å $=6.6d_{(200)}$; (e) 19.18 Å $=6.8d_{(200)}$; (f) 19.74 Å $=7.0d_{(200)}$.

In summary, we have shown experimental and theoretical evidence for the presence of well-controlled nonintegral structural modulation in GaAs/AlAs superlattices. This *nonrandom* incommensurability of the superlattice with the underlying lattice is explained by introducing the concept of controlled roughness.

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