STRUCTURAL FLUCTUATIONS AND RANDOMNESS IN GaAs-$Al_xGa_{1-x}As$ SUPERLATTICES

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We discuss the use of X-ray and Raman scattering to probe structural disorder in aperiodic GaAs-$Al_xGa_{1-x}As$ superlattices, including random and quasiperiodic examples. Evidence is found for the presence of monolayer-thick steps at the interfaces. The X-ray data appear far more sensitive to this type of disorder than the acoustic phonon spectra obtained by Raman scattering.

I. INTRODUCTION

The high degree of control offered by modern thin-film growth techniques such as Molecular Beam Epitaxy (MBE) opens up many interesting opportunities for basic studies in solid state physics.[1] In this context attention has focussed recently on new classes of aperiodic thin-film structures in which the different layers are deposited according to some non-repeating sequence.[2-4]

Of particular interest are the so-called quasiperiodic superlattices reported by Merlin et al.[2] These heterostructures are experimental realizations of one-dimensional incommensurate potentials giving rise to properties quite unlike those of normal periodic systems.[5-7] For example, one of the most interesting predictions[8] is that of a hierarchical band structure exhibiting a complex self-similar spectrum of gaps. This unusual electronic structure is the result of the characterestic scaling symmetry of such systems. A related area of interest concerns studies of Anderson localization in disordered media utilizing random thin-film superlattices.[3,5]

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It is crucial in all of these studies to be able to distinguish the deliberately introduced aperiodic structure from structural disorder which is inherent to the growth process. The latter may include, for example, variations in layer thickness due to growth rate fluctuations, interface roughening, or atomic interdiffusion.[9] In this paper we present X-ray and Raman scattering results on a number of periodic and aperiodic superlattices in order to probe structural fluctuations. We find that high resolution measurements of the X-ray line-width[10] provide an accurate measure of the amplitude of growth fluctuations. Raman measurements of the the acoustic phonon modes, on the other hand, are found to be more sensitive to the sequencing of layers than low-angle X-ray diffraction. Thus the two techniques provide complementary structural information on aperiodic superlattices.

II. EXPERIMENTAL

Epitaxial films of GaAs-$Al_xGa_{1-x}As$ ($x \approx 0.30$) superlattices were grown on GaAs substrates in the (100) orientation using a VG-V80H MBE system.
Three types of superlattices were prepared:

(i) Periodic, with alternating layers of $Al_xGa_{1-x}As$ (A) and GaAs (B).
(ii) Quasiperiodic,[2,11] with layers deposited according to the Fibonacci sequence [ABAABABA...].

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(iii) Random, with layers A and B selected according to a random-number generator.

In this last example the probability of selecting A or B was weighted in the ratio τ:1, where τ is the golden mean, $(\sqrt{5} + 1)/2$. This was done in order to maintain approximately the same overall ratio of A and B layers in (ii) and (iii) in an attempt to produce a type of randomized Fibonacci structure. In all samples $d_A \approx 2d_B \approx 50Å$ and a total of 300 layers (either A or B) were deposited resulting in film thicknesses of approximately 1μm.

High-resolution (00ℓ) X-ray diffraction scans were obtained on a Huber diffractometer fitted with Ge(111) monochromator and analyzer crystals. This gave a resolution of $2\times10^{-3}Å^{-1}$ full width at half maximum (FWHM). The X-ray source was a Rigaku RU-200 operating with a molybdenum rotating anode (λ=0.71Å).

Raman spectra were recorded on a Spex double monochromator in the $z(x',x')\bar{z}$ backscattering configuration, where z is normal to the layers and x' is along the [110] direction. This geometry allows scattering only by longitudinal acoustic (LA) phonons with wavevector parallel to [001].[12] The excitation frequency corresponded to λ_L=4765Å.

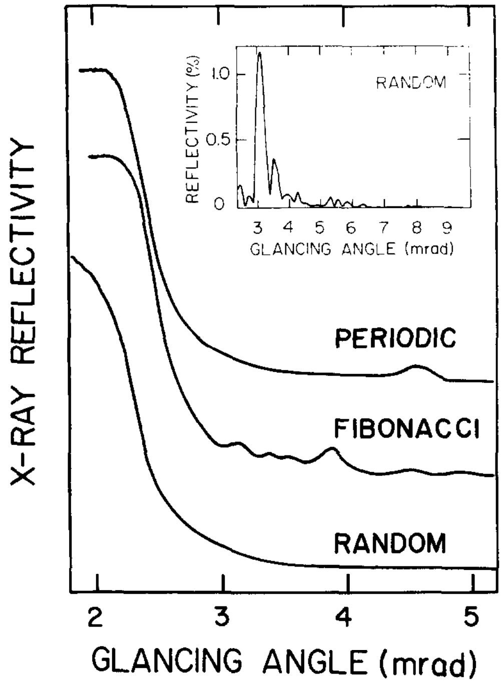

FIGURE 1. Experimental low-angle X-ray reflectivity curves for periodic, quasiperiodic (Fibonacci) and random GaAs-$Al_xGa_{1-x}As$ superlattices. The curves are offset for clarity. The reflectivity rises to 100% at low glancing angles. Inset: Calculated X-ray reflectivity curve for random superlattice using multiple scattering formulation described in the text. Optical parameters: $\delta_{GaAs} = 1.75 \times 10^{-6}$, $\delta_{(Al,Ga)As} = 1.33\times10^{-6}$ and $\beta = 2.03\times10^{-7}$ for both types of layers.

III. RESULTS AND DISCUSSION

Traditionally, low-angle X-ray scattering is used to probe long-range correlations in artificial multilayer materials. Fig. 1 compares the low-angle X-ray scattering profiles for the three different superlattices described in the previous section. The overall shape of the profiles is dominated by total external reflection of X-rays[13] when the glancing angle of incidence, θ, is less than the critical angle, $\theta_c=\sqrt{2\delta}$. Here δ is the real part of the complex index of refraction for X-rays, $n=1-\delta-i\beta$.

The reflectivity data for $\theta>\theta_c$ contain information about the relative sequencing of the superlattice layers. The periodic case is the simplest to analyze since it shows peaks only at $k=2\pi\ell/c_0$, where c_0 is the superlattice period (d_A+d_B), $k=(4\pi\sin\theta)/\lambda$ and ℓ is an integer. For ideally abrupt interfaces (square wave modulation), odd-integer peaks are favored. The ℓ=1 peak can be seen in Fig. 1.

The quasiperiodic case is also relatively straightforward to analyze since the diffraction profile can be shown[2,14] to exhibit sharp peaks at:

$$k = \frac{2\pi}{d}(m+n\tau) \qquad (1)$$

where $d = \tau d_A + d_B$.

Moreover, the most intense peaks occur in a geometric progression:[2,11]

$$k_p = \frac{2\pi}{d}\tau^p \qquad (2)$$

where p is an integer. Various analytical expressions for the intensities of diffraction peaks in quasiperiodic lattices have been derived in the kinematic (single scattering) approximation. These have been quite successful[11] in reproducing the most prominent features of the diffraction patterns. However, because of the dense nature of the quasiperiodic reciprocal space (Eq. 1) we felt that it would be more accurate to include the possibility of multiple scattering of X-rays in the interior of the film. We have therefore implemented a numerical procedure to calculate the X-ray reflectivity from any sequence of multilayers, periodic or non-periodic, which takes into account the multiple reflections and transmission at all the superlattice interfaces. The method also turns out to be useful for treating random sequences.

The numerical procedure is an extension of that proposed by Parratt[15] and further developed by Underwood and Barbee[16] for their X-ray and UV optical studies of layered synthetic microstructures. The method is based on solving iteratively a Fresnel-like recursion relation for the reflected amplitude at the interface between the jth and (j+1)th layers:[16]

$$R_{j,j+1} = a_j^4 \left(\frac{R_{j+1,j+2} + F_{j,j+1}}{R_{j+1,j+2}\,F_{j,j+1} + 1}\right) \qquad (3)$$

Here, a_j is an attenuation coefficient and the Fresnel coefficients are defined by:

$$F_{j,j+1} = \frac{E_j^R}{E_j} = \frac{g_j - g_{j+1}}{g_j + g_{j+1}} \qquad (4)$$

and $g_j = (\tilde{n}_j^2 - \cos^2\theta)^{1/2}$, where $\tilde{n}_j$ is the complex refractive index of the j'th layer.

The inset of Fig. 1 shows the results of such a calculation for the random superlattice of 300 layers described in section II. The appearance of relatively sharp features in this calculated reflectivity profile strikingly illustrates a major difficulty in preparing a realistically random medium from a finite number of elements such as the layers in a thin-film:[17,18] the finite size introduces "noise" in the form of discrete Fourier components which mimic those of systems with long-range order.[19] The structure appearing in the calculated reflectivity profile for the random superlattice (inset, Fig. 1) suggests that at the level of 1% reflectivity it should be possible to observe the sharp features experimentally. However, the measured X-ray reflectivity curve for the random case (see Fig. 1) is completely smooth in the region $3<\theta<10$ mrad showing no structure whatsoever.

A possible reason for this discrepancy is the presence of interface fluctuations in the superlattices which lead to interpenetration of neighboring layers. Measurements of the X-ray peak widths for periodic and quasiperiodic samples, shown in Fig. 2, reveal interface fluctuations at the level of ±1 atomic monolayer per ~100Å layer pair (A+B). The fact that different samples grown under different conditions (even on different MBE systems) show the same degree of broadening suggests that its origin is intrinsic rather than an artifact of growth such as evaporation-source instabilities or inaccuracies in the shutter operation. We believe that interface steps (in the form of growth islands)[20] are the most probable cause of the broadening. We are in the process of checking this idea directly by high-resolution transmission electron microscopy. It is interesting to note that recent photoluminescence studies[21] on high quality GaAs-$Al_xGa_{1-x}As$ superlattices also concluded that interface steps at the level of no greater than a monolayer were present in the samples which could account for discrete split peaks in the luminescence spectra.

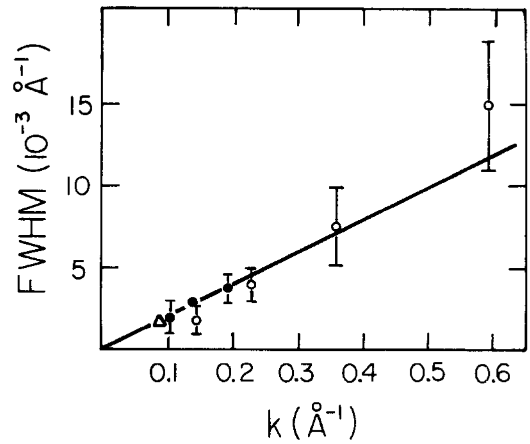

FIGURE 2. X-ray peak widths (FWHM) for periodic (Δ) and quasiperiodic (•) superlattices of GaAs-$Al_xGa_{1-x}As$. For comparison, some previous measurements[24] on a quasiperiodic GaAs-AlAs sample (o), grown at the University of Michigan have been included.[2,11] The straight line represents the expected theoretical behavior: FWHM = ak, where a is a constant determined by the RMS interface fluctuation amplitude. The peak widths have been corrected for instrumental broadening.

Finally, we turn briefly to another technique that is widely used to characterize semiconductor heterostructures, Raman scattering. The technique provides structural information through modulation of the photoelastic coefficient.[12] Measurements of the LA phonon spectrum in the quasiperiodic superlattice (not shown) show pairs of peaks whose mid-frequencies correspond to phonons at wavevectors equal to those of the prominent peaks in the X-ray diffraction profile (Fig. 1, $\theta>\theta_c$), as reported in Refs. 22 and 23. A Raman spectrum of the random superlattice (Fig. 3) also shows discrete structure, due to finite-size effects.[17,18] The features are somewhat broader than in the quasiperiodic spectrum. Comparison of Fig. 3 with the X-ray measurements in Fig. 1 (lowermost curve) indicates that, in the presence of interface disorder, the Raman technique is much more sensitive to the form of the layering sequence than X-ray scattering. The latter shows experimentally only a featureless profile for the random superlattice.

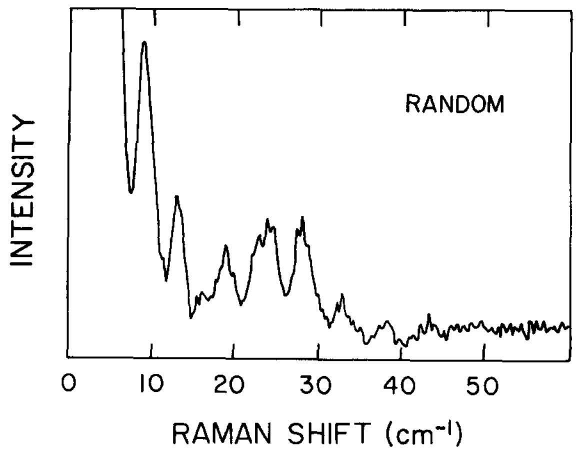

FIGURE 3. Raman spectrum of the GaAs-$Al_xGa_{1-x}As$ random superlattice.

IV. CONCLUSIONS

We have implemented a multiple scattering approach for structural analysis of aperiodic superlattices. Preliminary work on random systems shows that the low-angle X-ray reflectivity is sensitive to interface fluctuations to the extent that monolayer steps completely smear out the fine structure expected from calculations of the "ideal" random superlattice. On the other hand, fine structure in Raman measurements of the LA phonon spectrum seems to be much less sensitive to this kind of disorder. We do not fully understand these differences but suggest that it is perhaps the excitation of a relatively extended normal mode spectrum that makes the Raman scattering mechanism less susceptible to local structural inhomogeneities.

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