Synchrotron X-Ray Study of a Fibonacci Superlattice

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Quasiperiodic ordering is studied in a GaAs-AlAs Fibonacci superlattice by high-resolution x-ray scattering. The data are consistent with the predicted dense set of diffraction vectors. Moderately large growth fluctuations in the sequential deposition of GaAs and AlAs layers do not appear to disturb seriously the quasiperiodic order. The effects of randomness are analyzed in a computer simulation.

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Recently there has been a good deal of controversy surrounding the applicability of quasiperiodic geometries to rapidly quenched Al-Mn alloys. First identifications of quasicrystal structure were made on the basis of electron diffraction data.[1–3] Subsequent measurements using x-ray scattering techniques at much higher resolution could be indexed in terms of a quasicrystalline pattern exhibiting icosahedral symmetry[4]; however, certain diffraction peaks were broadened while others remained sharp. These effects have been interpreted as evidence for a random packing of units with icosahedral symmetry.[5] Recently, the role of disorder in quasicrystals has been discussed in terms of phononlike and phasonlike strains.[6]

It is of interest in this context to study high-resolution x-ray scattering from a model system in which quasicrystalline order is established. This has recently been achieved[7] by the deposition of GaAs and AlAs layers in a Fibonacci sequence by means of molecular-beam epitaxy (MBE) techniques.

Even under the most stringent experimental conditions currently available, it is not possible to make a perfectly ordered quasiperiodic structure: Some randomness is inevitably introduced into MBE-grown material by the method of deposition. A surprising finding of this study is that the disorder does not appear to disrupt seriously the overall coherence of the quasiperiodic sequence. This result suggests that the unusual physical behavior associated with quasiperiodic ordering, for example, the hierarchy of electronic bands and associated "critical" nature of wave functions,[8] may be preserved in the presence of significant levels of randomness. In order to gain more insight into this question we have modeled the effects of disorder on the x-ray scattering by introducing interface randomness into a computer simulation of the ideal Fibonacci superlattice.

Figure 1 shows schematically the ordering of GaAs and AlAs layers in a Fibonacci sequence to form the one-dimensional quasiperiodic heterostructure described previously.[7] The Fibonacci sequence leads to a structure which has an incommensurability ratio given by the golden mean, $\tau = (1+\sqrt{5})/2$, parallel to the layering axis. The thicknesses of the two "building blocks" A and B (see Fig. 1) were also chosen to be approximately in this ratio. Strictly, this is not necessary in order to achieve a quasiperiodic structure[7]; the

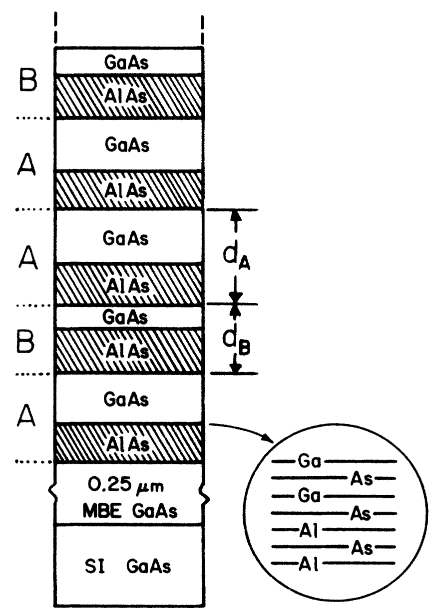

FIG. 1. Schematic arrangement of GaAs and AlAs layers deposited in a Fibonacci sequence: *ABAABABA* . . . . The AlAs strata are nominally of identical thicknesses (~ 17 Å); $d_A \simeq 59$ Å, $d_B \simeq 37$ Å.

key factor is the arrangement of layers according to a Fibonacci sequence.

The Fourier transform of the ideal Fibonacci sequence consists of a dense set[3,7] of components such that diffraction peaks are expected at all wave vectors in the reciprocal space defined by $k=2\pi/[(\tau d_A+d_B)\times(m+n\tau)]$, where n and m are integers; $d_A \simeq 59$ Å and $d_B \simeq 37$ Å are the respective thicknesses of the building blocks A and B shown in Fig. 1. Experimentally, every increase in instrumental resolution is thus expected to reveal new, weak, resolution-limited peaks in what was previously unresolved background. This aspect is well illustrated by comparison of the data shown in the inset of Fig. 2, taken with longitudinal resolution 0.03 $Å^{-1}$ FWHM, with the data shown in the main body of Fig. 2, a portion of a scan with resolution 0.0015 $Å^{-1}$ FWHM. The data were obtained on bending-magnet beam line X22 at the Brookhaven National Synchrotron Light Source with use of flat Ge (111) monochromator and analyzer crystals.

The data shown in Fig. 2 are part of a more extensive set of $(00l)$ scans performed at low angles ($k \lesssim 0.1$ $Å^{-1}$), and around the 002 and 004 GaAs/AlAs peaks at $k \approx 2.2$ and 4.4 $Å^{-1}$, respectively. In general, we observe no peak broadening that could not be accounted for by the instrumental resolution function, which at the higher angles was dominated by dispersion. Moreover, the relative intensities of the peaks in the measured diffraction profile agree remarkably well over a wide range of wave vectors with those calculated from an ideal GaAs/AlAs Fibonacci superlattice model. The model consisted of $\sim 13\,200$ atomic layers of Ga, As, and Al appropriately sequenced; no temperature factors or polarization corrections were applied. The fit with a small portion of the calculated profile, which was convoluted with the instrumental resolution function, is shown in Fig. 2.

A general feature of the diffraction pattern of the GaAs/AlAs Fibonacci superlattice is the enhancement of the intensities of peaks at wave vectors close to the even-order peaks of GaAs (002, 004, etc.); see inset of Fig. 2. This enhancement is caused by the almost identical lattice parameters of GaAs and AlAs, a factor which is also important in the achievement of excellent epitaxial lattice matching in the directions perpendicular to the quasiperiodic layering. Thus, the quasiperiodic arrangement in this particular superlattice is determined primarily by the *chemical* sequencing of al-

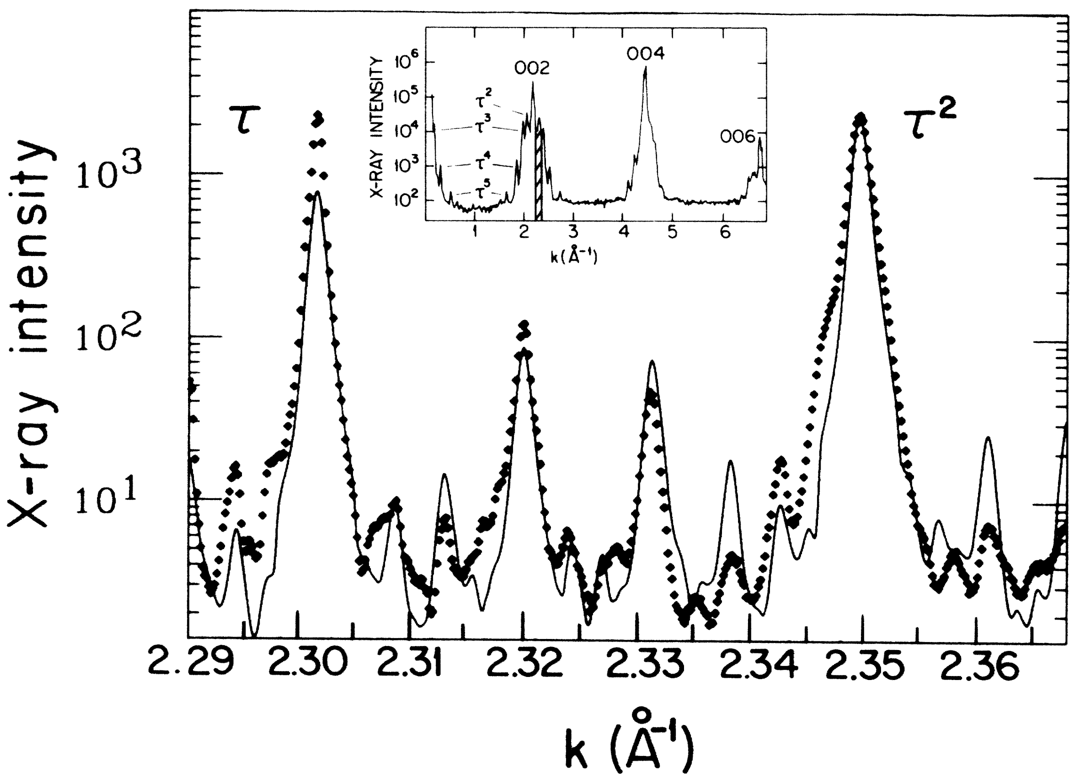

FIG. 2. $(00l)$ diffraction profiles of the GaAs/AlAs Fibonacci superlattice. The dots represent high-resolution synchrotron x-ray data and the line is calculated from the ideal model described in the text. Inset: low-resolution $(00l)$ scan showing the overall appearance of x-ray scattering and indexing of strong "Fibonacci" peaks as powers of τ. The shaded region indicates the range of the high-resolution scan shown in the main figure.

most equally spaced (100) atomic planes (see Fig. 1).

In view of the importance of compositional sequencing in the formation of a quasiperiodic superlattice by MBE techniques, it is somewhat surprising that randomness introduced by fluctuations in the evaporation rates and timing of the source shutters does not seriously compromise the fit with the ideal (defect-free) quasiperiodic structure (see Fig. 2). In particular, even in the presence of what we estimate to be a roughly $\pm 5\%$ fluctuation in layer thickness resulting from the combined uncertainty in the MBE flux and intervals for opening and closing the various source shutters, the peaks are neither appreciably smeared out nor reduced in number relative to the predicted ideal pattern at this resolution.

In order to investigate this issue in more detail we carried out a computer simulation in which the effects of growth fluctuations are mimicked by the adding or subtracting of a single Ga/As or Al/As bilayer from the GaAs and AlAs strata with a predetermined probability, α, while still generating succeeding strata according to a Fibonacci sequence. This modification leads to a fluctuation of the individual thicknesses of building blocks A and B shown in Fig. 1. The way in which the quasiperiodic model with randomness is constructed leads to an uncertainty in the phase of the structure factor growing like $\sqrt{N}$, where N is the number of strata. Thus the "average lattice" is not preserved. In a nominally periodic superlattice ($ABAB$ . . .) this type of growth fluctuation would give rise to symmetrical broadening of the superlattice peaks.[9]

In the absence of translational symmetry, specifically for the case of a quasiperiodic structure, it is far from obvious what will be the effect of randomness. Thus, we have resorted to the computer simulations in order to analyze these effects. The most important result (see Fig. 3) is that the intense diffraction peaks are generally affected relatively little by even substantial amounts of disorder. The diffraction pattern still appears as a dense set of peaks and the intensities of the weaker peaks can be either enhanced or diminished by the introduction of randomness. We conclude from these unexpected findings that local disruption of the quasiperiodic order does not smear out or remove large numbers of peaks from the diffraction pattern. Furthermore, randomness even at the level of $\alpha = 0.2$ (one fault every ~ 130 Å on average) does not result in noticeable peak broadening on the scale of our instrumental resolution (1.5×10^{-3} Å^{-1}). We note here that the insensitivity of the most intense diffraction peaks to mistakes in the quasiperiodic sequence has also been discussed analytically by Lu and Birman[10]; however, the nature of the defects that they considered (transposition of A and B blocks at random) is not relevant to the MBE deposition method considered in this work.

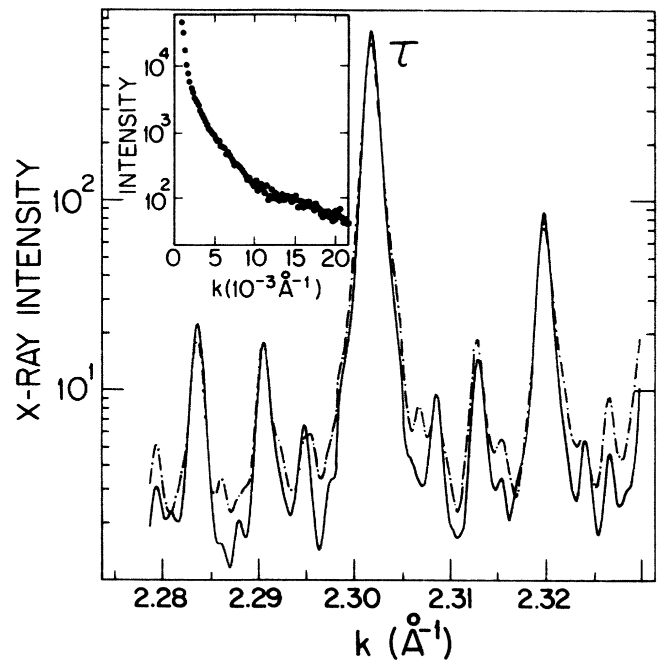

FIG. 3. The effect of growth fluctuations on the $(00l)$ diffraction profile of the quasiperiodic superlattice. Solid line: ideal Fibonacci structure; broken line: including growth fluctuations with a probability of $\alpha = 0.1$ as described in the text. Inset: low-angle portion of synchrotron data showing contribution of large-$g_\perp$ peaks.

Further insight into the effects of disorder in the quasiperiodic superlattice may be gained by our considering the form of the modulation function $S(k)$ which determines the intensity of each peak labeled by the integers (n,m). For $d_A = \tau d_B$, which is approximately obeyed in our case, $S(k)$ is given by[11]

$$S(k) = N(\sin z_{nm})/z_{nm}, \tag{1}$$

where $z_{nm} = \frac{1}{2}|g_\perp|\tau^2(1+\tau^2)^{-1/2}$ and $g_\perp = (2\pi/\tau) \times (1+\tau^2)^{-1/2}(m\tau - n)$. Thus the envelope of the intensity pattern falls off as $1/g_\perp$. This has interesting consequences for the low-angle portion of the diffraction pattern which consists entirely of contributions from high orders of m and n (of opposite sign), i.e., large-$g_\perp$ peaks. The inset of Fig. 3 shows indeed that the measured x-ray diffraction profile in this low-angle region is almost completely featureless, as expected since the profile consists of weak overlapping peaks. It is surprising that these very high-order satellites are not severely attenuated by the growth fluctuations.

A further interesting effect occurs in the opposite limit of small $g_\perp$. In the ideal Fibonacci superlattice these peaks are predicted by Eq. (1) to be relatively intense. One such set of peaks is indexed by consecutive Fibonacci numbers $(n,m) = (2,1)$, $(3,2)$, $(5,3)$, (8,5), etc. for which n/m approaches τ. It is easy to show that the diffraction wave vectors in this case can be expressed as a geometric progression, $k=[2\pi/(\tau d_A+d_B)]\tau^p$, where p is an integer.[7] This series of small-$g_\perp$ peaks is found to dominate the general form of the diffraction profile (see inset of Fig. 2) and as such can be used as a fingerprint of quasiperiodic structure. In addition, the results of our computer modeling show that this particular subset of peaks is broadened and diminished progressively with decreasing $g_\perp$ (increasing k) when growth fluctuations of the type described above are included[6]; however, the broadening is difficult to observe at the resolution limit of our experiments. Thus the high-order Fibonacci peaks ($p \gg 1$) are indicative of a high degree of quasiperiodic order in the superlattice in much the same way as the high-order Bragg satellites in a periodic heterostructure are associated with atomically sharp interfaces.

In summary, we have investigated the diffraction pattern of a quasi one-dimensional Fibonacci superlattice using x-ray scattering. At low resolution the diffraction pattern is dominated by small-$g_\perp$ peaks whose indices (n,m) are consecutive Fibonacci numbers. The richness of the quasiperiodic structure is revealed at high resolution where the data are consistent with the predicted dense set of diffraction vectors. Imperfections of the superlattice in the form of interfacial compositional disorder are found to affect peaks strongly only in the limit of $g_\perp \to 0$. Our findings suggest that the unusual physical properties of true quasicrystals will be largely preserved in the presence of random defects. There are also important implications for future experiments on *single crystals* of quenched Al-Mn in that the observation of a dense set of diffraction peaks would be a major test of the quasicrystalline structure.

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