Quasiperiodic GaAs-AlAs Heterostructures

R. Merlin, K. Bajema, and Roy Clarke
Department of Physics, The University of Michigan, Ann Arbor, Michigan 48109

and

F.-Y. Juang and P. K. Bhattacharya
Solid State Electronics Laboratory, Department of Electrical Engineering and Computer Science, The University of Michigan, Ann Arbor, Michigan 48109

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We report the first realization of a quasiperiodic (incommensurate) superlattice. The sample, grown by molecular-beam epitaxy, consists of alternating layers of GaAs and AlAs to form a Fibonacci sequence in which the ratio of incommensurate periods is equal to the golden mean τ. X-ray and Raman scattering measurements are presented that reveal some of the unique properties of these novel structures.

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In recent years there have been many theoretical studies of one-dimensional (1D) Schrödinger equations with quasiperiodic (incommensurate) potentials.[1-3] The interest here stems partly from the fact that the Bloch theorem is inapplicable and also that this problem represents, in some sense, an intermediate case between periodic and disordered 1D solids. Quasiperiodicity leads to spectra that are Cantor sets having not only pure-point components (localized eigenstates) and components with absolutely continuous measure (extended eigenstates), but also singular continuous components with chaotic extended states.[3] Unlike random or commensurate 1D solids, for which all states are either localized or extended, quasiperiodic potentials admit the existence of a mobility edge and allow for a "metal-insulator" transition to take place at a critical strength of the modulation.[3] In addition, the nature of the spectrum may depend quite dramatically on the incommensurability ratio.[3]

Quasiperiodic 1D potentials arise in the description of specific properties of many physical systems such as, e.g., incommensurate conducting linear chains,[4] and periodic solids[5] and superconducting lattices[6] in a uniform magnetic field. Experimentally, the possibility of revealing the expected richness of the problem has, however, been very limited. The widespread application of 1D Hamiltonians in the description of artificially grown heterostructures suggests a convenient experimental realization of quasiperiodicity. GaAs-$Al_xGa_{1-x}As$ superlattices are particularly appealing in this respect since their parameters can easily be tailored to meet a specific need. Although conditions are normally chosen to achieve a *periodic* superlattice, in principle, a quasiperiodic structure could be obtained by imposing an incommensurate modulation of the alloy composition or doping. It is unlikely, however, that in such a method the growth conditions could be controlled sufficiently well as to arrive precisely at the desired value of the incommensurability ratio.

In a recent work,[7] Levine and Steinhardt describe a new class of incommensurate lattices that they have termed "quasicrystals."[8] Here, we show that an extension of their ideas leads to a simple procedure that allowed us to grow a quasiperiodic GaAs-AlAs superlattice with an incommensurability ratio precisely given by the golden mean $\tau = (1+\sqrt{5})/2$. To illustrate this procedure, and also because of its relevance later, we focus now on the particular subclass of 1D quasicrystals that derives from the Fibonacci sequence.[7] Following Levine and Steinhardt, the "Bravais lattice" of such quasicrystals is given by the set of points $z_{i+1} = z_i + r_i$, where $\{r_i\}$ is the Fibonacci sequence of intervals $\{d_a d_b d_a d_a d_b \ldots\}$ with $d_a/d_b = \tau$.[7] This lattice has been shown to be quasiperiodic with two linearly independent periods of ratio equal to τ and, moreover, to exhibit self-similarity.[7] We find that, even for $d_a/d_b \neq \tau$, the quasiperiodic properties are preserved although not the self-similarity. More precisely, one can show using a recursion relation for the structure factor that for *all* $d_a \neq d_b$, the Fourier spectrum of a Fibonacci lattice consists of δ-function peaks at $k = 2\pi d^{-1}(m + m'\tau)$, where m and m' are integers and $d = \tau d_a + d_b$ is the "average" lattice parameter (this result, in a slightly different form, is given in Ref. 7 for the particular case $d_a/d_b = \tau$).

The procedure to grow what we will refer to as a *Fibonacci superlattice* now becomes clear: one needs only to attach a basis to the Fibonacci lattice. More generally, the procedure involves defining two distinct building blocks and having them ordered in a Fibonacci manner. The building blocks can each be composed of one or more layers of different materials and can have arbitrary thicknesses. Obviously, quasiperiodic superlattices with period ratios other than τ can be derived from other 1D-quasicrystal subclasses; all possible subclasses have been classified by Levine and

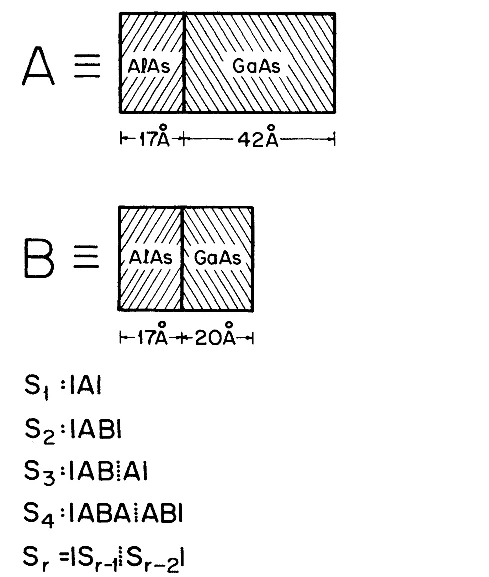

FIG. 1. The two building blocks of the Fibonacci superlattice investigated in this work. Also shown are the first four Fibonacci generations and the rule to derive generations of arbitrary order.

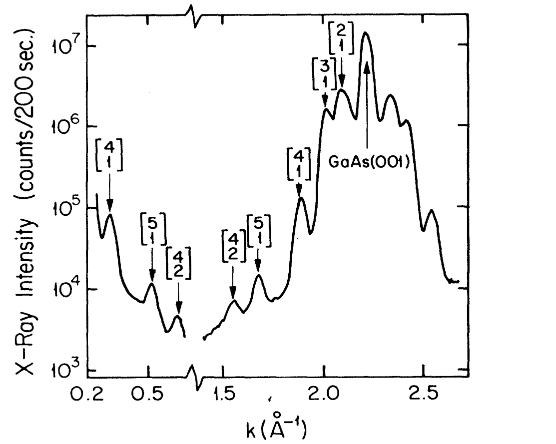

FIG. 2. Room-temperature x-ray diffraction pattern of the Fibonacci superlattice for **k** perpendicular to the layers. The arrows with labels $[{}^p_n]$ indicate the positions of $k_{p,n} = 2\pi d^{-1} n \tau^p$ and corresponding satellite reflections of the GaAs (001) peak (the substrate and superlattice peaks are not resolved in this order). Note that $k_{p+1,n} = k_{p,n} + k_{p-1,n}$.

Steinhardt.[7]

The two building blocks of the Fibonacci superlattice grown for this work are shown schematically in Fig. 1. Blocks A and B consist nominally of (17 Å AlAs)-(42 Å GaAs) and (17 Å AlAs)-(20 Å GaAs). The sample was grown by molecular-beam epitaxy on (001) GaAs. The Fibonacci sequence can be described in terms of a series of generations that follow the rules indicated in Fig. 1. Our sample consists of thirteen generations and has a total thickness of ~ 1.85 μm.

Figure 2 shows the x-ray diffraction pattern of the Fibonacci superlattice for **k** along [001]. The spectrum shows superlattice reflections ($k < 0.7$ Å^{-1}) and also satellites of GaAs(001) (at $k \approx 2.223$ Å^{-1}). The most striking feature of these data is the fact that all but the weakest peaks occur in a geometric progression with τ as the common ratio.[9] These results are consistent with a numerical calculation of scattering intensities which predicts the strongest peaks to lie at $k_{p,n} = 2\pi d^{-1} n \tau^p$, where n and p are integers (these peaks and corresponding satellites are labeled $[{}^p_n]$ in Fig. 2). From these data, we further determine that $d = \tau d_a + d_b \approx 132.9$ Å which is in very good agreement with the nominal value of 132.5 Å. The x-ray results can also be used to obtain an experimental value for τ; we find $\tau_{\rm exp} = 1.630 \pm 0.015$.

A further probe of the quasiperiodic nature of the Fibonacci superlattice is provided by Raman scattering, which gives insight into the phonon density of states. Using this technique, we have investigated the spectrum of longitudinal-acoustic (LA) phonons propagating along [001].[10] The problem of determining the spectrum of LA phonons is of interest because, for *piecewise-constant* modulations as in our case, it can be shown to map onto the problem of solving a quasiperiodic Schrödinger equation (this also applies to transverse-acoustic modes which will not be considered here because their scattering is forbidden[10] in the geometry used in the experiments). The link between the two problems is easily established if one substitutes $\rho_i \omega^2 / K_i$ by $2m/\hbar^2 (E - V_i)$ in the LA wave equation

$$-\omega^2 \rho u = \frac{d}{dz}\left(k \frac{du}{dz} \right), \tag{1}$$

where ρ_i and K_i are the constant values of the density and elastic constant for a given layer, and ω the LA frequency; m and E are the mass and eigenenergy of a particle moving in the piecewise-constant potential defined by V_i.

Raman scattering by LA phonons originates in the modulation of the corresponding photoelastic coefficient.[10] In our case it is convenient to separate the contributions due to localized and extended eigenstates. For the latter, one can use results from Refs. 2 and 10 to obtain the following expression for the scattered intensity:

$$I(\omega) \propto \left| \textstyle\sum_{k'} (q - k') P_{k'} u_{q-k-k'} \right|^2 [n(\omega) + 1]/\omega. \tag{2}$$

Here k is the Bloch index[2] of the phonon with frequency ω, $n(\omega)$ is the Bose factor, q is the scattering

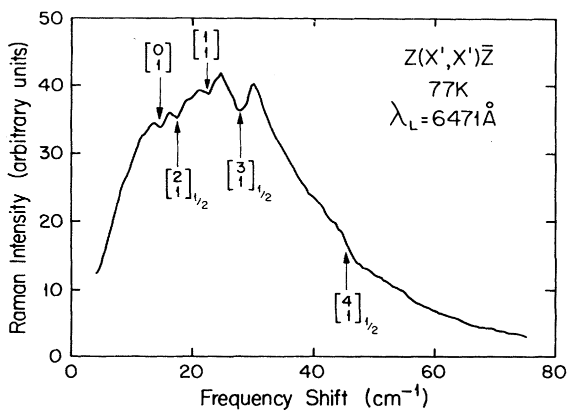

FIG. 3. Raman spectrum of the Fibonacci superlattice showing scattering by LA phonons propagating along [001] [the intensity has been divided by $n(\omega)+1$, where $n(\omega)$ is the Bose factor]. The features labeled $[{}^{p}_{n}]$ and $[{}^{p}_{n}]_{1/2}$ are dips occurring at $\omega_{p,n}=ck_{p,n}$ (see text) and at $\frac{1}{2}\omega_{p,n}$ which reflect associated gaps in the density of states. The backscattering geometry $z(x',x')\bar{z}$ corresponds to polarizations of the incident and scattered light along $x'=[110]$, with the wave vector of the incident photon along $z=[001]$.

wave vector, and $P_{k'}$ and $u_{q-k-k'}$ are the corresponding Fourier components of the photoelastic coefficient and the phonon amplitude. Periodic superlattices show nonzero Fourier components at $2\pi d_0^{-1}m$ (d_0 is the period and m is an integer) and the Raman spectrum consists of a series of discrete lines that correspond to phonons with $k=|q+2\pi d_0^{-1}m|$.[10] For quasiperiodic structures, the δ functions of the Fourier spectrum densely fill the real axis. Accordingly, the Raman spectrum associated with extended modes should be a weighted density of states with weighting factors defined in Eq. (2). The same applies to localized eigenmodes if we set $k=0$ in Eq. (2).

The measured Raman spectrum of the Fibonacci superlattice is shown in Fig. 3. The scattering geometry $z(x',x')\bar{z}$ allows only LA modes.[10] As expected from the discussion above, Fig. 3 shows a continuum with clearly resolved "dips" that we ascribe to gaps in the density of states of LA modes. Quasiperiodic structures should exhibit an infinite number of gaps, the principals of which are associated with the largest Fourier components of the modulation.[3] We do indeed find a close correspondence between the x-ray pattern and the Raman spectrum in that the dips in the latter occur at positions approximately given by $\omega=ck_{p,n}$ and $ck_{p,n}/2$ (labels $[{}^{p}_{n}]$ and $[{}^{p}_{n}]_{1/2}$ in Fig. 3), with $c\cong 4.42\times10^5$ cm/s. This is consistent with the fact that the elastic constants of GaAs and AlAs are very similar; in a perturbation approach,[10] the frequencies $ck_{p,n}$ and $ck_{p,n}/2$ correspond to midgap values associated with the "reciprocal lattice vectors" $2k_{p,n}$ and $k_{p,n}$. Further, the value of c from the experiments is close to the calculated value of 4.96×10^5 cm/s for the longitudinal sound velocity of the superlattice, based on known elastic constants of GaAs and AlAs.[10]

In summary, we have demonstrated a new kind of heterostructure: a Fibonacci superlattice exhibiting quasiperiodic order. Preliminary results of x-ray and Raman scattering experiments reveal striking features associated with the quasiperiodic nature of the structure, in particular, the occurrence of spectral singularities that follow a power-law behavior. Realization of such well-controlled quasiperiodic structures holds promise for a wide range of experimental studies in transport, lattice dynamics, magnetic ordering, and superconducting behavior.

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Note added.—After this manuscript was submitted, we learned of a recent paper by Zia and Dallas where a very elegant derivation of the structure factor of 1D quasicrystals is presented.[11] The result $k=2\pi d^{-1}(m+m'\tau)$ for arbitrary $d_a\neq d_b$ can be obtained by use of their projection method on a *rectangular* (instead of a square) lattice.

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[8] In two or three dimensions, it is important to distinguish between *quasicrystals* and incommensurate structures resulting from incommensurate modulations of periodic lattices. However, the distinction disappears in the one-dimensional case.

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