High-Resolution X-Ray-Diffraction Spectra of Thue-Morse GaAs-AlAs Heterostructures: Towards a Novel Description of Disorder

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We present the first analysis of high-resolution x-ray-diffraction spectra of finite-size Thue-Morse GaAs-AlAs superlattice heterostructures, which show essential properties of singular continuous measures. The implications for future investigations of disordered systems are considered.

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The experimental discovery of quasicrystals[1] in 1984 has opened a new field of research to both experimentalists and theoreticians. In this field, the importance of deterministic structures having controlled aperiodic disorder is being increasingly recognized. This is why one-dimensional deterministic sequences generated by substitutions or finite automata[2-4] are now widely used mathematical objects to build such structures. In particular, 3D multilayer heterostructures having two kinds of layers arranged according to the Fibonacci sequence were first defined and effectively made as early as 1985 by molecular-beam epitaxy[5,6] (MBE) and consequently investigated by x-ray and neutron diffraction,[6-9] Raman scattering,[10,11] etc.

Prompted by all of these studies, an extension of such methods to *nonquasiperiodic* systems soon began (for a review, see for example, Ref. 12), with special interest in the Thue-Morse sequence,[13-16] and its mathematical and physical properties.[17-23] For instance, in 1987 a Thue-Morse superlattice heterostructure was made for the first time and investigated by Raman scattering.[10]

Let us recall that the Thue-Morse sequence which can be generated using a two-automation[2,4,17] can also be defined on a two-letter alphabet $\{0,1\}$ using the following substitution rule σ:

$$\sigma(0)\rightarrow 01,\quad \sigma(1)\rightarrow 10\,. \tag{1}$$

Starting, for example, with 0 one obtains, by successive application of this rule, after $n=4$ iterations a sequence of length $2^n=16$: 0110100110010110 and so on. It is well known that the Thue-Morse sequence has a singular continuous Fourier transform[16,24,25] and this fact, as we shall see below, focuses interest for the first time on a purely measure-theoretic property and its possible physical role in the properties of the high-resolution x-ray-diffraction spectra of Thue-Morse superlattice heterostructures.

A "measure" describes how a certain quantity is "attributed" to a set[26]—the most usual illustration resorts to the mass. After the famous decomposition theorem due to Lebesgue, any measure μ can be decomposed in a unique fashion into three primitive types of measures,

$$\mu=\mu_{\mathrm{AT}}+\mu_{\mathrm{SC}}+\mu_{\mathrm{AC}}\,, \tag{2}$$

the so-called atomic (AT), singular continuous (SC), and absolutely continuous (AC) measures. One can say[27] that to the measure μ of a set, or of an interval, is associated in a unique fashion an increasing right continuous function φ which is some sort of primitive of μ:

$$\mu(]a,b])=\varphi_\mu(b)-\varphi_\mu(a)\,. \tag{3}$$

An example of *atomic* measure is Dirac's measure, also called by physicists the "Dirac delta function." Then, with a mass at the point $x=a$, the function φ is a step function [$\varphi(x)=0$ if $x<a$; $\varphi(x)=1$ if $x>a$]. It is *not* continuous. The Fourier transform of the Fibonacci sequence has an atomic measure with a countable number of peaks.

Lebesgue's measure is *absolutely continuous,* and the associated φ function is continuous and differentiable. [$\varphi(x)=x$]. The Fourier transform of a random sequence as well as of the Rudin-Shapiro sequence has an absolutely continuous measure.

It is a *singular continuous* measure which is associated with the function φ called a "devil's staircase"[28] (previously known as "Lebesgue's singular function"). φ is continuous, and the measure has no point masses, but it is carried by a Cantor set. Both absolutely and singular continuous measures have long been called "diffuse measures."

A deterministic sequence being given, we shall be interested in the decomposition according to Lebesgue's theorem of the measure associated with its Fourier transform and, in a physical realization of this sequence, with the possible evolution, depending on physical parameters, of each of its three primitive components, AT, AC, and SC. We shall now turn to the specific case of the high-resolution x-ray study of a Thue-Morse multilayer heterostructure.

The Thue-Morse superlattice heterostructures, having 2^n layers, were grown on a GaAs(001) substrate by molecular-beam epitxy (MBE). The deposition rate was about 1 Å/sec. The lattice simply consisted of AlAs (A) and GaAs (B) layers. The thicknesses of alternating layers, d_A and d_B, were determined by the intensity oscillation of the specularly reflected electron beam. The values of d_A and d_B were finally designed to be $d_A=d_B=5a_0$, where a_0 is the average constant of the cubic "zinc-blende" lattice of AlAs and GaAs.

The measurement of the x-ray-diffraction pattern was

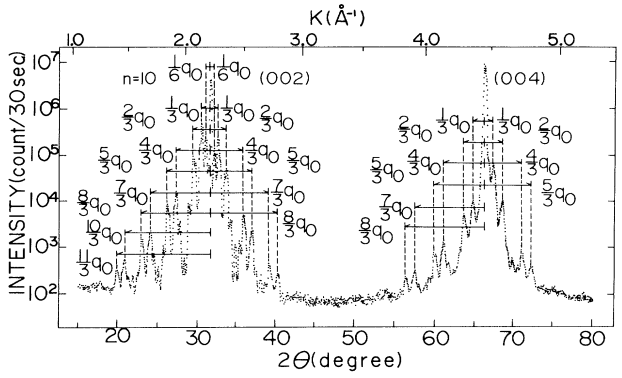

FIG. 1. X-ray-diffraction pattern of a GaAs-AlAs Thue-Morse superlattice heterostructure with 2^{10} layers (see text).

made using a triple-axis spectrometer with Cu $K\alpha_1$ radiation (50 kV, 250 mA) monochromatized by (002) reflection from a pyrolytic graphite crystal. A divergent slit of 0.15° and receiving slit of 0.10° were employed as an x-ray optical system. The detailed procedure of the measurement is nearly the same as that of the measurement in the Fibonacci lattices and has been described elsewhere.[6,8,9]

The observed diffraction pattern is given in Fig. 1 for $n=10$, where K is the magnitude of the scattering vector $4\pi\sin\theta/\lambda$. One observes numerous peaks superimposed on the fundamental (002) and (004) reflections referred to the average structure of GaAs and AlAs. In order to investigate the spectra in more detail, a high-resolution measurement was carried out using a GaAs (002) monochromator and a Si (111) analyzer.

The high-resolution diffraction patterns just above the scattering angle (2θ) of the GaAs (004) reflection are given in Figs. 2(a) and 2(b), for $n=10$ and 7, respectively, where we take as the unit of q, $q_0=2\pi/5a_0$, where q_0 is the wave number associated with the thickness of alternating layers $d_A=d_B=5a_0$. The resolution is so high that the (004) reflections of the grown heterostructure and the GaAs substrate are well separated around $2\theta \sim 66$. Comparison with the corresponding high-resolution spectra of Refs. 8 and 9 composed of a dense set of Bragg peaks shows that here the peaks are generally broader, and the underlying self-similar pattern, governed in the Fibonacci case by powers of τ^{-1}, less immediately obvious.

As shown in Figs. 2(a) and 2(b), the peaks on the high-resolution spectrum, which are well resolved, can be labeled by $(2k+1)/3.2^p$, with k and p integers, with an accuracy of over $\frac{1}{200}$. It has been known for a long time (see, e.g., Ref. 20) that the intensity of the Fourier transform of the Thue-Morse heterostructure of finite length $L=2^n$ is

$$I_n(q)=2^{2n}\prod_{j=0}^{n-1}\sin^2(2^j\pi q)\,. \tag{4}$$

In the limit where $n\to\infty$, this intensity, which is a Riesz product,[26] defines a singular continuous measure and is a "pure" case of the Lebesgue decomposition theorem. It can be easily realized from Eq. (4) that $q=\frac{1}{3}$ yields identical values for all the factors of the product. Also, defining the scaling properties of the intensity with sample size by

$$I_n(q)=L^{\alpha_n(q)}\,, \tag{5}$$

it can be proven[20] that $\alpha_n(\frac{1}{3})$ is, in fact, independent of n and the largest scaling exponent. As a consequence one can predict that when n increases, only those peaks having an abscissa q of the form $(2k+1)/3.2^p$, with k and p integers, will keep a sufficient intensity (these are the values of q whose difference from $\frac{1}{3}$ is a dyadic $l/2^m$, with l and m integers), and they will become increasingly dense. This is exactly what we observe. The fractal nature[28] of the spectral properties is obvious.

This can be understood yet in another fashion. In previous work,[19] it has been proved that the energy spectrum of a 1D classical harmonic chain made of identical springs and of masses of two different kinds arranged after the Thue-Morse sequence is a Cantor-like set, and that the value of the integrated density of states on each of the gaps is precisely $(2k+1)/3.2^p$, with k and p integers (see also Refs. 29 and 30). Also, it was indicated,[29] using perturbative methods, that the peaks in the Fourier transform of the sequence itself generate and label the gaps of the energy spectrum of a tight-binding discrete Schrödinger equation with its couplings determined by this very same sequence—closely related, via a trivial mapping to the mass and spring equation. This reasoning then leads to the same result.

Further observation of Figs. 2(a) and 2(b) clearly shows that line shape and height evolve in a characteristic fashion upon increase of n: Lines become thinner and $\alpha_n(q)$ increases with n for a given q. The intensities of the x-ray-diffraction pattern were carefully measured, and then normalized in the same fashion as the (004) reflection, which goes as L^2. The background level was estimated from the diffraction profile of the GaAs substrate with no deposition. So, after the relevant processing of the data, $\alpha_n(q)$ could be measured from the $I_n(q)$ values so obtained. A few calculated [from Eqs. (4) and (5)] and measured values of $\alpha_n(q)$ are summarized in Table I. The uncertainty is estimated to be less than 0.06, about 4%, due in part to peak overlap. Note that, for an atomic measure ("Bragg peaks"), one would have $\alpha_n(q)=2$ for all n and all q in this situation. A detailed line-shape analysis will be presented elsewhere.

X-ray-diffraction patterns from perfect crystals are composed exclusively of Bragg peaks whose positions, labeled by three integers (hkl), allow, together with extinction rules and intensity measurements, a complete reconstitution of the structure. Patterns from incommensurate samples, also composed of Bragg peaks, will take at least four integers ($hklm$) to label their positions.[31] In our singular continuous case, the peaks are

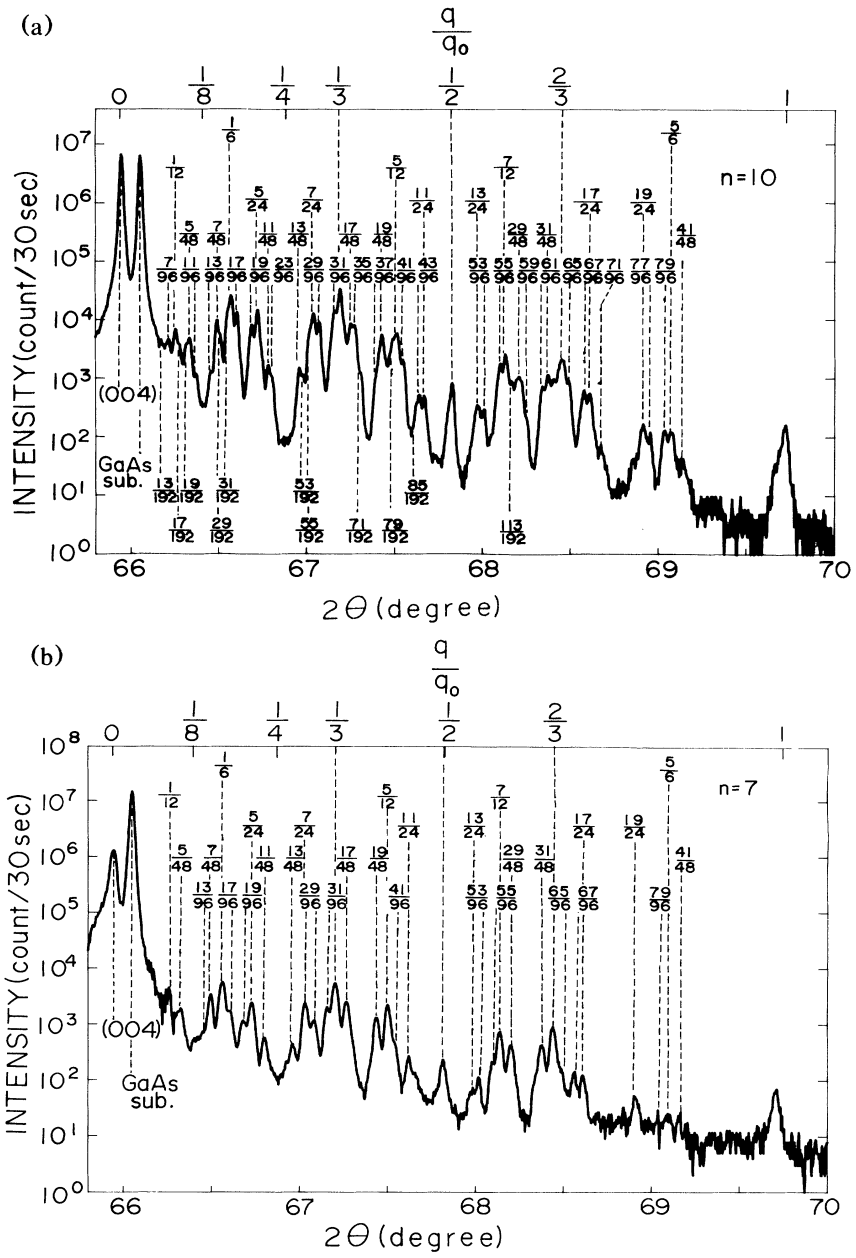

FIG. 2. High-resolution x-ray-diffraction pattern of a GaAs-AlAs Thue-Morse superlattice heterostructure with (a) 2^{10} layers and (b) 2^7 layers observed just above the scattering angle (2θ) of the GaAs (004) reflection. The dashed lines indicate peak assignment to $2k+1/3.2^p$ (k and p integers) in units of q_0 (see text). Note the change in peak height and shape with sample size.

labeled by very specific rationals, but line shape and height evolution as a function of wave vector and sample size also carry information which can be specific of the controlled disorder sequence generating the diffraction pattern—at variance with the classical situation. It is too early to decide whether a one-to-one correspondence between x-ray-diffraction spectra of such heterostructures and their generating sequence can be established, which would then allow the accurate understanding of a large new class of disordered systems.

In conclusion, to the best of our knowledge, this is the very first time that x-ray-diffraction spectra having the essential properties of a singular continuous measure —even if these are finite-size samples—have been obtained and understood. We have shown that this opens a whole new field in the study of disordered systems, and

TABLE I. Examples of values of $\alpha_n(q)$ calculated and measured from the x-ray-diffraction patterns of Fig. 2 (see text).

	$n=10$		$n=7$	
q/q_0	Calculated	Measured	Calculated	Measured
$\frac{1}{3}$	1.5849	1.58	1.5849	1.52
$\frac{1}{6}$	1.4265	1.45	1.3585	1.34
$\frac{5}{12}$	1.4580	1.42	1.4082	1.40
$\frac{7}{24}$	1.4327	1.46	1.3674	1.31

that careful and detailed analysis of their x-ray-diffraction spectra in terms of peak position as well as of line shape and height can yield accurate information on certain nonconventional structural properties.

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