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RAMAN SCATTERING BY ACOUSTIC PHONONS AND STRUCTURAL PROPERTIES OF FIBONACCI, THUE-MORSE AND RANDOM SUPERLATTICES

R. MERLIN, K. BAJEMA, J. NAGLE* and K. PLOOG*

Department of Physics, The University of Michigan, Ann Arbor, MI 48109-1120, U.S.A.

**Max-Planck-Institut für Festkörperforschung Heisenbergstr. 1, D-7000 Stuttgart 80, F.R.G.*

Des études structurales de superréseaux GaAs-AlAs incommensurables et désordonnés ont été réalisées par des mesures de diffusion Raman par les phonons acoustiques. Les propriétés du facteur de structure des superréseaux de type Fibonacci et Thue-Morse sont discutées.

We report structural studies of incommensurate and random GaAs-AlAs superlattices using Raman scattering by acoustic phonons. Properties of the structure factor of Fibonacci and Thue-Morse superlattices are discussed in some detail.

Non-periodic layered structures and, in particular, random and Fibonacci superlattices (FSL's) have recently received much attention.¹⁻⁴ The motivation for this is largely the fact that these structures are a realization of well known one-dimensional (1D) models showing features quite unlike those of periodic systems.⁴⁻⁸ The interest in random superlattices focuses on the problem of Anderson localization.^{2,4} FSL's are 1D analogs of *quasicrystals*,⁹ with wave behavior characterized by a self-similar hierarchy of gaps and critical (or chaotic) eigenstates.⁵⁻⁷ Structures based on *automatic* sequences have been also considered in the literature.¹⁰ Thue-Morse superlattices (TMSL's) belong to this group.

Raman scattering (RS) has been extensively applied to the study of acoustic phonons in periodic semiconductor structures.¹¹ Such studies provide information mainly on the structural properties of superlattices and, to a lesser extent, on the frequency spectrum of sound waves.¹¹ In this report we concentrate on the structural aspects of RS in layered systems. We review recent works^{12,13} on FSL's and present new results on GaAs-AlAs random and Thue-Morse structures.

The samples used in this study consist of sequences of two building blocks A (GaAs) and B (AlAs) of thicknesses $d_A = d_B = 20\text{Å}$. They were grown by molecular beam epitaxy on (001) GaAs substrates. Raman spectra were obtained 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 only allows scattering by longitudinal acoustic (LA) phonons with wavevector along [001].¹¹

In the photoelastic continuum model, the intensity for RS by LA phonons is given by:¹¹

$$I(\Omega) \propto \left\langle \left| \int_{-\infty}^{+\infty} e^{-iqz} P(z) \frac{\partial U}{\partial z} dz \right|^2 \right\rangle_{\Omega}, \quad (1)$$

where $U(z)$ is the amplitude of the mode with frequency Ω , $P(z)$ is the local photoelastic coefficient $P^{12} = P_A, P_B$ and q is the scattering wavevector. In GaAs-AlAs and other systems, the P^{12} -modulation dominates over the relatively weak modulation of the LA sound velocity.¹¹

The phonons can be approximated by plane waves and Eq. (1) reduces to ($k_B T \gg \hbar \Omega$):

$$I[\Omega(K)] \propto |P_{q-K}|^2, \quad (2)$$

where K is the Bloch wavevector and P_k is the Fourier transform of $P(z)$. Eq. (2) establishes the link between RS and structural studies. For a given P_k , it describes scattering by phonon doublets with $K = |k \pm q|$. In periodic superlattices, $P_k \propto L \delta_{k, k_n}$ with $k_n = 2\pi n(d_A + d_B)^{-1}$; n is an integer and L is the total thickness of the structure. This leads to equally spaced doublets at $\Omega_n = c|k_n \pm q|$ (c is an average sound velocity). For non-periodic systems with $d_A = d_B = l$, like our samples, it is convenient to introduce the sequence $\{\alpha_j\}$ where $\alpha_j = 0$ if the j^{th} layer is A and $\alpha_j = 1$ if it is B. The expression for P_k in terms of the α_j 's is ($k \neq 0$):

$$P_k = (P_B - P_A)[1 - \exp(-ikl)](-ik)^{-1} S(k), \quad (3)$$

with the structure factor

$$S(k) = \sum_j \alpha_j e^{i(klj)}. \quad (4)$$

The properties of $S(k)$ for Fibonacci, Thue-Morse, and particular random sequences are discussed in the following.

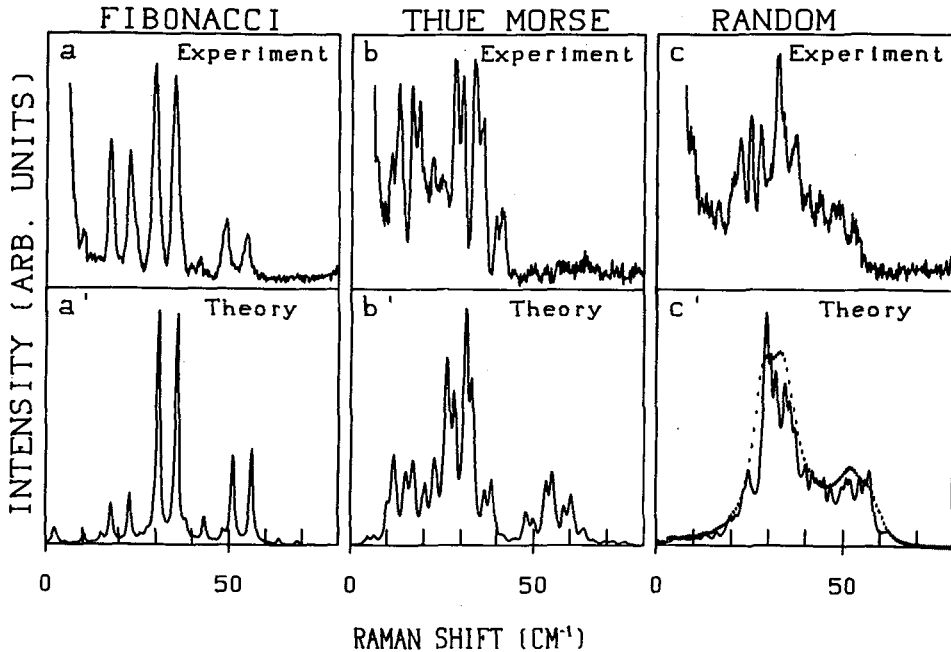


Figure 1: Comparison between measured and calculated [Eq. (1)] Raman spectra of the Fibonacci (a,a'), Thue-Morse (b,b') and random (c,c') superlattices. The dashed curve in (c') corresponds to $L = \infty$. The scattering geometry is $z(x', x')z$. $T = 300^\circ \text{K}$ and the laser energy is $\omega_L = 2.602 \text{ eV}$.

Fibonacci Superlattices. – The Fibonacci sequence can be described as the limit of generations that obey the rule $\sigma_r = \sigma_{r-1} \oplus \sigma_{r-2}$ with $\sigma_1 = \{0\}$ and $\sigma_2 = \{01\}$. This gives, e.g., $\sigma_5 = \{01001010\}$. The resulting structures are incommensurate with two basic periods that are

in a ratio given by the golden mean $\tau = (1 + \sqrt{5})/2$.^{1,9} An analytical expression for $S(k)$ can be derived using Elser's projection method.¹⁴ The result is:

$$S(k) = \frac{L}{2l\tau^2} \frac{\sin(\pi\tau^{-1}n)}{n} \delta_{k, k_{mn}}, \quad (5)$$

where $k_{mn} = 2\pi l^{-1}\tau^{-1}(m\tau - n)$. Eq. (5) gives a dense set of δ -function peaks. $S(k)$ is largest for $n = 0$, but $P(k)$ [Eq. (3)] vanishes at the corresponding k -values (this is not the case if, e.g., $d_A = 2d_B$). The next maximum is $n = 1$ leading to $k = 2\pi l^{-1}\tau^{-1}, 2\pi l^{-1}\tau^{-2}$ for $|k| < 2\pi l^{-1}$. More generally, it can be proved that the strongest peaks of P_k follow the geometric progression $k_p = 2\pi l^{-1}\tau^p$ (with integer p).^{12,13} Phonon doublets at midfrequencies given by τ^p -progressions are the characteristic signature of Raman spectra of FSL's.^{12,13}

Thue-Morse Superlattices.— Thue-Morse generations are defined by $\sigma_r = \sigma_{r-1} \oplus \sigma_r^\dagger$ where σ^\dagger is the complement of σ ; $0^\dagger = 1$ and $1^\dagger = 0$. The first four generations are $\sigma_1 = \{0\}, \sigma_2 = \{01\}, \sigma_3 = \{0110\}$ and $\sigma_4 = \{01101001\}$. The Thue-Morse sequence is *not* quasiperiodic, but automatic.¹⁰ $S(k)$ shows an infinite number of irreducible periods. For $kl = 2n\pi, S$ grows $\propto L$ as in periodic systems and, for $kl = (2n+1)\pi, S = 0$. Other values of k can be shown to satisfy the recursion relation:

$$S_r(k) = [1 - \exp(ikl2^{r-2})]S_{r-1}(k), \quad (6)$$

which is valid for $L \rightarrow \infty$. The set of k 's for which $S \neq 0$ has the property that $S \propto L^\gamma$ with $\gamma < 1$. The highest exponent is $\gamma = \ln(3)/\ln(4)$ for $kl = \pi/3, 2\pi/3$. The associated phonon doublets are expected to dominate the spectra of samples with $d_A = d_B$.

Random Superlattices.— The simplest random sequence is obtained by flipping a coin. This gives equal probabilities for A and B and zero short range correlations. For $L \rightarrow \infty$, one finds¹⁵

$$\langle |S^2(k)| \rangle = \frac{L^2}{4l^2} \delta_{k, k_n} + \frac{L}{4l} \quad (7)$$

with $k_n l = 2n\pi$. The first term on the right corresponds to the structure factor of a regular lattice of period l while the second term is the constant incoherent background. The introduction of correlations leads to incoherent scattering that depends on k . For instance, the random version of FSL's, as defined by a three state Markov process,¹⁶ gives:

$$\langle |S^2(k)| \rangle = \frac{2L}{l\tau^5} \sin^2(kl/2) [1 - 2\tau^{-3} \sin^2(kl/2) - \tau^{-2} \cos(2kl) - \tau^{-1} \cos(3kl)]^{-1}, \quad (8)$$

with maxima at $kl \simeq 2\pi\tau^{-1}, 2\pi\tau^{-2}$. In finite samples, fluctuations respect to the $L \rightarrow \infty$ limit can be quite important. An example is shown below.

Results.— In Fig. 1, we compare Raman spectra of our Fibonacci, Thue-Morse and random samples with calculations using Eq. (1). The superlattices consist of 377, 256 and 377 blocks, respectively. The random structure [Fig. (1) c,c'] is the disordered counterpart of the FSL [Fig. (1) a,a']; it was grown according to the Markov process considered above. The continuum model¹¹ describes well the positions of the Raman peaks, but not their relative intensities. This problem, also noticed in periodic systems, is most likely due to the breakdown of the local assumption for very thin layers. The FSL shows major doublets following a power-law (τ^p) behavior, in agreement with the discussion above. The spectrum is more complex for the TMSL [Fig. (1) b, b']. The strongest lines can be identified in terms of a small set of wavevectors giving the largest γ 's of $S(k)$ [Eq. (6)] (a detailed analysis of the Thue-Morse case will be presented elsewhere). The narrow features in the spectrum of the random sample are *noise* due to the finite size of the structure. This is evident in the comparison with the $L \rightarrow \infty$ limit [Eq. (8)] shown in Fig. (1) c'.

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