RAMAN SPECTROSCOPY OF ACOUSTIC PHONONS IN FIBONACCI SUPERLATTICES

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We report on resonant and non-resonant Raman scattering by acoustic phonons in Fibonacci GaAs-AlAs superlattices. Spectra off-resonance show doublets centered at frequencies that follow a power-law behavior, in good agreement with predictions of a continuum model. Resonant data show a weighted density of states revealing the expected rich structure of gaps in the phonon spectrum. It is proposed that the electronic excitation involved in the resonant process is a surface state of the superlattice.

Superlattices based on the Fibonacci sequence exhibit quasiperiodicity with two lengths that are in a ratio given by the golden mean $\tau = (1 + \sqrt{5})/2$.[1,2] The properties of these modulated materials, closely related to one-dimensional (1D) quasicrystals,[3] have been extensively discussed in recent experimental[1,2,4,5] and theoretical[6-19] studies. In this work, we report on a Raman scattering (RS) investigation of longitudinal-acoustic (LA) phonons in GaAs-AlAs Fibonacci superlattices (FSL's). The spectra provide separate information on: (i) the superlattice structure factor (as with the 'folded'-phonons of the periodic case)[20], and (ii) the frequency spectrum of LA modes, depending on the excitation energy ω_L. Specifically, the expected rich structure of gaps in the 1D density of states[8,12,15,19] is revealed by resonant data; off-resonant spectra exhibit phonon doublets that follows a power-law behavior reflecting the self-similarity[1-3,14,18] of the reciprocal lattice. The latter finding are shown to be consistent with previous x-ray measurements.[2]

The FSL used in this work has been described previously.[1,2] It corresponds to generation thirteen of the Fibonacci sequence (377 elements), and it was grown by molecular-beam epitaxy on (001) GaAs. The two building blocks are $A \equiv$ [17 Å AlAs - 42 Å GaAs] and $B \equiv$ [17 Å AlAs - 20 Å GaAs]. Raman spectra were recorded in the $z(x',x')\bar{z}$ backscattering configuration where z is normal to the layers and x' is along the [110] direction. In the acoustic region, this geometry only allows scattering by LA phonons with wavevectors parallel to [001].[20]

Figure 1 shows Raman spectra of the FSL for two laser energies ω_L. Results on other superlattices (with different parameters) are qualitatively very similar. The top and bottom spectra correspond, respectively, to resonant and non-resonant conditions; as shown by the reflectivity data in the inset, ω_L = 1.916 eV is close to a critical point for optical transitions (at ~ 1.89 eV) while ω_L = 2.409 eV falls in a relatively featureless region of the spectrum. The differences between resonant and off-resonance results are striking. The former exhibit a complex lineshape with 'dips' that are ascribed to gaps in the LA density of states.[21] Within experimental resolution, the dips do not shift when ω_L is tuned across the range 1.83 - 1.94 eV. The off-resonance trace is dominated by doublets resembling Raman results in periodic superlattices.[20] As for the latter, the doublet-splitting in the FSL is $\cong q\bar{c}$, where $\bar{c}$ is the superlattice sound velocity.[20] In contrast to the periodic case, the centers of the doublets are not equally spaced but closely follow a geometric progression with τ as the common ratio. This property, implying periodicity in a logarithmic scale, reflects the self-similarity of the Fibonacci ordering.[1-3,14,18] Other than the major doublets, the data reveal weaker lines throughout the whole spec-

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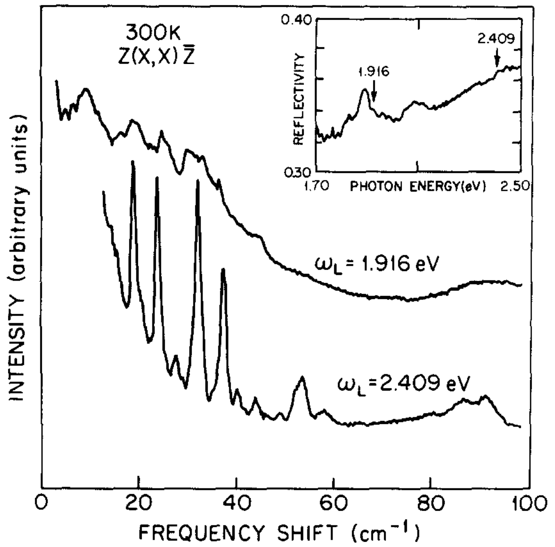

Figure 1: Typical resonant (ω_L = 1.916 eV) and non-resonant (ω_L = 2.409 eV) Raman spectra of the Fibonacci superlattice showing LA scattering. The scattering geometry is $z(x',x')\bar{z}$. Inset: normal reflectivitiy data. The scattering resonance is due to the electronic transition associated with the peak at ~1.89 eV.

tral range. This feature relates to the dense set of δ-function peaks that characterizes the structure factor of FSL's.[1-3,14,18]

The photoelastic continuum model[20] provides a bases for understanding the link between doublets and the structural properties of the FSL. Within this model, the scattering intensity is approximated by:[22]

$$I[\Omega(\kappa)] \propto \kappa^2 |P_{q-\kappa}|^2 [n(\Omega(\kappa))+1]/\Omega(\kappa), \quad (1)$$

where Ω is the frequency of the phonon, $n(\Omega)$ is the Bose factor, P_κ is the Fourier transform of the photoelastic coefficient $P^{12}(z) = P^{12}_n$ in the n-layer, and κ is the Bloch index. For a given P_k, Eq. (1) describes scattering by phonon doublets with $\kappa = |q \pm k|$; since the P_k's form a dense set,[1-3] all modes are in principle allowed. In our sample, for which $d_A/d_B \cong \tau$, the largest components of the modulation (strongest features in the x-ray pattern)[2] correspond to $k_p = 2\pi d^{-1}\tau^p$ with integer p ($d = \tau d_A + d_B$; d_A and d_B are the thicknesses of building blocks A and B).[1-3,14] This leads to doublets at $\Omega \cong \bar{c}\,|q \pm 2\pi d^{-1}\tau^p|$ accounting for the origin of the geometric progression. Calculations using the full expression for $I[\Omega(\kappa)]$[20] support the above approximation. The exact results exhibit doublets only slightly shifted from the frequencies predicted by Eq. (1).[22]

A comparison between our resonant data and a calculation of $\Omega(\kappa)$ indicates that the positions of the 'dips' correlate with major gaps in the 1D phonon spectrum.[21] Based on this correlation, the resonant scattering is ascribed to a weighted density of states of [001] LA modes. The situation here differs from the periodic counterpart in several respects. For the latter, resonances lead primarily to changes in the relative intensities of the doublets.[20] The development of asymmetric lineshapes and interference-type behavior, as reported in Ref. 20, do not relate to our case because these effects are strongly dependent on ω_L. Wavevector non-conserving scattering by zone-edge phonons[20,23] is also unrelated; both LA and transverse-acoustic modes are involved and the doublets remain as the dominant spectral features.

The origin of the resonant scattering is not well understood. The quasi-periodicity of FSL's allows scattering by all [001] phonons, but such a general statement does not account for the differences with the non-resonant data. An interesting possibility is that the relevant electronic state is an <u>intrinsic</u> gap excitation localized at the surface. As discussed in Ref. 12, FSL's may contain as many surface states as states of the bulk. Calculations of the electronic spectrum and further experimental work are being pursued to test this idea.

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