Confined optical phonons in piezoelectric [311] GaInAs/AlAs superlattices probed by Raman scattering

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Abstract. We present a resonant Raman scattering investigation of optical phonons in strained Ga0.85In0.15As/AlAs superlattices grown along [001] and [311], the latter presenting built-in piezoelectric fields. The Raman configuration required to separate the deformation potential and Fröhlich contributions to the scattering are described and evidenced in resonant and non-resonant experiments.

1. Motivation
Sound manipulation in the THz range is interesting for the control of optical and electronic properties. The link between sound and optoelectronics is given by the electron-phonon interaction [1]. The latter proceeds through relatively weak mechanisms: deformation potential and, in materials which lack inversion symmetry, the piezoelectric coupling [1]. Photons and acoustic phonons can be confined in resonant cavities providing an alternative strategy for enhancing the coupling between light, charge and sound [2, 3, 4]. The intrinsic mechanism in these structures remains, however, the same. Piezoelectric heterostructures grown under internal strain in low-symmetry directions present built-in electric fields. It is the case of GaInAs/AlAs multilayers grown along [n11] directions (electric fields of the order of $10^5$ V/cm) [5] and [0001] GaN/GaN heterostructures (up to $10^6$ V/cm) [6]. A giant phonon generation and detection efficiency in the presence of built-in electric fields has been reported for the latter structures [7]. The modulation of these huge built-in electric fields could lead, in fact, to an additional mechanism of electron-acoustic phonon interaction, conceptually connected to the Fröhlich coupling operative for optical phonons, and potentially important in strained piezoelectric heterostructures [8, 9, 10].

To evidence this mechanism we study [001] and [311] GaInAs/AlAs multilayers (without and with built-in fields respectively), for which high quality samples can be grown by molecular beam epitaxy (MBE) [9]. We concentrate here in the optical phonon Raman spectra of these samples. In heterostructures with built-in electric fields the selection rules for scattering by longitudinal
optical (LO) phonons are modified, displaying in addition a non-trivial dependence of Raman intensity with laser power [8]. Raman scattering can thus be used as a tool to probe the presence of built-in electric fields, and to understand its role in the process of inelastic scattering of light. The aim of this short communication is to describe an experimental configuration able to separate the deformation potential and Fröhlich interactions in strained superlattices grown along [311]. Resonant and non-resonant optical phonon Raman spectra are presented and analyzed in terms of the Raman tensors and selection rules applicable in the proposed scattering geometry.

2. Selection rules

The Raman cross-section is given, in terms of the Raman tensor $R$, by $d\sigma_R \propto |\hat{e}_s \cdot R \hat{e}_i|^2$. $R$ describes the specific vibrational mode, while $\hat{e}_i$ and $\hat{e}_s$ stand for the incident and scattered light polarizations, respectively. The Raman tensors corresponding to the deformation potential interaction for incidence along [001] can be found, e.g., in Ref. [1]. For the less common [311] direction, on the other hand, they are given by [12]:

$$ R_{[311]}^{[01\bar{1}]} = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & -d & d \\ -d & 0 & 0 \\ d & 0 & 0 \end{pmatrix}, \quad R_{[311]}^{[23\bar{3}]} = \frac{1}{\sqrt{22}} \begin{pmatrix} 0 & 3d & 3d \\ 3d & 0 & -2d \\ -2d & 0 & 0 \end{pmatrix}, \quad R_{[311]}^{[31\bar{1}]} = \frac{1}{\sqrt{11}} \begin{pmatrix} 0 & d & d \\ d & 0 & 3d \\ d & 3d & 0 \end{pmatrix} \quad (1) $$

Here the sub-indices represents the phonon polarization. Along [001] there are two transversal (TO) and one longitudinal (LO) optical modes. For [311] there is one pure transversal (TO, [01\bar{1}]), one quasi-transversal (QTO, mainly in [23\bar{3}]) and one quasi-longitudinal mode (QLO, mainly in [311]). For the LO and QLO phonons there is also a Fröhlich contribution, which is represented by a diagonal Raman tensor [11]

$$ R_F = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & a \end{pmatrix}. \quad (2) $$

To separate the Fröhlich ($a$) and deformation potential ($d$) contributions specific incident and scattered light polarizations can be chosen such that, for a certain $y = \cos(\theta)x' + \sin(\theta)y'$ polarization, $y \cdot R_{LO}y = 0$. $x', y'$ are the crystal axes used to define Equation 1. For [001], this gives the crystallographic directions $x^{[001]} = (100), y^{[001]} = (010)$. For [311], on the other hand, we obtain

$$ x^{[311]} = (10, -15 + 11\sqrt{5}, -15 - 11\sqrt{5}) $$

$$ y^{[311]} = (-2, 3 + \sqrt{5}, 3 - \sqrt{5}) \quad (3) $$

which correspond to an angle of $\theta \approx 56$ respect to $x', y'$. The Raman cross sections for these polarizations are summarized and compared to [001] in Table 1. It follows that only the Fröhlich (deformation potential) interaction contributes in the parallel yy (crossed xy) configuration.

Though the presence of the heterostructure does not modify in general the bulk selection rules discussed above, it introduces new selection rules related to the symmetry of the vibrations with respect to the center of the layers [12]. The confined optical modes can be divided into even $B$ ($1^{st}, 3^{rd}, ...$) and odd $A$ ($2^{nd}, 4^{th}, ...$) confined modes. It turns out that only even $B$ modes are allowed through deformation potential, while only odd $A$ modes contribute mediated by the Fröhlich interaction [12].
3. Experimental results
Details of the samples can be found in Ref.[9]. They are 21Å/78Å(23Å/84Å) Ga$_{0.85}$In$_{0.15}$As/AlAs superlattices (SLs) grown along [001]/[311]. The AlAs layers are only weakly strained (∼ 0.05%), while Ga$_{0.85}$In$_{0.15}$As is under a large (∼ 1%) compressive strain. The built-in electric fields in the Ga$_{0.85}$In$_{0.15}$As layers can be estimated for the [311] structure to be ∼ 4.6 × 10$^4$V/cm [5].

Figure 1 shows the GaAs-like optical phonon Raman spectra ($T = 80$ K) taken with 5mW of laser power fociZalized into a ∼ 30µm spot. The out-of-resonance spectra (bottom, 2.54 eV) for both parallel and cross polarizations show clearly the selection rules stated in the previous section. For the cross (xy) configuration, only the first and third confined LO (QLO for [311]) modes are seen which can be related to the deformation potential interaction. For parallel (yy) configuration, on the other hand, only the even order confined LO (QLO) modes are seen related to the Fröhlich interaction. This applies to both [001] and [311] growth directions, implying that the axes defined by Equation 3 allow, indeed, the separation of the two scattering mechanisms.

Figure 1 also displays the corresponding near-resonance (1.83 eV) spectra (the fundamental

| Configuration | [001] | [311] |
|---------------|-------|-------|
| $z(x,x)z$     | $0$   | $a$   |
| $z(x,y)z$     | $0$   | $d$   |
| $z(y,y)z$     | $0$   | $a$   |

Table 1. Raman cross sections (Porto’s notation) for incidence along [001] and [311]

Figure 1. Optical GaAs-like phonon Raman spectra out-of (2.54eV) and near-resonance (1.83eV) with the fundamental electronic transition of the superlattice. LO, QLO, QTO, and IF stand for longitudinal, quasi-longitudinal, quasi-transversal, and interface optic phonon modes, respectively. The inset shows the confined modes peaks position (symbols) and the optical dispersion relation (continuous curve).
gap of the SL’s is around $\sim 1.7\text{eV}$). The deformation potential contribution (odd order modes) can be seen to contribute in crossed polarizations in agreement with the above consideration. In contrast with the out-of-resonance case, however, it is observed that the second confined LO (QLO) mode dominates the spectra both in parallel and cross polarizations. This can be understood by the fact that the Fröhlich interaction requires a finite phonon wavevector and thus is strongly favored by the presence of impurities and defects, specially under resonant conditions [13, 14]. For each sample we show also in the inset to Figure 1 the peak positions associated to the confined LO and QLO modes (full squares), as a function of the corresponding effective momentum. We also show in this graphs the position of peaks associated to the coupling of interface phonons (IF) and higher order confined modes (open circles) which correspond approximately to the even order confined optical vibrations [15]. The peak positions closely map the bulk GaAs optical dispersion for each growth direction (shown with a continuous curve), further confirming the mode assignment.

Note the presence of low-energy shoulders in the QLO peaks of the [311] sample, marked by arrows in the near-resonance spectrum. High quality samples grown in the [311] direction develop a corrugation of the surfaces [16]. This corrugation induces an in-plane folding of the phonon bands allowing the observation of optical phonons with a finite in-plane wavevector, separated from the main peak by a certain energy given by the dispersion relationship and the effective quasi-momentum. Shields et al. [16] reported this separation for unstrained GaAs/AlAs SLs grown along [311] to be $\sim 1\text{cm}^{-1}$. We found a separation of around $\sim 2.8\text{cm}^{-1}$: the difference could be related to a different period for the corrugation produced by the internal strain.

4. Conclusions
We have described the Raman geometry appropriate to separate the deformation potential and Fröhlich terms contributing to the electron-optical-phonon interaction in SLs grown along [311]. The corresponding selection rules have been verified through the observation of confined optical phonons of different parity in non-resonant and resonant Raman scattering experiments. These results are relevant to study the effect of built-in electric fields on the Raman scattering process in strained piezoelectric superlattices.

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