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Magnetic-field-enhanced Raman scattering by interface phonons in p-type modulation-doped multiple quantum wells

J Kraus†, G Weimann‡ and K Panzlaff§||

† Physikalisches Institut der Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany
‡ Walter-Schottky-Institut der TU München, Am Coulombwall, D-85748 Garching, Germany
§ Abteilung Optoelektronik der Universität Ulm, Oberer Eselsberg, D-89069 Ulm, Germany

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Abstract. We have investigated the coupling of hole intersubband transitions and phonons in p-type modulation-doped GaAs–$Al_xGa_{1-x}As$ multiple quantum wells by means of resonance Raman spectroscopy. Magnetic fields with flux densities up to $B = 14$ T were applied, oriented parallel to the growth direction. For $B = 0$ T the spectra can be interpreted in terms of a Fano interference of the zone-centre quantum well LO phonon with a quasi-continuum of hole transitions between the lowest and the second excited heavy-hole subband. For $B > 4$ T we observe the coupling of a hole intersubband transition with interface phonons. The character of the latter modes is deduced from a comparison of the excitation energies with values calculated within a dielectric continuum model as well as from their behaviour under photoexcitation. Our measurements indicate an interaction of the Fröhlich type, where the strength of the coupling seems to increase with the in-plane wavevector of the excitations. The ionized impurities in the barriers dominate the relaxation of wavevector conservation in the scattering process.

1. Introduction

The most typical application of Raman spectroscopy in solid state physics concerns the investigation of zone-centre optical phonons. Strictly speaking this implies complete translational invariance of an ideal crystal. In semiconductor multiple quantum wells (MQWs) and superlattices the translational symmetry is intentionally reduced in one dimension, the growth direction of the sample, and normally unintentionally reduced in lateral directions due to imperfections of the structures (interface roughnesses or impurities). The latter give rise to wavevector-non-conserving light scattering in such structures, especially to the appearance of interface phonons at finite in-plane wavevector $q_\parallel$ [1] or to disorder-induced scattering from modes of the whole folded acoustic phonon dispersion [2], even under conditions of vanishing $q_\parallel$-transfer by the scattered photons. Magnetic fields perpendicular to the layers lead to a strong enhancement of the signal strength of these wavevector-non-conserving light scattering processes [3, 4].

We report here on the observation of a magnetic-field-enhanced Raman signal from interface phonons coupled to hole intersubband transitions in p-type modulation-doped MQWs. Previous investigations—already described in [5]—could be completed by measurements at higher magnetic fields and under conditions of a photo-induced variation

|| Present address: ANT Nachrichtentechnik GmbH, Gerberstraße 33, D-71522 Backnang, Germany.

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of the hole density in the quantum wells; we have arrived now at new interpretations and at a better understanding of the experimental observations.

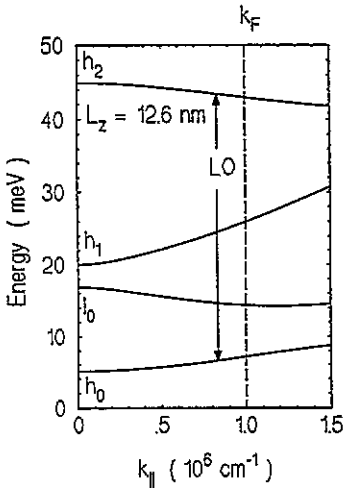

Figure 1. Calculated dispersions of valence subbands for a well width $L_z = 12.6$ nm ($x = 0.43$). Heavy- and light-hole subbands are denoted according to their character at $k_\parallel = 0$. The limits of the single-particle quasi-continuum of (h_0–h_2) transitions—vertical in the $k_\parallel$-space—are defined by the subband distances at $k_\parallel = 0$ and $k_\parallel = k_F$. At $k_\parallel = 0.83 \times 10^6$ cm^{-1} the subband distance coincides with the energy of the quantum well LO phonon.

Comparing the behaviour of intersubband transitions in n- and p-type doped MQWs makes apparent some very striking differences, which are mainly caused by the strongly nonparabolic dispersions of the valence subbands (figure 1). The dependence of the energy differences between valence subbands on the in-plane wavevector $k_\parallel$ has the consequence that the spectra of single-particle–hole intersubband excitations have finite widths even under experimental conditions, which realize vanishing in-plane wavevector transfer $q_\parallel$. The widths of the single-particle continua of hole intersubband transitions vertical in $k_\parallel$-space are determined by the variation of the subband distances between $k_\parallel = 0$ and $k_\parallel = k_F$ (k_F: is the Fermi wavevector). The wavevector dependence of the subband distance becomes evident in a conspicuous difference of the linewidths of transitions between valence subbands of a curvature with the same or the opposite sign [7] and can be experimentally investigated in more detail by means of resonance Raman spectroscopy [6]. The finite width of the single-particle spectrum subsequently results in a strong Landau damping of collective excitations and in a dominating single-particle character of hole intersubband transitions [8].

The energy differences between conduction subbands are nearly independent of $k_\parallel$, and transitions reveal a discrete spectrum for $q_\parallel = 0$. Collective effects are very evident in n-type samples from the energy difference between charge density excitations (CDE) and spin density excitations (SDE) observed in configurations with the polarizations of the incoming and outgoing photons parallel (polarized spectrum, CDE) and perpendicular to each other (depolarized spectrum, SDE), respectively [9]. This energy difference is caused by direct Coulomb interaction and reveals the macroscopic electric fields associated with intersubband transitions observed in the polarized spectrum. This so-called depolarization field gives rise to the dominant mechanism of the coupling of collective intersubband CDEs to the longitudinal optical (LO) phonon of the well material [9].

The very small strength of the interaction between hole intersubband excitations and

phonons in p-type samples, already mentioned in [5] and demonstrated in more detail in the present paper, is completely consistent with the above-mentioned strongly suppressed collective character of these excitations. The finite widths of the hole intersubband transition spectra result in a type of coupling which can be described in terms of a Fano interference [10] between the discrete phonon line and the quasi-continuum of hole excitations (compare, e.g., [11, 12]).

Under application of a strong magnetic field, oriented perpendicular to the layers, the in-plane motion of the carriers is quantized and the single-particle continua of hole intersubband transitions are replaced by transitions between discrete Landau levels. The mixing of heavy- and light-hole subbands, which determines the nonparabolicity of the valence subband dispersion at $B = 0$ T, results also in a very complicated Landau level scheme [13] in comparison to the nearly linear field dependencies of the quantized conduction subband states. With increasing flux density B the energy differences between intersubband excitations observed in the polarized and depolarized configurations decrease [5], and vanish completely at fields where the split components are well separated.

The possibility of tuning the intersubband transition energies by varying B gives us the ability to perform a very detailed investigation of the interaction with phonons, where, in contrast to the zero-field case, consideration of the coupling of two discrete transitions can be assumed to be adequate. The energies of the unperturbed excitations and the coupling strength can be deduced. This was done in the evaluation of our previous results [5] and was continued in the interpretation of spectra under application of higher magnetic fields. Three features are most striking in this context: a difference in the frequency of the unperturbed coupling phonon compared to that of the zone-centre LO phonon of the well material; the existence of a (nearly) non-coupling quantum well phonon; and again the small interaction strength—compared to that of n-type samples—which, however, seems to increase with the in-plane wavevector of the coupling excitations. We interpret the non-coupling quantum well phonon, whose energy coincides with the bulk LO phonon energy of GaAs, as a zone-centre phonon. The energy of the unperturbed coupling phonon suggests interpretation as an interface phonon at finite $k_\parallel$.

Some of our conclusions concerning the character of the coupling and the non-coupling phonon are in contrast to our earlier interpretations given in [5, 14]; however, they seem to be well verified by our new measurements at high magnetic fields as well as by experiments where the relative contributions of wavevector-conserving and wavevector-non-conserving scattering processes could be varied by illumination of the sample with photons of an energy above the band gap of the barrier material.

2. Experimental details

Our experiments were performed in backscattering configuration under resonant excitation using a tunable cw dye laser (power density $P_L < 200$ W cm^{-2}). The scattering geometry is described in the standard notation $z(e_i, e_s)\bar{z}$, where e_i and e_s are the linear (x, y) or circular $(+, -)$ polarization of the incident light. The samples were immersed in superfluid helium within a superconducting split coil ($B \leqslant 8$ T) or a solenoid magnet ($B \leqslant 14$ T), respectively. The use of the latter type of magnet system required experimental access by fibre optics, with the disadvantages of an increased continuous background underlying the spectra and a loss of information from the light polarization. The scattered light was dispersed with a triple spectrograph (DILOR XY) and detected by an intensified diode array. Our experiments were performed on two nearly identical p-type modulation-doped MQWs with 10 periods of GaAs wells of nominal widths $L_z = 10.0$ nm, separated by spacers of a

thickness $L_s = 24.5$ nm from the Be-doped layers in the centre of the $Ga_{1-x}Al_xAs$ barriers. Our two samples differ in the widths L_d of the doped layers, the Be concentrations n_{Be} and the x-values of the barrier materials (sample A: $L_d = 3.5$ nm, $n_{Be} = 1.25 \times 10^{18}$ cm^{-3}, $x = 0.43$; sample B: $L_d = 15.0$ nm, $n_{Be} = 0.30 \times 10^{18}$ cm^{-3}, $x = 0.32$). Most of the results described in the following were observed on sample A, the sample with higher interface quality as regards structure roughness, as verified by Raman spectroscopy [15]. For this sample a hole density of $p = 1.6 \times 10^{11}$ cm^{-2} was established from Raman spectroscopic data [8] performed under nearly the same experimental conditions (temperature, laser power densities) as realized in our present investigations. A nearly identical carrier concentration can be assumed to be realized in sample B, because of the closely corresponding total number of dopant atoms in the barrier centre layer.

Sample illumination with photons of an energy above the barrier band gap, which allows the variation of the hole concentration in the quantum wells, was performed using an Ar^+ laser. Both simultaneous illumination and illumination alternating in time with the detection of the Raman signal (delay times $\Delta t \geqslant 0.3$ ms) were experimentally realized. Investigations making use of the latter experimental set-up were mainly performed on sample B, where the slight differences concerning the doped barrier regions turned out to be advantageous. More details of the set-up will be published elsewhere [16].

3. Results and discussion

Because the features of the single-particle spectra of hole intersubband transitions differ considerably for zero and high magnetic fields, the two cases are discussed separately in subsections 3.1 and 3.2. We first describe our observations at $B = 0$ T; subsequently we discuss the evidence of magnetic-field-enhanced scattering by interface phonons. Our interpretation of the modes under discussion as interface phonons is confirmed by Raman spectroscopic investigations under photoexcitation (subsection 3.3).

3.1. $B = 0$ T

According to subband calculations performed in an anisotropic envelope function approximation for our sample geometry (see [6, 8] for more details), the energy of the zone-centre LO phonon of the well material GaAs (36.7 meV) lies within the continuum of single-particle transitions between the lowest (h_0) and the second excited heavy-hole subband (h_2) for well widths $L_z \approx 13.0$ nm (figure 1). Effects of an interference of the LO phonon line with the single-particle continuum of the intersubband transitions can be studied under conditions of a resonantly enhanced (h_0–h_2) Raman signal only, i.e. with the energy of the incoming laser photons in resonance with the energy difference between the second excited valence and conduction subbands h_2 and c_2, respectively. Unfortunately the Raman signal is superimposed upon by a strong luminescence transition $c_2 \rightarrow h_0$ (figure 2). This prevents a direct observation of the intersubband excitation, but the upper and lower limit of the single-particle quasi-continuum of (h_0–h_2) transitions, defined by the subband distances at $k_\parallel = 0$ and $k_\parallel = k_F$ can be deduced from Raman spectra under photoinduced variation of the hole density in the well and from experimental data at high magnetic fields.

As shown in [17] for n-type samples, the actual carrier densities in the quantum wells of modulation-doped samples change with the laser power density: at increasing power density the carrier concentration in the well decreases. Raman spectra observed with high and low laser power differ in all features (e.g., collective effects [17]) which depend on the carrier concentration. In our case the difference of two spectra normalized to the

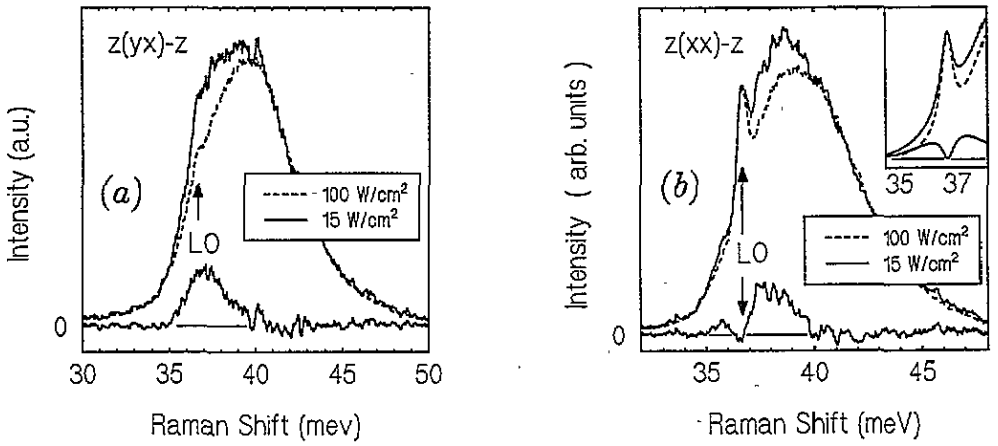

Figure 2. (a) Spectra of sample A measured in the allowed configuration $z(yx)\bar{z}$ with laser energy 1.747 eV ($B = 0$ T). The intensities are normalized to the respective laser power densities. The Raman signal of the quantum well phonon (LO) and of the quasi-continuum of (h_0–h_2) transitions is superimposed by luminescence ($c_2 \rightarrow h_0$), which prevents a direct observation of the intersubband excitation. The difference spectrum (lowest spectrum) displays the changes of the Raman signal of intersubband transitions caused by the photoinduced decrease of the hole density in the well at high laser power density. (b) Corresponding spectra observed in the forbidden configuration $z(xx)\bar{z}$. The difference spectrum shows a dip close to the energy of the GaAs LO phonon. The inset displays the result of a simulation according to equation (4).

respective laser power densities (figure 2(a)) should display the changes in the Raman signal of the (h_0–h_2) transitions, mainly caused by the shift of k_F (figure 1), while the underlying (c_2–h_0) luminescence background should be less strongly affected by the variation of the hole density. From the difference spectrum shown in figure 2(a) the width of the single-particle spectrum of 4–5 meV can be estimated, where the onset at about 35.5 meV should correspond to the (h_0–h_2) subband distance at $k_\parallel = k_F$.

A nearly exact value of the (h_0–h_2) subband distance at the zone centre can be deduced from spectra at high magnetic fields. Landau levels with quantum numbers $N = -2$ (in the notation of Altarelli [18]) do not intermix with neighbouring light-hole levels [13]. The energy difference between (-2) Landau levels is nearly independent of the magnetic flux density and can be identified with the separation of the corresponding heavy-hole subbands at $k_\parallel = 0$. In Raman spectra (not shown here) we could observe such a component of the (h_0–h_2) excitation at a Raman shift of 40.0 meV at $B = 6.5$ T [5] in the configuration $z(+,+)\bar{z}$.

A (h_0–h_2) subband distance of 40.0 meV at $k_\parallel = 0$ corresponds to a well width of $L_z = 12.6$ nm, which is significantly larger than the nominal value (10.0 nm), and also larger than the well width (11.0 nm) deduced from the evaluation of the (h_0–h_1) transition performed in [8]. The increase in the values of the well width L_z deduced from the observed transitions with the energy of the participating subbands can be interpreted as an indication of a continuous variation of the Al concentration x at the GaAs–$Al_xGa_{1-x}As$ interfaces rather than an abrupt transition. Possible consequences of component roughness of interfaces for in-plane phonon modes will be discussed at the end of the next section.

The estimated hole density of $p = 1.6 \times 10^{11}$ cm^{-2} determines the Fermi wavevector (1.0×10^6 cm^{-1}). The calculated (h_0–h_2) subband distance at $k_\parallel = k_F$ (35.8 meV, figure 1) agrees well with the low-energy onset of the difference spectrum in figure 2(a). A (h_0–h_2)

subband distance, which corresponds to the energy of the GaAs LO phonon (36.7 meV) is realized at $k_{\parallel} = 0.83 \times 10^6$ cm^{-1} (figure 1).

Effects of an interference of the discrete phonon line with the quasi-continuum of hole excitations could only be observed in the configuration $z(xx)\bar{z}$ (figure 2(b)). The difference signal of the two spectra, again measured under variation of the laser power density (figure 2(b)), shows a dip very close to the energy of the zone-centre LO phonon of bulk GaAs (minimum position: 36.5 meV). This behaviour can be well interpreted by means of Fano's theory [10] applied to inelastic light scattering experiments. The Raman spectrum can be described by [19, 12]

$$I(\eta, \tilde{q}) = I_e \frac{(\tilde{q} + \eta)^2}{1 + \eta^2} \tag{1}$$

where

$$\eta = \hbar \frac{\omega - \Omega_p - \Delta\Omega}{\Gamma} \tag{2}$$

and

$$\tilde{q} \propto T_p / T_e. \tag{3}$$

Ω_p represents the frequency of the discrete phonon line, $\Delta\Omega$ the renormalization due to interaction with the quasi-continuum of hole excitations (the real part of the phonon self-energy), and Γ the linewidth. The so called lineshape or asymmetry parameter $\tilde{q}$ is proportional to the ratio of the scattering amplitudes of the phonon (T_p) and the electronic transitions ($T_e = T_e(\hbar\omega)$). $I_e = I_e(\hbar\omega)$ corresponds to the spectrum of (h_0–h_2) intersubband transitions, which is affected by the photoinduced changes of the hole density. If the energy dependence of $I_e(\hbar\omega)$ in a range Γ about $\Omega_p + \Delta\Omega$ is weak, $\tilde{q}$ can be assumed to be constant ($T_e \approx T_e(\Omega_p + \Delta\Omega)$). Strongly asymmetric lineshapes can be observed if $\tilde{q}$ is not purely imaginary and $|\tilde{q}|$ is close to unity.

Difference spectra of the type displayed in figure 2(b) can be simply simulated under assumption of a broad quasi-continuum $I_e(\hbar\omega)$ of arbitrary shape, whose strength is reduced to a fraction a:

$$\Delta I(\hbar\omega) = I_e(\hbar\omega) \frac{(\tilde{q} + \eta)^2 - a(\tilde{q}' + \eta)^2}{1 + \eta^2} \tag{4}$$

with

$$\tilde{q}^2 / \tilde{q}'^2 = a. \tag{5}$$

For $\eta = 0$ one finds a zero crossing in the calculated difference spectrum which coincides with the minimum position of $\Delta I(\hbar\omega)$ if $\tilde{q}$ is purely imaginary (inset of figure 2(b)), symmetric lineshape). The sign and absolute value of $\Delta\Omega$ are determined by the difference in the number of hole transitions of an energy above and below $\hbar\Omega_p$ and by their coupling strengths. The zero-crossing position in the measured difference spectrum agrees within the experimental resolution with the energy of the zone-centre LO phonon $\hbar\Omega_p$. The nearly vanishing value of the phonon self-energy $\Delta\Omega$ and especially the independence of $\Delta\Omega$ on photoinduced changes of the spectrum of hole transitions indicate a very small coupling strength. The interaction mechanism can be deduced from the polarization selection rules being of Fröhlich type.

The slightly asymmetric lineshape of the LO phonon, especially at low laser power density, and the small shift of the minimum position of the dip in the difference spectrum compared to $\hbar\Omega_p$ is a consequence of a small real component of the lineshape parameter $\tilde{q}$.

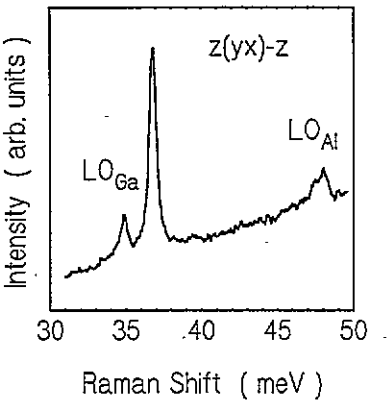

Figure 3. Spectrum of sample A observed in the allowed configuration $z(yx)\bar{z}$ with laser energy 1.778 eV. The peaks labelled LO_{Ga} and LO_{Al} are the GaAs-like and AlAs-like LO phonons of the barrier material, respectively.

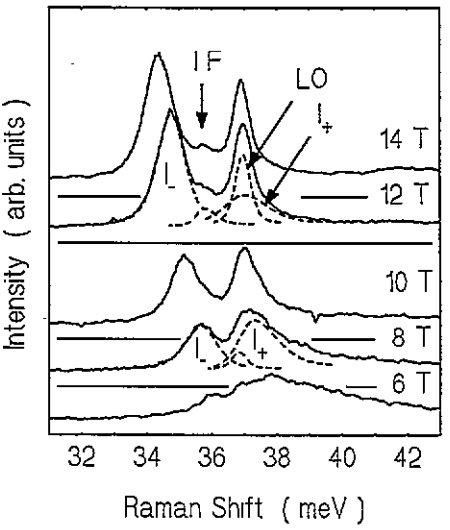

Figure 4. Raman spectra of the coupled-phonon intersubband excitations (I_+, I_-) at different flux densities B, observed with laser energy 1.750 eV. The light polarization is undefined because of the use of fibre optics. The peak labelled IF is interpreted as an interface phonon. The dashed lines indicate the different contributions to the spectra, where for $B \geqslant 12$ T the I_+ mode and the signal of a non-coupling quantum well LO phonon (labelled LO) are hardly separable.

With the dye laser energy far from the case of resonant light scattering by (h_0–h_2) transitions the phonon lineshape becomes symmetric ($T_e \to 0$, $|\tilde{q}| \to \infty$).

Besides the Raman signal of the quantum well LO phonon two additional vibrational modes can be observed in allowed configuration. Their signal strength increases when the energy of the dye laser approaches the barrier band gap. These modes can be interpreted as the GaAs-like and AlAs-like LO phonons of the barrier material $Al_xGa_{1-x}As$ (figure 3). The measured line positions (34.8 meV and 48.0 meV, respectively) are in good agreement with energies expected from the nominal x-value of sample A [20]. The width and the asymmetric lineshape of the AlAs-like mode can be seen as an indication for contributions of light scattering by AlAs-like interface phonons [1, 3], while the GaAs-like mode seems to be a pure zone-centre excitation.

3.2. $B > 0$ T

Two components of coupled modes (labelled I_+, I_- in figure 4) can be clearly resolved at flux densities $B > 4$ T, where the in-plane dispersion of the valence subbands is quantized into more or less well separated states [13]. Besides these coupled-phonon intersubband excitations we observe a Raman signal from an obviously almost non-coupling LO phonon, whose energy exactly coincides with the bulk value of GaAs. From the resonance behaviour (figure 5(b) in [5]) this line can also be identified as an excitation of the quantum well. Finally, for fields $B \geqslant 11$ T a further line is observed at a Raman shift of 35.6 meV

(labelled IF in figure 4), which will be interpreted as an interface phonon. The latter excitation does not show any indication of an interaction with the intersubband transition. The spectra of the coupled excitations can be measured in the configuration $z(-,-)\bar{z}$ [5]. The Landau quantum numbers of the initial and final states of the intersubband transition are $N = 1$, the quantum number of the only occupied state at high flux densities ($B > 8$ T) [13]. From the polarization selection rules, Fröhlich interaction must again be assumed to be responsible for the coupling [21], in agreement with the measurements at $B = 0$ T.

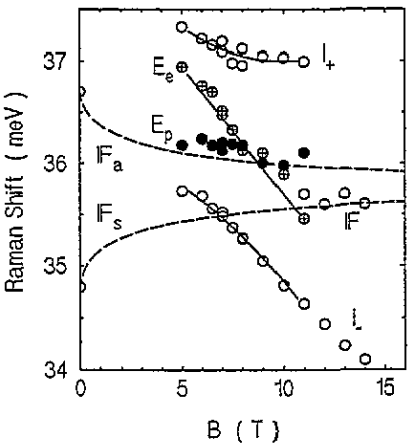

Figure 5. Peak positions of Raman excitations as a function of the magnetic flux density B (○). The energies $E_p = \hbar\Omega_p$ (●) and E_e (⊕) of the unperturbed excitations are obtained by a fit of equation (6) to the spectra of the coupled modes (I_+, I_-). The solid lines are guides to the eye. The dashed lines are the calculated dispersions of an antisymmetric (IF_a) and a symmetric branch (IF_s) of interface phonons [23], displayed as a function of B by identification of $k_\parallel$ with the inverse of the magnetic length $l_0(B)$. They describe well the positions of the coupling phonon E_p and of the line labelled IF in figure 4.

In figure 5 all observed peak positions—with the exception of the field-independent position of the non-coupling GaAs LO phonon line—are displayed as a function of the magnetic flux density B as open circles.

As described in [5] we have analysed the spectra of the coupled modes in terms of the Green function formalism, where the Raman spectrum $I(\hbar\omega)$ of two interacting excitations is proportional to [22]

$$I(\hbar\omega) \propto \mathrm{Im}\left(\sum_{i,j=e,p} T_i \langle i|G(\hbar\omega)|j\rangle T_j\right) \tag{6}$$

with the Green function operator

$$G(\hbar\omega) = \begin{pmatrix} \tilde{E}_p - \hbar\omega & V \\ V^* & \tilde{E}_e - \hbar\omega \end{pmatrix}^{-1}. \tag{7}$$

The energies $\tilde{E}_i$ are assumed to be complex: $\tilde{E}_i = \hbar\Omega_i + \mathrm{i}\Gamma_i$. V is the matrix element for the decay of the unperturbed excitation i of frequency Ω_i and damping Γ_i into excitation j. The T_i are the scattering amplitudes of the unperturbed excitations.

For each magnetic field we have evaluated a large number of spectra measured with the dye laser energies close to the (c_2–h_2) subband distance. The scattering amplitudes T_e and T_p were found to vary with laser energy as a consequence of the resonance behaviour

of the scattering efficiency [5]. The hole intersubband transition energy $\hbar\Omega_e$ varies as a result of slight fluctuations in the well widths of the order of about one monolayer [15]. The energies $\hbar\Omega_e(B)$ given in figure 5 are results of fits of equations (6) and (7) to Raman spectra measured at the respective resonance maximum. The coupling strength $|V|$ and the phonon energy $\hbar\Omega_p$ were found to be independent of the dye laser photon energies, as expected. At the crossing point of $\hbar\Omega_p$ and $\hbar\Omega_e(B)$ at about $B = 8$ T (figure 5) our fits yield a coupling strength $|V| = 0.9$ meV, which seems to increase slightly with B ($\mathrm{d}|V|/\mathrm{d}B \leqslant 0.08\ \mathrm{meV\,T^{-1}}$). Finally we found that $\hbar\Omega_p$ does not coincide with the energy of the zone-centre GaAs LO phonon. $\hbar\Omega_p$ is slightly shifted by up to about -0.7 meV (filled circles in figure 5).

We interpret the coupling phonon $\hbar\Omega_p$ and the peak at 35.6 meV, observed for $B \geqslant 11$ T, as the magnetic-field-enhanced Raman signal from interface phonons at finite in-plane wavevector $k_\parallel$, which evolve out of the GaAs quantum well LO phonon and the GaAs-like LO phonon of the barrier material at $k_\parallel = 0$, respectively. The in-plane dispersion of interface phonons can be calculated from a dielectric continuum model [23]. Six branches of interface phonon dispersion curves correspond to the zone-centre TO and LO frequencies of the GaAs-like and AlAs-like vibrational modes of the well and barrier material. With respect to the centre of the GaAs-layer the macroscopic potentials of the modes are symmetric or antisymmetric. We have calculated the interface phonon dispersions for a well width of $L_z = 10.0$ nm (the nominal well width of our sample) in the approximation of infinite barrier widths.

Following [3] we assume that the extent of the relaxation of the kinematic selection rule is determined by the inverse of the magnetic length $l_0 = (\hbar c/eB)^{1/2}$. Interface modes at a maximum in-plane wavevector equal to l_0^{-1} should be able to participate in the scattering process. In figure 5 the dashed lines give the interface phonon energies at $k_\parallel = l_0^{-1}$ displayed as a function of B. The upper branch is an antisymmetric mode (IF_a), which evolves out of the GaAs quantum well LO phonon, the lower branch is a symmetric mode (IF_s), which starts at the energy of the GaAs-like LO phonon of the barrier at $k_\parallel = 0$.

The energies of the coupling phonon $\hbar\Omega_p$ and the peak positions of the line labelled IF in figure 4 approximately coincide with the calculated positions of the antisymmetric and symmetric interface phonons, respectively. This supports the interpretation of the origin of these modes given above. On the other hand, the mechanism of interaction (Fröhlich-type) would predict a coupling of the intersubband excitation to the symmetric mode, and not to the antisymmetric one, at least in the limit $k_\parallel \to 0$. This is in contrast to our experimental result, where we can observe an interaction with interface phonons of the antisymmetric branch only. This contradiction need not violate our interpretation of the character of the coupling phonon. The consideration of non-abrupt interfaces could be supposed as a possible solution of the problem mentioned here. Indications for a considerable component roughness of the interfaces in our sample exist, as described in the foregoing section in the context of our calculations of intersubband transition energies. Interface optical phonons and related electron–phonon interactions in GaAs–$Al_xGa_{1-x}As$ systems with gradients of the chemical composition at the interface were recently discussed in the literature [24]. From the considerations described in [24] we draw the conclusion that, in the case of non-abrupt interfaces, the discrimination of symmetric and antisymmetric modes is futile. If we assume that the coupling strengths of interface phonons to hole intersubband transitions are mainly determined by the proportion of the mode amplitudes within and outside the quantum well, it seems to be reasonable that the branch which evolves out of the GaAs quantum well LO phonon couples most strongly.

Considerable work was done to investigate the nature of optical phonon modes in

layered semiconductor structures, i.e. of confined and interface phonons (see, e.g., [25, 26]), and to give an estimation on their contributions to effects of electron–phonon interaction [27, 28, 29]. These theoretical investigations show that the relative contributions of interface phonons increase with decreasing well width and under application of a magnetic field. The important or even dominant role of interface phonons in the electron–phonon interaction in quantum wells could be shown experimentally by Raman studies of the nonequilibrium phonon population generated by intrasubband relaxation of photoexcited hot electrons [30, 31]. Magnetopolaronic effects on impurity transition energies in quantum wells [32] and magnetophonon resonance measurements on heterojunctions [33] also suggest strong electron interaction with interface phonons [34, 35].

3.3. Raman spectroscopic investigations under photoexcitation

It was stated in [36], that interface phonons can be distinguished from zone-centre vibrational excitations of layered semiconductor structures by measuring their behaviour under photoexcitation. Photoexcitation was found to lead to a strong suppression of the nominally forbidden scattering by interface phonons [36]. In our experiments photoexcitation was realized by a second laser (Ar^+ laser), which emits photons of an energy above the barrier band gap ($\lambda = 514$ nm). Following the model described in [37] we assume that electron–hole pairs created in the barriers are separated by the built-in electric field: the electrons move into the QWs reducing the density of the two-dimensional hole gas by recombination, while the holes move towards the Be-doped barrier layers. The effects of sample illumination can be observed up to some milliseconds after an Ar^+ laser light-pulse, because of the finite lifetime of the photoexcited holes in the potential minimum of the barrier centre region [38]. Under conditions of simultaneous illumination (figure 6) as well as alternation of illumination and detection (figure 7) we observed a suppression of the light scattering by the coupled modes and an increase of the signal strength of the non-coupling quantum well phonon. The degree of suppression of the coupled excitations, which are non-zone-centre modes according to our interpretation, depends on the intensity of the Ar^+ laser (figure 6) or on the delay time Δt between illumination and detection (figure 7). In the context of these results it should be stated that split components of the (h_0–h_1) intersubband transition, with line positions far from the energy range of optical phonons, experience a line narrowing and an intensity increase with decreasing delay time Δt in contrast to the behaviour of the coupled modes displayed in figure 7 [16]. The spectra of figure 7 were measured on sample B, where the larger width of the doped layer reduces the rate of tunnelling of the photoexcited holes from the potential minimum in the barrier to the quantum well, and enlarges the duration of the illumination effect.

We see our results to be in agreement with interpretations given in [36]. The authors of this paper investigated the quenching of the intensity of the Raman signal from interface phonons with increasing laser power density in a nominally undoped sample. As a possible explanation of this effect they assume a neutralization of charged impurities in their nominally undoped samples by trapping of photoexcited carriers. This reduces the efficiency of impurity-induced scattering. For the case of our modulation-doped samples we have to modify this model slightly. We assume a smoothing of potential fluctuations which are caused by the statistical disorder of the ionized impurities in the doped barrier layer [39] by the photoexcited holes as a result of compensation or screening. The signal strength of non-zone-centre modes, which require a loss of translational symmetry parallel to the interface, is reduced in favour of wavevector-conserving light scattering.

Our results give new aspects to the open discussion about the nature of the structural

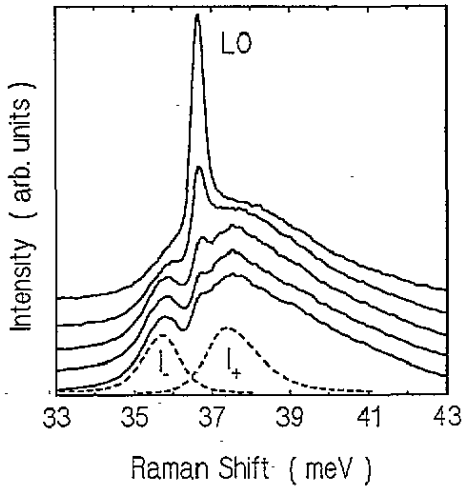

Figure 6. Raman spectra of the coupled-phonon intersubband excitations I_+, I_- (sample A, $B = 7$ T, $z(x,-)\bar{z}$, dye laser energy: 1.750 eV) under conditions of simultaneous illumination with photons of an energy above the barrier band gap (Ar^+ laser, $\lambda = 514$ nm). The power densities increase from the lowest to the uppermost spectrum: 0, 0.03, 0.2, 3 and 10 W cm^{-2}. The dashed lines indicate the contributions of the I_+ and I_- modes. The peak labelled LO is the non-coupling quantum well LO phonon.

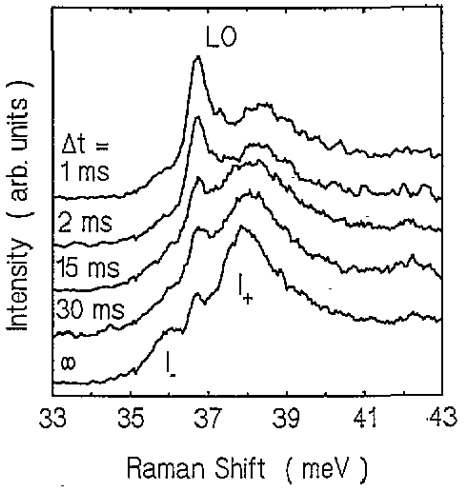

Figure 7. Raman spectra of the coupled-phonon intersubband excitations I_+, I_- (sample B, $B = 7$ T, $z(xx)\bar{z}$, dye laser energy: 1.759 eV) under conditions of previous pulsed illumination with photons of an energy above the barrier band gap (Ar^+ laser, $\lambda = 514$ nm, power density $\leqslant 10$ W cm^{-2}, Δt: delay time between illuminating pulse and Raman detection).

defects responsible for the relaxation of the momentum conservation condition in the process of Raman scattering by interface phonons [40]: the influence of the charged impurities on the scattering process dominates in the case of our doped samples, while islands of well width fluctuations must be assumed to be of less importance. Effects of sample heating by the Ar^+ laser can be excluded considering the small power densities used.

The signal of the coupled modes completely disappears under simultaneous illumination at sufficient intensity, while the peak of the non-coupling quantum well phonon is intensified.

We attribute the latter signal to wavevector-conserving scattering processes. From the lack of any indications of interaction with hole intersubband transitions we have to draw the conclusion that the strength of the coupling of zone-centre quantum well phonons to hole intersubband transitions is nearly vanishing. The strength of this Fröhlich-type interaction seems to increase with the in-plane wavevector $k_\parallel$, and becomes obvious in the field-enhanced Raman signal of the coupled non-zone-centre excitations.

4. Conclusions

We have investigated the interaction of hole intersubband transitions and phonons in p-type modulation-doped MQWs under application of magnetic fields up to $B = 14$ T. The evaluation of Raman spectra observed at high flux densities suggests a coupling of hole intersubband transitions to interface phonons and an increase of the strength of the Fröhlich-type interaction with the in-plane wavevector. The extent of the relaxation of the in-plane wavevector conservation seems to be correlated to the inverse of the magnetic length l_0. The energy of the unperturbed coupling phonon and the line position of a second peak, observed for $B \geqslant 11$ T only, are well described by the calculated energies of interface phonons at $k_\parallel = l_0^{-1}$, which evolve out of the GaAs- and GaAs-like LO phonons of the well and barrier material. The existence of a non-coupling quantum well LO phonon is explained by wavevector-conserving light scattering and the decrease of the coupling strength with vanishing in-plane wavevector.

Our interpretation of the coupling modes as non-zone-centre excitations is confirmed by investigations of their behaviour under photoexcitation, which is able to suppress the signal. These measurements elucidate the mechanism of wavevector-non-conserving light scattering: Potential fluctuations due to ionized impurities in the doped barrier regions are assumed to be smoothed under photoexcitation.

For $B = 0$ T the Raman spectra can be interpreted in terms of a Fano interference of the zone-centre GaAs-LO phonon of the quantum well with the quasi-continuum of $(h_0–h_2)$ transitions. The strength of the Fröhlich interaction mechanism must again be assumed to be very weak as in the case of wavevector-conserving light scattering at high flux densities.

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