Electron-phonon coupling in the two phonon mode ternary alloy \( \text{Al}_{0.25}\text{In}_{0.75}\text{As}/\text{Ga}_{0.25}\text{In}_{0.75}\text{As} \) quantum well

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Abstract. – We have investigated the infrared transmission of a two-dimensional (2DEG) electron gas confined in a \( \text{Al}_{0.25}\text{In}_{0.75}\text{As}/\text{Ga}_{0.25}\text{In}_{0.75}\text{As} \) single quantum well in order to study the electron optical phonon interaction in a two phonon mode system. Infrared transmission experiments have been performed in both the perpendicular Faraday (PF) and tilted Faraday (TF) configurations for which the growth axis of the sample is tilted with respect to the incident light propagation direction and to the magnetic field direction. The experimental results lead to question the validity of the concept of polaron mass in a real material.

Introduction. – Based on the Fröhlich interaction mechanism, polaronic effects have attracted much interest both on the theoretical and on the experimental side [1]. In polar semiconductors, the propagation of longitudinal optical (LO) phonons produces a macroscopic electric field which couples -via the Fröhlich interaction- to the electron motion. This interaction is predicted to give rise to the Resonant Magneto Polaron (RMP) effect between the \( |n = 1 > \) Landau level and the \( |n = 0 + 1\text{LO} > \) state, \( n \) being the Landau level index. This RMP effect, in polar materials, is predicted to induce an anti-crossing of the cyclotron resonance (CR) transition when the CR frequency \( \omega_c = eB/m^* \), where \( B \) is the magnetic field and \( m^* \) is the electron effective mass, is tuned through the LO phonon frequency [2,3]. It has however never been directly observed and different studies on a \( \text{Ga}_{0.47}\text{In}_{0.53}\text{As} \) heterostructure [4], and more recently, on a GaAs quantum well [5] and on a InAs quantum well [6], indicate that no interaction affects the CR mode in the vicinity of the LO phonon energy. It has recently been shown, based on infrared transmission measurements performed on single GaAs quantum wells (QW) doped with carrier concentrations ranging from \( 6 \times 10^{11} \) to \( 12 \times 10^{11} \) \( \text{cm}^{-2} \) [7], that the CR mode does not couple to the LO phonon of the quantum well but instead, couples to the hybrid inter electric sub-band plasmon - LO phonon mode that develops in a doped

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quasi two dimensional layer. These results have been explained using a semi classical model based on the dielectric function formalism \[8\]. This model reproduced the experimental data in an accurate way without assuming any specific electron-phonon interaction. The observed coupling is of dielectric origin. In these GaAs QW studies, the inter electric sub-band energy was approximately twice larger than the LO phonon energy. It is then of fundamental importance to know whether these findings are specific of the GaAs QW or are indeed more general. We report here on results obtained with a \(Al_{0.25}In_{0.75}As/Ga_{0.25}In_{0.75}As\) QW sample differing from the GaAs QW by two main characteristics: the phonon structure is a two mode system and the inter sub-band energy is close to the GaAs-like LO phonon energy. After describing the sample and the experimental details, we present the experimental results, their interpretation and discuss the results.

Samples and experimental details. – The sample \(HM617\) is a single 30 nm wide \(Ga_{0.25}In_{0.75}As\) quantum well grown by molecular beam epitaxy on a (001) GaAs substrate and sandwiched between two \(Al_{0.25}In_{0.75}As\) barriers. To achieve a good electron mobility, a step graded buffer \(Al_{1-x}In_xAs\) with \(x\) ranging from 0.15 to 0.85 was grown on the substrate to obtain a quasi complete strain relaxation in the quantum well. The sample is unintentionally doped \(9\) and the electron density \(n_s\) determined from low temperature transport measurements is of \(2.8 \times 10^{11} \text{cm}^{-2}\) with a mobility of \(20 \text{m}^2\text{V}^{-1}\text{s}^{-1}\). \(Ga_{0.25}In_{0.75}As\) is a two-phonon mode system with two distinct phonon types, InAs-like phonons with energy \(\hbar \omega_{TO} = 21.7 \text{meV}\) and \(\hbar \omega_{LO} = 22.4 \text{meV}\), and GaAs-like phonons with energy \(\hbar \omega_{TO} = 27.9 \text{meV}\) and \(\hbar \omega_{LO} = 29.5 \text{meV}\). Preliminary calculations indicate that the inter electric sub-band energy, for this particular sample, should be of the order of the GaAs-like LO phonon energy, a situation which has never been explored yet.

The experiments are performed with a Fourier transform Brucker IFS-113 spectrometer and the signal is detected by a silicon bolometer located behind the sample at a temperature of 1.8 K. To avoid interference effects, the sample has been wedged with an angle of 2°. Magneto infrared experiments have been performed in an absolute way by using a rotating sample holder which allows to switch \textit{in situ} between the sample and a reference (in the present case a GaAs substrate). We can then determine the absolute transmission of the sample which is the ratio of the transmission of the sample at a given field by the transmission of the reference at the same value of the magnetic field. Magnetic field dependant absorptions, of electronic origin, are more visible on the relative transmission spectrum which is the ratio of the absolute transmission at a given magnetic field by the absolute transmission at zero field. In the tilted Faraday (TF) configuration, for which the normal to the sample surface is tilted by an angle \(\theta\) with respect to the magnetic field and to the light propagation direction, the angle is imposed mechanically by a wedged sample holder. The CR absorption is measured using a superconducting magnet at magnetic fields up to 13 T. The interpretation of the experimental data is accomplished by comparing them with the results of a multi-dielectric simulation of the entire sample structure by an appropriate model \(8\). This is essential because in the optical phonon range of energy, spectra can be strongly distorted by interference effects independent of any electron-phonon interaction. Probably the most studied III-V material is the GaAs quantum well for which transmission results are always obscured by the strong absorption of the substrate. The \(Ga_{0.25}In_{0.75}As\) quantum well system is more attractive to study polaronic effects because the optical phonon energies of this compound are below the GaAs reststrahlen band: this allows to study the infrared absorption through the interesting optical phonon range of energy.

Experimental results. – In the perpendicular faraday (PF) configuration, the spectra exhibit a characteristic field dependent CR absorption line (see black dots in fig. 1). This
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Fig. 1 – Experimental CR spectra (black dots) and simulated spectra (solid lines) of sample HM617 measured at $T = 1.8$ K in PF configuration. The InAs-like TO phonon is indicated by the vertical solid line, the InAs-like LO phonon by the vertical dash-dotted line, the GaAs-like TO phonon the vertical dashed line and the GaAs-like LO phonon by the vertical dotted line. The shaded region of energy corresponds to the reststrahlen band of the GaAs substrate.

Fig. 2 – Experimental CR spectra (black dots) and simulated spectra (solid lines) of sample HM617 measured at $T = 1.8$ K in TF configuration for $\theta = 15^\circ$.

CR absorption is tuned through the entire range of InAs-like and GaAs-like optical phonon energies without being affected by neither the InAs-like LO phonon (dash-dotted line in fig. 1) nor the GaAs-like LO phonon (dotted line in fig. 1) of the quantum well. Therefore, in this PF configuration, the expected polaron coupling is not observed. This is in accordance with results obtained on GaAs QW [7]. At low fields below $\approx 10$ T, the simulation (solid lines in fig. 1 and fig. 2) provides a fitted value of the effective mass $m^* = 0.038 m_0$ in agreement with previous electrical detection of CR experiments performed on the same type of samples [10]. When the magnetic field is tuned to 13 T, the CR absorption is observed above the reststrahlen band of the GaAs substrate with a significant increase of the mass $m^*$ which, in the present case, reaches a value of 0.0395 $m_0$. This may be due to band non parabolicity effects known to be significant for this compound.

The infrared transmission measured in the TF configuration shows a very different behavior (see black dots in fig. 2): When the CR energy is well below the GaAs-like optical phonon range of energy, we observe a single electronic absorption that pins at an energy of 27.2 $\pm$ 0.3 meV while a second absorption appears at an energy of 28.3 $\pm$ 0.4 meV. This second absorption then enters the obscured region of the GaAs substrate reststrahlen band while a third absorption appears above this band. For a given perpendicular component of the magnetic field, the minima of transmission, measured in the TF configuration above the reststrahlen band of the substrate, is well above the one measured in the PF configuration (see spectra at 13 T in fig. 1 and at 12.55 T in fig. 2 and full dots in fig. 3) which is surprising. Indeed the cyclotron energy is usually believed to depend only on the perpendicular component of the magnetic field which
should lead to a lower energy of the CR than the one measured in the PF configuration as this is observed in GaAs QW [7]. Moreover, the width of this second absorption is much bigger than the one of the CR mode measured in the PF configuration in the same range of energies.

We argue that all the observed effects are due to the coupling of the CR mode with the hybrid inter sub-band plasmon - LO phonon modes as observed in a GaAs single quantum well [7]. In the present study however, the polar slab containing the 2DEG has two distinct phonon modes (InAs-like and GaAs-like) which are both coupled to the plasmon inter sub-band with the inter sub-band energy close to that of the GaAs like LO energy.

Interpretation of the results. – To explain these data, we have calculated the optical transmission of the whole structure using the multi-dielectric simulation [8] and the exact layer sequence for our sample. The calculation consists in evaluating for each layer N with a dielectric tensor $\varepsilon_N(\omega)$, the transfer matrix $M_N$. This requires to solve the Maxwell equations in order to determine the appropriate modes of propagation of light $k_i$ inside the layer. There are four modes $k_i$ for each layers. Writing the conservation relations of the tangential components of both electric and magnetic fields at each interface enables us to determine the $4 \times 4$ transfer matrix characterizing a layer. If this layer does not contain free charges, $\varepsilon_N(\omega)$ is then diagonal and the $4 \times 4$ transfer matrix decomposes into two blocks of $2 \times 2$ matrices related to the transverse electric (TE) and transverse magnetic (TM) modes respectively. For a doped layer in TF configuration, all the components of $\varepsilon_N(\omega)$ and therefore of the transfer matrix are non zero and the TE and TM modes are mixed. The transmission of the whole structure can then be calculated as soon as the dielectric tensor of the 2DEG in the TF configuration has been evaluated.

The main effect of the in-plane component of the magnetic field is to couple the x- and z- part of the wave function describing the electron gas. This coupling can be evaluated using perturbation theory in the case of a square quantum well. The use of a parabolic confinement [11,12], characterized by an inter sub-band energy $\hbar\Omega$, provides an exact solution of this problem. It is this model, based on the Drude formalism, that we have used to evaluate, neglecting retardation effects, all components of the dielectric tensor $\varepsilon_{QW}(\omega)$ describing the quantum well and then to simulate the transmission.

The results of such a calculation for the PF configuration are presented in fig. [solid lines]. When the cyclotron energy is tuned through the GaAs-like optical phonon range of energy, the electronic absorption splits around the GaAs-like TO phonon frequency (see spectrum at 9 T in fig. [1]). This behavior was first attributed to a dielectric artifact [13,14] as the TO phonon is a natural pole of the dielectric function describing the 2DEG, but this effect seems to be more intrinsic. As the dielectric calculation, in which no electron-TO phonon interaction is considered, cannot reproduce the experimental spectra, the observed discontinuity of the cyclotron energy at the TO energy is a sign of an interaction of the CR mode with the GaAs-like TO phonon mode of the Ga$_{0.25}$In$_{0.75}$As quantum well. The same effects were already observed in this kind of compounds [4] and in GaAs QW [7]. Therefore it seems that in doped QWs, there exists in general an interaction between the cyclotron mode and the TO phonon modes. This deserves to be worked out on theoretical grounds. The average electron scattering time deduced from the fitting procedure is $\tau_{CR} = 6 \times 10^{-12}$ s which corresponds to a mobility of $27 \text{ m}^2\text{V}^{-1}\text{s}^{-1}$ in agreement with the mobility deduced from transport measurements.

The use of the TF configuration enables to excite the 2DEG with a non vanishing component of the incident electric field perpendicular to the quantum well. In this case, the response of the electronic system involves the $\epsilon_{\alpha z}(\omega)$ ($\alpha = x, y, z$) component of the dielectric tensor.
For instance, the $\epsilon_{zz}(\omega)$ component describing the 2DEG confined in a two phonon mode polar slab can be formulated, ignoring for simplicity the imaginary part, as [8,15]:

$$
\epsilon_{zz}(\omega) = \epsilon_0 \prod_i \frac{\omega_{p,i}^2 - \omega^2}{\omega_{p,i}^2 - \omega_0^2} + \frac{\omega_p^2(\omega_{e,i}^2 - \omega^2)}{\omega^4 - \omega^2(\Omega^2 + \omega_0^2) + \omega_{zz}^2\Omega^2}
$$

(1)

where $\epsilon_0$ is an average static dielectric constant taken to be equal to 13.85, L and T refer to the longitudinal and transverse phonons while the subscript i refers to the relevant GaAs-like and InAs-like modes, $\omega_p$ is the plasma frequency, $\Omega$, the inter sub-band frequency, $\omega_e$, the cyclotron frequency and $\omega_{e,i} = eB_\perp/m^*$, with $B_\perp$ being the perpendicular component of the magnetic field. This component of the dielectric tensor has 4 zeros corresponding to longitudinal solutions: for low values of the magnetic field, they correspond to the CR like and InAs-like modes, $\omega$ the cyclotron frequency and $\omega_{e,i} = eB_\perp/m^*$, with $B_\perp$ being the perpendicular component of the magnetic field and to three hybrid inter electric sub-band plasmon-LO phonon modes of the quantum well. For higher values of the magnetic field, the evolution of these modes is far from trivial: they are displayed, as full lines, in fig. 3, as a function of $B_\perp$ for a tilt angle of 15°. One can compare these results with the observed minima of transmission (full dots in fig. 3). Though all non diagonal elements of the dielectric tensor play also a role in the calculation of the transmission, the main contribution is provided by $\epsilon_{zz}(\omega)$. Therefore the agreement between the experimental results and the solutions of Eq. 1 is very good.

In the TF configuration, we impose, to fit the experimental data, that the effective mass for a given perpendicular component of the magnetic field is the same as the one deduced in the PF configuration for the same value of the magnetic field. We then fit the inter sub-band energy $h\Omega$ together with the inter sub-band scattering time ($\tau_{IS}$). The deduced inter sub-band energy is $h\Omega = 28.5$ meV. The fitted spectra are obtained with a value of $\tau_{IS} = 3 \times 10^{-12}$ s, much shorter than $\tau_{CR}$ resulting in a larger width for the corresponding transmission structure. This line width dominates the absorption in the GaAs-like optical phonon energy range. This situation is different from the previously measured absorption in the GaAs QW [7] where the coupling involved the GaAs-like phonon hybrid mode with a width considerably less than that of the inter sub-band plasmon observed in this report. The reason is that the hybrid mode which couples to the CR transition has a phonon character when $h\Omega \gg h\omega_{LO}$ whereas it acquires an inter sub-band transition character when $h\Omega \lesssim h\omega_{LO}$ as in the present case. This inter sub-band transition is known to appear with a much larger line width [16] than that observed for the resonance transition between non dispersive levels as Landau levels or phonons.

Despite small discrepancies, the multi-dielectric simulation in the TF configuration reproduce the main observed features, which are the anti-crossing behavior below the GaAs-like LO phonon energy with the pinning of the CR absorption, and the appearance of a third absorption at an energy higher than the observed cyclotron energy in PF configuration. This means that, though simple, our model contains all the physics necessary to explain the observed behavior without invoking any electron-LO phonon interaction.

Therefore, the good agreement between the theory and the experiment allows to assign unambiguously the origin of the observed anti-crossing in the TF configuration. It is clearly due to the dielectric coupling of the CR with each of the inter sub-band plasmon-LO phonon hybrid modes. Though, due to the small oscillator strength of the InAs-like mode and the relatively large width of the cyclotron line, this coupling is not observed for the lower hybrid mode around 22 meV, it is clearly visible around the second hybrid mode (27.8 meV). Therefore, from the present results, and those previously obtained in GaAs QW [7], one can say that, in 2D doped QW (at least in III-V compounds), whatever is the phonon structure, the carrier density or the relative value of the inter sub-band energy with respect to the LO
Discussion of the results. – As discussed in the preceding section, the model used to evaluate the dielectric function neglects retardation effects: the dielectric function is therefore formulated at \( k \to 0 \). This \( k \)-dependence has been studied by Peeters et al. \([17]\) in this system and it appears to be significant for \( k \)-vectors of the order of \( 10^6 \) cm\(^{-1}\), decreasing rapidly below this value. In the present experiments, performed in the infra-red region, the real part of the \( k \)-vector at the CR frequency is of the order of \( 10^4 \) cm\(^{-1}\) and therefore neglecting retardation effects in the derivation of the dielectric response is well justified.

Since when deriving the multi-dielectric model of transmission \([8]\), one has to solve the Maxwell equations for each layer, we are indeed using the polariton picture with a non \( k \)-dispersive dielectric function. What is not included in the model is any electron-phonon interaction which should appear in the dielectric response function. In the case of the Fröhlich interaction, it could be included through the memory function approach \([18]\) which predicts an anti-crossing of the CR transition around the LO energy in the PF configuration which should be significant for the carrier concentration used here: this RMP effect is however not observed. Since it is directly related to the concept of polaron mass, this concept itself can be questioned in real doped systems.

It is easy and instructive to extend the present model to the 3-dimensional case by letting \( \hbar \Omega \to 0 \) while keeping \( \omega_0^2 \) constant. As an example, in this limit, the zeros of Eq. 1, for a one-phonon mode system at \( B = 0 \) are simply the well known \( \omega_{\pm} \) modes found for the plasmon-phonon coupling in a polar semiconductor, as first reported by Mooradian et al. \([19]\).
Conclusion. — In conclusion, infrared transmission experiments have been performed on a single $\text{Al}_{0.25}\text{In}_{0.75}\text{As}/\text{Ga}_{0.25}\text{In}_{0.75}\text{As}$ quantum well to investigate the nature of the electron-phonon coupling in a two phonon modes system. In PF configuration, we observe no sign of interaction of the CR mode with the GaAs-like LO phonon. Instead, we observe signs of a coupling with the TO phonon of the quantum well. As in pure GaAs single quantum well, but changing the physical parameters of the problem, we observe for the first time, the coupling of the CR mode with the two hybrid GaAs-like LO phonon-electric sub-band modes of the quantum well with free carriers. This effect is of pure dielectric origin and is not due to any specific electron-phonon interaction.

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