Electromodulation spectroscopy in midinfrared spectral region: The band offset study for GaSb-based quantum wells

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Abstract. Electromodulation spectroscopy (photoreflectance and electroreflectance) is an excellent technique to study optical transitions in quantum well (QW) structures but the development of this technique in the midinfrared spectral region is still limited due to various reasons. In this work we report our recent progress in the development of contactless electroreflectance spectroscopy in midinfrared and discuss some aspects and perspectives of this technique in the context of its application to study optical transitions in GaSb-based QWs. Especially, we have focused on the band offset issue in GaInAsSb/GaSb QWs since the accurate study/verification of band gap discontinuities at QW interfaces is a crucial point in the optimization of laser structures grown on GaSb substrate and operating in the 1.8-3.0 µm spectral range.

1. Introduction
Semiconductor lasers operating in the midinfrared regime are attractive light sources for various applications including remote gas sensing, pollutant detection, medical procedures or laser spectroscopy. GaSb-based quantum wells (QWs) are one of the most promising material systems to achieve light emission in this spectral range [1-6]. In general, the band structure of a GaIn(As)Sb/GaSb QW (i.e., the valence band position) can be predicted within the “model-solid” theory [7], but the accuracy of the valence band offset determination within this theory can be comparable with the band gap discontinuities at QW interfaces (~0.1 eV versus <0.3 eV). It means that an optimal design of GaSb-based QW for a particular application is not a simple task.

From the viewpoint of laser applications, type-I QWs with a deep confinement potential for electrons are required. Such a situation is realized in GaInSb/GaSb QWs [8-11], but the emission wavelength from this system is limited to <1.8 µm because of the compressive strain. InAsSb/GaSb system is more promising from the viewpoint of strains but the InAs/GaSb interface is type-III [12] and according to the “model-solid” theory no hole confinement is expected for InAsSb/GaSb QWs. It makes these QW unpromising within the classical approach to design the laser structures and therefore it is the GaInSb/GaSb QW system that is mainly explored and optimized. For example, the incorporation of arsenic into GaInSb leads to significant changes in strains and small changes in the energy gap of free standing GaInSb. It means that the GaInAsSb/GaSb QW with small amount of arsenic, is a promising way to achieve type-I QWs with the emission wavelength longer than 1.8 µm and sufficiently deep confinement potential for electrons and holes. However, it is very difficult to predict the optimal content of GaInAsSb/GaSb QWs due to some inaccuracies in the “model-solid”
theory, strain model and material parameters. It means that particular GaInAsSb/GaSb QWs have to be investigated experimentally. One of the most powerful approach to verify the type of QW as well to find the band gap discontinuities at QW interfaces is electomodulation spectroscopy [photoreflectance (PR) or contactless electroreflectance (CER)] supported by theoretical calculations. Recently, this approach has been applied by us many times to study dilute nitride QWs such as GaNAs/GaAs [13], GaInNAs/GaAs [14, 15], GaNAsSb/GaAs [16-19], and GaInNAsSb/GaAs [20-23] QWs. The application of electromodulation (EM) spectroscopy to investigate the conduction (valence) band offset in dilute nitride QWs with various alloy contents was fully justified since any predictions of the valence band offset within the “model-solid” theory do not work in this case [24]. In the case of GaSb-based QWs, it is expected that the “model-solid” theory can be successfully applied to predict the valence band offset but the absolute accuracy of this prediction can be unsatisfactory. Therefore, the application of EM spectroscopy to study the valence band offset in GaSb-based QWs is also required.

The aim of this paper is to show the usefulness of EM spectroscopy to study the band structure of GaInAsSb/GaSb QWs and discuss some aspects of EM spectroscopy in the midinfrared spectral region.

To date the EM spectroscopy was used very rarely in the midinfrared spectral region because of weaker band bending modulation in narrow band gap semiconductors as well as some technical problems which are associated with weaker sensitivity of detectors in this spectral region. Recently, the application of PR spectroscopy has been developed up to 5 μm for bulklike samples [25, 26]. Unfortunately, the QW transitions are usually 1-2 magnitudes weaker than barrier (bulklike) transitions. It makes the EM spectroscopy of QW transitions more difficult and challenging. In addition, in the case of PR spectroscopy of QW transitions, an interference of QW-related resonances with a below band gap oscillation can be observed and thereby the analysis of QW transitions can be difficult or impossible [27-29]. Such a situation is often observed for GaAs-based structures grown on n-type GaAs substrate [27] and sometimes can be also observed for structures grown on semiinsulating substrates [28]. Recently, this situation was observed for GaSb-based structures, i.e. GaInSb/GaSb QWs [11]. In the case of GaAs-based structures, it has been shown that the oscillation-like signal can be eliminated if CER spectroscopy is applied instead of the PR one [27, 28]. This effect is attributed to a different electromodulation mechanism in the two techniques [27]. A similar situation is expected for GaSb-based structures. It means that the development of CER spectroscopy in the midinfrared spectral region (λ > 2.0 μm) is strongly motivated. In this paper we focus on CER spectroscopy since this technique seems to be the most promising for GaSb-based QWs grown on n-type GaSb substrates.

2. Experimental details

The CER measurements were performed in the so-called ‘bright configuration’ [30], see in Fig. 1, where the sample was illuminated by white light instead of monochromatic light as it takes place in the standard configuration, i.e. so-called ‘dark configuration’. In the CER measurements the sample is mounted in a semitransparent capacitor illuminated by light from a halogen lamp (100 W) at normal incidence as shown in Fig. 1. In order to eliminate unneeded visible and ultraviolet illumination of the sample an edge filter (e.g. a silicon wafer) is mounted behind the halogen lamp. The reflected light is dispersed through a 0.55 m focal length single grating monochromator and detected by thermoelectrically cooled p-i-n InAs photodiode. The signal measured by photodiode has two components: i) the DC component which is proportional to \( I_0 R \), and ii) AC component which is proportional to \( I_0 \Delta R \). Both DC and AC components are measured with a lock-in amplifier (\( I_0 \) is the intensity of reflected light). A computer records both the AC and DC signals and next the AC signal is divided by the DC component giving the CER spectrum, \( \frac{\Delta R}{R}(E) \), where \( E \) is the photon energy of the incident beam. The capacitor system is built of a top electrode made of a copper mesh and the bottom electrode made of a copper solid block. The semitransparent top electrode was kept at a distance of approx. ~0.1-0.4 mm from the sample surface while the sample itself was glued to the bottom copper.
electrode. A home made generator of square AC voltage was used to generate the AC field inside the capacitor. A maximum peak-to-peak alternating voltage of 1.8-2.5 kV with the frequency of 280 Hz was used for the modulation.

The QW structure selected to this paper was grown on a (100) oriented n-GaSb substrate by solid source molecular beam epitaxy in an EIKO system at Wurzburg University. The structure consists a 500-nm thick GaSb buffer layer, a 21 nm thick Ga_{0.24}In_{0.76}As_{0.08}Sb_{0.92} quantum well, and a 100-nm thick GaSb cap layer. Relevant details of the sample growth were described in our previous paper [31].

Figure 1. Experimental set-up for CER measurements in the so-called ‘bright configuration’. H – Halogen lamp; S – Slit; L1, L2, L3 – Lenses; F1, F2 – Filters; BS – Beam Splitter; C – Capacitor; M – Monochromator; D – Detector; NV – Lock-in; G – Generator; PC – Personal Computer.

3. Analysis of contactless electroreflectance spectra

Figure 2 (a) shows room temperature CER spectrum for 21 nm thick Ga_{0.24}In_{0.76}As_{0.08}Sb_{0.92}/GaSb QW. This spectrum is dominated by GaSb bulklike signal at the energy of ~ 0.725 eV. Below this signal three CER resonances are clearly observed. These resonances are attributed to optical transitions in the Ga_{0.24}In_{0.76}As_{0.08}Sb_{0.92} QW region. It is worth noting that without any comparison with theoretical calculations it can be concluded that this QW is type-I since optical transitions between QW excited states are observed in addition to the ground state transition, which is observed at the energy ~ 0.56 eV. In order to determine the value of the conduction (valence) band offset, energies of QW transitions have to be extracted from CER spectrum and compared with theoretical calculations for various values of the band offset.

In order to extract energies of the optical transitions, a standard fitting procedure has been applied, see e.g. Refs. [13, 18]. CER resonances were analyzed using the low-field electromodulation Lorentzian lineshape functional form [32]

$$\frac{\Delta R}{R}(E) = \text{Re} \left[ \sum_{j=1}^{n} C_j e^{i\theta_j} \left( E - E_j + i\Gamma_j \right)^{-m_j} \right],$$

where $n$ is the number of the optical transitions and spectral functions used in the fitting procedure, $C_j$ and $\theta_j$ are the amplitude and phase of the line shape, and $E_j$ and $\Gamma_j$ are the energy and the broadening parameter of the transitions, respectively. The term $m_j$ refers to the type of optical transitions in question: $m_j = 2, 2.5, 3$ for an excitonic transition, a three-dimensional one-electron transition, and two-dimensional one-electron transition, respectively. It is worth noting that this formula is appropriate at low temperatures for a high quality structures. Usually at room temperature and/or for a sample with high inhomogeneities first derivate Gaussian line shape is more appropriate. In our case we assume that $m = 3$. It corresponds to a one electron transition in two dimensional system and is very close to the third derivative Gaussian line-shape, i.e., the most appropriate line-shape at room temperature. The fitting curves are shown by thick solid lines in Fig. 2 (a) together with the moduli of
individual resonances (dashed lines) obtained according to Eq. 2. The identification of CER resonances was possible on the basis of theoretical calculations.

\[
\Delta \rho_j(E) = \frac{|C_j|}{\left[\left(E - E_j\right)^2 + \Gamma_j^2\right]^{1/2}}.
\]

(2)

![Figure 2](image)

Figure 2. (a) Room temperature contactless electroreflectance spectrum for 21 nm wide Ga\(_{0.24}\)In\(_{0.76}\)As\(_{0.08}\)Sb\(_{0.92}\)/GaSb QW. (b) Energy-band diagram in real space for compressively strained GaInAsSb/GaSb QWs.

4. Calculations of quantum well transitions

The calculations of QW energy levels have been performed within the framework of the effective mass approximation. The influence of strain on the band structure is taken into account, but excitonic effects are neglected since it is expected that at room temperature for narrow gap semiconductors they are negligible. The biaxial strain was calculated based on the Pikus-Bir Hamiltonian [33]. For the \(E_0\) critical point in GaInAsSb (\(k = 0\)) the hydrostatic component of the strain, \(\delta E_H^c\), shifts the valence and conduction bands. This total shift corresponds to the change in the bandgap energy due to a hydrostatic deformation, which is proportional to the deformation potential \(\alpha\), that is measured. The total shift should be divided between the valence and conduction bands proportionally to the potential constants \(\alpha_V\) and \(\alpha_C\), where \(\alpha = \alpha_V + \alpha_C\). The shear component, \(\delta E_S\), removes the valence band degeneracy, giving a separate \(E_{HH}\) (related to heavy holes) and \(E_{LH}\) (related to light holes). The energy of conduction and valence bands can be expressed in terms of the unstrained value as

\[
E_c = E_c^0 + \delta E_H^c,
\]

\[
E_{v,HH} = E_v^0 + \delta E_H^0 + \delta E_S^0,
\]

\[
E_{v,LH} = E_v^0 + \delta E_H^0 - \delta E_S^0 + \ldots,
\]

where \(E_c^0\) and \(E_v^0\) correspond to the energy of the conduction and valence bands at \(k = 0\), respectively. The values of the \(\delta E_H^c\), \(\delta E_H^0\), and \(\delta E_S^0\) are given by following formulas

\[
\delta E_H^c = 2\alpha^c \left(1 - \frac{C_{12}}{C_{11}}\right)
\]

(4a)
where \( \varepsilon \) is the in-plane strain, \( C_{11} \) and \( C_{12} \) are elastic stiffness constants, and \( b \) is the shear deformation potential. All the material parameters for Ga\(_{1-x}\)In\(_x\)As\(_{1-y}\)Sb\(_y\) have been obtained by linear interpolation between the parameters of a relevant binary semiconductor [12] according to Eq. 5.

\[
Q(x, y) = (1 - x) \cdot y \cdot Q_{GaSb} + (1 - x) \cdot (1 - y) \cdot Q_{GaAs} + x \cdot y \cdot Q_{InSb} + x \cdot (1 - y) \cdot Q_{InAs}.
\]  

The bandgap energy of GaInAsSb was calculated according to the formula given in Ref. [34]. Note that the energy gap of GaInAsSb can be adjusted to the experimental value of the QW ground state transition since the energy of the ground state transition does not vary significantly with the change in the conduction (valence) band offset. The conduction- and valence-band offsets, \( Q_c \) and \( Q_v \), are defined by Eq. 6 as

\[
Q_c = \frac{\Delta E_c}{(\Delta E_c + \Delta E_v)} \times 100\% \quad \text{(6a)}
\]

\[
Q_v = (1 - Q_c) \times 100\% \quad \text{(6b)}
\]

where \( \Delta E_c \) and \( \Delta E_v \) are the conduction- and valence-band discontinuities at the heterojunction for unstrained materials, as illustrated in Fig. 2 (b). From a laser device perspective, the most interesting values are the band gap discontinuities with the strain corrections, shown in Fig. 2 (b) as \( \Delta E_c^* \) and \( \Delta E_v^{\text{HH}} \). Generally, the most appropriate approach to heterojunction band offsets is to determine the \( Q_c \) (or \( Q_v \)) since a GaInAsSb/GaSb QW can be grown on both GaSb and InAs substrates. For identical QWs grown on different substrates, the \( Q_c \) does not vary whereas the \( \Delta E_c^* \) and \( \Delta E_v^{\text{HH}} \) discontinuities vary due to different lattice strains. Moreover, the value of \( \Delta E_c^* \) and \( \Delta E_v^{\text{HH}} \) discontinuities can be calculated if the unstrained ‘natural’ offset, \( Q_n^* \), is known. Thus the \( Q_c \) value is a more universal parameter. In this paper, both the \( Q_c \) and \( \Delta E_c^* \) and \( \Delta E_v^{\text{HH}} \) discontinuities are analyzed.

Figure 3 (a) shows theoretical calculations of QW transitions for the 21nm wide Ga\(_{0.76}\)In\(_{0.24}\)As\(_{0.08}\)Sb\(_{0.92}\)/GaSb QW as a function of the conduction band offset together with experimental data (horizontal dashed lines). On the basis of such a plot the nature of QW transitions...
can be identified and the conduction (valence) band offset can be found. The analysis in Fig. 3 (a) shows that the best agreement between experimental data and theoretical calculations takes place when the $Q_C$ equals $-90\%$. It means that the unstrained Ga$_{0.76}$In$_{0.24}$As$_{0.08}$Sb$_{0.92}$/GaSb interface is of type I for both electrons and holes. As was mentioned for QW laser design, the relevant parameters are the band gap discontinuities in the strained system, namely the values of $\Delta E^*_C$ and $\Delta E^*_{VE}$ for the conduction and heavy-hole QWs, respectively. These values can be determined knowing the $Q_C$, for the unstrained GaInAsSb/GaSb interface and the strain-related shifts, as shown in Fig. 2 (b).

According to this figure, it has been found that the $\Delta E^*_C$ and $\Delta E^*_{VE}$ equals 132 and 52 meV, respectively, see Fig. 3(b). The $\frac{\Delta E^*_C}{\Delta E^*_C + \Delta E^*_{VE}}$ ratio for the Ga$_{0.76}$In$_{0.24}$As$_{0.08}$Sb$_{0.92}$/GaSb interface ($Q_C^{III}$) equals 72%. It means that this quantum well has a good confinement potential for both electrons and holes and is promising from the viewpoint of laser applications. For light holes this QW is of type II mainly due to the compressive strain in this system.

5. Summary and outlook

In summary, it has been shown that CER spectroscopy can be used to study energies of QW transitions in the midinfrared spectral region at room temperature. For Ga$_{0.76}$In$_{0.24}$As$_{0.08}$Sb$_{0.92}$/GaSb QW, QW transitions related to both the ground and excited states can be clearly observed in CER spectra. The experimental QW transition energies were compared with theoretical predictions based on an effective mass formalism model. The calculations were carried out for various $Q_C$ to obtain a match between theory and measured data. A good agreement between experimental data and theoretical calculations was observed for $Q_C \sim 90\%$. In this way, it was concluded that the Ga$_{0.76}$In$_{0.24}$As$_{0.08}$Sb$_{0.92}$/GaSb QW is of type I for both electrons and heavy holes; the $\Delta E^*_C$ and $\Delta E^*_{VE}$ equals 132 and 52 meV, respectively. It means that EM spectroscopy, especially CER technique, is a very useful tool to investigate/verify the band structure of the active part in GaSb-based lasers. In our opinion, a future development of EM spectroscopy in the midinfrared spectral region is very perspective and needed. We suppose that the following issues/problems will be very important in this development:

i) **The intensity of probing beam:** Since the sensitivity of midinfrared detectors is worse than Ge and Si detectors, one way to improve EM measurements in midinfrared spectral region is to increase the intensity of probing beam. Global lamps seem to be the most common and promising light emitters in midinfrared but the power density from tungsten lamps (common halogen lamps) is higher than the power density attainable from globar lamps. Therefore, in the region of quartz bubble transparency (<4 µm) tungsten sources seem to be better than globar sources.
ii) **Temperature of measurements:** One of the best advantages of EM spectroscopy is its high sensitivity at room temperature, i.e., the operating temperature of electronic devices. In the case of narrow semiconductors, the band bending modulation at room temperature is usually much weaker than the possible band bending modulation in GaAs-, InP- or GaN-based structures. In order to enhance band bendings and their modulation, the measurement temperature can be lowered. The temperature of liquid nitrogen is easily available and cheap in operation. Therefore, we expected that the development of midinfrared EM spectroscopy will be associated with the lowering of measurement temperature to the liquid nitrogen temperature, which is not an obstacle in a standard characterization of semiconductor structures.

iii) **Fourier transform in infrared FTIR:** The recent development in the application of Fourier transform in infrared to measure photoreflectance spectra [25, 26] shows that it could be a very promising way to obtain EM spectra in the midinfrared spectral region. To date, only bulk-like transitions have been detected by using this approach. QW related transitions are usually one or two magnitudes weaker than bulk-like transitions. Therefore, the measurement of QW transitions by FTIR EM is a challenge. In the case of conventional CER spectroscopy, we have achieved sensitivity better than $5 \times 10^{-6} \Delta R/R$ at $\sim 1.6-2.5 \mu m$, see Fig. 2 (a). Such a sensitivity in FTIR EM should guarantee successful measurements of QW transitions.

iv) **Spectral resolution:** In general, the broadening of CER and PR resonances is a result of the thermal broadening of the optical transitions and the spectral resolution of the experimental setup. In the case of electromodulation methods, such as CER and PR, the modulation of internal electric field can be a source of an additional broadening of CER (PR) resonances. This additional broadening depends on the magnitude of electromodulation inside the sample. In practice, two CER (PR) measurements at two significantly different modulation amplitudes (various modulation voltages for CER and pumping beam intensities for PR) are enough to estimate the importance of modulation-related broadening for optical transitions. So far, we have observed that this broadening is less than a few meV. At room temperature this broadening is usually much smaller than the thermal broadening of optical transitions and therefore can be neglected. At cryogenic temperatures the modulation-related broadening can be important and thus the amplitude of electromodulation should be selected/controlled very carefully. However, it is not expected that this issue will be very important for narrow gap semiconductors since in this material system there are much smaller amplitudes of electromodulation.

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