Contactless electroreflectance approach to study the Fermi level position in GaInNAs/GaAs quantum wells

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A fruitful approach to study the Fermi level position in GaInNAs/GaAs quantum wells (QWs) has been proposed in this paper. This approach utilizes contactless electroreflectance (CER) spectroscopy and a very simple design of semiconductor structures. The idea of this design is to insert a GaInNAs quantum well (QW) into a region of undoped GaAs layer grown on *n*-type GaAs substrate. The possible pinning of the Fermi level in the GaInNAs QW region modifies band bending in this system. In CER spectra both QW transitions and GaAs-related Franz-Keldysh oscillations (FKOs) are clearly observed. The analysis of QW transitions allows one to determine the band gap discontinuity at GaInNAs/GaAs interface whereas the analysis of FKOs allows one to determine the built-in electric field in the GaAs cap layer, and, finally, one is able to find the Fermi level pinning in GaInNAs QW region. © *2007 American Institute of Physics*.
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I. INTRODUCTION

Substitution of a group V element in group III-V compounds with small amounts of nitrogen at alloylike concentrations causes dramatic changes in the band structure[1–4] and optical properties of the III-V host. The most important effects are a large reduction of the fundamental band gap energy (100–150 meV per % of nitrogen)[5] and a significant increase in the electron effective mass.[6,7] Moreover, nitrogen typically generates point defects and deteriorates the optical properties of III-V compounds. It is generally accepted that low luminescence efficiency indicates the existence of nonradiative recombination centers or traps inside the band gap of dilute nitrides. In the case of N-containing III-V compounds (so-called dilute nitrides) many different traps can exist inside the band gap, depending upon the growth method, growth conditions, and alloy composition. The exact nature of these centers is still unknown. Additional questions also remain: How do these defects influence the Fermi level pinning in dilute nitrides? Is the material, which is nominally undoped, *n*-type or *p*-type in character? The Fermi level pinning in diluted nitrides is a challenging subject of investigation from both a fundamental and practical point of view. In this paper a fruitful approach to investigate this problem has been proposed. This approach utilizes electromodulation (EM) spectroscopy[8] and a special design of semiconductor structures. We name this design as a "modified Van Hoof structure" since a similar structure has been proposed by Van Hoof *et al.*[9] to investigate the Fermi level pinning at the GaAs surface.

Figures 1(a) and 1(b) show the layer sequence and band bending in the unmodified and modified Van Hoof structures, respectively. The difference between the original Van Hoof structure is that an additional epilayer exists inside the undoped GaAs region (a GaInNAs layer in this case). For a common Van Hoof structure, a homogenous electric field is expected inside the undoped GaAs layer. The value of this field can be estimated with knowledge of the thickness of GaAs epilayers and the Fermi level pinning at the GaAs surface and at the GaAs/GaAs (*n*-type substrate) interface,[9,10] as shown in Fig. 1(a). In addition, the built-in electric field can be measured by EM spectroscopy since the EM spectra exhibit Franz-Keldysh oscillations (FKOs) for bulklike transitions in the regime of medium and high internal electric fields. The electro-optic energy, which corresponds to the period of FKOs, is related to the amplitude of built-in electric fields.[10] Originally this approach has been applied to determine the Fermi level pinning at semiconductor surfaces.[9,10]

In the case of a modified Van Hoof structure, the thickness of epilayers and the Fermi level pinning at the GaAs surface and at the GaAs/GaAs (*n*-type substrate) interface are known, but the electric field inside the undoped GaAs layers cannot be estimated as easily since the GaInNAs layer can modify band bending in this system. In order to predict the electric field in the modified Van Hoof structure, the band gap discontinuity at the GaInNAs/GaAs interface and the Fermi level pinning in the GaInNAs layer must be known. On the other hand, the Fermi level pinning in GaInNAs can be estimated with knowledge of the electric field and band

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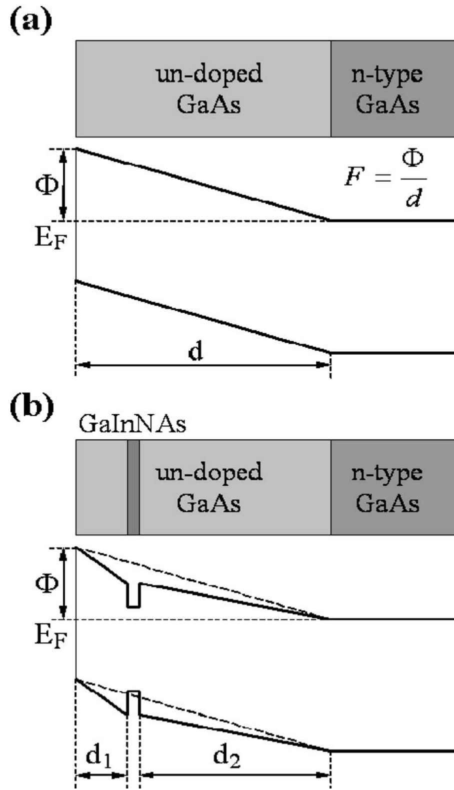

FIG. 1. Layer sequence and a sketch of band bending in (a) unmodified and (b) modified Van Hoof structures; the dashed line corresponds to a situation when the band bending is not modified by the QW whereas the solid line corresponds to a situation when the QW influences band bending in this structure.

gap discontinuity at the GaInNAs/GaAs interface (see Fig. 2). EM spectroscopy can be applied to measure both the electric field inside GaAs layers and the band gap discontinuity at the GaInNAs/GaAs interface if the GaInNAs layer is sufficiently thin (i.e., GaInNAs is a quantum well). The application of EM spectroscopy to study optical transitions in GaInNAs/GaAs quantum wells (QWs) and FKOs in GaAs layers allows us to determine the Fermi level pinning in GaInNAs. In this paper, this concept was applied to study the Fermi level position in the GaInNAs QW region with low and high indium contents.

II. EXPERIMENTAL DETAILS

The samples used in this study were grown on n-type (100) GaAs substrates by solid-source molecular beam epitaxy in a Varian Mod Gen-II system. Gallium and indium were supplied by SUMO effusion cells. A valved arsenic

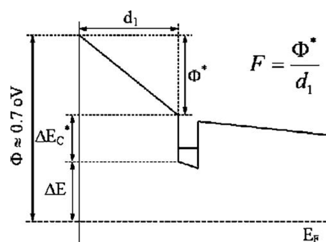

FIG. 2. Band bending in the modified Van Hoof structure containing a GaInNAs quantum well.

cracker supplied As_2. The GaInNAs QW was grown at a substrate temperature of 440 °C measured by pyrometry. An arsenic-to-gallium overpressure of 20× was supplied during the GaInNAs QW growth. Nitrogen was supplied by a modified SVT Associates plasma cell operating at a rf of 13.56 MHz. Nitrogen gas of 5N (99.999%) purity was filtered through a <1 ppb Pall Mini-Gaskleen purifier to minimize oxygen contamination. The cell was operated with 300 W input power and a nitrogen gas flow of 0.5 sccm. Other details of the growth are given elsewhere.[11] Two samples, one with low and one with high In content and almost the same nitrogen content, are analyzed in this paper. The structure for the two samples consists of a GaInNAs QW grown on a 300 nm thick GaAs buffer capped by a 50 nm thick GaAs layer. The QW thickness and content for the two samples were determined by using high-resolution x-ray diffraction and secondary-ion mass spectrometry.[11] It has been concluded that the low In content sample is ~7.0 nm thick QW with ~10% In and ~2.1% N whereas the high In content sample is ~7.5 nm thick QW with ~28% In and ~2.2% N.

A conventional experimental setup with a tungsten halogen lamp (150 W) as a probe light source, a 0.55 m monochromator, and InGaAs $p-i-n$ photodiode was applied for obtaining contactless electroreflectance (CER) spectra. Samples were mounted in a capacitor. The top electrode of the capacitor is a copper-wire mesh, which is semitransparent for light. This electrode was kept at a distance of 0.1–0.3 mm from the sample surface while the sample itself was fixed on the bottom copper electrode. A maximum peak-to-peak alternating voltage of ~1.8 kV was applied. The frequency of the ac voltage was 285 Hz. Phase sensitive detection of CER signal was made using a lock-in amplifier. Other relevant details of CER measurements are described in Ref. 12. The amplitude of electromodulation inside the sample appears close to the sample surface[12] and is weak (few or few tens of kV/cm) since most of the applied voltage drops in the air gap between the top electrode and the sample surface. Both the probe light and the electromodulation by the external electric field can be treated as a weak perturbation of the band structure for this system, especially at room temperature.

III. RESULTS AND DISCUSSION

Figures 3(a) and 3(b) show CER spectra for GaInNAs/GaAs QWs with low and high indium content, respectively. Two regions of optical transitions are visible for theses samples. The first region, below 1.4 eV, is associated with optical transitions between energy levels confined in the GaInNAs/GaAs QW. The second one, at 1.42 eV with strong FKOs, is associated with bulklike absorption inside GaAs epilayers. As it was mentioned in the Introduction, the analysis of QW transitions allows us to determine the band gap discontinuity at GaInNAs/GaAs interface, whereas the FKO analysis allows us to determine the built-in electric field inside GaAs epilayers.

QW transitions: CER resonances associated with QW transitions were analyzed using the low-field electromodulation Lorentzian line shape functional form,[13] as in our previ-

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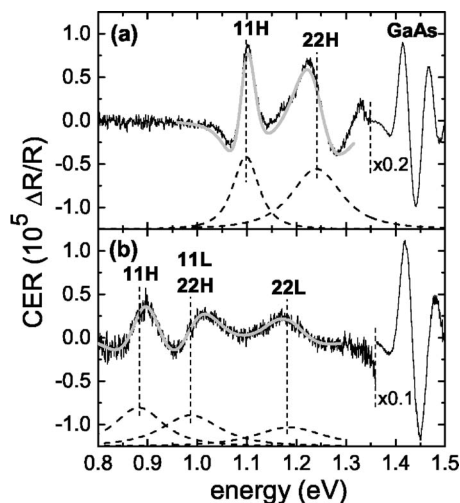

FIG. 3. Room temperature contactless electroreflectance spectra for modified Van Hoof structures with (a) $Ga_{0.9}In_{0.1}N_{0.021}As_{0.979}$/GaAs QW and (b) $Ga_{0.72}In_{0.28}N_{0.022}As_{0.978}$/GaAs QW.

ous papers.[14–17] The dashed lines in Fig. 3 represent the modulus of individual CER resonances (individual QW transitions). The notation $klH(L)$ in this figure denotes the transition between the kth heavy-hole (light-hole) valence subband and the lth conduction subband. The identification of the resonances is possible on the basis of the calculations performed in the framework of the effective mass approximation with appropriate material parameters. In the calculations, the conduction band offset (Q_C) can be treated as a free parameter. Note that the Q_C is the "so-called" chemical band offset and is defined by

$$Q_C = \frac{\Delta E_C}{\Delta E_C + \Delta E_V} \times 100\%, \quad (1)$$

where ΔE_C and ΔE_V are the discontinuities for conduction and valence bands of unstrained materials, respectively.[14,15] The conduction band offsets for the two samples, which are discussed in this paper, was analyzed in detail in Refs. 16 and 17. The band gap discontinuities at GaInNAs/GaAs interfaces were calculated, taking into account strain in the GaInNAs layer and material parameters after Refs. 18 and 19 (see details in Refs. 14 and 15). For the QW with low In content the conduction band offset was estimated to be ~55%, and the conduction and valence band discontinuities for this Q_C were calculated to be 194 and 202 meV, respectively.[16] In the case of GaInNAs/GaAs QWs with high In content and ~2% of N, the conduction band offset was investigated many times and is known to be ~80%.[20] For the QW with high In content, which is investigated in this work, the conduction band offset was estimated to be ~80%.[17] For this Q_C the conduction and valence band discontinuities were calculated to be 460 and 153 meV, respectively. The band gap line-up for the low and high indium content QWs is shown in Fig. 4.

GaAs absorption: A small increase of the FKO periodicity with the rise of indium content is seen in Fig. 3. In order to eliminate a possible photovoltaic effect,[10] the GaAs-related FKOs have been measured in dark configuration utilizing another experimental setup, which was described in

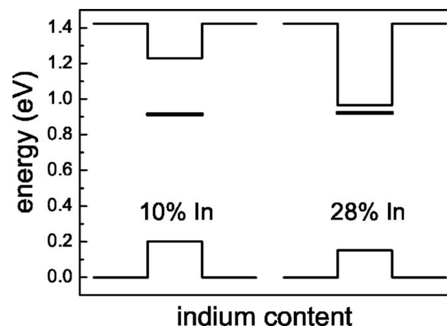

FIG. 4. Band gap line-up for $Ga_{0.9}In_{0.1}N_{0.021}As_{0.979}$/GaAs QW and $Ga_{0.72}In_{0.28}N_{0.022}As_{0.978}$/GaAs QW together with the Fermi level position in the QW region.

Ref. 21. Figure 5(a) and 5(b) show CER spectra in the vicinity of GaAs-related absorption for samples with low and high indium content, respectively. The oscillatory behavior at $E > E_0$, where E is photon energy and E_0 is band gap energy, can be described by an electro-optic function, whose asymptotic form[22] can be written as

$$\frac{\Delta R}{R}(E) \sim \cos\left[\frac{4}{3}\left(\frac{E-E_0}{\hbar\theta}\right)^{3/2} + \frac{\pi(d-1)}{2}\right], \quad (2)$$

where d is the dimensionality of the critical point and $\hbar\theta$ is the electro-optic energy. The extrema are given by

$$n\pi = \varphi + \frac{4}{3}\left[\frac{(E_n - E_0)}{\hbar\theta}\right]^{3/2}, \quad (3)$$

where n is the index of the nth extrema, φ is an arbitrary phase factor, and E_n is the energy of the nth extrema. The electro-optic energy is given by

$$(\hbar\theta)^3 = \frac{e^2\hbar^2F^2}{2\mu}, \quad (4)$$

where μ is the reduced interband effective mass for the electron and heavy-hole pair in the direction of electric field. If μ is known [μ is $0.055m_0$ for the (100) direction in GaAs after

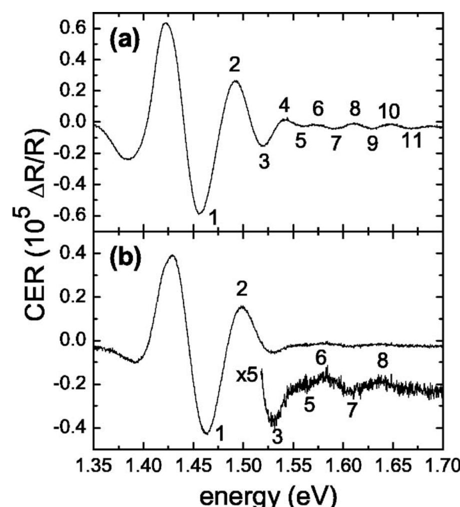

FIG. 5. Room temperature contactless electroreflectance spectra for modified Van Hoof structures with (a) $Ga_{0.9}In_{0.1}N_{0.021}As_{0.979}$/GaAs QW and (b) $Ga_{0.72}In_{0.28}N_{0.022}As_{0.978}$/GaAs QW in the vicinity of GaAs-related absorption.

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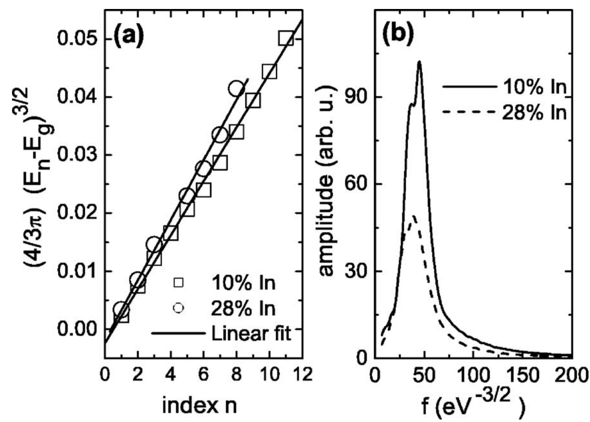

FIG. 6. Analysis of built-in electric fields: (a) a standard approach and (b) the Fourier transform approach.

Ref. 18], the built-in electric field, F, can be directly evaluated from a plot of $(4/3\pi)(E_n-E_g)^{3/2}$ as a function of index n [see Fig. 6(a)]. It has been calculated that the slope yields electric fields of ~58 and ~64 kV/cm for the samples with low and high indium content, respectively.

Sometimes the simple analysis of FKO extrema can be inappropriate since the GaAs signal can be composed of two or more FKOs with different periods. For the modified Van Hoof structures different built-in electric fields are expected in the cap and buffer layers [see Fig. 1(b)]. In such a case it is more appropriate to use a method that takes the Fourier transform (FT) of the EM spectrum[23] to obtain the respective oscillation periods of transitions in the cap and buffer layer. Figure 6(b) shows FT spectra of GaAs-related FKOs. Two separate peaks are resolved in FT spectra. However, it has been concluded that the two peaks are associated with light-hole–electron (LH) transition and heavy-hole–electron (HH) transition in the same GaAs layer since the frequency of LH and HH peaks yields the same electric field within the experimental error (10% In: ~55 kV/cm for the LH transition and ~56 kV/cm for the HH transition; 28% In: ~64 kV/cm for the LH transition and ~63 kV/cm for the HH transition). The electric fields, extracted from FT analysis, are in good accordance with those extracted from the conventional analysis, i.e., the analysis of FKO extrema. This finding is expected for FKOs originating from one layer with a homogeneous built-in electric field. In such a case, the LH contribution does not disturb HH-related extrema. Only small dimpling is observed in the FKO signal (see CER spectra at 1.55–1.6 eV and Refs. 10 and 23).

The measured electric fields are much higher than the electric field that is expected for unmodified Van Hoof structure (~19 kV/cm). It proves that the GaInNAs QW significantly modifies band bending in this system. The CER signal usually originates from the section of the sample that is close to the surface.[12] Therefore, the measured electric field is attributed to the GaAs cap layer in these samples. Much smaller built-in electric fields are expected for the GaAs buffer layer according to the following equation:

$$\Phi \approx F_1 d_1 + F_2 d_2, \tag{5}$$

where F_1 and F_2 are the built-in electric fields in the GaAs cap and buffer layers, respectively [see also Fig. 1(b)]. d_1 and d_2 are the thicknesses of the GaAs cap and buffer layers, respectively. Φ is the potential difference between GaAs surface and GaAs/(GaAs n-type substrate) interface. In the case of GaAs-based Van Hoff structures, like those studied in this paper, this difference was studied many times[10] (we assume that Φ at room temperature is 0.7 eV). Note that for other materials, an unmodified Van Hoof structure can be measured in order to determine this value.

On the basis of Eq. (5), it has been calculated that the electric fields in the GaAs buffer layer are ~14 and ~13 keV/cm for samples with low and high indium content, respectively. In this regime of electric fields any FKOs are rather unexpected. In the low-field regime a Lorentzian- or Gaussian-like resonance at 1.42 eV is usually observed. In this case, no signal from the GaAs buffer layer is resolved in CER spectra since the electomodulation in the buffer layer, i.e., far from the sample surface, is very weak.[12] Usually, it is easy to probe a buffer layer by photoreflectance (PR) spectroscopy because of the different mechanism of electromodulation in PR,[12] i.e., a generation of electron-hole pairs in the buffer and substrate layers and hence the electromodulation of band bending also at the GaAs (buffer)/GaAs (substrate) interface (far from the sample surface). But in order to determine the Fermi level position in the QW region for the modified Van Hoof structures, a signal from the GaAs cap layer and QW region should be observed first and foremost. A possible signal from the GaAs buffer layer can complicate the analysis of the built-in electric field. Note that in the case of PR spectroscopy, QW transitions and GaAs-related oscillations were observed many times for GaInNAs/GaAs QW structures,[24–26] but the Fermi level position in the GaInNAs QW region has never been determined for these samples since the origin of FKOs was not as obvious and easy to interpret. In the case of CER spectroscopy, the interpretation of the FKO signal is much easier. In addition, PR spectra for GaAs-based structures grown on n-type substrate usually exhibit a below-band-gap oscillation, which complicates or makes impossible the analysis of QW transitions and GaAs-related signal.[27–29] CER spectra are free of the below-band-gap oscillation[28,29] due to different electromodulation mechanism and therefore this technique is excellent to study both the QW transitions and built-in electric field in the GaAs cap layer.

The Fermi level position in the GaInNAs QW has been evaluated with knowledge of the electric field in the GaAs cap layer and band gap discontinuity in GaInNAs/GaAs QW, according to Fig. 2, and this position is plotted in Fig. 4. Two very interesting findings are visible in this figure. The first finding is that the Fermi level is located at the same energy, i.e., ~0.9 eV above the GaAs valence band. The second one is that the material becomes n-type with the rise of indium content. The last finding is in accordance with recent investigations of the Fermi level stabilization energy in group III nitrides.[30] We believe that systematic investigations of the Fermi level pinning in dilute nitrides, within the experimental approach proposed in this work, will provide a lot of information that can help us to understand fundamental properties of dilute nitrides. Our investigations for series of

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GaInNAsSb/GaAs QW samples with various contents are under way and will be presented in another paper.

IV. CONCLUSIONS

In conclusion, a very fruitful approach to study the Fermi level position in the GaInNAs QW region has been proposed in this work. This method utilizes CER spectroscopy and very simple design of the semiconductor structure. In this work the Fermi level pinning has been determined for as-grown GaInNAs/GaAs QWs with low (10%) and high (28%) indium content and nitrogen content of ~2.1% –2.2%. It has been observed that for as-grown GaInNAs/GaAs QW samples the Fermi level is located at the same energy, i.e., ~0.9 eV above the GaAs valence band. In addition it has been recognized that GaInNAs material becomes *n*-type with the rise in indium content since the Fermi level in the GaInNAs region shifts to the conduction band.

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