A new optical front-end compensation technique for suppression of spurious signal in photoreflectance spectroscopy using an antiphase signal

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A new optical front-end compensation technique to suppress the unwanted, spurious signal in photoreflectance (PR) spectroscopy is developed. In this approach an optical compensation signal, which is amplitude-matched to and in antiphase to the spurious signal, is introduced and directed to the photodetector. The combination of the compensation signal and the spurious signal results in a dc output from the photodetector which is thereafter suppressed by the lock-in amplifier, leaving only the true PR signal to be recovered and amplified. A high spurious signal suppression efficiency is demonstrated and the advantages of the technique are discussed. © 2010 American Institute of Physics. [doi:10.1063/1.3368598]

I. INTRODUCTION

Modulation spectroscopy is a powerful experimental technique in the investigation of semiconductor materials. Modulation spectroscopy can provide information on both the fundamental ground states and the excited states. Due to the derivative nature of the modulation spectra, they are sensitive to the critical points of the joint density of states and the unwanted background is suppressed. Photoreflectance (PR) spectroscopy is a kind of electromodulation spectroscopy in which the reflectivity of the sample is modulated by the screening of the built-in electric field by photoexcited electron-hole pairs. PR has several advantages as it is contactless, nondestructive, and experimentally simple and has been widely adopted in the study of semiconductor materials.

II. EXPERIMENTAL CONCEPT AND SET-UP

Figure 1 shows a schematic diagram of a typical PR setup. A laser beam is used to excite electron-hole pairs in a sample. The electron-hole pairs screen the built-in electric field in the sample and result in a change in the reflectivity (ΔR) of the sample. ΔR is probed by a probe beam. The reflected probe beam is detected and converted to electrical signal by a photodetector. The laser beam is usually modulated by a mechanical chopper or an acousto-optic modulator with a modulation frequency ω. ΔR can then be retrieved and recovered by a lock-in amplifier (LIA) according to the modulation frequency. As shown in Fig. 1, the optical signal S that reaches the photodetector consists of three contributions: S = I·R + I·ΔR·M(ωt) + SPE·M(ωt), where I is the intensity of the probe beam, R the unaffected reflectivity, and M(ωt) the modulation function which has a square waveform with the minimum value as 0 and the maximum value as 1. $SP_E$ includes the scattered laser excitation beam (EB) and the photoluminescence (PL) excited by EB. Note that there is actually a phase difference between the scattered EB and the PL, since the PL has a buildup trail after the laser beam is on and has a quenching trail after the laser beam is off. However, the luminescence buildup time is usually at the magnitude of picosecond and the quenching time is usually at the magnitude of nanosecond in semiconductors, which are negligible when a mechanical chopper which works at a frequency of a few hundreds or thousands Hz is used. The optical signals reach the photodetector and are converted into electrical signals and the electrical signals are sent to a LIA. The first term, I·R, is a dc signal and results in a dc detector output which will be rejected by LIA. The second term, I·ΔR·M(ωt), and the third term, $SP_E·M(ωt)$, have the modulation frequency ω dependence and produce ac detector outputs which can be recovered and amplified. $I·ΔR$ corresponds to the change in the reflectivity ΔR, while $SP_E$ is a spurious signal since it has nothing to do with ΔR. The spurious signal is a common problem in the PR spectroscopy. It distorts the PR spectra, and even submerges the PR signal if it is much stronger than the PR signal, which is usually the case in semiconductor nanostructures. There have been many approaches to suppress the spurious signal in PR spectroscopy, such as sweeping PR, dual chopped PR, electrical front-end compensation (EFEC) PR, employing a subtraction scheme with two detectors, and using a Fourier-transform spectrometer. These techniques either require complicated configurations, or have additional requirements on the samples or the electronic components. In this work we propose a new optical front-end compensation (OFEC) technique to suppress the spurious signal in PR spectroscopy. The OFEC PR setup is experimentally simple and a high spurious signal suppression efficiency can be achieved. The key idea of OFEC PR is, as shown in Fig. 2, to use an additional laser beam to produce an optical signal that is amplitude-matched to and in antiphase to the spurious signal.

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produced by the excitation laser and convert it into a dc signal, which then can be rejected by the phase-sensitive LIA. This paper presents the principle, the implementation, and the operation of OFEC PR. As a demonstration, PR spectra of a GaAs/AlGaAs single quantum well (SQW) sample are shown and discussed. Comparison of OFEC PR with other spurious signal suppression methods is made and the advantages of OFEC PR are discussed.

Figure 3 shows the schematic block diagram of the experimental configuration. The setup is similar to the normal PR setup reported in Ref. 8. The probe beam comes from a 150 W tungsten lamp and is dispersed by a 0.3 m monochromator. The laser beam is divided into two beams, one for excitation (EB) and the other for compensation (CB). EB and CB pass through different positions on the chopper blades. EB is directed to the sample to produce modulations and coincides with the probe beam spot on the sample. CB is also focused onto the sample but away from the probe and EB beams. Note that CB also excites the sample and produces a fluorescence excited by CB.

The optical signal that reaches the detector consists of four contributions

\[ S = I \cdot \Delta R \cdot M(\omega t) + I \cdot R + S_{PE} \cdot M(\omega t + \theta), \]

(1)

where \( S_{PC} \) is the compensation signal produced by the CB. Like \( S_{PE} \), \( S_{PC} \) includes the scattered CB and the luminescence excited by CB. \( \theta \) is the phase difference between \( S_{PE} \) and \( S_{PC} \) since the EB and CB pass through different positions on the chopper blades. By carefully choosing the position of CB on the chopper blades by adjusting the position of mirror M1 in Fig. 3, \( \theta = \pi \) can be achieved. Through tuning of the CB intensity by adjusting the variable neutral density filter (VNDF) in Fig. 3, we can adjust the intensity of spurious signal produced by CB to be equal to that produced by EB, that is, \( S_{PE} = S_{PC} \). Under such conditions Eq. (1) becomes

\[ S = I \cdot \Delta R \cdot M(\omega t) + I \cdot R + S_{PE,C}. \]

(2)

That is, the spurious signal term and the compensation signal term in Eq. (1) merge into one dc term \( S_{PE,C} \) in Eq. (2) which, together with the second dc term \( I \cdot R \), is rejected by the LIA. Only the term \( I \cdot \Delta R \cdot M(\omega t) \) which corresponds to the change in reflectivity caused by photoexcitation, contains the modulation frequency and is recovered and amplified by LIA.

The compensation operation procedure is as follows: (1) Block the probe beam and the CB. The LIA shows only the spurious signal, \( S_{PE} \). (2) Block the probe beam and EB, then the LIA shows the compensation signal produced by CB. Adjust the position of CB on the chopper blades by changing the position of mirror M1 to get the signal produced by CB to be in antiphase to the spurious signal produced by EB. (3) Block the probe beam and unblock EB and CB, adjust the VNDF so that the amplitudes of \( S_{PE} \) and \( S_{PC} \) match each other and results in a zero LIA output. (4) Unblock the probe beam and measure the PR spectra as usual. Note that it is easy to switch between the normal PR and the OFEC PR by blocking/unblocking CB.

As a demonstration of the OFEC PR technique, a GaAs/AlGaAs SQW sample grown by metal-organic chemical-vapor deposition was used. A 25 nm AlGaAs/2 nm GaAs/25...
nm AlGaAs SQW was grown on a semi-insulating GaAs substrate. On the top of the structure a 5 nm GaAs layer was deposited to protect the sample from oxidation. The nominal Al composition is 0.54. The sample was mounted in a cryostat and the temperature of the sample was kept at 77 K during measurements.

III. RESULTS AND DISCUSSION

Figure 4 shows the PR spectra (I·ΔR) of the sample obtained at 77 K using the normal PR and the OFEC PR. The LIA sensitivity was set to 5 nA for normal PR and 1 nA for OFEC PR. The data are presented in the form that their values correspond to the original detector output and conversions have been performed between the LIA output and the detector output. The value of the corresponding LIA output is also marked on the right y-axis. The wavelength of the laser used for modulation is 635 nm and the laser photon energy is indicated in Fig. 4 by an arrow below the OFEC PR spectrum. Both spectra show the same features. Note that a PR resonance at an energy higher than the laser photon energy is observed, indicating that the detectable wavelength range of the PR spectroscopy is not limited by the laser wavelength. Such limitation usually occurs in emissionlike spectroscopy such as PL. The normal PR spectrum shows a high base line of the spurious signal background, which consists of both the scattered laser beam and the luminescence from the sample. The magnitude of the spurious signal is even larger than that of the PR signal. The spurious signal dramatically distorts the PR spectrum when performing the normalization process. Although the spurious signal caused by the scattered laser can be removed by placing a long-pass optical filter with a cutoff wavelength longer than the laser wavelength before the detector, it will sacrifice the detectable wavelength range by limiting it to a wavelength longer than the laser wavelength. By using the OFEC PR configuration, the spurious signal recovered by LIA is greatly suppressed in the PR spectrum due to the compensation effect of CB. The magnitude of the recovered spurious signal decreases from ~1.2 nA in the normal PR spectrum to ~0.013 nA in the OFEC PR spectrum, a reduction of two orders in magnitude. The reduction in the spurious signal allows a more sensitivity range on the LIA, which increases the reliability of the signal and improves the system sensitivity. In the experiments the amplitude of the PR signal is about ~0.2 nA. A 5 nA LIA sensitivity range was chosen for the normal PR while a smaller 1 nA LIA sensitivity range, which is closer to the PR signal amplitude, was chosen for the OFEC PR. The features of the OFEC PR spectra are similar to the normal PR spectra except for the spurious signal background, indicating that the CB has no effect on the reflected probe beam since there is no overlap between them.

Figure 5 shows the (a) OFEC PR, (b) reflectance, (c) normalized PR, and (d) PL spectra taken at 77 K. Note that the PR spectrum was taken with a red laser with a wavelength of 635 nm while the PL spectrum was carried out with a green laser with a wavelength of 532 nm. The PL spectrum shows two main emission peaks, the peak at 1.511 eV corresponds to the GaAs substrate and the emission at 1.891 eV is attributed to the confined SQW state. The normalized PR spectrum shown in Fig. 5(c) is obtained by dividing the OFEC PR spectrum (I·ΔR) over the reflectance spectrum (I·R). Note that since the spurious signal is suppressed, it is easy to obtain the real-time normalized ΔR/R directly in the mea-
measurement system. This can be achieved by either employing a servomechanism driven VNDF at the probe beam or using a second voltmeter/current amplifier to obtain the reflectance data. PR features around 1.5 eV are attributed to GaAs-related transitions and Franz–Keldysh oscillations due to the built-in electric field in the GaAs substrate/layer. However, the assignment of these PR features is beyond the scope of this paper. The PR resonance at 1.916 eV comes from the SQW confined state. Note that for the SQW confined state there is a difference of 25 meV between the energy determined from the PR spectrum and that determined from the PL spectrum. This can be attributed to the effect of carrier localization caused by the conduction band minima that form due to well width fluctuations. This effect is especially significant in thin QWs and at a low temperature, which is the case for our study (well width=2 nm and temperature =77 K). The PR resonance at 2.208 eV comes from the AlGaAs barriers, which does not appear in the PL spectrum due to the indirect band gap of the barriers.

As mentioned before, several techniques have been developed to suppress the spurious signals in PR spectroscopy. In the dual chopped scheme both the pump beam and the probe beam are chopped at different frequencies using two optical choppers and the signal detection is taken at the sum frequency or the difference frequency. The scheme suffers from signal losses and the synchronization requirements between the two optical choppers add complexity to the system. Sweeping PR (Ref. 3) technique replaces the optical chopper with an acousto-optic modulator and sweeps the pump beam spot with respect to the probe beam spot to produce a dc PL signal. However if the sample is inhomogeneous a residual ac PL signal will remain at the modulation frequency and distort the PR signal. The double detectors subtraction approach requires two detectors and any mismatch between the two detectors will result in residual spurious signal. The EFEC PR (Ref. 10) adopts a phase shifter/amplifier to produce and feed an electrical compensation signal into the LIA differential input. This requires that the PR signal fed into the LIA input must be a voltage signal and the LIA must have a differential input. The time response characterizations of the photodetectors need to be taken into account in the compensation signal generation, which results in complexity in the compensation processes. The Fourier-transform approach uses inverse Fourier transform to reject the pump laser and the PL signal. The PR signal is obtained by calculating the inverse Fourier transform of an interferogram. It is difficult to perform real-time normalization in such a process. The OFEC PR presented here has several advantages. First, the implementation of OFEC PR is experimentally simple. Only a few mirrors and lens are needed to add to an existing standard PR setup to turn it into an OFEC PR setup so the additional cost is minimal. Second, the compensation of the spurious signal is implemented within the optical domain so there are no additional requirements for the photodetectors and electronic instruments. Lastly, it is easy to realize real-time normalization in the OFEC PR setup.

IV. SUMMARY

In summary, a new, low cost, and easy-to-implement OFEC technique is developed for suppression of spurious signal in PR spectroscopy. An additional compensation beam is applied to produce signal that is in antiphase to the spurious signal and converts it into a dc signal which is thereafter rejected by the LIA. High spurious signal suppression efficiency up to two orders of magnitude is achieved. The advantages of OFEC PR lie in its high spurious signal suppression efficiency without signal loss and experimental simplicity.

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