Note: Improved sensitivity of photoreflectance measurements with a combination of dual detection and electronic compensation

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A setup is described for performing photoreflectance (PR) measurements using two photodetectors, wherein the photodetectors need not be identical. The second detector monitors the photoluminescence and scattered pump laser background signal in real time. It is then eliminated from the measured PR signal using electronic circuits that compensate for amplitude and phase differences between the background signals from the two detectors. The technique overcomes the adverse effect of short-term fluctuations in the pump laser intensity. The signal-to-noise ratio is shown to improve significantly, enabling measurement of weak PR signals. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4704087]

Photoreflectance spectroscopy (PR) is an important tool for the study of electronic band structure (EBS) and non-destructive characterization of semiconductors, especially quantum confining heterostructures such as quantum wells (QW) and superlattices. In PR one perturbs the sample using a pump beam which is typically a laser. The photon energy of the pump is fixed and is chosen to be larger than the bandgap of the material under study. The absorbed pump photons generate electron-hole pairs which in turn modify the built-in electric field at the surface or interfaces. The consequent dielectric function change results in a reflectivity change $\Delta R$, which is measured using a second probe beam of variable photon energy. $\Delta R$ peaks around energies where critical points occur in the EBS. This enables the determination of the fundamental and higher direct bandgap energies. The relative change in reflectivity is typically small ($\Delta R/R \sim 10^{-2} - 10^{-4}$) and therefore one has to periodically chop the laser and perform phase sensitive detection of the signal proportional to $\Delta R$, which arises at the chopping frequency, using a lock-in amplifier (LIA). Since the PR signal scales as a fractional power of the pump intensity, to detect weak signals one may need to increase the pump laser intensity considerably. This poses a problem since typically in direct bandgap materials of high crystalline quality, the pump laser also generates strong photoluminescence (PL) at the chopping frequency due to radiative recombination of the electron-hole pairs. A large background signal due to PL, and possibly scattered pump laser that might have leaked through the optical filters used, forces one to measure the PR signal at a poorer sensitivity scale on the LIA, which is undesirable. The more serious problem is that since PL output mostly varies linearly with laser intensity therefore small short-term fluctuations in the laser intensity, which do not directly affect the PR signal, can create variations in the background signal that are comparable in magnitude to the PR signal. Such noise can render measurement of weak PR signals very difficult.

There have been several proposals for reducing this background and they can be broadly divided into four categories. The first category involves spectral filtering of the PL signal using a second monochromator, a solution which is both expensive and leads to a loss of PR signal strength. The second category involves optical compensation. Here, the PL signal is made dc, for example, by sweeping the laser across the probe beam spot rather than chopping it. In another case this is achieved by introducing a third chopped light beam of appropriate intensity and out of phase with the background in order to nullify it. The third beam is generated either by a second laser or a reflected fraction of the pump laser, but these do not necessarily ensure improvement in the signal to noise (S/N) ratio. The third category involves suppression of the background using signal detection/processing techniques. An example is dual chopping where the pump and the probe are both chopped and the PR signal is detected at the summed chopping frequency, thereby avoiding the PL signal which arises at the pump chopping frequency. This technique suffers from noise due to phase jitter between the two chopping processes. Another such technique involves Fourier transform based detection which is useful mainly in the infrared. Simultaneous detection of signal over a range of wavelengths using an array detector can reduce measurement time to seconds. This can make PR measurements insensitive to pump beam intensity fluctuations over time scales longer than seconds and has been shown to work well for large signal strengths. The fourth category involves pure electrical compensation. Here, a fixed voltage equal to the PL background signal is subtracted, but it cannot compensate for fluctuations and drift in the background and so the S/N ratio does not improve. A dual detector technique has also been proposed to subtract out a separately detected PL background, but it required two identical detectors placed close to each other so as to record identical PL signals. In this Note we report on a scheme for nullifying the PL background signal in PR which involves a combination of dual detection and electronic compensation. It overcomes the requirement of identical detectors and also compensates for fluctuations...
and drift in the background, which helps improve S/N ratio considerably.

Figure 1 shows a schematic of our experimental setup for PR spectroscopy. The probe beam is obtained by dispersing light from a 100 W tungsten lamp using a 0.5 m focal length monochromator with a bandpass of 0.6 nm. The pump was a 532 nm frequency doubled YAG laser. The conventional PR measurement mode uses a single infrared sensitive Si photodiode detector D1. The ac part of the signal from D1, after the current-to-voltage (I-to-V) conversion stage, is measured using a LIA and has the component proportional to \( I_0 \)R. The high crystalline quality of the sample gives rise to a large PL background signal of \( \sim 18.5 \) mV. In this wavelength range, the average dc signal \( \propto I_0 R \) in the absence of the pump beam was \( \sim 160 \) mV. The separately measured PL spectrum is also shown.

Figure 2 shows signal from a low temperature PR measurement on a GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As QW sample (well width 4.6 nm) grown using metal-organic vapor phase epitaxy. Its PL spectrum, measured separately, is also shown. This PR spectrum was measured using the conventional single detector approach. In the PR spectrum, one can easily identify dominant features associated with the first electron-heavy hole (e1lh1) and electron-light hole (e1lh1) transitions in the QW. Although the pump power was only \( \sim 50 \) \( \mu \)W on the sample, it resulted in a relatively large background signal \( \sim 18.5 \) mV, with root-mean-squared (rms) fluctuation of \( \sim 74 \) mV. While this corresponds to only a 0.4% change in the background, it is comparable in magnitude to signals corresponding to the weaker spectral features, making their measurement impossible. In the above wavelength range, in the absence of the pump beam the average dc signal was \( \sim 160 \) mV. A large background also introduces both noise and errors in the measurement of the dc signal, which ideally should just be proportional to \( I_0 R \).

To study this background signal in PR and fluctuations therein, we again used the optical arrangement shown in Fig. 1 but this time using both the detectors D1 and D2. However, unlike in Fig. 1, we measured signals from D1 and D2 separately, just after the I-to-V converter stages, using two different LIA. D2 is a UV-sensitive Si photodiode, deliberately chosen to be different from D1. It is positioned so as to completely avoid the reflected probe beam. With the probe beam switched off but the chopped pump laser beam on, the signal detected by the two detectors were recorded as a function of time. The total duration was kept similar to the time required for acquiring the data in Fig. 1. These signals are plotted in Fig. 3. It is evident that the fluctuations in the two signals are correlated in time and similar in magnitude. They are also comparable to the background signal in Fig. 1 and fluctuations therein. This suggests that the observed fluctuations in the PR signal background did not arise from the detection process, but instead has an optical origin. Since the probe beam was off and scattered pump laser was blocked, this background is due to the total PL output from the sample. The PL output has an angular spread \( \propto \cos^2 \theta \), where \( \theta = 0^\circ \) is normal to the

![Figure 1](image1.png)

**Figure 1.** Schematic of the setup for PR spectroscopy involving two detectors D1, D2 and electronic compensation. The detectors need not be identical. Also shown are the paths of the pump laser beam and the probe whose wavelength can be varied. L are focusing lenses and F are laser blocking long-wavelength pass filters. The dashed curve represents the angular dependence of the PL intensity. Signal from D2, which does not see the probe beam, goes through additional compensation electronics before being added to the signal from D1. The resultant signal is fed to a lock-in amplifier (LIA).

![Figure 2](image2.png)

**Figure 2.** Raw PR spectrum (signal \( \propto I_0 \Delta R \)) from a GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As QW sample, measured using a conventional arrangement involving only the detector D1 in Fig. 1. The high crystalline quality of the sample gives rise to a large PL background signal of \( \sim 18.5 \) mV. In this wavelength range, the average dc signal \( \propto I_0 R \) in the absence of the pump beam was \( \sim 160 \) mV. The separately measured PL spectrum is also shown.

![Figure 3](image3.png)

**Figure 3.** Time dependence of signals measured simultaneously from the two detectors D1 and D2 in the configuration shown in Fig. 1, by using two lock-in amplifiers. Signals were measured just after the current-to-voltage converter stages. The laser pump intensity was the same as in Fig. 2, but the probe beam was absent.
sample surface and is therefore detected by both detectors. The fluctuations in the PL signal are due to small short-term variation in the pump laser intensity.

The above observation suggests that by using signals from D2 it is possible to eliminate the background from the PR signal. This requires the background measured by D1 and D2 to be identical in magnitude and phase. However it is not necessary to have identical detectors, and that too specially mounted, to get identical background signal strengths. Instead, we have used electronic compensation circuits to do this. Not requiring identical detectors is very useful because for instance if one is measuring the main PR signal with a photo-multiplier tube as D1, one can then use an inexpensive and easy to mount Si photo-diode as D2 for PL background correction. In this measurement scheme, shown in Fig. 1, the signal from D2 in the absence of the probe beam is made identical in magnitude and ±180° out of phase with the signal from D1. The sum of these two signals is then measured by a LIA which will now record a PR signal without a PL background when the probe beam is switched on. This will also eliminate the problems with measuring the true dc signal proportional to IoR.

For conditioning the signal from D2 it is first passed through an inverting attenuator/amplifier and phase shifter. The circuit is shown in Fig. 4(a), its gain can be varied between −0.22 and −10.22. The 100 kΩ, 10 turn variable resistor is used to finely adjust the gain such that output at Vout1 matches the voltage output of D1 in magnitude when the probe beam is absent. If the output at Vout1 is exactly ±180° out of phase with the voltage output from D1 in the absence of the probe beam, then these two outputs can be fed directly to the voltage adder shown in Fig. 1. Else the signal from D2 needs to pass through the phase shifter. This circuit is also shown in Fig. 4(a) and has two parts. The first part with output at Vout2 provides a variable phase shift. The second part with output at Vout3 adds an additional 180° phase shift when required. The variable phase shift θ depends on the resistor Rp and capacitor Cp values as \( \theta = -2\tan^{-1}(2\pi f R_p C_p) \), where f is the signal frequency which in our case is the same as the laser chopping frequency. The variation of θ with Rp for certain values of Cp and f = 175 Hz is plotted in Fig. 4(b).

Figure 5 shows the PR spectrum of the GaAs/Al0.3Ga0.7 As QW sample measured using the two detector plus electronic compensation scheme. Although the pump power was the same as in Fig. 1, the background signal in this case is negligible. The smaller signals here could be measured on the 100 μV sensitivity scale of the LIA which provides better resolution compared to the 20 mV scale on which one was forced to take the data shown in Fig. 2. More importantly, the rms noise riding on the signal is now only ~2.3 μV, which represents a factor of more than 32 improvement in the S/N ratio compared to the data in Fig. 1. This enables better observation of weaker features in the PR spectra and one can now clearly see two additional features in the spectrum of this sample which are associated with the two-dimensional continuum edges of the heavy-hole and light-hole excitons. To conclude, the dual detection plus electronic compensation scheme for PR spectroscopy is an easy to implement and cost-effective solution for overcoming the problem of PL backgrounds in PR. It can help improve S/N ratio significantly by nullifying the detrimental influence of short-term variation in the pump laser intensity, thereby enabling measurement of weak PR spectral features.

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