Double AC Photoreflectance Spectroscopy of Semiconductors

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Abstract—We report a new way of making photoreflectance (PR) measurements to overcome the problem of photoluminescence (PL) that is often encountered in low temperature PR measurements. In conventional PR, the probe beam is dc while the pump beam is chopped and in phase detection is done at that chopping frequency. At low temperatures a large PL signal arises at the chopping frequency of the pump beam (a laser) and swamps the PR signal. We overcome this problem by chopping both the pump beam as well as the probe beam at two different frequencies and detecting the PR signal at the sum frequency. This way we avoid the PL signal which now comes at a frequency which is not the frequency of detection. In this paper we discuss the details of this technique and present some low temperature PR data on epitaxial GaAs and GaAs-In,Ga1-x,As-GaAs strained layer quantum wells along with fitted lineshapes to show the feasibility of this technique.

I. INTRODUCTION

MODULATION spectroscopy is today an important experimental technique for the study of semiconductors and their microstructures [1]. One of the most popular modulation techniques in use is photoreflectance (PR) because it is a contactless measurement technique requiring no special mounting. Herein a laser beam is used for modulation. The absorbed laser beam creates electron hole pairs at the surface of the sample which in turn modulates the surface electric field. An alternating electric field modulates the dielectric function of the material which shows up as a change in reflectivity of the material. The reflectivity of the material is then probed as a function of energy using another light beam whose energy can be varied continuously. The spectrum obtained by this technique has a derivative like nature whereby an alternating reflecting surface can be measured. The spectrum of the incident photon energy is then given by

\[ R_s(E) = R_{so}(E) + \frac{2}{\pi} \Delta R_s(E) \sin (\omega_1 t) \]  

(1)

where \( R_{so}(E) \) is the reflectivity of the sample in the absence of the pump beam. \( \Delta R_s(E) \) is the change in reflectivity due to the pump beam. We have neglected the other dc term \( (\Delta R_s(E)/2) \) in (1) as it is much smaller than \( R_{so}(E) \). The probe beam intensity is dc and so the signal \( S_1 \) from the detector will be given by

\[ S_1 = I_0(E)R_{so}(E) + \frac{2}{\pi} I_0(E)\Delta R_s(E) \sin (\omega_1 t) \]

(2)

where \( I_0(E) \) is the probe beam intensity and includes the measurement system’s response to incident photon energy including that of the lamp, the monochromator, the optics and the detector. Thus if one locks on to the frequency \( \omega_1 \), one can measure a signal proportional to \( 2/\pi I_0(E)\Delta R_s(E) \). Then dividing this spectrum by another simple reflectivity spectrum which yields \( 2/\pi I_0(E)R_{so}(E) \), one gets the required PR spectrum in the form of \( \Delta R_s/R_{so} \) as a function of energy. However the problem with this technique is that at low temperatures the pump beam excites PL from the sample which therefore comes at the same frequency at which detection is being made. This PL signal can be just an order of magnitude smaller than the direct probe beam signal \( 2/\pi I_0(E)R_{so}(E) \), but since \( 2/\pi I_0(E)\Delta R_s(E) \) signal is normally three to four orders of magnitude smaller than the \( 2/\pi I_0(E)R_{so}(E) \) (as \( \Delta R_s/R_{so} \sim 10^{-3} - 10^{-4} \), therefore...
the PR signal is completely swamped by the PL background. To overcome this problem we adopted the double ac PR technique (henceforth to be referred to as DACPR) wherein both the pump and the probe beam are now chopped at frequencies $\omega_1$ and $\omega_2$ respectively. The probe beam is now a rectangular waveform and considering its first harmonic only, the signal $S_2$ from the detector will be given by

$$S_2 = \frac{2}{\pi} I_0(E) \sin(\omega_2 t + \phi) \times R_s(E)$$

(3)

where $\phi$ is the phase difference between the first harmonic of the ac component of reflectivity and that of the probe beam at $t = 0$. Upon expansion using (1), (3) becomes

$$S_2 = \frac{2}{\pi} I_0(E) R_{so}(E) \sin(\omega_2 t + \phi)$$

$$+ \frac{2}{\pi^2} I_0(E) \Delta R_s(E) \cos((\omega_1 + \omega_2)t + \phi)$$

$$+ \frac{2}{\pi^2} I_0(E) \Delta R_s(E) \cos((\omega_1 - \omega_2)t - \phi).$$

(4)

Thus if in-phase detection is made with a reference signal at frequency $\omega_1 + \omega_2$ ($\omega_1 - \omega_2$ will have larger 1/f noise), which is generated by the same chopper that chops the pump and the probe beam, then one can measure a signal proportional to $(2/\pi^2) I_0(E) \Delta R_s(E)$. However, since we are no longer locked onto the frequency $\omega_1$ at which the PL signal arises, it does not hamper our measurement. Again dividing this spectrum by a $(2/\pi) I_0(E) R_{so}(E)$ spectrum we get a $1/\pi(\Delta R_s/R_{so})$ spectrum as a function of energy. We therefore find that the signal in a DACPR spectrum is $1/\pi$ times that in a conventional PR spectrum.

III. EXPERIMENT

Fig. 1 shows a schematic of the setup for the DACPR measurement. A 4-mW He-Ne laser filtered by a line filter, which selects the 632.8-nm line, acts as the pump beam. After being reflected by the mirror M1 it is chopped at frequency $\omega_1$ by the set of slots on the inner side of the chopper blade. The chopper blade has two sets of slots on it, six slots on the outer side and five slots on the inner side so that the chopping frequency of the pump beam is $\omega_1 = 5\omega_0$, where $\omega_0$ is the frequency of the chopper motor. The chopped laser beam is then focussed by the lens L2 before being made incident on the sample by mirror M2. The sample sits on a vertical mount on the cold head of a closed cycle helium refrigerator which can cool the sample down to 10-K. The probe beam is obtained by passing light from a 250-W quartz-tungsten-halogen lamp through an ac motor driven 1/8 in monochromator. The probe beam is then chopped by the outer slots of the chopper blade at a frequency $\omega_2 = 6\omega_0$. Thereafter it is focussed by lens L1 and made incident on the sample after reflection from mirror M3. After being reflected from the sample the probe beam is focussed by lens L3 onto a Si detector. F1 is a low pass optical filter which acts as an order sorting filter in front of the monochromator exit slit. F2 is also a low pass optical filter which blocks the scattered laser light from reaching the detector but passes the probe beam. The current signal from the detector is then converted into a voltage signal by the current preamplifier and is then passed through a unity gain band pass filter tuned at $\omega_1 + \omega_2$ having a Q of 50. The band pass filter is necessary because the probe beam is no longer dc and although we are not locked at the frequency at which the probe beam is being chopped nevertheless it saturates the input amplifier stage of the lock-in amplifier making it impossible to go down to $\mu$V scale on it. After the band pass filter the signal is fed to a lock-in amplifier, the reference to which is a rectangular wave at frequency $\omega_1 + \omega_2$, generated by the chopper controller from $\omega_1$ and $\omega_2$ itself. The lock-in amplifier is interfaced to a computer which reads and records the data.

In an actual experiment, the PR signal was of the order of 30 mV in the samples studied ($\Delta R_s/R_{so} \sim 10^{-4}$), while the PL signal at 12 K was $\sim 4.5 \mu$V, more than two orders of magnitude larger than the PR signal. This made it impossible to make a conventional PR measurement on these samples at 12 K, but by using the DACPR technique we were able to make measurements down to 12 K. However we find that the
PL signal as well as the probe beam has a component at the frequency $\omega_1 + \omega_2$ which has a definite phase relationship to the reference being fed to the lock-in amplifier. This background, at frequency $\omega_1 + \omega_2$, is of the same order of magnitude as the PR signal. We suspect that this arises due to the nonuniformity of the chopper blades, which therefore gives rise to a Fourier component at frequency $\omega_1 + \omega_2$ since it also happens to be the 11th harmonic of $\omega_0$, the frequency of the chopper motor. Since the intensity of the probe beam is dependent on the incident photon energy, so is this background. To overcome this problem we make two sets of measurements. In the first set, the probe and the pump beam are focussed on the same spot and we get a reading which has the PR signal as well as the background. In the next measurement the pump beam is moved away from the spot where the probe beam is focussed. This way we measure only the background and not the PR signal. The actual PR data is then obtained by subtracting the background readings from earlier set which had background and signal.

IV. RESULTS AND DISCUSSION

We have used the DACPR technique to investigate different semiconductor samples. Here we present data on two of them which have large PL signal at low temperatures, making it impossible to do a conventional PR measurement on them. Fig. 2(a) shows the conventional single ac PR spectrum of a GaAs-In$_x$Ga$_{1-x}$As-GaAs strained layer quantum well ($L_w \sim 100 \text{ Å}, x \sim 0.2$) at room temperature and Fig. 2(b) shows the PR spectrum of the same sample using the DACPR technique. We see that the main spectral features seen in the conventional PR spectrum are faithfully reproduced in the DACPR spectrum but the background in the latter has more noise. The increased noise arises mainly from the additional electronics in the form of a band pass filter that the DACPR technique requires, also the final spectrum is obtained by subtracting two spectra and hence the noise gets added.

The feature due to transition between the $n = 1$ conduction band state to the $n = 1$ heavy hole state in the quantum well is clearly identifiable in both the spectra and is denoted by elhh1. For a bound system like electrons and holes in a quantum well, the PR spectrum would be a first derivative spectrum [1]. Using the expression for the dielectric function which has been shown to have a Lorentzian broadening [10], one gets to a lineshape function of the form [1], [5]:

$$L(E) = \text{Re} \left[ \frac{A \exp(i\theta)}{(E - E_0 + i\Gamma)^m} \right]$$

with $m = 2$. In the above equation, $\theta$, $E_0$ and $\Gamma$ are the phase factor, transition energy, and the broadening parameter respectively and $A$ is a constant. This function (with $m = 2$) fits the elhh1 transition quite well (not shown here) and gives the values for $E_0$, $\Gamma$ and $\theta$ as 1.18 eV, 5.3 meV, 1.61 rad, respectively, in the conventional PR spectrum and 1.18 eV, 5.5 meV, 1.84 rad, respectively, in the DACPR spectrum. The feature at 1.42 eV is due to the GaAs barrier layer. The feature to the left of the GaAs signature could be due to interference effects [10]. These along with the other features between 1.38 and 1.2 eV are currently under investigation.

Fig. 3 shows the low temperature (12 K) DACPR spectrum of the same quantum-well sample whose room temperature spectrum is shown in Fig. 2(a) and (b). The phase change of $\pi$ of the low temperature spectrum w.r.t. the room temperature data is an artefact due to electronics which was corrected for in the spectra measured later. At 12 K, the total PL signal
from this sample was 4.5 mV while the DACPR signal was 12 μV for the 1lh1 transition. Thus although a conventional PR signal would have been about three times larger than the DACPR signal, still it would have been about two orders of magnitude smaller than the PL signal and so it was not possible to do a conventional PR measurement. However in the DACPR spectrum one can clearly see the features due to transition between the states in the quantum well some of which are more pronounced than what they were at room temperature. The structure near 1.52 eV is due to GaAs. The inset shows the lineshape function of (5) with $m = 2$, fitted (dashed lines) to the 1lh1 transition data, which was taken with a slower scan speed and a larger time constant at a different spot on the sample. The best fit values of $E_0$ and $\Gamma$ were found to be 1.267 eV and 6.6 meV, respectively. The transition energies indicate strain relaxation in the well layer.

Fig. 4 shows the 12-K DACPR spectrum of epitaxial GaAs. The inset shows a room temperature conventional PR spectrum of the same sample. Here too, a large PL signal prevented the conventional PR measurement at 12 K. In the DACPR spectrum of this sample at 12 K, we see none of the Franz-Keldysh oscillations that were seen at room temperature. For a transition involving a 3-D critical point like that at the band gap of GaAs, the lineshape function for low modulating fields is proportional to the third derivative of the dielectric function [1], [4], [5]. With a Lorentzian broadening of the dielectric function, the lineshape function has the same form as that given by (5), with $m = 2.5$. But if the transition is excitonic in nature then one expects a first derivative spectrum [1]. With the appropriate Lorentzian form of the dielectric function for discrete excitons [5] one again gets a lineshape function of the form given in (5) with $m = 2$. We tried to fit (5) to the low temperature GaAs data using both $m = 2$ and $m = 2.5$. While trying to fit the data with (5) a linear term was added to take care of the background. We find that in spite of the sample being doped, the data is better fit by $m = 2$ (shown as dashed lines in the figure) than by $m = 2.5$ (not shown) indicating an excitonic transition. The best fit values of $E_0$ and $\Gamma$ were found to be 1.516 ± 0.004 eV and 9.7 meV, respectively, at 12 K.

V. Conclusion

In conclusion, we have found a new solution to overcome the problem of photoluminescence in PR measurements at low temperatures. At room temperature or for samples which show negligible PL, the conventional PR technique is certainly superior to the DACPR technique. But at low temperatures the DACPR technique can extract signals where conventional PR fails to give any. We have also shown that the DACPR data can be fitted very well with established theoretical lineshape functions for various physical processes. The present drawback regarding noise can be can be improved upon by using better electronics.

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References

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