Time Resolved Pump-Probe Reflectivity in GaAs and GaN

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ABSTRACT
We have theoretically studied the time evolution of the Pump-Probe Reflectivity (PPR) signal. Two different types of carrier distribution function scenarios are studied, first a thermalized hot carrier distribution and second, a non-thermalized carrier distribution and both carrier distributions cool via several carrier-scattering mechanisms. We also study the effect of the carrier trapping or recombination and cooling on the PPR signal time evolution. The calculation also includes the effect of Sommerfeld factor and pump induced modulation of the probe reflectivity due to band filling (BF), band gap renormalization (BGR) and free carrier absorption (FCA). We find that in the case of thermalized carrier distribution, with comparable carrier cooling and decay rate, the PPR signal weakly depends on the FCA and strongly depends on the BGR and BF. The calculated absorption curves show that absorption can become negative at high enough carrier densities. We further find that, the temporal rise of the PPR signal is related to the cooling of the carriers while the decay of the signal is related to the carrier decay in a complex way. The interplay between BGR and BF effects decide the final shape (temporal evolution) of the PPR signal. The two cases studied here are relevant to the scenarios in GaAs (thermalized) and GaN (non-thermalized) and may be used to distinguish between the role played by the carrier-LO phonon scattering in the two materials.

Keywords: time resolved, reflectivity, carrier trapping, bandgap renormalization, band filling, carrier cooling.

1. INTRODUCTION
In the last few decades the study of the non-equilibrium carrier dynamics in semiconductors has been a very rich and an active subject. Several techniques have been used to study the carrier dynamics temporally as well as spectrally. The time resolved Pump-Probe Reflectivity (PPR) technique has proved to be one of the fast, simple yet powerful means to study the carrier cooling and carrier trapping dynamics. There have been numerous PPR measurements reported to study carrier lifetime and carrier capture in the defect engineered materials, like Low Temperature Molecular Beam Epitaxy (MBE) grown GaAs (LT-GaAs), ion implanted GaAs, and other relatively new materials, like GaN. The PPR experimental results in the literature are often found to display quite different behavior under otherwise similar conditions and sample preparation and measurements and the interpretation of the experimental results is sometimes not very straightforward. An extensive detailed theoretical study of the PPR signal under various conditions of carrier relaxation was made recently.

In this paper we have tried to calculate the effect of the carriers generated by the pump pulse on the reflectivity of the probe pulse, delayed in time with respect to the pump pulse. In the calculation we have included the effect of Sommerfeld factor and pump induced modulation of the probe reflectivity due to band filling (BF), band gap renormalization (BGR) and free carrier absorption (FCA). The material systems of relevance here are GaAs and GaN and pertaining to these two materials we consider two conditions of carrier distribution. In first one, the carriers are assumed to form a hot, thermalized energy distribution during excitation pump pulse duration itself and the distribution then cools via phonon emission. This scenario is expected in the case of high density excitation in GaAs. In the second case, the carriers are assumed to essentially remain in a non-equilibrium, non-thermal state while they relax. This scenario is possible when the carrier-Longitudinal Optical (LO) phonon interaction is stronger than carrier-carrier scattering, as is likely in GaN even at moderately high densities. The LO-phonon energy in GaN is 2.5 times higher (~92meV) than the LO-phonon energy in GaAs (~36meV) and the Fröhlich coupling of electrons with LO phonons is stronger by a factor of ~14 in GaN compared to GaAs. We thus expect that the PPR response may be used to distinguish the two modes of carrier relaxation. In the case considered here, we assume that the carrier decay time is larger than the carrier relaxation due to LO-phonon scattering mechanism. The extraction of the carrier decay time from the PPR curve is very complex. The notion often expressed in the literature that the decaying part of the PPR signal with double exponential structure essentially represents the cooling and decay of the carriers is found to be not correct in
general. Additionally, our study shows that the effect of BGR on the time evolution of the signal can be quite strong and the BGR influences the overall shape of the delay dependence of the PPR signal. The BGR and BF effects compete with each other and determine the magnitude and signature of the PPR signal. We have made a comparative study of the effect of BGR and BF on the sign of the PPR signal here. In section 2 we briefly describe the model used and the relevant effects included in the calculation of the PPR signal. In section 3 we present the results of the calculations for GaAs.

2. MODEL

The PPR signal is obtained by modulating a relatively high intensity femtosecond pulsed pump beam incident on the sample surface and detecting in the lockin type of detection setup, an unmodulated weaker intensity reflected beam (probe beam) appropriately delayed with respect to the pump beam, from the same spot on the sample. The interband transitions in the semiconductor due to the incident pump beam create charge carriers which are mainly responsible for the PPR signal thus generated. The response of the semiconductor to the incident light pulses can be modeled by studying the changes in the complex dielectric constant of that semiconductor. These changes are governed by various carrier scattering and decay mechanisms. This change in the dielectric constant is related to the refractive index of the semiconductor and can be modeled by considering the absorption change due to band filling (BF), the band gap shift due to the presence of carriers (band-gap renormalization or BGR), Plasma screening effect, enhancement of absorption due to the Coulomb Enhancement factor (CEF) and intraband absorption of photons by the free carriers. The relevant model has already been described by us. The essential theoretical formalism to calculate PPR was available in the literature in terms of BF, BGR and FCA effects, but was not extensively applied for the study of the delay dependence of the PPR. We now describe briefly the essential idea of the model used by us to calculate the effect of each of the above mechanisms on the PPR signal. The typical semiconductor considered here is GaAs, with its two valence bands.

The change in the reflectivity is due to the change in the complex refractive index \(\eta = \eta_r + i\eta_i\). In general, the major contributions to the change in \(\eta(\nabla \omega)\) can be written as,

\[\Delta \eta(\nabla \omega) = \Delta \eta_{\text{BF+BGR}} + \Delta \eta_{\text{FCA}}\]

(1)

where, \(\nabla\) is the Planck’s constant divided by \(2\pi\) and \(\omega\) is the photon frequency (energy) of the reflected probe beam.

The changes in the real part of the refractive index are related to the imaginary part of the refractive index by the well known Kramers-Kronig (KK) relations. The imaginary part of the refractive index is directly related to the absorption in the material. The detailed formulae for the calculation of the absorption changes are described elsewhere. The modified absorption due to the carrier induced band filling (BF) and the BGR effect is given by,

\[\alpha_{\text{BF+BGR}}(\nabla \omega, E_g) = \sum_{i=l,h} \frac{\alpha(\nabla \omega, E_g') (1 - f_s(E_{i\text{hole}})) - f_c(E_{i\text{electron}}))}{f_c(E_{i\text{electron}})}\]

(2)

where, \(f_c\) and \(f_s\) are the electron and hole distribution functions respectively and \(E_g = E_g' + \Delta E_g\), \(E_g'\) is the bandgap without renormalization \((E_g' = 1.423\ \text{eV at 300 K})\) and \(\Delta E_g\) is the band gap shift due to BGR. For a thermalized distribution, \(f_c\) and \(f_s\) are the Fermi-Dirac distributions of the respective carriers with appropriate quasi-Fermi energies in the case of thermalized carrier distribution. The change in refractive index is given by,

![Figure 1. Room temperature absorption curves for GaAs at carrier density 10^{18} \text{cm}^{-3}. The curves \(\alpha(\nabla \omega, E_g)\), \(\alpha(\nabla \omega, E_g')\) and \(\Delta \alpha\) are shown as continuous, dash and dotted curves respectively. (see the text for details).](image-url)
In Fig.1 and Fig.2, we show the calculated curves of $\alpha(V_0E_g')$ (pump beam OFF), $\alpha(V_0E_g)$ (pump beam ON) and also change in $\alpha(\Delta\alpha)$ at two different excitation densities at zero delay. These are shown as continuous, dashed and dotted curves respectively. The curves are shown for two densities $\sim 10^{18}$ cm$^{-3}$ and $\sim 10^{19}$ cm$^{-3}$. The carrier decay time and carrier cooling time are assumed to be 1ps each. There are several interesting features which can be seen from these curves. The curves in the ON and OFF condition of the pump beam show lot of difference owing to Eq.2 above. Due to band gap renormalization (BGR) the curves in the ON condition are shifted in energy to the lower side. The renormalized bandgap ($E'_g$) and bandgap at no excitation ($E_g$) are also shown in the figure. The change in absorption ($\Delta\alpha$) is positive near the bandedge for lower densities ($10^{18}$ cm$^{-3}$) at initial times. However, at higher densities the entire absorption curve shifts to the negative side near the bandedge. To explain this in more detail, let’s consider the condition soon after the carriers are excited. At early times, if the carrier temperature is large enough, $\Delta\alpha$ at larger energies ($E>\omega$) will contribute negatively to $\Delta\eta$ and subsequently the reflectivity will be negative. The electron and hole occupancy factors are smaller than unity for all energies at early times which

$$\Delta\eta_{BF+BGR}(\omega) = \left(\frac{\partial\alpha_{BF+BGR}(\omega)}{2\pi}\right) \int_{E_g}^{\infty} \frac{\alpha_{BF+BGR}(\omega,E_g')}{(E^2 - (\Delta\omega)^2)} dE - \int_{0}^{E_g} \frac{\alpha_{BF+BGR}(\omega,E_g)}{(E^2 - (\Delta\omega)^2)} dE$$

(3)

Where $P$ is the Cauchy principal value of the integrals and $c$ is the velocity of light.

The carrier distribution functions are used to calculate the absorption changes taking into account the various other effects, such as BGR, BF, CEF and FCA. The changes in the reflectivity are calculated using the reflectivity in the pump beam ON and OFF case.

### 3. RESULTS

The effect of BF and BGR on the temporal and spectral evolution of the PPR signal is quite complex. The PPR signal shows a negative signal at very early times at high enough excitation densities, becomes positive at intermediate times and at longer times it again becomes negative with a decaying signal. Although the signal decay is purely related to the carrier decay, the direct extraction of the decay time from this PPR curve is not possible. In Eq.2 above, we have the absorption coefficient calculated using the modified bandgap due to the presence of the carriers. This shifts the bandgap to the lower energy value. Thus the absorption coefficient in general different. We have calculated the absorption

![Figure 2](image2.png)

Figure 2. Room temperature absorption curves for GaAs at carrier density $10^{19}$ cm$^{-3}$. The curves $\alpha(V_0E_g')$, $\alpha(V_0E_g)$ and $\Delta\alpha$ are shown as continuous, dash and dotted curves respectively. (see the text for details).

![Figure 3](image3.png)

Figure 3. Room temperature absorption curves for GaAs at carrier density $10^{18}$ cm$^{-3}$. The curves $\alpha(V_0E_g')$, $\alpha(V_0E_g)$ and $\Delta\alpha$ are shown as continuous, dash and dotted curves respectively at 5ps delay. (see the text for details).
makes $\alpha (\nabla \omega, E_g')$ become negative. It is clear from Fig.1 and Fig.2 that the $\Delta \alpha$ is positive at smaller densities while it is negative at higher densities. Therefore, this will lead to a negative contribution to $\Delta \eta$ and reflectivity. Thus $\Delta \eta$ is negative for very short delays and high carrier temperatures. As the carriers cool, they begin to fill the states situated close to the band edge. This makes the electron-hole occupancy factors approach nearly unity if the carrier density is large enough. This is the Band Filling (BF) effect. At this point now, the integral will contribute positively to $\Delta \eta$ and reflectivity. At higher energies $\Delta \alpha$ is positive. This is seen in Fig.1, Fig.2 and Fig.3. At longer time delays the BGR effect vanishes and $\Delta \alpha$ remains negative near the band edge and positive at higher energies. This is shown in Fig.3. Finally, as the carriers decay, the overall magnitude of $\Delta \eta$ decreases and the reflectivity signal vanishes eventually.

A detailed study of the effects of various scattering mechanisms has already been reported. We now consider here the time evolution of the non-thermal carrier distribution. This case is more relevant for the GaN. However, as an illustrative example we have considered here GaAs case. The time evolution of the thermalized distribution and the nonthermalized distribution is clearly different. The difference mainly comes from the way the scattering mechanisms play role at various times. In Fig.4 we have plotted the time evolution of the nonthermalized distribution of carriers in GaAs at 300K with excitation and detection energy at 1.55eV (~18meV pulse-width) and with a carrier decay time of 1ps. The distribution clearly shows a peak at the zero delay, which is due to the excitation pulse, and the carriers cool by mainly with the emission of several LO-phonons. The subsequent peaks of the LO-phonons are visible at later times shown at 50fs, 500fs, 1ps and 3.5ps by dashed, dotted, dash-dotted and dash-double-dotted curves respectively. Due to the non-thermalized distribution of carriers at various energies from the bandedge, it is clear that the PPR signal in the case of nonthermal carrier evolution will look spectrally very different from the one obtained from thermalized carrier distribution. The LO-phonon emission time in GaAs is about 200fs for electrons while for in GaN it is about 20fs. Thus there is an order of magnitude difference in the two materials. If the other scattering mechanisms are not as fast the LO-phonon scattering time, then the carrier distribution is expected to be non-thermal for a long time during the cooling process. In fact the carriers will cool mainly via LO-phonon emission mode only. Such a scenario is possible in GaN. Thus for a non-degenerate measurement of PPR signal it should be possible to distinguish between the two cases of carrier relaxation discussed here.

4. CONCLUSIONS

We have studied the time evolution of the pump-probe reflectivity in GaAs and GaN. Two scenarios of carrier relaxation are considered here. In the case of thermalized carrier relaxation, it was found that the signal evolution is quite complex and it is not possible to extract the decay time from the measured signal in a straight forward way. The effects of Band Filling (BF) and Band-Gap Renormalization (BGR) play a complex role in determining the shape and sign of the signal. The absorption curves are used to explain the observed behavior. The non-thermalized distribution shows several peaks in the energy at various times, due to LO-phonon replicas. The effect of the shape of the carrier distribution function on the spectral shape of the reflectivity curve can be used to distinguish between the GaAs and GaN cases, where in the latter case the LO-phonon mechanism is very dominant and the carrier relaxation is expected to be predominantly non-thermal.
REFERENCES


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