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Photomodulation of the coupled plasmon–LO phonon of GaAs surfaces

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Abstract

Photomodulation of the coupled plasmon–LO phonon modes has been employed to determine the change in both the surface charge density and the depletion electric field as a function of photomodulation beam (PMB) intensity. The samples are two pieces of highly doped (001) n-type GaAs. The total surface charge density has been obtained as a function of the photomodulating intensity using the dependence of the unscreened LO phonon on the depletion width. We are able to separate the impact of the PMB on the surface electric field from the impact on the depletion width. This allows a separate determination of the change in depletion electric field, which reaches ∼73% of its original value at the highest intensity used for PMB.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Photomodulation (PM) provides a dynamic non-contact technique for varying the depletion electric field (E_s) and its related parameters within the surface depletion region of a doped semiconductor [1]. On the other hand, the forbidden LO phonon Raman scattering (RS), i.e. the electric field induced Raman scattering (EIRS) [2], is a very sensitive microscopic probe for detecting any perturbation of E_s at semiconductor surfaces. Photomodulation Raman spectroscopy (PM-RS) is a combination of these two techniques and provides a powerful spectroscopic technique which enables one to characterize the semiconductor surfaces and measure their electrical parameters that are important for semiconductor devices. The main idea of PM-RS is based on the optical perturbation of E_s by illuminating the sample with a second beam of light while RS is in progress. This perturbation should affect the EIRS intensity within the depletion region, which has width L.

The PM (with photon energy greater than the band gap of the material) creates electrons and holes within the entire penetration depth of the photomodulating beam (PMB), δ_{PMB}. The built-in surface field, which is a linearly decreasing function of the depth (z) from the surface, is given by $|E_s| = qNL\varepsilon^{-1}(1-z/L)$, where $q = |e|$ is the unit charge, N is the doping density, and ε is the dielectric constant. This field separates the PM-created carriers within the depletion region and those that travel to the surface and partly neutralize the surface charges. This, in turn, decreases the depletion electric field E_s and the band bending, leading to a decrease in the intensity of the forbidden LO phonon. Furthermore, the diffusion of photoexcited carriers from beyond the depletion region (i.e. $\delta_{PMB} > L$) also contributes to decreasing the field at the surface, depending on the range of their diffusion (length) during their lifetimes. Carriers that are generated at an even greater depth (far from the depletion region, $\delta_{PMB} \gg L$) mainly recombine before they can reach the surface and do not contribute to the Raman signal. On the other hand, the reduction in the surface charge density through PM also leads to a decrease in L. This effect plays an important role in affecting the scattering within the depletion region when L is smaller than the penetration depth of the incident Raman light (δ_R).

The measurement of interfacial electronic properties of semiconducting materials using PM-RS may be helpful in designing better electronic devices. Using this technique, Talaat *et al* [3] were first to use these dynamic measurements of RS intensity. By following the increase in the intensity of the forbidden LO after the PMB is turned off, they succeeded in calculating the surface state (trap) discharge time in n-type GaAs. In their work they also showed that the maximum response of the forbidden LO to the PM process is achieved when $\delta_{PMB} < L < \delta_R$. All of the photogenerated carriers in this case can contribute to neutralize the field at the surface and furthermore to decrease the scattering length, which is limited to L. From these results it was concluded that the PM process affects E_s and the scattering length when $L < \delta_R$, which is the case for highly doped samples. In this work the differentiation between these two effects will be investigated to allow a quantitative determination of the change in the value of E_s during the PM process.

It is known that, in highly doped polar crystals, the plasma oscillation and the bulk LO phonons couple via their macroscopic electric fields [4]. To observe these coupled modes, δ_R must be larger than L (which represents an important parameter for these measurements). The Raman signal in this case carries surface unscreened LO information (in the depletion region) and bulk information (the coupled LO-plasmon modes). The Raman signal in these measurements is less affected by the surface field, but it is sensitive to any change in the depletion width L, which controls the scattering length of the unscreened LO and the coupled modes. These measurements can allow the experimental separation of the impact of the surface electric fields from the effect of the depletion width on the Raman signals. Hence one can separate the effect of the PM process on E_s from that on the depletion width.

PM-RS has been applied to the LO–plasma coupling for the (001) surface of an n-type GaAs single crystal. In this case the allowed unscreened LO is observed in cross-polarization configuration and depends mostly on the depletion width L. On the other hand, the EIRS (forbidden LO) is observed in parallel-polarization configuration and depends on both the surface field E_s and the depletion width L (as a scattering length). By comparing the resulting PM-RS of the forbidden LO to the allowed LO, we are able to separate the effect of the PMB on the depletion width from its effect on the surface field.

2. Experimental details

The samples studied were single crystals of (001) n-type GaAs of two different electron concentrations ($N_e = 5.8 \times 10^{17}$ cm^{-3} with $L \approx 500$ Å and $N_e = 1.1 \times 10^{18}$ cm^{-3} with $L \approx 260$ Å). The surface quality is essential, since surface damage and contamination may cause a shift and broadening of the coupled mode [5]. For the two samples, the Raman penetration depth, δ_R, is larger than the depletion width for all wavelengths used, which allows observation of the coupled plasmon modes. To carry out the PM-RS experiment, the PMB was incident on the samples at the same time that the Raman measurements were in progress.

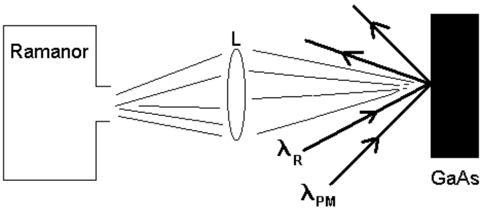

Figure 1. A schematic of the incident PMB and the RS beam.

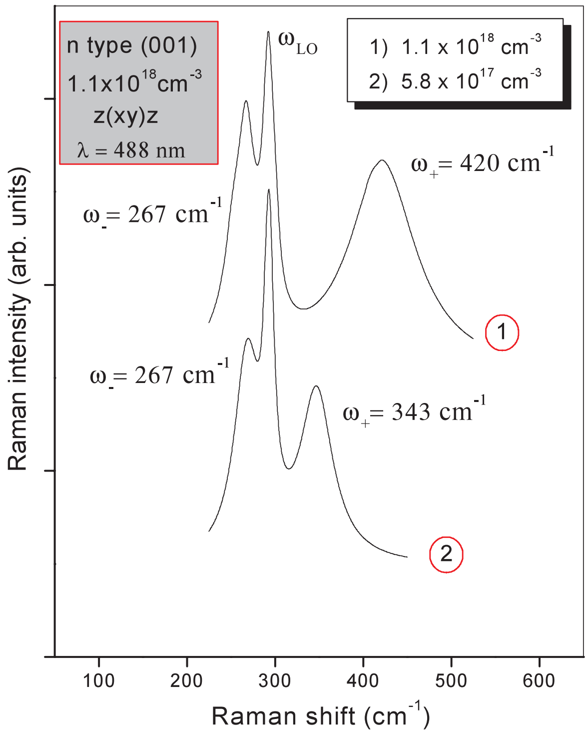

Figure 2. Raman spectra of n-GaAs in back-scattering geometry from the (001) surface at two different carrier concentrations. For sample 1, $\omega_- = 267$ cm^{-1} and $\omega_+ = 420$ cm^{-1}; for sample 2, $\omega_- = 267$ cm^{-1} and $\omega_+ = 343$ cm^{-1}.

The PMB was carefully aligned on the same scattering spot in all the experiments. A schematic diagram of the incident PMB and the RS beam is shown in figure 1. This arrangement gave us the best experimental conditions for observing the effect of modulation of the band bending. In the case of non-coincidence of the PMB with the Raman beam (spatially separated), the process may become diffusion dependent. In all the experiments, each recorded spectrum is an average of at least five time measurements.

The values of the depletion widths quoted above were taken from [6]. The penetration depths for all wavelengths used in this work were calculated from $\delta = l/4\pi\kappa$, using the available data for the extinction coefficient κ of GaAs [7].

3. Coupled plasmon–LO phonon modes

The RS spectra for the two samples, using $\lambda = 4880$ Å with intensity 15 W cm^{-2} at the sample surface, are shown in figure 2. In addition to the two coupled plasmon–LO phonon modes ω_-

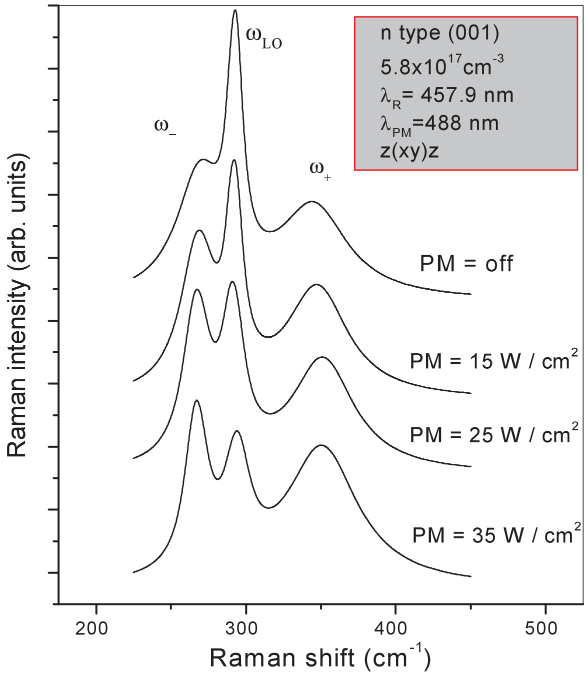

Figure 3. PM-RS spectra of n-GaAs (5.8×10^{17} cm^{-3}) in the cross-polarization configuration from the (001) surface at three different values of I_{PM} (as indicated) and with no PM.

and ω_+, one also observes the unscreened LO phonon mode at 292 cm^{-1}, which originates from the surface depletion region. The peak for the lower coupled mode ω_- has nearly the same frequency for the two samples ($\approx$267 cm^{-1}), while that of the higher coupled mode ω_+ shifts from 343 cm^{-1} for the low-doped sample to 420 cm^{-1} for the high-doped case. The depletion width becomes smaller with increasing N_e—a fact that is reflected in the decreasing intensity of the unscreened LO phonon modes and the increasing intensity of the coupled modes with increasing doping.

4. PM of the coupled plasmon–LO phonon modes

The PM-RS results for the sample with $N_e = 5.8 \times 10^{17}$ cm^{-3}, using $\lambda_R = 4579$ Å and PMB of $\lambda_{PMB} = 4880$ Å at three different intensities I_{PM} (15, 25 and 35 W cm^{-2}), are shown in figure 3, together with the case without a PMB. The spectra were obtained at room temperature in the cross-polarization $z(xy)z$ configuration and the intensities of the PMB were measured at the surface of the sample. In these experiments, carriers were generated inside as well as outside the depletion region, L, for both samples, since $\delta_{PMB} > L$ in both cases. It is observed that, with increasing I_{PMB}, the scattering intensity of the coupled modes increases without a change in the frequency shift, while that of the unscreened LO phonon decreases. The same observations hold for the other sample with $N_e = 1.1 \times 10^{18}$ cm^{-3} under the same conditions.

These results could be explained according to the fact that the reduction in the depletion width is a direct result of the decrease in the surface charge density resulting from PM. This is reflected as a decrease in the scattering length for the unscreened LO mode and at the same time an increase in the scattering length for the coupled modes. The constancy of the frequency

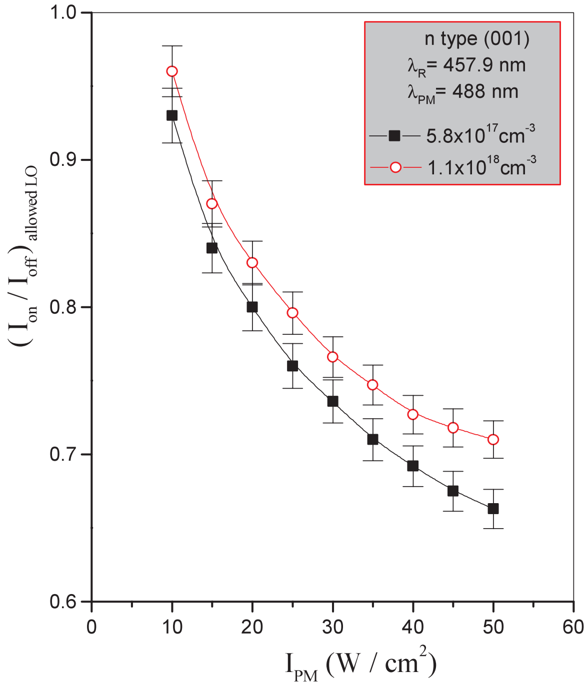

Figure 4. The normalized intensity I_{on}/I_{off} of the allowed LO phonon for two different carrier concentrations of n-GaAs as a function of I_{PM}.

of the coupled modes with respect to I_{PMB} indicates that there is no significant change in the carrier concentration within the bulk. This is consistent with the results obtained by Krost *et al* [8] for the photoexcited plasmon–LO phonon modes in a ZnSe/GaAs interface. They showed that there is no change in the coupled mode frequencies for GaAs for intensities less than 10^4 W cm^{-2}.

Figure 4 shows the variation in the normalized scattering intensity (I_{on}/I_{off}) of the allowed unscreened LO phonon versus the intensity I_{PMB} ($\lambda_{PMB} = 4880$ Å with $\delta_{PMB} \sim 900$ Å) for the two highly doped samples of (001) surface. It is observed that the variation in I_{PM} has a stronger effect on the sample with lower doping (larger L) than the sample with higher doping (smaller L), since the number of electron–hole pairs generated inside the depletion region is larger in the former case, leading to a stronger PM effect.

When the PM light illuminates the sample long enough for the carrier populations to reach steady state, the surface charge density σ_0 (majority carrier) is reduced by a factor of p_0, which is the surface minority carrier density generated by the PMB. Accordingly, the net surface charge density is given by $\sigma = \sigma_0 - p_0$. The intensity of the unscreened LO phonon, I_{LO}, within the depletion region is proportional (to a first-order approximation) to L/δ_R [9]. Consequently $(I_{on}/I_{off})_{LO} = L_{on}/L_{off}$ at constant δ_R and, since $\sigma = N_e L$, we may write

$$\sigma = \sigma_0(I_{on}/I_{off}).$$

This relation is used to determine the change in the surface carrier density as a function of the PMB intensity. Accordingly, the total surface charge density σ ($N_e = 5.8 \times 10^{17}$ cm^{-3}) changes from 2.9×10^{12} cm^{-2} (the value of σ_0) to $(1.92 \pm 0.038) \times 10^{12}$ cm^{-2}, while σ ($N_e = 1.1 \times 10^{18}$ cm^{-3}) changes from 3.9×10^{12} cm^{-2} to $(2.77 \pm 0.055) \times 10^{12}$ cm^{-2} on changing I_{PMB} from 0 to 50 W cm^{-2}.

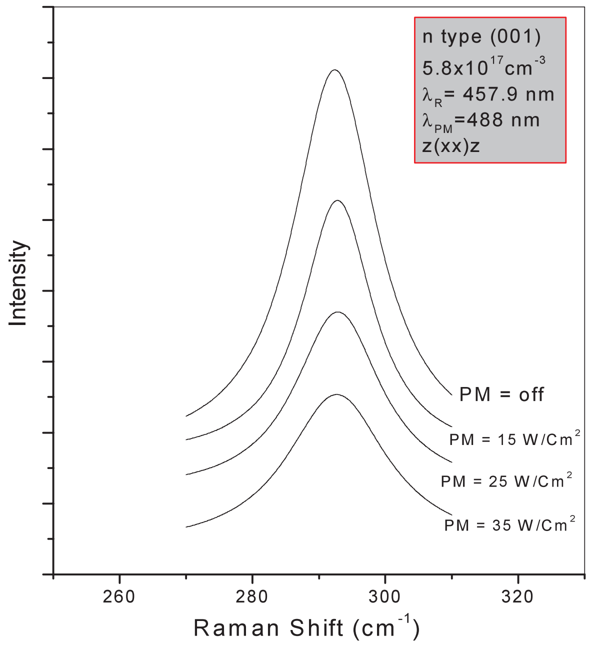

Figure 5. PM-RS spectra of the forbidden LO for n-GaAs (5.8×10^{17} cm^{-3}) in the parallel-polarization configuration from the (001) surface at three different values of I_{PM}.

To compare the above results with the effect of the PM on field-induced LO phonon scattering, the PM-RS spectra in the forbidden-polarization $z(xx)z$ configuration of the sample with $N_e = 5.8 \times 10^{17}$ cm^{-3} were measured, using the same λ_R (4579 Å) and λ_{PMB} (4880 Å). Figure 5 shows some of these spectra at three different selected intensities, I_{PM} (15, 25 and 35 W cm^{-2}), together with the case without a PMB. These results reflect a strong effect of the PMB on the intensity of the forbidden LO that is proportional to the square of the surface field (E_s^2) and the depletion width L as a scattering length ($L < \delta_R$). For comparison, in figure 6 we show the normalized intensity of the allowed LO, I_A ($=L_{on}/L_{off}$), together with that of the forbidden LO, I_F ($=E_{on}^2 L_{on}/E_{off}^2 L_{off}$), as a function of I_{PM}, where E_{on} and E_{off} are the values of E_s with the PMB on and off, respectively. From the figure it is clear that the effect of the PM on the forbidden LO is much stronger than that on the allowed LO. This is obviously due to the strong dependence of the forbidden LO on the depletion field, in addition to its dependence on the depletion width, as was mentioned earlier.

The above comparison shows that the variations in the surface field as a function of I_{PMB} can be extracted (separated) and calculated simply using the following relation:

$$\frac{E_{on}}{E_{off}} = \left(\frac{I_F}{I_A}\right)^{1/2}$$

which is presented in the inset of figure 6. These results show the ability of the PM process to decrease the surface field to about 73% of its original value at the largest intensity used for PMB (50 W cm^{-2}), which illustrates the success of this process in affecting significantly the surface charges even for such highly doping samples.

In conclusion, we have shown that the photomodulating light has significantly modulated the coupled plasmon–LO phonon of GaAs. Using the dependence of the unscreened LO phonon on the modulated depletion width, we were able to calculate the net surface charge

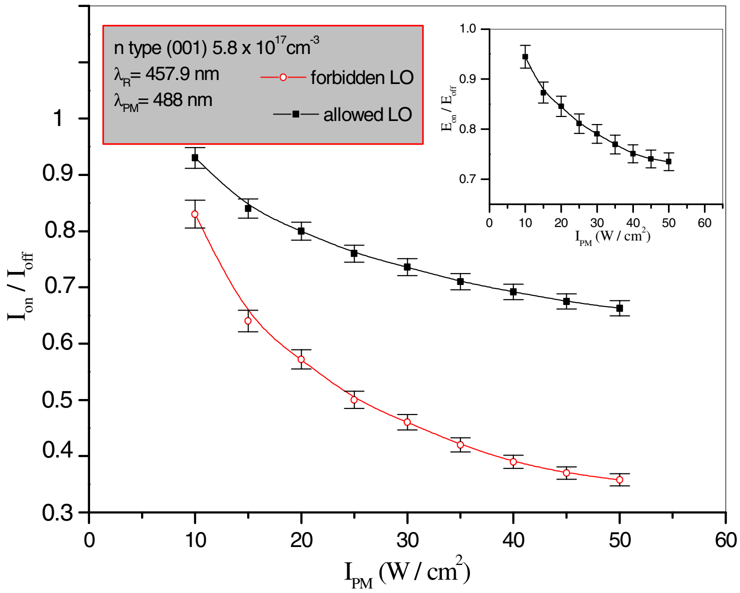

Figure 6. The normalized intensity I_{on}/I_{off} of the allowed and forbidden LO phonons for n-GaAs (5.8×10^{17} cm^{-3}) as a function of I_{PM}. The variation of E_{on}/E_{off} as a function of I_{PM} is shown in the inset.

density as a function of PMB intensity. Also, this PM helps us to differentiate the effect of PMB on the depletion field strength from the effect on the depletion width. As a result, we were able to obtain the relative change in the depletion field with the variation in PMB intensity.

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