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Photoreflectance modulation mechanisms in GaAs-$Al_xGa_{1-x}As$ multiple quantum wells

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Photoreflectance (PR) measurements performed in conjunction with photoluminescence excitation spectroscopy have allowed the modulation mechanisms that are responsible for the PR effect in GaAs-$Al_xGa_{1-x}As$ multiple quantum wells to be determined. This study indicates that the modulation of the interband excitonic transitions, rather than the band-to-band transitions, is observed in the PR spectrum. The spectral form of the PR line shape is discussed in terms of the optical modulation of the exciton energy gap, exciton lifetime, and integrated oscillator strength.

INTRODUCTION

Although room-temperature photoreflectance (PR) measurements have been shown to be valuable for the characterization of the alloy composition, quantum well width, and interfacial quality of GaAs-$Al_xGa_{1-x}As$ multiple quantum wells (MQW),[1-4] the nature of the modulation mechanisms that give rise to the PR effect remains poorly understood. For example, recent studies have shown that the line shapes of the PR spectra recorded at 77 K[2] and room temperature[2,3] are well described by the third derivative with respect to energy of the dielectric function of a two-dimensional critical point. It is well known that the third-derivative functional form (TDFF) is appropriate for the description of electroreflectance (ER) line shapes that arise from nonexcitonic interband transitions in bulk materials.[5] The conclusion that the PR spectrum of MQW is dominated by the modulation of nonexcitonic interband transitions is somewhat surprising in view of the wealth of experimental information that indicates that the room-temperature absorption spectra of quantum wells are dominated by excitonic transitions.[6-11] This apparent discrepancy, as well as other fundamental uncertainties regarding the origin of the PR effect in quantum wells, suggests that the following questions need to be answered: (a) Is the optical transition that is being modulated in the PR effect excitonic or band to band in nature? (b) How does the modulation of the dielectric function give rise to the experimentally observed line shapes? (c) What is the mechanism by which the dielectric function is modulated by the pump beam? In order to provide answers to these questions, we have simultaneously performed photoluminescence excitation (PLE) spectroscopy and PR measurements. The former spectroscopy measures the efficiency with which PL can be excited by radiation of different frequencies. In MQW, the PLE spectrum has been shown to be roughly proportional to the imaginary part of the dielectric function.[12,13] In contrast, in PR the modulation of the real and the imaginary parts of the dielectric function is measured via periodic changes in the reflectivity R that arise from a chopped optical pump beam.[1-4] Because of the intimate relationship between the real and the imaginary parts of the dielectric function, and because the PLE spectrum has been shown to exhibit sharp excitonic peaks,[6,9,12] a comparison of the results obtained from both of these spectroscopies places constraints on models that describe the PR effect, and has allowed the above-mentioned questions regarding the origin of the PR effect to be answered. In particular, by comparing peak positions between PR and PLE features, we can determine whether the observed PR features arise from excitonic transitions. This determination places constraints on the formalism which can be used to describe the observed data and sheds light on the nature of the modulation mechanisms giving rise to the PR signal. It also allows us to address the apparent discrepancy between recent studies that suggest that PR spectrum is dominated by nonexcitonic interband transitions[2,3] and the wealth of investigations that indicate that the interband optical spectrum of MQW is dominated by excitonic transitions.[6-13]

RESULTS AND DISCUSSION

A GaAs-$Al_{0.3}Ga_{0.7}As$ multiple-quantum-well sample with quantum well and barrier widths of 200 and 150 Å, respectively, has been studied by photoluminescence (PL), PLE, and PR. The thickness of the MQW was approximately 2 μm and the sample was capped with 3000 Å of $Al_xGa_{1-x}As$. The top 1000 Å of the cap layer was Si-doped to 2×10^{16} cm^{-3} to ensure that the MQW experienced a weak electric field from the surface space-charge layer. The PR and PLE measurements were performed simultaneously with a cw dye laser pumped by an Ar-ion laser and operated with Styryl 9 dye. A combination of 5145 and 4880 Å wavelengths from the argon-ion laser was employed as the modulation (or pump) source for the PR measurements. Simultaneous PR and PLE measurements were performed at temperatures between 6 and 250 K.

Shown in Fig. 1(a) is the PLE spectrum obtained at 6 K with the indicated PL detection window. The photoluminescence spectrum (not shown) exhibited a narrow electron–heavy-hole exciton with a linewidth of less than 1 meV, and the PL detection window of the PLE was in the region of the spectrum where absorption due to donor–bound-exciton and valence-band–ionized-donor transitions occur.[14] The lowest energy peak in the PLE arises from a donor-related absorption process. The three sharp peaks labeled in the figure as 11H, 11L, and 12H arise from interband *excitonic* transitions.[12,13] The first number of these labels denotes the conduction subband in-

dex while the latter number and letter specify the valence subband index and whether it is of light (L) or heavy (H) hole character. In the discussion that follows, the energies of these sharp excitonic peaks will be referred to as the exciton energy gaps. The PR spectrum obtained from this sample is shown in Fig. 1(b). The energies of the critical points, denoted by the arrows in Fig. 1(b), were determined by the three-point fitting procedure developed by Aspnes, assuming an excitonic transition.[5] Because there are several overlapping PR lines and because of the uncertainty in the critical point type, this procedure is accurate to within 0.2 meV. A comparison of the spectra shown in Fig. 1(a) and Fig. 1(b) indicates that the energies of the critical points determined from the PR spectrum and excitonic peak positions of the PLE spectrum are nearly identical. We have also performed PR and PLE measurements at higher temperatures, up to 250 K, and observe in all cases a similar agreement (within 1 meV at 250 K) between the excitonic peak energies observed in the PLE spectrum and the critical point energies obtained from the PR spectrum. Because the difference in energy between the peak positions of the excitonic peaks observed in PLE and the PR features is much less than the binding energies of the $11H$, $11L$, and $12H$ excitons ($\sim$8 meV),[15] we conclude that the PR spectrum arises from an optical modulation of *excitonic* transitions rather than a modulation of band-to-band transitions. Therefore, we must try to understand the PR line shapes in terms of modulation of a dielectric function appropriate for excitons rather than one for nonexcitonic interband transitions.

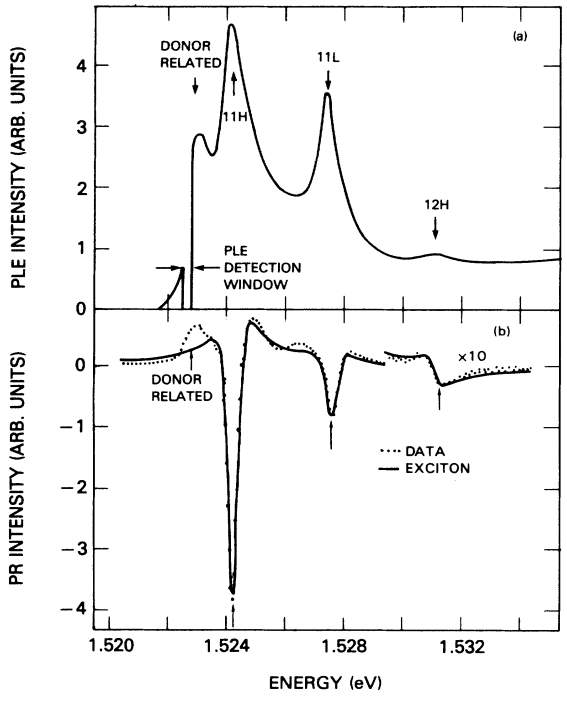

FIG. 1. (a) PLE spectrum obtained at 6 K from a MQW with well widths of 200 Å. The transitions are labeled according to the scheme discussed in the text. The energy of the PL detection window is indicated by the horizontal arrows. (b) The dotted curve displays the PR spectrum obtained simultaneously with the PLE data from (a). The arrows denote the positions of the energy gaps and the solid curve indicates the fit to the PR spectrum when Eqs. (1)–(3) are employed.

Previous studies in bulk material have suggested that the PR effect can arise from an optical modulation of the dielectric function by a variety of phenomena, including heating, free-carrier screening of excitons, and changes in the magnitude of the surface electric field.[16] These mechanisms lead to a modulation of the dielectric function through changes in the exciton energy gap, exciton lifetime (or spectral linewidth), and the integrated absorption strength of the exciton.

In general, the modulation of the dielectric function is related to the normalized change in the reflectivity measured in the PR experiment and is given by[17]

$$\Delta R/R = \alpha \Delta\varepsilon_1 + \beta \Delta\varepsilon_2 , \tag{1}$$

where α and β are the Seraphin coefficients, and $\Delta\varepsilon_1$ and $\Delta\varepsilon_2$ are the changes in the real and the imaginary parts of the dielectric function resulting from a change in the intensity of the pump beam P. Near the fundamental gap of the MQW, β is expected to be near zero, thus in this regime only $\Delta\varepsilon_1$ contributes to PR.[17] For excitonic transitions, $\Delta\varepsilon_1$ can be expressed as[5]

$$\Delta\varepsilon_1 = \frac{\partial\varepsilon_1}{\partial E_g}\left(\frac{\partial E_g}{\partial P}\right) + \frac{\partial\varepsilon_1}{\partial\Gamma}\left(\frac{\partial\Gamma}{\partial P}\right) + \frac{\partial\varepsilon_1}{\partial I}\left(\frac{\partial I}{\partial P}\right) , \tag{2}$$

where E_g is the exciton energy gap, Γ is a phenomenological broadening parameter, and I is the integrated intensity of the excitonic transition. If the bound excitonic transition exhibits a Lorentzian absorption profile, a form that is expected for quantum wells with little inhomogenous broadening due to well width variations and in cases of weak exciton-phonon coupling,[18,19] then the spectral form of the dielectric function is given by[20]

$$\varepsilon - 1 = \varepsilon_1 + i\varepsilon_2 \sim \frac{-I}{E - E_g + i\Gamma} . \tag{3}$$

This dielectric function leads to PR line shapes composed of a *sum* of the spectral forms shown in Figs. 2(b)–2(d). The relative contributions to the PR line shape arising from the linewidth [Fig. 2(b)], energy gap [Fig. 2(c)], and intensity [Fig. 2(d)] modulation mechanisms are determined by the magnitude of the bracketed coefficients in Eq. (2). These coefficients will be functions of the pump intensity and the subband indices of the excitons. However, the assignment of a physical significance to the relative magnitudes of the bracketed coefficients in Eq. (2) is only justified when the PR line shapes are not determined by a mixing of the real and imaginary parts of the dielectric function or by averaging effects that may arise as a consequence of a nonuniform perturbation.[21] Such effects are not expected to be important in determining the spectral forms of the PR lines reported in this paper because of the insensitivity of the PR spectra to changes in either the intensity of the pump or probe beams.

Shown in Fig. 1(b) is a fit of the PR spectrum with the model described by Eqs. (1)–(3). Except in the region of the spectrum where donor-related absorption processes

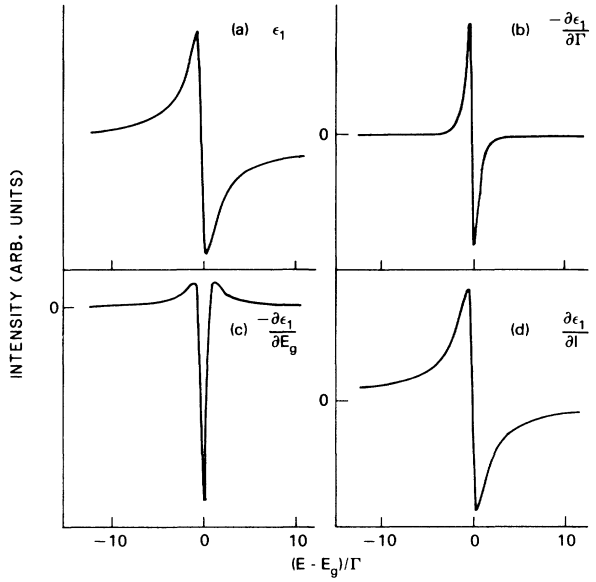

FIG. 2. (a) The real part of the dielectric function of a Lorentzian oscillator; (b)–(d) derivatives of the real part of the dielectric function shown in (a) with respect to the indicated variable. Some of the functions have been multiplied by -1 to allow an easy comparison of their spectral forms with that observed experimentally and shown in Fig. 1.

occur, this procedure yields excellent agreement with the observed line shape. The values of the exciton energy gaps and linewidths employed in the fit are in good agreement with those estimated from the PLE spectrum. We have also found that the fit employing the TDFF was poor. This occurrence is not surprising in view of the excitonic nature of the transitions observed in PR.

Having established the model of Eqs. (1)–(3), we can now address the question of the modulation mechanisms. A comparison between the relative magnitudes of the bracketed coefficients determined by the fit indicates that the spectral form of the PR lines from the $11H$ and $11L$ excitons of Fig. 1(b) is primarily determined by an energy-gap modulation. This conclusion is verified easily by comparing the spectral form of the PR spectrum from the $11H$ and $11L$ excitons with Fig. 2(c). A comparison of the line shapes shown in Fig. 2 with the PR spectrum from the $12H$ exciton indicates that both the integrated intensity [Fig. 2(d)] and lifetime [Fig. 2(b)] modulation mechanisms exhibit spectral forms that are in qualitative agreement with experiment. However, because only the intensity modulation mechanism yielded a good fit to the PR data when the value of Γ determined from the PLE spectrum was employed, we conclude that the contribution from the lifetime modulation mechanism to the PR line shape of the $12H$ exciton must be small.

The spectral form of the PR line shapes indicates that the relative magnitudes of the bracketed coefficients of Eq. (2) are different for the parity-allowed $11H$ and $11L$ transitions and the parity-forbidden $12H$ transitions. While this difference is not easily explained by an optically induced modulation of a screening or heating phenomenon, it is explained naturally by optically induced changes in the surface electric field. Specifically, electroabsorption measurements have indicated the sensitivity of the *exciton energy gap* of the $11H$ and $11L$ transitions to an applied electric field.[11] These experimental observations are in good agreement with effective mass calculations.[11,22] In addition, it has been shown experimentally that the *intensity* of the parity-forbidden excitonic absorptions is very sensitive to the magnitude of the electric field in the quantum well.[23] The intensity modulation arises because the electric field distorts the subband envelope wave functions and results in a violation of the interband excitonic selection rule.[22] Several other observations in our measurements support the idea that the PR experiment probes the optical modulation of the dielectric function that arises from changes in the surfaces electric field. First, we observe a saturation in the intensity of the 6-K PR signal for pump intensities greater than 0.04 W cm^{-2}. This behavior is difficult to explain by either the heating or screening modulation mechanisms which are expected to become larger under high optical pump excitations. Second, at our highest pump laser intensities of 0.4 W cm^{-2}, the modulation of the free-carrier concentration is less than 10^7 cm^{-2}, orders of magnitude smaller than that predicted to be necessary for a significant renormalization of the exciton energy gap by many-body effects.[24]

Although the formalism given by Eqs. (1)–(3) is sufficient for the description of the low-temperature PR spectra reported in this paper, we expect it to be inadequate under *any* situation that results in an excitonic absorption profile that is not well described by the dielectric function given in Eq. (3). Such situations are easily envisioned in MQW samples that exhibit large inhomogeneities in quantum well widths. Another example of the limited applicability of the dielectric function described by Eq. (3) occurs at high temperatures. Specifically, theoretical studies have suggested that the high-temperature limit of the excitonic absorption spectrum in a system with weak exciton-phonon coupling is described by a Gaussian absorption profile and not a Lorentzian as assumed in Eq. (3). In agreement with this suggestion, room-temperature absorption measurements of GaAs-$Al_xGa_{x-1}As$ multiple quantum wells have been shown to exhibit excitonic absorption spectra that are described by Gaussian line shapes.[10] We have explored this issue in relation to the PR data and find that the fit to the excitonic model of Eqs. (1)–(3) begins to fail at higher temperatures. An example of this is shown in Fig. 3 for PR data (dotted curve) taken at 150 K. The dashed line represents a fit of the data to Eqs. (1)–(3) and it is clear that an excitonic dielectric function with a Lorentzian absorption profile is inadequate at 150 K. However, we find that a dielectric function with a Gaussian absorption profile along with Eqs. (1) and (2) yields a good fit to the PR spectra obtained at temperatures above 150 K. This result is in agreement with previous work on the nature of excitonic lines at high temperatures.[10,19] A detailed discussion of the evolution of the dielectric function from one exhibiting a Lorentzian absorption profile to one characterized by a Gaussian profile, will be presented elsewhere.[25]

Also shown in Fig. 3 is the fit to the data employing the

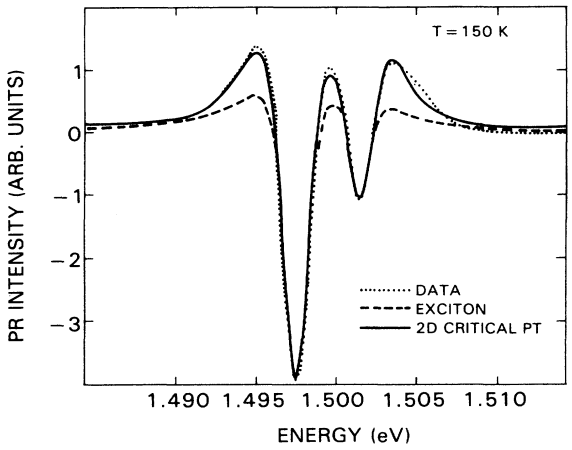

FIG. 3. A comparison between the PR spectrum observed at 150 K (dotted) and fits to the data when either Eqs. (1)–(3) (dashed) and the TDFF of a two-dimensional critical point (solid) are employed.

TDFF as applied to a two-dimensional critical point (solid curve). It is interesting to note, in agreement with the 77 and 300 K measurements of Ref. 2, that the TDFF provides a reasonable fit to the data at 150 K. This shows that although the TDFF is physically inappropriate to describe the PR spectrum of excitons in MQW, it does mimic Eqs. (1) and (2) with an excitonic dielectric function having a Gaussian absorption profile. However, it is important to remember that even at high temperatures, the PR spectrum still arises from an optical modulation of excitonic transitions and *not* from an optical modulation of interband transitions at a two-dimensional critical point. In this temperature range, the application of Eqs. (1) and (2) with an excitonic dielectric function with a Gaussian absorption profile is appropriate.

In conclusion, the optical transitions being probed by PR in MQW are excitonic in nature and the detailed form of the PR line shape allows the relative contributions that arise from the modulation of the exciton energy gap, exciton lifetime (spectral linewidth), and integrated oscillator strength to be estimated. This study has indicated that the low-temperature PR signal from the low-lying parity-allowed excitonic transitions arises primarily from a modulation of the exciton energy gap. In contrast, the integrated intensity modulation mechanism is shown to be dominant for the parity-forbidden transitions. These observations suggest that the optical pump beam causes a modulation of the dielectric function by changing the magnitude of the surface electric field experienced by the quantum well.

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