Exciton localization in $In_xGa_{1-x}As/GaAs$ quantum wells observed by temperature-modulated photoluminescence

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Temperature-modulated photoluminescence is used to study the recombination processes in $In_xGa_{1-x}As/GaAs$ single and multiple quantum wells between 8 and 70 K. The measured spectra correspond to the temperature derivative of the photoluminescence signal. We have compared the experimental data with calculations based on the temperature-dependent localized and delocalized exciton energy, density, and linewidth, and have found good agreement between theory and experiment. It is shown that temperature-modulated photoluminescence spectroscopy can be used to obtain the exciton binding and localization energies.

INTRODUCTION

Modulation spectroscopy is an often-used and effective technique to study semiconductors, bulk and layer structures alike.[1] The advantage of this type of optical spectroscopy is associated with the resulting sharp, derivativelike modulated signal which can be advantageously used to study critical points, band offsets, and other important semiconductor parameters. Modulation spectroscopy has been most often combined with reflection and absorption spectroscopies.[2] In this publication, we report results obtained using temperature-modulated *photoluminescence* (TMPL) spectroscopy. The basic idea of TMPL is similar to other modulation spectroscopies: while modulating the temperature of the sample, the photoluminescence (PL) spectrum and its temperature-modulated component are simultaneously measured.[3] This technique, in addition to producing sharp, derivativelike spectra, also illustrates a further virtue of modulation spectroscopy: it allows the separation and identification of compound luminescence processes, thereby providing information on binding energies, broadening mechanisms, and other *temperature-dependent* parameters.

In the past, TMPL has been used to study the nature of the recombination mechanism in GaP,[3] temperature-dependent tunneling of excitons,[4] and exciton binding energies in $In_xGa_{1-x}As/InP$ quantum wells.[5] Temperature modulation has also been observed, although unintentionally, in other photoluminescence experiments, such as optically detected cyclotron resonance[6] and optically detected magnetic resonance.[7]

In this paper, we will present experimental results and model calculations describing the temperature evolution of the TMPL spectra in $In_xGa_{1-x}As/GaAs$ single and multiple-quantum-well samples between 8 and 70 K. It will be shown that exciton localization plays a significant role in the temperature dependence of the photoluminescence process.

EXPERIMENTAL ARRANGEMENT

The general features of a TMPL experiment are similar to a conventional photoluminescence setup, except for the modulation of the sample temperature. A number of different techniques have been described in the literature to achieve temperature modulation.[1] Some of these include heating strips,[8] direct current heating,[9] and microwave heating.[10] While in previous TMPL measurements we have used a miniature heater to modulate the temperature of the sample,[3] here we report experimental results based on a more convenient and easily adaptable method of *laser-beam heating,* displayed in Fig. 1. The sample, mounted on a sapphire substrate, was attached to the cold finger of a variable temperature cryostat. The front surface of the sample was illuminated by a 0.5-mW HeNe laser and the resulting luminescence emission was focused on the entrance slit of a 0.75-m monochromator. The HeNe laser was *not* modulated but, instead, a second laser beam from an argon ion laser ($\lambda=514.5$ nm) was mechanically chopped at 9 Hz, and focused through the sapphire onto the back side of the sample. This modulated Ar laser beam provided the required temperature modulation of the sample. Using 40 mW of laser power, the amplitude of the temperature oscillations was estimated to be approximately 1 K by comparing these TMPL measurements to measurements done using a calibrated miniature heater.[5] The HeNe-laser-induced PL

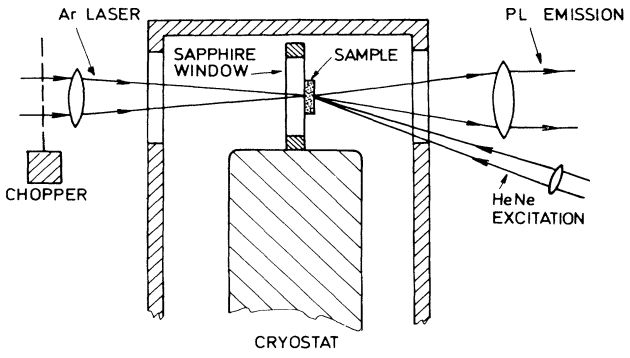

FIG. 1. Schematic diagram showing the (Ar^+) laser induced modulation of the sample temperature during the TMPL experiment.

and its temperature-modulated component, ΔI_{PL}, were detected by a Si photodiode. The modulated signal was amplified by a lock-in amplifier whose reference input was provided by the mechanical chopper. The magnitude of the relative signal, $\Delta I_{PL}/I_{PL}$, varied from sample to sample, but was in the order of 10^{-2}–10^{-3}. Both the PL and the TMPL signals were stored in a personal computer, which also controlled the wavelength drive of the monochromator.

SAMPLES

Undoped single and multiple quantum wells of $GaAs/In_xGa_{1-x}As/GaAs$ were grown by molecular-beam epitaxy on Cr-doped GaAs(100) substrates under As-rich conditions. A total of 15 samples were studied. Sample *A* discussed in the text refers to an $GaAs/In_xGa_{1-x}As/GaAs$ single quantum well, where $x=0.16$ and the quantum well width is 67 Å. Sample *B*

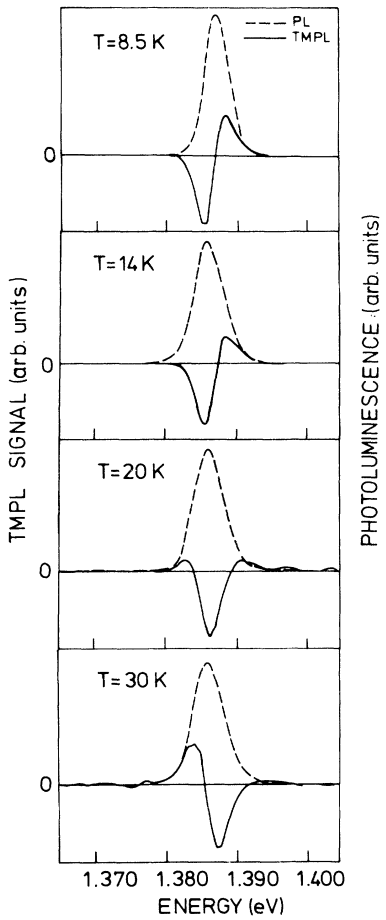

FIG. 2. Photoluminescence (dashed curve) and temperature-modulated photoluminescence (TMPL) spectra of an $In_xGa_{1-x}As/GaAs$ single quantum well at different temperatures.

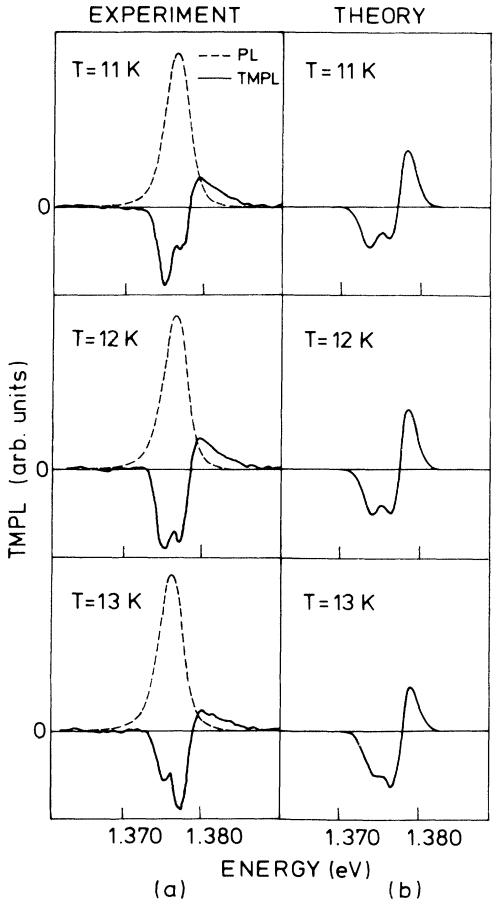

FIG. 3. (a) Measured PL and TMPL spectra of an $In_xGa_{1-x}As/GaAs$ MQW having three nominally identical wells ($L_z=100$ Å). (b) Calculated TMPL spectra for a *two-well* MQW, having their well widths differ by approximately one monolayer, 3 Å.

refers to a GaAs/$In_xGa_{1-x}As$/GaAs multiple quantum well, having a mole fraction of $x=0.15$ and three uncoupled wells, each with a well width of 100 Å and a barrier width of 320 Å.

RESULTS

(a) Single quantum wells. Typical TMPL spectra for an $In_xGa_{1-x}As$/GaAs single quantum well (sample *A*) at four different temperatures are shown in Fig. 2, together with their respective PL spectra. As can be seen in these figures, while there is no noticeable change in the PL spectra, the TMPL signals differ significantly at the various temperatures. The general features of the modulated spectra are as follows: For temperatures below approximately 20 K and above 30 K the TMPL spectra correspond to the *first derivative* of the PL emission. Between 20 and 30 K the signal changes phase from a positive high-energy lobe at low temperatures (<20 K), to a negative one at high temperatures (>30 K). The exact temperature where the phase change occurred varied from sample to sample, however for single quantum wells this phase change always occurred between 20 K and 30 K.

(b) Multiple quantum wells. The TMPL spectra of most, but not all, multiple-quantum-well samples display additional fine structure which show strong temperature dependence. An example of such spectra, for a sample containing three nominally identical wells (sample *B*), is shown in Fig. 3(a). While the PL spectra are indistinguishable at the different temperatures, the TMPL spectra show distinct fine structure. Below 10 K and above approximately 20 K the signal is first-derivative-like, and similar to single quantum wells. However, between 10 and 20 K, the TMPL signal exhibits additional structure, usually separated by a few meV's. The fine structure is related, as we shall discuss below, to the slight differences between the individual wells.

THEORY

(a) Single quantum wells. As we have discussed previously,[5] the TMPL signal is proportional to the *temperature derivative* of the PL emission. At low temperatures, the PL process is dominated by excitonic transitions and hence the TMPL signal is determined by the temperature dependence of the exciton energy $E_0(T)$, the broadening parameter $\Gamma(T)$, and the exciton density $N(T)$ according to the expression

$$\Delta I_{\rm PL}(T)=I_{\rm PL}(T+\Delta T)-I_{\rm PL}(T)\simeq\frac{dI_{\rm PL}(T)}{dT}\Delta T\simeq\left[\frac{\partial f(\Gamma,N,E_0)}{\partial\Gamma}\frac{\partial\Gamma}{\partial T}+\frac{\partial f(\Gamma,N,E_0)}{\partial N}\frac{\partial N}{\partial T}+\frac{\partial f(\Gamma,N,E_0)}{\partial E_0}\frac{\partial E_0}{\partial T}\right]\Delta T\ , \tag{1}$$

where $\Delta I_{\rm PL}$ is the measured temperature-modulated photoluminescence signal, $I_{\rm PL}(T)$ is the PL intensity at a temperature T, and $f(\Gamma,N,E_0)$ is the line-shape function of the PL emission which can be approximated either by a Lorentzian or a Gaussian profile.[11] In order to evaluate the relative importance of the three terms in Eq. (1) we need to know the temperature dependence of the exciton energy E_0, the exciton density N, and the broadening parameter Γ.

The term proportional to $\partial E_0/\partial T$ in Eq. (1) can be calculated using the Varshni[12] equation, which describes the temperature dependence of the exciton energy, and is given by

$$E_0(T)=E_G-\frac{\alpha T^2}{(\beta+T)}\ , \tag{2}$$

where E_G, α, and β are known constants.[13]

The temperature derivative of the exciton density $N(T)$ can be calculated by assuming thermal equilibrium between the excitons and the free carriers:

$$N=n_0e^{-E_B/kT}\ , \tag{3}$$

where E_B is the (well-width-dependent) exciton binding energy.

The luminescence linewidth $\Gamma(T)$ was recently measured by Lee, Koteles, and Vassell for excitons in GaAs quantum wells.[14] They showed that at temperatures below 150 K, the exciton-acoustic phonon, and the exciton-ionized impurity interactions determine the exciton linewidth, which is given (in meV) by

$$\Gamma(T)=\Gamma_0+(1.47\times10^{-3})T+\Gamma^+e^{-\varepsilon/kT}\ , \tag{4}$$

where Γ_0 is the temperature-independent linewidth due to inhomogeneous fluctuations, Γ^+ is the linewidth due to the fully ionized impurity scattering, and ε is the impurity binding energy averaged over all possible locations of the impurities. In GaAs, Γ^+ is estimated to be 0.75 meV, and ε to be 10 meV.[14]

We are now in a position to compare the predicted line shapes with the experimental spectra. Above 30 K, the term proportional to the temperature derivative of the exciton energy,

$$\frac{\partial f}{\partial E}\frac{\partial E}{\partial T}=\frac{\partial f}{\partial E}\left[\frac{\alpha T^2}{(\beta+T)^2}-\frac{2\alpha T}{\beta+T}\right]\ ,$$

dominates the line shape. This yields a first-derivative-like line shape, in good agreement with the experimentally observed spectra, as discussed below. At temperatures below 15 K on the other hand, the temperature derivative of the exciton *density*, which increases as $1/T^2$ with decreasing temperature, becomes the dominating factor in Eq. (1). This term generates a TMPL line shape which is a (wavelength-independent) constant times the PL emission and quite unlike the observed (inverted) first-derivative-like spectra. The observed first derivative spectrum is an indication of a *blue* shift of the PL emis-

sion with increasing temperature. Therefore, in order to correctly describe the TMPL process, additional contributions must be included in the model. We proceed by considering the relevance of exciton localization on the TMPL spectra.

It has been suggested by a number of authors that at low temperatures, the PL spectrum is a combination of localized (or bound) and delocalized (free) excitonic emissions.[15–17] Assuming that the PL spectrum is the superposition of localized and delocalized excitonic emissions, Eq. (1) has to be recalculated using

$$I_{\mathrm{PL}}=N_1 f(\Gamma,E_1)+N_2 f(\Gamma,E_2)\ , \tag{5}$$

where N_1 (E_1) represents the density (energy) of the bound excitons, while N_2 (E_2) denotes the free excitons. Using this model, the temperature derivative of the density can be written in the following form:

$$f_1\frac{dN_1}{dT}+f_2\frac{dN_2}{dT}=N_1\frac{E_1}{kT^2}[f(\Gamma,E_1)-f(\Gamma,E_2)]\ . \tag{6}$$

In deriving Eq. (6), we have assumed $dN_1=-dN_2$, that is the ΔT induced decrease in the bound exciton density (dN_1) is equal to the increase in the free exciton density (dN_2). This is valid when the exciton *localization* energy $\Delta E=E_1-E_2$ is much less than the exciton binding energy. Equation (6) can be further approximated if we expand (f_1-f_2):

$$N_1\frac{E_1}{kT^2}(f_1-f_2)\approx N_1\frac{E_1}{kT^2}\left[\frac{df}{dE}\right]\Delta E\ . \tag{7}$$

Using Eq. (7), the calculated TMPL line shape is in excellent agreement with the experimental results (see below). We conclude, therefore, that the first-derivative-like low-temperature TMPL signal is an indication of the existence of localized and delocalized excitons in the recombination process.

Figure 4 displays the calculated TMPL line shapes at various temperatures, together with the respective experimental results. Gaussian line shapes were used and the exciton localization energy ΔE was the only fitting parameter. The values of the other parameters are taken from the literature and are given in Table I. The agreement between theory and experiment is surprisingly good, considering the approximations used in deriving the model. Although the value of the fitting parameter ($\Delta E=2$ meV) compares well with that measured by Delalande, Meynadier, and Voos[17] in $GaAs/Al_xGa_{1-x}As$, the purpose of the fitting procedure was to clarify the nature of the recombination process, rather than to estimate the fitting parameter *per se*.

(b) Multiple quantum wells. In multiple quantum wells the PL spectrum is a superposition of the spectra originating from the individual wells. If the wells are not identical (for example, if there are slight variations in the well widths) the PL spectrum can broaden and information about the individual wells will be lost. Temperature-modulated PL, on the other hand, reveals such fluctuations through the fine structure appearing in the TMPL spectra.

TABLE I. Values of parameters used in calculating theoretical TMPL line shape for single quantum wells.

Parameter	Value
ΔE	2 meV (fitting parameter)
α	5.4×10^{-4} eV/K (Ref. 19)
β	204 K (Ref. 19)
E_B	8 meV (Ref. 20)
ε	10 meV (Ref. 14)
Γ^+	0.75 meV (Ref. 14)

In order to illustrate this point let us consider two wells of slightly different widths: $L_1=100$ Å and $L_2=103$ Å. The PL emission from this multiple-quantum-well (MQW) structure will be the superposition of two emissions separated by approximately 2 meV. If

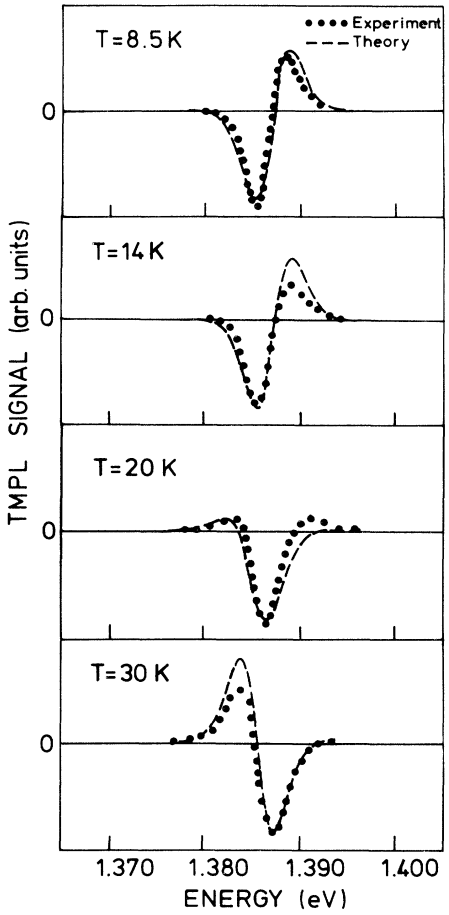

FIG. 4. Comparison of the experimental (dotted curve) and calculated TMPL spectra of the $In_xGa_{1-x}As/GaAs$ single quantum well. Calculated line shape was based on a single fitting parameter, the exciton localization energy. Other parameters are given in Table I.

the linewidth is 3–4 meV or larger (which is common in the $In_xGa_{1-x}As/GaAs$ system), the PL spectrum will consist of a *single* broadband. The TMPL spectroscopy, on the other hand, can resolve the individual emissions. In Fig. 3(b) we display the calculated TMPL spectra for the above discussed example. For this calculation the binding energies and the broadening parameters were chosen to be identical for both wells ($E_1=E_2=7$ meV, $\Gamma_1=\Gamma_2=3$ meV) and the PL emission peaks were shifted by 2 meV (this corresponds to a monolayer fluctuation in the well width). The calculated TMPL spectra, which are the sum of the two modulated line shapes, clearly highlight the individual emissions, in qualitative agreement with the measured TMPL spectra of Fig. 3(a). Temperature-modulated photoluminescence, therefore, may be used to resolve compound PL emissions in semiconductor quantum wells. A quantitative analysis of the TMPL line shapes for MQW's will be discussed in a forthcoming publication.[18]

CONCLUSION

We have measured the temperature-modulated photoluminescence spectra of single- and multiple-quantum-well $In_xGa_{1-x}As/GaAs$ samples between 8 and 70 K. The measured spectra correspond to the temperature derivative of the photoluminescence signal. The calculated line shapes, based on the temperature dependence of the exciton energy, density, and linewidth, are in good agreement with the observed spectra. The fitting procedure can be used to obtain the exciton binding and localization energies. The TMPL spectra of some MQW samples, unlike the conventional PL spectra, exhibit additional fine structure which is shown to be due to fluctuations of the quantum well width.

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