Temperature dependence of exciton-capture at impurities in GaAs/Al_xGa_(1-x)As quantum wells

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Abstract: In this paper we present an investigation of the exciton capture process in GaAs/Al_{0.3}Ga_{0.7}As quantum wells using a picosecond time-resolved photoluminescence technique. We demonstrate that there are significant differences in the capture mechanism for narrow quantum wells in comparison to bulk material. In particular the initial capture efficiency is shown to increase with temperature. This behaviour is understood in terms of the role of localisation of the free exciton in the potentials caused by the interface roughness. Higher temperatures destroy this localisation process which otherwise limits the total capture rate for the exciton to the impurity. The effect of localisation on capture is also shown to be stronger for narrower wells. We conclude that the relative weakness of bound exciton recombination in the near bandgap luminescence of doped quantum wells can in part be understood by the reduction of capture efficiency due to localisation.

1. Introduction

The low temperature optical emission in bulk GaAs from moderately doped down to all but the highest purity samples is dominated by recombination from impurity bound excitons [1,2]. The dominance of the bound emission over the free exciton recombination is simply understood in terms of the large capture cross section for the free exciton at an impurity. If however we now compare the situation in confined GaAs/AlGaAs systems the recombination is dramatically different. In particular, for narrow quantum wells the free exciton emission remains the dominant component up to impurity concentrations as high as 10^{17} cm^{-3} [3]. Clearly there is a fundamental difference in the capture mechanism in a confined system as compared to bulk material. Despite this dramatic difference there has been little or no work on capture of excitons at impurities either in low dimensional systems or even in bulk GaAs. Work on bulk material has been carried out to a limited extent for Si and Ge [4,5], for the purpose of this report we shall make use of the results available in the literature for these materials to make a comparison with our data for the confined system.

The process of capture of a free exciton (FE) by a neutral impurity (we shall not discuss ionised impurity capture here) to form a bound exciton (BE) can be characterised by a capture cross section σ . This term is a measure of the probability of an exciton to be captured by an impurity and relax to the ground state of the bound exciton, as such it is independent of the capture mechanism assumed and describes a fundamental property of the impurity in the material. For the purpose of discussion we assume the capture process to be a cascade relaxation process, whereby the exciton is initially trapped in an excited state of the bound exciton and subsequently relaxes via phonon emission. This model is somewhat analogous to the model proposed by Lax for the capture of a free particle in a Coulombic potential [6]. Since however both the impurity and the exciton are neutral species and the attractive potential therefore not a simple Coulombic one, it is inappropriate to apply the details of the Lax model to the present case. The details of the attractive potential and the resulting pattern of bound states is in fact exactly what is physically described by the statistical idea of the capture cross section. From this argument one quickly sees that the capture cross section will be strongly dependent on impurity, a result clearly demonstrated by the work on bulk Si [4,5].

A consequence of the cascade model is that capture will in effect only occur to those bound states lower in energy than the thermal energy $k_B T$ from the free particle continuum. Excitons captured to states below this threshold rapidly relax to the ground states while those in higher lying states are thermally re-emitted. The simplest model takes no account of the relaxation rates from excited levels to the ground state, however, provided that this relaxation is rapid in comparison to the observed decay rates, the population will predominantly reside in the ground state [7]. This description allows us to make a simple approximation in modelling the capture mechanism by treating it as a two level system, the exciton is either free or bound and the capture describes the transition rate between these two levels. The occupancy of excited states is however important since this determines the thermal emission from bound to free states, which in turn represents the temperature dependence of the capture process. Again since the pattern of excited states and the rate of relaxation between them is unknown it is impossible to predict the exact temperature dependence expected for the capture mechanism. Clearly, with increasing temperature thermal emission will increase and the capture efficiency correspondingly decrease. The work by Hammond and Silver [4] and by Feenstra and McGill [5] on bulk Si has shown that the temperature dependence of the capture cross section can be fitted by a relation of the form $\sigma \propto 1/T^\chi$, where $\chi = 0.5 - 4$ depending on the impurity measured.

The reduced efficiency with which excitons are captured at impurities in the confined system has in general been associated with, (although not explicitly discussed), the restriction of the free exciton mobility to a two dimensional (2D) plane. By limiting the movement of the exciton in this way the probability that the exciton approaches an impurity is reduced in proportion to the loss in dimensionality ($\sigma(2D) \propto \sigma(3D)/R$, where R is the average separation between impurities). This result has even greater significance in 1D and 0D systems, where the probability for extrinsic recombination will as a result become almost negligible. In addition since the same argument can be applied to non-radiative centres this has led to the conclusion that low dimensional structures, in particular quantum wires should provide the basis for highly efficient semiconductor lasers. The ratio of free exciton to bound exciton luminescence observed in a time integrated spectra is a complex function of not only capture rate but also of recombination lifetime. The latter variable determining how long the capture process continues. The predicted lifetime for low dimensional excitons is significantly shorter than that for bulk GaAs [8,9]. This again implies a significant reduction in the total capture to the bound exciton and hence to the integrated spectral strength. In practise very short lifetimes have not been observed for samples grown using standard techniques [10,11], arguments for weaker bound exciton recombination based only on lifetime arguments are therefore inadequate.

In this paper we report an investigation of the temperature dependence of the capture rate for acceptor doped quantum wells of different well width. In apparent contradiction to the theoretical arguments presented above and to the experimental work on bulk Si, the initial capture rate is found to increase with temperature. Furthermore this phenomena is also enhanced for narrower well widths. We discuss how this result can be understood in terms of the localisation of the free exciton in narrow quantum wells due to interface roughness.

2. Experimental Method

The study described requires well-resolved free and bound exciton peaks to enable a clear analysis of the kinetics even at elevated temperatures. We therefore restrict the discussion in this paper to acceptor doped GaAs/Al_{0.3}Ga_{0.7}As quantum wells with well width of 100Å or 150Å. The samples have been prepared using molecular beam epitaxy (MBE) with non-interrupted growth. The Be dopant was introduced into the central 20Å of the well only at a concentration of $1 \times 10^{10} \text{ cm}^{-2}$. The low temperature photoluminescence experiment was carried out at 2K, time resolved measurements were made using a Hamamatsu synchroscan streak camera with a temporal resolution of about 20ps. Pulsed excitation was provided by a dye laser synchronously pumped using a mode-locked argon laser to give pulse lengths of around 5ps duration.

3. Results and Discussion

Fig. 1. illustrates the initial development in the PL signal for the BE emission of a 100Å QW at a series of different temperatures. The rate of onset of the emission corresponds to the rate of capture of resonantly excited free excitons at the neutral Be impurity, given by:

$$\frac{dN_{BE}}{dt} = C_{FE} \times N_{FE} \times (A_0 - N_{BE}) \quad (1)$$

where C_{FE} is the capture rate, A_0 the neutral Be acceptor concentration and N_{FE} , N_{BE} the concentrations of free and bound excitons respectively. The initial capture rate to the BE is clearly seen to increase with

temperature. The time taken to reach the maximum intensity of the BE emission decreases from 200ps at the lowest temperature (2K) to less than 50ps at 40K. The shape of the onset also alters with increasing temperature indicating that the capture mechanism is itself modified with increasing temperature. The analysis of these results to give a quantitative value for the capture rate is made difficult by this latter point. If the capture rate is fast in comparison to the decay rate one can expect an exponential onset for the emission, the time constant for which defines the capture rate. Given the small change in signal strength and the inconsistent deviation from exponential behaviour the direct analysis proves to be inaccurate and a rather poor measure of the behaviour. A more accurate analysis of the average capture rate can be made from the time integrated spectra. As discussed in the introduction the ratio of the BE to the FE reflects the capture strength of the exciton at the impurity. To assess the capture rate we must however take into account the FE lifetime during which the capture process is able to occur. We therefore make an estimate of the capture rate by taking the ratio of the BE to FE intensities and dividing by the observed free exciton decay time, this gives a probability per unit time of capture from the FE to the BE. This value can be compared to the usual definition of the capture rate by calculating the rate per impurity, giving the quantity we call C_Q :

$$C_Q = \frac{BE}{FE} \cdot \frac{1}{\tau_{FE} \cdot n_i} \quad (2)$$

Assuming a bulk density of 10^{17}cm^{-3} we calculate the capture rates shown in Fig. 2 for both a 100Å QW and also an 150Å well with comparable doping densities. The results are plotted on a log-log scale to illustrate the suggested power law dependence. For comparison the results for capture of excitons to an indium acceptor in silicon as taken from reference [5] are plotted. At higher temperatures the derived capture rate is in remarkably good agreement with the power law dependence found for bulk material, the exponent calculated assuming a least squares fit to the data within the linear region gives a value of $x=2.6$ for the 150Å well and $x=3.9$ for the 100Å well. As yet it is not understood why there should exist a stronger temperature dependence for capture in a 100Å QW as compared to the 150Å well. The low temperature behaviour, in contrast shows a strong deviation from the expected trend. The initial decrease in capture efficiency with temperature is relatively small, this anomalous behaviour continues up to a definite threshold above which the trend returns to the simple power law dependence. This threshold occurs between 15 and 20K for the 100Å well and somewhat lower, between 10 and 15K, for the 150Å well. The data presented in Fig. 1. do not clearly illustrate this threshold behaviour, instead the onset appears to continue to become faster even at temperatures above which the data of Fig. 2. indicate a significant reduction in average capture efficiency. The discrepancy between these sets of data is due to the fact that Fig. 1. illustrates only the initial capture behaviour. At these short times following the laser pulse, the environment in which capture takes place may be very different to later times. In particular since we resonantly excite free excitons they are essentially 'cold' i.e. they have no kinetic energy, capture to the bound state initially occurs before the excitons are fully thermalised to the lattice temperature, as a result the initial onset is less sensitive to temperature than capture at later times. We are left with the question as to what limits the capture rate at low temperatures? We suggest that both sets of data can be understood in terms of the role of localisation at interface roughness on the capture process.

The capture rate C_{FE} is related to the total capture cross section σ by the thermal velocity V_{th} such that:

$$C_{FE} = \sigma_{FE} \cdot V_{th} \quad (3)$$

An analysis of the capture process must therefore also take into account the temperature dependence of the thermal velocity :

Figure 1. Temperature dependence of the onset in BE emission for a 100Å acceptor (Be) doped quantum well, excitation is resonant with the FE. The increasing initial slope with temperature indicates a faster capture rate of the free exciton at the acceptor.

Figure 2. Temperature dependence of capture rate for both the 100Å and 150Å quantum wells, for comparison the results for capture at an Indium acceptor in bulk Si is also shown. Good agreement with a power law dependence of capture rate on temperature is achieved at high temperatures, at low temperatures the data indicates a much weaker dependence on temperature. This effect is due to the localisation of the free exciton at interface roughness which inhibits the capture.

4. Conclusion

The temperature dependence of the capture rate has been measured for acceptor doped GaAs/AlGaAs quantum wells of 100Å and 150Å well width. The low temperature behaviour is found to contradict the trend expected from the standard capture model based on cascade relaxation from excited states. The mechanism for this anomalous behaviour is believed to be the localisation of the free exciton thereby inhibiting capture at low temperatures. With increasing temperature there is a thermal activation behaviour of the localised free exciton such that there is a dramatic increase in the mean thermal velocity. The increase in thermal velocity is the dominant contribution to the capture rate at lower temperatures, at higher temperatures the decrease in capture cross section becomes the dominant term and a typical negative power law dependence can then be observed.

5. References

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$$V_{th} = \left\{ \frac{8KT}{\pi m_{ex}} \right\}^{1/2} \quad (4)$$

where V_{th} is the mean thermal velocity of the free exciton and m_{ex} is the exciton mass. The capture rate is found to be suppressed at low temperatures due to the effect of localisation on the free exciton thermal velocity. The localisation energy suggested by the temperature at which the behaviour deviates from the power law dependence is approximately consistent with the Stoke's shift observed in these samples. As a result there are two competing responses to increasing temperature. The capture cross section decreases with increasing temperature as is required by the cascade capture model, however, at low temperatures the dominant mechanism is the thermal activation behaviour observed in the thermal velocity of the FE. The results of Fig.1. as suggested are sensitive to the initial capture rate which is in turn more sensitive to the effects of localisation. The cold exciton is easily localised inhibiting further capture to an impurity, increasing lattice temperature has a relatively weak effect on this initial process. This process, if indeed limited by the localisation, is determined by the hopping probability between localised islands, such a process is known to have a $T^{-1/4}$ temperature dependence[12], such a low value of the exponent is indeed suggested by the low temperature data of Fig.2.