NRC Publications Archive Archives des publications du CNRC

Temperature dependence of ultrasharp luminescence lines in AlyIn_{1-y}As/AlxGa_{1-x}As self-assembled quantum dots

Raymond, S.; Fafard, S.; Charbonneau, Sylvain

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. /
La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version
acceptée du manuscrit ou la version de l'éditeur.

For the publisher's version, please access the DOI link below. / Pour consulter la version de l'éditeur, utilisez le lien
DOI ci-dessous.

Publisher's version / Version de l'éditeur:

<https://doi.org/10.1139/p96-862>

Canadian Journal of Physics, 74, suppl.1, pp. 216-219, 1996

NRC Publications Archive Record / Notice des Archives des publications du CNRC :

<https://nrc-publications.canada.ca/eng/view/object/?id=b07c61f7-f84d-44f4-9732-6edf051ed296>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=b07c61f7-f84d-44f4-9732-6edf051ed296>

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at

<https://nrc-publications.canada.ca/eng/copyright>

READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site

<https://publications-cnrc.canada.ca/fra/droits>

LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at

PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the
first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la
première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez
pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

21
sent RS

**Temperature dependence of ultra-sharp luminescence lines in
 $\text{Al}_y\text{In}_{1-y}\text{As}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ self-assembled quantum dots**

S. Raymond*, S. Fafard, and S. Charbonneau

Institute for Microstructural Sciences, National Research Council,

Ottawa, Ontario, Canada, K1A 0R6.

(August 1995)

* Also at Department of Physics, University of Ottawa, Ottawa, Ontario, Canada K1N 6N5.

PACS # 78.47, 78.55.

Ensembles of ~ 600 $\text{Al}_y\text{In}_{1-y}\text{As}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ self-assembled quantum dots (QDs) are investigated using photoluminescence (PL) and time-resolved PL (TRPL) in the visible. At very low excitation intensities, the PL spectrum shows multiple ultranarrow luminescence lines (FWHM ~ 200 μeV), which are attributed to the ground state transition of a few dots (4 or less). The temperature and intensity evolution of these sharp lines is then monitored. The temperature-dependent measurements show that the linewidth and lifetime of the narrow lines remain constant up to the onset of thermionic emission. Intensity-dependent measurements show that for high excitation density the collective background, emitted by the ensemble of QDs, is enhanced relative to the amplitude of individual ultranarrow lines.

Des ensembles d'environ 600 îlots quantiques d' $\text{Al}_y\text{In}_{1-y}\text{As}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ sont étudiés grâce à des mesures de photoluminescence (PL) et de photoluminescence résolues dans le temps (TRPL). Lorsque l'intensité d'excitation est faible, le spectre de PL montre une série de raies très étroites (~ 200 μeV) résultant de l'émission de quelques îlots (4 ou moins). L'influence de la température sur ces raies d'émission est étudiée, et les résultats montrent que leurs temps de vie et leurs largeur restent constants jusqu'à ce qu'on observe de l'émission thermionique. De plus, on montre qu'une haute densité d'excitation favorise l'émission de fond collective des îlots au détriment des raies individuelles.

I. Introduction

Recently, the spontaneous island formation in the Stranski-Krastanow growth mode of highly strained semiconductors have been exploited to produce self-assembled $\text{Al}_y\text{In}_{1-y}\text{As}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ QDs with emission in the visible (red) [1-3]. Non-resonant PL experiments, performed on ensembles with a different number of QDs, exposed the sharpness of the single-dot emission forming the smooth Gaussian lineshape which is often observed when probing a larger number of slightly different QDs [2]. However, the influence of the temperature and of the intensity of excitation remains to be investigated in the case where sharp QD lines can be observed. In particular, the linewidth and lifetime temperature dependence are crucial to determine if the system is truly of 0-Dimensional nature. For a QD, these two quantities should be temperature independent, whereas other temperature dependencies are expected for higher dimensionalities [4-5]. Also, the intensity dependence might reveal some effects typical of 0-D systems, such as renormalization of the emission energy due to multiple-carrier effects at the moment of recombination. Therefore, we present measurements of temperature and intensity dependence of the sharp spectral PL lines associated with the individual QD emission.

II. Experimental details

The sample used for this work was grown by molecular beam epitaxy using the spontaneous island formation in the Stranski-Krastanow growth mode [6]. From substrate to top, the layers grown are: GaAs (100) buffer, 200 nm $\text{Al}_{0.35}\text{Ga}_{0.65}\text{As}$ barrier layer, $\text{In}_{0.55}\text{Al}_{0.45}\text{As}$ QD layer, 30 nm

$\text{Al}_{0.35}\text{Ga}_{0.65}\text{As}$ barrier layer and a thin GaAs cap layer. The QD layer is composed of a 2.1 nm quantum well, called the wetting layer (WL), on top of which QDs are formed. The QDs thus produced have a diameter of 17 ± 2 nm, a thickness of 3.2 nm and their areal density is $200 \mu\text{m}^{-2}$. More detailed sample and growth descriptions have been published elsewhere [1,3]. In this paper, we report mainly on small ensembles of QDs obtained from the above sample. These ensembles were achieved by defining $3 \mu\text{m}^2$ mesas, resulting in a small group of ~ 600 QDs being probed [2].

For this work, the PL measurements were performed using an Ar^+ ion pumped CW DCM dye laser system, emitting at 1.96 eV as an excitation source. The PL was dispersed by a 0.75 m double spectrometer and detected by a 25 mm diameter cooled imaging photomultiplier tube (IPMT). Combined with a position computer, the IPMT gives the time and location of given photon events, allowing electronic gating of the signal and precise detection-energy selection[7]. The resolution of the PL system was 80 μeV in the red. For TRPL experiments, the optical excitation was provided by a frequency-doubled mode locked $\text{Nd}^{3+}:\text{YAG}$ laser synchronously pumping a dye laser, producing 5 ps pulses with a repetition rate of 4 MHz. The time resolution of this system was 100 ps.

III. Results and discussion

Fig. 1 shows the steady state PL spectra obtained for an unetched part of the sample. The laser was focused to a spot $\sim 80 \mu\text{m}$ in diameter, thus probing a population of 10^6 QDs. The PL scan shows three peaks at 1.887, 1.96 and 1.993 eV respectively. The first peak has been assigned to QD emission, the shoulder observed at 1.96 eV is attributed to WL PL, while the peak at 1.993

eV comes from barrier luminescence. The QD peak has a smooth Gaussian shape with a full-width-at-half-maximum (FWHM) of 46 meV. This smooth shape has been shown to result from the convolution of a great number of slightly different QDs, each emitting a narrow luminescence line at a slightly different energy. Indeed, previous experiments [2] have shown that for a small number of QDs probed (~ 600), sharp spectral lines start appearing on top of the smooth Gaussian spectrum, which seemed to confirm the 0-D nature of the Density of states (DOS).

Fig. 2 shows the QD PL spectrum obtained for a mesa ensemble at different temperatures with an excitation power of $250 \text{ nW}/\mu\text{m}^2$. The spectra have been shifted to align the sharp lines at all temperatures. The spectrum exhibits several sharp spectral features of similar magnitude with a FWHM ranging from 190 to $400 \mu\text{eV}$. Other mesas excited above the barrier material ($E_{\text{exc}}=2.0 \text{ eV}$) with very low power densities ($25 \text{ nW}/\mu\text{m}^2$) have shown some spectral features of $\sim 100 \mu\text{eV}$. Each sharp feature corresponds to the PL trace of a few QDs, typically one to six [1], emitting at similar wavelengths. The non-zero PL signal observed in between adjacent lines is due to the finite intrinsic emission linewidth of each single QD. This collective background has to be distinguished from the dark count signal since the former is a property of the QD population and the latter is a property of the set-up. Spectral features for which the intensity level remains well above the collective emission background show very little broadening as the temperature is increased up to 70K. Some lines, such as G, do show some broadening, although much less than the increase in the thermal energy ($k_B T$). This small effect can be attributed to the background noise in the experiment since at 70K the PL intensity has decreased by a factor of 10 due mostly to thermionic emission [8] and several lines are barely seen above the dark count level for such small excitation intensities. These temperature-independent linewidth are in contrast with the case of a quantum

well where the 2-D density of state permits a continuum of energy leading to thermal broadening of the emission line. The difference between the two cases lies in the density of states (DOS) and the behavior in temperature observed here clearly is the one expected for a δ -function like density of states with no carrier population in the radiative excited-state.

Figure 3 shows the temperature evolution of the time-decay of a single sharp line up to 60 K. The TRPL traces were obtained using non-resonant excitation with the set-up electronically gated to detect photons at the wavelength of the peak indicated in the inset of the figure. The decay traces obtained are single-exponential over two orders of magnitude, and the observed decay times do not vary below 60 K. A single-exponential fit yields a lifetime of 400 ps in this temperature range, while at 60K the decay time speeds up to 340 ps due to thermionic emission above the barrier material, allowing other relaxation mechanisms to occur such as non-radiative recombination near the edge of the mesa. Temperature dependent measurements done on 2D and 1D systems have shown longer lifetimes with increasing temperature[4]. This effect was attributed to thermal population of high k-vector states which cannot decay radiatively. Here however, the DOS has no dispersion and the lifetime therefore remains constant until thermionic emission occurs. These results are also consistent with theoretical calculations done for QDs with large level spacings [5].

Fig. 4 Shows the evolution of the PL as a function of excitation power at low temperature. The low power spectrum ($150 \text{ nW}/\mu\text{m}^2$) shows similar features as in fig. 2a. As the intensity is increased, most of the sharp spectral features (A to F) observed lose amplitude with respect to the collective background, and in some cases even disappear completely, the most striking example being peak H. Lines that remain identifiable tend to stay narrow, although some of them do show

evidence of broadening (peak B for example). In some cases, higher intensities have little effect (J, K, L) or even cause the emergence of new features. Line I for example is barely detected at low intensity, but progressively rises above the background as the intensity is increased. These observations cannot be related to temperature effects, since fig. 2 demonstrates that increasing the temperature does not change the spectrum significantly. An increase in the photocarriers population would be more likely to be at the origin of the new spectral features. For example, the presence of carriers in the QD at the moment of recombination is expected to have an effect on the emission energy [9]. The emission can be redistributed at different energies and this would be seen as a rise of the background in the CW spectrum, and in some cases new QD emission lines can cause the emergence of new spectral features. Neglecting the dynamics in the higher energy states, the steady-state population in the QD ground state (GS) would be given by the product of the photogenerated carrier flux, $\Phi(I_{\text{ex}})$, and the lifetime in the QD GS, τ_{GS} . For $\tau_{\text{GS}} = 400$ ps and for the intensity used of $150\text{nW}/\mu\text{m}^2$, $1.5\mu\text{W}/\mu\text{m}^2$ and $4.5\mu\text{W}/\mu\text{m}^2$, $\Phi(I_{\text{ex}})\tau_{\text{GS}}$ is respectively 0.003, 0.03, and 0.08 per QD¹. The observed spectral modifications could also be explained in part, if at the lowest intensities only some of the QDs emit, and the remaining of the QDs start to contribute only at higher excitation densities.

¹ A 30% reflection at the sample surface and a 0.5% absorption in the wetting layer were included in the calculation of $\Phi(I_{\text{ex}})$.

IV. Conclusion

We have shown that the lifetime and linewidth of sharp spectral lines, obtained in the PL spectrum of small ensembles of QDs, do not change up to the onset of thermionic emission, thus clearly demonstrating the 0-D nature of the confinement. Also, for low temperatures it was shown that the sharp lines lose amplitude with respect to the collective background as the excitation intensity is increased.

Acknowledgments

Some of us (S.F. and S.R.) would also like to acknowledge NSERC for their support.

PL intensity (a.u.)

PL intensity (a.u.)