

Self-assembled InAs/GaAs quantum dots studied with excitation dependent cathodoluminescence

Y. Tang, D. H. Rich,^{a)} I. Mukhamezhanov, P. Chen, and A. Madhukar

Photonic Materials and Devices Laboratory, Department of Materials Science & Engineering, University of Southern California, Los Angeles, California 90089-0241

(Received 13 April 1998; accepted for publication 9 June 1998)

We have examined the optical properties of self-assembled InAs quantum dots (QDs) with polarization sensitive and time-resolved cathodoluminescence (CL) techniques. The InAs QDs were formed via self-assembly during molecular beam epitaxial growth of InAs on unpatterned GaAs(001). CL spectra exhibited a two-component line shape whose linewidth, intensity, and peak positions were found to be temperature and excitation dependent. The two components are found to be consistent with state filling of the QDs, resulting in emission involving ground state and excited state excitonic transitions. The luminescence intensities and lineshapes of the QD and wetting layer (WL) excitonic transitions were analyzed with constant excitation and time-resolved CL for various temperatures and excitation levels to study the thermal activation, re-emission, and recombination kinetics of carriers. Thermal quenching of the QD ground state and excited state components in the 105–175 K range is correlated with a rise in the WL emission, showing that the WL carrier distribution is fed partially by thermal reemission of carriers from the QDs. A more rapid thermal quenching of the QD and WL excitonic emissions in the 185–300 K range is consistent with thermal reemission of carriers into the GaAs matrix. Time delayed CL spectra show that carriers in the excited states rapidly feed the ground state during the decay phase of the luminescence, further confirming that the two components are derived from the same QD. The CL decay time for the QD luminescence was found to be wavelength and temperature dependent, owing to thermal reemission into the WL. The CL decay times ranged from 0.1 to 2.0 ns. A polarization anisotropy was found in CL spectroscopy, revealing the importance of the stress anisotropy in both the formation of QDs and their subsequent optical emission. © 1998 American Institute of Physics.

[S0021-8979(98)02218-X]

I. INTRODUCTION

The molecular beam epitaxial (MBE) growth of coherent three-dimensional (3D) islands has recently drawn a great deal of attention, owing to its great potential for applications in lasers and infrared detectors that are based on the utilization of optically active quantum dots (QDs).^{1–6} Under certain MBE growth conditions the formation of a high-density strained pyramidal InAs QDs on GaAs has been demonstrated and studied by several groups with photoluminescence (PL).^{2–10} The InAs self-assembled quantum dots (SAQDs) form with a substantial variation in size, thereby yielding a unique spectrum of confined δ -like states for each QD. The inhomogeneity in height and lateral size of the SAQDs results in a broad (40–60 meV) luminescence line shape, whose width depends on the level of excitation and number of QDs which are excited.⁶ For SAQDs grown on unpatterned substrates, a partial linear polarization anisotropy in photoluminescence has been previously observed and was attributed to the asymmetry of the island shape.^{9,10} Various attempts have been made to control the size-shape morphology and spatial arrangement of InAs SAQDs. Experiments have shown that SAQDs, at the later stages of growth,

have a tendency toward lateral size equalization, owing to a strain-dependent incorporation and elastic interaction of islands through the substrate.^{2,11} Attempts for size equalization have been made via vertical self-organization by utilizing the strain field in the cap layers induced by 3D islands as a driving force.² Recently, a lateral selectivity in the control of InAs SAQD formation has been demonstrated by the growth of InAs on patterned GaAs(001) substrates.¹² Recent work has focused on the temperature and excitation dependence of the luminescence properties of InAs SAQDs.^{7,8,13–17} Conflicting interpretations have arisen over the interpretation of multiple peaks which can appear in the broad luminescence lineshape.⁸ Time-resolved PL studies have further explored the temperature dependence of the decay time and relaxation kinetics of excited carriers in SAQDs.^{14,18–21}

In this work, we have examined the optical properties of SAQDs grown on unpatterned substrates with spatially, spectrally, and temporally resolved cathodoluminescence (CL) techniques. We have performed a detailed analysis of a sample which exhibits multiple and distinct emission features in its CL line shape to clarify issues of QD size distributions and state filling effects. Polarization-sensitive detection is further utilized to examine the presence of strain and probe the character of the hole states in the optical transitions. The CL approach can provide simultaneous spatial and spectral information which is ideally suited for the study of

^{a)} Author to whom correspondence should be addressed; electronic mail: danrich@almaak.usc.edu

nanostructures grown on patterned substrates. We have examined and compared the recombination and relaxation kinetics of carriers confined to SAQDs, the temperature dependence of the luminescence line shape and energy shifts, and excitation dependence of the polarization anisotropy and spectral line shapes.

II. SAMPLES AND EXPERIMENTAL SETUP

Self-assembled InAs quantum dots structures were grown by MBE on planar GaAs(001) $\pm 0.1^\circ$ substrates. A thick high quality GaAs buffer layer was first grown. Details of the growth and surface morphology as a function of growth for similar samples have been described elsewhere.² A sample was grown with a ~ 1.74 monolayer (ML) InAs deposition at 500 °C and 6×10^{-6} Torr As_4 pressure. Under these conditions the initial GaAs(001) surface showed a $c(4 \times 4)$ reconstruction. Growth of other samples showed that the reflection high-energy electron diffraction (RHEED) pattern becomes spotty after ~ 1.57 ML InAs deposition, revealing the 2D to 3D morphological transition. Previous atomic force microscopy (AFM) and scanning tunneling microscopy (STM) studies of the uncapped samples confirmed that the coverage of ~ 1.57 ML is the transition coverage for 2D–3D growth.^{2,11} These studies also showed that the 3D InAs islands coexist with small 2D clusters for an InAs delivery of ~ 1.74 ML.² The density of the SAQDs was determined by AFM and is $\sim 450 \mu\text{m}^{-2}$. The lateral size of the island ranges from 11 to 21 nm, with 90% of the islands falling in the 13–17 nm range. A 50-nm-thick GaAs capping layer was grown by migration-enhanced epitaxy at 400 °C, resulting in buried InAs QDs suitable for optical studies. A recent AFM study by Garcia *et al.* has shown that a large anisotropic redistribution of the InAs island material can occur upon deposition of the GaAs capping layer, thereby significantly altering the size and shape distributions of the buried InAs QDs.²²

The CL experiments were performed with a modified JEOL-840A scanning electron microscope (SEM) using a 10–15 keV electron beam with probe current ranging from 0.1 to 10 nA.²³ The temperature of the sample was varied from 87 to 300 K. Constant excitation CL spectra were obtained with a 0.275 m spectrograph with a Si CCD array detector. In CL imaging and time-resolved experiments, a Si avalanche photodiode was used. The method of delayed coincidence in an inverted single photon counting mode was employed for the time-resolved study with a time resolution of ~ 100 ps.²³ Electron beam pulses of 50 ns width with a 1 MHz frequency were used to excite the sample. Linearly polarized cathodoluminescence (LPCL) measurements were performed by detecting luminescence with a rotatable vacuum linear polarizer mounted in the SEM chamber. Luminescence was detected with the electric field (\mathbf{E}) along either the $[110]$ or $[\bar{1}\bar{1}0]$ directions to evaluate the presence of an anisotropy caused by strain or quantum confinement.

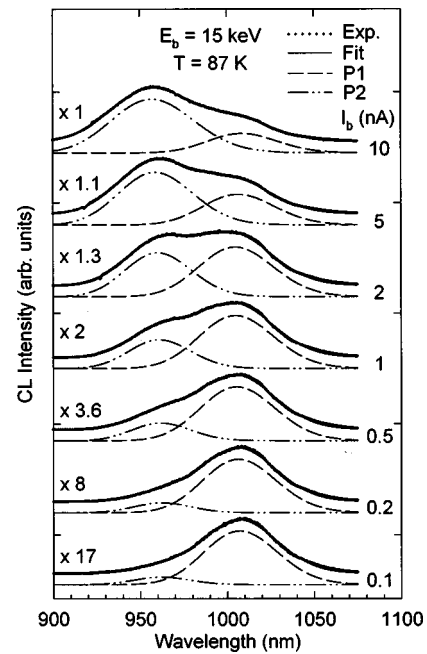


FIG. 1. CL spectra (dots) acquired for various electron beam currents, I_b , ranging from 0.1 to 10 nA. Spectra were decomposed into two Gaussian components represented by dash (P1) and dot-dash (P2) lines which are shown offset below the data. The fits are represented by solid lines running through the data.

III. RESULTS AND DISCUSSION

A. Excitation-dependent CL

Spatially averaged CL spectra were acquired while the electron beam rastered over a $128 \times 94 \mu\text{m}^2$ area with various probe currents, I_b , as shown in Fig. 1 for a temperature (T) of 87 K. The CL spectra exhibit a broad asymmetric line shape possessing a small shoulder on the shorter wavelength side of the lineshape for $I_b < 0.1$ nA. As the beam current is increased from 0.1 to 10 nA, this shoulder grows and evolves into a distinct peak. We have decomposed the spectra into two components P1 and P2 by fitting the spectra to the sum of two Gaussian components, each with a variable full width at half-maximum (FWHM) and variable peak energy position. The decomposition into P1 and P2 is indicated by dash and dotted curves in the figure. For a fixed beam current, the relative contribution of the higher energy component, P2, increases as the temperature increases. The two components in the luminescence spectra are attributed to two distinct excitonic transitions, originating from the ground-state (P1) and excited states (P2) of individual QDs. Similar results have been observed in excitation dependent PL studies of InAs SAQDs.⁷ The large FWHM is due to the usual statistical nature of the SAQD ensemble, which results in large ($> \sim 10\%$) QD size variations during growth.

Results of the fits are plotted in Fig. 2 for various beam currents at $T = 87$ K. The peak energy positions and integrated intensities of the P1 and P2 components are found to vary as the excitation level is increased from $I_b = 0.1$ to 10 nA. The intensity of the ground state transition, P1, begins to saturate when $I_b \approx 1$ nA. This e -beam excitation level corresponds to a steady-state average occupation of about 2.7

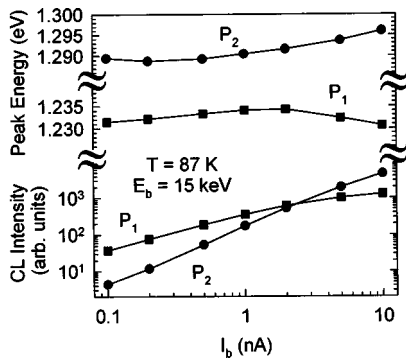


FIG. 2. The P1 and P2 peak energies and integrated intensities for various beam currents.

electron-hole pairs per dot.²⁴ P1 becomes blueshifted by ~ 3 meV as the current increases from 0.1 to 1 nA, and then redshifts by ~ 4 meV over the 1 to 10 nA range. The initial blueshift may be due to an enhanced carrier collection in smaller QDs possessing larger effective band gaps as the excitation level is increased, thereby causing a shift in the weighted ensemble towards larger energies. The subsequent redshift is evidence for a Coulomb interaction between excitons in the same QD, as the ground and excited state excitonic transition energies are expected to be reduced when the dots become occupied with multiple excitons.^{25,26} Owing to the strong localization of excitons in QDs, biexciton binding energies of ~ 5 meV are expected,^{25,26} consistent with our observed redshift. The excited state component P2 monotonically shifts to the blue by ~ 7 meV over the 0.6 to 10 nA range, suggesting that additional excited state filling occurs at higher excitation levels. This effect is in competition with the Coulomb interaction, which should also reduce the energy of the excited states. The FWHM increases only slightly from 60.5 to 62.1 meV for P1 as probe current increases from 0.1 to 1 nA and after that decreases to 57.2 meV at $I_b = 10$ nA. However, the FWHM of P2 increases monotonically from 51.0 to 75.2 meV with increasing probe current over the $0.1 \leq I_b \leq 10$ nA range, consistent with the filling of additional excited states in the dots. A blue shift of the P2 peak should occur as the occupation of high-energy excited states is expected to simultaneously broaden the distribution. The P1 and P2 intensities are found to cross at $I_b \approx 2$ nA, indicating the increasing population of the excited states once the ground state excitonic transitions saturate.

B. Temperature-dependent CL and thermalization in SAQDs

We have examined the temperature dependence of the CL spectra for the temperature range $87 \leq T \leq 300$ K. The integrated CL intensities of components P1, P2, and the InAs wetting layer (WL) transition are shown in a log of CL intensity versus $1/T$ plot in Fig. 3. As the temperature is increased from ~ 105 to 150 K, a decrease in the P1 and P2 intensities occurs with an attendant increase in the WL emission intensity. The WL emission occurs at 1.423 eV for $T = 103$ K. The decrease in the P1 and P2 emission with increasing temperature is evidently due to the increase in ther-

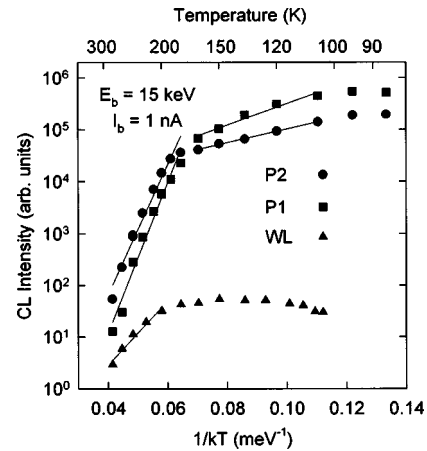


FIG. 3. The P1, P2, and wetting layer (WL) integrated CL intensities for various sample temperatures. A log of intensity vs $1/kT$ plot is shown from which activation energies are obtained from slopes of lines fit to the data. The WL intensities are shown reduced by a factor of 10 for clarity.

mal reemission of captured excitons in the QDs prior to their recombination in the QDs. Once re-emitted into the WL and GaAs barrier, the carriers can recombine through other radiative and nonradiative channels. One such channel is the WL emission, which shows a corresponding small increase in its integrated intensity (Fig. 3) as the P1 and P2 emissions decrease in the $105 \leq T \leq 150$ K range. The complementary behavior of the QD and WL emissions provides direct evidence for thermal transfer to the WL in this temperature range.

The CL intensity of the P1 and P2 peaks exhibits an Arrhenius-type dependence whose activation energy varies with the temperature range. To estimate the activation energy, we have performed linear fits of the log of intensity versus $1/T$ data for two different temperature ranges, as shown by the lines in Fig. 3. In the $105 \leq T \leq 175$ K range, the activation energies, E_1 and E_2 , for P1 and P2 emission are 48 and 30 meV, respectively. In the $180 \leq T \leq 300$ K range, E_1 and E_2 are 333 and 287 meV, respectively. We attribute the different activation energies to two fundamentally different thermal reemission processes occurring in the different temperature ranges. In the lower temperature range, excitation of single electrons or holes to the WL is expected to occur, owing to the slight rise in the WL emission in this range. The results show that $E_1 > E_2$ in both ranges, which should be expected given that holes involved in the P1 emission have larger confinement energies than those involved in the P2 emission. The magnitude of these activation energies is related to the energy difference between the electron and hole levels of the WL and QD. Previous calculations have shown that for QDs emitting in the 1.20–1.30 eV energy range, the electron levels between the QD ($|000\rangle_e$) and WL are separated by 20–80 meV.⁷ Likewise, the excited state hole levels ($|001\rangle_h$) and WL hole levels are possibly separated by 50–100 meV.⁷ Thus, the measured activation energies in the lower temperature range are roughly consistent with energies required for single particle excitations of electrons and/or holes from confined QD states to the WL continuum levels. Once carriers have been re-emitted into the WL they may recombine nonradiatively through defects, be

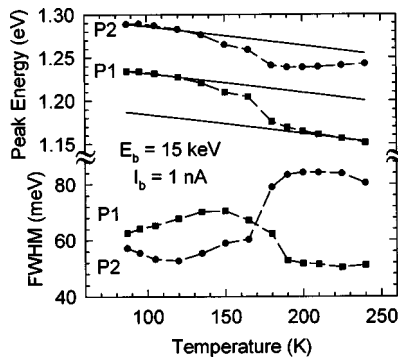


FIG. 4. The P1 and P2 peak energies and FWHM of each component vs temperature. The predicted InAs band gap vs temperature (i.e., Varshni's law) is shown with solid lines.

captured by QDs, or recombine radiatively in the WL. The latter most again gives rise to the increase in the measured WL emission in this temperature range.

The activation energies in the 180–300 K temperature range are much closer to the difference in energy between the GaAs excitonic transition (~ 1.50 eV) and the P1 and P2 transitions. Thus, it is apparent that a direct excitation of single particles and excitons to the GaAs barrier should be possible in this temperature range. Likewise, carriers in the WL can also be thermally reemitted to the surrounding GaAs matrix, thus also giving rise to its quenching in this temperature range. A linear fit is also illustrated for the log of the WL intensity versus $1/T$ in Fig. 3, resulting in an activation energy E_3 of 143 meV, roughly consistent with the energy difference between the GaAs and WL luminescence energies throughout the 180–300 K range.

Further results of the fits are shown versus temperature in Fig. 4. The solid lines in the top panel show the expected shifts of the peak position based on the Varshni law²⁷ for the band gap shift of InAs with temperature. The rapid deviation from the Varshni dependence in the 140–180 K range suggests that a thermal redistribution of excitons into different QD groups, possessing different size/shape distributions, occurs. We suggest that additional groups of QDs may be accessed through thermal re-emission and diffusive transport in the WL. Since the P1 emission energy is decreasing with temperature at a rate faster than the Varshni prediction, we conclude that larger QDs comprise this new group, thereby accounting for the redshift. From 170–240 K, the P1 emission again follows the Varshni dependence. A previous temperature dependent PL study of InAs SAQDs has likewise shown evidence for thermally assisted carrier transfers to QDs possessing different size distributions.⁸ We expect that each new group of QDs involved in the thermal redistribution will exhibit similar P1 and P2 emissions, owing to ground state and excited state excitonic transitions. The resulting P1 and P2 energies will depend on largely unknown details, such as the strain, size, and shape morphology of the various groups of buried QDs that are thermally accessed at higher temperatures.

The further deviation, however, of the P2 emission from the Varshni dependence throughout the 170–240 K range may indicate that a thermal redistribution of excitons occu-

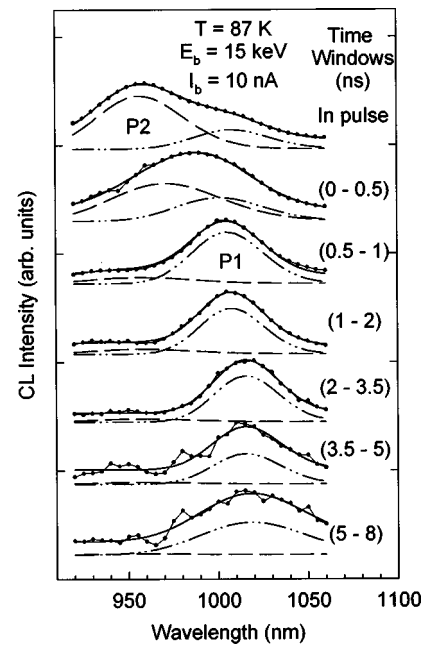


FIG. 5. Time-delayed CL spectra acquired for various time windows shown during the decay phase. The decomposition into the P1 and P2 components are shown.

pying high-energy states occurs. Several, nearly degenerate and closely spaced higher lying hole levels in InAs QDs have been predicted,^{7,18} and these states may have energy spacings less than kT for this temperature range. At higher temperatures, the weight of populated excited states in the QDs is expected to shift towards higher energies, accounting for the blueshift of P2. Further corroboration of this effect is observed in the lower panel of Fig. 4, which shows the FWHM of each peak versus temperature. A rapid decrease and increase of the FWHM for P1 and P2, respectively, is observed in the 140–180 K range, where the peak energies shift most rapidly. The broadened P2 emission in the 170–240 K range is also consistent with a thermal redistribution into higher lying hole states in this range.

C. Time-resolved CL measurements

The carrier relaxation kinetics have been studied with time-resolved CL measurements. A series of CL transients were taken with the wavelength ranging from 920 to 1060 nm at a temperature of 87 K. Time delayed spectra of the P1 and P2 emission were reconstructed from the CL transients, as shown in Fig. 5. The spectrum labeled *in pulse* was acquired with the time window centered in the middle of the 50 ns excitation pulse, thereby obtaining a spectrum similar to the constant excitation CL measurement in Fig. 1. Spectra are shown for various time windows during the decay phase. A rapid redshift is seen to occur, indicating that carriers are relaxing rapidly to lower energy states. Using the same fitting procedure employed for the constant excitation CL spectra, we have decomposed the time-delayed spectra into P1 and P2 components, as shown in Fig. 5. The rapid redshift is now observed to be due to a differing rate in which the P1 and P2 emissions decay. The P2 emission has noticeably dissipated to a weak background in the 1 to 2 ns time win-

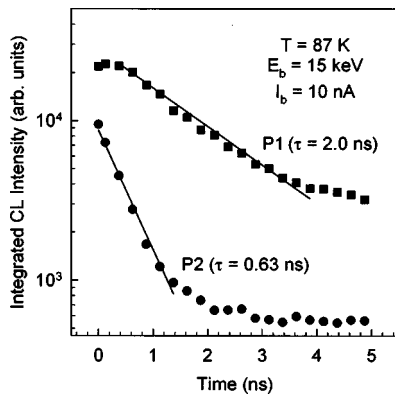


FIG. 6. Integrated P1 and P2 intensity vs time transients. Solid lines are fit to the data in a semilog plot to obtain the decay times.

dow. A redshift of ~ 15 nm is observed for the P1 emission as the exciton decay proceeds towards equilibrium.

The integrated intensities of the P1 and P2 emissions are shown as a function of time in Fig. 6. The P2 peak decays rapidly with a decay time of 0.63 ns, as obtained by the linear fit in the 0–1.2 ns range. The P1 emission, however, exhibits a much slower decay with a decay time of 2.0 ns, as obtained from a fit in the 0.7–4 ns range. A delay of the P1 decay is also observed in the 0–0.7 ns range. This delay is direct evidence for carrier feeding from the excited states (P2 emission) to the ground state (P1 emission) during the decay phase. Carriers in the excited states can recombine through radiative emission or relax to the ground state, provided the ground state is empty. The temporary supply of excess carriers from the P2 states will feed the P1 ground state such that it is fully occupied for ~ 0.7 ns, after which it exhibits the usual exponential decay behavior.

Similar time-resolved CL measurements were performed with varying temperatures ranging from 87 to 200 K. The lifetimes are found to reduce from 2.0 to 1.2 ns for P1 and from 630 to 90 ps for P2, as shown in Fig. 7. Theoretical studies predict that the intrinsic carrier lifetime of excitons in QDs should increase linearly with temperature in the high temperature range.²⁸ In this temperature range, it is also possible however for defect-induced nonradiative recombination

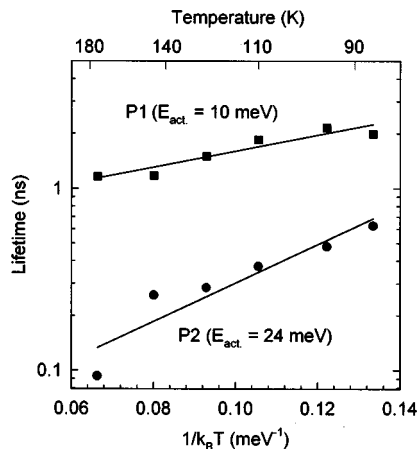


FIG. 7. Lifetimes of P1 and P2 components vs $1/kT$. Activation energies are obtained from the linear fits, as shown.

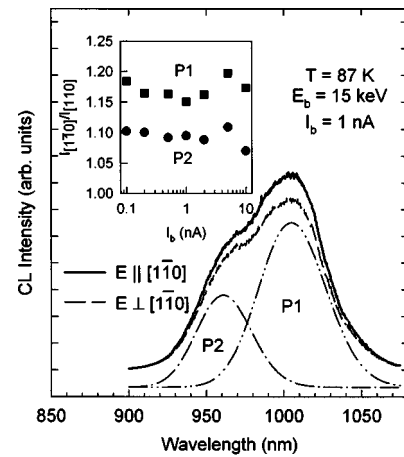


FIG. 8. Linearly polarized cathodoluminescence (CL) spectra acquired with electric field, E , parallel and perpendicular to the $[1\bar{1}0]$ direction. The inset shows the polarization ratio as a function of electron beam current.

and thermal re-emission to occur, as previously discussed, which will reduce the measured decay time. Carriers in QDs re-emitted to the WL and GaAs barriers can be captured by defects prior to recapture, thereby increasing the decay rate. The re-emission rate of carriers increases as temperature increases, which increases the CL decay rate at higher temperatures. The thermal activation energies are estimated with an Arrhenius fit, as shown in Fig. 7, and they are 10 and 24 meV for P1 and P2, respectively. These values are again consistent with re-emission of QD carriers into the WL for the 105–180 K range.

D. Linear polarization measurements

Previously, linearly polarized cathodoluminescence (LPCL) measurements have been employed to study the influence of strain on the optical properties of strained $(\text{InP})_2/(\text{GaP})_2$ quantum wires.²⁹ The presence of a strain may effect the relative admixtures of heavy- and light-hole (hh and lh) characters in the valence-band wavefunctions, thereby potentially inducing polarization effects in the excitonic luminescence. Such measurements can be employed to help distinguish between states possessing varying degrees of hh and lh admixtures.

LPCL spectra of the QD luminescence are shown in Fig. 8. A partial linear polarization along the $[1\bar{1}0]$ direction is observed, as evidenced by a $\sim 13\%$ increase in the integrated CL emission for the $E \parallel [1\bar{1}0]$ detection orientation, relative to $E \perp [1\bar{1}0]$. A similar polarization behavior in photoluminescence from SAQDs has been reported by Nabetani *et al.*,⁹ who observed a linear polarization anisotropy in PL by a factor of ~ 2.9 . They attributed the polarization anisotropy to the elongation of quantum dot along the $[110]$ direction as evidenced by their TEM, STM, and RHEED results.⁹ The average structural anisotropy in the elongation of the InAs islands was observed to be $L_{1\bar{1}0}/L_{110} \approx 1.13$, where L is the maximum length of the island along the corresponding (110) direction.⁹ Another study by Saito *et al.*¹⁰ showed a structural anisotropy of 1.94 and PL polarization ratio of 1.37.

We have decomposed the LPCL spectra into P1 and P2 components as in our previous excitation-dependent analysis in order to evaluate differences in anisotropy between the two components. The decomposition of the CL spectra shows polarization ratios, $I_{1\bar{1}0}/I_{110}$, of 1.17 and 1.09 for P1 and P2, where $I_{1\bar{1}0}$ and I_{110} are the integrated CL intensities for light with its field parallel to the [110] and $[1\bar{1}0]$ directions, respectively. These ratios are essentially independent of the excitation level, as shown in the inset of Fig. 8, despite the large variation in the P1 and P2 relative intensities with beam current (Figs. 1 and 2).

There are a few interesting ramifications worth mentioning. The polarization of the luminescence is primarily determined by the character of the hole wavefunction involved in the excitonic transition. Since P1 and P2 possess different polarization anisotropies, it is reasonable to conclude that these transitions involve hole states possessing different admixtures of hh and lh characters. That is, we argue that P1 and P2 should involve excitonic transitions with different hole states, each possessing a different energy and hh/lh character. Second, the existence of an anisotropy which is constant over a wide range of e -beam excitation suggests that the SAQDs possess a homogeneous or roughly constant shape and strain distribution. Randomly oriented SAQDs would be expected to exhibit no polarization anisotropy. TEM measurements of the uncapped islands in a 1.74 ML InAs sample prepared with similar growth conditions show pyramidal islands with primarily {302} side facets, independent of island size.³⁰ If the facets and island shapes were to vary considerably with island size, we would expect an excitation dependent polarization anisotropy. Likewise, the strain distribution in and surrounding an island will depend on its shape and not on its size.⁷ Variations in strain distribution can markedly affect the polarization anisotropy in nanostructures.²⁹ Thus, the constant polarization anisotropy over a wide range of excitation is consistent with islands that exhibit a roughly constant shape, strain, and orientation distribution, while the size variation of the ensemble leads to the inhomogeneous energy broadening.

IV. SUMMARY AND CONCLUSIONS

We have examined the optical properties of self-assembled InAs QDs with excitation dependent, polarization sensitive, and time-resolved cathodoluminescence spectroscopy techniques. The InAs QDs were formed via self-assembly during MBE growth of InAs on unpatterned GaAs(001). CL spectra exhibited a two-component line shape whose linewidth, relative intensity, and peak positions were obtained by decomposition into two Gaussian components and found to be temperature and excitation dependent. The two components are found to be consistent with state filling of the QDs, resulting in emission involving ground state and excited state excitonic transitions. Various blueshifts and redshifts in the peak positions of the decomposed gaussian components for increasing e -beam current are interpreted as a state filling of excited states and Coulomb effects involving multiple exciton effects at higher levels of excitation.

The intensities of the QD components and wetting layer (WL) excitonic transitions were analyzed as a function of temperature and excitation level to study the thermal activation and re-emission of carriers. Thermal quenching of the QD ground state and excited state components in the 105–175 K range is correlated with a rise in the WL emission, showing that the WL carrier distribution is fed partially by thermal re-emission of carriers from the QDs. A more rapid thermal quenching of the QD and WL excitonic emissions in the 185–300 K range is consistent with thermal reemission of carriers into the GaAs matrix. An activation energy analysis was performed to confirm the correct energetics of these thermalization processes. A polarization anisotropy was found in CL spectroscopy, revealing the importance of the stress anisotropy in both the formation of QDs and their subsequent optical emission. Ground state and excited state excitonic transitions were found to exhibit differences in polarization anisotropy, revealing that the transitions likely involve different hole states, each possessing different admixtures of heavy- and light-hole characters. Time delayed CL spectra show that carriers in the excited states rapidly feed the ground state during the decay phase of the luminescence, further confirming that the two components are derived from the same QD. The CL decay time for the QD luminescence was found to be temperature dependent, owing to thermal reemission into the WL. These results illustrate the complexity of the spectral and temperature dependence of the carrier relaxation in InAs SAQDs, and should provide a foundation for further studies of the optical properties of such systems.

ACKNOWLEDGMENTS

This work was supported by the Army Research Office, Office of Naval Research, Air Force Office of Scientific Research, and the National Science Foundation (RIA-ECS). The authors wish to thank Dr. R. Heitz for fruitful discussions.

- ¹S. Guha, A. Madhukar, and K. C. Rajkumar, *Appl. Phys. Lett.* **57**, 2110 (1990).
- ²Q. Xie, N. P. Kobayashi, T. R. Ramachandran, A. Kalburge, P. Chen, and A. Madhukar, *J. Vac. Sci. Technol. B* **14**, 2203 (1996).
- ³A. Madhukar, Q. Xie, P. Chen, and A. Konkar, *Appl. Phys. Lett.* **64**, 2727 (1994).
- ⁴S. Fafard, D. Leonard, J. L. Merz, and P. M. Petroff, *Appl. Phys. Lett.* **65**, 1388 (1994).
- ⁵J.-Y. Marzin, J.-M. Gerard, A. Izrael, and D. Barrier, *Phys. Rev. Lett.* **73**, 716 (1994).
- ⁶M. Grundmann, J. Christen, N. N. Ledentsov, J. Bohrer, D. Bimberg, S. S. Ruvimov, P. Werner, U. Richter, U. Gosele, J. Heydenreich, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, P. S. Kop'ev, and Z. I. Alferov, *Phys. Rev. Lett.* **74**, 4043 (1995).
- ⁷M. Grundmann, N. N. Ledentsov, O. Stier, J. Bohrer, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Z. I. Alferov, *Phys. Rev. B* **53**, R10 509 (1996); M. Grundmann, O. Stier, and D. Bimberg, *Phys. Rev. B* **52**, 11 969 (1995).
- ⁸L. Brusaferrri, S. Sanguinetti, E. Grilli, M. Guzzi, A. Bignazzi, F. Bogani, L. Carraresi, M. Colocci, A. Bosacchi, P. Frigeri, and S. Franchi, *Appl. Phys. Lett.* **69**, 3354 (1996).
- ⁹Y. Nabetani, T. Ishikawa, S. Noda, and A. Sasaki, *J. Appl. Phys.* **76**, 347 (1994).
- ¹⁰H. Saito, K. Nishi, S. Sugou, and Y. Sugimoto, *Appl. Phys. Lett.* **71**, 590 (1997).
- ¹¹N. P. Kobayashi, T. R. Ramachandran, P. Chen, and A. Madhukar, *Appl. Phys. Lett.* **68**, 3299 (1996).

- ¹²A. Konkar, A. Madhukar, and P. Chen, *Appl. Phys. Lett.* **72**, 220 (1998).
- ¹³R. Steffen, T. Koch, J. Oshinowo, F. Faller, and A. Forchel, *Surf. Sci.* **361/362**, 805 (1996).
- ¹⁴F. Adler, M. Geiger, A. Bauknecht, F. Scholz, and H. Schweizer, *Phys. Status Solidi A* **164**, 431 (1997).
- ¹⁵A. Patane, M. G. Alessi, F. Intonti, A. Polimeni, M. Capizzi, F. Martelli, M. Geddo, A. Bosacchi, and S. Franchi, *Phys. Status Solidi A* **164**, 493 (1997).
- ¹⁶H. Lee, W. Yang, and P. C. Sercel, *Phys. Rev. B* **55**, 9757 (1997).
- ¹⁷Y. T. Dai, J. C. Fan, Y. F. Chen, R. M. Lin, S. C. Lee, and H. H. Lin, *J. Appl. Phys.* **82**, 4489 (1997).
- ¹⁸F. Adler, M. Geiger, A. Bauknecht, F. Scholz, H. Schweizer, M. H. Pilkuhn, B. Ohnesorge, and A. Forchel, *J. Appl. Phys.* **80**, 4019 (1996).
- ¹⁹W. Yang, R. R. Lowe-Webb, H. Lee, and P. C. Sercel, *Phys. Rev. B* **56**, 13 314 (1997).
- ²⁰J. H. Sandmann, S. Grosse, G. V. Plessen, J. Feldmann, G. Hayes, R. Phillips, H. Lipsanen, M. Sopanen, and J. Ahopelto, *Phys. Status Solidi A* **164**, 421 (1997).
- ²¹R. Heitz, A. Kalburge, Q. Xie, M. Grundmann, P. Chen, A. Hoffmann, A. Madhukar, and D. Bimberg, *Phys. Rev. B* **57**, 9050 (1998).
- ²²J. M. Garcia, G. Medeiros-Ribeiro, K. Schmidt, T. Ngo, J. L. Feng, A. Lorke, J. Kotthaus, and P. M. Petroff, *Appl. Phys. Lett.* **71**, 2014 (1997).
- ²³H. T. Lin, D. H. Rich, A. Konkar, P. Chen, and A. Madhukar, *J. Appl. Phys.* **81**, 3186 (1997).
- ²⁴The relationship between the carrier occupation in the QDs, n_q , and I_b is given by $n_q \approx I_b E_b \tau_r L_D / (3 e E_g V_e A_D)$, where E_b is the e -beam energy (15 keV), E_g is the GaAs band gap (1.52 eV), V_e is the e -h excitation volume at 15 keV ($4.2 \mu\text{m}^3$), L_D is the carrier diffusion length ($0.5 \mu\text{m}$), τ_r is the e -h lifetime (0.5 ns), and A_D is the density of QDs ($450 \mu\text{m}^{-2}$). Using the values appropriate for our experimental conditions, we get $n_q \approx 2.7 I_b$ (nA).
- ²⁵Y. Z. Hu, S. W. Koch, M. Lindberg, N. Peyghambarian, E. L. Pollock, and F. F. Abraham, *Phys. Rev. Lett.* **64**, 1805 (1990).
- ²⁶S. Raymond, P. Hawrylak, C. Gould, S. Fafard, A. Sachrajda, M. Potemski, A. Wojs, S. Charbonneau, D. Leonard, P. M. Petroff, and J. L. Merz, *Solid State Commun.* **101**, 883 (1997).
- ²⁷Y. P. Varshni, *Physica (Amsterdam)* **34**, 149 (1967); InAs parameters were obtained from Landolt–Bornstein, in *Numerical Data and Functional Relationships in Science and Technology*, New Series, III/17a, edited by O. Madelung (Springer, Berlin, 1982), p. 297.
- ²⁸D. S. Citrin, *Superlattices Microstruct.* **13**, 303 (1993).
- ²⁹D. H. Rich, Y. Tang, and H. T. Lin, *J. Appl. Phys.* **81**, 6837 (1997).
- ³⁰Q. Xie, Ph.D. dissertation, University of Southern California, 1997.