Cathodoluminescence wavelength imaging of μm-scale energy variations in InAs/GaAs self-assembled quantum dots

D. H. Rich,[a] C. Zhang, I. Mukhametzhanov, and A. Madhukar
Department of Materials Science and Engineering, Photonic Materials and Devices Laboratory, University of Southern California, Los Angeles, California 90089-0241

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Cathodoluminescence wavelength imaging of InAs/GaAs self-assembled quantum dots (SAQDs) was performed to study the spatial variation in the spectral line shape of the broadened quantum dot (QD) ensemble. The line shape was found to vary on a scale of $\sim\mu$m, revealing attendant variations in the size distribution of SAQD clusters on this spatial scale. Energy variations in clusters of SAQDs are found to exhibit a spatial correlation with the efficiency of luminescence and the activation energy for thermal re-emission of carriers. A reduction in the energy variation of the QD clusters occurs when the thickness of the spacer layers in vertically self-organized samples is reduced or the number of stacks is increased. © *2000 American Institute of Physics.* [S0003-6951(00)01823-4]

Scanning probe microscopy studies of the InAs island formation reveal the stochastic nature of the island formation, as the average size, variation in size, and distance between neighboring islands vary with coverage.[1] The length scale on which the island size fluctuates is determined by the average interisland separation distance, which ranges from 50 to 300 nm, and depends on the initial InAs coverage. Such fluctuations are evident in spatially resolved experiments that measure the discrete δ-like luminescence spectra of multiexcitonic transitions in isolated quantum dots (QDs).[2]

In this letter, we report on the observation of a length scale on which the size distribution of SAQD clusters vary. We employ cathodoluminescence wavelength imaging (CLWI) to enable a spatial mapping of the peak energy of the emission from the ensemble of QDs. We show that subtle variations in the SAQD line shape for clusters on a scale of $\sim\mu$m can be detected with CLWI. We have examined the spatial correlation between the energy of interband transitions, CL luminescence efficiency, and the activation energy for thermal re-emission of carriers from the SAQDs on a μm scale. We further explore the relationship between the number of layers, interlayer spacing, and the magnitude of the QD peak energy variations in multilayered vertically self-organized (VSO) samples.

The SAQD samples were grown by molecular beam epitaxy (MBE) in which a total of 1.74 monolayers (ML) of InAs were deposited for each layer at a 500 °C substrate temperature and resulted in a lateral QD density of ~350 μm^{-2}. In the multilayered VSO samples, 20- and 36-ML-thick GaAs spacers were grown at 400 °C by migration enhanced epitaxy (MEE) after each InAs deposition. The samples were capped with 170 ML MEE-grown GaAs. The CL experiments were performed with a modified JEOL-840A scanning electron microscope (SEM).[3] CL spectroscopy was performed with an InGaAs linear array detector. CLWI is accomplished by acquiring a series of discrete monochromatic images, constructing a local spectrum at all 640×480 scan points within the image, and determining the wavelength, $\lambda_m(x,y)$, at which there is a peak in the CL spectrum at each scan point (x,y).

We illustrate these results first with a stack plot of CL spectra acquired from a *two-layer* InAs sample whose spacer thickness is 36 ML. The 21 CL spectra in Fig. 1 are acquired locally along an arbitrary scan line of 57 μm in length by

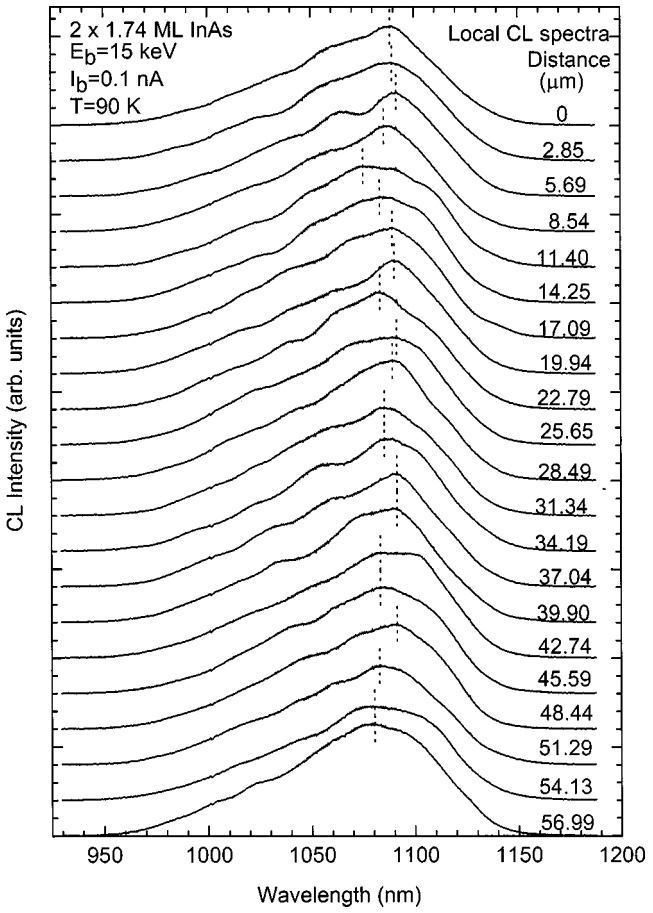

FIG. 1. Stack plot of CL spectra acquired locally for an electron beam movement along an arbitray line of the two-layer SAQD sample. The distance along this line and the peak position (vertical dashed lines) are indicated.

[a]Electronic mail: danrich@almaak.usc.edu

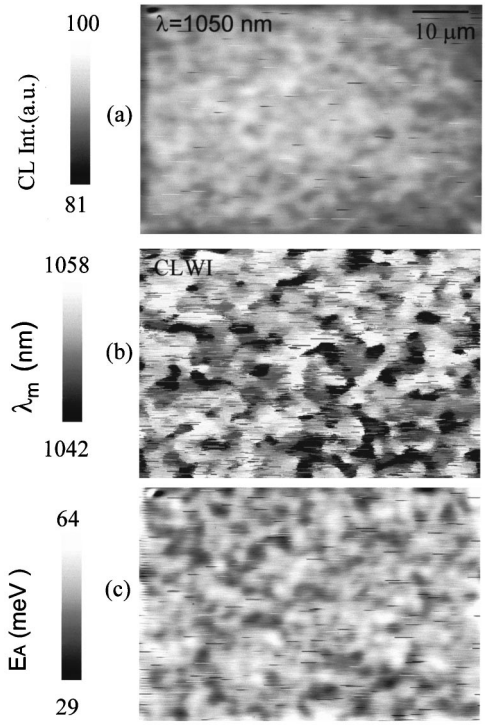

FIG. 2. Monochromatic CL image (a), CLWI image (b) and activation energy image (c) for the single layer SAQD sample.

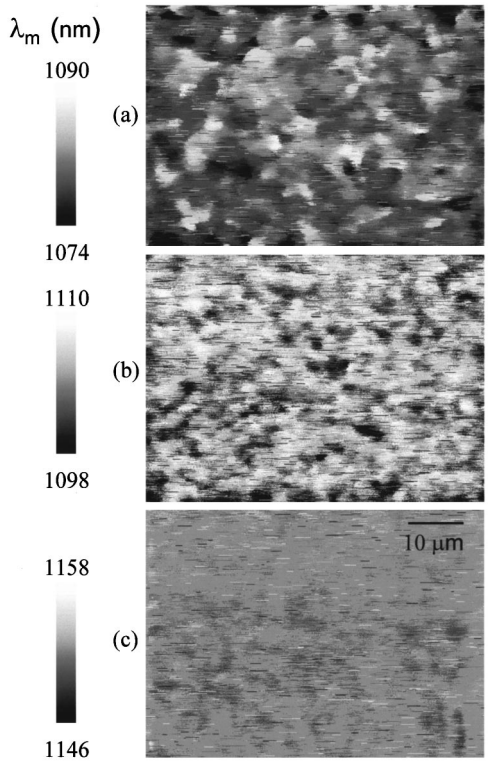

FIG. 3. CLWI images of the two layer with 36 ML spacer, five layer with 36 ML spacer, and five layer with 20 ML spacer samples in (a)–(c), respectively.

fixing the electron beam at each point, whose relative distance is indicated in the figure. The line shape of each spectrum is observed to change at each position. The center of gravity of the spectral line shape is roughly independent of the position. However, small changes in the line shape give rise to a change in the peak position, λ_m. The spectra further appear to be composed of multiple components whose relative intensities change from one location to another. In further analysis, a dashed vertical line is marked at the point in each spectrum which has a maximum in intensity. It is therefore evident that the peak wavelength position, λ_m, varies along this line, owing to changes in the relative intensity of all components that comprise the spectra. Similar subtle variations in lineshape are observed for the single-and five-layer samples.

Conventional monochromatic CL imaging reveals ~10% variation in the intensity of luminescence in the SAQD samples, with domain sizes on the order of a few microns, as shown in Fig. 2(a) for the single layer sample ($\lambda = 1050$ nm). We estimate a spatial resolution of ~ 0.5 μm under the present CL imaging conditions. In order to examine spatial variations in λ_m, we have performed CLWI for four InAs SAQD samples composed of a single layer, two layer with a 36 ML spacer, five layer with a 36 ML spacer, and five layer with a 20 ML spacer, as shown in Figs. 2 and 3. The samples were maintained at a temperature of 90 K. An electron beam of 15 keV and 1 nA was used. A false-color scale, with a color bar indicating the wavelength range, is shown for each image. Variations on a scale of $\sim \mu$m in the peak wavelength position are observed in which domains of constant color are clearly visible.

In order to better understand such variations and their associated correlation, we have examined spatial variations in the temperature dependence of the luminescence. This is accomplished by recognizing that a thermally activated process is responsible for the reduction in the luminescence efficiency of the SAQDs as the temperature is increased. Previously, we have measured an activation energy associated with an Arrenhius behavior for SAQD samples.[4] In this analysis the logarithm of the CL intensity versus $1/T$ yields a nearly straight line from which a constant thermal activation energy, E_A, is obtained from the slope. We have extended this analysis to an imaging technique by determining E_A for all 640×480 points in a CL image by acquiring discrete monochromatic CL images in the 180 to 250 K range, thereby enabling a spatial mapping of E_A. Along with the monochromatic image, and CLWI image, we show an *activation energy image* in Fig. 2(c). The simultaneous acquisition of these three images for a fixed region of the sample enables an evaluation of the degree of spatial correlation of the associated optical properties measured.

An evaluation of the degree of correlation is illustrated by plotting the CL intensity, λ_m, and E_A versus distance along an arbitrary line, as shown in Fig. 4. The line scan analysis reveals a clear correlation between a red-shift in λ_m, an increase in E_A, and an increase in CL intensity, where peaks and dips in each of these three scans coincide. This correlation, while not perfect, illustrates a strong coupling in the optical behavior to the structural properties of the QDs on a μm-scale, which is the length scale on which these variations occur.

The intensity variations reflect size and shape dependent variations in the carrier capture into the QDs, spatial variations in thermally induced re-emission of carriers, and spatial variations in the density of localized defects, which can act

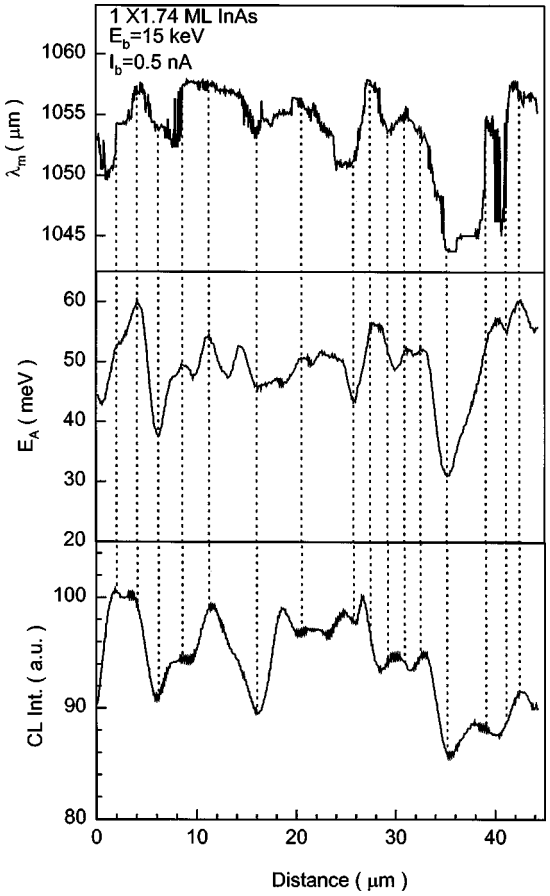

FIG. 4. Line scan analysis of the single-layer sample showing the peak wavelength (λ_m), activation energy (E_A), and CL intensity vs distance along an arbitrary line. The vertical dashed lines illustrate the spatial correlation in peaks and dips for these scans.

as nonradiative recombination sites. On a scale of a few microns the images reveal a tendency for InAs QDs to form constant energy clusters, within the typical cation migration length of $\sim\mu$m for In during MBE growth. The strong correlation between peaks and valleys in λ_m, E_A, and intensity can be understood by a simple model, which connects the size of the QDs with the expected rates of thermal re-emission. Larger QDs, yielding a relative red-shift in λ_m, also exhibit a larger confinement energy, owing to the larger energy separation between single particle electron and hole levels of the QDs and the single particle electron and hole levels of the wetting layer (WL). This in turn should lead to a larger E_A, which, in the limit of Boltzmann statistics, should represent the energy barrier for single particle excitation from confined QD levels to the WL levels. An E_A of $\sim$40–50 meV is consistent with the energy difference between the QD and WL electron and hole levels, as determined by theoretical calculations[5] and previous estimates based on thermal quenching of the QD luminescence.[4] Finally, a larger barrier for thermal re-emission would also lead to a reduced re-emission rate, a larger steady-state carrier population in these larger QDs, and thus lead to a larger CL intensity, which is the third aspect of the correlation.

The domain size for the correlations in λ_m, E_A, and intensity are on the scale of a few microns. For an InAs delivery of 1.74 ML, the density of the SAQDs, as determined by a previous atomic force microscopy (AFM) measurement, is $\sim$350 μm^{-2}.[1] We estimate that the blue- and red-shifted regions depicted in Figs. 2(b) and 3(a) represent regions containing $\sim$300–3000 QDs. The energy shifts represent a subtle change in the size and shape distribution of QDs, as reflected by the change in weight of individual components that comprise a CL spectrum as the *e*-beam is moved from one domain to another (i.e., as in Fig. 1). The size scale of the domains is identical to the expected In cation migration length of $\sim$1 μm, thus allowing for the likelihood of small mass transfers from one domain to another during growth. The stochastic nature of self-assembly may lead to fluctuations in the distributions of QD clusters on a $\sim$1–10 μm scale, owing to the coverage-dependent size distribution of SAQDs and the likelihood for surface inhomogeneities to generate local variations in In diffusion and coverage.

We finally observe that the growth of five-layer samples resulted in marked changes in the spatial variation of λ_m, as seen in Figs. 3(b) and 3(c) for the 36 and 20 ML spacers, respectively. The standard deviations in the wavelength fluctuation ($\delta\lambda_m$) for the CLWI images of the single, two- (36 ML spacer), five- (36 ML spacer), and five-layer (20 ML spacer) samples are 4.4, 3.2, 2.3, and 0.88 nm, respectively. The length scale of the fluctuations in λ_m and $\delta\lambda_m$ both decrease for the latter two VSO samples. This behavior is consistent with the improved uniformity that is expected to occur in the upper layers of VSO structures owing to the strain-driven In migration and subsequent formation of In islands above buried islands. A further decrease in $\delta\lambda_m$ in going from the sample with a 36 ML spacer to the sample with a 20 ML spacer is due to the larger strain fields in the 20 ML spacer which enhance the QD size uniformity and is also possibly due to a larger interlayer electronic coupling of QDs in samples with smaller spacer thicknesses. Photoluminescence of these structures shows a reduced linewidth relative to the single layer samples, consistent with the improved uniformity.[6]

In conclusion, we have observed a length scale of $\sim\mu$m on which energy and intensity fluctuations occur for SAQDs. Energy variations in clusters of SAQDs are found to further exhibit a spatial correlation with the efficiency of luminescence and the activation energy for thermal re-emission of carriers. Multilayered SAQD samples exhibit a reduction in the energy variations, owing to the stress-directed migration of In during growth.

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