Carrier transport properties in the vicinity of single self-assembled quantum dots determined by low-voltage cathodoluminescence imaging

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We propose a method to investigate the carrier transport properties in the ultrathin wetting layer of a self-assembled quantum dot (QD) structure using low-voltage cathodoluminescence (CL) imaging. Measurements are performed on diluted InAs/InP QDs in order to spatially resolve them on CL images at temperature ranging from 5 to 300 K. The mean ambipolar diffusion length extracted from CL intensity profiles across different isolated bright spots is about 300 nm at 300 K. This gives an ambipolar carrier mobility of about 110 cm²/V s. Temperature investigation reveals a maximum diffusion length near 120 K. © 2009 American Institute of Physics. [DOI: 10.1063/1.3072613]

With the development of solid-state optoelectronic devices based on single self-assembled quantum dots (QDs) such as nanolasers or single photon sources, evaluation of their individual properties is fundamental. As well as the study of single dot emission, investigation of carrier transport from their generation point in the barrier to their capture into one specific dot.

Figure 1 shows the schematic of the experiment. A low-voltage electron beam generates excess carriers in the barrier layer just below the surface of the QD structure. Most of them relax very rapidly into the WL in the picosecond range, then diffuse through it, and finally get captured into the QDs where they can recombine and emit light. Other carriers are either captured by surface defects near the beam impact or recombine in the barrier and WL. So, we can assume that carrier diffusion takes place mainly in the WL when the scanning electron beam is far from the dot. Furthermore, the density of electron-hole pairs generated in this confined region close to the surface is very high and much greater than the barrier majority carrier concentration. Thus, only ambipolar diffusion lengths of excess carriers in the WL can be determined. The diffusion length is deduced from CL intensity profile I(R) over the QD with R as the distance between the probe and the QD.

in comparison with the diffusion length in III-V bulk semiconductors. Under these conditions, if the CL intensity comes only from one QD’s emission, the limited resolution distance becomes the carrier diffusion length before capture into one specific dot.

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FIG. 1. (Color online) Schematic of the experiment. Excess carriers generated in sub-10-nm volume of the barrier are mainly captured by the WL and diffuse through it before their final capture into a QD. Diffusion length is deduced from CL intensity profile I(R) over the QD.
The investigated structure is made of InAs self-assembled QDs grown by solid-source molecular beam epitaxy on a semi-insulating InP(001) substrate. In order to spatially resolve QD luminescence, a specific growth procedure was implemented to produce diluted QDs, whose density is reduced to about 1 QD/μm². The resulting dots are truncated pyramids with a typical lateral size of 30–40 nm and height of 2–5 nm. The InAs WL below the QDs has a mean thickness of 2 ML. QDs are capped with a thin 20 nm InP layer to allow excess carriers generated near the surface to reach the WL region. CL measurements were performed in a Zeiss Supra 55 VP field emission scanning electron microscope equipped with a continuous helium flow cryostat providing temperature from 5 to 300 K. The luminescence was dispersed by a 30 cm grating monochromator and detected by a cooled InGaAs cathode photomultiplier tube within the 0.3–1.65 μm spectral range. CL micrographs were performed using a long pass cutoff filter at 1.1 μm and a 1 kV focused electron beam at several probe currents \( I_b \). At 1 kV, \( \phi_p \) is about 3 nm and according to Monte Carlo simulations electron-hole pairs are injected into sub-10-nm volumes in the InP barrier providing a quasiapoint source of carriers.

Figure 2 shows a CL image of such an array of InAs/InP QDs obtained at 1 kV, 300 K, and for \( I_b = 3 \) nA. The bright spots are correlated with the emission of individual dots. This has been confirmed by observing complementary features on WL and QD CL images of the same area at low temperatures and by comparing the density of bright spots with QD density estimated by AFM measurements on an uncapped sample. The measured spectrum of the same sample is presented (inset). We can clearly see the WL emission around 1 μm and the QD emission starting at 1.1 μm and extending up to 1.65 μm. The dashed area on the image represents the wavelength range removed using the optical filter.

Figure 3(a) shows the CL image over a specific region obtained at higher magnification and its related CL intensity profile for \( I_b = 3 \) nA. When the electron beam is scanned above one QD, the CL signal is nearly constant since carriers are captured efficiently into this dot. For an excitation spot away from the dot, the CL signal decreases since part of the excess carriers recombine in the WL. For ambipolar diffusion, the local density of electrons and holes is obtained by solving the continuity equation under a steady state regime. If we assume that dots act as perfect sinks, the diffusion current will be more pronounced toward the nearest dot direction. A one-dimensional diffusion equation can be used to obtain the approximate distribution of carriers from the generated point at \( r = 0 \) to the dot position at \( r = R \).

This is a well known diffusion problem with trivial limit conditions at \( r = 0 \), diffusion current equals generation rate, and at \( r = R \), carrier density in the WL is zero. The carrier diffusion current evaluated at the dot position is given by \( I(R) \sim \text{csch}(R/L_a) \), where \( L_a \) is the ambipolar diffusion length. This expression is valid for \( R \gg \phi_p, Re, \gamma, R_0 \), the QD radius. We assume that the QD CL signal is linearly proportional to this diffusion current at \( r = R \). The mean \( L_a \) value is then obtained through the fit of 15 CL intensity profiles across isolated bright spots taken randomly at different sample areas. Figure 3(b) presents mean carrier diffusion length values as a function of probe current and carrier generation rate at 300 K. For the lowest carrier injection at 3 kV and \( I_b = 0.42 \) nA, we found \( 300 \pm 50 \) nm, which is in accordance with another group value taken at low-level injection. It decreases down to 120 ± 30 nm and then slightly increases at higher injections. Enhanced nonradiative carrier recombination by Auger processes in the WL is suggested as an explanation for the reduced diffusion length observed at higher excitation current. The slight increase might be attributed to enhanced carrier diffusion into the InP barrier as the WL state filling becomes more important. One can note that the calculated standard deviation (\( \Delta L_a = 50 \) nm) is much larger than the mean deviation in the lateral size of our QD ensemble (about 15 nm). This high \( \Delta L_a \) value suggests that there exist local carrier transport inhomogeneities in the WL, most probably resulting from interface roughness, InAs, and alloy composition and strain induced QD size effects. Ambipolar diffusion length is related to the carrier mobility by this \( L_a = \sqrt{\mu q T e} \), where \( \tau \) is the carrier lifetime in a WL in the absence of QDs (recombina-

FIG. 2. CL intensity micrograph of a low QD density InAs/InP sample, acquired at 1 kV and 300 K using a long pass filter cutoff at 1.1 μm. The luminescence on the map corresponds to the QDs emitting between 1.1 and 1.65 μm as shown by the CL spectrum (inset).

FIG. 3. (Color online) (a) Zoom on a submicrometer bright spot associated with a single QD and its CL intensity profile at 300 K for \( I_b = 3 \) nA. The ambipolar diffusion length \( L_a \) is deduced from the \( \text{csch}(R/L_a) \) fitting curves (solid line). (b) \( L_a \) values as a function of beam intensity and carrier generation rate at 1 kV and 300 K. One measure is made at 3 kV and 0.42 nA as indicated. Error bars correspond to the calculated mean standard deviation.
We obtain an ambipolar mobility value of about 110 cm²/(V s) for the 300 nm InAs quantum well, which is comparable in thickness to our WL. If we consider this value for the 300 nm InAs quantum well to be nearly constant at low temperatures, we can estimate the diffusion length at 300 K using low-voltage CL imaging with submicron spatial resolution. This method provides a direct measurement of the carrier diffusion length in the WL. An ambipolar carrier mobility of about 110 cm²/ (V s) is estimated from the 300 K $L_a$ value. The $T$-dependence of the diffusion length is difficult to model but our results give valuable insights on possible scattering mechanisms in this InAs/InP system.

In conclusion, we investigated the transport properties of excess carriers in the WL of a diluted InAs QD structure using low-voltage CL imaging with submicron spatial resolution. This method provides a direct measurement of the carrier diffusion length in the WL. An ambipolar carrier mobility of about 110 cm²/ (V s) is estimated from the 300 K $L_a$ value. The $T$-dependence of the diffusion length is difficult to model but our results give valuable insights on possible scattering mechanisms in this InAs/InP system.

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