Stark shift in electroluminescence of individual InAs quantum dots

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We have fabricated light-emitting-diode heterostructure devices, in which a layer of InAs self-assembled quantum dots is embedded, with an active area of submicron size. In the electroluminescence spectra of these devices, we observed isolated narrow peaks due to emission from individual dots. From the shift of the peaks in an electric field (the quantum confined Stark effect), we show that the ground and excited states in the dots have different spatial alignments of the electron and hole. © *2000 American Institute of Physics.* [S0003-6951(00)02526-2]

There has been considerable interest in InAs/GaAs self-assembled quantum dots (SAQD) in recent years (see Ref. 1 for a comprehensive review). They provide an almost ideal system for investigating zero-dimensional excitons because the small size of the dots results in fully quantized, discrete energy spectra for electrons and holes, with large interlevel spacings compared with the Coulomb interaction energy of localized carriers. However, in a SAQD *ensemble*, there is a large inhomogeneous broadening of the energy spectrum due to fluctuations in dot parameters, ~50–100 meV, overwhelming any excitonic effects which are on a finer scale. Therefore, to study excitonic phenomena, it is necessary to resolve the signal from an *individual* dot.

Tunneling transport[2] and photoluminescence[3,4] (PL) studies have shown that single-dot spectroscopy is an excellent tool for local investigations of physical phenomena on a fine energy scale. In this letter, we report on optical studies of individual SAQD by means of electroluminescence (EL) as an alternative to PL. EL spectra from SAQD, with features attributed to individual dots, have been reported earlier.[5] However, there was considerable doubt as to whether any particular feature in the measured spectra could be identified with the emission from a single dot. In this letter, we show the conditions under which a sharp feature in the EL spectrum can be interpreted unambiguously as arising from a single dot. As the bias increases, we observe evidence for the occupation of excited states and also the shift in energy of the emission lines, the quantum-confined Stark effect.

To allow injection of carriers, a layer of SAQD was embedded in the intrinsic region of a *p-i-n* light-emitting-diode (LED) heterostructure. The band profile of the heterostructure under flatband conditions, which correspond to an external bias $V \approx 1.5$ V applied to the diode, is shown in Fig.

1(a). The sample was grown by molecular beam epitaxy on a (100) semi-insulating (SI) substrate. A layer of InAs SAQD, nominally 1.8 monolayers thick, was embedded in the center of a 90 Å wide GaAs quantum well with 30 Å thick AlAs confining barriers, sandwiched between two nominally undoped GaAs spacer layers, 200 and 300 Å thick, respectively. The intrinsic region is surrounded by graded *n*-type and *p*-type contact layers. The layers were grown at 550 °C, and there was a growth interrupt before the QDs were grown at 520 °C. The average dot diameter of 10 nm and height of 3 nm, as well as the areal density of 2×10^{11} cm^{-2}, were established by scanning tunneling microscopy and cross-sectional transmission electron microscopy.
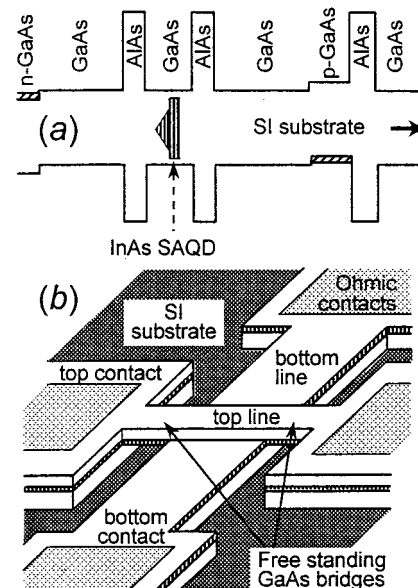

FIG. 1. (a) The band profile of the GaAs/AlAs light-emitting-diode heterostructure under flatband conditions; (b) Schematic diagram of the small-area device.

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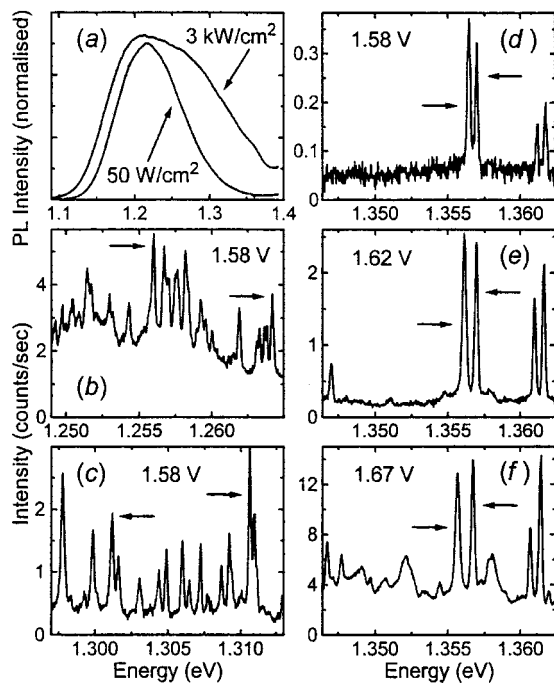
FIG. 2. (a) Normalized PL spectra from the dot ensemble in a large-area sample at low and high pumping intensities; (b)–(f) Representative EL spectra from a small-area device: (b)–(d) Spectra in various spectral regions at the same forward bias $V = 1.58$ V; (d)–(f) Spectra in the same spectral region at various biases. Arrows indicate the peaks for which the Stark shift is plotted in Fig. 3.

A crucial condition for observing EL from individual SAQD is to have a LED device with an active area of submicron size. This was achieved by fabricating cross-shaped devices,[6] shown schematically in Fig. 1(b), by optical lithography and wet etching, using a selective etch solution which preferentially removes GaAs. The AlAs layers, including the one between the p-type contact layer and the substrate, act as selective etch stops. The cross is made of two GaAs bars, one etched in the top and the other in the bottom contact layer, with the active device area of submicron dimensions defined by their intersection. Note that the top bar is undercut to form freestanding bridges.

The EL spectra at 4.2 K were dispersed by a high-resolution spectrometer and registered by a cooled charge coupled device array, typically with a resolution of $\approx 50 \, \mu$eV and an accumulation time of 3 min. PL spectra, excited by an Ar$^+$ laser and registered with a cooled Ge detector, were taken for comparison. Under low pumping intensity, the PL spectra from a large-area sample exhibit a broad line of approximately symmetric shape, with a maximum amplitude at ≈ 1.2 eV and a full-width half maximum (FWHM) linewidth of ≈ 100 meV, i.e., the emission from the dot ensemble, as shown in Fig. 2(a). With increasing pumping, a distinct higher-energy feature arises in the spectra, corresponding to emission from the excited states in the dots.

Figures 2(b)–2(d) show representative EL spectra from a small-area device at the same positive bias V in several spectral regions which correspond to the emission from ground (b) and excited (c)–(d) states. At all energies, we observe a set of sharp features which are reproducible for a given device and reflect the existence of a discrete spectrum of individual dot states. At low energies, which correspond to emission from dot ground states, see Fig. 2(b), the features are rather dense and it is impossible to resolve individual peaks: even a cursory examination of the spectrum shows that most features consist of at least two or three peaks. Moreover, the features emerge together with a steadily increasing background. Very probably, the background is related to the emission from the contact layers rather than from the quantum dots; however, the existence of the background EL is another reason why we *cannot* say that any observed sharp feature corresponds to the emission from a particular individual dot. In the presence of the background, *any* sharp feature may reflect statistical fluctuations in the density of states of the dot ensemble and involve several dots with similar emission energies. Similar EL spectra from SAQD exhibiting poorly resolved individual features have been reported earlier.[5]

At higher energies and at the same applied bias, Fig. 2(c), the density of the features is smaller, and they are better resolved, probably because the number of dots emitting in this range is smaller; the background is also much lower. Note that this energy range corresponds to emission from the excited states in the dots; therefore, at this particular bias the excited states are occupied either with electrons, or with holes, or both.

The appearance of the spectra is drastically different in the energy range of the tail of the emission from the excited states, as shown in Fig. 2(d). We clearly observe narrow (FWHM down to $\approx 100 \, \mu$eV) emission lines, with approximately 3–5 lines in a 10 meV energy range, which emerge from a *zero* background. Note that the observed FWHM is *not* limited by the spectrometer resolution. Figure 2(e) shows that with increasing bias, the line intensity grows up by an order of magnitude with only a small change in the background. Therefore, it is very improbable that more than one dot is involved in the emission; only in this case can we unambiguously attribute each particular line to the emission from a single dot.

A further increase in bias results in a further increase in the line intensity, and also in the appearance of additional features, as shown in Fig. 2(f). Some of the extra features are rather broad. They may either be composed of several broader unresolved dot transitions, or be due to transitions which include extended states in the wetting layer or in the GaAs matrix.

Increasing bias also causes a shift in the peak energy, the fingerprint of the quantum confined Stark effect. The positions of a few representative peaks as a function of bias are shown in Fig. 3. At lower energies, corresponding to emission from the ground states, the peaks all exhibit a near-linear Stark shift of similar magnitude, as shown in Fig. 3(a). The sign of the shift, which agrees with that recently observed by means of photocurrent (absorption-type) spectroscopy of SAQD,[7] indicates that the hole wave function is localized close to the top of the dot, above the wave function of the electron, an alignment opposite to that predicted by most theoretical models. (See Ref. 7 for a detailed discussion.)

The magnitude of the Stark shift gives an estimate of the spatial separation of the electron and hole wave functions
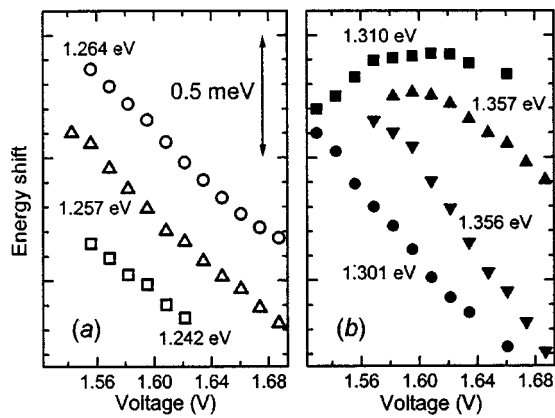
FIG. 3. Stark shift as a function of bias for several representative peaks in the spectral regions of ground- (a) and excited-state (b) transitions. Labels indicate approximate energy positions of the peaks at $V = 1.58$ V. Note that the flatband conditions correspond to $V \approx 1.5$ V.

$\Delta_{e-h} = (3.5 \pm 1)$ Å which is in excellent agreement with the value of (4 ± 1) Å found for Δ_{e-h} under flatband conditions in Ref. 7. Note that in our EL experiment, the *p-i-n* diode is under *forward* bias, and the electric field experienced by the dots is much smaller than that under the *reverse* bias applied to the *p-i-n* devices in the photocurrent experiments[7] (i.e., our device is always close to flatband). Consequently, the Stark shift remains linear in *V* in our experiments, in contrast to the quadratic shift reported in Ref. 7.

The Stark shift is less straightforward for the excited-state transitions. This can be easily seen for the lines shown in Figs. 2(d)–2(f) which clearly diverge with increasing bias. The positions of these lines are shown by up and down triangles in Fig. 3(b). Moreover, a reversal of the sign of the shift is observed for some lines; an example of this behavior is shown in Fig. 3(b) by solid squares. The bias at which the differential Stark shift disappears corresponds to the symmetric alignment of the electron and hole wave functions for the states involved in this particular transition; therefore, under flatband conditions, the alignment is opposite to that observed for the majority of transitions. Unfortunately, because we have a few tens of active dots in our sample, we cannot say for certain that the electron–hole alignment is different for ground- and excited-state transitions for the *same* dot. Nonetheless, we believe it is very probable because the ground state emission lines always show a linear Stark shift, and the evidence for the opposite alignment is restricted to some of the lines in the region of excited states.

The problem of the electron–hole alignment in the growth direction in the SAQD is rather complex because of the effect of lateral confinement. Note that the excited states in the SAQD arise due to the *lateral* confinement,[7] while the observed alignments result from the change in the wave functions in the *growth* direction. Therefore, our results show that for the SAQD, the spatial variables in the Hamiltonian are not separable; to account for our observations, detailed calculations are required, taking into account the precise shape and composition of the dots.

To conclude, we have successfully fabricated submicron-size, light-emitting diodes with an embedded layer of InAs self-assembled quantum dots. We have clearly observed the emergence of well-resolved, narrow emission lines in the electroluminescence spectra due to individual dots. The behavior of the observed quantum confined Stark effect indicates that the relative alignment of electron and hole wave functions in the dots may be different for ground and excited states.

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