Random telegraph noise in photoluminescence from individual self-assembled quantum dots

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We have performed photoluminescence microscopy studies of spatially resolved quantum dots grown by the Stranski-Krastanow technique. A small fraction of the dots (one in about a thousand) has been found to exhibit an intermittence, with their emission intensity switching between discrete levels on a time scale of seconds. The statistics of the switching corresponds to a random telegraph signal and the frequency increases superlinearly with excitation power density. The intermittence can be irreversibly suppressed by strong illumination. Emission spectra of these dots show significant differences between the on-state and the off-state. The observed switching behavior is attributed to mobile photoactivated nonradiative recombination centers.
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Random switching between discrete levels is known as random telegraph noise. Such switching may affect observable properties in many different ways and have different origins.[1–3] This has been observed in various systems such as in resonant tunneling diodes,[4] metal-oxide-semiconductor structures,[5] metallic nanoconstrictions,[5] tunnel barriers,[6] single atoms,[7] and in single molecules.[8] In solids, random telegraph noise has most often been observed via electrical effects and has usually been attributed to defects.[3] In atoms and molecules, switching has been detected via light emission.[7,8] Recently the switching of the luminescence intensity of nanocrystallite quantum dots has also been reported[9,10] and it has been attributed to an intrinsic ionization-neutralization process.[9,11] These quantum dots were fabricated by chemical means and supported by, but not coherently bonded to, a substrate. We describe here the phenomenology of random switching of luminescence in self-assembled quantum dots, coherently embedded in a semiconductor. This is another of the main types of nanostructures with possible applications in quantum dot-based devices.[12] In this case the experimental data are consistent with a modulation of the radiative efficiency of the dot originated by switching defects.

The samples were grown by metal-organic vapor phase epitaxy, and the quantum dots were formed by the Stranski-Krastanow growth mode.[13] The quantum dot material is InP and the barrier material is GaInP, which is lattice matched to the GaAs substrate. The GaInP layers are typically *n*-type with a carrier concentration of $2\times10^{16}\,\mathrm{cm}^{-3}$. The InP thickness was adjusted to be slightly above the critical thickness of quantum dot formation (1.5 monolayer). The quantum dots are shaped like truncated pyramids on a hexagonal base with a typical height of 15 nm.[14] The results that we will report here correspond to samples having a GaInP capping layer of 300 nm, unless otherwise indicated. The quantum dots are thus fully strained.[15] The lateral quantum confinement energies in these dots are about 10 meV for electrons and 1 meV for holes and the exciton lifetime is about 1 ns.[16] A detailed optical study of individual quantum dots has been reported in Ref. 16 and a theoretical study in Ref. 17.

The quantum dots were excited with the 488 nm line of an Ar^+ laser or the 532 nm line of a frequency-doubled YAG laser, focused with conventional optics. The excitation power densities were typically 1–10 $\mathrm{W/cm}^2$. The photogenerated carriers were generated mainly in the barrier material then captured by the dots, where they eventually recombined. The luminescence from the sample was collected through an optical microscope. For imaging, the wavelength was selected using either a long-wavelength–pass filter or a tunable interference filter with a band-pass of 10 nm. A CCD camera was used for detection. The spatial resolution of the system was about 1.4 μm and the emission from individual quantum dots was easily resolved, due to the low density of quantum dots (10^6–$10^7\,\mathrm{cm}^{-2}$) in our samples. The temporal resolution in our experiments was given by the integration time of the images. The shortest integration time that we could use, keeping an acceptable signal-noise ratio, was 8 ms in the best case, but typically 120 ms. In order to obtain spectrally resolved movies we used a low-resolution monochromator in combination with a video camera. The emission spectra of the quantum dots (within the above mentioned power density range) corresponded to no state-filling conditions.[16] The measurement temperature was typically 7 K.

Figure 1(a) shows an image of the emission from the quantum dots. The image has been obtained using a detection energy of 1.66 eV, corresponding to the quantum dot emission. Well separated quantum dots are clearly observed. Although most of the dots continuously emit light, we found that there are some exceptions. Figures 1(b) and 1(c) show magnified images of the marked area of Fig. 1(a) taken at two different times. In one case one of the quantum dots is weakly emitting (''off'' state) while in the other it is emitting with an intensity similar to that of the other dots in the image (''on'' state). A trace of the emission intensity of this quantum dot is displayed in Fig. 2. It can be seen that the intensity switches between two discrete levels on a time scale of seconds.

In order to determine if the switching is random, it is necessary to analyze the statistics of the plateau durations in the intensity traces. If that is the case, the probability density (p) of the occurrence of an on-plateau with duration t should obey[3]

$$p(t)=\tau^{-1}\exp(-t/\tau), \tag{1}$$

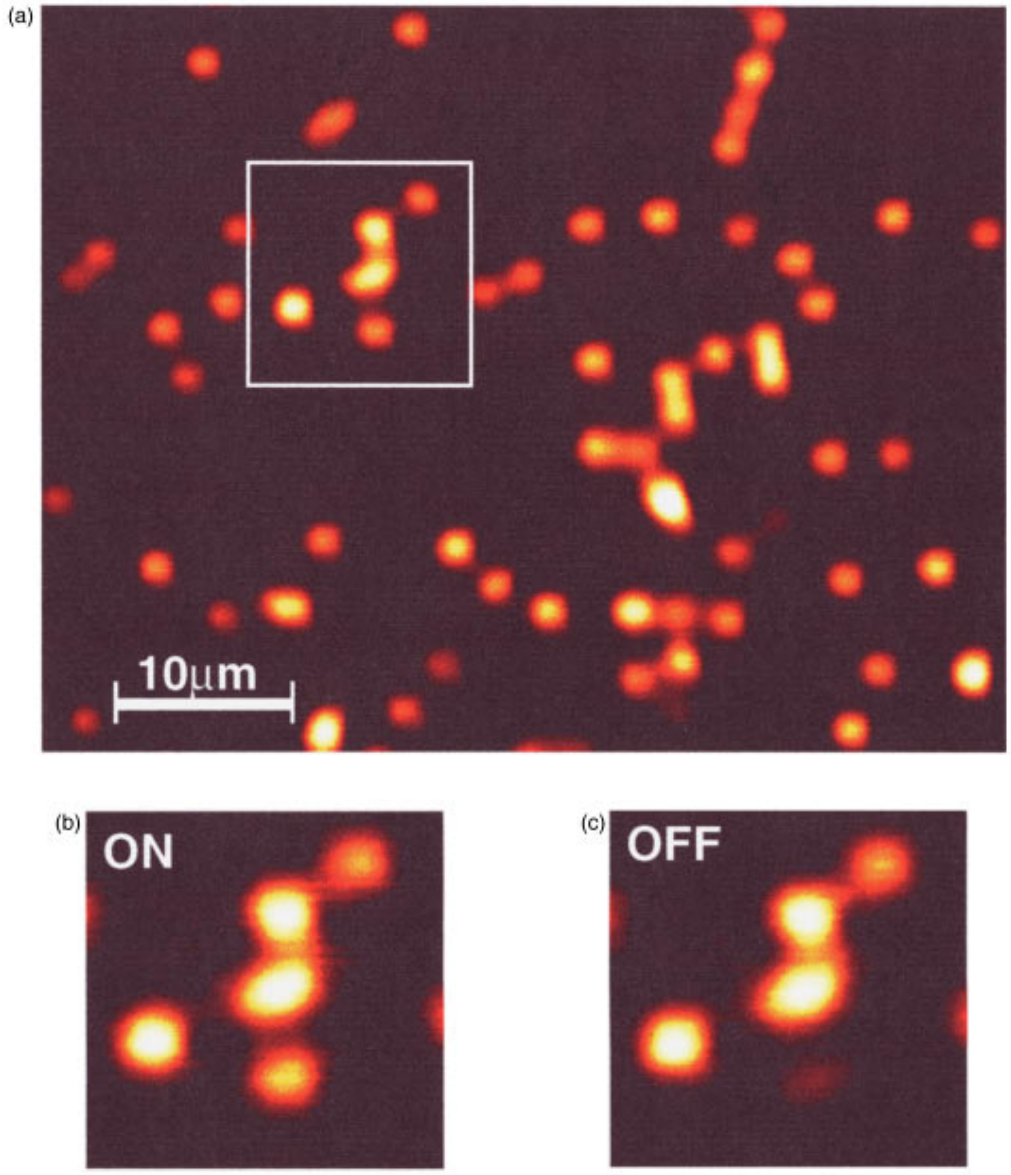

FIG. 1. (Color) (a) An image of the emission from a sample containing quantum dots. The photon energy monitored was 1.66 eV, corresponding to the quantum dot emission. Magnified images of the marked area recorded at two different times are also shown [(b) and (c)]. In one of them the emission from one of the quantum dots is absent (OFF) while in the other it is emitting (ON).

where τ is a characteristic time length for the dot in the on-state. The same behavior should be obeyed for the off-plateaus, but with a different τ. Histograms of the number of on- and off-plateaus as a function of the duration are shown in Fig. 2, along with exponential fits to the data, for the case of the switching dot of Fig. 1. As can be seen, the data are consistent with the random switching described by Eq. (1) and we thus conclude that we see a random telegraph signal in the emission intensity of this quantum dot.

More than 100 000 dots have been inspected (in samples with a capping layer) and about 100 of them have been found to exhibit switching. It has to be kept in mind, though, that intermittences having time constants below 40 ms or above 20 s would typically not be detected. We have observed switching dots in samples grown on both *n*-type and *p*-type substrates (with the Fermi level differing about 0.2 eV in the vicinity of the dots). Samples without a capping layer have also been found to contain switching dots, and in a proportion higher than in capped samples. Although most switching dots switch between two levels, there are exceptions. Some dots were found to switch between three levels and we have also observed a dot switching between four levels. We concentrate here on the most common type of switching dots having two levels. It should be mentioned that most of the switching dots retain their characteristics even after repeated cycling between room temperature and low temperatures.

Figure 3(a) shows traces of a switching quantum dot at different excitation power densities. The characteristic switching rates (τ_{on}^{-1} and τ_{off}^{-1}) are displayed in Fig. 3(b) as a function of the excitation power density. At sufficiently low power the switching times are of the order of minutes and an extrapolation of the data taken at low power shows that at zero excitation power there is no switching. This suggests that the switching is actually induced (rather than being monitored) by the excitation light. The emission intensity in the on-state scales linearly with excitation power density,

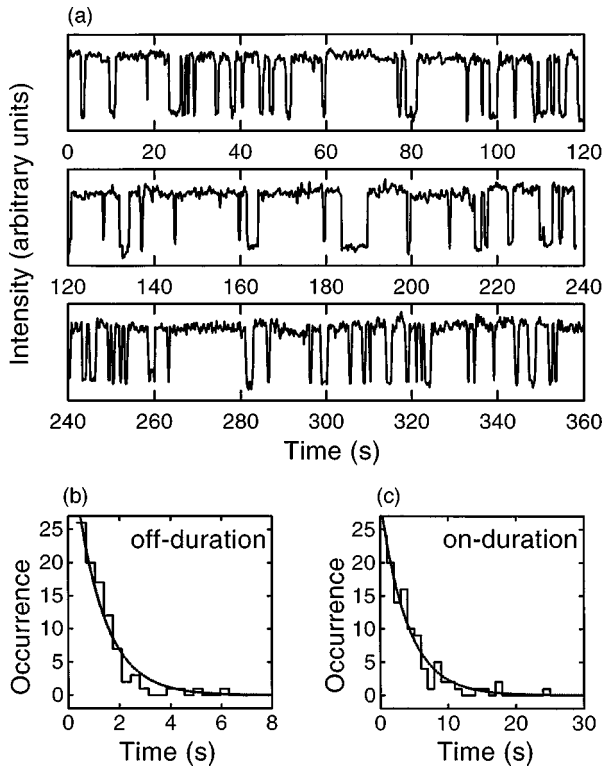

FIG. 2. (a) A trace of the emission intensity of the switching quantum dot of Fig. 1 as a function of time. The emission intensity switches between two discrete levels. (b) and (c) The histograms of the occurrence of plateaus of time length t (both off and on) as a function of t are plotted. Theoretical fits assuming random switching are included.

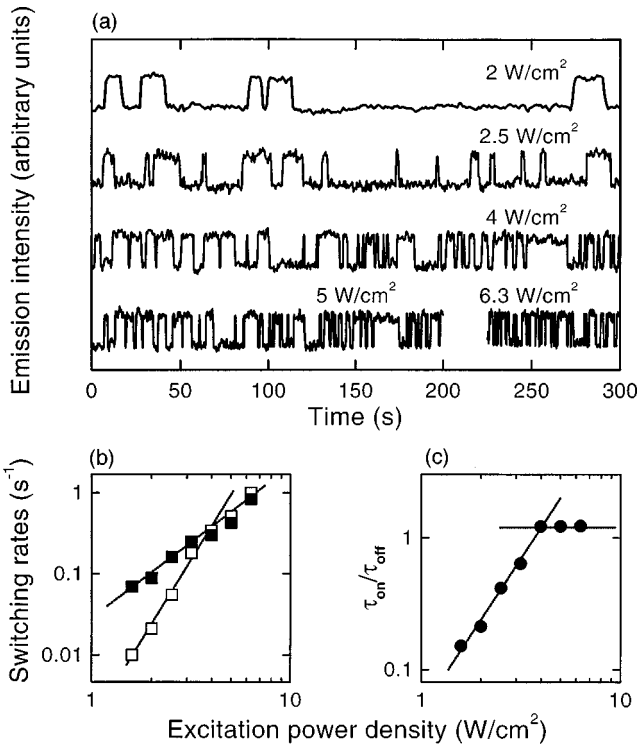

FIG. 3. (a) Examples of emission traces of a switching quantum dot for different excitation power densities. The emission intensities were normalized by the excitation power density. The characteristic switching rates of the on- and off-states (τ_{on}^{-1} and τ_{off}^{-1}, represented by full and open symbols, respectively) are plotted in (b) as a function of the excitation power density P. The ratio between the switching times (τ_{on}/τ_{off}) is displayed in (c). As a reference, the slopes corresponding to P^4 and P^2 and to P^2 and P^0 are displayed in (b) and (c), respectively.

indicating that the number of carriers captured by the quantum dot is linear with excitation power density (in this power range). In contrast, the switching rates are strongly superlinear with excitation power density [see Fig. 3(b)], suggesting that the switching process involves multiple particles. In Fig. 3(c) we have plotted the ratio of the mean time-on with the mean time-off (τ_{on}/τ_{off}). The ratio increases with excitation power density, this behavior being opposite to the photodarkening observed in nanocrystallite quantum dots.[9] At sufficiently high excitation power density it saturates.

At even higher excitation power densities the quantum dots irreversibly stop switching and stay permanently in the on-state, even if the excitation power is again reduced. The required excitation power is of the order of 50 W/cm^2, which is high enough to clearly observe state-filling features in the emission spectrum of the quantum dots.[16] After a dot stops switching we have found no reliable way to turn it back to a switching situation. These effects from the excitation power density have been found for all switching dots. Similar effects (although in this case involving a final off-state) have been observed in single molecules.[8]

If we increase the temperature in the range of 4–30 K we find that the switching rate increases [Fig. 4(a)]. The emission intensity has carefully been kept constant in these experiments in order to avoid effects of the excitation power density (Fig. 3). It is not sufficient to keep the excitation power density constant, since we have found that the capture into the dots is quite temperature dependent. It is possible to derive an activation energy of switching of about 10 meV from these experiments. Such an activation energy is, however, quite ambiguous since the dots become unstable at higher temperatures (30–60 K). An example of a dot which goes from a switching behavior to a nonswitching behavior (but not irreversible) is shown in Fig. 4(b) at a temperature of 57 K. Such instabilities are in fact very common at high temperatures, and we also have examples of dots going from nonswitching to switching behavior as well as dots suddenly changing their rate of switching (data not shown). What is common for all investigated dots is that the switching rate increases with temperature, before such instabilities occur.

In Fig. 5 we show emission spectra of a switching dot, both in the off-state and the on-state, as a function of excitation power density. Previous investigations of quantum dots of this type (InP in GaInP) have shown that the emission spectrum always contains multiple lines, also at very low excitation power density.[16] The switching dot (Fig. 5) has an emission spectrum which is similar to nonswitching dots. The emission energy is the same and also here multiple emission lines are present. A comparison of the energies of the different lines in the off- and the on-state shows no energy shift. There are clear signs of state-filling in the spectra for the off-state. Also for the on-state we see some transfer to the lowest energy peaks at the lowest excitation power density.

We performed an additional experiment in which the carriers were generated directly in the dot. To do that, we used a Ti:sapphire laser with a photon energy of 1.72 eV, that is

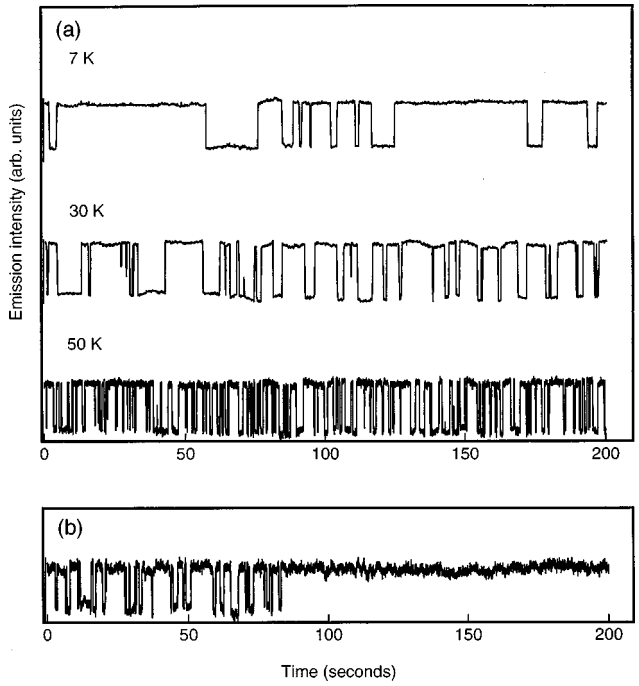

FIG. 4. (a) Traces of a switching quantum dot at different temperatures illustrating the increase of switching frequency with temperature. (b) Example of a dot which goes from a switching state to a nonswitching state at high temperature (57 K).

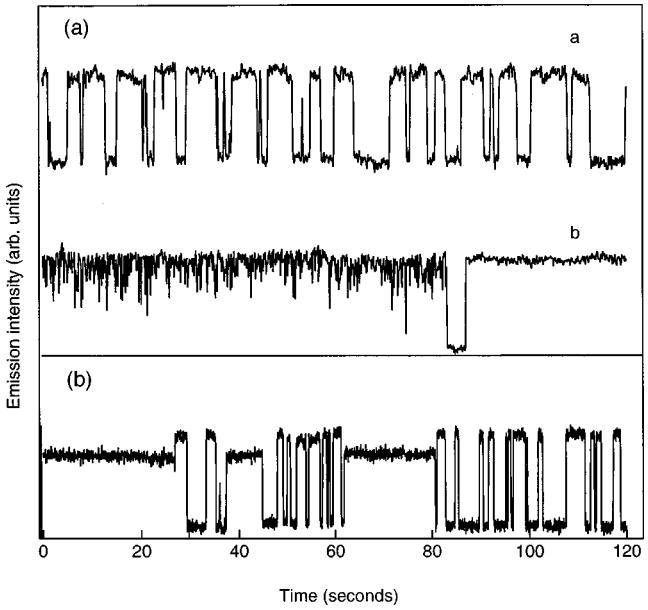

FIG. 6. (a) Upper trace corresponds to a normal switching dot at a temperature of 7 K. After increasing the excitation power density about 10% (which quenched the switching) the dot became noisy. This is shown in the lower trace which was taken a few minutes after the upper one. After a brief time in the off-state the dot returned to the on-state without excess noise. (b) Trace of a dot switching between three levels.

lower than the absorption edges of the barrier material and of the wetting layer. In these conditions, in which no capture process was involved, the switching behavior was still observed. Thus, we can conclude that the effect is related to a modulation of the radiative recombination efficiency, rather than to a modulation of the carrier capture.

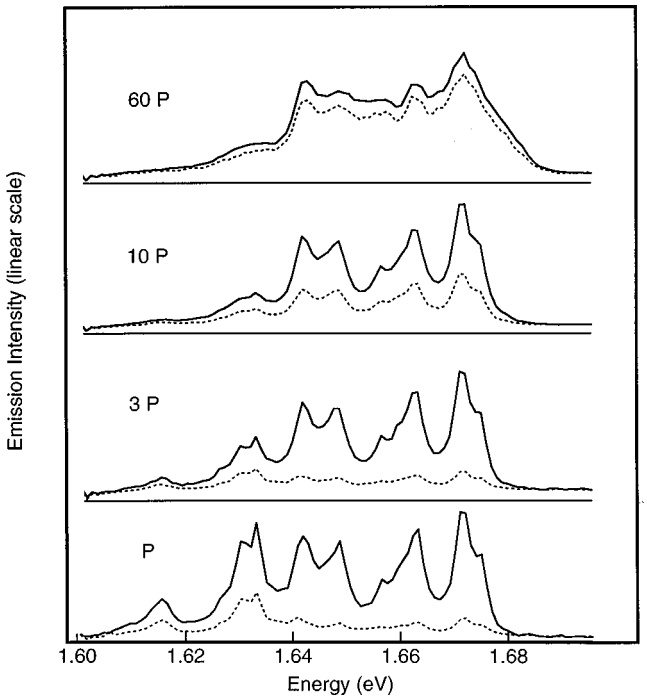

FIG. 5. Spectra of a switching quantum dot for different excitation power density. Full lines correspond to the dot in the on-state and dashed lines correspond to the dot in the off-state. The spectral resolution was 1 meV. *P* corresponds to an excitation power density of about 0.5 W/cm^2.

In our case, the modulation of the radiative efficiency causing the switching cannot be related to an effect intrinsic to the dots, since most dots do not switch. We attribute the switching to a native defect in the neighborhood of the dot. The defect would have two metastable configurations corresponding to the off-state and to the on-state, respectively, where in the off-state configuration the defect acts as a nonradiative center. The observed occurrence of switching dots would correspond to a defect density of the order of $10^{15}\,\mathrm{cm}^{-3}$. The transition between the two configurations is photoactivated (Fig. 3) and the defect is also mobile (since the switching can be irreversibly turned off by intense illumination). Such photoactivated, mobile, and nonradiative defects are by no means without precedent and have been individually observed using photoluminescence topography.[18,19] They are indeed a common feature in photodegradation of semiconductor heterostructure lasers.[20] Several models for a photoactivated mobility have been proposed, one of which is the phonon-kick model.[21] In this model the defect is activated via phonons generated by carrier recombination. In our case the phonons would more likely be generated by the relaxation of carriers in the dot. The superlinear intensity dependence of switching (Fig. 3) is then a sign of a multiphonon process of activation.[21] Increasing the temperature also gives a supply of phonons which will enhance the transition between the two configurations of the defect (Fig. 4). We do not see any Stark shift between the emission spectra in the on-state and in the off-state (Fig. 5). Thus, most likely, the transition between the two configurations does not involve a change in the charge state. For increasing excitation power the defect-related nonradiative re-

combination would saturate and the emission intensity in the off-state would become rather similar to that of the on-state (Fig. 5). Upon sufficiently strong illumination the defect moves far away from the dot, and has no effect on the emission.

Based on this model, we designed an experiment where we gently quenched the switching (that is, we increased the excitation power density only about 10% over the threshold for quenching to the on-state for a few seconds). When we subsequently observed the emission intensity of the dot, we found that initially the emission intensity was much more noisy than for nonswitching dots [Fig. 6(a)]. During the measurement the dot returned once to the off-state and then settled down permanently in an on-state (without excess noise). Although we have no explanation for the return to the off-state, we conclude from this experiment that the responsible defect moved only a small distance away from the dot, and that it still was affecting the emission from the dot (seen as noise) for some time. This resembles the noise behavior observed in electrical nanodevices when a defect diffuses near the active region.[5]

The model suggested here is quite crude, but it does give a framework for discussion and it gives guidelines for future experiments. The fact that there are switching dots having several levels [Fig. 6(b)] indicates that a unified model is going to be very complicated and, most likely, several types of defects are involved.

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