Submicrosecond correlations in photoluminescence from InAs quantum dots

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Photon correlation measurements reveal memory effects in the optical emission of single InAs quantum dots with time scales from 10 to 800 ns. With above-band optical excitation, a long-time scale negative correlation (antibunching) is observed, while with quasi-resonant excitation, a positive correlation (blinking) is observed. A simple model based on long-lived charged states is presented that approximately explains the observed behavior, providing insight into the excitation process. Such memory effects can limit the internal efficiency of light emitters based on single quantum dots, and could also be problematic for proposed quantum-computation schemes.

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I. INTRODUCTION

A variety of memory effects have been reported in the optical emission of single semiconductor quantum dots. Many of these effects occur on millisecond time scales, including blinking, two-color blinking, and spectral diffusion. Evidence suggests that millisecond blinking, seen only in a small minority of quantum dots, is caused by nearby defects. However, we have previously reported a much faster type of blinking that occurs in a majority of quantum dots subject to resonant optical excitation. This blinking behavior appears as a positive correlation in two-photon coincidence measurements. Blinking has also been observed in diamond color centers, semiconductor nanocrystals, and in molecules. In our case, the correlation time scale depends on the laser excitation power, but can vary from less than 10 ns to at least 800 ns.

This paper presents a more detailed study of these fast memory effects. In addition to presenting data on the blinking of quantum dots under resonant excitation, we report negative photon correlations with time scales greater than 100 ns for quantum dots under pulsed excitation with the laser tuned above the band gap of the host semiconductor. From a physical viewpoint, studying these complimentary memory effects can provide information about the states of a quantum dot, the transitions between them, and especially the nature of the optical excitation process. From a practical viewpoint, blinking effects reduce the efficiency of quantum-optical devices based on single dots, and also might be problematic for proposed quantum-computation schemes that involve optical control.

Both negative and positive correlations over long time scales can be explained reasonably well through a simplified model presented later in this paper. Memory effects imply multiple long-lived configurations of the quantum dot, and our analysis suggests that these are likely states with differing total charge. As explained below, negative correlations occur because above-band excitation injects electrons and holes into the dot separately, whereas positive correlation (blinking) occurs because resonant excitation injects electrons and holes together, in pairs.

II. SAMPLES

Sample A, the principal sample used in this study, has been described in Refs. 16 and 18. It contains self-assembled InAs quantum dots (about 25 μm²) embedded in the middle of a GaAs spacer layer, and sandwiched between GaAs/AlAs distributed-Bragg-reflector mirrors, grown by molecular-beam epitaxy. The quantum dots were grown at a relatively high temperature, which leads to intermixing between the InAs and surrounding GaAs, shortening the emission wavelength to ≈900–950 nm. Pillars [Fig. 1(a)] with diameters ranging from 0.3 μm to 5 μm and heights of 5 μm were fabricated in a random distribution by chemically assisted ion-beam etching (CAIBE) using sapphire dust particles as etch masks. The resulting microcavities, exhibiting three-dimensional photon confinement, have quality factors of ≈1000 and spontaneous-emission rate enhancement (Purcell) factors as high as 5. The purpose of the optical microcavity was to enhance the photon collection efficiency and to decrease the spontaneous-emission lifetime through the Purcell effect. For this study, the enhanced collection efficiency was valuable, since the data collection rate in photon correlation measurements is proportional to the square of the efficiency.

Sample B, described in Ref. 8, provided additional data for Figs. 5 and 7 below. This sample contained quantum dots (about 11 μm²) embedded in simple mesa structures (0.2–0.4 μm diameter) without optical cavities. The emission wavelengths of these dots were shorter (860–900 nm).
III. EXPERIMENT

The main features of the experimental setups used for acquiring photon correlation data are shown in Fig. 1. This type of setup, known as a Hanbury Brown and Twiss (HBT) setup, has become a common tool for studying the dynamics of single-quantum systems, including quantum dots. This measurement technique can be used to characterize the performance of single-photon devices and is more generally useful in studying spectral emission lines and determining how they are connected. For the measurements presented below, several setups were used, but in all cases the samples were held in a cryostat at temperatures ranging from 4–15 K and excited from a steep angle (about 54°) by 3 ps pulses every \( T_{\text{rep}} = 13 \) ns from a mode-locked Ti:sapphire laser. The emission was collected by a lens (numerical aperture = 0.5) and imaged onto a pinhole to define a collection region, \(~= 6 \mu m\) wide, on the sample. A single linear polarization was selected by a half-wave plate followed by a fixed polarizer. The emission was then spectrally filtered using a diffraction grating in a monochromator-type configuration, providing spectral resolutions from about 0.1 nm to 0.35 nm for the various setups. This filter allows one to collect from a single spectral line of the quantum-dot emission.

The HBT portion of the setup consists of a beam-splitter with each output leading to a photon counter. The photon counters were EG&G SPCM avalanche photodiodes having about 200 s\(^{-1}\) dark counts. The timing resolution varied from about 400 ps to 1.3 ns, depending on how narrowly the light was focused onto the detector active areas. Coincidence electronics, consisting of a time-to-amplitude converter followed by a multichannel analyzer computer card, generated a histogram of the relative delay \( \tau = t_2 - t_1 \) between photon detections at the two counters (\( i = 1, 2 \)) at times \( t_i \).

The two types of memory effects we have observed appear in the photon correlation measurements shown in Fig. 2. The peaks at \( \tau = nT_{\text{rep}} \) correspond to events for which one photon was detected from some pulse \( m \), and a second photon was detected from pulse \( m + n \). The area of the central peak at \( \tau = 0 \) gives information about photon number statistics within a single pulse. The side peaks at \( \tau \neq 0 \) give information on how the emission from different pulses is correlated. One can also interpret the data in terms of conditional probability: for \( n > 0 \), the area of peak \( n \) is, to an excellent approximation, proportional to the probability of detecting a photon from pulse \( m + n \) given that a photon was previously detected from pulse \( m \). If the configuration of the quantum dot is eventually completely randomized, then the peak areas at \( n \to \infty \) are proportional to the \textit{a priori} probability of detecting a photon (the probability of detecting a photon without information about prior events).

Both histograms were obtained from the same quantum dot on sample A, dot 1. The emission was collected from a bright spectral line at about 932 nm, shown in Fig. 3. In Fig. 2(a), the excitation laser was tuned above the band gap of the GaAs material surrounding the quantum dot, and the excitation intensity was chosen so that the collected emission intensity was far below its maximum value. The decrease of the side peaks near \( \tau = 0 \) indicates a long-term (27 ns) anticorrelation between photons in consecutive pulses. This effect indicates that the quantum dot is unlikely to emit a photon if it has emitted another photon in the previous pulse. In Fig. 2(b), the excitation laser was resonant with an excited level of the quantum dot at 904 nm. Such resonances are found through a photoluminescence excitation (PLE) measurement. The photoluminescence intensity is monitored as a function of laser wavelength, and typically two or three peaks are found. The rise of the side peaks in the photon correlation histogram near \( \tau = 0 \) is opposite from the behavior in (a), and indicates a blinking of the quantum dot at the particular wavelength being collected. By “blinking” we mean that once the quantum dot has emitted one photon, its probability of emitting a photon in the next pulse increases,
compared with the \emph{a priori} case. Alternatively, if blinking is
defined as low-frequency noise in the emission intensity, this
noise can be inferred from Fig. 2(b) through the Fourier-
transform relation between the intensity autocorrelation and
the power spectral density. Such blinking behavior suggests
that the quantum dot switches between a configuration that
can emit light at the wavelength of our spectral filter, and one
or more other configurations that cannot emit at this wave-
length (but could possibly emit at other wavelengths).

The fact that emission from the same quantum dot can
have either positive or negative correlations over large time
scales, depending on the laser wavelength, implies that there
is an important difference in how carriers are injected into
the quantum dot for above-band and resonant excitation.

Figures 3(a) and 3(b) show photoluminescence spectra
from dot 1. The measurements in Fig. 2 were performed on
the bright emission line at 932 nm. In (a), the excitation was
at 750 nm, above the GaAs band gap, and many emission
lines appear. Some of these are probably from the same
quantum dot (trion and biexciton lines, for example) while
others could be from other quantum dots. In (b), resonant
excitation at 909 nm was used, and most of the other peaks
have disappeared, demonstrating the selective nature of reso-
nant excitation. Figure 3(c) shows data obtained by sending
light from the main emission line through a Michelson inter-
ferometer. This setup is described in Ref. 16 and is similar to
the one in Ref. 32. This measurement, performed without a
polarizer, reveals fine structure though an oscillation or
“beating” in the interference fringe contrast as the path
length is varied, indicating that the emission line is actually a
doublet with a 13 \( \mu \text{eV} \) splitting. Further measurements have
shown that the two components have orthogonal linear po-
rarizations, as in Ref. 33. The existence of such a splitting
suggests that this line originates from a neutral-exciton tran-
sition rather than a charged-exciton (trion) transition, which
would be polarization degenerate in the absence of a mag-
netic field.34 For the measurements in Fig. 2, just one com-
ponent of the doublet was used, selected through polariza-

tion.

Several photon correlation measurements taken at dif-

dent excitation powers are shown in Fig. 4. In this figure, only
the normalized areas of the peaks from the measured histo-
grams are shown, plotted versus peak number, with peak 0 at
\( \tau=0 \). For an ideal \( g^{(2)} \) measurement, the normalized peak
areas can be written as

\[
g^{(2)}[n] = \frac{\langle n_1[m]n_2[m+n] \rangle}{\langle n_1[m] \rangle \langle n_2[m] \rangle}, \tag{1}
\]

where \( n_i[m] \) is the number of photons measured on detector
\( i \) from pulse \( m \). The light source is assumed to be stationary:
a shift in \( m \) does not change the expectation values in the
numerator or denominator. The histogram peak areas ob-
tained from our setup are approximately proportional to the
numerator as long as the mean count rates are small com-
pared with the inverses of the relevant dead times (\( \approx 50 \text{ ns} \)
for the photon counters, \( \approx 1 \mu \text{s} \) for the electronics). The
effects of these dead times can also be corrected through cali-

Fig. 4. Normalized peak areas from photon correlation histo-
grams plotted vs peak number (circles), and two-sided exponential
fits (lines) using Eq. (2). (a) Results for dot 1. The lower plots
(empty circles) are from data obtained under above-band excitation,
measured at four different excitation powers, resulting in the indi-
cated count rates on one detector. The data sets are shifted vertically
for clarity. The upper plot (filled circles) is from data obtained un-
der resonant excitation at 908.6 nm. (b) Results for dot 2 under
resonant excitation at 905 nm, measured at four different excitation
powers, as indicated.

bination relative to scattered laser light. The denominator can
be calculated from the mean count rates on the detectors.

For the quantum dots we have studied, when absolute
normalization using measured count rates was performed,
the peak areas \( g^{(2)}[n] \) converged to 1 as \( n \to \infty \), and were
well fit by a simple two-sided exponential function:

\[
g^{(2)}[n \neq 0] = 1 + g_1 \exp[-(|n| - 1) \text{rep} / \tau_b], \tag{2}
\]

where \( g_1 \) and \( \tau_b \) are fitting parameters that characterize the
amplitude and time scale of the memory effect, respectively.
This shows that there were no additional blinking effects
occurring with time scales greater than \( \tau_b \), up to \( \approx 100 \text{ s} \).
The fits using Eq. (2) typically have errors that can be ex-
plained in terms of statistical Poisson \( \sqrt{N} \) fluctuations in the
measured peak areas. As discussed below, this suggests that a
two-state Markov process is sufficient to describe the ob-
served memory effects. When absolute normalization was
not possible (due to insufficient count-rate data), relative nor-
malization was performed by fitting Eq. (2) to the data with
an additional fitting parameter, a normalization coefficient
multiplied by the entire right-hand side. This procedure was
necessary for the data in Fig. 4, while absolute normalization
was possible for the data from dots 3, 4, 6, and 7 in Figs. 6
and 7, presented below.

Both the negative and positive correlation effects in Fig. 4
display a strong dependence on laser excitation power. The
lower plots in Fig. 4(a) show data obtained from dot 1 under
above-band excitation, while the excitation power was varied
to produce mean count rates on one photon counter of 25, 50,
100, and 200 kcps (kilocounts/second), as indicated. The up-
per plot shows data obtained for the same dot, using the
same setup, with resonant excitation at 908.6 nm. The mea-
surements in Fig. 4(b) were taken from dot 2, another quan-
where \( I_0 \) and \( I \) are constants characterizing the saturation intensity and power, respectively. The normalized excitation power is \( p = P/P_0 \). This behavior is typical of an “incoherent” excitation process. On the other hand, a simple two-level system without dephasing excited on resonance is expected to undergo Rabi oscillations, characteristic of “coherent” excitation. This behavior can in fact occur in quantum dots when an isolated sharp resonance with little background can be found in the PLE spectrum. Partially coherent behavior is shown in Fig. 5(c) for a quantum dot on sample B emitting at 877 nm and excited at 864 nm. Oscillations in the emission intensity are seen as the laser power is increased. However, the quantum dots discussed elsewhere in this paper did not show this effect when excited at the chosen PLE resonances.

Finally, Figs. 6 and 7 summarize the behavior of the memory time scale \( \tau_b \) and amplitude \( g_1 \), both defined in Eq. (2), as a function of normalized excitation power for several quantum dots. In Fig. 6, dot 3 was excited at 750 nm, above the GaAs band gap. Figure 6(a) is a log-log plot showing the fitted value of \( \tau_b/T_{\text{rep}} \) as a function of \( p \). The points approximately follow a line of slope \(-1\), indicating that the blinking rate \( 1/\tau_b \) is approximately proportional to the excitation power. The memory time scale can be as long as 130 ns for small excitation powers, and less than one pulse period for large powers. For large enough powers, the memory time scale becomes so short that even the innermost peak areas in the photon correlation histogram have areas close to 1, and it becomes difficult to extract a value for \( \tau_b \). Figure 6(b) is a semilog plot showing the amplitude \( g_1 \) as a function of excitation power. A smaller data set from dot 1, corresponding to the plots in Fig. 4(a), is also shown. Figure 7 shows similar data for dots 2, 4, 5, 6, and 7 under resonant excitation. Various parameters of these quantum dots are summarized in Table I. Dots 2, 4, 5, and 6 are from sample A, while dot 7 is from sample B. The blinking rate \( 1/\tau_b \) is again approximately proportional to excitation power below the saturation regime (\( P/P_0 < 1 \)). The memory time scale was as long as 770 ns for dot 4 under weak excitation power. The blinking amplitude \( g_1 \) shows similar behavior as with above-band excitation, except that it is positive. The curve fits in these two figures are based on the theoretical model to be introduced next.

\[
\frac{I}{I_0} = 1 - e^{-P/P_0}, \quad (3)
\]

where \( I \) is the measured intensity, \( P \) is excitation power, and \( I_0 \) and \( P_0 \) are constants characterizing the saturation intensity and power, respectively. The normalized excitation power is \( p = P/P_0 \).
where $p_i$ was in state $i$ and $g_i$ is the probability to be in state $i$ before the pulse. One can then calculate $g(2)[n]$, obtaining a two-sided exponential function as in Eq. (2) with parameters,

$$
\begin{align}
\left(\begin{array}{c}
p_1 \\
p_2
\end{array}\right) & \rightarrow \left(\begin{array}{cc}
1-a & b \\
ap & 1-b
\end{array}\right) \left(\begin{array}{c}
p_1 \\
p_2
\end{array}\right),
\end{align}
$$

where $p_i$ is the probability to be in state $i$, and $a$ and $b$ are the $1 \to 2$ and $2 \to 1$ transition probabilities, respectively. We next assume that if we have detected a photon immediately after an excitation pulse, the system must have ended in state 1. This is reasonable in our experiment, since we spectrally select an emission line corresponding to a unique transition. If a photon was detected from pulse 0, the system evolves to

$$
\begin{align}
\left(\begin{array}{c}
p_1[m] \\
p_2[m]
\end{array}\right) & = \frac{1}{a+b} \left(\begin{array}{c}
b
\end{array}\right) + \frac{a(1-a-b)^m}{a+b} \left(\begin{array}{c}
1 \\
-1
\end{array}\right),
\end{align}
$$

where $p_i[m]$ is the probability of the system to be in state $i$ after pulse $m$. Finally, let $\eta_i$ be the probability of emitting a photon immediately after an excitation pulse, given the dot was in state $i$ before the pulse. One can then calculate $g(2)[n]$, obtaining a two-sided exponential function as in Eq. (2) with parameters,

\begin{table*}[h]
\centering
\caption{Parameters of the studied quantum dots. $\lambda_0$ and $\lambda_{exc}$ are emission and excitation wavelengths, respectively.}
\begin{tabular}{|c|cccccccc|}
\hline
dot No. & 1 & 2 & 3 & 4 & 5 & 6 & 7 \\
$\lambda_0$ (nm) & 932 & 920 & 919 & 938 & 940 & 921 & 876 \\
$\lambda_{exc}$ (nm) & 750/904 & 905 & 750 & 913 & 904 & 892 & 858 \\
$P_0$ ($\mu$W) & 1/30 & 190 & 70 & 3.1 & 115 & 28 & 446 \\
$C_1$ & 1.21/0.85 & 0.58 & -0.96 & 0.956 & 0.956 & 0.72 & 0.88 \\
$C_2$ & 0.58/1.6 & 2.9 & 0.51 & 0.97 & 0.84 & 2.2 & \\
\hline
\end{tabular}
\end{table*}

\begin{align}
\tau_b & = \frac{-T_{rep}}{\ln(1-a-b)}, \\
g_1 & = \frac{a(\eta_1-\eta_2)}{b\eta_1+a\eta_2}.
\end{align}

If $\eta_1 > \eta_2$, $g_1$ is positive, as observed experimentally with resonant excitation. If $\eta_1 < \eta_2$, $g_1$ is negative, as observed with above-band excitation. Another important parameter is the internal efficiency, that is, the probability of emitting a photon after any given excitation pulse. The result is

$$
\eta_{tot} = \frac{\eta_1+a\eta_2}{a+b}.
$$

We next consider the nature of states 1 and 2. They can have lifetimes approaching 1 $\mu$s, so they cannot be optically active states such as excitons, biexcitons, and trions, which have nanosecond lifetimes. For quantum dot 1 discussed above, spectroscopy suggests that the studied emission line is a neutral-exciton transition, and thus state 1 is the neutral ground state (empty dot). For state 2, the “dark” state, recent spectroscopy literature on self-assembled quantum dots\textsuperscript{37–39,34} and chemically synthesized nanocrystals\textsuperscript{40} suggest two main possibilities: a “dark exciton” or a charged state. Dark excitons are electron-hole pairs with spins oriented such that optical recombination is forbidden. Although these states can have microsecond lifetimes in chemically synthesized nanocrystals with typical diameters less than 5 nm, it is unlikely that they could be long lived in our self-assembled quantum dots. Self-assembled dots can have reduced symmetry, so that the dark-exciton transition is not entirely forbidden.\textsuperscript{34} Additionally, if the energy splitting between the bright and dark exciton levels is proportional to the inverse cube of the dot radius,\textsuperscript{40} spin-flip processes might occur much faster in our samples, with typical dot radii of 30 nm. Dark excitons in InAs quantum dots have not yet been adequately studied, and we cannot reject this possibility with certainty. Nevertheless, we favor the other possibility that state 2 is a charged state. This choice allows one to explain all of the observed memory effects with a single model. Charge fluctuations in quantum dots under continuous-wave above-band excitation have been observed through correlations between exciton and trion photon emissions,\textsuperscript{26} and we have seen similar behavior in our samples. The assumption below is that the extra charges reside inside the quantum dot.
FIG. 8. Energy levels and transitions for the blinking model described in the text: (a) a short period of laser excitation followed by (b) a long relaxation period.

An alternative possibility not considered here is that extra charges could be associated with impurities outside the quantum dot; this would not explain the negative correlations observed with above-band excitation.

The model is shown schematically in Fig. 8. Level 0 is the neutral ground state and level X is the single-exciton state. Level $e^-, h^+$ is a charged state. There could in reality be more than one charged state involved significantly with the dynamics, but we consider only one charged state in this model. Level $X^-$ is a trion (charged-exciton) state. Some of these levels can have degeneracies, but in these cases we are interested only in the total occupation probabilities.

The system evolves in two steps. First, a strong optical field is applied for a short-time duration $\Delta t$, during which upward transitions are induced. Two of these transitions, with rates $\alpha$ and $\beta$, change the total charge of the dot from neutral $\rightarrow$ charged and from charged $\rightarrow$ neutral, respectively. These transitions correspond physically to the capture of single electrons and holes from the surrounding region into the quantum dot. The third transition, with rate $x$, brings the quantum dot from 0 to X. This transition could occur either through resonant excitation or through the capture of an entire electron-hole pair. During the second step, which could last several nanoseconds, the system relaxes back down to levels 0 and $e^-, h^+$ through electron-hole recombination. This is assumed to be a charge-conserving process. If a relaxation occurs from X to 0, a photon is emitted at the special wavelength that our setup detects.

This model can be solved to give the following parameters for the general Markov process in Eq. (4):

\begin{align}
a &= \frac{\alpha}{\alpha + \beta} (1 - e^{-(\alpha + \beta) \Delta t}), \\
b &= \frac{\beta}{\alpha + \beta} (1 - e^{-(\alpha + \beta) \Delta t}), \\
\eta_1 &= 1 - a - e^{-(\alpha + x) \Delta t}, \\
\eta_2 &= b.
\end{align}

To fit this model to the experimental data, we make a further assumption that the rates $\alpha$, $\beta$, and $x$ are proportional to the excitation power $p$. Inserting Eqs. (8) into Eq. (7) gives the saturation behavior,

\begin{equation}
\eta_{\text{sat}} = \frac{\beta}{\alpha + \beta} (1 - e^{-(\alpha + \beta) \Delta t}).
\end{equation}

Comparing this with the empirical Eq. (3) allows the assignment $(\alpha + x) \Delta t = P/P_0 = p$. We then introduce two fitting constants, $C_1 = (x - \beta)/(x + \alpha)$ and $C_2 = \alpha/\beta$. Substituting these and Eqs. (8) into Eqs. (6) gives the final result:

\begin{equation}
\tau_b = \frac{T_{\text{rep}}}{(1 - C_1)p},
\end{equation}

\begin{equation}
g_1 = C_2 \left(\frac{e^{C_1 p} - 1}{e^{p} - 1}\right).
\end{equation}

The parameter $C_1$ has special importance, since it determines whether electrons and holes are more often added individually or in pairs. If they are added individually ($C_1 < 0$), Eq. (10b) predicts negative correlation ($g_1 < 0$) in the detected photons. If they are added more often in pairs ($C_1 > 0$), Eq. (10b) predicts positive correlation ($g_1 > 0$). These behaviors have simple qualitative explanations. It follows from Eqs. (1) and (2) that the quantity $1 + g_1$ is proportional to the conditional probability, given that a photon was detected from pulse 0, that a second photon will be detected from pulse 1. When a first photon is detected, the dot is empty immediately afterward. For single-carrier injection, two injections must occur before another photon can be emitted, and thus it is unlikely that another photon will be emitted from pulse 1 if the injection rate is small. The opposite argument applies for the injection of entire electron-hole pairs. If a photon was emitted from pulse 0, it is especially likely that another photon will be emitted from pulse 1, since only a single additional pair needs to be injected. Otherwise, the dot could be charged, in which case it will appear dark at the selected wavelength.

The lines in Figs. 6(a) and 7(a) were obtained by fitting Eq. (10a) to the measured blinking time scales. The values of $C_1$ obtained from these fits are summarized in Table I. As expected from the preceding discussion, $C_1$ is negative for above-band excitation and positive in all cases for resonant excitation. Substantial variation in the value of $C_1$ is seen among the different dots under resonant excitation, however. $C_1$ is much closer to its maximum value of 1 for dots 4 and 5, than for dots 2 and 6. In our model, $C_1$ characterizes how “clean” the resonant excitation process is, or in other words how rarely extra charges are added and removed from the quantum dot. Perhaps the differences among these dots are related to the emission wavelength. It is also possible that for one or more of these dots, the studied emission line could be a trion (charged exciton) transition. In those cases our model should still apply (after changing the labels in Fig. 8), but it would not be surprising if the values of the fitting parameters were different.

The curves in Figs. 6(b) and 7(b) were obtained by fitting Eq. (10b) to the data using the values of $C_1$ already obtained and using $C_2$ as a fitting parameter. The fits do not match the data perfectly, a sign that the model is too simple. The saturation regime ($P/P_0 > 1$) is difficult to model accurately,
since a large number of states are involved. The model does
correctly predict that $g_1$ tends to zero for large excitation
powers, as observed in the data. The parameter $C_2$ has prac-
tical importance, since according to Eq. (9), the maximum
internal efficiency at the selected wavelength is $\eta_{\text{max}}$
$= \beta/(\alpha + \beta) = 1/(1 + C_2)$. Using this formula with the fitted
values of $C_2$ may not give a quantitatively correct efficiency
in the saturation regime, where the model is least accurate.
Nevertheless, we expect the efficiency of a single-photon
device to suffer as a result of charge fluctuations, even far
into the saturation regime. The disappearance of memory
effects in photon correlation measurements at large excita-
tion power probably does not imply a lack of charge fluctua-
tion. Rather, in this case the charge of the quantum dot is
almost completely randomized after each pulse, with $\tau_b$
$< T_{\text{rep}}$. For each excitation pulse that leaves the dot in a
charged state, a photon cannot have been emitted at the
neutral-exciton wavelength, and hence the efficiency is re-
duced.

It may seem surprising that the charge of a quantum dot
should ever change under resonant excitation, for which the
excitation energy is tuned below the GaAs band gap and
below the InAs wetting layer band edge. Our previous stud-
ies on sample B provide evidence that this does, in fact,
occur. In Ref. 41, Fig. 1(a) shows a photoluminescence spec-
trum under above-band excitation of the dot 7 included in the
present study. Through photon-counting cross-correlation
measurements similar to those described in Ref. 26, we have
identified a neutral-exciton line, a biexciton line, and at least
two prominent charged-exciton lines. Under resonant excita-
tion, the charged-exciton lines become much weaker com-
pared with the above-band case. This does not imply that
states containing a single charge do not occur under resonant
excitation, but rather, if charged states occur, they are not
excited efficiently by the laser. This is a demonstration of the
selective nature of the excitation process. However, when the
excitation power is increased, the charged-exciton lines be-
come more prominent. This is apparent in Fig. 2(b) of Ref. 8.
Since the charged-exciton states decay radiatively into
single-charge states, this shows, first, that charged states can
occur even under resonant excitation. Second, this shows
that the selectivity of the excitation process decreases when
the laser power is increased into the saturation regime.

The exact cause of charge fluctuations under resonant exci-
tation requires some speculation. It could be related to the
“wetting layer tail” observed in PLE spectra. This feature has
been attributed to continuum states associated with the
combined wetting layer–quantum dot system.30 If these
states are not localized to a single-quantum dot, electrons
and holes excited into these states by a laser pulse could be
captured by different quantum dots, changing their charges,
for example. It has also been suggested that an Auger-type
process, which would allow an electron to escape from the
quantum dot, could play an important role in the relaxation
of electron-hole pairs from excited states.42,43 Two-photon
processes are an unlikely mechanism when the excitation
power is far below saturation. This is because the blinking
rate $1/\tau_b$ is observed to vary approximately linearly with the
excitation power.

V. CONCLUSIONS

We have observed submicrosecond memory effects in
quantum-dot photoluminescence on two samples, one with
optical microcavities and one without. The observed correla-
tions were positive when the excitation laser was resonant
with excited levels of the quantum dots, and negative when
the laser frequency was tuned above the GaAs bandgap.
These memory effects imply the existence of multiple long-
lived states, which are most likely states with different total
charge. If this interpretation is correct, it is apparently diffi-
cult to prevent the charge of a semiconductor quantum dot
from changing during optical excitation.

Fluctuations in the charge of a quantum dot could pose
serious difficulties for proposed applications. Light emitters
based on single quantum dots will have reduced internal ef-
ficiencies at a particular wavelength. Even though the blink-
ing seems to disappear at large excitation powers in photon
correlation measurements, our model suggests that the effi-
ciency still suffers, due to the presence of multiple configu-
rations. Optically induced charge fluctuations could also be
problematic in schemes for quantum computation that in-
volve optical control of single charges or excitons.

Better stability might be achieved using laser excitation
resonant with the fundamental exciton transition. We did not
attempt this because of the difficulty of removing scattered
laser light, but at least one group has performed single-dot
measurements under these conditions.44 Another option
might be to design the optical excitation process so that the
transition rate into an unwanted configuration is much
smaller than the transition rate out of it. In this case, the
quantum dot would spend most of its time in the desired
configuration. Finally, if charge fluctuations are, in fact, re-
lated to continuum states, then fabrication of quantum dots
with a low density and without a wetting layer might be
advantageous.

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