Time-Resolved and Time-Integrated Spectroscopy Studies of The Optical Properties of Silicon Quantum Dots.

Lap Van Dao, Xiaoming Wen, Peter Hannaford
ARC Centre of Excellence for Coherent X-Ray Science, Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, Melbourne, Australia 3122

ABSTRACT

Using time-resolved and time-integrated photoluminescence and spectrally resolved two-colour three-pulse photon echo spectroscopy we study the quantum confinement and dephasing properties of near spherical Si QDs with an average size of 4.3 nm. Filling of the low energy states and parabolic confinement of the quantum dot structures can be inferred from the dependence of the photoluminescence intensity on the detection wavelengths. A dephasing time of 1–1.8 ps which is slightly dependent on the quantum dot energy states can be measured. We show that the dephasing time of the electrons in the quantum dots is strongly influenced by the density of excited carriers.

Keywords: Optical properties of low-dimensional structures, Ultrafast optical measurements, Laser spectroscopy.

1. INTRODUCTION

At present the growth and study of quantum structures based on silicon, the most technologically important material for electronic device in the twentieth century, is attracting much attention. Efficient light emission from Si quantum structures has led to the current interest in silicon for optoelectronic devices, but the origin of the light emission from an indirect band-gap semiconductor is still under debate.

Because of the finite degeneracy of quantum dot (QD) states, state filling effects can be observed when only a few carriers populate the lowest dot states [1,2]. The relaxation of excited carriers from two-dimensional quantum states to zero-dimensional QD states or also from high energy QD states is very efficient due to Coulomb scattering [2] and the low energy states are always fully occupied when the QDs are excited with photons which have a higher energy than that of the QD ground states. The discrete energy states of QDs reduce the available phase space and the allowed scattering possibilities are restricted to only the very narrow QD resonance, which leads to an excitation dependence of the dephasing of the QD states [3]. Band filling and excitation-induced dephasing are also interesting topics for studying the optical properties of QDs.

While measurement of linear optical properties such as photoluminescence or absorption is used to determine the quality of structures, nonlinear coherent optical studies can provide valuable information about the coherence properties and dynamics of carriers in quantum structures which is important for the fabrication of many optoelectronic devices. Although the nonlinear signal intensity is rather low for semiconductor quantum dot (QD) samples and experimental difficulties arise from the dispersion of the dot size there are a number of experiments which can be considered for studying the optical properties with coherent laser pulses [4-7]. Four-wave-mixing (FWM) experiments [4], pump-probe spectroscopy [6], time resolved photoluminescence [8,9], and photon echo experiments [7,10] have been applied to the measurement of the dephasing properties of quantum dot systems.

In this paper we report studies of the linear and nonlinear properties of a silicon quantum dot ensemble. Using time-resolved and time-integrated photoluminescence and spectrally resolved two-colour three-pulse photon echo spectroscopy we study the quantum confinement and dephasing properties of near spherical Si QDs with an average size of 4.3 nm.

2. EXPERIMENTS

The silicon QD superlattices were fabricated by alternate deposition of silicon oxide and silicon-rich-oxide (SRO) with differing thicknesses. Details of the fabrication process have been reported elsewhere [11, 12]. The samples used for
In this study, the average dot size is 4.3 nm, and they are deposited on a sapphire substrate with a dot density of about $10^{12}$-$10^{13}$ dots/cm$^2$.

In the time-resolved and time-integrated photoluminescence (PL) experiment, the PL signal is dispersed by a 0.27 m grating spectrometer and detected with a photomultiplier tube (PMT). For the time-resolved measurements, the response time of the PMT is reduced to reach a resolution of about 5 ns, which is much shorter than the decay time of the signals (in the microsecond range) and the signal is recorded by a 500 MHz (5 Gs/s) digital oscilloscope or a boxcar system.

In the time-resolved and time-integrated measurements, the three-peak photon echo measurements use three sequential ultrashort pulses with wave vectors $k_1$, $k_2$, and $k_3$ and a pulse duration of about 100 fs. These pulses are applied to the sample. Three beams with time delays $t_{12}$ (between pulses $k_1$ and $k_2$) and $t_{23}$ (between pulses $k_2$ and $k_3$) are aligned in a triangular configuration and focused by a 20 cm focal length lens on the same spot (diameter 300 $\mu$m) where the sample is placed [13]. The nonlinear signal is recorded in the phase-matching direction $k_4 = k_3 - k_2 - k_1$.

The ultrafast pulses are generated by two independently tunable optical parametric amplifiers (OPA) pumped by a 1 mJ amplified pulse (80 fs, 800 nm, 1 kHz) with wavelengths in the range 600-850 nm.

### 3. RESULTS AND DISCUSSIONS

#### 3.1 Time resolved photoluminescence measurements

The time-integrated photoluminescence of the sample shows a very broad band (~350 meV) signal around 750 nm without structure [11]. The excitation intensity is about 10 mJ/cm$^2$ which suggests that we can generate more than 10 electrons/dot. This allows investigation of the carrier relaxation dynamics between distinct QD levels starting with several fully occupied QD levels. The PL intensities are measured at different wavelengths and the ratio of the intensity at each short wavelength ($I_\lambda$) to the intensity at 850 nm ($I_0$), which would be seen as luminescence from the ground dot level, is recorded.

Figure 1 shows the PL intensity at different wavelengths with a step size of 50 nm for the first 10 $\mu$s after excitation. The rise times of all recorded signals are limited by the temporal resolution of the detection system. This indicates that the capture time is shorter than 5 ns. The maximum of the PL intensity reflects the “filling” of the dot levels by photon-excited carriers and depends on the density of states of given dot levels. In short times after excitation, when the PL intensity is expected to reach a maximum, the influence of recombination and relaxation is very small and the dependence of the maximum PL intensity can be determined by

$$I_{PLi} \sim \eta_i N_i \sim \eta_i D_i / (1 + \alpha_i D_i / I),$$

where $I$ is laser intensity, $\eta_i$ is the quantum efficiency, $\alpha_i D_i$ is the saturation factor which depends on the optical properties of level $i$, and $D_i$ is the density of states of level $i$. For a structure with level degeneracy $g_i$ and dot density $D_d$, we can write $D_i = g_i D_d$.

The QD energy levels can be described by a harmonic oscillator model with parabolic quantum confinement [11]. The potential function can be written as $V_j = \hbar \omega_j r^2 / 2$, where $j$ represents an electron or a hole, $\omega_j$ is the frequency, $m^*$ is...
the effective mass, and \( r \) is the radial coordinate of the dot. The eigenenergies for a single electron or hole are \( E_{\text{lin}}^j = \hbar \omega_j (2l - |m| + 1) \) where \( j \) is the radial quantum number, \( m \) is the angular momentum quantum number, and \( \hbar \) is Planck’s constant. We can set \( i = 2l - |m| \), in which case the eigenenergy is of the form \( E_i^j = \hbar \omega_j (i + 1) \). If we take the spin degeneracy into account the degeneracy of each state is \( g_i = 2(i + 1) \), with \( i = 0, 1, 2, \ldots \). When the capture rate is independent of the dot energy level we find the ratio \( \frac{I_{\lambda_{\text{max}}}}{I_0_{\text{max}}} \sim g_i \), where \( I_{\lambda_{\text{max}}} \) is the maximum of the PL intensity of the ground dot level (850 nm) and \( I_0_{\text{max}} \) is the maximum of the PL intensity at a given wavelength. The degeneracy is close to \( g_n = 2(n+1) \), which shows that a parabolic potential is a good approximation for these QD structures.

3.2 Spectrally resolved two-colour three-pulse photon echo measurements

The time evolution of the nonlinear signal spectrum in the phase-matching direction \( \mathbf{k}_4 = \mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1 \) versus coherence time \( t_{12} \) is shown in Fig. 2 for a fixed \( t_{23} = 2 \) ps. The intensity of the signal at the peak wavelength with a spectral window of 1 nm is plotted against \( t_{12} \) in Fig. 2(A-D). The wavelength of the two pump pulses is fixed at 600 nm and the peak wavelength of the probe pulse is 640 nm (Fig. 2a), 670 nm (Fig. 2b), 705 nm (Fig. 2c) and 740 nm (Fig. 2d). The observed signal decays very rapidly for positive \( t_{12} \) and a peak shift (the shift of the signal maximum in time from \( t_{12} = 0 \)) is observed. Because the pulse sequence \( \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3 \) (\( t_{12} < 0 \)) favours the rephasing processes the observed signal is a photon echo signal. The peak shift and the fast decay of the signal for \( t_{12} > 0 \) results from the inhomogeneous broadening of the dots ensemble. The inhomogeneous broadening washes out the signal generated by free induction decay because of interference between the homogenous broadening modes [14]. An oscillation of the signal with time \( t_{12} \) is observed which is very clear for probe wavelengths of 705 nm and 740 nm. The period of the oscillations varies slightly with the wavelength of the probe pulse and is about 50-60 fs corresponding to an energy splitting of 70-90 meV. Photon echo signals are generated when the wavelength difference between the pump and probe pulses is much larger than the spectral bandwidth (~10 nm) and the observed spectra are broader than the spectrum of the probe pulse which indicates a long dephasing time of QD states and a phase memory during the relaxation from the excited state to the probed state. A broadening of the photon echo signal on the blue side of the probe spectrum for a long probe wavelength is related to the occupation of the high energy states. The photon echo intensity versus time delay \( t_{12} \) has an effective

![Figure 2: Time evolution of the signal spectrum versus coherence time \( t_{12} \) for fixed \( t_{23} = 2 \) ps. The wavelength of the two pump pulses is fixed with peak wavelength at 600 nm and the peak wavelength of the probe pulse is 640 nm (Fig. 2a), 670 nm (Fig. 2b), 705 nm (Fig. 2c) and 740 nm (Fig. 2d). The 2D plot (A-D) is the intensity of the signal at peak wavelength of the probe pulses with a spectral window of 1 nm versus delay \( t_{12} \).](image-url)
dephasing time $T_2$ [18]. When the two pulses $k_1$ and $k_2$ overlap in time a transient population grating with wave vector $k_2 - k_1$ is created. The probe pulse is diffracted by this grating in the $k_4$ direction and this also contributes to the observed signal. The lifetime of the population grating is related to the lifetime and diffusion time of the excited carriers [15].

To obtain more quantitative information about the origin of the decay processes versus $t_{23}$ we choose different combinations of laser wavelengths. The sapphire substrate thickness is 1 mm and the FWM signal from the sapphire substrate is very strong when the three pulses overlap in time; therefore we are not able to analyze the signal from the silicon dots for the case $t_{23} < 300$ fs. Figure 3 shows the time evolution of signal spectra versus delay $t_{23}$ for different laser wavelengths. In one set of measurements the wavelength of the two pump pulses is fixed at 600 nm and the signals are recorded for the three wavelengths of the probe pulse of 640 nm (Fig. 3a), 705 nm (Fig. 3b) and 740 nm (Fig. 3c). In the other set the wavelength of the probe pulse is fixed at 670 nm and the wavelength of the pump pulse is tuned to 600 nm (Fig. 3d), 640 nm (Fig. 3e) and 670 nm (Fig. 3f). Two decay components, with a short and a long (> 10 ns) decay time, can be seen from the time evolution of the signal intensities (Fig. 3A-F). The slow decay component is found to persist across the emitted photoluminescence band of the QDs and to be independent of the detuning of the laser pulses.

![Figure 3: Time evolution of the signal spectrum versus population time $t_{23}$ and the spectral intensity at peak wavelength of the probe pulse for different combinations of laser wavelengths.](image-url)
4. CONCLUSIONS

Using time-resolved and time-integrated photoluminescence and two-colour three-pulse photon echoes we have studied the dephasing time, the state-filling effect and the carrier lifetime of silicon quantum dot energy states. Filling of the low energy states and parabolic confinement of the quantum dot structures can be inferred from the dependence of the photoluminescence intensity on the excitation intensity. The emitted intensities from different quantum dot levels decay with a stretched exponential function in the range 2 – 100 µs depending on the observation wavelength and the dot size. A dephasing time of 1 – 1.8 ps which is slightly dependent on the quantum dot energy states can be measured. We show that the dephasing time of the electrons in the quantum dots is strongly influenced by the density of excited carriers.

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