Coherent nonlinear optical spectroscopy of single quantum dot excited states

E. T. Batteh,[a] Jun Cheng, Gang Chen,[b] and D. G. Steel[c]
Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan 48109-1120

D. Gammon, D. S. Katzer, and D. Park
Naval Research Laboratory, Washington, D.C. 20375-5347

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We report a coherent nonlinear optical signature of the excited states of two quantum dots. By comparing the nonlinear spectra with the linear photoluminescence excitation spectrum, a clear identification of excited states is possible. © *2004 American Institute of Physics.*
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Zero-dimensional confinement achieved in semiconductor quantum dots (QDs) leads to a delta-function-like density of states. Early linear experimental optical studies based on photoluminescence excitation (PLE) identified the discrete spectra and identified states which are likely to be part of the excited state manifold.[1–4] However, PLE resonances could arise from interdot relaxation which also has been identified in coherent nonlinear optical (CNO) studies.[5,6] The identification of excited states requires CNO techniques that can differentiate between energy transfer from a higher-energy state of one QD to a lower-energy state of a second QD and the case where both states are in the same QD. CNO spectroscopy is able to identify excited states of a particular QD. In this letter, we use nondegenerate CNO spectroscopy to improve our confidence in identifying excited states. This knowledge is important not only to provide a more complete understanding of the physics of these QDs but excited state luminescence has also been critical in read-out experiments to identify the quantum state of the system.[7]

The GaAs QD sample used in this study is naturally formed by interface fluctuations and is discussed in detail in Ref. 1. The zero-field selection rules are linear due to island elongation along the $[\bar{1}10]$ axis (Π_x polarization is defined as being aligned along the direction of elongation).[4]

The photoluminescence (PL) spectrum for the apertured sample is shown in Fig. 1 (lower trace) for excitation at $\sim$1631 meV. The excitation field is Π_x polarized. In the PL spectrum, two states of interest, A and B, are indicated by the arrows in Fig. 1. The spectrometer is set at each of the two arrows, and the PLE spectra of these states is measured. Figure 1 displays the two spectra with the open (solid) circles representing the PLE from state A (B), and the spectra are offset vertically for clarity. Similar spectra result when Π_y polarized excitation is used (data not shown here). The discrete lines seen in the PLE spectra are similar to what has been reported recently[1–4] as the excited states of single QDs.

Following the work done in Ref. 5, the CNO signal is obtained using two independently tunable, frequency stabilized (to $\sim$1 MHz), continuous-wave fields with a mutual coherence bandwidth of $\sim$3 MHz. Each of the two fields $[E_1(\Omega_1)$ and $E_2(\Omega_2)]$ is amplitude modulated at a different frequency so that the CNO signal, created by the third-order polarization of the form $E_i^* E_i E_j$, is homodyne detected with field E_j at the difference frequency of the modulators. Unlike PLE which is typically associated with linear absorption, the CNO response can report on much more complex behavior including optically induced coherence,[8] interdot interactions,[6,9] and intradot coupling (this work).

In the degenerate studies ($\Omega_1 = \Omega_2$), the CNO spectrum identifies the individual resonances. In the nondegenerate studies, Ω_1 is tuned to one of the resonances, Ω_2 is scanned over the spectrum, and the CNO response is homodyne detected with either or both of the transmitted fields. In the nondegenerate response, one can identify both the saturation of the resonantly excited state and interactions between states.

For this system of ground and excited state excitons, two ground state excitons $|01\rangle$ and $|10\rangle$ and two excited state excitons $|01\rangle'$ and $|10\rangle'$ can be excited from the crystal ground state $|00\rangle$ using either σ_- (dashed) and σ_+ (solid)

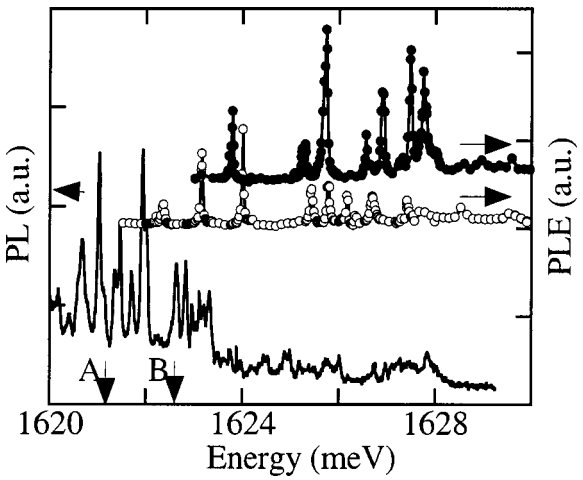

FIG. 1. PL and PLE spectra. The PL spectrum (lower trace) is compared with the PLE spectra of resonances A (open circles) and B (closed circles). The narrow resonances confirm the discrete QD spectrum.

[a]Present address: General Dynamics, 3300 Plymouth Road, Ann Arbor, MI 48105; electronic mail: elizabeth.batteh@gd-ais.com
[b]Present address: Bell Laboratories Lucent Technologies, 600 Mountain Avenue, Murray Hill, NJ 07974.
[c]Electronic mail: dst@eecs.umich.edu

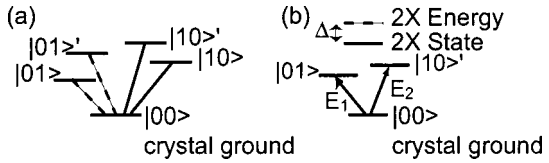

FIG. 2. (a) Ground state excitons $|01\rangle$ and $|10\rangle$ and excited state excitons $|01\rangle'$ and $|10\rangle'$ can each be excited from the crystal ground state $|00\rangle$ using σ_- (dashed) or σ_+ (solid) polarized light. (b) Experimental configuration for cross-polarized fields where selection rules allow for the formation of a two-exciton (2X) state at an energy Δ from the two-exciton (2X) energy.

polarized fields as shown in Fig. 2(a). The notation $|ij\rangle$ identifies which of the two electrons ($m_j = \pm 3/2$) is excited from the crystal ground state.

The model for cross-polarized excitation fields with E_1 resonant with $|01\rangle$ and E_2 resonant with the $|10\rangle'$ is shown in Fig. 2(b), analogous to the model described in Ref. 8 but with the ground and excited exciton states as levels $|01\rangle$ and $|10\rangle'$, respectively, and the two-exciton state made up of these two single exciton states. Similar to the results of Ref. 8, a peak will occur in the CNO signal that is homodyne detected with E_2 only when a Coulomb interaction occurs between the two single excitons. The following theory demonstrates that these results can be interpreted independently of an energy transfer model, leading to the conclusion of the resonant excitation of the excited state of a single QD.

To compare the CNO spectral features that distinguish between a resonant phenomenon and energy transfer, the third-order CNO response homodyne detected with E_2 is calculated for both cases using the density matrix formalism in the rotating-wave approximation.[10] α and β are two excitons with $\omega_\alpha < \omega_\beta$ where ω_i is the energy of exciton i. In the calculation, nearly degenerate (within the linewidth) orthogonal exciton states are included to keep the model consistent with the states occurring within a QD. The results are shown in Fig. 3.

The degenerate spectrum for α and β is shown in the lowest trace of Fig. 3. The upper two spectra [Figs. 3(a) and 3(b)] show the nondegenerate CNO response for the case where α and β are related through energy transfer from β to α. The arrows in Fig. 3 indicate the position of E_1 in each case. A CNO response is measured at E_1 in each spectra due to the presence of the nearly degenerate orthogonal exciton states. To study the relationship between α and β, the CNO response is examined at β when E_1 is resonant with α and vice versa. When E_1 is resonant with the higher-energy state β, the CNO signal is also measured at α. However, when E_1 is resonant with the lower-energy state α, no response is measured at β. A clear dependence of the CNO response on the position of E_1 is seen.

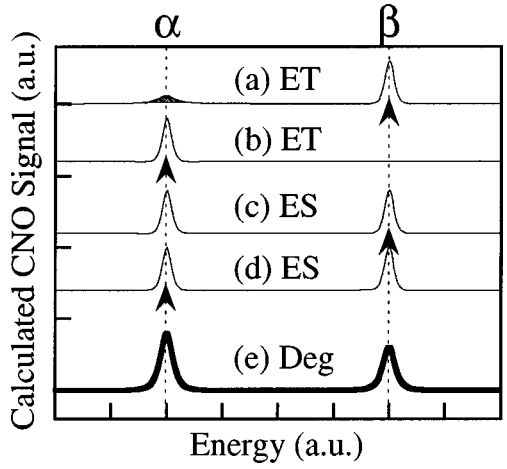

FIG. 3. Comparison of theoretical calculations. (e) The degenerate CNO spectrum showing the two resonances. (a) and (b) are the nondegenerate spectra for energy transfer (ET) from β to α with the arrows indicating the position of E_1. In this case, CNO signal occurs at a lower energy than E_1. For the case when β is the excited state (ES) of α, the two spectra are identical whether E_1 is resonant with (d) α or (c) β. Therefore, nondegenerate CNO spectroscopy with $\Omega_1 < \Omega_2$ can be used to distinguish between energy transfer and excited state behavior. The positive signal at E_1 is due to the CNO response from nearly degenerate orthogonal exciton states.

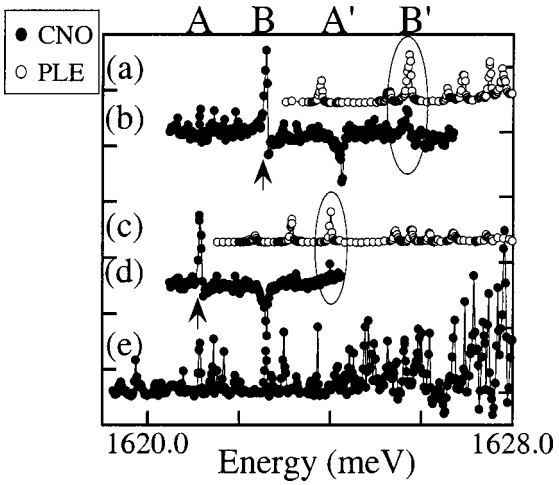

FIG. 4. Comparison of the PLE and CNO spectra for two states A and B. The arrows indicate both the position of E_1 for the CNO experiments and the spectrometer position for the linear experiments. For the CNO experiment, the excitation fields are cross-linearly polarized, and the CNO signal is homodyne detected with E_2. A peak appears in each of the CNO spectra [(b) and (d)] at E_1 due to the presence of the orthogonal ground state exciton. A second peak at higher energy also appears (A′ or B′) which coincides energetically with peaks in the linear PLE spectra [(a) and (c)], supporting the claim of excited state. The degenerate CNO signal is shown in (e). Negative signal indicates interactions between QDs.

The spectra in Figs. 3(c) and 3(d) indicate the CNO response when β is the excited state of α with the arrows showing the position of E_1. The CNO response occurring at E_1 results from the nearly degenerate orthogonal exciton state, and the relationship between α and β is understood by studying the CNO response that occurs away from E_1. In the excited state case, positive CNO signal occurs independent of the position of E_1.

Comparing the two spectra with $\Omega_1 < \Omega_2$ [Figs. 3(b) and 3(d)], the lack of CNO signal in Fig. 3(b) indicates the presence of energy transfer, whereas the existence of CNO signal in Fig. 3(d) indicates the resonant behavior of an excited state. Therefore, nondegenerate CNO spectroscopy with $\Omega_1 < \Omega_2$ is useful to distinguish between energy transfer and resonant excited state behavior.

Shown in Figs. 4(b) and 4(d) are the experimental nondegenerate CNO spectra for E_1 fixed at a ground state (A or B from Fig. 1 as indicated by the arrows) and E_2 scanned. The data are taken using linearly cross-polarized excitation fields, and the CNO signal is homodyne detected with E_2. In each of the two spectra, a peak appears at higher energy (~ 3 meV) than E_1. These peaks, labeled A′ and B′ are

tentatively assigned as the excited states of A and B, respectively. We also note the presence of an inverted resonance, corresponding to induced absorption, in both traces [Figs. 4(b) and 4(d)] at approximately 1622.6 meV and 1624.2 meV, respectively. This feature, similar to the biexciton signature discussed earlier,[11] is clearly not accounted for in the model above. We believe it is due to interdot coupling to be discussed in detail elsewhere.[9] Also noted in the two spectra are additional positive peaks that occur near resonances A and B. These peaks indicate the presence of the second ground state exciton (of opposite pseudo-spin) which have already been studied and discussed elsewhere.[8]

To confirm the excited state claim, the nondegenerate CNO signal is compared with the linear PLE [Figs. 4(a) and 4(c)] for B and A, respectively. At the same spectral position as the peak seen in the nondegenerate CNO spectra, a peak appears in the PLE spectra for both resonances (circled area). We believe that this is a strong indication that the particular peak seen in the nondegenerate CNO signal is nonlinear experimental evidence of an excited state of a single QD.

For both A and B, there were other resonances in the PLE spectra which did not have a detectable CNO signal. Based on the above discussion, this lack of a CNO signal indicates the absence of an interaction between the two excitons, such as Coulomb coupling. This difference, then, between PLE and the CNO response may indicate that the PLE resonances arise from energy transfer from a second dot, as we reported earlier.[5,6]

Identifying the CNO signature of the excited state extends the experimental demonstration of the predictions of the master equations for the low-dimensional electronic systems and further confirms their similarity to the atomic systems. These results will also be important if excited states in QDs become useful in the read-out process in quantum computing as discussed by Di Vincenzo and Loss[12] and demonstrated by Monroe *et al.*[7]

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