High-resolution spectral hole burning in InGaAs-GaAs quantum dots

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We report the use of continuous wave spectral hole burning to perform high-resolution spectroscopy of the homogeneous linewidth of self-assembled InGaAs-GaAs quantum dots at low temperature. We use this technique to examine the power broadening behavior of the homogeneous InGaAs-GaAs quantum dot line. We find that at a temperature of 9.8 K and over the majority of the pump powers considered, the spectral hole signal is well fit by a single Lorentizian line shape. Analysis of the power broadening yields a full width at half maximum of 0.74 μeV for the homogeneous linewidth and a corresponding coherence time T_2 of 1.76 ns.
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The basic physical properties of self-assembled semiconductor quantum dots (SAQDs), which lead to their atomic-like energy structure, make them a model semiconductor two-level system and of particular interest for numerous device applications. Potential quantum information applications in which excitonic-carrier populations in SAQDs are coherently controlled have recently become of considerable interest. As a result, the homogeneous line shape of the optical transition in SAQDs is an important subject of a variety of experimental and theoretical investigations. Traditionally the extremely broad inhomogeneous line shapes of SAQD ensembles have made it difficult to use linear optical measurements to resolve the relatively narrow homogeneous linewidth. Measurements to extract the homogeneous lineshape buried within the inhomogeneous spectra have been carried out on either a single quantum dot level[1,2] or in the time domain, by measurement of the decay of the "photon echo" after ultrafast excitation.[3] In this paper we present low temperature measurements of the homogeneous line shape using continuous wave spectral hole burning.[4] This technique has the advantage of sampling a small window of quantum dot transition energies in a large ensemble of SAQDs. While this technique has been applied to various solid state systems such as CdSe nanocrystals,[5] its application to the InGaAs SAQD system in our experimental geometry represents a novel application of this experimental method.

The samples employed in this study contain a single layer of InGaAs-GaAs SAQDs in an undoped semiconductor ridge waveguide. A schematic of the waveguide structure is shown in Fig. 1(a). The waveguide serves to increase the interaction length of the laser fields with the weakly absorbing SAQDs. Photoluminescence data of the SAQDs in our samples (shown in Fig. 1) indicate a ground state transition wavelength of 1008 nm at a temperature of 10.0 K, while room temperature data (not shown) yield a ground state transition at 1150 nm. We also note that Fig. 1 indicates the inhomogeneous line of the SAQD ensemble has a full width at half maximum (FWHM) of approximately 50 nm at 10.0 K. For spectral hole measurements the waveguides are mounted in a modified sample-in-vacuum He^4 flow cryostat with a base temperature of 9.8 K. At the experimental temperature the center wavelength for these measurements of 1064 nm ensures that the experiment examines only the ground state of the SAQDs.

The spectral hole measurements are performed by coupling light from two narrow-frequency external cavity diode lasers into the waveguide containing the SAQDs. The instantaneous linewidth for these lasers is <200 kHz, and we observe a heterodyne linewidth between the two lasers of ~4 MHz for an integration time of ~500 ms. The first of these sources remains at a fixed frequency and acts as the pump, burning a spectral hole in the inhomogeneous SAQD spectra. The second beam serves as the probe and is swept in frequency relative to the pump. The co-linear arrangement dictated by the waveguide geometry prevents the spatial discrimination of the probe signal from the much stronger pump light. Instead, signal discrimination is achieved by using a heterodyne technique. The beam from the probe laser is split into two beams, one beam is frequency offset from the other by an amount $\omega_r \sim 61$ MHz with an acousto-optic modulator. This frequency shifted beam is coupled into the waveguide and serves as the probe. The unperturbed beam from the same source is routed to an InGaAs photodiode at the waveguide output. This beam serves as a local oscillator reference for the probe. The resulting probe/reference beat note in the photodiode at ω_r is free of any dc contamination from the pump. The pump is intensity modulated at a frequency $\omega_p \sim 17.1$ kHz with an electro-optic modulator. This second modulation at ω_p produces a differential transmission signal ($\Delta T/T$) that is observed as frequency sidebands on the probe/reference beat note at $\omega_r \pm \omega_p$. This differential transmission signal is then recorded as a function of pump-probe laser

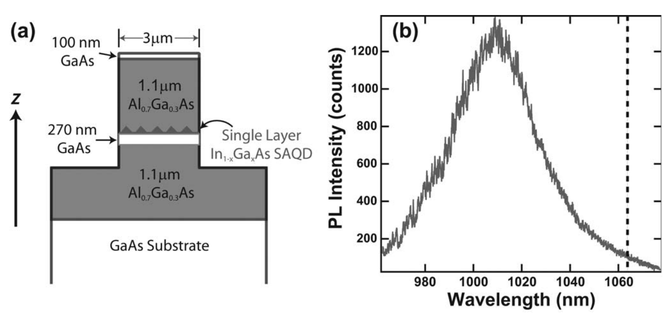

FIG. 1. Schematic of sample structures is shown in panel (a). The z axis is the growth direction for the sample. Panel (b) shows photoluminescence spectra taken at 10.0 K with an excitation power of 1.0 μW at 780 nm. The dashed line indicates the experimental wavelength of 1064 nm for the differential transmission measurements.

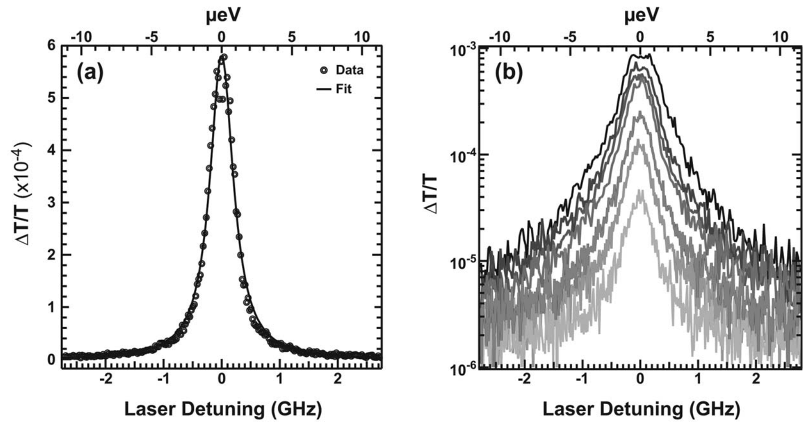

FIG. 2. Differential transmission signal ($\Delta T/T$) for single and multiple pump powers taken at 9.8 K. Panel (a) shows the $\Delta T/T$ for a pump power of 77 pW, along with a Lorentzian fit to the data. Panel (b) shows selected $\Delta T/T$ data for pump powers ranging from 2.5 (bottom curve) to 208 pW (top curve). The data in panel (b) show a uniformity in the line shape, despite a clear increase in the line width with increasing pump power.

detuning using a lock-in amplifier or directly with a radio frequency spectrum analyzer. The heterodyning of the probe and reference also provides amplification needed to observe the $\Delta T/T$ signal at the low probe beam powers which are required to limit self broadening of the spectral hole. In all measurements reported here the pump and probe are linearly co-polarized and, consistent with previous reports, we observe the absorption signal only when the pump/probe polarization is perpendicular to the growth direction z.[6]

The differential transmission data for an intermediate pump power is shown in Fig. 2(a) along with a fit to a Lorentzian. We note that the Lorentizian fit to $\Delta T/T$ signal is found to be numerically superior to a Voigtian line shape expected in the presence of inhomogeneous contributions. Unlike other studies using this technique we see no other evidence for either satellite peaks or the presence of inhomogeneous contributions to the line shape. Figure 2(b) shows InGaAs SAQD differential transmission signals taken at 9.8 K for a range of pump powers, all of which also display a uniform Lorentzian line shape. In addition to the uniformity of the line shape, evidence of power broadening is also visible in the data in Fig. 2(b). The power broadening behavior of a two-level system is described by a square-root dependence of the linewidth with pump power[7,8]

$$\Delta E_b = \Delta E_h (1 + P/P_0)^{1/2}. \quad (1)$$

In Eq. (1), ΔE_b is the power broadened peak FWHM, ΔE_h is the homogeneous FWHM, P is the pump power, and P_0 is the saturation pump power. A fit of the data to Eq. (1) permits the extraction of the homogeneous linewidth ΔE_h from the measured power broadened spectral hole linewidth ΔE_b.

In Fig. 3 the FWHM is shown over an extended range of pump powers along with a fit to Eq. (1). The fit in Fig. 3 yields $\Delta E_h = 0.74 \pm 0.09$ μeV, or equivalently, $T_2 = 1.76 \pm 0.21$ ns.[9] While the value of P_0 is subject to systematic errors and uncertainties in the pump power such as the coupling efficiency, the value for the homogeneous FWHM (ΔE_h) is robust against pump power uncertainties. The uncertainties in ΔE_h result from changes in the pump-probe laser detuning and are reflected in the error bars of Fig. 3 and the subsequently derived value for the coherence time.

From time-resolved photoluminescence (TRPL) data shown in Fig. 4 we are able to determine the radiative recombination time for the SAQDs examined in our spectral hole measurement. A radiative lifetime of 970 ± 100 ps is extracted by fitting the TRPL data with a single exponential convolved with the experimentally determined instrument response. While the uncertainty in the instrument response, compounded by systematic errors in the fitting limit the absolute accuracy, the 970 ± 100 ps value for the lifetime gives a radiatively limited T_2 value of 1.9 ± 0.2 ns and indicates that the T_2 determined by our power broadening analysis is near or just below the radiative limit. Previous studies of InGaAs quantum dots have reported homogeneous FWHMs (ΔE_h) in the range of 1.6–2.0 μeV, with corresponding coherence times T_2 between 680 and 830 ps.[1–3] In contrast, our results indicate a significantly narrower energy width of $\Delta E_h < 1$ μeV and a correspondingly longer T_2, despite the fact that we are working at a higher experimental temperature. We believe that the difference between our measurement and previous reports arises from small but significant physical differences in the experimental methods and systems. For example, in Ref. 3 the time-resolved four-wave-mixing studies were carried out on a doped system and due to the pulse bandwidth a much wider section of the inhomogeneous linewidth (2 nm) was examined, in comparison to the narrow range considered here. By designing the experiment to probe a narrow frequency range at the ground state energies of the SAQDs, we circumvent dephasing mechanisms induced by the inhomogeneous distribution of dipole moments that will pollute the homogeneous lineshape and

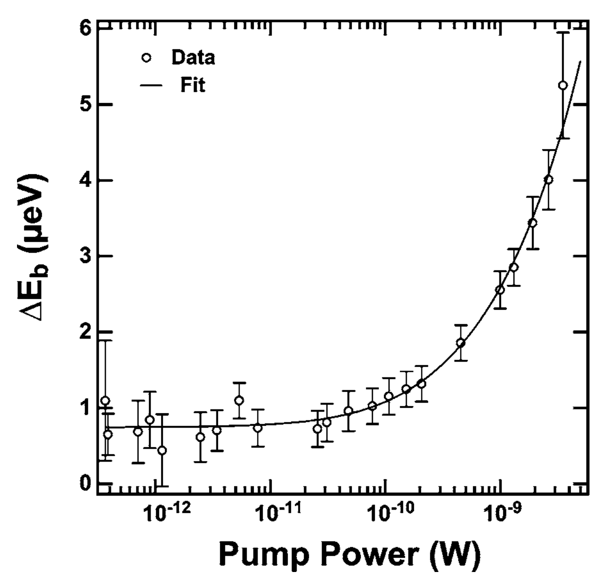

FIG. 3. Spectral hole full width at half maximum (ΔE_b) vs the pump power through the waveguide. Data are fit to $\Delta E_b = \Delta E_h (1 + P/P_0)^{1/2}$ and yield $\Delta E_h = 0.74 \pm 0.09$ μeV and $P_0 = 90 \pm 30$ pW.

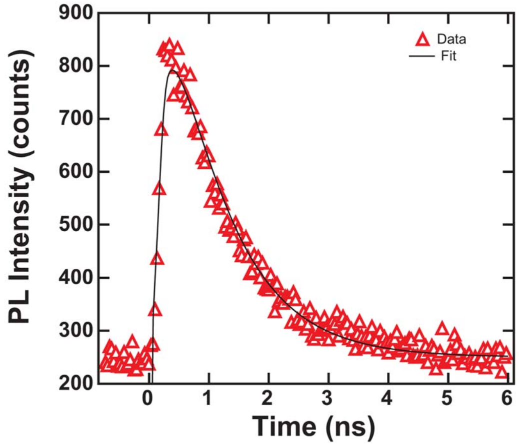

FIG. 4. Time-resolved photoluminescence data for SAQDs spectra at 1064 nm with a spectral resolution of 1.2 nm. Spectra was taken using a 1.0 μW average excitation power from a mode-locked Ti:Sapphire laser with a center wavelength of 780 nm and a 1.0 nm pulse bandwidth. Fit to data is a single exponential convolved with the instrumental response. Fit yields a radiative lifetime of 970±100 ps.

the subsequent T_2 value.[10,11] Other factors, such as static electric fields or spectral diffusion, that may contribute to the observed spectral hole width are mitigated by the intrinsic test structure and the underlying physics of the spectral hole burning technique.[12]

In conclusion, we have demonstrated the novel application of spectral hole burning to perform high resolution spectroscopy of SAQDs embedded in a ridge waveguide. This continuous wave pump-probe technique permits the differential transmission of a narrow energy range of SAQDs to be examined. The experimentally observed spectral holes are well fit by a Lorentzian line shape, and display power broadening behavior with increasing pump intensity. Analysis of the power broadening indicates a homogeneous FWHM of 0.74 μeV and a corresponding T_2 time of 1.76 ns. These results indicate a longer InGaAs-GaAs SAQDs coherence time compared with previously reported results.

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[1]M. Bayer and A. Forchel, Phys. Rev. B **65**, 041308 (2002).
[2]A. Högele, S. Seidl, M. Kroner, K. Karrai, R. J. Warburton, B. D. Gerardot, and P. M. Petroff, Phys. Rev. Lett. **93**, 217401 (2004).
[3]P. Borri, W. Langbein, S. Schneider, U. Woggon, R. L. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. Lett. **87**, 157401 (2001).
[4]M. Sargent, III, Phys. Rep. **43**, 223 (1978).
[5]P. Palinginis, S. Tavenner, M. Lonergan, and H. Wang, Phys. Rev. B **67**, 201307 (2003).
[6]K. L. Silverman, R. P. Mirin, S. Cundiff, and A. G. Norman, Appl. Phys. Lett. **82**, 4552 (2003).
[7]A. E. Siegman, *Lasers* (University Science Books, Sausalito, CA, 1986).
[8]L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms*, 2nd ed. (Dover, New York, 1987).
[9]The relation between the FWHM ΔE_h and the coherence time T_2 is given by $\Delta E_h = \hbar\Delta\omega = 2\hbar/T_2$. In the radiative limit, the lifetime is half the coherence time $(T_1 = \frac{1}{2}T_2)$.
[10]P. Borri, W. Langbein, S. Schneider, U. Woggon, R. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. B **66**, 081306 (2002).
[11]P. Borri, W. Langbein, S. Schneider, U. Woggon, R. L. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. Lett. **89**, 187401 (2002).
[12]R. Oulton, J. J. Finley, A. D. Ashmore, I. S. Gregory, D. J. Mowbray, M. S. Skolnick, M. J. Steer, S. Liew, M. A. Migliorato, and A. J. Cullis, Phys. Rev. B **66**, 045313 (2002).